Radiation Exposure by Natural Radionuclides in Drinking Water in Upper Austria

A Radioanalytical and Hydrogeological Research and Evaluation in an International Context

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Eingereicht von
Dipl.-Ing.
VALERIA GRUBER

Betreut von
Univ.-Doz. Dipl.-Ing. Dr. Franz Josef Maringer
Univ.-Prof. Dipl.-Ing. Dr. Martin Gerzabek

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Für meine Eltern

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(Trink dich satt am Wasser der hellen Quelle!)

Apuleius, *Der goldene Esel*

DAS BESTE ABER IST DAS WASSER.

Pindar, *Olympische Oden*

Contents

Abstract

The present thesis gives a fundamental overview on the topic of natural radionuclides in drinking water by presenting the technical basics and the current legal situation in Austria, Europe and the world with applied directives and recommendations as well as a summary of existing studies on this topic in different countries. The principle items of the study are the 350 water samples taken in the province of Upper Austria from 2004 to 2006. The samples were taken directly at wells and at consumers' houses and several times within the water flow, to get both hydrogeological-radiometric basis data and the effective exposure of the public. The samples were analyzed for different radionuclides (^{3}H, ^{222}Rn, ^{226}Ra, ^{228}Ra, ^{238}U, ^{210}Po and ^{210}Pb, gross alpha, gross beta) by LSC and ICP-MS in the laboratory. Secondary all samples of the detailed sampling phase were measured on-site for radon by the mobile liquid scintillation instrument Triathler. This method was implemented, tested and verified by comparison measurements within the thesis and will be established as a standard procedure for drinking water regulations monitoring in Upper Austria.

Correlations between different radionuclides were surveyed, and dependencies of radionuclide concentrations in drinking water on geological and hydrogeological factors were studied. This provides a basis for predictions of dose assessments for the public without measuring all radionuclides in the water. The activity concentration gradient within the flow of water from the well to the consumer and the impact of different water treatment techniques was investigated, which is especially interesting for precaution or remediation measures. Correlation to other drinking water parameters (pH-value, electric conductivity, temperature) and other elements (some heavy metals and metals) were surveyed. Finally dose calculations and estimations with dose models according to applied standards and alternative models were carried out. The results of the study provide a well-defined basis for adaptation and extension of legal regulations and for implementation of intervention measures. Therefore recommendations for radionuclide parameters for drinking water assessment have been worked out. These recommendations should help to standardize and simplify the experts' evaluation at official drinking water proceedings.

The various results and knowledge of this thesis about natural radioactivity in drinking water in Upper Austria were discussed according to different guidelines, directives and studies of Austria, Europe and the world.

Kurzfassung

Die Dissertation liefert einen grundlegenden Überblick über das Thema natürliche Radionuklide im Trinkwasser durch Darstellung der fachlichen Grundlagen und der aktuellen rechtlichen Situation in Österreich, Europa und international mit den derzeit gültigen Verordnungen, Richtlinien und Empfehlungen sowie eine Zusammenfassung von Studien zu diesem Thema in verschiedenen Ländern. Das Kernstück der Arbeit bilden 350 Wasserproben, die im Bundesland Oberösterreich zwischen 2004 und 2006 genommen wurden. Die Probennahme erfolgte innerhalb einer Wassereinheit sowohl direkt an den Quellen und Brunnen, als auch aus der Leitung beim Verbraucher, um einerseits hydrogeologisch-radiometrische Basisdaten zu erhalten und andererseits die effektive Exposition der Bevölkerung durch Radioaktivität im Trinkwasser abschätzen zu können. Die Proben wurden mittels LSC und ICP-MS im Labor auf verschiedene Radionuklide analysiert (^{3}H, ^{222}Rn, ^{226}Ra, ^{228}Ra, ^{238}U, ^{210}Po and ^{210}Pb, Gesamt-Alpha, Gesamt-Beta). Zusätzlich wurden die Proben vor Ort mit dem mobilen Flüssigszintillationsgerät Triathler auf ^{222}Rn gemessen. Diese Methode wurde im Rahmen dieser Arbeit adaptiert, getestet und durch Vergleichsmessungen verifiziert, und wird in Zukunft als Standardmethode für Trinkwasser-Monitoring in Oberösterreich eingeführt.

Es wurden Korrelationen zwischen den einzelnen Radionukliden und Abhängigkeiten der Radionuklidkonzentrationen im Trinkwasser von geologischen und hydrogeologischen Einflussfaktoren untersucht. Die Ergebnisse der Untersuchungen dienen als Basis, für Prognosen von Dosisabschätzungen für die Bevölkerung, ohne alle Nuklide im Wasser einzeln zu bestimmen. Aktivitätsverteilungen innerhalb einer Wassereinheit von der Quelle bis zum Verbraucher und der Einfluss von Wasseraufbereitung auf den Radionuklidgehalt im Wasser wurden ermittelt. Zusätzlich wurden die radiometrischen Ergebnisse mit Trinkwasserparametern (pH-Wert, Temperatur, Leitfähigkeit) und anderen im Wasser enthaltenen Elementen (einige Schwermetalle und Metalle) korreliert. Aus den ermittelten Radionuklid-Aktivitätskonzentrationsergebnissen im Trinkwasser wurden Dosisberechnungen und Dosisabschätzungen mit Dosismodellen entsprechend den gültigen Richtlinien, aber auch mit alternativen Modellen durchgeführt, bewertet und diskutiert.

Die Ergebnisse bieten eine Basis für die Überarbeitung und Erweiterung der rechtlichen Richtlinien und für die Durchführung von Eingreifmaßnahmen. Dafür wurde eine Empfehlung für die Radionuklid-Parameter zur Beurteilung von Trinkwasser erarbeitet. Diese soll helfen, die Arbeit der Gutachter in behördlichen Verfahren zu vereinfachen und zu standardisieren. Die Ergebnisse und Erkenntnisse dieser Dissertation über natürliche Radionuklide in Trinkwasser in Oberösterreich wurden in Hinblick auf unterschiedliche Richtlinien, Verordnungen und Studien aus Österreich, Europa und im internationalen Kontext diskutiert.

Chapter 1

Introduction

1.1 General Background

Drinking water is the most important food. Therefore its availability, quality and regulation are delicate and important topics.

> ”Water has to be suitable for drinking or using without endangering human health.” (Republik Österreich, 2001)

In Austria **ground water** is the major source of drinking water. 99% of the Austrian drinking water originates from ground water – half from pore groundwater sources in vales and basins, the other half from karst and crevice groundwater sources in the mountains (Lebensministerium, 2008a, Umweltbundesamt Wien, 2007b). 66% of the Austrian population is fed by **227 water supplies** with an average water take out $>$ 1000 m^3 per day or a supply of more than 5000 persons. This means a water output of 428.8 million m^3 per year (BMGFJ, 2007). 13% of the Austrian population uses drinking water from **private wells** (Umweltbundesamt Wien, 2007b). Radionuclides of the natural decay chains ^{238}U, ^{232}Th and ^{235}U are ubiquitous in the earth’s crust and therefore also available in ground and drinking waters.

So for this purpose it is fundamental to have an overview and hence reasonable regulations about **natural radioactivity in drinking water**.

In the EU the legal framework for regulation of radioactivity in drinking water is the **European Drinking Water Directive 98/83/EC** (European Commission, 1998) which was published in December 1998 and should be implemented by the member states within two years of coming into force of the directive (Risica & Grande, 2000). In the directive a minimum requirement on the quality of drinking water and water intended for human consumption is appointed. For radioactivity two indicative standard parameter limits are established – Tritium activity concentration of 100 Bq/l and total indicative dose TID (effective dose from radionuclides in drinking water except ^{3}H, ^{40}K, radon and radon progenies) of 0.1 mSv/a. The legal framework for exposure from natural radionuclides in drinking water in Austria is the **Drinking Water Regulation – TWV** (Republik Österreich, 2001) which implements the European Drinking Water Directive 98/83/EC (European Commission, 1998). The appointment and the evaluation of the TID are specified in the Austrian Standard ÖNORM S 5251 (Austrian Standards Institute, 2005b). The required measurement techniques (e.g. decision limit), sampling site and the evaluation methods including examples are specified there. Generally only the radionuclides ^{226}Ra and ^{228}Ra are taken into account for dose calculation of drinking water in Austria. Beside this Austrian standard there is a lack of regulation concerning other radionuclides e.g. ^{210}Po and ^{210}Pb.

Besides the TID several different reference values and guidelines exist concerning activity concentrations of radionuclides (WHO, EU, ICRP – see Chapter 3.1) and all these recommendations afford high responsibilities of the countries to establish their individual and detailed limitations and regulations. Furthermore from the practical point of view it is fundamental for the experts and operators to have directives not only for the total indicative dose but also for activity concentration values for different radionuclides (e.g. ^{226}Ra, ^{228}Ra, ^{238}U, ^{222}Rn, ^{210}Po, ^{210}Pb).

This lack of regulation concerning radionuclides like ^{210}Po and ^{210}Pb leads to different interpretations of measuring values in official proceedings and to individual use of different international recommendations (EC, WHO) by experts. This fact and the upcoming ”Ordinance on exposure due to natural radiation sources” (Republik Österreich, 2008) lead to a **drinking water pilot study in Upper Austria**. The project was one part of a two-part project ”*Strahlenexposition durch Trinkwasser in Oberösterreich – 2004–2006*” (Radiation exposure caused by drinking water in Upper Austria – 2004–2006), with this part ”*Bevölkerungsexposition*” (**Population radiation exposure**) (Gruber et al., 2006) and the other part ”*Strahlenexposition von Beschäftigten in oberösterreichischen Wasserwerken*” (**Radiation exposure of workers in Upper Austrian drinking water supplies**) (Ringer et al., 2006b).

This thesis is based on the ”Population radiation exposure” project supported and funded by the *Amt der oberösterreichischen Landesregierung, Direktion Umwelt und Wasserwirtschaft, Abteilung Umweltschutz, Strahlenschutz* (Office of the Upper Austrian Government, Department Environmental Protection). The sampling and measurements were carried out by the *Amt der oberösterreichischen Landesregierung, Strahlenschutz* in cooperation with the University of Natural Resources and Applied Life Sciences (BOKU), Low Level Counting Laboratory Arsenal. Scientifically the project was worked out in the frame of this thesis by Valeria Gruber, BOKU and published in a final report (Gruber et al., 2006) and a booklet for the Upper Austrian population (Gruber et al., 2007). The thesis was supported by the *National Metrology Institute of Austria (BEV)* in the course of the cooperation agreement of the Low Level Counting Laboratory. In this thesis the measurement results of the Upper Austrian drinking water project were scientifically evaluated to develop a comprehensive scientific view on the radioactivity of drinking water resources in Upper Austria. Besides the situation in Upper Austria was compared and discussed with results and studies in other countries and a general overview and contribution to the topic natural radioactivity in drinking water was given.

1.2 Goals of this Thesis

In the framework of this thesis several **aims and questions** are tried to be achieved, answered and discussed:

- Survey of a hydrological-radiometric basis data net of ground and spring waters used as drinking waters in Upper Austria
- Survey of population radiation exposure in Upper Austria caused by drinking water
- Discussion of existing national and international drinking water directives and recommendations
- Developing of own recommendations and guidelines concerning sampling and monitoring and defining parameter values for different radionuclides

To achieve these goals several questions have to be answered and steps must be accomplished:

- Testing of different measurement methods for radionuclides in drinking water and installing a routine measurement program for drinking water monitoring in Upper Austria
- Is there a correlation between different radionuclides in drinking water?
- Is there a correlation between radionuclide activity concentrations in drinking water and other physical or chemical parameters or other elements like (heavy) metals?
- Are radionuclide activity concentrations in drinking water dependent on geology and hydrogeology?
- Is there a difference between radionuclide activity concentrations in drinking water occurring from different groundwater, deep wells, private wells or spring water?
- Are radionuclide activity concentrations higher at wells or springs than at tap water at the consumers?
- Does water treatment in water supplies impact radionuclide activity concentrations in drinking water?
- Are the drinking waters in Upper Austria harmless for the population's health concerning natural radioactivity or are radiation protection measures necessary?
- Are the national and international drinking water directives and recommendations applicable and reasonable for Upper Austria and for the experts' work?

1.3 Proceedings and Presentations

Parts of results of this thesis will be also published in journals and were also presented and disucssed at different conferences and published in the proceedings.

Refereed papers

Gruber, V., Maringer, F.J., Landstetter, C., Radon and Other Natural Radionuclides in Drinking Water in Austria: Measurement and Assessment, Applied Radiation and Isotopes, Article in press, online published since 26.1.2009 (doi:10.1016/j.apradiso.2009.01.056)

Conference contributions and papers

Gruber, V., Maringer, F.J., Landstetter, C., Natural Occuring Radionuclides in drinking water: Standards, Measurement techniques and practical implementation, 5th International Conference on Radionuclide Metrology, Low Level Radioactivity Measurement Techniques, 22.–26. September 2008, Braunschweig, Germany (Oral)

Gruber, V., Maringer, F.J., Natural Radioactivity in Drinking Water: Regulations and Data in Austria and Europe, in: Regional and Global Aspects of Radiation Protection, IRPA Regional Congress for Central and Eastern Europe, 24.–28. September 2007, Brasov, Romania (Oral)

Gruber, V., Maringer, F.J., Kaineder, H., Sperker, S., Exposure and Regulation of natural radionuclides in drinking water in Austria, International Conference on Environmental Radioactivity: From Measurements and Assessments to Regulation, International Atomic Energy Agency (IAEA), Vienna, Austria, 23.–27. April 2007, IAEA Proceedings series (accepted)

Gruber, V., Maringer, F.J., Kaineder, H., Sperker, S., Exposition durch Radionuklide im Trinkwasser. Strahlenschutz aktuell, 40(2), 17–24; ISSN 1993-6273, 2006 – ÖSRAD conference, 22.6.2006 Vienna, Austria (Oral)

Gruber, V., Maringer, F.J., Kaineder, H., Brettner-Messler, R., Sperker, S., A survey of radioactivity in drinking water in upper Austria. In: European International Radiation Protection Association (Eds.), Second European IRPA Congress on Radiation Protection, 15.–19. May 2006, Paris, France

Gruber, V., Maringer, F.J, Katzlberger, C., Kaineder, H., Brettner-Messler, R., Survey of radioactivity in drinking water in Upper Austria by LSC and gammaspectroscopy. In: LSC2005, Advances in Liquid Scintillation Spectrometry, 17.–21. October 2005, Katowice, Poland (Oral)

1.4 Overview of the Configuration and Content of the Thesis

In this **Introduction** (Chapter 1) the motivation and background for the realization of this thesis are described and the goals, purposes and questions are discussed. Besides the conferences and proceedings in which parts of the thesis have already been presented are listed.

Chapter 2 (**General**, page 7) is designed to give an introduction to relevant topics concerning this thesis. Physical fundamentals to radioactivity and water are given, as well as biological effects of radioactivity and properties of radionuclides, relevant for this thesis, are discussed.

In Chapter 3 (**Drinking Water Regulations and Studies**, page 31) the existing regulations, directives and recommendations for natural radioactivity in drinking water in Austria, Europe and the world are collected and demonstrated. Additionally former studies about natural radioactivity in drinking water in Austria are presented and discussed. As an overview also the regulatory situation and existing studies in other selected countries of the world are surveyed.

General hydrogeological considerations and the drinking water and hydrogeology situation in Upper Austria are studied in Chapter 4 (**General Hydrogeology and Hydrogeology of Upper Austria**, page 45). Different ground water bodies and hydrogeological classifications of Upper Austria are presented, which are used in later chapters for the interpretation of results.

In Chapter 5 (**Materials and Methods**, page 55) the general realization of the practical part of the thesis is illustrated in detail – the selection of measurement points, sampling phases, sampling procedures and analytical methods.

For a better demonstration, comparability and interpretation of the activity concentration results a classification was implemented. This classification and further processing of the results is discussed in Chapter 6 (**Classification of Activity Concentrations**, page 75)

The following 10 chapters represent the results of the thesis concerning different questions and purposes: First the results of the survey sampling are represented in Chapter 7 (**Results of the Survey Sampling**, page 81). The results of the detailed sampling phase are discussed in Chapter 8 (**Results of the Detailed Sampling**, page 89), and in Chapter 9 (**Results of the "Sacred Wells"**, page 115) the results of the selected so called "sacred wells" are illustrated. In these 3 chapters the activity concentrations of the analyzed radionuclides are discussed according to the classification; median and maximum concentrations are displayed and cumulative frequency distributions are determined. Also the differences between the results of the survey and detailed sampling phase are discussed.

In Chapter 10 (**Comparison and Verification of Measurement Methods**, page 121) the on-site radon liquid scintillation counting measurement method (Triathler) is tested with different cocktails and Triathlers and sampling methods and bins are surveyed concerning their impact on radon activity concentration. Additionally the analyzed nuclide activity concentrations by different methods and laboratories are compared and discussed.

In Chapter 11 (**Correlations of Radionuclides**, page 143) selected sampling points with one or more higher nuclide activity concentration are demonstrated and discussed for radionuclide correlations or single occurrences. On the other hand general radionuclide correlations are surveyed with the measurement results of this thesis for scientific reasons, but also for the purpose of radiation protection, because of possible simplification of measurement methods or dose assessments.

The variation of the activity concentration of water on the way from the wells to the consumers and the influences of water treatment, elevated tanks and pipes are surveyed in Chapter 12 (**Activity Concentration Distributions in Water Units**, page 173).

Beside nuclide activities, also a couple of other parameters were analyzed in the water samples collected for this thesis: In Chapter 13 (**Impact of Physical Drinking Water Characteristics on Radioactivity**, page 189) pH value, electric conductivity and water temperature of the water samples are discussed and the correlation to nuclide activity concentrations is surveyed. Concentrations of several other non radioactive elements were analyzed in the water samples, particulary (heavy) metals. The results of these analyses and the correlations to the detected radionuclide activity concentrations are discussed in Chapter 14 (**Relation of Heavy Metals and Some Other Elements to the Radioactivity of Drinking Water**, page 197).

In Chapter 15 (**Natural Radioactivity in Water Regarding Geology and Hydrogeology**, page 209) the analyzed nuclide activity concentration results of the thesis are related to the geology where the samples were taken. Different geological and hydrogeological characterizations of Upper Austria are used for discussion. Additionally the difference of nuclide activity concentrations of waters from drilled wells and springs are surveyed.

The last chapter (Chapter 16 – **Dose Calculations**, page 243) of the results is related to dose assessments for the Upper Austrian population caused by natural radionuclides in drinking water. Dose estimations are done and discussed according to different directives, recommendations and other dose models.

Finally, the results of the thesis are summed up and interpreted in Chapter 17 (**Conclusions, Interpretation and Prospects**, page 267). Final statements and conclusions to the results are given and recommendations for drinking water sampling and directives are worked out. Further reasonable studies and steps are discussed in the chapter Prospects.

The references used within this thesis are listed in the **Bibliography**.

The thesis is completed with the List of Figures, the List of Tables and the tables with all analyzed radionuclide activity concentrations in the water samples, as well as the relevant legends in the **Annex**.

Chapter 2

General

2.1 Radioactivity, Physical

The atomic nucleus is made up of 2 kinds of nucleons – protons and neutrons. The total number of nucleons determines the mass of the atom and is called mass number A. The number of protons is the atomic number Z. Atoms with different numbers of neutrons but the same numbers of protons in their nuclei are called isotopes. The ability of atomic nuclei to convert spontaneous (without exterior action) under emitting **ionizing radiation** is called **radioactivity**. Ionizing Radiation has the property of producing ion pairs. Different modes of ionizing radiation exist: Alpha, beta and gamma radiation, X-rays, Auger electrons, cosmic radiation (p, n, light nuclei,. . .). . . In this thesis only alpha, beta and gamma radiation are of interest and therefore only these are discussed here.

Alpha radiation is a highly ionizing form of particle radiation with a high linear energy transfer (LET) and low penetration depth in matter, consisting of **helium nuclei** (2 protons, 2 neutrons). When an alpha particle is emitted, the atomic number of the atom decreases by 2, as the atom loses 2 protons. The new element (Equation 2.1) of high atomic masses (proton rich) has the energetic potential for emitting alpha particles. The emitted particle must have enough energy to get over the potential barrier in the nucleus (about 25 MeV) (Figure 2.1, Bröcker (1993)). Alpha particles can escape this barrier by the process of quantum tunneling (Bröcker (1993), Magill & Galy (2005), Figure 2.1). Figure 2.1 is explained in German, the green region is the potential curve, the red curve is the probability distribution of the alpha particle and the blue line demonstrates the energy level of the alpha particle (E_α).

$$(A,Z) \rightarrow (A-4, Z-2) +^4_2 He + \Delta E \tag{2.1}$$

$$E_\alpha = \frac{m_d}{m_d + m_\alpha} \cdot \Delta E \tag{2.2}$$

A	. . . mass number
Z	. . . atomic number
He	. . . alpha particle (Helium-nucleus)
ΔE	. . . liberated energy of the decay
E_α	. . . kinetic energy of the alpha particle
m_d	. . . mass of the daughter nuclide
m_α	. . . mass of the alpha particle

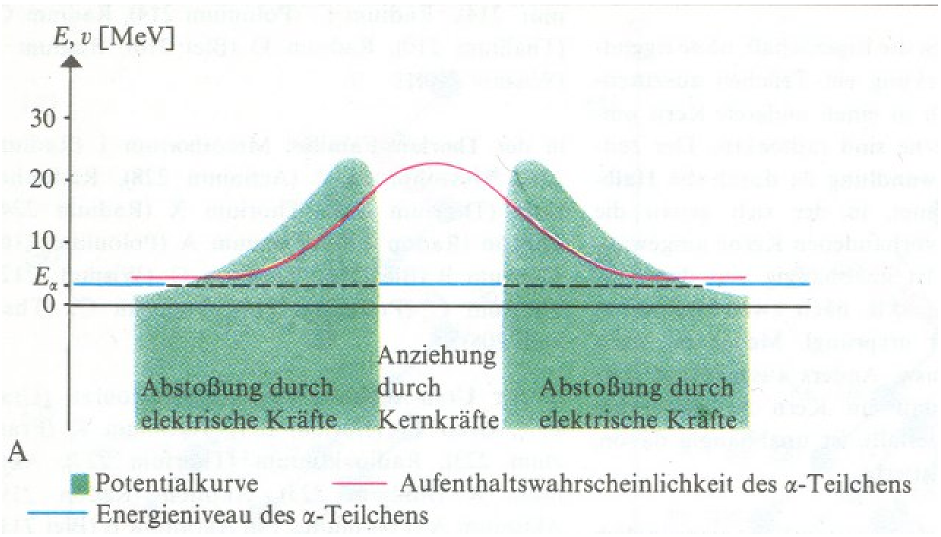

Figure 2.1: *Principle of alpha decay (Bröcker, 1993)*

The alpha particle receives the kinetic energy E_α (Equation 2.2) due to conservation of momentum. The energy E lies among 4 to 10 MeV (von Buttlar & Roth, 1990), all alpha particles of one isotope have the same discrete energy (monoenergetic), and so they have a **discrete, characteristic spectrum**.

Because of their charge and large mass, alpha particles are easily absorbed by materials and can reach only a few centimeters in air. They can be absorbed by the outer layers of human skin and thus are not generally dangerous to life, unless the source is ingested or inhaled. In this case they can be harmful, because of their high mass and strong absorption.

Beta radiation is a particle radiation of high energy **electrons** or **positrons**. Two forms of beta decay exist. β^--decay occurs, when the nuclide has an excess of neutrons. In the process a neutron is converted into a proton, an electron and an electron-type antineutrino are emitted (Equation 2.3). In nuclides where the neutron to proton ratio is to low, the nucleus may become more stable by β^+-decay. A proton is converted into a neutron, a positron and a neutrino are emitted (Equation 2.4). The atomic number increases by 1 (β^-) or decreases by 1 (β^+).

$$\beta^- : (A, Z) \rightarrow (A, Z+1) + e^- + \overline{\nu_e} \tag{2.3}$$

$$\beta^+ : (A, Z) \rightarrow (A, Z-1) + e^+ + \nu_e \tag{2.4}$$

A . . . mass number
Z . . . atomic number
e^- . . . electron
e^+ . . . positron
$\overline{\nu_e}$ . . . antineutrino
ν_e . . . neutrino

Beta particles have a **continuous spectrum** (different from alpha particles) of energies among zero and a maximum value (total decay energy, $T_{\beta,max}$), characteristic for that nuclide (Figure 2.2). The available energy of the decay is distributed among the electron and the antineutrino (positron and the neutrino). The maximum intensity of the Beta-spectrum is located among $1/2T_{\beta,max}$ und $1/3T_{\beta,max}$ (von Buttlar & Roth, 1990). The maximum energies of the beta particles range from 10 keV to 4 MeV (Magill & Galy, 2005). In Figure 2.2 the beta-spectrum of ^{40}K is illustrated with the energy in keV on the x-axis and the number of beta particles with the particular energies on the y-axis. The maximum energy of the beta particles of ^{40}K is about 400 keV.

Beta Particles can reach a few meters in air and can penetrate human tissue a few milimeters.

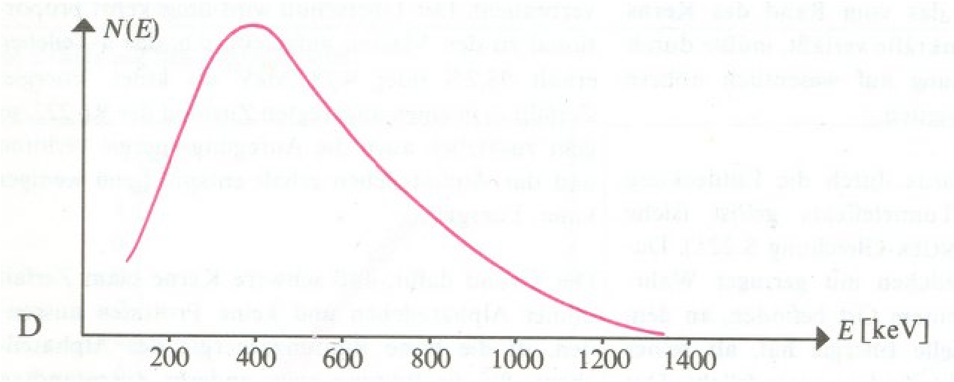

Figure 2.2: *Continous beta spectrum of the nuclide ^{40}K (Bröcker, 1993)*

An alpha or beta decay process may leave the product nucleus in an excited state. A nucleus in an excited state can return to the ground state most commonly by **emitting electromagnetic radiation** called **gamma radiation**. A gamma transition is characterized by a change in energy of the nucleus without change in Z or A (Equation 2.5). Gamma rays can penetrate the human body.

$$(A, Z)^* \rightarrow (A, Z) + \gamma \tag{2.5}$$

A	... mass number
Z	... atomic number
*	... excited state
γ	... photon

Radioactive decay is a statistical process in which the decay rate is proportional to the number of radioactive nuclei of a particular type present at any time t. The constant of proportionality (λ), termed the **decay constant**, is the probability of decay per unit time interval. It is related to the **half-life** ($T_{1/2}$) of a radionuclide (Equation 2.9), which is the time required for the decay of one half of the original number of its nuclei present The **activity** (a) is the number of decays per unit time interval. The activity of a number of radioactive atoms (N) is $N\lambda$. The rate of depletion is equal to the activity as long there is no new supply of radioactive atoms. (Equations 2.6– 2.8) (Ivanovich & Harmon, 1982)

$$\frac{dN}{dt} = -N \cdot \lambda = a \tag{2.6}$$

$$N_{(t)} = N_{(0)} \cdot e^{-\lambda \cdot t} \tag{2.7}$$

$$a_{(t)} = a_{(0)} \cdot e^{-\lambda \cdot t} \tag{2.8}$$

$$\lambda = \frac{ln2}{T_{1/2}} \tag{2.9}$$

$N_{(0)}$ is the number of radioactive atoms at time t=0; $N_{(t)}$ is the number of remaining radioactive atoms at some later time t.

The unit of the activity (a) is **Becquerel** (Bq), named after the French physicist Henri Becquerel (1852–1908), one of the discoverers of radioactivity. One Becquerel is one transformation per second in average [1 Bq = 1 s^{-1}].

The former unit was **Curie** (Ci), named after Marie Curie (1867–1934) and Pierre Curie (1859–1906), who discovered radium in 1898. 1 Ci = $3.7 \cdot 10^{10}$ Bq, which corresponds to the activity of 1 g ^{226}Ra.

The **specific activity** or **activity concentration** is related to mass [Bq/kg]; for liquids or gases usually the activity concentration related to volume [Bq/l, Bq/m^3] is used.

The **radioactive decay law** mentioned above takes into account only the decay of one single, separate nuclide transforming into a stable daughter nuclide. But very often the daughter product of a nuclear decay is also radioactive. This is termed

radioactive decay chain, and the decay and the appearance have to be considered together (Equation 2.10).

$$\frac{dN_i}{dt} = \lambda_{i-1} \cdot N_{i-1} - \lambda_i \cdot N_i \tag{2.10}$$

N_i . . . radioactive nuclei of the nuclide i at time t
λ_i . . . decay constant of the nuclide i

After some intermediate steps for the mother-daughter-transformation follows (Stolz, 2005):

$$a_2 = \frac{\lambda_2}{\lambda_2 - \lambda_1} \cdot a_{10} \cdot (e^{-\lambda_1 \cdot t} - e^{-\lambda_2 \cdot t}) = \frac{T_1}{T_1 - T_2} \cdot a_{10} \cdot (e^{-\frac{l_{n2}}{T_1} \cdot t} - e^{-\frac{ln2}{T_2} \cdot t}) \tag{2.11}$$

For multiple nuclides the **Bateman equation** can be applied:

$$a_n = a_{10} \cdot T_1 \cdot \left(K_1 \cdot e^{-\frac{ln2}{T_1} \cdot t} + K_2 \cdot e^{-\frac{ln2}{T_2} \cdot t} + \ldots + K_n \cdot e^{-\frac{ln2}{T_n} \cdot t}\right) \tag{2.12}$$

$$K_n = \frac{T_n^{n-2}}{(T_n - T_1) \cdot (T_n - T_2) \cdot \ldots \cdot (T_n - T_{n-1})} \tag{2.13}$$

a_n . . . activity of nuclide n
a_{10} . . . activity of the mother-nuclide at time t=0
$\lambda_{1,\ldots n}$ . . . decay constant of nuclide 1, . . . n
$T_{1,\ldots n}$ . . . half time of nuclide 1, . . . n

The formula above (Equation 2.13) shows that the time to reach **radioactive equilibrium** (a1/a2=const.) depends on the half-lifes of the parent and the daughter. Three cases can be distinguished. **Secular equilibrium** ($T_1 >> T_2$) arises if the half life of the parent is much longer than the one of the daughter – e.g. ^{226}Ra (T_1 = 1600a) and ^{222}Rn (T_2=3.8d). **Transient Equlibrium** ($T_1 \geq T_2$) occurs if the half-life of the daughter is of the same order but smaller than that of the parent – e.g. ^{140}Ba (T_1 = 12.75d) and ^{140}La (T_2 = 1.68d). If the half life of the parent is shorter than the one of the daughter ($T_1 < T_2$ **no equilibrium** arises. The daughter activity grows to a maximum and then decays with its own characteristic half-life – e.g. ^{146}Ce (T_1 = 13.5m) and ^{146}Pr (T_2 = 24.2m) (see Figure 2.3)

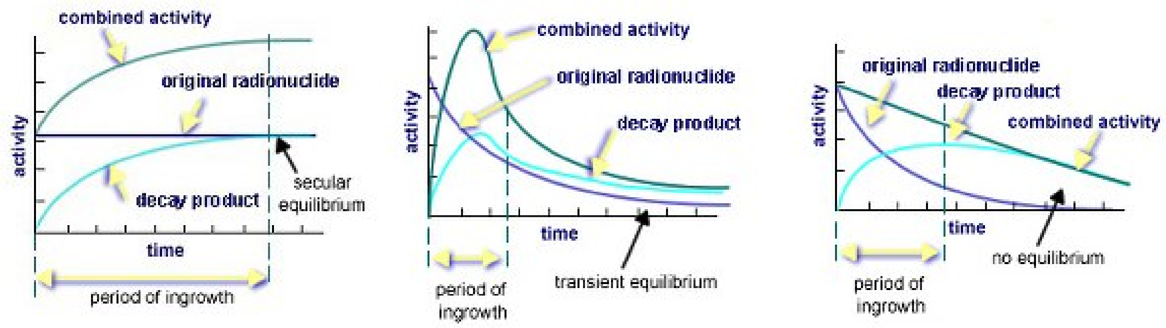

Figure 2.3: *Three cases of radioactive equilibrium – secular equilbrium, transient equilibrium and no equilibrium (EPA, 2009)*

2.2 Biological Effects of Ionising Radiation

After Eisenbud & Gesell (1997), Koelzer (2001), Magill & Galy (2005), von Buttlar & Roth (1990).

Ionising Radiation can ionise or excite atomes while passing through tissue. There are various quantities to specify the dose received and the biological effectiveness of that dose:

The **absorbed dose** (D) is the amount of the radiation energy (δE) absorbed per unit mass of material (δm). The unit of absorbed dose is Joule per kilogram, called after the British physicist Louis Harold Gray (1905–1965) 1 **gray** (Gy), the former unit is the Rad (rd). 1 Gy = 1 J/kg = 100 rd

It is useful in dosimetry to define an average dose for a tissue or organ D_T (Equation 2.14).

$$D_T = \frac{dE_T}{dm_T} \tag{2.14}$$

The **absorbed dose rate** is the rate at which an absorbed dose is received (Gy/s).

The **biological effect** of radiation depends not only on the energy deposited by radiation in an organism, but in addition on the type of radiation and the way in which the energy is deposited along the path of the radiation. So therefore the **linear energy transfer** (LET) is defined. It describes the mean energy deposited per unit path length in the absorbing material. The unit of the LET is keV/µm. So for the same absorbed dose, the biological effect of alpha particles or neutrons (high LET) is much greater than of beta or gamma rays (low LET). For characterisation of this difference in biological effects of different types of radiation, the **radiation weighting factor** w_R was established (Table 2.1) and is published in the ICRP recommendations 103 (ICRP, 2008). This recommendation was published in February 2008 and replaced the ICRP recommendation 60 (ICRP, 1991). For neutrons in

Table 2.1: *Radiation weighting factors for common radiations (ICRP, 2008)*

Type and energy range	**Radiation weighting factor,** w_R
Photons, electrons and muons, all energies	1
Protons, other than recoil protons, energy $>$ 2 MeV	5
Alpha particles, fission fragments, heavy nuclei	20

the new ICRP recommendations 103 (ICRP, 2008) an energy dependent continous function for w_R is defined.

The **equivalent dose** (H_T) shows the correlation between the absorbed dose and the biological effects. So the absorbed dose is multiplied by the weighting factor (w_R) of the radiation. If there are several types of radiation (R) present, the equivalent dose in the tissue (T) is the weighted sum over all contributions (Equation 2.15, (ICRP, 2008). The unit of the equivalent dose is also Joule per kilogram, because of the unit free weighting factor. For differentiation the unit of the equivalent dose is named after the Swedish doctor and physicist Rolf M. Sievert (1896–1966) 1 **sievert** (Sv). The former unit is rem (radiation equivalent man); 1 Sv = 100 rem.

$$H_T = \sum_R (w_R \cdot D_{T,R}) \tag{2.15}$$

The **equivalent dose rate** is the rate at which an equivalent dose is received, expressed for example in Sv/s or Sv/h.

The equivalent dose is always related to a defined tissue or organ. Different tissues and organs show different sensitivities to radiation, depending on their cell cleavage frequency and their cell renewal frequency. To take these effects into account, the equivalent doses in different tissues must be weighted (Table 2.2, ICRP (2008)). The equivalent dose (H_T) in tissue or organ T multiplied by **this tissue weighting factor** (w_T) is called the **effective dose** (E) (Equation 2.16, ICRP (1991)). In the new ICRP recommendations 103 the absorbed dose and the equivalent dose are calculated for women and men and for determination of the effective dose an averaged equivalent dose is used (ICRP, 2008).

$$E = \sum_T (w_T \cdot H_T) \tag{2.16}$$

Table 2.2: *Tissue weighting factors (ICRP, 2008)*

Tissue or Organ	**Tissue weighting factor,** w_T
Gonads	0.08
Bone marrow (red)	0.12
Colon	0.12
Lung	0.12
Stomach	0.12
Bladder	0.04
Breast	0.12
Liver	0.04
Esophagus	0.04
Thyorid	0.04
Skin	0.01
Bone Surface	0.01
Salivaries	0.01
Brain	0.01
Remainder	0.12

The sum of the relative weighting factors is one, this means that the sum of the weighting risks for the organs is numerical equal to the risk for the whole body (Tschurlovits, 2000).

Irradiation by gamma radiation outside the body causes only a dose during the period of irradiation. But by an intake through ingestion or inhalation some radionuclides can remain inside the body and irradiate the tissues for years. The total radiation dose in these cases depends on the half-life of the radionuclide, its distribution in the body, and the rate at which it is eliminated from the body. On the basis of mathematical models doses can be calculated with consideration of the following intake each year. The resulting total effective dose delivered over a lifetime is called the **committed effective dose**. The ICRP (European Commission, 1996) has published so called **effective dose coefficients**, values for committed doses following the intake of 1 Bq of a radionuclide via ingestion and inhalation. These coefficients have been calculated for members of the public at six standard ages and for intake by adult workers. The unit of the effective dose coefficient is Sv/Bq. The received dose via ingestion or inhalation of a radionuclide can be calculated as a product of the incorporated activity and the effective dose coefficient. In the Austrian *Allgemeine Strahlenschutzverordnung* these effective dose coefficients were adopted (Republik Österreich, 2006).

The **effective half life** ($T_{1/2,eff}$) is a degree of how long an incorporated radionuclide remains in the body (Haberer, 1989). The effective half life is composed of the **physical half life** ($T_{1/2,phys}$) and the **biological half life** (Equation 2.17). The physical half life has been described before. The biological half life ($T_{1/2,bio}$) is the time required for half of the amount of a certain substance to be removed from an organ or the whole body in a natural way (physiological excretion). (Diehl, 2003, Gerzabek, 2005)

$$\frac{1}{T_{1/2,eff}} = \frac{1}{T_{1/2,phys}} + \frac{1}{T_{1/2,bio}} \tag{2.17}$$

In the case of radionuclides with a long physical half life, the remaining time of the radionuclide in the body is dependent only on the biological half time. In case of a short physical half life the remaining time in the body results of an interaction of physical life time and physiological excretion. The biological half life and thus the effective half life of a radionuclide can differ for a certain organ or the whole organism. For example the biological half life of ^{131}I in the thyroid is 80 days, in the other organs only 7 days. The biological half time is also age dependent like the effective dose coefficients described above. (Diehl, 2003)

Radiation can affect people's health in two different ways called **deterministic effects** and **stochastic effects**. Deterministic effects are characterised by a threshold, below this threshold no damage is recognized – above it, the damage increases with dose. Deterministic effects are the acute radiation syndrome, which occurs immediately after an irradiation with high doses and damages, which occur at a later time, but induce no cancer (opacity of lens, vitiation of fertility). Immediate Symptoms after a whole body irradiation can be recognized above a dose between 0.5 and 1 Gy. In the range of 4 to 5 Gy 50% mortality would be expected, if the whole body dose goes up to 10 Gy, the mortality would reach 100%. (Eisenbud & Gesell, 1997)

Stochastic effects seem to have no threshold, only the probability of occurring increases with the dose. But different models for the dose-response relationship for induction of stochastic effects (see Figure 2.4) exist (Eisenbud&Gsell, 1997, Diehl, 2003). In Figure 2.4 some of these models are illustrated, extrapolated from known high doses to the region of low doses (Curve A describes a linear extrapolation, curve B a linear-quadratic extrapolation and curve C with a threshold level) (Diehl, 2003). Main stochastic effects are cancer, leukaemia and hereditary defects. These effects can appear years or decades after irradiation and can not be distinguished from ”common” cancer. (Tschurlovits, 2000)

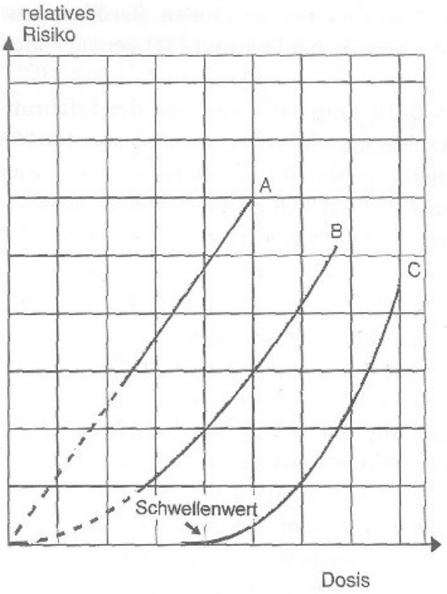

Figure 2.4: *Different models for dose-response curves (Diehl, 2003)*

2.3 Natural and Artificial Radioactivity

Radiation exposure of human beings is caused by **natural** and **artificial radiation sources**. Later in this work only the natural radioactivity in drinking water is taken into account, nevertheless in this chapter also the important artificial radiation sources are mentioned.

Another differentiation of radiation exposure of human beings is **external** (affected from outside) **exposure** and **internal exposure** by ingestion (drinking water, food) or inhalation (Diehl, 2003). In Figure 2.5 the sources and distributions of average radiation exposure for the world population are illustrated (WHO, 2004).

Figure 2.6 shows the average annual radiation dose of the Austrian population caused by different sources with a total of 4.6 mSv. The largest dose contribution is caused by inhalation of radon (about 2 mSv/a), about 1.3 mSv/a is caused by applications in medicine, about 1 mSv/a is caused by natural external radiation (cosmic and terrestric). About 0.29 mSv/a are caused by ingestion of natural radionuclides, including drinking water. Contributions to the annual dose of the population by other exposures are very small (applications in research and technology, Chernobyl accident, nuclear weapon tests and enrichment of natural occurring radioactive material (NORM) without radon).

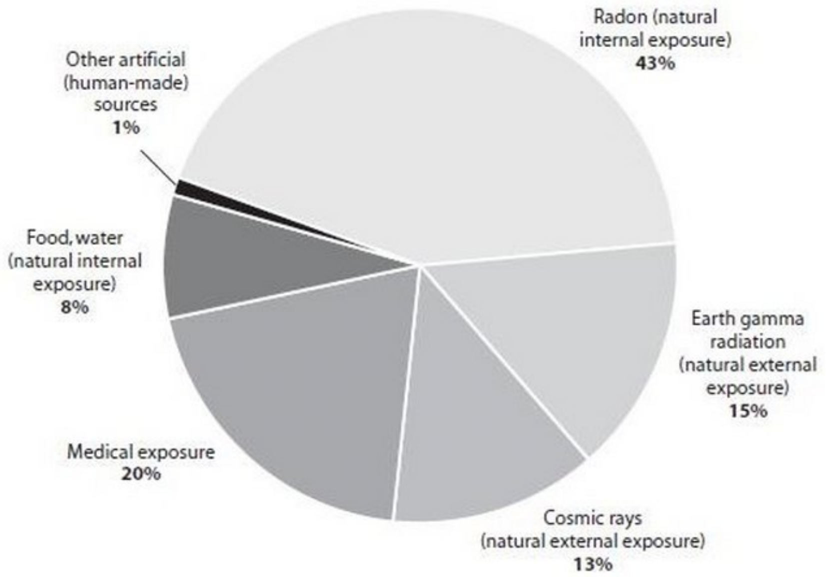

Figure 2.5: *Sources and Distribution of average radiation exposure for the world population (WHO, 2004)*

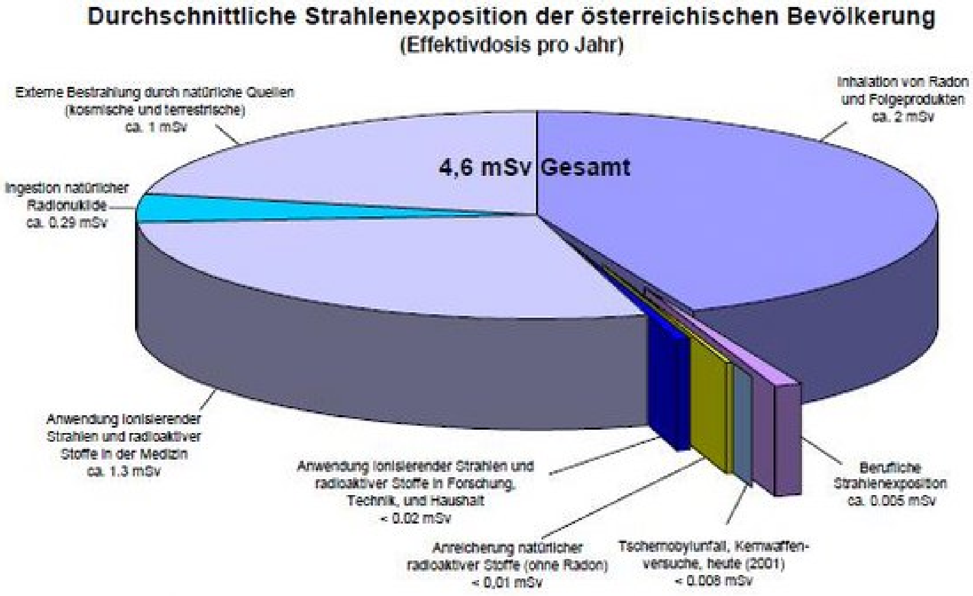

Figure 2.6: *Average annual radiation exposure of the Austrian population (Mück, 2001)*

The major contribution of artificial sources is the application of ionising radiation and radionuclides in medicine (X-ray diagnostics, radiotherapy, nuclear medicine). Other artificial sources are applications of ionising radiation and radionuclides in technology, research and homes, operation of nuclear power plants and the fallout from nuclear weapon tests in the 1950s and 1960s and nuclear accidents (most notably Chernobyl 1986).

The sources of natural radioactivity are divided into **cosmic radiation** (from sun and outer space) and **terrestrial radiation** (ground, building materials), which cause external exposure, and incorporated natural radioactive materials (Diehl, 2003, von Buttlar & Roth, 1990). Furthermore a classification based of the origin of the radionuclide exists: **cosmogenic** and **primordial radionuclides**. Cosmogenic radionuclides are accumulated currently by nuclear reactions of the cosmic radiation with aerosols. Of the 22 identified cosmogenic nuclides (Table 2.4) only four (^{3}H, ^{7}Be, ^{14}C, ^{22}Na) are significant from the perspective of doses to humans. Primordial radionuclides were produced at the origin of our solar system and because of their long half-life they still exist (for example ^{40}K, ^{87}Rb, ^{238}U, ^{235}U, ^{232}Th) (Table 2.4). ^{238}U, ^{235}U and ^{232}Th are the mother nuclides of the **three radioactive decay chains**, which generate most of the important natural radionuclides (for example ^{226}Ra, ^{228}Ra, ^{222}Rn), and each ends up in a stable lead isotope. The chains of radioactive elements are called **uranium series** (originates with ^{238}U, Figure 2.7), **thorium series** (originates with ^{232}Th, Figure 2.7) and **actinium series** (originates with ^{235}U, Figure 2.7). A fourth familiy, the neptunium series, which originated with the parent element ^{241}Pu, is known to have existed at one time, but this nuclide has a half-life of only 14 years and existed only briefly after its formation. The only surviving member of this family is the nuclide ^{209}Bi, which has a half life about $2 \cdot 10^{1}9$ years. From the 17 nonseries, primordial nuclides (Table 2.4) only ^{40}K and ^{87}Rb are of most interest. (Eisenbud & Gesell, 1997)

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000) has estimated that the global average annual human exposure from natural sources is 2.4 mSv/a (Table 2.3).

Table 2.3: *Average radiation dose from natural sources (UNSCEAR, 2000, WHO, 2004)*

Source	**Worldwide average annual effective dose (mSv)**	**Typical range (mSv)**
Cosmic rays	0.4	0.3–1.0
Terrestrial gamma rays	0.5	0.3–0.6
Internal exposure Inhalation (mainly radon)	1.2	0.2–10
Ingestion (food and drinking water)	0.3	0.2–0.8
Total	**2.4**	**1–10**

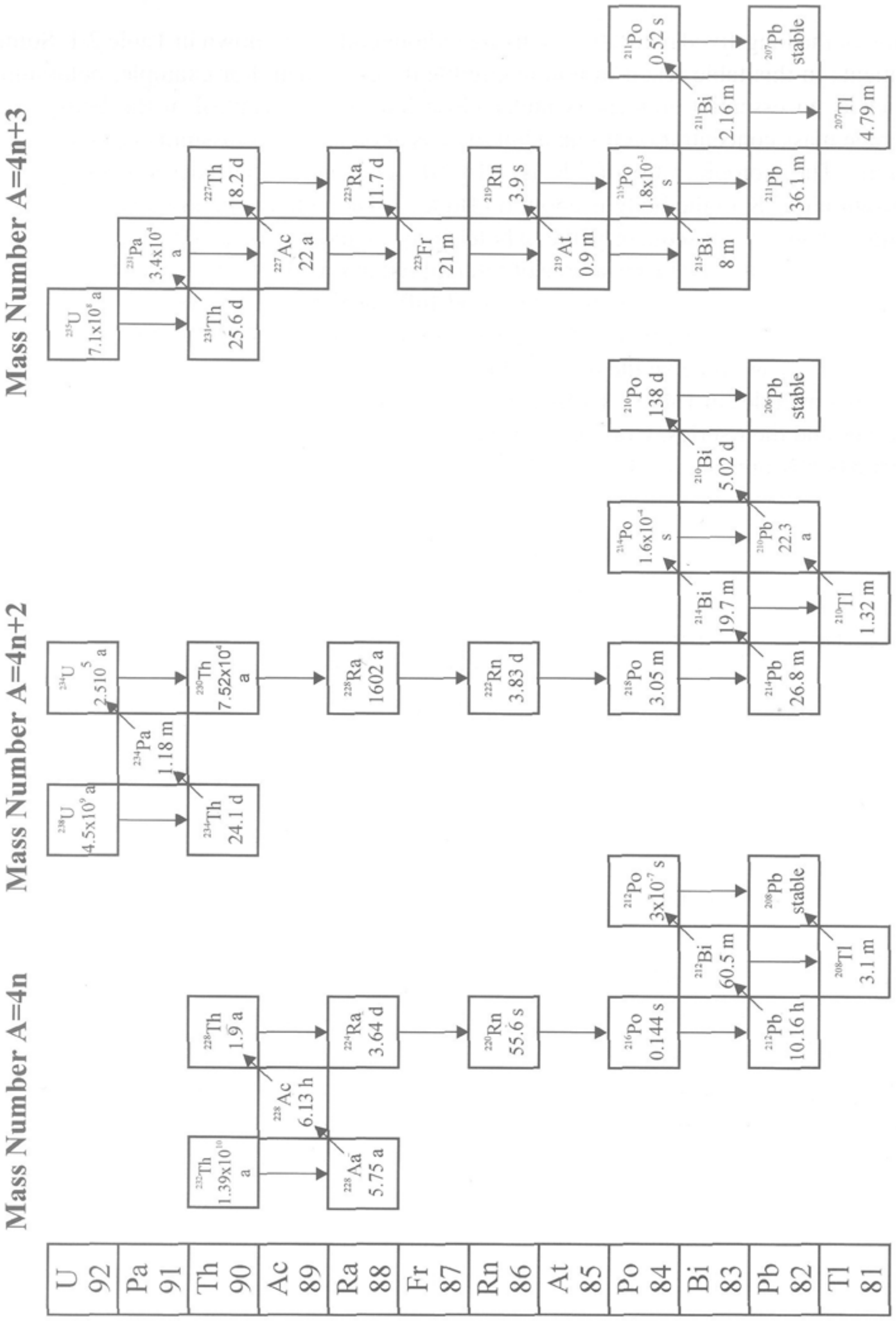

Figure 2.7: *The three radioactive decay chains – thorium series (originates with ^{232}Th), uranium series (originates with ^{238}U), and actinium series (originates with ^{235}U) (Valkovic, 2000)*

Table 2.4: *Nonseries Primordial Radionuclides (Eisenbud & Gesell, 1997)*

Radionuclide	Half-life (y)	Major radiations	Typical crustal concentration (Bq/kg)
^{40}K	$1.26 \cdot 10^{9}$	β, γ	$6.3 \cdot 10^{2}$
^{87}Rb	$4.80 \cdot 10^{10}$	β	$7 \cdot 10^{1}$
^{147}Sm	$1.05 \cdot 10^{11}$	α	$7 \cdot 10^{-1}$
^{176}Lu	$2.20 \cdot 10^{10}$	e^{-}, γ	$4 \cdot 10^{-2}$
^{138}La	$1.12 \cdot 10^{11}$	β, γ	$2 \cdot 10^{-2}$
^{187}Re	$4.30 \cdot 10^{10}$	β	$1 \cdot 10^{-3}$
^{144}Nd	$2.40 \cdot 10^{15}$	α	$3 \cdot 10^{-4}$
^{115}In	$6.00 \cdot 10^{14}$	β	$2 \cdot 10^{-5}$
^{50}V	$6.00 \cdot 10^{15}$	γ	$2 \cdot 10^{-5}$
^{123}Te	$1.20 \cdot 10^{13}$	X rays	$2 \cdot 10^{-7}$
^{142}Ce	$>5.00 \cdot 10^{16}$	Not reported	$<1 \cdot 10^{-5}$
^{152}Gd	$1.10 \cdot 10^{14}$	α	$7 \cdot 10^{-6}$
^{192}Pt	$1.00 \cdot 10^{15}$	α	$3 \cdot 10^{-6}$
^{113}Cd	$>1.30 \cdot 10^{15}$	Not reported	$<2 \cdot 10^{-6}$
^{174}Hf	$2.00 \cdot 10^{15}$	α	$2 \cdot 10^{-7}$
^{190}Pt	$6.90 \cdot 10^{11}$	α	$7 \cdot 10^{-8}$
^{209}Bi	$>2.00 \cdot 10^{18}$	α	$<4 \cdot 10^{-9}$

2.4 The Substance Water

Earth is the only planet in our solar system, of which a huge part (app. 70%) of the surface is covered with liquid water (Bliefert, 1994). On earth, water occurs in 3 states of aggregation (solid, liquid, gaseous), dependent on temperature and pressure, as shown in the **phase diagram** (Figure 2.8). The physical properties of water are highly dependent on temperature and pressure and show various **anomalies**.

Most liquids contract while freezing. In contrast water expands and has its highest density (volume minimum) under normal pressure at approximately 4 °C (**density anomaly**, Figure 2.9). Also the melting- and freezing point of water are clearly different to analogue built compounds of the adjacent elements of the same group of the periodic table (H_2S, H_2Se, H_2Te). The melting points of these compounds are in the range of -60°C to -2°C and the freezing points -86°C to -49°C. In contrast water boils under normal conditions at 100°C and freezes at 0°C. The anomalies of water are caused by the build-up of the water molecule and the ability of building hydrogen bonds. One molecule of water has two hydrogen atoms covalently bonded to a single oxygen atom and is geometrically angulated by 105°. Since oxygen and hydrogen have different electronegativities, the O-H-bond is polarized, the water molecule builds a dipole. The water molecules build up hydrogen bonds, because of interactions of the electron pairs at the oxygen atom with hydrogen atoms of other water molecules. One water molecule overall can be involved in four hydrogen bonds because of its two hydrogen atoms and electron pairs (”tetrahedral short-range order”). (Bliefert, 1994)

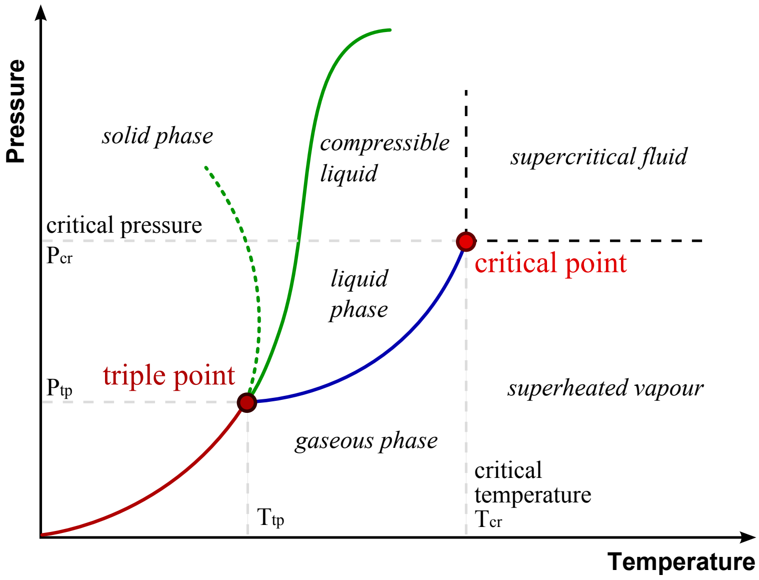

Figure 2.8: *Phase diagramm of water (Wikimedia Foundation Inc., 2008b)*

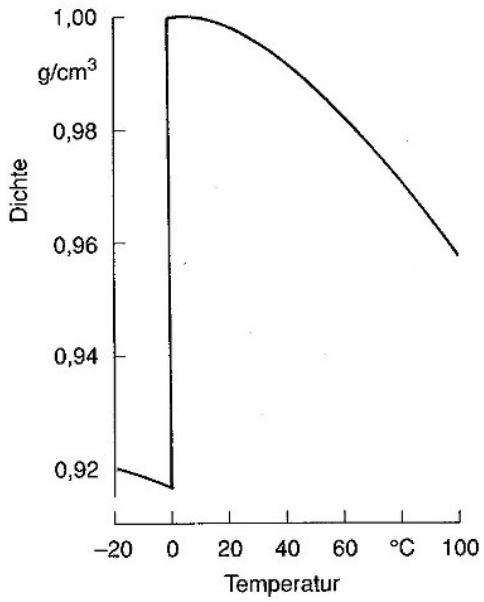

Figure 2.9: *Anomaly of water – density of water as a function of temperature (Bliefert, 1994)*

Because of its **dipol character** water is a very strong solvent, dissolving many types of substances. Therefore in nature water never occurs purely, without dissolved minerals, gases or organic matter (Deflorin, 2004).

Minerals are solved into water because of the contact of ground or surface water with soil and rocks. The chemical elements are usually ionized thereby and occur as cation (positively charged ion) or anion (negatively charged ion). The most common cations in ground or surface waters are sodium (Na^+), potassium (K^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}). The most common anions are bicarbonates (HCO_3^-), sulfates (SO_4^{2-}), chlorides (Cl^-) and silicates (SiO_4^{4-}). The most common ions in water occur in the range of part-per-million (ppm), which means that one milligram of an element is solved in one litre of water (mg/l). Besides, marginal elements exist in water in the range below 1 ppm, but above 1 part-per-billion (ppb – equates to µg/l) and trace elements in the scale of below 1 ppb. Marginal elements are e.g. Al, As, Ba, B, Be, Br, Cr, F, I, Fe, Li, Mn, P, Rb, Sr, Ti, V, Zn. Trace elements are among others Cd, Co, Cs, Au, Hg, Ni, Se, Ag, Sn, Tl, Wo, U (Sullivan et al., 2005). In Chapter 14 more details are discussed about these elements solute in water. Regulations about substances in water are discussed in Chapter 3.

An important parameter in this context is the relative acidity or alkalinity of water, which is determined by measuring the **pH value**. The pH value of water is affected by different chemical reactions with different minerals of ground and rocks, but also with the atmosphere. The pH value for natural water can range from 6 to 9 (Sullivan et al. (2005), see also Chapter 13).

Two-thirds of the human body consist of water and the cells consist of 80% water. The daily water demand of a person amounts to approximately 35 g/kg, related to the body weight. For a 70 kg person this leads to about 2.5 l drinking water per day, thus 50 000 to 60 000 l in a life. (Bliefert, 1994)

Drinking water is water which is applicable for human consumption and usage. Therefore a number of directives and quidelines exist, which are discussed in Chapter 3.

2.5 Properties and Effects of Certain Radionuclides

2.5.1 Radon

Radon (Z=86) is an inodorous, tasteless and colourless **noble gas**, occurring as nonpolar, monatomic molecules and is inert for practical purposes. ^{222}Rn, referred to as **radon**, originates from the ^{238}U decay series when ^{226}Ra decays by α-emission and has a half life of 3.82 days. ^{220}Rn, called **thoron**, with a short half life of 55.6 seconds, is a progeny of the ^{232}Th decay series. ^{219}Rn, **actinion**, is a member of the ^{235}U chain and decays most rapidly, with a half-life of about 3.92 seconds. Due to the short half life of ^{219}Rn and the fact that the percentage of ^{235}U in natural uranium is only 0.7%, no important amount of ^{219}Rn can be originating. ^{222}Rn has a greater opportunity than the shorter-lived radon isotope ^{220}Rn to escape into the atmosphere, therefore the thoron concentration in air is usually essential lower than the radon concentration. All 3 radon-isotopes are α-emitters. (Eisenbud & Gesell, 1997, Firestone & Ekström, 2008, Nero, 1988)

Radon **originates** in the grain of rock where the mother radium is located. When a radon atom originates near the surface of a grain, it can recoil into a pore between grains. The amount of radon that reaches the pores is described by the **emanation fraction** (NRC, 1999). Typically emanation fraction ranges from 10–40% in soils and bedrocks, and up to 70% in clay. The proportion of radon that emanates is dependent on the mineral grain size, composition, cracking and degree of weathering, and on how the radium atoms are located and the radium content (Clavensjö & Åkerblom, 1994). The radon atom has a kinetic energy of 86 keV caused by recoil of the α-decay of the radium atom, and therefore can cover different distances, dependent from density and compound of the material, until the energy is transferred to the surrounding material. In the grain of rock this distance is about 0.02–0.07 µm, in water 0.1 µm and in air 63 µm (Nazaroff et al., 1988). Usually the pores between grains of material contain a mixture of air and water, and only the radon atoms in these pores are available in soil gas and groundwater. (Nazaroff et al., 1988, Nero, 1988)

The radon concentration in the earth's crust in average is 400 fg/kg. The levels of radon in air vary widely according to the geological nature of the ground, being low in areas of basalt and high in areas rich in granite. Thoron concentrations are much lower and the radiation doses delivered to the population by ^{220}Rn are less than one tenth of those that result from radon. (Taylor, 2002)

Radon has a high solubility in cold water. This solubility decreases with increasing temperature (Cothern & Smith (1987), see Chapter 13.1). Elevated radon levels in groundwater have been found in crystalline rock formation, especially granite, whereas sedimentary rock usually yields lower levels. (Eisenbud & Gesell, 1997)

According to Böhm (2002) the radon concentration in ground water is influenced by different factors like uranium concentration in the host rock, permeability of the aquifer and the form of the covering layers.

Different studies in the USA report that **private well sources** and small water supplies tend to be higher in radon than large public water supplies. Private well sources and small water supplies incline to be in aquifers with low capacity, which often are uranium bearing granite, metamorphic rocks or fault zones. Large public water supplies tend to use high capacity sand and gravel aquifers, which have usually low uranium content (Eisenbud & Gesell, 1997, NRC, 1999). In areas with high radon activity in water, indoor radon concentrations can be elevated and the population using private wells may have higher radon exposures (Nero, 1988). This knowledge has been confirmed by surveys in member states of the European Union. Some groundwaters showed elevated radon concentration, particularly in regions with crystalline rocks. High concentrations are often related to individual drilled wells, but sometimes also to waterworks using water origination in rock or soil aquifers.

Radon concentrations in drinking water could be radiologically significant if they expose the population to enhanced doses and should not be disregarded from the radiation-protection point of view (European Commission, 2001). UNSCEAR and the National Research Council estimate, that a radon activity concentration of 1000 Bq/l in tap water increases the indoor radon activity concentration by 100 Bq/m^3 in average. The increase depends on various parameters such as the total consumption of water in the house, the volume of the house and the ventilation rate. (European Commission, 2001)

For the population radon in **tap water** causes exposure by **ingestion** and **inhalation**. UNSCEAR estimated the committed effective dose from ingestion of radon in water is 10^{-8} Sv/Bq for an adult (UNSCEAR, 1993), the National Research Council presented a dose conversion factor of $0.35 \cdot 10^{-8}$ (NRC, 1999). Estimations on the annual committed effective dose absorbed by an adult by ingestion of water containing 1000 Bq/l vary between 0.2 mSv and 1.8 mSv, depending on the annual water consumption and the range of conversion factors used (European Commission, 2001). Considering the ingestion and inhalation pathways, the annual effective dose caused by water containing 1000 Bq/l radon is, according to current knowledge, very comparable to that caused by an indoor radon concentration of 200 Bq/m^3 – the designed level established in Recommendation 90/143/Euratom (European Commission, 1990), which is also implemented in Austria in the standard ÖNORM-S 5280-2 (Austrian Standards Institute, 2003).

The inhalation exposure to radon progenies arising from the release of radon from potable water is of greater significance than the ingestion exposure (Nazaroff et al., 1988). So this may be a major problem for workers in water supplies (see studies: Ringer et al. (2006a,b), Trautmannsheimer (2002))

^{222}Rn was the first occupational respiratory carcinogen to be identified, because of high mortality from respiratory diseases among underground metal miners in the Erz mountains of eastern Europe since the early 20th century. Evidence of radon and **lung cancer** is available from about 20 epidemilogic studies of underground miners. (BEIR VI, 1999)

The results and deliverables of recent studies show, that the exposure to the noble gas radon and its decay products is the dominant source of exposure to ionizing radiation in most countries (WHO, 2000). Smoking multiplies the risk of Rn-induced lung cancer enormously (EPA, 1993). Darby et al. (2005) combined the results of 13 important European studies, which results in an indicated increase of lung cancer risk of 16% per 100 Bq/m^3 indoor radon activity concentrations. The inhaled radon becomes deeply lodged in the lung and especially the radon progenies can radiate and damage the cells and their DNAs of mucous membranes, bronchi and other pulmonary tissue. The ionizing radiation affecting the bronchial epithelial cells is believed to initiate the progress of carcinogenesis (EPA, 1993).

2.5.2 Radium

Radium (Z=88) is an **earth-alkaline metal** and behaves chemically similar to barium and calcium (IAEA, 1984). In nature four radium isotopes with mass numbers 223, 224, 226 and 228, originating from the decay of different thorium isotopes, exist. ^{223}Ra originates from the ^{235}U decay series (Figure 2.7) and has a short half life of 11.44 days. ^{224}Ra has also a short half life of 3.66 days and originates from ^{232}Th series (Figure 2.7), as well as ^{228}Ra, with a half life of 5.75 years. The Radium isotope with the longest half life is ^{226}Ra with 1600 years. It originates from the ^{238}U decay series (Figure 2.7). Besides ^{228}Ra, which decays by β-decay, all Radium isotopes are α-emitters. (Firestone & Ekström, 2008)

^{226}Ra is a very **rare metal**, nevertheless it is present in all rocks and soil in variable amounts. The radium concentration in the earth's crust in average is 900 pg/kg. (Taylor, 2002)

The IAEA (1990) reports normal ^{226}Ra concentrations in soil of about 3–140 Bq/kg. Much higher concentrations are found in certain regions in India, Brazil and New Zeeland (IAEA, 1984). Few data exist about normal ^{228}Ra concentrations in soil, because of required more advanced measurement technique to measure ^{228}Ra, but it seems to have the same magnitude than ^{226}Ra concentration (Diehl, 2003).

Igneous rocks tend to contain higher radium concentrations than sandstones and limestones. ^{226}Ra is generally in approximate equilibrium with ^{238}U. Although there is more ^{232}Th than ^{238}U in nature on an activity basis, there are geochemical factors which cause local concentrations of uranium, which often results in greater amounts of ^{226}Ra relative to ^{228}Ra in water. The radium content of surface water is low compared to most groundwaters. (Eisenbud & Gesell, 1997)

Dissolved radium sorbs quickly to solids and does not migrate far from its place of release to groundwater. Campos et al. (1986) reported that the annual mobilization rate of ^{226}Ra by groundwater solubilization is on the order of 10^{-7}. One of the principal reasons for the slow rate of mobilization was shown to be tenacity with which radium is sorbed on clays and organic matter (Eisenbud & Gesell, 1997).

In the United States the Environmental Protection Agency (EPA) conducted a survey of radioactivity in 1000 drinking water supply systems which obtain water supply from groundwater. The activity concentration average of all samples is 15 Bq/m^3 (15 mBq/l) ^{226}Ra and 26 Bq/m^3 (26 mBq/l) ^{228}Ra. The fact that the average values for ^{228}Ra are a factor of two larger than for ^{226}Ra is probably because of the larger minimum reporting level for ^{228}Ra (Eisenbud & Gesell, 1997, EPA, 1991). Early data suggested that under normal circumstances the ^{228}Ra content of food, water and human tissue is from one-half to one-fourth of the ^{226}Ra content (UNSCEAR, 1966). More recent data suggest that ^{228}Ra content of food is only slightly less than the ^{226}Ra content (Eisenbud & Gesell, 1997)

Since earth-alkaline elements are highly electropositive (2^+ valence state) radium is not prone to hydrolysis over a wide pH-range and is present in solution mainly as ionic species [Ra^{2+}] (Wisser, 2003).

The absorption of radium from the human gastrointestinal tract is assumed to average 20% (ICRP, 1996). Radium absorbed from the gastrointestinal tract deposits mainly in the **skeleton** (70–90%), where it may substitute calcium and strontium in human bone cells and it remains intact and can cause radiation damages by emitting particles (Krieger, 2002, Taylor, 2002). The dose conversion factors for ^{228}Ra are high, for instance more than 10 times higher than for ^{238}U. The effective dose of regions with average radium activity is about 7 µSv/a (Krieger, 2002).

2.5.3 Uranium

Uranium (Z=92) is the **heaviest naturally occurring element** and exists as a silvery-white, ductile and slightly **paramagnetic metal** and is a little softer than steel (Wisser, 2003). Natural uranium consists of three isotopes with mass numbers 234, 235 and 238. ^{238}U is present in the earth's crust in the isotopic ratio of 99.28% and is usually in radioactive equilibrium with ^{234}U, which is present in the isotopic ratio of 0.0058%. ^{235}U is present in the isotopic ratio of 0.71% (Eisenbud & Gesell, 1997, U.S. Department of Health and Human Services, 1999). All uranium isotopes slowly decay by emitting particles, with half life $4.47 \cdot 10^9$ years for ^{238}U, $2.455 \cdot 10^5$ years for ^{234}U and $7.038 \cdot 10^8$ years for ^{235}U (Firestone & Ekström, 2008). With percental isotopic ratios and half lifes an mass ratio ^{235}U/^{238}U is calculated to about 1/138 and an activity ratio to about 1/21.5 (Deflorin, 2004).

Uranium is a member of the **actinide family**, a series of elements similar to the lanthanide (rare earth) family in some respects. Chemically uranium resembles elements of Group VI of the periodic table and thus has some marked similarities to chromium, molybdenum and tungsten. Uranium is an electropositive element and the uranium atom has 6 electrons exterior to the radon electron core, a feature which permits four oxidation states: U^{3+}, U^{4+}, U^{5+} and U^{6+}. In nature only the 4 and 6 oxidation states are stable and of interest in geochemistry. The common uranium ions in the 4 and 6 oxidation states are U^{4+} and $(UO_2)^{2+}$ respectively (IAEA, 1988). The $(UO_2)^{2+}$ builds different soluble complexes with anions like CO_3^{2-}, SO_4^{2-} and Cl^- which are found in ground and surface waters. If $(UO_2)^{2+}$ is available soluble, sorption with organic matter or FeO(OH) is likely.

Uranium is found in all **rocks and soils**, in common rock types **uranium concentrations** range from 0.5 to 4.7 ppm. Acid igneous rocks for example contain concentrations 100 times higher than the ones in ultrabasic igneous rocks. (Eisenbud & Gesell, 1997)

Uranium is also contained in food and occurs in minerals as a less water soluble compound. Therefore **deposits of soil particles** on **leaves** and **fruits** are of significance for **uranium ingestion**, particularly in vegetables with craggy and crinkly surface like parsley and green cabbage. Fine soil particles (diameter up to 40 µm), which are hardly washable, have higher activities (factor 10) than larger ones (40–1000 µm). Mushrooms also show higher uranium concentrations than most other food. (Diehl, 2003)

The intake of uranium through **drinking water** can be a small or large fraction of the total intake depending on concentrations in local water supplies (Eisenbud & Gesell, 1997). According to the WHO, the levels in drinking water are generally less than 1 µg/l, although concentrations as high as 700 µg/l have been measured in private supplies. In circumstances in which uranium is present in a drinking water source, the majority of intake can be contributed by drinking water (WHO, 2004). The amount of uranium which has been measured in drinking water in different parts of the United States by EPA is generally below 1.5 µg for every litre of water. EPA surveyed that the levels of uranium of water in different parts of the United States are extremely low in most cases (U.S. Department of Health and Human Services, 1999).

Diehl (2003) reported regional highly fluctuating ^{238}U activity concentrations in **tap water** in **Germany** from 0.5 to 310 mBq/l.

The International Atomic Energy Agency (IAEA) reports in its technical report No. 284 (IAEA, 1988) the uranium content of terrestrial groundwater in a range between 0.5 to 2.5 ppb (accords to µg/l) and up to 100 ppb in or near uraniferous deposits. Wisser (2003) reports characteristic concentrations for ^{238}U and ^{234}U in aquifers in range from 10–100 mBq/l, which is about 3 times higher than the IAEA range. In areas with uraniferous granitic intrusions – e.g. in Finland – ^{238}U activity concentrations higher than 100 Bq/l were detected (Salonen & Huikuri, 2002). Krieger (2002) reports an annual intake of ^{238}U in regions of average soil activity of about 5 Bq/a.

Uranium generally shows a **log-normal distribution** in the natural waters of background areas. Most frequent positive correlations of uranium with other constituents and elements in natural waters are with SiO_2, HCO_3^-, PO_4^{3-}, SO_4^{2-}, F, Cl, Na, K, Mg, Ca, Sr, V, Se, Mn, Rn, humic matter, pH, hardness, specific conductivity, and dissolved solids (see Chapter 14 and 13). (IAEA, 1988)

Uranium contents in ground water are highly dependent on chemical reduction and oxidation conditions of the environment, which yields high differences in uranium contents in ground waters.

Uranium has **no known metabolic function in the human body** and is considered to be non-essential. Due to its chemical properties it is, like other heavy metals, **nephrotoxic**. The radiotoxic effect of uranium is regarded to be low. The daily intake of uranium is estimated to be 1–2 µg in food and 1.5 µg in drinking water. It has been calculated that 90% of the ingested uranium is excreted in urine within 24 hours after intake. The remaining fraction is excreted within the following weeks after uptake and only a small part is accumulated in the skeleton, tissues and organs. About 70% of the uranium contained in the human body is accumulated in the skeleton, 30% in soft tissue (Krieger, 2002). According to different studies (e.g. Limson-Zamora et al., 1998) the only chemical toxic effect due to the ingestion of uranium via drinking water is reversible damage to the kidney. (U.S. Department of Health and Human Services, 1999, Wisser, 2003)

2.5.4 Tritium

Tritium (^{3}H) is a **cosmogenic radionuclides** and is currently accumulated by a nuclear reaction of the cosmic radiation with aerosols of the atmospheric gases. The most important reaction for natural tritium production is the interaction of a neutron with an atmospheric nitrogen atom (Equation 2.18). Besides in this reaction stable carbon-12 (^{12}C) is formed.

$$^{14}_{7}N +^{1}_{0} n \rightarrow^{3}_{1} H +^{12}_{6} C \tag{2.18}$$

Tritium decays by **beta decay** with the half life of 12.3 years to ^{3}He. $72 \cdot 10^{15}$ Bq ^{3}H is formed in the atmosphere annually, the total amount of ^{3}H on earth is estimated to about $1.3 \cdot 10^{15}$ Bq. Tritium was also built artificial by nuclear weapons test in the 1950s and 1960s and deliverd to the atmosphere. Today tritium is also an byproduct in the nuclear industry and also delivered from there with the sewage and in the atmosphere.

Ground water out of great depths is tritium free, rain water has an activity concentration of about 1 Bq/l. The daily intake of ^{3}H by adults via water and food is about 0.1 Bq and about 50 Bq per year (Diehl, 2003).

With the tritium activity concentration in water an age estimation of the water can be carried out. As mentioned above, tritium free water comes from great depths and is older than 60 years (before nuclear weapons tests). Tritium with an high actvity concentration is about 20 years old because the highest tritum concentration existed in the atmosphere at this time. For water with a tritium activity concentration of 1–2 Bq/l a normal exchange with the atmosphere exists and is young water.

In the presence of oxygen tritium oxidizes to HTO (**tritiated water**). Tritiated water is the most abundant chemical form of tritium in the environment. HTO's properties are very similar to those of water because of their relatively small difference in atomic weight. HTO is taken up by organisms and environmental media far more readily than molecular tritium. A small fraction of tritium is incorporated into organic molecules exposed to tritium gas and HTO. This organically bound tritium (OBT) has a different metabolism than HTO. When tritiated water enters the body it acts just like normal water, spreading throughout the body and delivering a uniform radiation dose to all soft tissues. Inside an adult's body tritium behaves 97% as HTO with a biological half life of about 10 days and 3% as OBT with a biological half life of about 40 days. (Diehl, 2003, McKone et al., 1997)

2.5.5 Lead-210 and Polonium-210

^{210}**Pb** is a radioactive isotope of lead (Z=82) and is a member of the ^{238}U decay series, separated from ^{222}Rn by six short lived α- and β-emitters. ^{210}Pb is a **β-emitter** with a half life of 22.3 years. The ^{210}Pb decays to ^{210}Po via the intermediate Bi-210 which has a 5 days half life by β-decay. ^{210}**Po** (Z=84) has a half life of 138 days and is an α**-emitter**.

^{210}**Pb** is **produced rapidly in the atmosphere** because of ^{222}Rn decay, but its long half life allows very little to decay in the atmosphere before it **precipitates** to earth surface by rain or snow. So plants with large specific surface – on which ^{210}Pb can be deposited – are enriched in this radionuclide. This is observed in case of tobacco leaves (Marsden & Collins, 1963). ^{210}Po is believed to enter tobacco by ingrowth in ^{210}Pb deposited on tobacco leaves from the atmosphere. Direct uptake of ^{210}Po from soil is probably not significant. ^{210}Po is exhaled from tobacco leaves while smoking and is inhaled. 10 cigarettes per day double the intake of ^{210}Po. (Diehl, 2003, Eisenbud & Gesell, 1997, Peres & Hiromoto, 2002)

Normal **annual intake** per person of ^{210}Pb and ^{210}Po is 30 and 50 Bq respectively (UNSCEAR, 2000). In contrast, nomads of Lapland have an annual ^{210}Pb and ^{210}Po intake of 140 and 1400 Bq respectively, because of their high reindeer consumption. Reindeers mainly live on lichenics, plants with a large specific surface. (Diehl, 2003, Krieger, 2002, UNSCEAR, 1982)

Radioactive disequlibria are found in the upper part of rocks and soil profiles from which ^{222}Rn diffuses. Atmospheric transport and deposition of ^{210}Pb cause a more consistant distribution than ^{226}Ra from which it derived. The ratio of ^{210}Pb to ^{238}U in surface soil is about 2. (Eisenbud & Gesell, 1997)

When ^{210}**Pb** is absorbed into the body, ingrowth of ^{210}Po can occur because ^{210}Pb is deposited in the **skeleton** (about 70%), from which it is removed slowly (half life of about 10^4 days).

Food or **water ingestion** is a more important contributor to blood levels of ^{210}Po than inhalation, even among smokers. Unlike other natural occurring α-emitters, ^{210}**Po** is distributed constant in the body after ingestion and deposits in **soft tissues** and not in bones. (Diehl, 2003, Eisenbud & Gesell, 1997, Krieger, 2002)

Chapter 3

Drinking Water Regulations and Studies

3.1 Regulations for Drinking Water

> ”Drinking water has to be digestible and must not be harmful. It has to be delicious and therefore it ought to be colourless, clear, cool, odourless as well as proper in flavour and origin. Besides its external properties the water must not show physical, chemical, bacteriological nor biological indications of contamination. Water which is inherently up to standard should be preferred to preprocessed water.” (ÖNORM M 6250 (Austrian Standards Institute, 1986), translated)

The **ÖNORM M 6250** (Austrian Standards Institute, 1986) appoints the standard for the quality of drinking water for public water supplies, which can be drawn on private water supplies analogously. The commitment of the objectives, guidance levels and limits is effected on the basis of recommendations of the World Health Organisation WHO (WHO, 1984). The requirements are in accordance with the actual chemical, hygienic and technical knowledge and are classified in chemical and physical (ÖNORM M 6250, Pkt.3) and bacteriological properties (ÖNORM M 6250, Pkt.4). Within point 3 it is distinguished between parameters with chemo-technical, chemo-hygienic and toxicological importance. Point 5 concerns with radioactivity and appoints a maximum ingestion of the population of radioactivity in drinking water of $2.7 \cdot 10^{-3}$ µCi (=99.9 Bq) per year. This can be achieved if radioactivity in drinking water does not exceed 3.3 pCi/l (=122.2 mBq/l).

The ”Quality of water for human consumption” is regularized in Austria in the **”Drinking Water Directive”** (Republik Österreich, 2001). §3(1) specifies:

> ”Water has to be appropriate for using or drinking without hazarding human health. This is guaranteed if
> 1. Water does not contain micro-organisms, parasites and substances of any kind in a number or concentration, which is a potential hazard of human health and
> 2 . complies with the minimum requirements defined in Annex I, part A and B, as well as in Annex I, part C defined requirements.” (Republik Österreich (2001), translated)

In Annex I of the Austrian drinking water directive parameter and parameter values are listed. Part A – Microbiological parameters, Part B – Chemical Parameters and Part C – parameters with indicator function (indicator parameters), including radioactivity (Table 3.1). Annex II regulates the monitoring of the ”analyzing parameters” and the analysis frequency, dependent on the amount of the delivered water per day.

Table 3.1: *Radioactivity (Indicator parameter) (Republik Österreich, 2001)*

Indicator parameter	**Parametric value**	**Unit**	**Note**
Tritium	100	Bq/l	
Total indicative dose	0.10	mSv/a	Excluding ^{3}H, ^{40}K, radon and radon progenies

The drinking water directive is an implementation of the **European drinking water directive 98/83/EC** (European Commission, 1998) which was published in December 1998. For radioactivity two indicative standard parameter limits are established – Tritium activity concentration of 100 Bq/l and total indicative dose TID (effective dose from radionuclides in drinking water except ^{3}H, ^{40}K, radon and radon progenies) of 0.1 mSv/a. Radioactivity is affiliate to indicator parameters, with values according to article 5 paragraph 2 only for monitoring purposes and representing no limits. According to article 8 of the directive any failure to meet the parametric values set in accordance with article 5 is immediately investigated in order to identify the cause and to test if this failure constitutes a potential danger to human health. If necessary, appropriate medial measures have to be carried out and the consumers shall be informed promptly.

The directive 98/83/EC (European Commission, 1998) does not appoint **monitoring frequency, monitoring methods** and the most relevant locations for monitoring points. But a draft by the European Commission exists (European Commission, 2005) which regulates the monitoring frequency, monitoring methods and monitoring of compliance with total indicative dose.

The total indicative dose is estimated with appointed reference concentrations (Table 3.2) and sum formula. The reference concentrations were calculated using the dose **conversion factors for adults** according to the **directive 96/29/Euratom** (European Commission (1996), see Table 6.1, Chapter 6).

Table 3.2: *Reference Concentrations for radioactivity in drinking water (European Commission (2005), extract)*

Origin	**Nuclide**	**Reference Concentration**
Natural	^{238}U	3.0 Bq/l
Natural	^{234}U	2.8 Bq/l
Natural	^{226}Ra	0.5 Bq/l
Natural	^{228}Ra	0.2 Bq/l

The appointment and the evaluation of the TID are specified in the **Austrian Standard ÖNORM S 5251** (Austrian Standards Institute, 2005b). The required measurement techniques (e.g. decision limit), sampling site and the evaluation methods including examples are specified there (Schönhofer & Maringer, 2005). Generally only the radionuclides ^{226}Ra and ^{228}Ra are taken into account for dose calculation of drinking water in Austria for geo-chemical reasons.

For all other radionuclides in drinking water only **recommendations** exist, but no legally appointed limit or guidance levels in Austria. The European commission (European Commission, 2001) recommends for ^{222}**Rn** that a reference level should be appointed above an activity concentration of 100 Bq/l, and with radon activity concentrations above 1000 Bq/l measures are justified. For the radon progenies ^{210}**Pb** and ^{210}**Po** the Commission recommends (European Commission, 2001) that above an activity concentration of 0.2 Bq/l and 0.1Bq/l respectively, it should be tested whether any measures are necessary.

Concerning ^{238}**U** no recommendations by the EC exist, but the World Health Organisation (**WHO**) recommends for example in the **drinking water guidelines** 2nd edition (WHO, 1993) a guidance level of 30 µg/l natural uranium, which complies with 0.37 Bq/l ^{238}U activity concentration. In the drinking water guideline 3rd edition (WHO, 2004) the guidance level for uranium was reduced to 15 µg/l (corresponding to 0.19 Bq/l). These guidance levels are applied to chemo-toxic effects of uranium, not on the radioactive exposure. A reference concentration for radiological properties is recommended by European Commission with a reference value of 3 Bq/l for ^{238}U (see Table 3.2, European Commission (2005)), but without taking into account the chemical toxicity.

The drinking water guidelines of the WHO, 3rd edition (WHO, 2004) defines guidance levels for radionuclides in drinking water (artificial and natural) and says that no deleterious radiological health effects are expected from consumption of drinking water if the concentrations of radionuclides are below the guidance levels (equivalent to a committed effective dose below 0.1 mSv/year). This corresponds with the European drinking water directive 98/83/EG (European Commission, 1998), which has been discussed above. The guidance levels are defined in Table 9.2 in the drinking water guidelines for adults with an annual water ingestion of 730 l. For ^{226}Ra this guidance level is 1 Bq/l, which is twice as high as the one defined in the European Commission (2005) (Table 3.2). On the other hand for ^{226}Ra it is 0.1 Bq/l, which is half the one recommended in European Commission (2005). For ^{238}U it is 10 Bq/l in the WHO (2004). So guideline levels do not correspond well in different publications and recommendations by different authorities.

For radon the WHO guidelines recommends that controls should be implemented if the radon concentration of drinking-water for public water supplies exceeds 100 Bq/l, which corresponds basically with the European Commission recommendation (European Commission, 2001) but is stricter.

The drinking water guidelines (WHO, 2004) also recommend a **screening method for drinking water** with screening levels – for drinking water below no further action is required – 0.5 Bq/l for gross alpha actitvity and 1 Bq/l for gross beta activity. If either of the screening levels is exceeded, then the specific radionuclides producing this activity should be identified and their individual activity concentrations should be measured and compared with guidance levels. If the dose is below 0.1 mSv/a the water is suitable and no further action is necessary. If the dose is above 0.1 mSv/a it had to be considered and when justified, remedial actions to reduce dose should be taken. If the total dose of 0.1 mSv/year is exceeded on aggregate, then the options available to the competent authority to reduce the dose should be examined (see WHO, 2004). This recommended **gross alpha** and **gross beta screening method** corresponds with the draft of the European Commission (European Commission, 2005) in which a gross alpha and gross beta screening activity concentration of 0.1 Bq/l and 1 Bq/l respectively is defined. The screening level of 0.1 Bq/l for gross alpha activity was defined in the WHO drinking water guideline 2nd editon (WHO, 1993), but has been enhanced in the newer edition. But the recommendations by the European Commission are only a draft and not implemented by now.

3.2 Drinking Water Studies in Austria

In the field of radioactivity in drinking water **several studies were carried out in Austria** and some papers and reports were published. Most of these studies deal with ^{222}Rn measurements in water, some of them with ^{226}Ra, mostly as an overview concerning Austria or with particular interests in selected regions.

As a first step of this thesis a fundamental research of literature and data of radioactivity in drinking water in Austria was carried out. The most relevant studies are presented in this chapter shortly and included in the references. Additionally aged data were collected, for example from the Landessanitätsdirektion, Lebensmittelaufsicht of the Government of Upper Austria (^{222}Rn measurements from 7 water supplies in the Mühlviertel) and data of the sampling of the of the existing routine water sampling programme ”Wassergüteerhebungsverordnung (WGEV)”.

The combination of the aged data and studies with the measurements and results of this study is difficult, because mostly the exact sampling point, sampling method and treatment is not documented. Nevertheless these results and research of these studies were used as a basis for the new survey and will be presented here.

A fundamental work and basis is the map ”Radon in Water in Austria” by **Friedmann** (Figure 3.1, Friedmann (2006, 2008)). This map collects data from several studies and measurements of ^{222}**Rn in spring and ground waters in Austria** since the beginning of the 20th century. About 6500 measurements exist in a data base, 1600 of these measurements are from before 1950, but verified by later ones (Friedmann, 1999). The motivation of the map is an assessment of the probability of occurrence of higher radon activity concentrations in spring and ground waters. The existing measurement results do not suffice for an estimation of entire Austria, so geological probabilities were used for extrapolations in regions without radon measurements. The ”radon risk” is classified in 3 classes:

- **Class 1** (green) – A probability of 85% that spring and ground waters (no deep wells) have a radon activity concentration of below 100 Bq/m^3
- **Class 2** (yellow) – A probability of 85% that spring and ground waters (no deep wells) have a radon activity concentration of below 300 Bq/m^3
- **Class 3** (red) – A probability of more than 15% that spring and ground waters (no deep wells) have a radon activity concentration of above 300 Bq/m^3

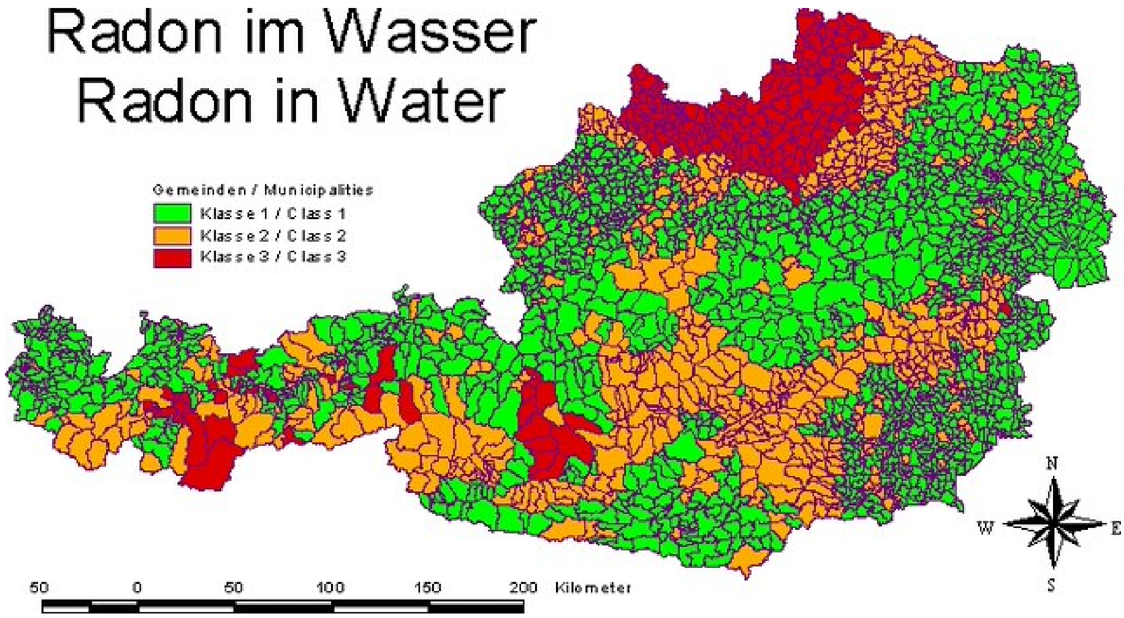

Figure 3.1: *Radon in Water in Austria (Friedmann, 2008)*

The map illustrates the regions in Austria with higher radon risk by the consumption of drinking water – mainly the Bohemian massif and the granite and gneiss regions of the central Alps. Other regions with special geological situations can also cause higher radon contents in water – former volcanism and subsurface transport of water across crevices and fault zones or a highly permeable ground, e.g. in alluvial fans. (Friedmann, 1999) For Upper Austria only the Mühlviertel (Bohemian Massif) is characterized bearing elevated radon activity concentrations in drinking water according to the map.

Relevant publications and data used for this map are for example by **Ditto** et al. Ditto et al. (1999) report about Radon-222 measurements in selected ground water samples of the existing routine water sampling programme **"Wassergüteerhebungsverordnung (WGEV)"**, which collects about 2000 water samples in Austria quarterly since 1993. The radon analyses for this programm were done by the BALUF (Federal Institute for Food Control and Research) Vienna and the BALU (Federal Institute for Food Control) Innsbruck by liquid scintillation counting. In the years 1999–2004 this project was continued and the analyses extended for ^{226}Ra. (Ditto, 2005)

Ditto et al. (1999) summarize the radon results of 1519 analyzed ground water samples with a mean value of 18.2 Bq/l, a median of 12.0 Bq/l and a maximum of 415.9 Bq/l. Only 5% of the surveyed drinking waters have radon activity concentrations above 50 Bq/l.

Figure 3.2 shows a **map of Upper Austria** with **classified radon measurements in drinking waters** (Amt der Oberösterreichischen Landesregierung, 1997). The sampling and analyses were carried out by the BALUF (Federal Institute for Food Control and Research), *Bundeskanzleramt Abt. VI* and the Office of the Upper Austrian Government, *Abt. Wasserbau and Umweltschutz, Lärm- und Strahlenschutz*. The map shows the measurement points in Upper Austria with circles of different sizes, which represent the measured radon activity concentrations. Most measurements were carried out in the Bohemian massif, only a few in southern Upper Austria. The radon activities in this region are very low compared to the Bohemian massif. At two points radon activity concentrations with about 1000 Bq/l were detected – both in the Bohemian massif –, one in the northern part of the Mühlviertel and one in the central region.

Schönhofer et al. did several studies and publications about ^{222}Rn and ^{226}Ra in ground and drinking waters in Austria (especially in Lower Austria) with regard to measurement methods, especially liquid scintillation counting. (Schönhofer, 1989, 1990, 1992, Schönhofer & Pock, 1996)

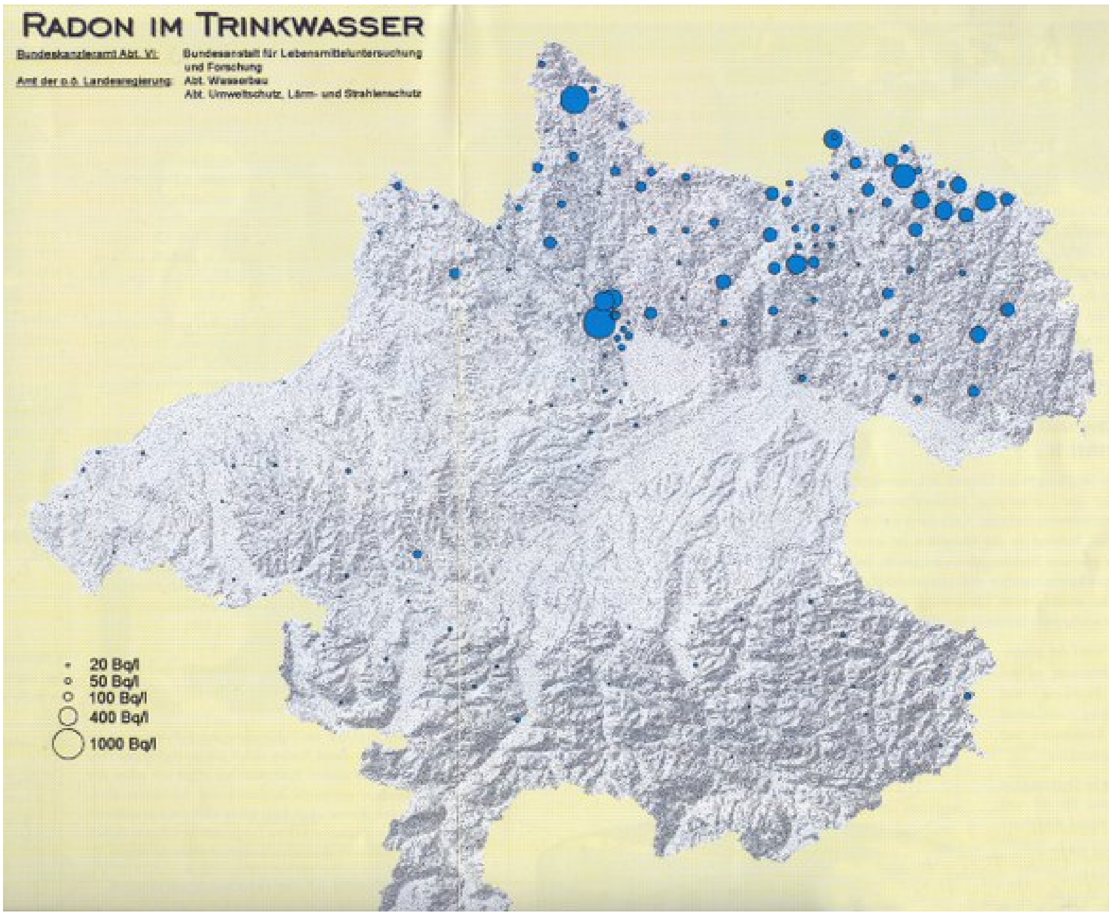

Figure 3.2: *Radon in drinking water in Upper Austria (Amt der Oberösterreichischen Landesregierung, 1997)*

There are also some studies taking into account also other radionuclides, for example ^{228}Ra and ^{238}U or the radon progenies ^{210}Pb and ^{210}Po. **Irlweck et al. (1997)** analyzed 40 tap water samples in different regions of Austria by liquid scintillation counting. The measured activity concentrations for ^{226}Ra are below 5 mBq/l. **Feigl-Heihs (1998)** realized these ^{210}Pb and ^{210}Po analyses within her master thesis. The detected activity concentrations for ^{210}Pb are in the range 1.5 to 102 mBq/l, for ^{210}Po 0.8 to 63.8 mBq/l.

Wallner & Steininger (2002) and Wallner (2002) surveyed about 65 drinking waters and 9 bottled mineral waters from different regions of Austria for ^{226}Ra, ^{228}Ra and ^{222}Rn by liquid scintillation counting. The highest ^{222}Rn activity concentration was detected in the Mühlviertel, Upper Austria with 644 Bq/l, all others were below 100 Bq/l. The highest ^{226}Ra activity concentration was detected in lower Austria with 111 mBq/l, all others are in the range $<$0.3 to 48 mBq/l. No ^{228}Ra activity concentrations above decision limit were detected in drinking waters.

Gegner (2002) analyzed 37 drinking water samples from different regions of Austria (selected because of geology) for their uranium content and uranium isotopes ratios. The detected ^{238}U activity concentrations range from 1 to 148 mBq/l, with one higher activity concentration (975 mBq/l) in a spa in Salzburg. The uranium contents in the analyzed drinking waters have a high fluctuation also in wells within only some kilometers distance and are not higher in the Bohemian massif.

Some surveys concerning radioactivity in drinking water in Austria were carried out with regard to special interest, like geology. **Schubert et al. (2003)** carried out a pilot study on radionuclides in ground water in crystalline bedrock in the Mühlviertel. They analyzed ^{40}K, ^{222}Rn, ^{226}Ra, ^{232}Th and ^{238}U in 4 different granite types (22 samples) of the lower Mühlviertel. The results are discussed in Chapter 4.

Krakhofer (2002) surveyed radon in ground and spring waters in the parts of Austria located north of the Danube in the framework of her diploma thesis. In this thesis the data base of the radon results in drinking waters by H. Friedmann (Friedmann, 1997) were processed with about 5000 data and analyzed for the northern part of Upper Austria (about 149 communities with measurement results). The results are surveyed with regard to the different geological zones, the radon indoor potential of the regions and uranium in river sediments (Thalmann et al., 1989).

Other Austrian studies deal with radioactivity in drinking water in a different point of view. **Staubmann (2002)** surveyed the techniques for the removal of natural radioactivity from drinking water and their distribution in Austria. **Schaffer (2005)** did a diploma thesis in this field. These studies were carried out in the framework of the EU project TENAWA – "Treatment Techniques for Removing Natural Radionuclides from Drinking Water" (Contract No F14 PCT 960054; Annanmäki (1999)).

3.3 Natural Radioactivity in Drinking Water – Situation in Other Countries

As discussed in Chapter 3.1 there are several directives and guidelines by different authorities (EU, WHO,…) regarding natural radioactivity in drinking water. But the directives and guidelines leave a high tolerance and high responsibility to the countries to define their own directives and implement them.

In this chapter an overview on the legal situation and studies about natural radioactivity in some selected countries should be given and discussed briefly.

The International Commission on Radiological Protection (ICRP) and The Council of the European Union have recommended that the Member States shall take action against radon in homes and at workplaces. Within the EU project European Research into Radon in Construction Concerted Action, **ERRICCA**, (Contract ERB F14P-CT96-0064, DG12-WSMN), the Topic Group on Legal and Building Code Impact was designated to study the current radon legislation and give advice regarding future enactment of laws and recommendations. On behalf of the Group, a questionnaire on radon legislation was sent out to nearly all European States and to selected non-European States. Questions were asked regarding reference levels for dwellings, workplaces and drinking water, and about regulations or recommendations for building materials and city planning. All 15 EU Member States, 17 non- EU European countries and 10 non-European countries responded to the questionnaire. (Åkerblom, 1999)

The results of this project for drinking water were actualized and should be briefly discussed here, together with regulations for other nuclides.

In **Germany** several studies were carried out in the field of natural radioactivity in drinking water since the 1970s (e.g. Aurand & Gans, 1991, Bünger & Rühle, 1993, Gellermann & Stolz, 1997, Rühle, 1996). Germany implemented the directive 98/83/EC in German law in 2001 with the drinking water directive (Bundesrepublik Deutschland, 2001). According to the EC directive, the maximum acceptable total indicative dose (TID) caused by radionuclides in drinking waters is 0.1 mSv/a, excluding ^{3}H, ^{40}K, radon and the radon decay products. The maximum acceptable concentration of ^{3}H is set to 100 Bq/l.

The **Bundesamt für Strahlenschutz (BfS)** performed measurements in German drinking waters and collected aged data about ^{210}Pb, ^{210}Po, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U and ^{238}U in drinking waters. 1734 drinking water samples were analyzed for ^{226}Ra in 13 federal countries, with a median value of 4.8 mSv/a, which is clearly higher than the UNSCEAR-value of 0.5 mSv/a. For other radionuclides predominantly water samples from mining regions of Saxony and Thuringia were analyzed, which is not representative for entire Germany. A nationwide survey program is running by the BfS (Bundesamt für Strahlenschutz, 2008).

A limit value of guidelines for maximum radon and radon progenies activity concentrations does not exist in Germany yet. Rühle (1996) reported activity concentration ranges of natural radionuclides in ground and spring waters (see Table 3.3) in Germany.

In the **USA** the Environmental Protection Agency (EPA) issued the National Primary Drinking Water Regulations (EPA, 2000) in December 2000, which became effective in December 2003. The regulation is only applicable to community water systems and it gives among others maximum contaminant levels (MCLs), which are defined as ”enforceable regulatory limits” with a gross alpha (excluding Rn and U) MCL of 555 mBq/l, beta particle and photon radioactivity of 40 µSv/a, ^{226}Ra + ^{228}Ra of 185 mBq/l, ^{210}Po is included in gross alpha, ^{210}Pb is explicitly not regulated and an MCL for Uranium of 30 µg/l. Therefore, it can be stated that the EPA approach is quite different from those of WHO and EU, because the suggested levels are neither ”indicator parameters” nor ”guidance levels”, but actual limits and are generally much stricter than EU and WHO requirements. (Nuccetelli & Risica, 2006)

(Hakonson-Hayes et al., 2002) reported uranium contents in wells in a region of northern New Mexico up to 1200 µg/l, which clearly exceeds the recommended MCL for uranium.

Table 3.3: *Ranges of activity concentrations of natural radionuclides in ground and spring waters in Germany (Rühle, 1996)*

Radionuclide	Activity concentration range (mBq/l)	Medium activity concentration (mBq/l)
^{3}H	<40–600	200
^{40}K	4–600	70
^{238}U	<1–400	5
^{226}Ra	<4–400	4
^{222}Rn and short lived progenies	$<2 \cdot 10^3$–$1.5 \cdot 10^6$	5600
^{210}Pb	<1–200	1
^{210}Po	<1–80	0.5
^{232}Th	<1–10	–
^{228}Ra	<1–30	–

In **Canada** the basic regulation for drinking waters is provided by the ”Guidelines for Canadian Drinking Water Quality”, which also include guidelines for radiological parameters. In addition, several Canadian Provinces have established their own drinking water standards with maximum acceptable concentrations for certain parameters adopted from the Health Canada guidelines. For instance the Province of Ontario stated the ”Ontario Drinking Water Standards Regulation” (Ontario Ministry of Environment and Energy, 2003) which deals with the standards that are used for water testing. The radiological standards were established as maximum acceptable concentrations (MAC), based on the committed effective dose of 0.1 mSv/a and an annual water intake of 730 litres. For example, the MAC for ^{238}U is defined with 4.0 Bq/l, ^{238}U also with 4.0 Bq/l, for ^{228}Ra with 0.5 Bq/l, for ^{226}Ra with 0.6 Bq/l and for the radon progenies ^{210}Pb and ^{210}Po with 0.1 and 0.2 Bq/l respectively (Wisser, 2003). These MAC therefore are slightly higher than the ones recommended in the draft by the European Commission (2005), but in the same range.

In **Switzerland** limit values for natural radionuclides in food exist since 1995 in the ”*Fremd- und Inhaltsstoffverordnung*” (Eidgenössisches Department des Inneren (EDI), 1995), in which maximum concentrations for radionuclides from the uranium-thorium decay chain for liquid food are defined. The maximum concentrations are defined for a group of radionuclides, and within the group for the sum of the detected activity concentrations. It is defined that when the maximum concentration is exceeded, the food is inapplicable for human consumption. Group I nuclides are ^{224}Ra, ^{228}Th, ^{234}U, ^{235}U and ^{238}U with the limit maximum concentration of 10 Bq/l; group II nuclides (^{210}Pb, ^{210}Po, ^{226}Ra, ^{228}Ra, ^{230}Th, ^{232}Th, ^{231}Pa) with 1 Bq/l.

In **Portugal** the Portuguese Decree Law n° 243/2001 defined guidance levels for drinking water for gross alpha, gross beta and ^{3}H concentrations with 0.1 Bq/l, 1 Bq/l and 50 Bq/l respectively. The approach taken in the guidelines for controlling the radiological hazards consists in an initial screening for gross alpha and gross beta, without regarding specific radionuclides. Lopes et al. (2006) conducted analyses in that field at about 50 drinking water samples and detected mainly gross beta and ^{3}H activity concentrations only below the guidance levels, and only 18% of the detected gross alpha activities exceeded the recommended level.

In **Italy** the European Council directive 98/83/EC was implemented in Italian law in 2001, and parameter values have been defined – 100 Bq/l for Tritium and 0.1 mSv/a total indicative dose excluding tritium, ^{40}K, radon and radon progenies, as recommended in the European Council directive. Since the implementation public authorities organized tap water surveys, like the one in Lombardia (Forte et al., 2007). In this study a wide range monitoring is tested in this region by measuring mainly gross alpha and beta activity concentration and checking compliance to derived limit values (0.1 Bq/l and 1 Bq/l for gross alpha and gross beta) by liquid scintillation counting, but also nuclide specific analyses.

In **Australia** the Australian Drinking water Guidelines (NHMRC, 2001) have recommended a dose of 1 mSv/a for radioactivity in drinking water in 2001 and screening levels have been derived from the recommended guideline dose – for human drinking water 0.5 Bq/l for gross alpha and 0.5 Bq/l for gross beta which is 5 times higher than recommended in the draft of the EC (2005) for gross alpha and only half the value for gross beta. Kleinschmidt (2004) developed a gross alpha and beta routine liquid scintillation screening method for local drinking water guidelines.

Several countries have **reference levels for radon in drinking water**, which are rather different and range from 50 to 1000 Bq/l and are either recommended reference or guidance levels or limits. Some countries have **enforced levels**, like **Sweden**, which has two enforced levels for public waters – 100 Bq/l at which the owners have to take measures, and 1000 Bq/l at which the delivery of water is not allowed. For private waters these levels are advisory. **Czech Republic** has one enforced level with 300 Bq/l and one advisory level, 50 Bq/l for public water, while private homeowners have two advisory levels, 1000 Bq/l and 200 Bq/l, corresponding to different remedial measures. **Finland** has one enforced level for public waters, 300 Bq/l and **Norway** an advisory level of 500 Bq/l. The **Slovak republic** has one advisory level for all drinking waters with 50 Bq/m3 and one enforced level (1000 Bq/l). **Russia** has an enforced level for all drinking waters, irrespective of patterns of ownership with 120 Bq/l. **Romania** has an enforced level of 300 Bq/l which is compulsory for public and private waters. In the UK there is an advisory level of 100 Bq/l for public waters. In the **USA** the EPA recommended that the value of the Alternative Maximum Contaminant Level (AMCL) for the average ambient radon concentration in drinking water to be set at 150 Bq/l, and that water supplies with levels in excess of the AMCL must be mitigated to at least the AMCL. Water supplies are considered to be public water supplies when serving 25 or more people. (Åkerblom, 1999)

Poland has a national standard for Radon with 74 Bq/l. Waters with an activity higher than 74 Bq/l should only be used under medical supervision. (Kozlowska et al., 1999)

Several studies all over the world were carried out in the field of **natural radioactivity in drinking water**. Some of them surveyed different nuclides in drinking water like ^{226}Ra, ^{228}Ra8 and ^{238}U. Only few studies exist about the radon progenies ^{210}Pb and ^{210}Po in drinking water. Most studies analyze radon in drinking or ground waters all over the world, but often in very specific regions and interests and often only with few analyzed samples. Almeida et al. (2004) analyzed 88 ground water samples from springs, dug wells and deep wells in Regiao dos Lagos, **Brazil** for ^{222}Rn, ^{228}Ra, ^{226}Ra and ^{238}U. The analyzed radon activity concentrations are very low, mostly beyond decision limit. The activity concentrations of the other nuclides are also mainly below recommended guidelines, except for ^{226}Ra. Godoy & Godoy (2006) also surveyed 220 ground water samples in Brazil for different radionuclides, including also ^{210}Pb and determined correlations between the nuclides and also to geology.

Waters used for drinking water out of 10 boreholes from the Valley of Toluca, **Mexico** were analyzed for ^{222}Rn, ^{226}Ra and ^{238}U by Segovia et al. (1999). The waters showed an average value of radon activity concentration of 2.2 kBq/m^3, ^{226}Ra between 0.01 and 0.2 Bq/m^3 and the ^{238}U activity concentrations were all below decision limit. The paper discusses the activity concentrations by geology and groundwater flow systems.

Zhuo et al. (2001) surveyed 553 groundwater samples in the Fujian Province, **China** for ^{222}Rn, ^{226}Ra, ^{228}Ra and uranium. They detected high radon activity concentration in the predominantly granite rock aquifers in this region.

As mentioned above, many studies exist all over the world in the field of ^{222}Rn in drinking and ground waters. The highest radon concentration in ground waters in Europe amounts to 77.5 kBq/l and has be detected in **Finland** because of geology (Salonen, 1994). There are several studies about radon in drinking water in Finland (e.g. Salonen, 1988, Salonen & Huikuri, 2002) and **Sweden** (e.g. Åkerblom & Lindgren, 1997, Erlandsson et al., 2001, Skeppström & Olofsson, 2005), which mainly deals with springs and drilled wells with high radon concentration because of geology.

In **Poland** studies on radon drinking water were carried out for different regions, e.g. the north-eastern hydro regions, where 643 samples were analyzed and 57 exceeded the guideline maximum level of 11 000 Bq/m^3, mainly because of geology (Zalewski et al., 2001) or the Suwalki region (Karpinska et al., 2002), where 61 water samples were analyzed for radon.

Studies about radon in drinking water (mostly only with a few sampling points and in a selected region) also exist for instance in **Saudi Arabia** (e.g. Alabdula'aly, 1999, Tayyeb et al., 1998), **China** (e.g. Xinwei & Xiaolan, 2004), **Mexico** (e.g. Villalba et al., 2005) and **Kenya** (e.g. Otwoma & Mustapha, 1998).

An example for a study of radon in drinking water with a total different point of view is the **Russian paper** by Voronov (2004). In this study it is searched for waters with a radon concentration higher than 185 Bq/l, because, according to the Russian mineral water classification, then waters are defined to be radon mineral curative waters with a high therapeutic effect. These so called radon-rich waters are mainly found in crystalline acid rocks that have exceeded uranium-radium mineralization.

Chapter 4

General Hydrogeology and Hydrogeology of Upper Austria

4.1 General Aspects of Ground Water

A groundwater system could be defined as ”the rock framework in the earth crust containing saturated groundwater with a water pressure larger than atmospheric pressure” (Nonner, 2003).

Groundwater can be formed only in rocks with pores or crevices. Such rocks with the ability to transport groundwater are called **aquifers**. In soils like sand and gravel the groundwater flows in the pores between grains, pebbles and rock particles (**pores groundwater**). In bedrocks the groundwater moves on bed interstices, crevices and cleavages (**crevice groundwater**). The volume and the flow velocity of groundwater in bedrocks are mostly lower than in soils except for karst groundwater. In some bed rocks (lime stones) with cohesive crevices and cleavages a karst water system is formed in which the **karst groundwater** flows with a velocity like in rivers (Bradl, 2005, Vogelsang, 1998).

The rise of groundwater table by precipitation may be relatively prompt or delayed, depending on the depth of the groundwater table and the rock composition of the unsaturated zone (Nonner (2003), Figure 4.1). In little permeable clayey soils a huge part of precipitation runs off overground. In high permeable sandy soils and karst every precipitation raises directly the ground water table. For extraction the ground water must be pumped to surface in drilled wells. In **artesian wells**, in contrast, pumping is not needed. The aquifer is covered by a non permeable layer, which declines under the ground water table. If this layer is drilled or punctuated the water rises to the ground water table. If this level is above earth surface an artesian well-spring originates there without any technical help.

In solid rocks the quantity of extractable water sources is much lower than in soils, because typically no general ground water level exists. (Vogelsang, 1998)

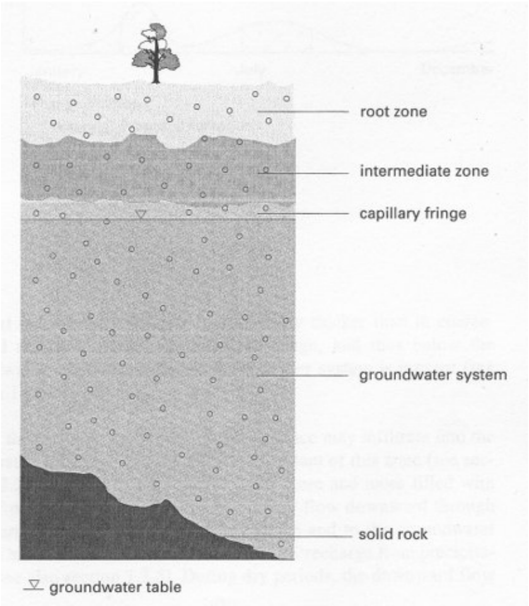

Figure 4.1: *Soil layers with ground water system (Nonner, 2003)*

The behaviour of water in aquifers is affected by three important factors. **Porosity**, which is defined as the percentage of pore or void space, determines how much water can be contained per unit volume of aquifer. Another important property is **permeability**, the degree to which the pores are connected to one another. The **hydraulic gradient** or slope of the aquifer affects the pressure, which, together with the permeability, determines the rate of water flow in the aquifer. (Eisenbud & Gesell, 1997)

The flow of water through porous rocks is specified by **Darcy's Law**. It says that the water flow is dependent on the pressure gradient and the permeability k of the soil or solid rocks. The permeability in different rocks and soils range from 10^{-8} (impermeable rocks with almost no pores like clay) to 10^{7} (sand, gravel). (Scheffer & Schachtschabel, 2002, Vogelsang, 1998)

As described above, ground water sources are dependent on geology. Different materials like soils (gravel, sand, clay) and solid rocks (sandstone, quartzite, karst) cause various origins and types of ground waters. In Chapter 4.3 a characterization of the hydrogeology of the investigation area Upper Austria is done.

Besides the geology different characteristics affect the ground water. One of them is the **catchment basin** of a well or aquifer. In soil a catchment basin is the area in which all not run off overground precipitations contribute to ground water regeneration. Ground water regeneration is only possible if the aquifer can accumulate the infiltrated water. If the maximum capacity is reached, the aquifer gives off the excess water to springs or streams, called receiving water.

Ground water is never a chemically pure H_2O but has a quantity of dissolved matter like Na, Mg, K, Cl, Si, S (see Chapter 13). **Water hardness** is dependent on the chemical equilibrium of lime content (Ca) of the water and the dissolved carbonic acid (H_2CO_3), classified in ”German hardness” (°dH). Regions with limy stones have ”hard” water with a high ”German hardness” (>20), because much lime is dissolved in the ground water. Water with a low ”German hardness” is only available in rocks without lime like pure gravels and sands, schist, granite and gneiss.

The **pH-value** (*potential hydrogenii*) of water indicates the hydrogen ions concentration in the water, which defines whether an aqueous solution reacts acidic or alkaline. In Central Europe the pH-values preponderantly range from 5 to 8, dependent on the material of the aquifer and the unsaturated zone. In pure siliceous sand and gravel usually acidic water (pH<6) occurs, in clays predominantly alkaline (pH>7) (Vogelsang, 1998). In Chapter 13 these parameters are discussed for the samples of this thesis.

Ground water tables additionally vary due to different influences like tides, atmospheric pressure, the seasonal circle and the surroundings. Low and high tide effects of the ground water are very small, usually only a few centimetres, and so is the influence of atmospheric pressure. The seasonal circle of the ground water is reliant on the precipitation quantity but also location-dependent from other factors. In Central Europe the ground water tables are high in winter and low in summer. The more permeable rocks are, the higher is the variation of the respective ground water table. Furthermore ground water deposits are affected by natural or artificial modification of the landscape or substratum (e.g. straightening of streams, earthquakes, detonations). (Vogelsang, 1998)

4.2 The Drinking Water Situation in Austria and Upper Austria

In Austria ground water is the most important source of drinking water. 99% of the Austrian drinking water originates from ground water – half of it from **pores groundwater** sources in vales and basins, the other half from **karst and crevice groundwater** sources in the mountains. 66% of the Austrian population is fed by 227 water supplies with an average water take out >1000 m^3 per day or a supply of more than 5000 persons. This means a water output of 428.8 million m^3 per year (BMGFJ, 2007). 13% of the Austrian population use drinking water from private wells (Lebensministerium, 2008a,b, Umweltbundesamt Wien, 2007b).

Upper Austria has access to a large quantity of groundwater, hence 100% of the Upper Austrian drinking water is obtained from ground water (Amt der Oberösterreichischen Landesregierung, 2008). About 90 000 **private wells** exist in Upper Austria, this means a quarter of the Upper Austrian homes are fed by private wells, which is the highest percentage in Austria compared to other provinces. The state of repair of these wells and the quality of the drinking water are under the responsibility of the owner respectively user (Amt der Oberösterreichischen Landesregierung, 2007).

The **quality of the Austrian ground water is excellent** according to international comparisons. To hold this high level in Austria legally defined quality requirements exist (e.g. Drinking water directive – Republik Österreich (2001)). In this directive also a monitoring for ground water quality is appointed (**WGEV – Water Quality Monitoring Ordinance**, see Chapter 5).

4.3 Hydrogeology of Upper Austria

As described above in Upper Austria (as in Austria) **three types of ground water** exist – pore groundwater, karst groundwater and crevice groundwater. Aquifers for **pore groundwaters** are mainly glacial terrace gravel as well as tertiary sand and gravel and are often covered with layers (e.g. loess clay). The flow velocity of the pore ground water is rather low (1 to 10 meters a day), therefore the duration of dwell in the ground is rather high and substances from the rocks can be solved in the water, which have an impact to the water (e.g. high water hardness). In Upper Austria pore groundwater occurs in the sediment filling of valleys and basins (e.g. *Eferdinger Becken*, *Linzer Becken*,...) and in the tertiary sand and gravel (*Kobernaußerwald*) (see Table 4.1). (Amt der Oberösterreichischen Landesregierung, 2008)

Karst ground waters are subsurface waters in karstified rocks like limestone, gypsum and dolomite. The storage capacity of ground water in karst is rather low, but nevertheless karst systems have a considerable amount of cavities and a high ground water regeneration rate. In karst systems the waters can have high flow velocities (up to some hundred meters per hour) but also high durations of dwell in the ground. In Karst systems mainly no covering soil layers exist, which have a filtering impact to pollution to the water. So karst ground waters are highly sensitive to pollution and acid deposition. In Upper Austria karst groundwater appears in the northern limestone Alps. (Amt der Oberösterreichischen Landesregierung, 2008)

Crevice ground water is ground water which is bounded at not karstified interstices in the rocks. Crevices are thereby either tectonic or relief interstices at the edges of valleys. Interstices mainly occur at inflexible rocks like gneiss, granites, carbonates or sandstones and also in ductile rocks like clay marls of the *Molasse*. Crevice groundwaters are usually less abundant because of reduced crevices in the depth. Flow velocities of crevice ground water can reach some meters per day. In Upper Austria crevice ground water occurs in the Bohemian massif and in the *Flysch*. (Amt der Oberösterreichischen Landesregierung, 2008)

According to Amt der Oberösterreichischen Landesregierung (2008) **16 different ground water bodies** exist in Upper Austria due to hydrogeological and hydrogeochemical characteristics (see Figure 4.2). Definable, hydrologic continuous ground water areas are called individual ground water bodies, while areas with localized structured but equably hydrogeological properties are combined to groups of ground water bodies. Beyond the ground water bodies are distinguished into surface near and depth ground water bodies. Surface near ground water participates in the active water cycle and is exposed to surface impacts (e.g. agricultural land utilisation) hence. Depth ground water is not directly impacted by surface infiltrations because of abundant covering and low permeable layers. (Amt der Oberösterreichischen Landesregierung, 2008)

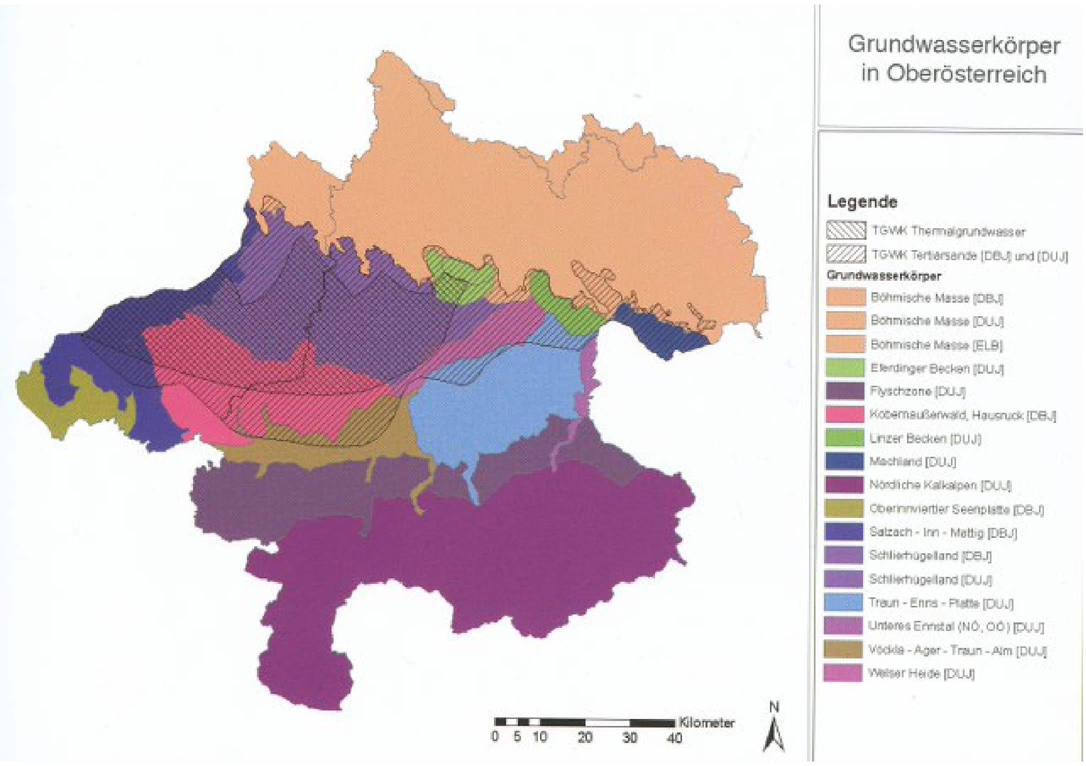

Figure 4.2: *Groundwater bodies in Upper Austria (Amt der Oberösterreichischen Landesregierung, 2008)*

The 16 ground water bodies in Upper Austria are listed in Table 4.1, characterised by individual or group of ground water bodies, surface near or depth, pore, crevice or karst ground water. Only tertiary sands and thermal water are depth water bodies, which are located mainly in the Upper Austrian Molasse.

This very detailed differentiation is not applied to the interpretation of all measurement results of this thesis, because too few measurements were carried out in each ground water body, nevertheless some discussion will be made on the basis of these characterisation in Chapter 15.

In the **hydrochemical comparison** of the **ground water bodies** in Upper Austria a clear difference is identifiable between the karst ground water body *Nördliche Kalkalpen* (Northern limestone Alps) and the crevice ground water body *Böhmische Masse* (Bohemian massif). This is noticeable because of a very low pH value in the Bohemian Massif (because of a high carbonic acid concentration in the crystalline) and very high pH values in the northern limestone Alps because of the lime rocks. These differences are also noticeable in the electric conductivity and water hardness – see Chapter 13. A simplified classification of ground water – pore (German: *Poren-Grundwasser*), karst (German: *Karst-Grundwasser*), crevice (German: *Kluft-Grundwasser*) can be made with the knowledge of the pH value (German: *pH-Wert*) and the electric conductivity (German: *elektr. Leitfähigkeit*) – of a ground water (Figure 4.3). Pore ground waters have higher concentrations of substances because of their low flow velocity and high contact time with the rocks and the higher contact area between water and rocks because of the small pores. The ground water body *Machland* shows the highest water hardness, the other basin ground waters also have hard water.

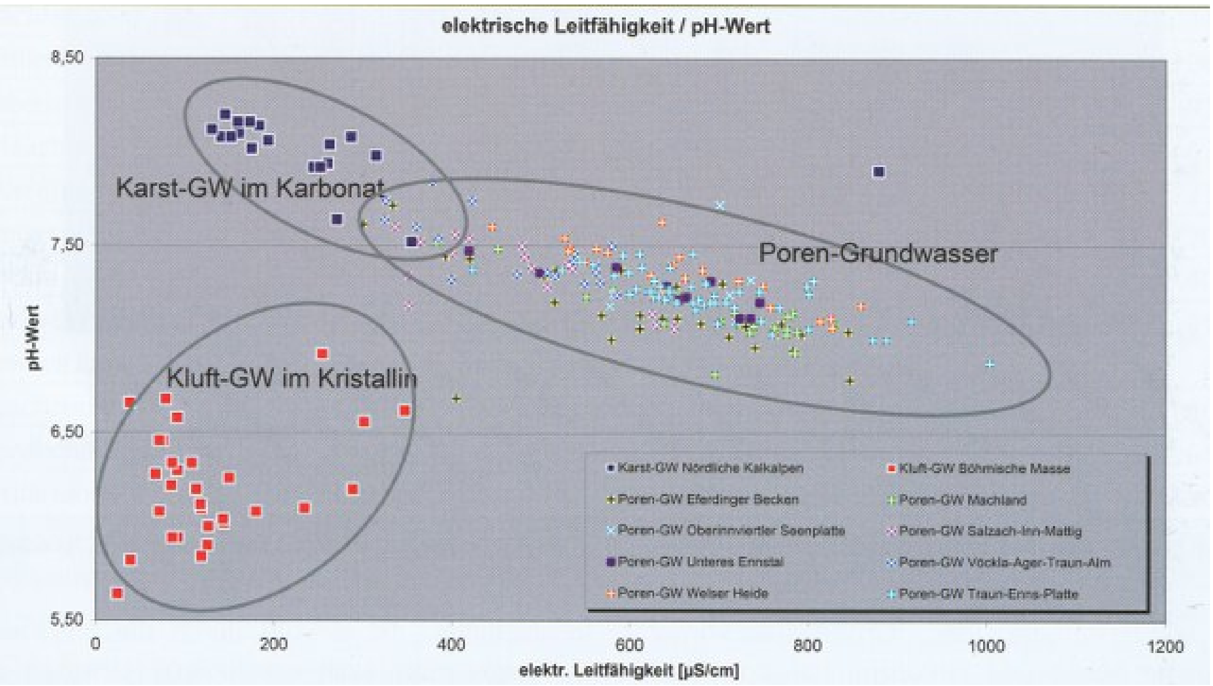

Figure 4.3: *pH-value and electric conductivity in different ground water bodies in Upper Austria (Amt der Oberösterreichischen Landesregierung, 2008)*

Additionally a hydrogeological map of Upper Austria shows the simplified fundamental geological formations and distribution of the different ground water occurrences (Figure 4.4, Vohryzka (1973b)). For the legend of this map, see Annex A1.

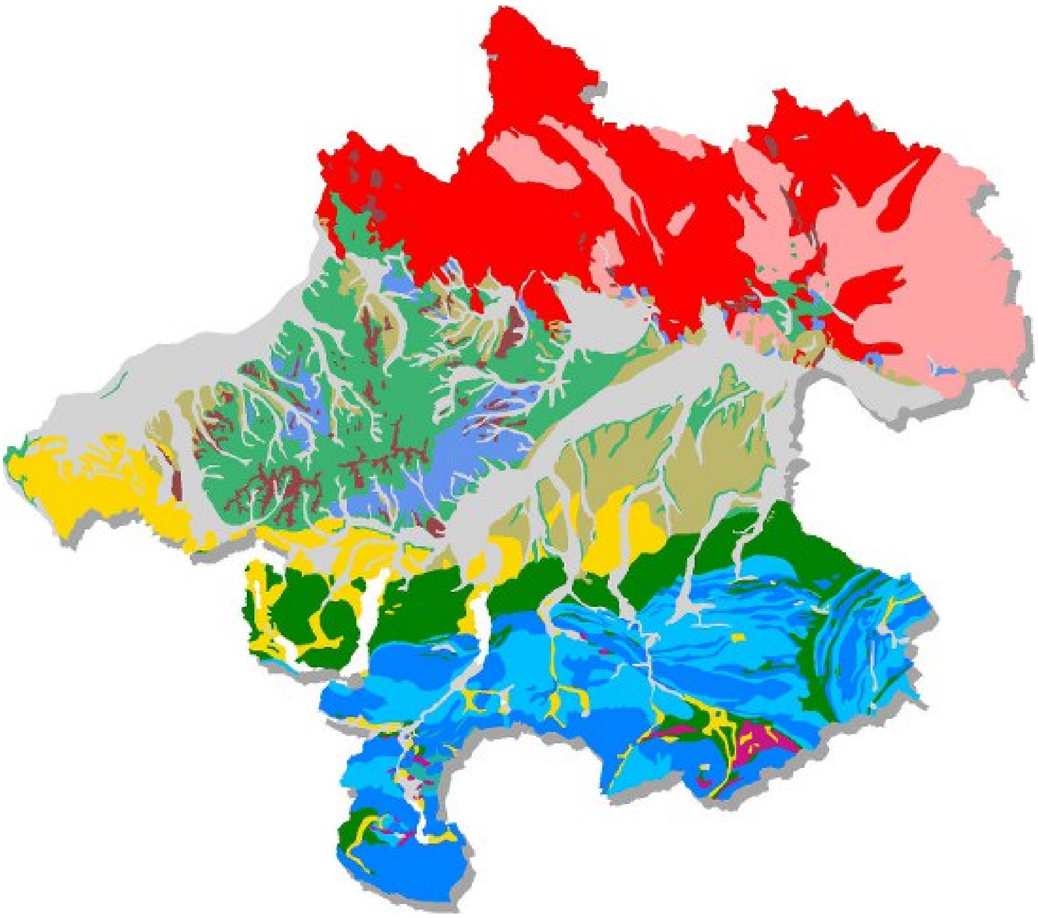

Figure 4.4: *Hydrogeological map of Upper Austria (Vohryzka, 1973b), for legend see Annex A1*

Vohryzka (1973b) described the hydrogeology of Upper Austria in detail in a map (Figure 4.4) and also in the explanatory notes. He distinguishes between 5 main hydrogeological zones:

1. Crystalline and overlayed tertiary and quaternary basins (**Bohemian Massif** – Mühlviertel and Sauwald)
2. **Limestone Alps** (1/5 of the area of Upper Austria – southern Upper Austria)
3. **Flysch** (foothills of the Alps – chalkstones)
4. **Molasse** (between Flysch and Bohemian Massif, sedimentary rocks, glacial deposits)
5. **Glacial and recent formations** (moraines, gravel bodies)

In the hydrological map these 5 main hydrogeological zones are characterized in more areas (see Figure 4.4 with caption). All zones are described in detail and again subdivided, but not discussed here. Some details will be taken into account in Chapter 15 at certain measurement points. Some measurement results of this thesis mainly will be characterised by a simplified characterization by **Vohryzka (1973b)** with following areas:

1. Weinsberger Granit (pink)
2. Gneise und Granite der Böhmischen Masse (red)
3. Tertiäre Schliere und Sande (medium blue and turquoise)
4. Junge Talfüllung, Austufe, Nieder- und Hochterasse (grey)
5. Deckenschotter (ochre)
6. eiszeitliche Moräne, Eisrandsedimente (yellow)
7. Mergel und Sandsteine der Flyschzone (green)
8. Dolomite der Nördlichen Kalkalpen (light blue)
9. Kalke der Nördlichen Kalkalpen (dark blue)
10. Hausruckschotter, Jüngere tertiäre Schotter (brown)

The results will be compared with the different classifications (Amt der Oberösterreichischen Landesregierung, 2008, Vohryzka, 1973b) and in Table 4.1 the correlation of the different classification is listed. The distinction is a hydrogeological characterization of the bedrock, not limited to geographic areas by Vorhyzka on the one hand and a geographic bounded characterization of different ground water bodies by Amt der Oberösterreichischen Landesregierung (2008).

In practice, many results are discussed and analyzed using only a very simplified classification because of statistical significance, especially in the detailed sampling phase:

- Upper Mühlviertel (Bohemian Massif)
- Central Mühlviertel (Bohemian Massif)
- Lower Mühlviertel (Bohemian Massif, Machland)
- Danube fault (Innviertel) (Molasse, Thermal water)
- Southern Upper Austria (Northern Limeston Alps, Flysch)

According to Vohryzka (1973b) the Bohemian Massif is mainly classified in *Weinsberger Granit* and all other Gneiss and Granites of the Bohemian Massif. There are **various different granite types** in the **Bohemian massif** according to the Geological map of Upper Austria (Geologische Bundesanstalt, 2006). The various Gneiss and Granite types have different **contents of Uranium and Thorium** and may therefore impact the radionuclides concentration of ground water originating from these bedrocks. There are few studies about the Uranium and Thorium contents in different Granite types of the Bohemian Massif. Schubert et al. (2003) surveyed the ^{222}Rn and ^{238}U concentration in ground water wells and springs in 4 different Granite types in the Bohemian massif (*Weinsberger Granit*, *Migmagranit*, *Altenberger Granit*, *Freistädter Granodiorit*) in a pilot project. The highest radon and uranium activity concentrations in ground water were measured in a drilled well in the *Weinsberger Granit* (in the village of the radon spa Bad Zell in the lower *Mühlviertel*), although the uranium concentration in the *Weinsberger Granit* is only about one fourth to one half of the one in the *Altenberger Granit* and *Migmagranit*, maybe because of the longer duration of dwell. According to Gerdes (1997) and Schubert et al. (2003) *Freistädter Granodiorit* have low uranium (1–3 ppm) and thorium (4–8 ppm) concentration, *Weinsberger Granit* 2–5 ppm U and 20–40 ppm Th. *Altenberger Granit* and *Migmagranit* of the eastern Mühlviertel shows high Th and U concentrations (*Altenberger Granit*: 8–11 ppm U, 14–18 ppm Th; *Migmagranit*: 5–15 ppm U, 12–31 ppm Th). Besides, a better correlation between ^{238}U and ^{222}Rn activity concentration in the water and the U and Th concentration in bedrock were surveyed in regions were mainly samples from springs were analyzed (*Freistädter Granodiorit*, *Altenberger Granit*) – maybe because at springs a greater mass of water with a longer duration of time streams out constantly, which yields a representative value for a larger catchment area compared to the surveyed wells in the *Migmagranit*. (Schubert et al., 2003)

In this thesis the results will mainly be characterized either as *Weinsberger Granit* or other Gneiss and Granites of the Bohemian Massif according to Vohryzka (1973b), but some selected waters will be surveyed for their bedrock in Chapter 15.

Table 4.1: *Overview of ground water bodies in Upper Austria (Amt der Oberösterreichischen Landesregierung, 2008)*

Name of ground water body (GWB)	**Character of GWB**	**Range**	**Type of ground water**	**Area (km^2)**	**Bedrock according to Vohryzka**
Böhmische Masse (Bohemian Massif)	Group	Surface near	predominant crevice	3505	1, 2, partly 3, 10
Eferdinger Becken	Individual	Surface near	predominant pores	120	4
Linzer Becken	Individual	Surface near	pores	96	4
Machland	Individual	Surface near	pores	112	4
Schlierhügelland	Group	Surface near	predominant crevice	1286	3, 4, 5, 10
Kobernaußerwald-Hausruck	Individual	Surface near	pores	916	3, 4, 5, 6, 10
Salzach-Inn-Mattig	Group	Surface near	predominant pores	630	4
Oberinnviertler Seenplatte	Group	Surface near	predominant pores	212	6
Welser Heide	Individual	Surface near	pores	205	4
Vöckl-Ager-Traun-Alm	Individual	Surface near	predominant pores	404	4, 6
Traun-Enns-Platte	Group	Surface near	predominant pores	811	3, 4, 5, 6
Unteres Ennstal	Individual	Surface near	pores	67	4
Flyschzone	Group	Surface near	predominant crevice	994	4, 6, 7
Nördliche Kalkalpen (Northern limestone Alps)	Group	Surface near	karst	2617	4, 6, 7, 8, 9
Tertiärsande	Group	Deep	predominant pores – depth ground water	3360	3, 4, 5, 10, partly 1, 2
Thermal water	Group	Deep	predominant karst – depth ground water	1625	3, 4, 5, 10

Chapter 5

Materials and Methods

5.1 Sampling

The drinking water sampling was carried out in **two phases**. First – to get a general idea – a survey with representatively distributed samples among Upper Austria. Afterwards more sophisticated samplings was operated based on defined selection criteria which are specified below.

5.1.1 Survey Sampling

The survey sampling was carried out by some institutions and departments for different objectives and feasibilities. So samples were taken at **water works** on the one hand and at **small and private water supplies** and **private wells** on the other. Additionally some locations of the existing routine water sampling programme *Wassergüteerhebungsverordnung* – **WGEV** (Water Quality Monitoring Ordinance) (Amt der Oberösterreichischen Landesregierung, 2008) were selected for sampling. The sampling points within this framework were selected in an optimized way to get overview data area-wide representatively in all geological areas. Special interest was spent to the Bohemian Massif (Mühlviertel).

Within the part of the project of the government of Upper Austria "Radiation exposure of workers in Upper Austrian drinking water supplies" (Ringer et al. (2006a,b), see Chapter 1.1) **50 water supplies** were selected for investigation (selection criteria: production rate >10 l/s in Upper Austria, <5 l/s in the Bohemian Massif). In the course of this project it was possible to take along water samples for this work. The requirements for water sampling were: at least 2 samples at representative locations within the water unit, preferential one directly at wells and one after water treatment and tanks directly before the first consumer. In practice it was not always possible to comply with these requirements in detail because of lack of time and huge distances between the wells and water supplies. But in general the sampling at water supplies provided a representative selection of samples for Upper Austria. A total of **79 water samples** were taken in the period between October

2004 and January 2005. Each sample consists of two litres in two standardized one litre glass bottles (AFNORM).

The second possibility for receiving water samples was within the **routine water sampling** of the **Austrian Agency for Health and Food Safety** (AGES), CC Hydroanalytic, Linz at **private wells** and **small water supplies** and communes. In the period between November 2004 and January 2005, **40 water samples** were taken at representative selected wells and small water supplies in different regions in Upper Austria.

In Austria the protection of groundwater is laid down in the Austrian Water Act and in regulations. In the framework of the **Water Quality Monitoring Ordinance** the status of the groundwater is measured nationwide four times per year at nearly 2050 monitoring sites for groundwater in porous media, in **fractured aquifers** and in **karstic areas** (WGEV). Responsibility lies by the Federal Ministry for Agriculture, Forestry Environment and Water Management in co-operation with the provincial authorities and the Federal Environmental Agency (Umweltbundesamt Wien, 2007a). For this work groundwater samples were collected at selected **86 sampling sites** within Upper Austria in the course of the monitoring program in December 2004.

A total of **205 water samples** were taken in the course of the above described survey sampling. The sampling under different conditions and objectives delivers miscellaneous insights regarding the research intention. Sampling in water works, small water supplies, communes and private wells gives a good overview about the radiation exposure of the Upper Austrian population caused by drinking water. On the other hand direct sampling at wells, springs and drills (WGEV) offered basis data about natural radioactivity in ground water in different geological areas and regions in Upper Austria.

5.1.2 Detailed Sampling

According to the survey sampling and analysis it was reasonable for the thesis objectives to take further samples. Because of capacity and cost limitations the additional sample number was limited to about 150. Therefore a **well-founded selection of sampling points** was required and so it was not possible to get samples from other institutions and routine sampling programmes. The samples were taken at well selected locations by myself and colleagues of the University of Natural Resources and Applied Life Sciences (**BOKU**) with assistance from the Environmental Department of the Government of Upper Austria.

The interesting regions for the detailed sampling were selected with help of geologists (Leichtfried, 2004) according to **specific criteria**:

- **Second sampling** of all sampling points with elevated activity concentration ($\geq$ class 3) in the survey sampling
- Sampling in communities at relevant geological faults: *Bayrischer Pfahl*, *Donaustörung*, *Haselgraben* (Leichtfried (2004); Figure 5.1)
- Sampling in communities with elevated **Uranium** and **Thorium** concentrations according to the Geochemical Atlas of Austria (Thalmann et al., 1989) (Figure 5.1)
- Sampling in communities in geological zones with rocks, expected to have elevated radon concentrations (*Hauptdolomit*, *Altenberger Granit*, *Weinsberger Granit* (Schubert et al., 2003, Vohryzka, 1973a)

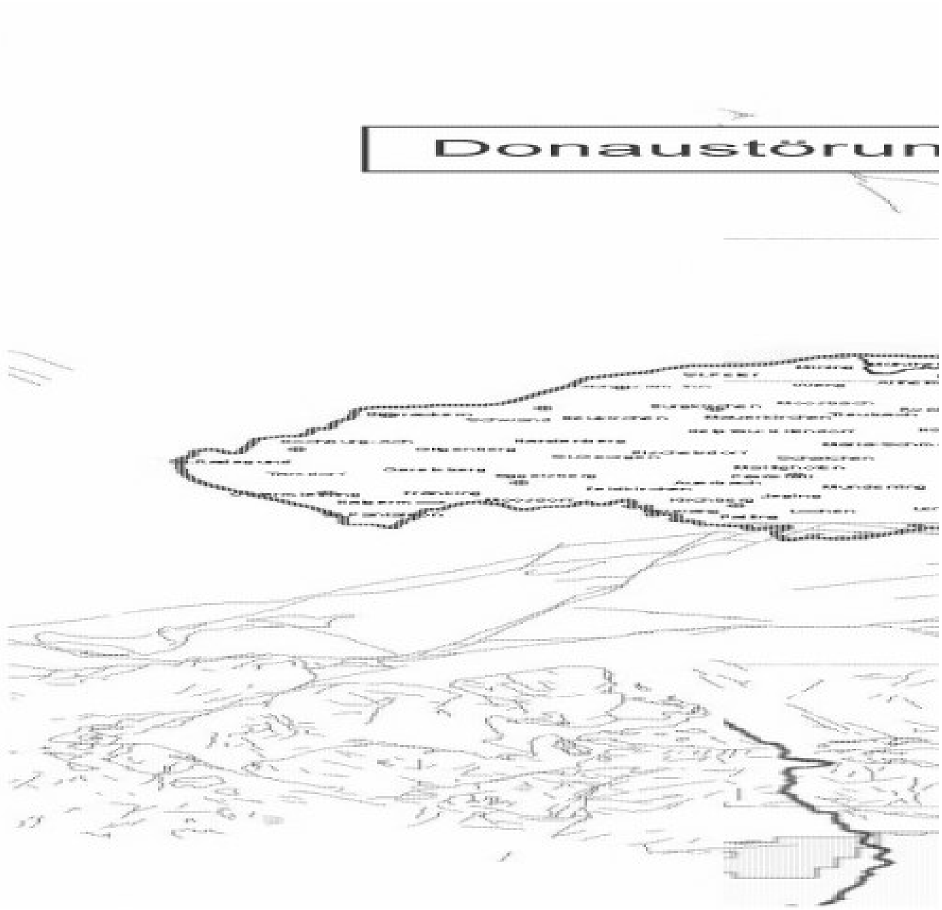

Figure 5.1: *Uranium in Riversediments (from: Geochemical Atlas of Austria (Thalmann et al., 1989), legend see Annex A1) and relevant geological faults in Upper Austria*

The detailed sampling was carried out in **5 sampling phases** from June 2005 to March 2006 in different communities selected in regard to the above mentioned criteria (Table 5.1).

Table 5.1: *Five sampling phases of the detailed sampling*

Date	Location	Communities	Samples
2005-06-20 – 2005-06-23	Central Mühlviertel Lower Mühlviertel	2 communities 3 communities	10 samples 27 samples
2005-07-05 – 2005-07-07	Innviertel (Donaustörung) Upper Mühlviertel	3 communities 2 communities	18 samples 11 samples
2005-08-24 – 2005-08-26	Lower Mühlviertel	6 communities	45 samples
2005-11-08 – 2005-11-10	Southern Upper Austria	6 communities	19 samples
2006-03-09	Southern Upper Austria	1 community	3 samples

Additionally 16 samples were taken at public springs and wells in the investigated regions which are regarded as being pleasant or curative within the population and characterized in a book by Hirsch & Ruzicka (2002) (**”Sacred Wells”**).

So in the framework of detailed sampling a total of **149 samples** were taken in **23 communities**.

The **procedure of sampling** in the detailed sampling phases was a learning process, because of appearing problems and extended questions within the thesis. In the first stage all samples were taken and afterwards analyzed in the laboratory by different methods for various nuclides. Because of the availability of two mobile liquid scintillation counters it was reasonable to measure radon on-site additionally to decide immediately on further samplings and measurements. Therefore the later phases were carried out with a measurement bus and **on-site radon measurements**. Besides, samples were taken at several points within one water unit, to survey the activity concentration gradient within the water flow from the well to the consumer.

The second sampling at the measurement points with elevated activity concentration in the survey sampling was realized because of detailed documentation (e.g. GPS coordinates). The further sampling points were selected within the regions of interest (above mentioned criteria) with help of the water attendants from the communities on the one hand and with contact to the responsible person of smaller water supplies or direct on-site contact to owner of private wells on the other.

The samples were taken **directly** at **springs**, **wells** and **drills** on the one hand to get hydrogeological radiometric basis data. Additionally samples were taken – as mentioned above – at **several points within the water flow** (elevated tank, water treatment facilities) to study the effects and the activity distribution. For radiation protection of the public, activity concentration in drinking water **at taps** in the consumers houses are significant. So additionally samples were taken directly at taps, after leaving water on for at least 10 minutes to measure fresh water.

On every sampling point at least samples for the on-site radon liquid scintillation measurement were taken. Therefore 20 ml glass or plastic vials were used. In these vials different scintillation cocktails for different methods (see below, Chapter 5.2.2) were pipetted before in the laboratory. On-site some ml of the water sample (dependent on the cocktail and the method) were directly pipetted in the vials, sealed and agitated. For verification usually at every sampling point some vials with different cocktails were taken. To avoid scintillation by sunlight the sampling was carried out carefully and covered or if possible inside. For transport from the well to the measurement bus (usually only some meters) a light tight container was used.

Figure 5.2: *Pipetting the water samples directly into measurement vials in a water supply*

Figure 5.3: *Sampling in the 2.5 l container for detailed radiometric analyzes*

Additionally 1 litre standardized glass bottles (AFNORM) were taken at some sampling points to analyze for radon by gammaspectrometry in the laboratory for verification. Besides at nearly every sampling point a water sample was taken in a 2.5 litre plastic container for detailed radiometric and chemical analysis in the laboratory. For stabilization and to avoid adsorption of radionuclides onto the container's material 20 ml nitric acid (65%) was added.

At some sampling points additionally **temperature**, **conductivity** and **pH-value** of the water were determined. Furthermore additional details about the sampling points were documented and **coordinates** recorded by **GPS**. This affords to relate the results with geology, geography, treatment techniques, sampling depths and other conditions at the sampling points.

Figure 5.4: *Equipment for the sampling procedure – GPS, writing utensils for documentation, vials and pipette*

5.2 Radiometric Analysis

The samples taken within the two sampling phases were analyzed for several radionuclides with **varying methods** and in **different laboratories**.

All samples of the **survey sampling** were analyzed for **gross alpha-beta activity**, **Radon-222** (^{222}Rn), **Radium-226** (^{226}Ra), **Tritium** (^{3}H) and **Uranium-238** (^{238}U) in the laboratory of the Austrian Agency for Health and Food Safety (AGES) Vienna, CC Radiation Protection and Radiochemistry. The samples of the **detailed sampling** were supplementary analyzed for **Radium-228** (^{228}Ra) and the radon progenies **Lead-210** (^{210}Pb) and **Polonium-210** (^{210}Po). **Gross alpha** and **gross beta** activity concentrations were measured in these cases instead of gross alpha-beta activity concentrations. Additionally Thorium-232 (^{232}Th) and diverse (heavy) metals were determined. ^{232}Th, ^{238}U and (heavy) metals were analyzed by inductively coupled plasma – mass spectrometry (ICP-MS), all other nuclides by liquid scintillation counting (LSC).

Furthermore all samples of the detailed sampling were measured **on-site** for ^{222}Rn by two mobile liquid scintillation counting instruments (Triathler). The samples which were taken in the water works during the survey were analyzed for ^{222}Rn by gammaspectrometry in the Low Level Counting (LLC) Laboratory Arsenal, University of Natural Resources and Applied Life Science. For verification and comparison to the LSC ^{222}Rn measurements some samples of the detailed sampling were analyzed for ^{222}Rn by gammaspectrometry in the LLC laboratory too. A few of these samples were evaporated and additionally measured by gammapectroscopy for verification and comparison of other radionuclides (^{210}Pb, ^{226}Ra, ^{228}Ra, ^{238}U).

Measurement systems for radioactive isotopes are principally based on the **interaction of ionizing radiation to matter**. Below the different analysis methods and procedures are discussed.

5.2.1 Inductive Coupled Plasma-Mass Spectrometry (ICP-MS)

Mass spectrometry is an analytical method to measure the **mass-to-charge ratio of ions** by generating a mass spectrum, which represents the masses of sample components.

A mass spectrometer consists of **three basic parts** – an ion source, a mass analyzer and a detector system. The technique of mass spectrometry is based on some major steps. First, ions have to be produced from the sample, which are then separated because of their differing masses and detected and generated proportional to their concentration in a mass spectrum. Many different techniques are used to ionize the sample material in the ion source for different types of samples (e.g. electron ionization, chemical ionization, thermal ionisation). In the analysis of this thesis **inductively coupled plasma** (ICP) is used for ionization. This technique is primarily used for metal analysis in liquid samples. However, solid samples can be analyzed using lasers or heated cells to vaporize the sample. Gas samples can be measured by direct introduction into the instrument.

Plasma is a gas that contains a sufficient concentration of ions and electrons to make the gas electrically conductive. The plasma which serves as the ion source of the mass spectrometer is generated in the ICP torch, where the analyzed atoms are converted into ions. The plasma is generated by passing argon through a series of concentric quartz tubes (the ICP torch) that are wrapped at one end by a radio frequency (RF) coil. Energy supplied to coil by the RF generator couples with the argon to produce plasma. The generated plasma has a temperature of about 6000°C.

Besides, the ICP-MS consist of components described below. To get the sample in the instrument, a **introduction system** exists, which consist of peristaltic pump, a nebulizer and a spray chamber. The introduction system brings the liquid sample into a form acceptable to the instrument's plasma (droplets with right velocity and right size e.g. diameter 10^{-6} m). The liquid droplets, which contain the sample matrix and the determining elements, are dried to a solid and then heated to a gas while entering the plasma. When the atoms pass through the plasma, they absorb more energy from the plasma and eventually release one electron to form a singly charged ion. These ions exit the plasma and enter the **interface region**. The interface region links the atmospheric pressure ICP ion source, which operates on a temperature about 6000°C, and the high vacuum mass spectrometer operating near room temperature. The interface consists of two inverted funnel-like devices called cones, which are typically made of nickel platinum and are mounted into a water-cooled metal housing to prevent damage from the heat of the plasma. The distance from the interface to the detector of the ICP-MS is typically about one meter. To avoid collision of the ions with gas molecules in the space between the interface and detector requires creating a vacuum. For this purpose different pumps are combined called the **vacuum system**. Immediately behind the interface a charged metallic cylinder acts as **ion lens**. This lens focuses the positive charged ions into a beam for transmission into the **quadropol**, since the charge on the lens is the same as the charge on the ions and the ions are repelled. The quadropol acts as a **mass filter** and sort the ions by their **mass-to-charge ratio**. It consists of 4 rods with about 20 cm lengths and 1 cm diameter and allows only one mass to pass through to the detector at any given time (Figure 5.5). This is done by the quadropol by setting up the correct combination of voltages and radio frequencies to guide the ions with the selected mass-to-charge ratio between its four rods. Ions without the selected ratio are ejected from the quadropol. The **mass spectrometer** can move to any mass-to-charge ratio needed to measure the elements of interest in a sample. The ions exiting the mass spectrometer hit the **active surface** of the **detector** and generate a measurable electronic signal. The detector is of the type of **dynodes**, where an electron releases each time an ion hits the dynode. The electron released from the first dynode hits the second dynode where more electrons are released. This **amplification by cascading** of electrons effects measurable pulses. By counting the pulses generated by the detector, the system counts the ions that hit the first dynode. The **computerized data system** is used to convert the measured signal intensities into concentrations of each element and generate a report of the results. Besides, all above discussed parts of the ICP-MS are under software control (e.g. ELAN by Perkin Elmer).

ICP-MS has many **advantages** over other technologies like AA (atomic absorption spectrometry) and ICP-OES (Inductively Coupled Plasma Optical Emission Spectrometry). It has low decision limits, little interferences and with quadrupole technique 35 elements can be measured in a sample in two or three minutes. (Perkin Elmer Instruments, 2001)

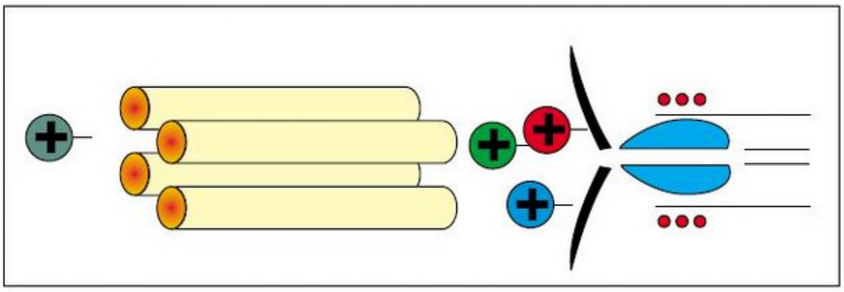

Figure 5.5: *Quadrupol mass filter (Perkin Elmer Instruments, 2001)*

Analysis of Uranium

The water samples were acidified with nitric acid (65%) (HNO_3) during the sampling and were measured without any chemical pre-treatment with the ICP-MS Elan DRC II by Perkin Elmer. Rhodium was used as an internal standard. The method provides a limit of detection well below 1 ng L^{-1} and a good repeatability tested at 3 different concentrations.

5.2.2 Liquid Scintillation Counting (LSC)

Liquid Scintillation Counting is mainly used for detection of low-energetic β-emitting isotopes like ^{3}H and ^{14}C, but α-emitting isotopes can also be detected with high efficiencies. The principle is based on a homogeneous distribution of the radioactive substance in a scintillation cocktail. The counting sample consists of the radioactive sample and the **scintillation cocktail**, which is composed of the scintillator and an organic solvent as carrier substance. The energy of a particle or radiation emitted by the sample material is transferred to the solvent molecules. Some of these exited solvent molecules transfer their energy to the scintillator molecules. This exited scintillator molecules return to ground state by emitting light flashes (**phenomenon of fluorescence**). These emitted photons are converted in an electrical pulse by being absorbed by the photocathode of a **photomultiplier tube** (PMT). This pulse is registered after suitable amplification as a count by electronic equipment corresponding to the emission of the particle or radiation. (ETH Zürich, 2007, University of Wisconsin, 2007, Valkovic, 2000)

A modern LSC instrument has a **multichannel analyzer** (MCA) which counts and sorts the pulses because of their amplitude into different channels. So a total spectrum is registered and a simultaneously measurement of more than one nuclide is possible by using selected and well defined channel ranges. Additionally many LSC instruments are able to distinguish between alpha and beta radiation. This **alpha-beta separation** is based on the fact, that alpha and beta emitting radionuclides produce different shapes at the PMT anode. These pulses are made up of two components – the prompt component and the delayed component. These components occur in different proportions in alpha and beta pulses, with the result, that alpha pulses are longer than beta pulses (Figure 5.7). The longer duration of alpha pulses is the basis of alpha-beta separation by pulse shape discrimination by pulse shape discrimination electronics. In the framework of this thesis the technique of **pulse shape analysis** (PSA) is used in the laboratory of the AGES. This method integrates the charge of the "tail" of the scintillation pulse and compares it with the total charge in the same pulse. Different settings of the PSA level assign the pulse into either a long (alpha-like) or a short (beta-like) category. Thus, different PSA settings allow pulses to be categorized according to their length (shape). Typically, increasing the PSA setting will direct more pulses toward the long or alpha category. (Passo & Cook, 1996)

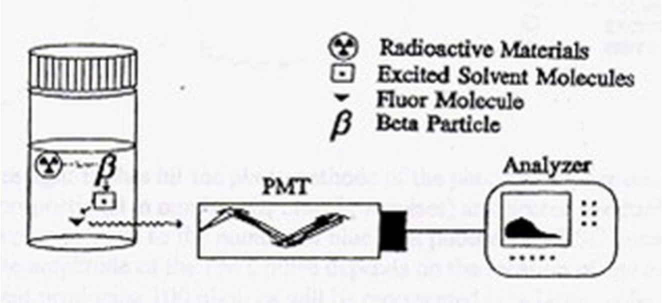

Figure 5.6: *Principle of LSC (University of Wisconsin, 2007)*

In LSC method the scintillator molecules can be regarded as the actual radiation detector in the counter. There are some major advantages of liquid scintillation counting like absent problems of sample self absorption, which is particular important for measurements of low energy beta emitters like ^{3}H and ^{14}C. The detection of emitted particles in LSC technique is highly efficient and may even approach 100% by a **4 π counting**, because of a complete surrounding of the radioactive material by the liquid scintillator. There is no radiation-backscattering from the detector and no absorption of radiation by air or a detector's window between the radioactive sample and the sensitive region of the detector. (Valkovic, 2000)

On the other hand a major disadvantage of LSC is their relatively poor energy resolution. Futhermore **quench effects** may reduce the counting efficiency and background radiation might negatively affect the measurement (Knoll, 2000). Quenching is a phenomenon caused by the energy loss in the process of the energy transfer inside the liquid scintillator and the counting efficiency decreases. Most materials are regarded as quenchers whose quenching strength depends on the material itself and its amount. Coloured samples for example can absorb already built photons (colour quenching). Quenching leads to a shift of a pulse height spectrum to lower energies. These phenomena should be taken into account, whenever the liquid scintillation measurement is performed. (ETH Zürich, 2007, Valkovic, 2000)

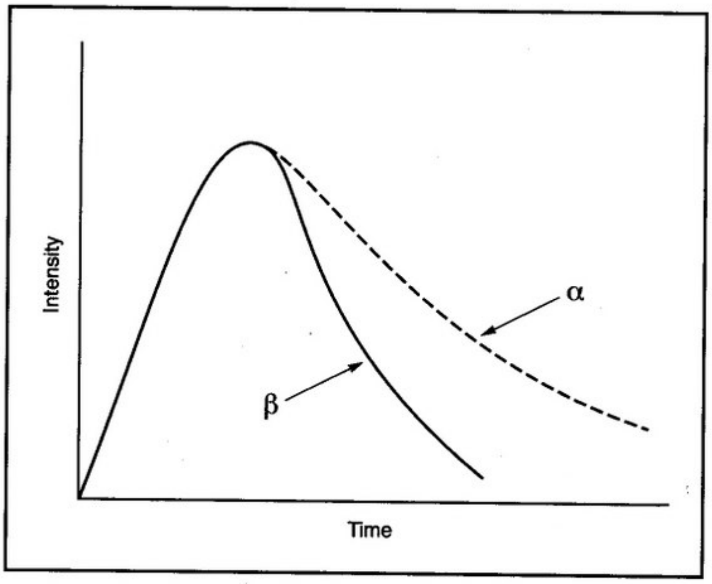

Figure 5.7: *Characteristic light pulse shapes of alpha and beta pulse in a liquid scintillator (Passo & Cook, 1996)*

There are different methods to correct these quenching effects, like the method of an **internal standard** sample. For this purpose a sample with a known counting rate and no quench effects (internal standard sample) is measured additionally to the sample with the unknown quench effect. Then the sample is mixed with the internal standard and measured again. The correction of the quenching can be calculated on the basis of the counting rates (ETH Zürich, 2007). Another method is to use a **quench curve**. The curve establishes the relationship between a quench parameter and the counting efficiency. This curve must be determined by counting a set of samples with the same activity but variable quenching. When the test samples are counted, the quench parameter is determined for each sample. To determine the activity of the measured sample the quenching correction curve can be used. The third method is the external standard method in which also an **external standard** quench curve is used. For this purpose an external gamma source is used for determining the relationship between a quench parameter (caused by the gamma source) and the counting efficiency. (Hidex OY, 2005, Valkovic, 2000).

For liquid scintillation counting technique an important part is the **scintillation cocktail**. Various cocktails with different solvents and scintillators exist for diverse applications. This issue is very miscellaneous and will not be discussed here in detail. More details are worked out for example in Frenzel (1999), Hidex OY (2005), Passo & Cook (1996), Valkovic (2000). One major differentiation is about cocktails application and properties like extraction cocktails or soluble cocktails (Frenzel, 1999, Hidex OY, 2005, Möbius-Ramamonjisa et al., 1998).

Analysis of Gross Alpha, Gross Beta and Tritium

The samples were analyzed for gross alpha, gross beta and Tritium by a liquid scintillation counter (Quantulus 1220 by Perkin Elmer) and different scintillation cocktails-water-mixtures:

- 7 ml water – 13 ml Hisafe 3 (by Perkin Elmer)
- 10 ml water – 10 ml Ultima Gold AB (by Perkin Elmer)

The pulse shape analysis (PSA) level setting for alpha-beta separation is 120. To determine Tritium an analysis range between channel 38 and 164 was chosen (referred to 1024 channels). Gross alpha activity was analyzed comparatively to an ^{241}Am standard in the range between channel 258 and 882. Gross beta activity was determined relatively to a ^{40}K standard in the same channel range.

Analysis of Radon-222 and Radium-226

For ^{222}Rn determination, 10 ml of the water sample are mixed with 10 ml of high efficiency mineral oil scintillator by Perkin Elmer, agitated and measured in equilibrium with ^{218}Po and ^{214}Po by LSC (Quantulus 1220 by Perkin Elmer) (Schönhofer, 1990). Thus the efficiency is nearly 300%.

From another 10 ml of the water sample ^{226}Ra is determined by removing the ^{222}Rn by shaking and waiting until ^{226}Ra is in equilibrium with ^{222}Rn (about 3 weeks) and using the same method as for measuring ^{222}Rn (Landstetter & Katzlberger, 2005).

Analysis of Radium-228, Lead-210, Polonium-210

The simultaneous measurement of ^{228}Ra and ^{210}Pb in one sample is not possible because of their similar beta energies (0.046 MeV and 0.064 MeV). Nevertheless these radionuclides can be determined out of one sample because of a previous **chemical separation** to get one fraction of ^{228}Ra and one fraction of ^{210}Pb and ^{210}Po. For that purpose 1 ml of $Pb(NO_3)$ solution (1.6 mg Pb^{2+}) was added to 1 l of water sample and the sample is evaporated to approximately 80 ml. This step should happen shortly after sampling because ^{222}Rn is removed in the process. By storing the sample for a longer time, ^{222}Rn is decaying with a half life of 3.82 d and ^{210}Pb (half life of 22.3 y) is built. They can not achieve radioactive equilibrium therefore the part of ^{210}Pb from ^{222}Rn has to be taken into account in the ^{210}Pb analysis. This can be done by using the ratio of the half lives of ^{222}Rn and ^{210}Pb (Equation 5.1, Katzlberger et al. (2001)). This results in 0.43 mBq ^{210}Pb from 1 Bq ^{222}Rn in 2 weeks of storage.

$$A_{Pb-210} \approx \frac{\tau_{Rn-222}}{\tau_{Pb-210}} \cdot A_{0\,Rn-222} \cdot \left(1 - e^{-\frac{ln2}{\tau_{Rn-222}} \cdot t}\right) \tag{5.1}$$

A_{Pb-210} . . . activity concentration of ^{210}Pb built from ^{222}Rn in time between sampling and measurement
$A_{0\,Rn-222}$ . . . activity concentration of ^{222}Rn
τ_{Rn-222} . . . half life of ^{222}Rn
τ_{Pb-210} . . . half life of ^{210}Pb
t . . . time between sampling und measurement

For continuing processing the 80 ml were further evaporated to near dryness and repeated with fuming HCl. The residue were dissolved in 15 ml 1 M HCl. The precipitation of Pb, Bi and Po was done at pH 1.5 with 1 M Na_2S. After centrifugation and filtration the filtrate is used for ^{228}Ra analysis. In this step also ^{234}Th (half life of 24.1 d) was separated from ^{228}Ra to avoid interference because of its beta energy.

The precipitate was dissolved in concentrated HNO_3 and fumed off with concentrated HNO_3 to delete sulphide and fumed off with HCl to build chlorides. The sample is evaporated to near dryness, dissolved in 2 ml 1 M HCl and 12 ml 1.5 M H_3PO_4 and stored in the refrigerator until ^{210}Bi is in equilibrium with ^{210}Pb. After 2 weeks ^{210}Bi and ^{210}Po selectively extracted with the organic extraction cocktail Polex and measured by LSC (Quantulus1220 by Perkin Elmer) with a PSA setting of 130. For determining the recovery of the lead precipitation 3.8 ml of the aqueous phase are diluted to 50 ml with 1% HNO_3 and measured by atom absorption spectrometry (AAS) (Landstetter & Katzlberger, 2005).

The filtrate for the ^{228}Ra analysis is diluted with 35 ml water and brought to pH 2. Afterwards 1 ml of 1 M H_2SO_4 is addes and radium is coprecipitated with $BaSO_4$ by adding 2 ml of $Ba(NO_3)_2$ under boiling conditions. After cooling, the precipitate is centrifuged, washed with water and dissolved under cooking with 5 ml of 0.25 M EDTA. After evaporation to a volume of about 2 ml the solution is mixed with 18 ml of the scintillation cocktail Hisafe 3 and measured by LSC. For determination of the chemical recovery via barium the exact volume of the solution is weighted out and 20 µl are diluted to 20 ml with 1% HNO_3 and measured by ICP-MS (Landstetter & Katzlberger, 2005).

Analysis of Radon-222 by Mobile LSC Triathler

As mentioned above, all samples of the detailed sampling were measured on-site by the mobile liquid scintillation counter **Triathler** (Hidex). No ^{222}Rn losses occur because of direct on-site pipetting of the water sample into the measurements vials and direct measurement without transport and decanting before measuring. Measuring ^{222}Rn in water with the mobile liquid scintillation counter Triathler was tested and verified in this thesis to determine a suitable method for future projects. This implies multiple measurements for comparison reasons with different methods.

The Triathler is a **mobile** liquid scintillation instrument with an **alpha-beta separation**. The detector type is a diffuse white reflector on aluminium. The energy range from 2 keV to 2000 keV and has an alpha counting efficiency of >80% for ^{222}Rn and a beta counting efficiency of >20% for ^{3}H. The lower decision limit for ^{222}Rn in water is 3 Bq/l. The instrument has an **instant quench correction** to count samples quench corrected without using quench standards. The alpha-beta separation in the Triathler uses the technique discussed above based on different light pulses duration of alphas and betas. Alpha and beta spectra show a number of pulses as a function of pulse energy. In addition to this, each pulse is collected to a two dimensional graph, one dimension (x-axis) shows the pulse height (total energy) while the second dimension (y-axis) indicates pulse length (Figure 5.9). This 2D graph helps to set correct parameters for separation (e.g. PLI – see below). For more technical details see the technical data sheet of the instrument. (Hidex OY, 2005)

The measurements were carried out with **two different Triathlers**. The older one (version 1.6) belongs to the Atomic Institute of the Austrian Universities (AI), the newer one (version 1.8) to the Environmental Department of the Government of Upper Austria (OOE). Within this thesis the compatibility of the two instruments was tested (see Chapter 10.3), and after verification both were used for the ^{222}Rn measurements. For this purpose both LSC instruments were operated in a measurement bus on-site (Figure 5.8). The two Triathler were connected to a notebook by a RS-232 C serial output interface and an USB adapter and controlled by the software CommFiler. For every measurement the total alpha and total beta counts, counts per minute, alpha-spectrum, beta-spectrum and a 2D spectrum (discussed above, see Figure 5.9) were recorded, stored and transferred to excel by macros for further processing and storing (Hidex OY, 2005).

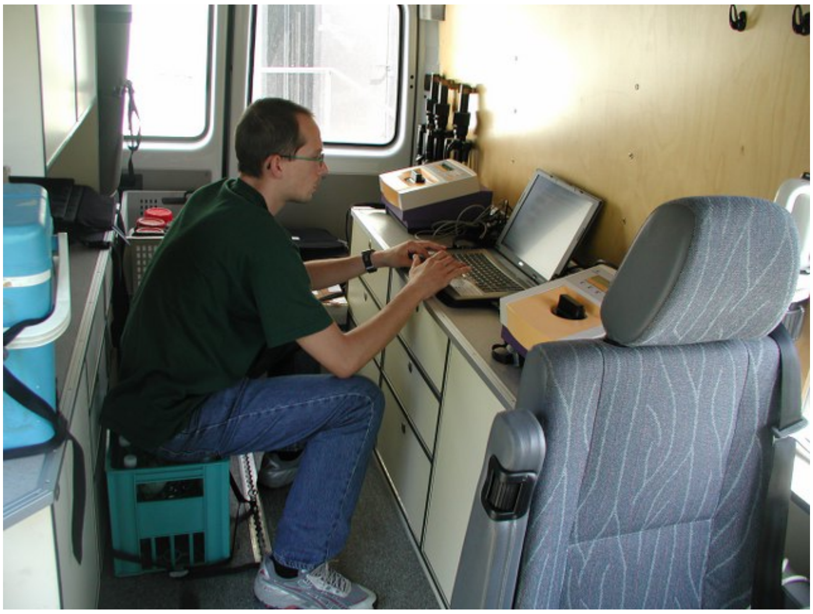

Figure 5.8: *Measurements with the mobile LSC-instruments Triathler on-site in a measurement bus*

For comparison and optimisation the analysis of the drinking water samples were carried out with **different scintillation cocktails** (Aqualight, Maxilight, Ultima Gold). Aqualight and Ultima Gold are water soluble cocktails by Hidex and Perkin Elmer. By using these cocktails the samples can be measured immediately. Maxilight – an extraction cocktail by Hidex – needs an at least 3 hour waiting time before measuring because radon has to migrate from the water into the cocktail. Also water samples with high percentage of solved and particulate substances (e.g. spa-water) can be measured with this cocktail without losses of counts by quench effects.

At nearly every sampling point the sample was mixed with Aqualight and Maxilight, Ultima Gold was used sporadically, only for comparison reasons.

As mentioned above for Triathler measurements 20 ml vials were used, and the water-cocktail mixture depends on the cocktails (Table 5.2).

Table 5.2: *Mixing ratios of water and the different used scintillation cocktails*

Name of cocktail	Mass of cocktail (ml)	Mass of water (ml)
Aqualight	12	8
Maxilight	10	10
Ultima Gold	2	10

For a sufficient measurement uncertainty (about 10%) a measurement time of 10 minutes is necessary.

For an optimal alpha-beta separation the **pulse length index** (PLI) and the **channel ranges (window)** for the analysis have to be determined for every Triathler and every cocktail by test measurements (Table 5.3). Normally these settings can be used for all measured water samples, however every spectrum should be controlled for right alpha-beta separation and right window ranges for alpha and beta counts (Figure 5.9).

The Triathler output for the described ^{222}Rn measurements are alpha counts per minute (cpm_{alpha}). The radon activity concentration (c_A) in Bq/l can be calculated by the Equation 5.2 including background countrate ($cpm_{background}$), water volume in the vial (V) (in litre l) and alpha-efficiency (ε). A correction factor of 3 is required in the equation because 3 alphas (^{222}Rn, ^{210}Po, ^{214}Po) are count during measurement (Frenzel, 2004).

$$c_A = \frac{cpm_{alpha} - cpm_{background}}{3 \cdot \varepsilon \cdot V \cdot 60} \qquad (5.2)$$

Numerous test measurements with radon free water and cocktails showed that the **background** is negligible. The alpha efficiency (ε) is according to the Triathler users' manual (Frenzel, 2005) about 80–90% – in these calculation 80% were used.

Table 5.3: *Window and PLI adjustment for the different Triathlers and cocktails*

Name of Cocktail	Name of Triathler	Alpha window	Beta window	PLI
Aqualight	AI	64–400	32–1000	384
Aqualight	OOE	480–1000	100–1000	384
Maxilight	AI	288–1024	32–1000	544
Maxilight	OOE	300–1000	100–1000	480
Ultima Gold	AI	64–400	32–1000	384
Ultima Gold	OOE	480–1000	100–1000	384

The **uncertainty** of the calculated radon activity concentration (Δc_A) in Bq/l is determined by error propagation (Equation 5.3).

$$\Delta c_{ARn} = \sqrt{\left(\frac{\Delta cpm_{alpha}}{180 \cdot \varepsilon \cdot V}\right)^2 + \left(\frac{-cpm_{alpha} \cdot \Delta\varepsilon}{180 \cdot \varepsilon^2 \cdot V}\right)^2 + \left(\frac{-cpm_{alpha} \cdot \Delta V}{180 \cdot \varepsilon \cdot V^2}\right)^2} \tag{5.3}$$

For the uncertainty of alpha counts the counting statistics uncertainty ($\sqrt{cpm_{alpha}}$) was estimated. For the uncertainty of the alpha efficiency 8% was used and for uncertainty of the water volume 0.3% was set according to the users' manual of the pipette (Eppendorf, unknown).

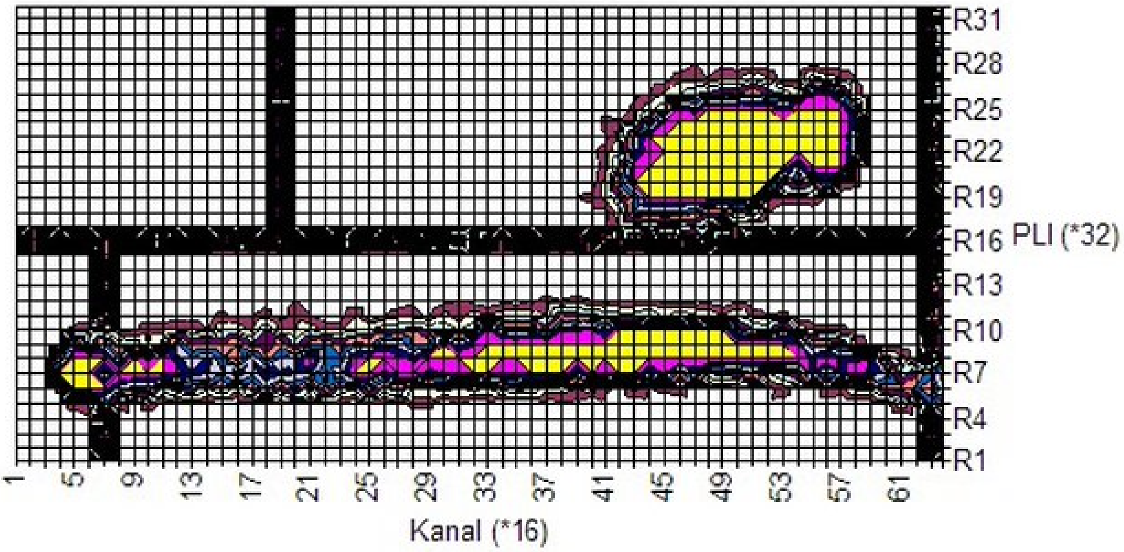

Figure 5.9: *Example for alpha beta separation with the Triathler*

5.2.3 Gammaspectrometry

Because of **interactions of photons with the detector material** secondary electrons originates because of **photo effect** (detector-atom absorbs the energy of the incident photon entirely), **Compton effect** (incident photons are scattered by electrons of the detector) or **pair production** (positron-electron pair is created by the incident photon) (Figures 5.10 – 5.12). After absorption of these particles in the detector, electric pulses accrue which are proportional to the electron energy. The pulse high spectrum is characterized by a series of approximately Gaussian shaped peaks superimposed on a continuum. Because of statistical variability of the elementary processes in the detector no sharp lines are obtained in the pulse high spectrum. In **gamma spectra** not only the characteristic peaks at the gamma energies occur, but also further maxima caused by different interaction of gamma radiation in the detector and the environment. This may complicate analysis and interpretation of the spectrum (Ivanovich & Harmon, 1982, Knoll, 2000, von Philipsborn, 1998).

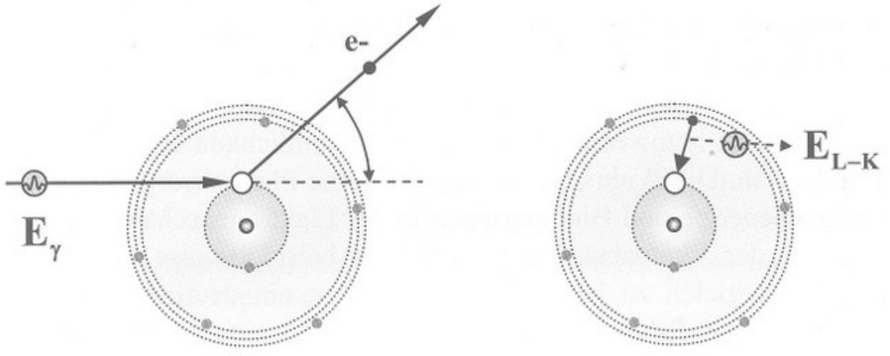

Figure 5.10: *Possible interactions of Photons with the detector material: photo effect (Krieger, 2004)*

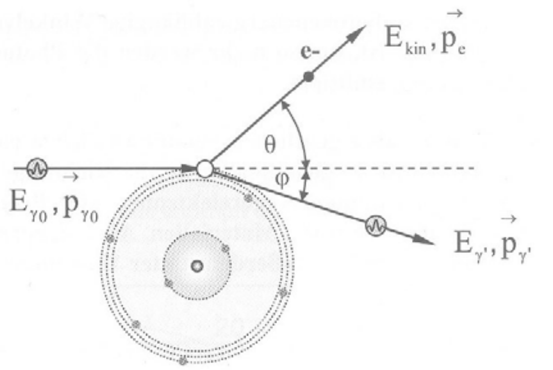

Figure 5.11: *Possible interactions of Photons with the detector material: Compton effect (Krieger, 2004)*

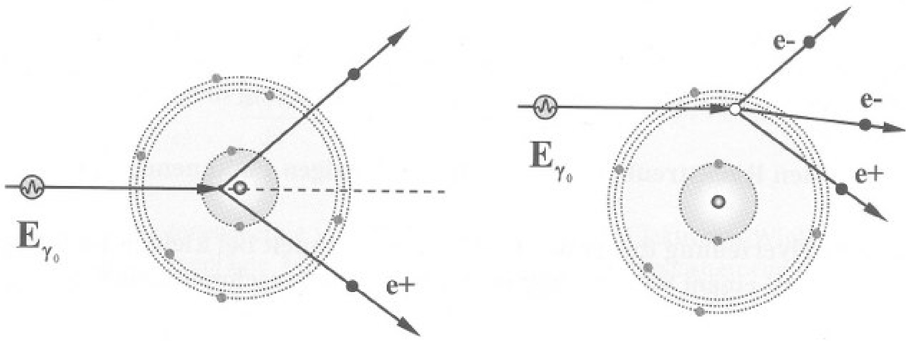

Figure 5.12: *Possible interactions of Photons with the detector material: pair production ((Krieger, 2004)*

Different types of **gamma detection instruments** exist, like scintillation counters (e.g. NaI detectors), ionisation chambers and semi conductor detectors. **Semiconductor detectors** are most frequently used for gammaspectrometry because of some advantages such as high energy resolution, good stability, excellent timing characteristics and simplicity of operation (Knoll, 2000). In the framework of this thesis only semiconductor detectors were used, so just this method is described below.

Semiconductor detectors have a *p-n* or *p-i-n* diode structure. The intrinsic (*i*) region is created by depletion of charge carriers when a reverse bias is applied across the diode. When photons interact within the **depleted region**, charge carries (positive holes and negative electrons) are freed and are swept to their respective collecting electrode by the electric field. The resultant charge is integrated by a charge sensitive **preamplifier** and converted to a **voltage pulse** with amplitude proportional to the original photon energy. Since the depletion depth is inversely proportional to net electrical impurity concentration, and since counting efficiency is also dependent on the purity of the material, large volumes of very pure material are needed to ensure high counting efficiency for high energy photons. (Valkovic, 2000)

The common materials for semiconductor detectors are **Germanium** (Ge) and **Silicium** (Si). There are three fundamental differences between Ge and Si detectors – the energy gap, the atomic number and the mobilities of the major carriers (see Table 5.4). Together with the purity they influence the thickness of the depletion region of a biased *p-n* junction. Less energy is needed to create an **electron-hole pair** in Ge than in Si. Thus, a Si detector may be used at room temperature. A Ge detector has to be cooled to 77 K to reduce the leakage current due to thermal generation of charged carriers to an acceptable level. So a Ge detector has to be operated inside a **vacuum chamber** and cooled to **liquid nitrogen temperatures**. The sensitive detector surfaces are thus protected from moisture and other condensable contaminates. For efficiency of a gamma detector a material with high photoelectric cross section should be used. The **photoelectric cross section** depends roughly on the fifth power of the atomic number Z, so Ge beats Si by one to two orders of magnitude. (Fettweis & Schwenn, 1998)

Table 5.4: *Some characteristics of Ge and Si detectors (Fettweis & Schwenn, 1998)*

	Ge (at 77 K)	**Si (at 300 K)**
Electron mobility (cm^2/Vs)	36000	1350
Hole mobility (cm^2/Vs)	42000	480
Energy needed to create 1 electron-hole pair	2.96 eV	3.62 eV
Atomic number Z	32	14
Forbidden energy gap	0.746 eV	1.115 eV

Gamma detectors must have a depletion layer thickness of several cm in order to enhance the probability of an interaction of a γ-ray with the sensitive detector material. In former times this was realized by compensating the excess acceptor ions of the p-type crystal with Li donor ions by a time-consuming process called lithium drifting. The big disadvantage of these detectors was, that the mobility of the Li^+ ions in Ge is so high that the detector had to be cooled not only to reduce the leakage current while using, but also to prevent the precipitation of Li. Uncooled, a detector would become damaged after a few hours. This fact also reduced the detectors dimension and thus its efficiency. Today **high purity Ge-detectors** (HPGe) are used. Large Ge crystals with low impurity levels of p or n type are grown (on 10^{10} atoms of germanium only one atom of impurity). These detectors can be stored at room temperature and have to be cooled only during operation. Detectors of different size or geometry are available like **planar detectors**, **coaxial detectors**, **well-type detectors** and **low energy germanium detectors**. Ge-detectors can also differ in the material of the entrance window (e.g. Al or Be) and the cryostat construction material. (Fettweis & Schwenn, 1998, Valkovic, 2000)

Analysis of Radon-222

^{222}Rn is not a γ-emitter, so instead of ^{222}Rn the short lived progenies ^{214}Pb and ^{214}Bi are measured by gammaspectrometry (gamma energies at 351.9 and 609.3 keV). For that purpose no further sample treatment is necessary. Waters samples are taken directly in 1 l AFNORM glass bottles on-site because the gamma detectors are calibrated to this geometry. So the samples are measured in these glass bottles for some hours on the detectors.

For ^{222}Rn measurements two coaxial Ge-detectors (p-type) (PGT DI718, Harshaw HP 045) with copper windows are used at the Low Level Counting Laboratory Arsenal. The relative efficiency of the detectors is 33.7% and 21.6% respectively, the energy spectra range from 200 to 3000 keV. The detectors are cooled with liquid nitrogen.

For analysis of the gamma spectra the software Genie 2000® by Canberra is used and for determining the ^{222}Rn activity concentration out of the gamma spectrum a laboratory made MS Access code is applied, which takes into account the sample geometry (filling height, density), detector geometry, coincidence summing losses, measuring time and radioactive decay from the sampling until the measurement date and time.

Analysis of ^{210}Pb, ^{238}U, ^{226}Ra, ^{228}Ra

For analyzing ^{210}Pb, ^{238}U, ^{226}Ra, ^{228}Ra by gammaspectrometry the water sample of the AFNORM glass bottle was evaporated after the radon measurement to concentrate the activity of the nuclides in the water sample and to reduce measurement time. The approximately one litre sample is filled in a 2 litre beaker with small plastic bag in it and than evaporated in a drier at 105°C. The residue is boxed, together with the plastic bag in a polystyrene measurement tin (64 mm diameter, 12 or 14 mm height) and sealed hermetic. The gamma detectors are calibrated to this geometry and the samples have to be measured for some days because of their less height and density. Instead of measuring ^{226}Ra directly, again progenies are measured (^{214}Bi with gamma energies at 609 and 1120 keV and ^{214}Pb with gamma energy at 352 keV), and therefore the sealed measurement tin has to wait 3 weeks before measuring, for arising radioactive equilibrium. ^{228}Ra is measured via ^{228}Ac with gamma energy of 911 keV.

For the measurements of ^{226}Ra and ^{228}Ra the above described coaxial detectors are used. For the measurement of ^{210}Pb and ^{238}U a planar Ge-detector (Silena NIGP 2010) is used with and an Beryllium window, an active area of 2000 mm and an energy range from 20 to 700 keV, because of the low gamma energies of these nuclides (46.5 and 63.6 keV).

The analysis of the spectra is carried out analogous as discussed above.

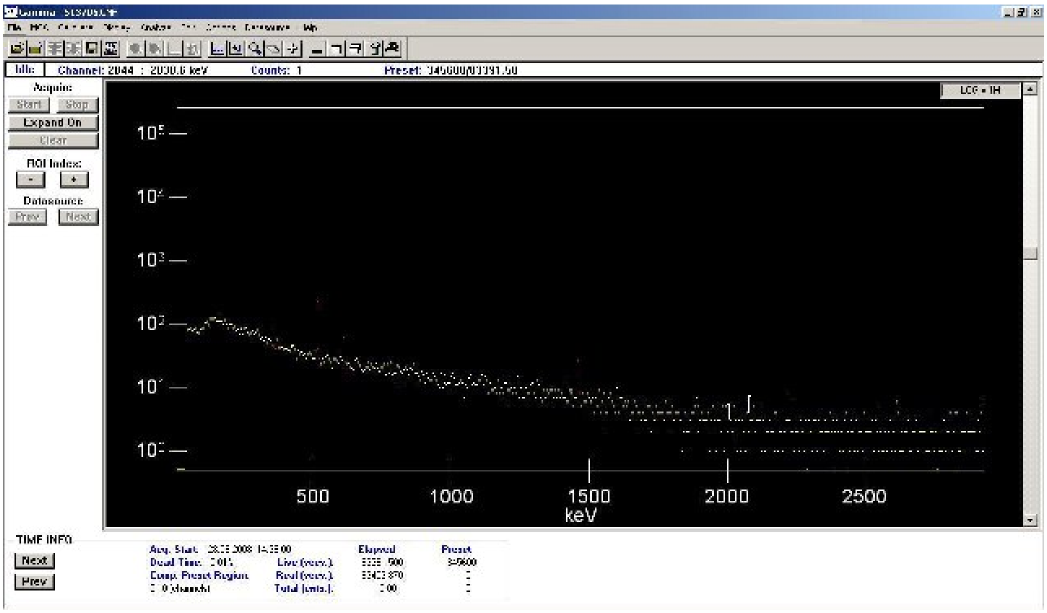

Figure 5.13: *Screenshot of the Genie 2000 program with a gamma spectrum*

Chapter 6

Classification of Activity Concentrations

As discussed in Chapter 5, **354 water samples** were taken in the framework of this thesis in Upper Austria (205 in the survey sampling, 149 in the detailed sampling). Figure 6.1 shows the survey sampling points in light blue and the detailed sampling points in dark blue.

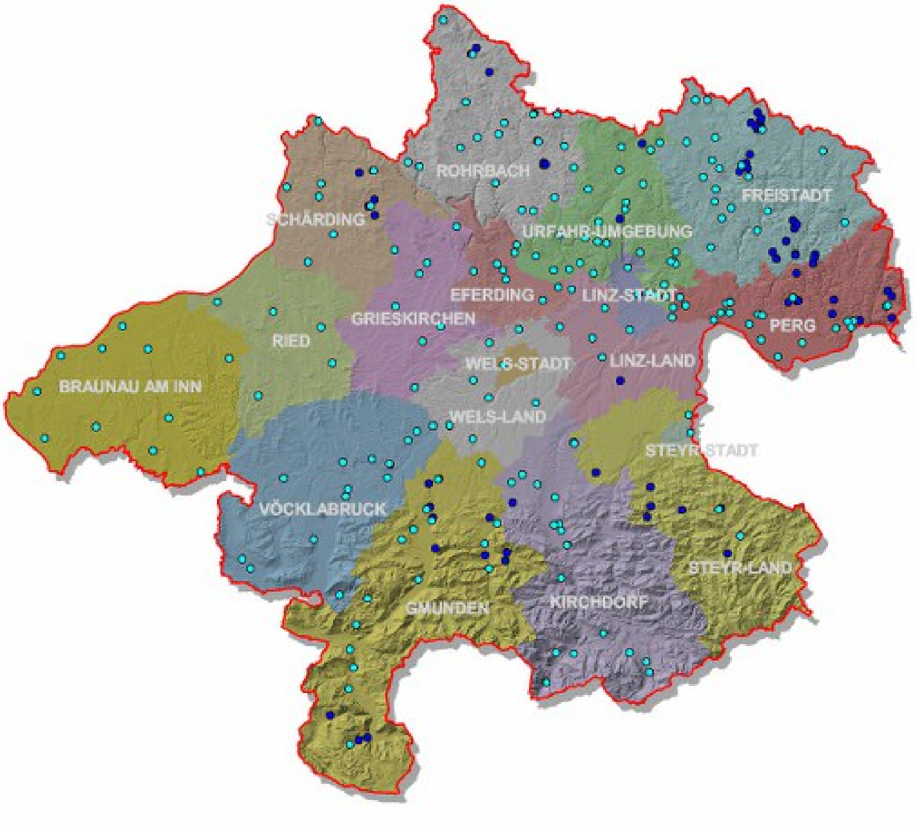

Figure 6.1: *The Province of Upper Austria with all measurement points of the two sampling phases*

For a better **demonstration**, **comparability** and **interpretation** of the natural radioactivity in drinking water measurement results a **classification** was implemented. The classification was defined for the activity concentrations of every measured nuclide in four classes relating to **applied standards**. For ^{3}H, ^{222}Rn and ^{238}U existing activity concentrations standards and recommendations were used, for all other nuclides activity concentration values were calculated out of existing dose standards. In the classification the **intersection** between class 2 and 3 is the standard or recommendation limit of the activity concentration of every nuclide. The intersection between class 1 and 2 identifies one tenth of this value, the intersection between class 3 and 4 the decuple (Tables 6.3 and 6.4).

For ^{3}**H** the activity concentration of 100 Bq/l was used according to applied standards (European Commission, 1998, Republik Österreich, 2001). For ^{222}**Rn** the activity concentration of 100 Bq/l was assumed by the "Commission Recommendation of 20 December 2001 on the Protection of the Public Against Exposure to Radon in Drinking Water" (European Commission, 2001). The used ^{238}**U** activity concentration value of 0.37 Bq/l followed the "Guidelines for Drinking-Water Quality, 2nd Edition" of the World Health Organisation (WHO, 1993). In these guidelines a guidance level for uranium of 30 µg/l is established, which corresponds to a ^{238}U activity concentration of 0.37 Bq/l. This guidance level is based on the chemo-toxic effect of uranium, not on the radioactive exposure. However, in the "Guidelines for Drinking-Water Quality, 3rd Edition" (WHO, 2004) the guidance level for uranium is halved to 15 µg/l, which corresponds to a ^{238}U activity concentration of 0.19 Bq/l. This level was not used in the classification. In Chapter 8 the ^{238}U classification is compared with these new guidance level applied on the measurement results to verify the dimension of discrepancy.

For **all other nuclides** the activity concentration values were calculated according to the existing dose limit for the **total indicative dose** (TID) of 0.1 mSv/a (European Commission (1998), Republik Österreich (2001), Table 6.2). This was done by dose conversion factors for adults (EC, 1996 Table 6.1) according to ÖNORM S 5251 (Austrian Standards Institute, 2005b) with an adult annual water consumption of 730 l, (see Chapter 16). This calculation results in activity concentrations for ^{226}**Ra** to 0.5 Bq/l and for ^{228}**Ra** to 0.2 Bq/l which correspond with the reference concentrations for radioactivity in drinking water given in the Draft of European Commission (2005) (see Chapter 2). For ^{210}**Po** and ^{210}**Pb** activity concentrations of 0.1 Bq/l and 0.2 Bq/l were calculated, which correspond with the Commission Recommendation for Radon in drinking water (European Commission, 2001). Tables 6.3 and 6.4 show the corresponding classification by effective dose. An activity concentration calculation for Tritium with a dose estimation of 0.1 mSv/a would yield 7600 Bq/l, because of its low dose conversion factor (WHO, 2004). The same calculation for ^{238}U yields 3 Bq/l, which corresponds with European Commission (2005) and WHO (2004), but this value takes into account only the radiological toxicity, not the chemical.

For **gross alpha** and **gross beta** activity concentrations 0.1 Bq/l and 1 Bq/l were determined according to the gross alpha and gross beta screening activity concentrations defined in European Commission (2005) given in the drinking water directive (European Commission, 1998). The WHO published screening levels for gross alpha activity of 0.5 Bq/l and also 1 Bq/l for gross beta activity (WHO, 2004) (Chapter 3.1). For **gross alpha-beta** activity concentration 1 Bq/l was estimated.

Table 6.1: *Dose conversion factors for adults for selected nuclides (European Commission, 1996)*

Nuclide	**Dose conversion factor (>17a) (Sv/Bq)**
^{226}Ra	$2.8 \cdot 10^{-7}$
^{228}Ra	$6.9 \cdot 10^{-7}$
^{210}Po	$1.2 \cdot 10^{-6}$
^{210}Pb	$6.9 \cdot 10^{-7}$
^{238}U	$4.5 \cdot 10^{-8}$
^{3}H	$1.8 \cdot 10^{-11}$

Table 6.2: *Dose Classification for ^{226}Ra, ^{228}Ra, ^{210}Po, ^{210}Pb*

Class	**Effective dose**
1	< 0.01 mSv/a
2	0.01 – < 0.1 mSv/a
3	0.1 – < 1.0 mSv/a
4	≥ 1.0 mSv/a

Table 6.3: *Activity concentration classification used in this thesis*

Gross alpha-beta	**Class**	**Activity concentration (Bq/l)**
	1	<0.10
	2	0.10 – 0.99
	3	1.00 – 9.9
	4	≥ 10.0

Gross alpha	**Class**	**Activity concentration (Bq/l)**
	1	<0.010
	2	0.010 – 0.099
	3	0.100 – 0.99
	4	≥1.00

Gross beta	**Class**	**Activity concentration (Bq/l)**
	1	<0.10
	2	0.10 – 0.99
	3	1.00 – 9.9
	4	≥10.0

^{3}H	**Class**	**Activity concentration (Bq/l)**
	1	<10
	2	10 – 99
	3	100 – 999
	4	≥1000

^{226}Ra	**Class**	**Activity concentration (Bq/l)**
	1	<0.050
	2	0.050 – 0.49
	3	0.50 – 4.9
	4	≥5.0

Table 6.4: *Activity concentration classification for all nuclides, part 2*

^{228}Ra	**Class**	**Activity concentration (Bq/l)**
	1	<0.020
	2	0.020 – 0.19
	3	0.20 – 1.9
	4	≥2.0

^{222}Rn	**Class**	**Activity concentration (Bq/l)**
	1	<10
	2	10 – 99
	3	100 – 999
	4	≥1000

^{238}U	**Class**	**Activity concentration (Bq/l)**
	1	<0.037
	2	0.037 – 0.369
	3	0.370 – 3.69
	4	≥3.70

^{210}Pb	**Class**	**Activity concentration (Bq/l)**
	1	<0.020
	2	0.020 – 0.19
	3	0.20 – 1.9
	4	≥2.0

^{210}Po	**Class**	**Activity concentration (Bq/l)**
	1	<0.010
	2	0.010 – 0.099
	3	0.100 – 0.99
	4	≥1.0

Chapter 7

Results of the Survey Sampling

All detailed results are listed in tables in Annex A2.

In the framework of the survey sampling **205 drinking water samples** were measured for ^{222}Rn, 191 are above decision limit (0.5–6.5 Bq/l). No sample was detected with a ^{222}**Rn** activity concentration in class 4, twelve samples are in class 3. The two highest ^{222}Rn activity concentrations (344 ± 38 and 342 ± 38 Bq/l) were detected in the same village in ”central Mühlviertel” (district of Urfahr-Umgebung), which are surveyed geologically in Chapter 15.12. Another nine samples with a ^{222}Rn activity concentration in class 3 were distributed in the area of Mühlviertel (districts of Rohrbach, Freistadt, Urfahr-Umgebung and Perg), one is located in the Innviertel (district Schärding), which may be related to the so called ”Danube fault” (see also Figure 15.4). This corresponds well with previous WGEV measurements, which also show higher radon activity concentrations in these regions (Ditto et al., 1999). Figure 7.1 illustrates the classified radon measurement points of the survey sampling – with green points for class 1, yellow points for class 2 and orange ones for class 3.

202 samples were analyzed for gross alpha-beta, ^{3}H, ^{238}U and ^{226}Ra. 3 samples of the survey sampling phase could not be analyzed for these nuclides because of problems in the laboratory at this time.

For **gross alpha-beta**, 18 samples are above decision limit (0.1–0.4 Bq/l). The decision limit is rather high for this purpose, compared to the classification concentrations. So in this case, a measurement point with a gross alpha-beta activity concentration above decision limit is related already to class 2. So for gross alpha-beta 17 samples were detected in class 2, one in class 3. This sample with the highest gross alpha-beta activity concentration (1.05 ± 0.40 Bq/l) is located in the lower Mühlviertel (Ried in der Riedmark, district Perg). The other measurement points with gross alpha-beta activity concentrations in class 2 are evenly spread among Upper Austria.

For ^{3}**H** 148 samples are above decision limit (0.7–0.95 Bq/l), but all in class 1.

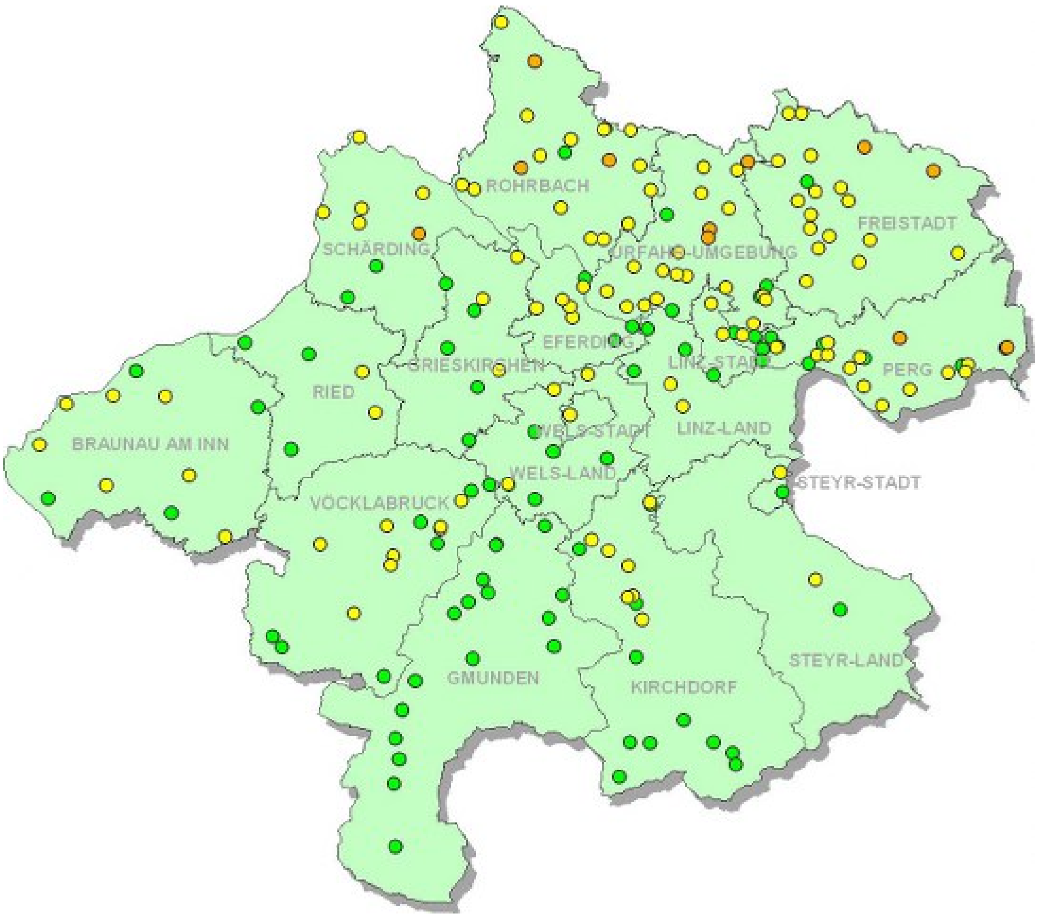

Figure 7.1: *Measurement points of the Survey sampling in Upper Austria classified for Radon results, legend see Annex A1*

For ^{226}**Ra** only 2 samples of 202 are above decision limit (0.030–0.045 Bq/l), one in class 2. This sample with the highest ^{226}Ra activity concentration (0.8 ± 0.4 Bq/l) is the same sample as the one with the highest gross alpha-beta activity concentration in the lower Mühlviertel. The second sample above decision limit is the sample with the highest radon activity concentration in the central Mühlviertel. The other sample in this village with highest radon activity concentration was unfortunately not analyzed for ^{226}Ra because of problems in the laboratory. So the correlation between high radon activity concentration and high ^{226}Ra activity concentration can not be verified. But this topic is discussed in Chapter 11.

185 of the 202 samples analyzed for ^{238}**U** were are above decision limit (0.12–0.62 mBq/l). No samples were detected in class 3 and 4, 15 samples are in class 2. These 15 samples are spread among Upper Austria (Figure 7.2). No correlation between higher radon activity concentrations and higher radon activity concentrations is noticeable in this context. None of these 15 samples with higher ^{238}U activity concentration shows a radon activity concentration of above 100 Bq/l. As mentioned above, this topic will be discussed in Chapter 15 and 11.

As mentioned in Chapter 3.1 in the "Guidelines for Drinking-water Quality, 3rd edition" (WHO, 2004) the **guidance level for uranium** is halved to 15 µg/l, which corresponds to a ^{238}U activity concentration of 0.19 Bq/l. Adopting this new level to the method of classification for this thesis (intersection between class 2 and 3: recommendation value, intersection between class 1 and 2: one tenth of this value, intersection between class 3 and 4: the decuple) yields slightly different results. One sample lies above 0.19 Bq/l in class 3, 36 samples in class 2.

These 36 samples are again distributed among Upper Austria, the highest measurement point (0.25 Bq/l) is located in the centre of Upper Austria (district Eferding). The 4 samples with the highest ^{238}U activity concentrations are all located in the central region of Upper Austria (districts Eferding, Wels-Land and Urfahr-Umgebung). This issue will be surveyed more detailed in Chapter 15 and was also detected by Gegner (2002), which is discussed in Chapter 3.2 and 8.

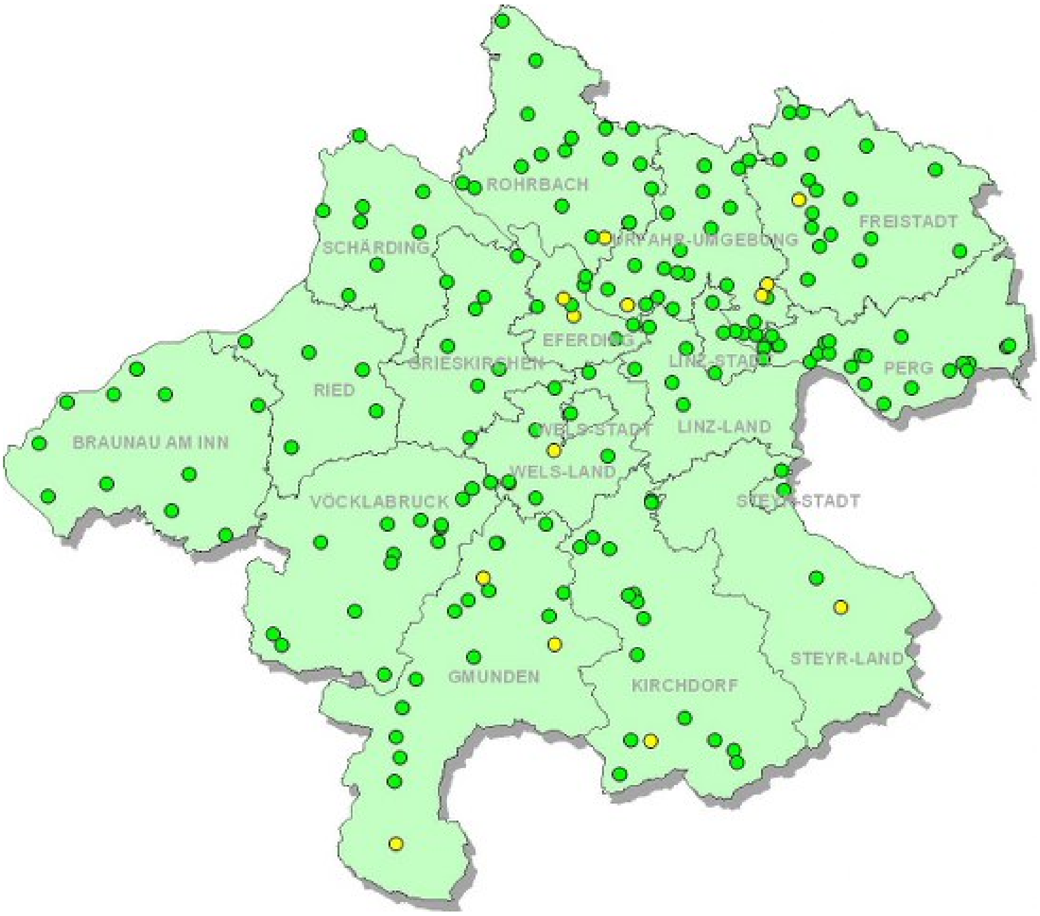

Figure 7.2: *Measurement points of the Survey sampling in Upper Austria classified for ^{238}U results, legend see Annex A1*

The WHO guidance levels for ^{238}U in drinking water are based on the chemo-toxic effect of uranium, not on the radioactive exposure. So it is debatable if it is reasonable to use the again halved WHO guidance level for radiation protection implementation. But the results show, that only one measured sample is above this very low WHO guidance level for ^{238}U activity concentration, and according to Figure 7.7 for about 99% of all drinking water samples in Upper Austria ^{238}U activity concentrations below 0.19 Bq/l are expected. (see below). So it currently seems that generally no high health risk is expected by ingestion of uranium in drinking water for the Upper Austrian population, even taking into account the chemo-toxic effect.

The measured activity concentrations of ^{222}Rn, ^{3}H and ^{238}U (above decision limit) are **log-normal distributed** within acceptable uncertainties (Figures 7.3-7.7). Log-normal distributions are very common in the statistical measurement of distributions of elements in geosphere (Ahrens, 1965) and of radon concentration in ground water (Zikovsky & Chah, 1990). Various studies approve the log-normal distribution of radon-activity in ground and drinking water (e.g. Rühle, 1996, Villalba et al., 2005, Zalewski et al., 2001). A survey of 236 drinking water samples in Quebec, Canada during a period of 24 years shows an approximately log-normal distribution of gross alpha activity (Zikovsky, 2006). A log-normal distribution of uranium in natural waters was also reported by the IAEA (1988).

The **cumulative frequency distribution** of ^{222}**Rn** activity concentrations of the survey sampling shows, that 95% of the samples are below 100 Bq/l. The median of the radon activity concentrations of the randomly distributed samples is about 13 Bq/l (Figure 7.3, Table 7.1), which correspond well with previous measurements (Friedmann, 1999, 2006) and Ditto et al. (1999), who analyzed about 1500 ground waters in Austria and cited a radon median of 12.0 Bq/l. Rühle (1996) reports an median of 1126 samples from total Germany of 5.6 Bq/l, which is much lower than the detected Austrian median.

According to Figure 7.3 for 99.9% of all drinking water samples radon activity concentrations below 600 Bq/l are expected. The "Commission Recommendation on the protection of the public against exposure to radon in drinking water" (European Commission, 2001) established that no remedial action should be required if the concentration is less than 100 Bq/l, and it is unlikely that water with a radon concentration exceeding 1000 Bq/l could be considered justifiable from the radiation protection point of view. In between national surveys might show that a higher reference level needs to be adopted for implementing a practical radon programme. It currently seems that 1000 Bq/l will not be achieved in Upper Austria, but 5% of the results of this survey sampling exceed 100 Bq/l. So it may be reasonable to adopt a higher reference level, especially in certain regions (e.g. Bohemian Massif) to avoid problems with authorities. This will be discussed in further chapters (15, 17).

Figure 7.4 shows the **frequency distribution** of the **radon** activity concentrations. The maximum frequency is detected at about 8 Bq/l.

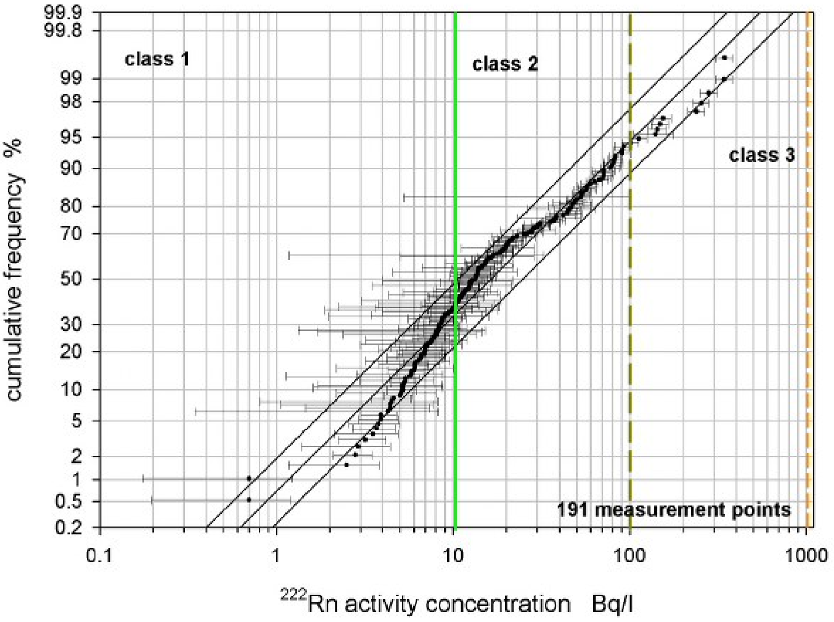

Figure 7.3: *Cumulative frequency distribution of the ^{222}Rn activity concentrations of the survey sampling*

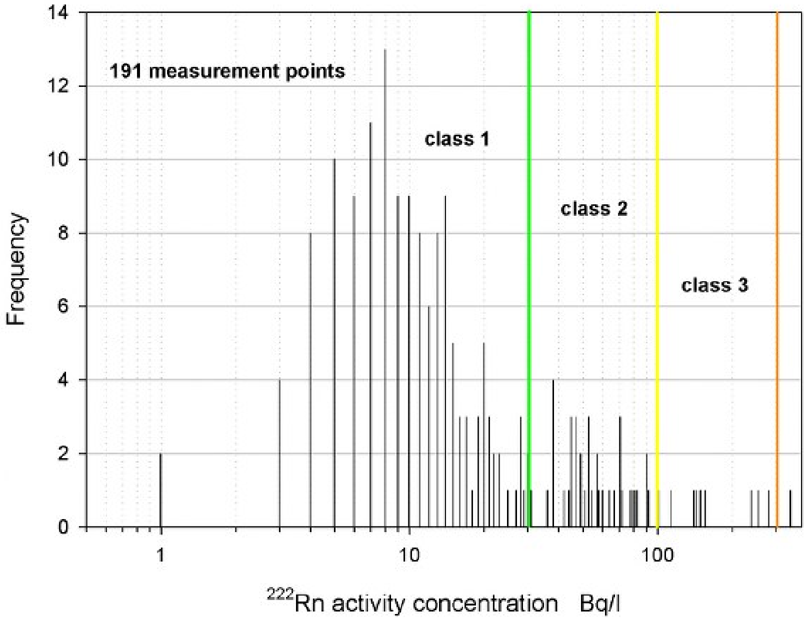

Figure 7.4: *Frequency distribution of the ^{222}Rn activity concentration of the survey sampling*

The **cumulative frequency distribution** of the ^{3}**H** activity concentrations of the survey sampling shows a median of about 1 Bq/l (Figure 7.5, Table 7.1). According to Figure 7.5 for 99.9% of all drinking water samples in Upper Austria tritium activity concentrations below 4 Bq/l are expected. The applied standards (European Commission, 1998, Republik Österreich, 2001) commit an indicative activity concentration for tritium of 100 Bq/l. So in this case no radiation protection problems are expected for the Upper Austrian population currently.

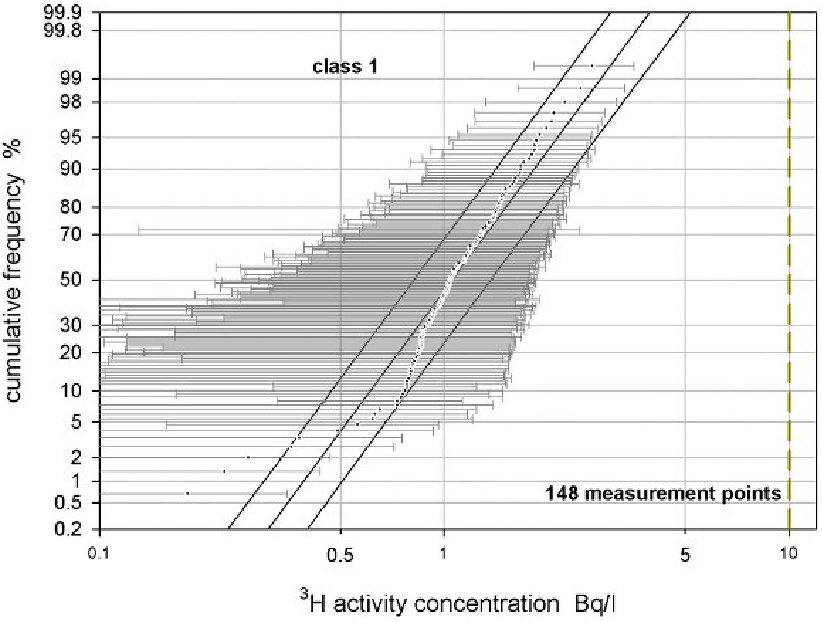

Figure 7.5: *Cumulative frequency distribution of ^{3}H activity concentrations of the survey sampling*

Figure 7.6 shows the **frequency distribution** of the **tritium** activity concentrations. The maximum frequency is detected at about 0.95 Bq/l.

It seems that the most analyzes drinking waters in Upper Austria are recent surface near waters with an exchange with the atmosphere (e.g. rain water) because of the ^{3}H activity concentration of about 1–2 Bq/l. The samples with ^{3}H activity concentration >DL may be deep waters with ages above 60 years (see Chapter 2.5.4).

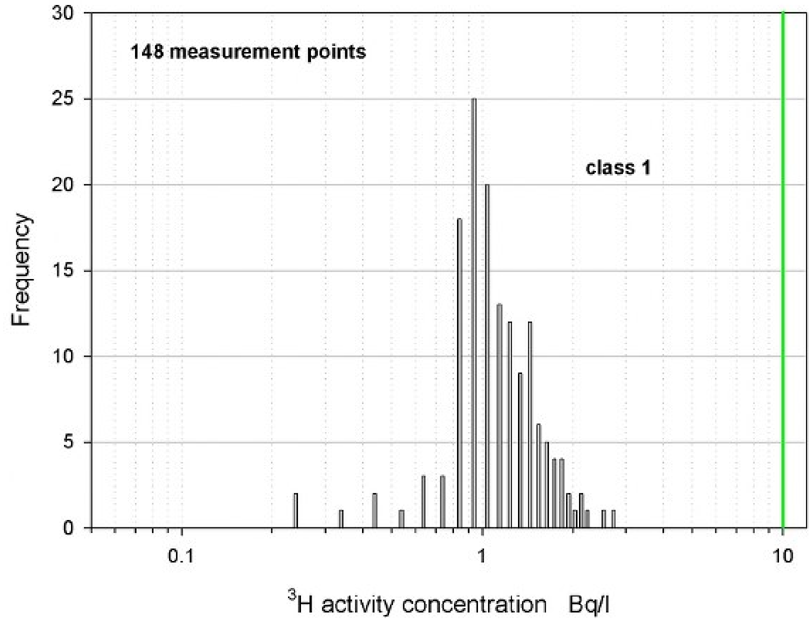

Figure 7.6: *Frequency distribution of the ^3H activity concentration of the survey sampling*

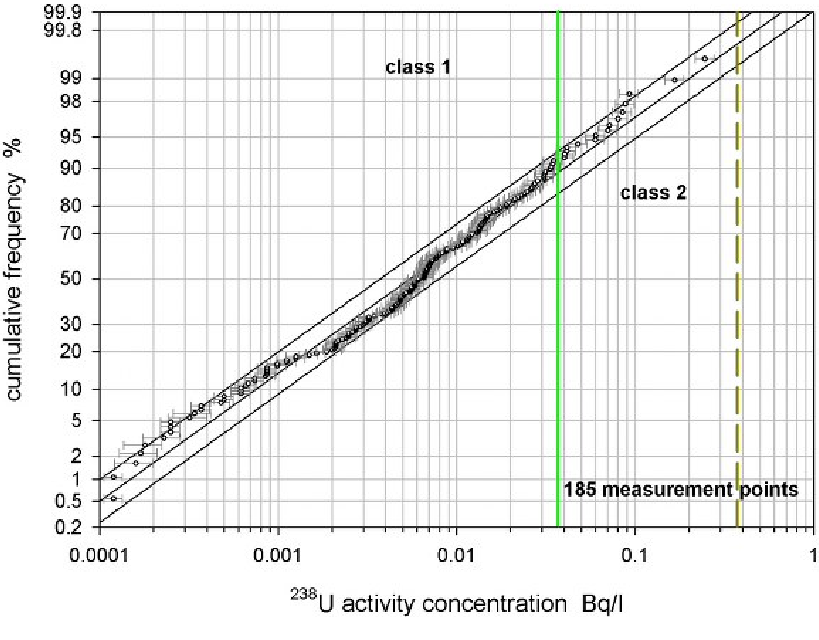

Figure 7.7: *Cumulative frequency distribution of ^{238}U activity concentrations of the survey sampling*

The **cumulative frequency distribution** of all measured ^{238}**U** activity concentrations of the survey sampling yields a **median** of about 6 mBq/l (Figure 7.7, Table 7.1). Figure 7.7 shows for 90% of all samples ^{238}U activity concentrations below

0.037 Bq/l (class 1). According to the distribution ^{238}U activity concentrations below 0.37 Bq/l are expected for about 99.5% of all drinking water samples in Upper Austria (guideline value in the "Guidelines for Drinking-water Quality, 2nd edition" of the World Health Organisation (WHO, 1993)). At least for about 99% of all drinking water samples in Upper Austria ^{238}U activity concentrations below 0.19 Bq/l are expected, which is the new recommended WHO guideline value in the "Guidelines for Drinking-water Quality, 3rd edition" (WHO, 2004).

Table 7.1: *Medium and maximum activity concentrations of the survey sampling for different radionuclides*

Radionuclide	**No. of measurements**	**Activity Concentration Median (Bq/l)**	**Activity Concentration Maximum (Bq/l)**
Gross alpha-beta	203	0.45	1.05
^{222}Rn	205	13	344
^{3}H	203	1.0	2.70
^{226}Ra	203	<DL (0.030–0.045)	0.08
^{238}U	203	0.006	0.25

Chapter 8

Results of the Detailed Sampling

All detailed results are listed in tables in Annex A3.

The **149 samples** taken in the detailed sampling phases were all analyzed for ^{222}Rn by Triathler but only partly for gross alpha, gross beta, ^{3}H, ^{226}Ra, ^{228}Ra, ^{210}Po, ^{210}Pb and ^{238}U because of laboratory and financial capacity reasons. The **analysis results** of **each individual nuclide** are discussed in the following chapter. Due to the different quantities of measurements per nuclide the number of measurements is listed in all figures. Additionally only results above desicion limits are included in the graphs, their number is also noted in the figures. Cumulative frequency distributions of activity concentrations are carried out only for the nuclides with more than 20 results above desicion limit. The classes are marked in all graphs for easier interpretation.

As mentioned before, activity concentrations in drinking water at taps in the consumers' houses are significant for the objective of radiation protection of the public. So in the graphs and in the interpretation activity concentrations **directly at the consumers' houses** are surveyed separately. 36 of the 149 samples were taken at sites where the water is directly available for the consumers (**tap water** from municipal water supplies, small water supplies or private wells).

Additionally to the graphs the measurement results are illustrated in **maps of Upper Austria** to give an overview and to survey for geographic correlations. Again the results are distinguished between consumers' houses and wells. Geological correlations are surveyed in Chapter 15.

The sampling locations are differentiated in **water units** and **private wells**. In all graphs and maps the sampling locations are made anonymous and referred to as **WVA** for water units and **H** for private wells and serially numbered. In all maps the water units are illustrated by **circles** and the private wells by **squares**. The results of the 16 samples, which were taken at public springs and wells in the investigated regions which are regarded as being pleasant or curative (see Chapter 5.1.2) are mapped and illustrated by a **triangle**. The results of these 16 samples are discussed additionally in a separate chapter (Chapter 9) (see legends in Annex A1).

Some samples were taken close-by others or at different sites in the same processing building. So in the illustrations in maps these points can be super-imposed. In this case the highest classified measurement point is shown.

8.1 Radon

As discussed in Chapter 5.1.2 Triathler measurements were carried out at all 149 measurement points of the detailed sampling with different cocktails and Triathlers (see Chapter 10.3). In each case the **maximum radon activity concentrations measured by Maxilight cocktails** are processed and used as ^{222}Rn results for discussion in the following chapters, because these results are verified well (Chapter 10.4). In a few cases no measurements by Maxilight cocktail were carried out, so the corresponding maximum Aqualight cocktail results are used.

In the framework of the nuclide specific analysis in the laboratory of the AGES Vienna ^{222}Rn was determined too, but the samplings for this analysis were not carried out especially for radon measurements and the sampling plastic containers were not fully radon tight. Thus, most of the ^{222}Rn activity concentrations measured in the AGES laboratory are clearly below the Triathler results (see Chapter 10.4) and are not further used in the following discussions. But these radon activity concentration results are processed for correction calculations for ^{210}Pb and ^{210}Po determination anyway (see Chapter 5.2.2).

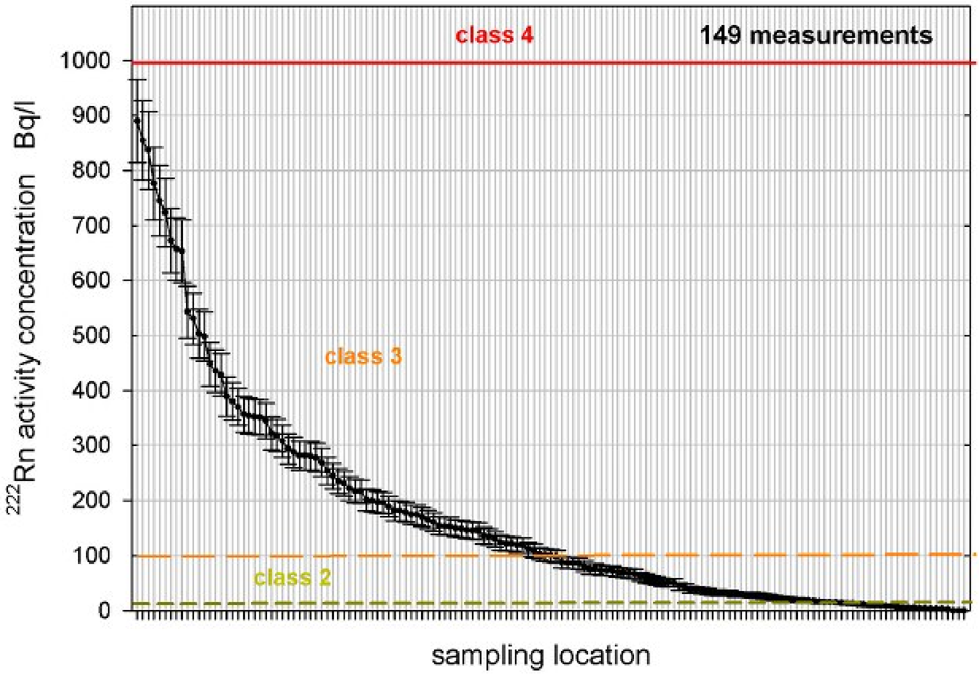

Figure 8.1: *Sorted ^{222}Rn activity concentrations of the detailed sampling*

From 149 measurement points, 146 ^{222}**Rn** activity concentrations are **above desicion limits** (about 1–2 Bq/l). No sample was detected with ^{222}Rn activity concentrations in class 4 (>1000 Bq/l), 74 samples are in class 3 (>100 Bq/l).The highest radon activity concentration (890 ± 76 Bq/l) was measured in a deep well in the lower Mühlviertel (WVA70, district Freistadt). The second and third highest activity concentrations (855 ± 73 Bq/l and 837 ± 71 Bq/l) were detected in the same village – in another deep well and in the reservoir. In Chapter 12 and in Chapter 15 a closer look will be spent to this results and region.

Figure 8.1 demonstrates the range of all measured ^{222}Rn activity concentrations in the detailed sampling and in Figure 8.2 these results are illustrated in a map of Upper Austria for geographic assignment.

All samples with radon activity concentrations in class 3 are located in the Mühlviertel (districts Perg, Urfahr Umgebung, Freistadt, Rohrbach) and in district Schärding (Danube fault). In southern Upper Austria, the alpine region, no higher ^{222}Rn activities were detected. This geographic distribution issue will be surveyed in Chapter 15.

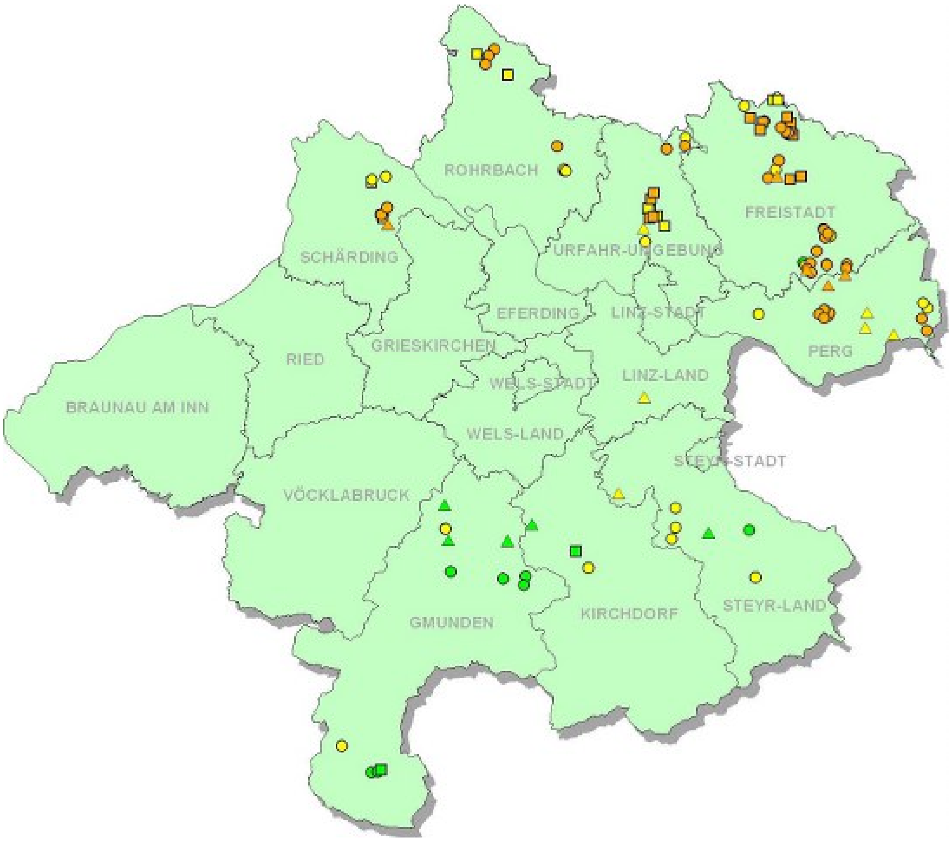

Figure 8.2: *All classified ^{222}Rn results of the detailed sampling demonstrated in a map of Upper Austria, legend see Annex A1*

In Figure 8.1 and 8.2 the results of all analyzed measurement points are demonstrated, not distinguishing between the different sampling locations (directly at wells, before and after water treatment, directly at the consumer). As mentioned above, for the radiation protection point of view for population exposure caused by radionuclides in drinking water mainly the activity concentration directly at the consumers' houses is of interest. The distribution of activity concentrations within the water flow from wells to the consumers are discussed in Chapter 12.

17 of the 36 water samples taken **directly at consumers' houses** have radon activity concentrations above 100 Bq/l (class 3). These sampling points are again located in the Mühlviertel and one in the Danube fault region (Figure 8.3). The highest radon activity concentration (745 ± 64 Bq/l) was detected in the **private well** in central Mühlviertel (district Urfahr Umgebung), which already showed the highest radon activity in the survey sampling (344 ± 38 Bq/l). The varying measurement results show supposedly the big radon losses by incautious sampling, transport and decanting (see Chapter 10.5). So it is likely that some ^{222}Rn results of the survey sampling are underestimated, although the median of the activity concentration distribution corresponds well with former measurements (Chapter 7). But maybe radon losses by incautious sampling, transport and decanting were problems in these measurements too. In Chapter 10.5 comparison and test measurements are carried out.

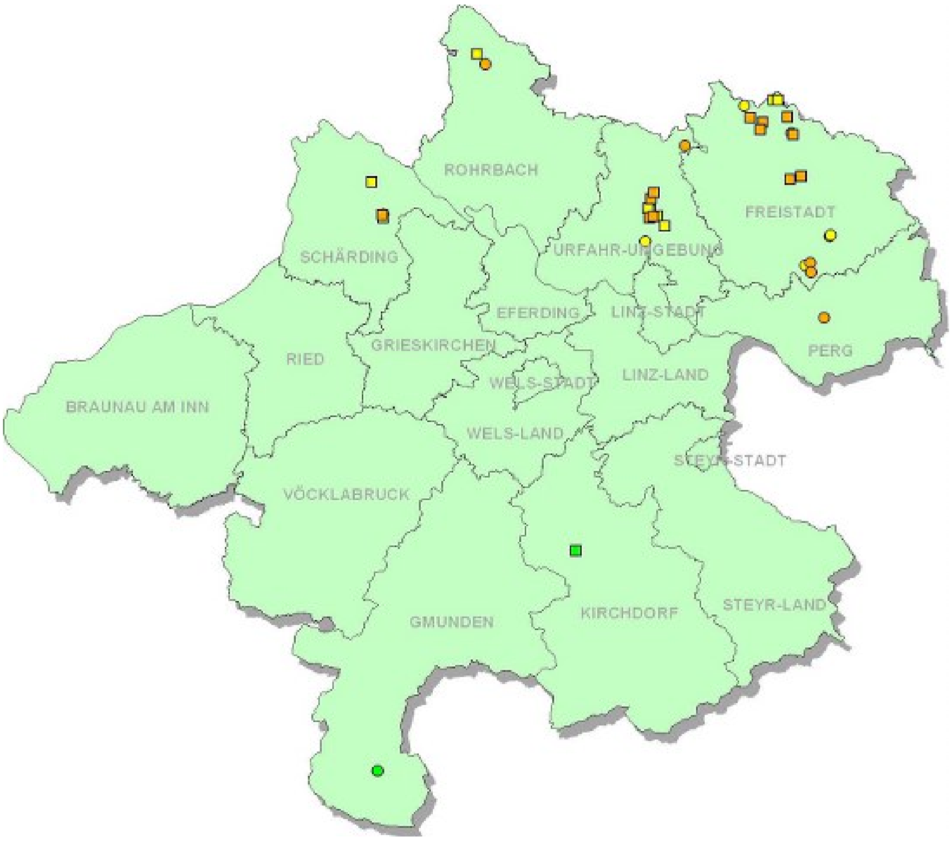

Figure 8.3: *Classified ^{222}Rn results taken directly at consumers' homes in a map of Upper Austria, legend see Annex A1*

At the measurement point (private well) with the second highest radon activity concentration within the survey sampling – which is located in the same village – the owner prohibited another sampling in the detailed sampling phase. So a sample was taken at the private well located next to this one, but the radon activity concentration measured there is only 38 ± 6 Bq/l. The reason might be, that in this region the **geology** is very **heterogeneous**, which is discussed in Chapter 15.12.

Figure 8.4 shows the range of the ^{222}Rn activity concentrations at **consumers' houses**. The figure demonstrates that most of the samples with higher radon activity concentrations were taken from private wells. WVA75 is a special case within a municipal water supply in the village in lower Mühlviertel with the highest measured radon activities and will be discussed in Chapter 12. It seems that for the purpose of radiation protection "problems" caused by radon activity concentration in drinking water will arise rather at **private wells**, where water is consumed directly, than at municipal water supplies. Similar results were also found in studies of other countries (e.g. Deflorin, 2004, Karpinska et al., 2002). Rühle (1996) reported the highest measured ^{222}Rn activity concentration in Germany in a deep well in the Fichtelgebirge with 1500 Bq/l, but the highest measured ^{222}Rn activity concentration at a private consumer was much lower with 550 Bq/l.

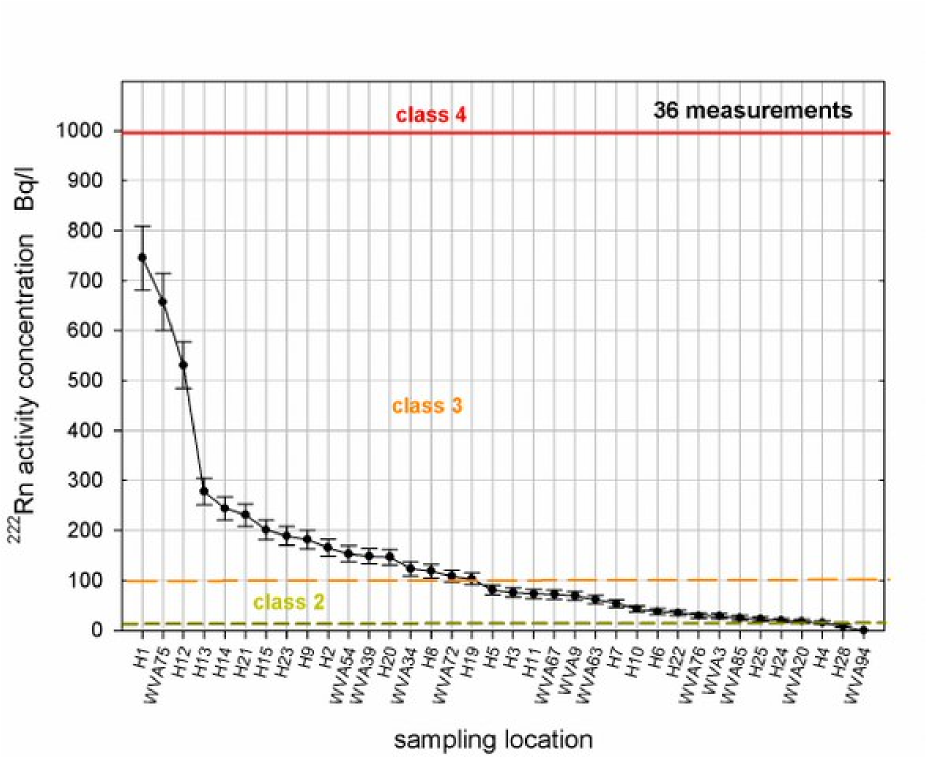

Figure 8.4: *Sorted ^{222}Rn activity concentrations directly at consumers' homes*

The **cumulative frequency distribution** of ^{222}Rn activity concentrations of the survey sampling, where the samples were randomly distributed, yields a median of about 13 Bq/l, which corresponds well with previous measurements (Chapter 7). The radon activity concentrations of the detailed sampling are consistently higher than the ones of the survey sampling. This results from immediate Triathler-measurements without radon losses on the one hand, and from not longer randomly distributed measurements points on the other. Sampling and measuring were just carried out in regions with elevated activity concentrations in the survey sampling and special geologic interests (see selection criteria in Chapter 5.1.2).

The cumulative frequency distribution of the ^{222}Rn activity concentrations of the detailed sampling results in a clearly higher median of 100 Bq/l (Figure 8.5). The radon activity concentrations of the detailed sampling are **not longer simply log-normal distributed**. The lower activity concentrations (up to 150 Bq/l) are log-normal distributed within small uncertainties, but at higher activity concentrations (from 200 Bq/l) the distribution curve rises strong. So it is possible that the radon activity concentrations results derive from **two distributions**, one for the lower and one for the higher activity concentrations. In Figure 8.5 the possible second distribution is illustrated in blue. An extrapolation of the first cumulative frequency distribution (black) results in an expectation of radon activity concentration above 1000 Bq/l for 10% of all drinking waters in Upper Austria. With the second cumulative frequency distribution only for 1% of the Upper Austrian drinking waters radon activity concentrations above 1000 Bq/l are expected. So with only an extrapolation of cumulative frequency of low activity concentration, the "potential radon problem" in drinking water may be overestimated.

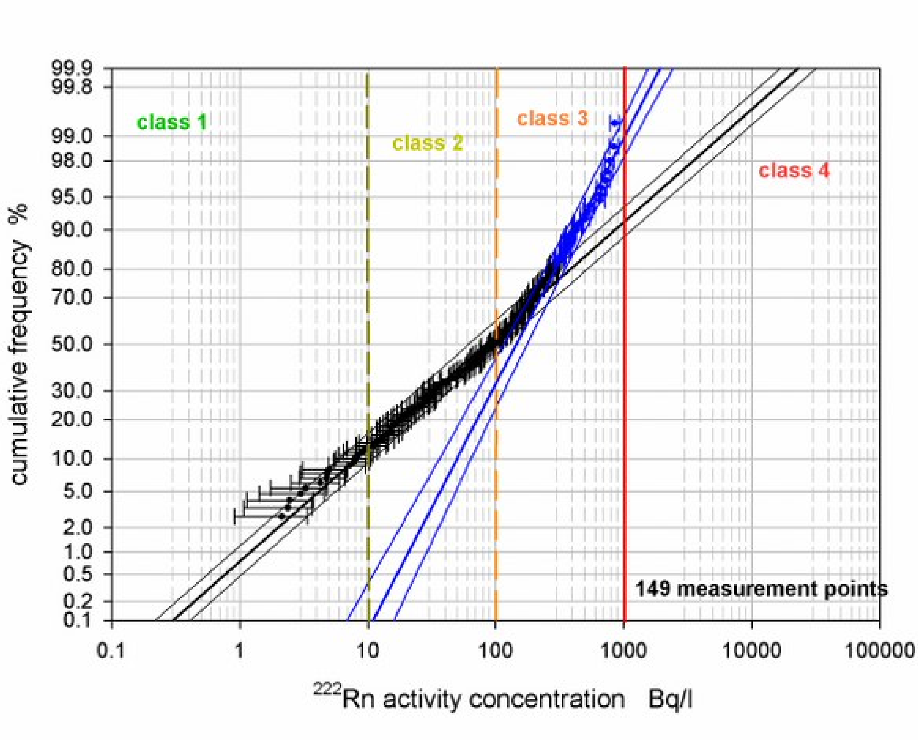

Figure 8.5: *Cumulative frequency distribution of ^{222}Rn activity concentrations of the detailed sampling*

The most likely reason for the not simple log-normal distributed cumulative frequency distribution are the not randomly distributed measurement points and the fact, that measurements were done at different sites within water flows. So the results are not fully comparable.

So, also **cumulative frequency distributions** are graphed for radon activity concentrations only at **consumers' homes** (Figure 8.6) and **directly at wells**, without water treatment (Figure 8.7). Some results of private wells are used for both distributions, when no losses between well and water tap are expected. Figure 8.6 shows a log-normal distribution of the radon activity concentrations with a slightly higher uncertainty because of few measurement points. The median of the distribution is about 55 Bq/l and 99.9% of all Upper Austrian tap waters are expected to be below 1000 Bq/l. Figure 8.7 shows a median of about 60 Bq/l and a similar rise in the curve at higher activity concentrations. So in spite of better comparability of the results, the rise appears anyway. So again the not randomly distributed measurement points among Upper Austria are the most reliable explanation. Possibly also an effect like a ”radon-saturation in water” is supposable, for water is not able to take up radon endlessly. Therefore more survey would be necessary, because no literature exists on this topic so far.

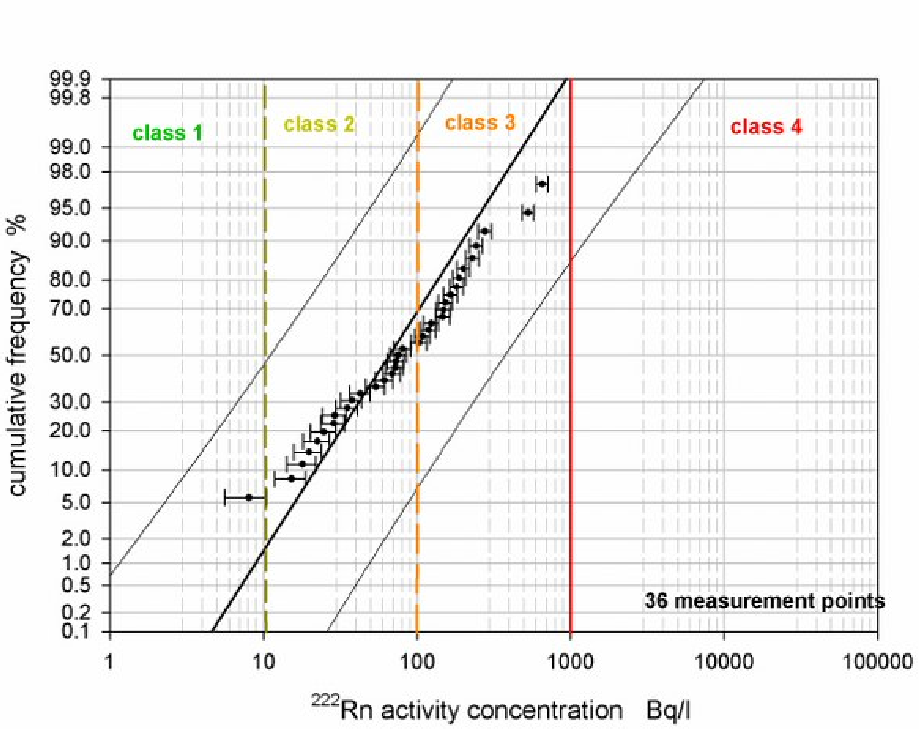

Figure 8.6: *Cumulative frequency distribution of ^{222}Rn activity concentrations at consumers' homes*

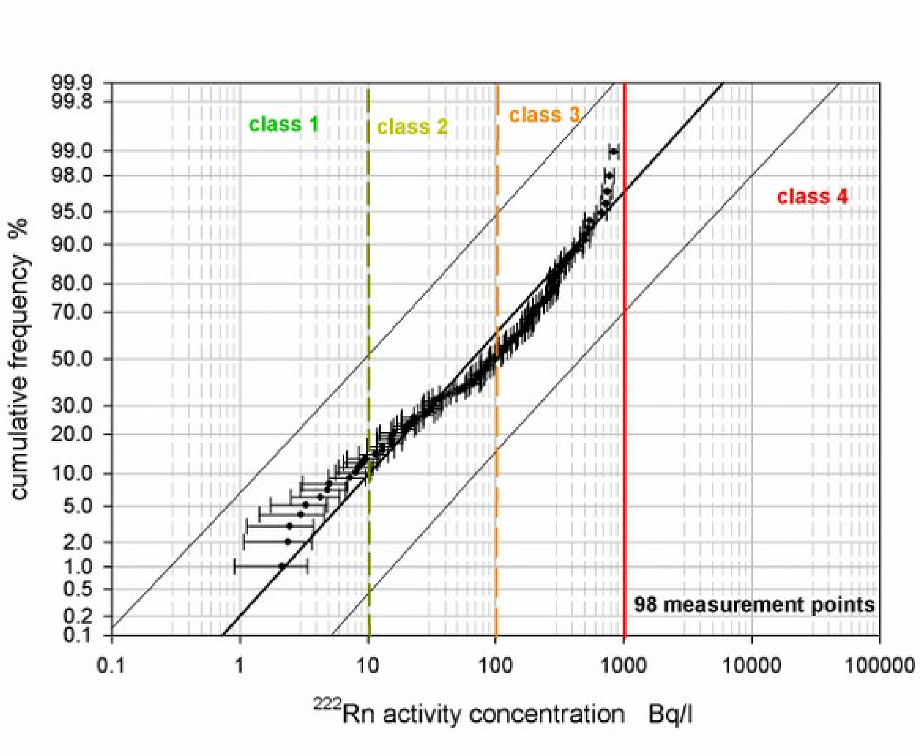

Figure 8.7: *Cumulative frequency distribution of* ^{222}*Rn activity concentrations at springs and wells*

8.2 Gross Alpha

129 samples were analyzed for gross alpha activity concentration in the laboratory of the AGES Vienna. **27** of these samples are **above desicion limit**, which varies from 0.073 to 0.17 Bq/l. The desicion limit is rather high for this purpose, compared to the classification concentrations (like in the gross alpha-beta analysis of the survey sampling, Chapter 7). A measurement point with a gross alpha activity concentration above decision limit is already related to class 2, mostly already to class 3. So for gross alpha 6 samples are in class 2, 20 in class 3 and one in class 4. In this case the measurement method is not ideal for the problem, because the desicion limits are too high and much more samples could be detected in class 2 or 3 with improved desicion limit. But gross alpha, gross beta or gross alpha-beta measurements are usually used for **screening methods** for general survey to decide if further nuclide specific measurements are necessary and therefore they should be simple and fast, even if the desicion limit increases thereby. The council Directive 98/83/EC (European Commission, 1998) points out, that if gross alpha and gross beta activities are less than 0.1 Bq/l and 1.0 Bq/l respectively, it can be assumed that the TID is less than 0.1 mSv/a. If the gross alpha and gross beta activity exceeds 0.1 Bq/l or 1.0 Bq/l, analysis for radionuclide specific radionuclides are required.

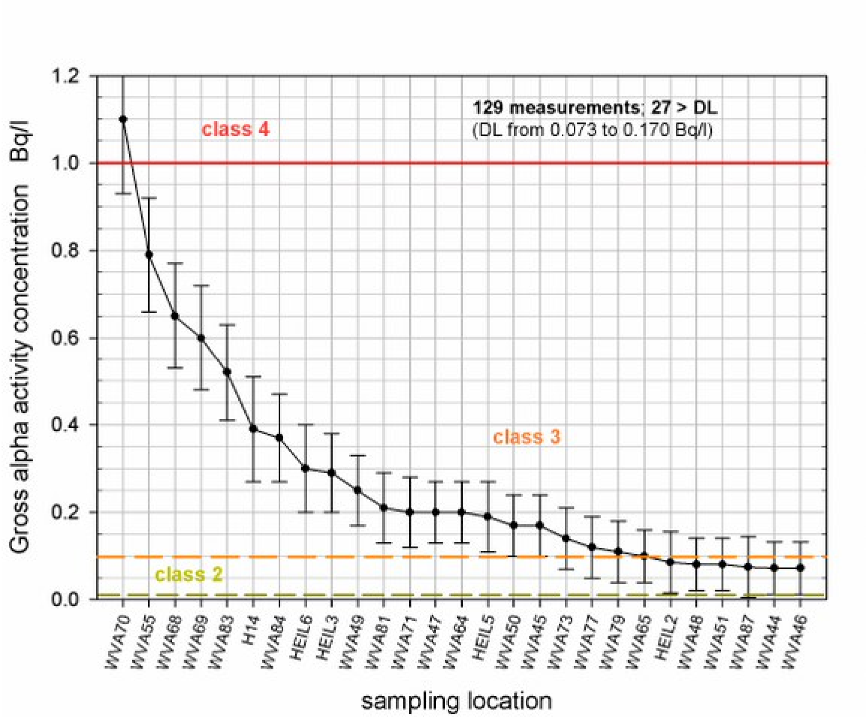

Figure 8.8: *Sorted gross alpha activity concentrations above desicion limit of the detailed sampling*

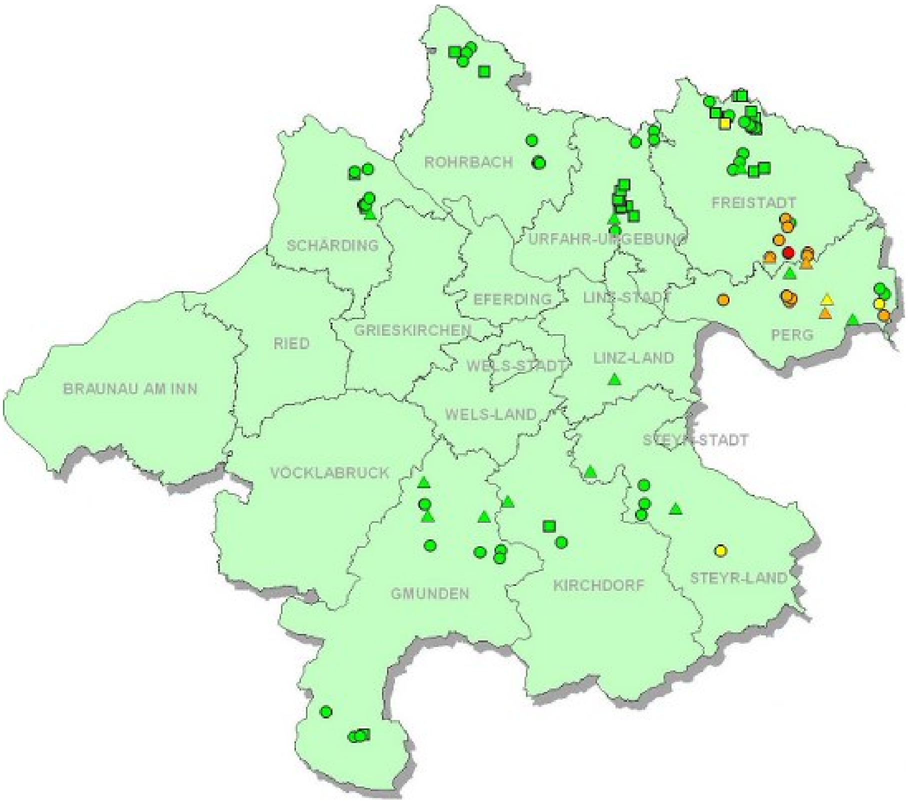

Figure 8.9: *All classified gross alpha results of the detailed sampling demonstrated in a map of Upper Austria, legend see Annex A1*

The sample in class 4 with the highest gross alpha activity concentration (1.10 ± 0.17 Bq/l) is the same as the one with the highest radon activity concentration – a deep well in the lower Mühlviertel (WVA70, district Freistadt) (Figure 8.8). The third and the fourth highest are located in the same village, the second highest in the neighbouring village. Generally all samples with gross alpha activity concentrations above 0.1 Bq/l are located in the lower Mühlviertel (Figure 8.9). Only one sampling point in class 3 was taken directly at a consumer's home (H14) (Figure 8.8 and 8.9).

The measured gross alpha activity concentrations in Upper Austria are **log-normal distributed** (Figure 8.10) with a **median** of about 0.2 Bq/l. Due to the mentioned decision limit problem and displaying only values above decision limit, only 30% of all measurements are below the reference limit of 0.1 Bq/l and for 2% of all Upper Austrian drinking waters gross alpha activity concentrations of more than 1 Bq/l are expected. Zikovsky (2006) analyzed 236 drinking water samples for gross alpha activity concentration in Quebec, Canada and found a much lower median of 0.052 Bq/l and a mean of 0.211 Bq/l. The mean value was calculated only with values above decision limit, but the median was determined using all values. The mean value of the samples above decision limit in Upper Austria is similar with 0.29 Bq/l, the median with all values is 0.12 Bq/l. So it seems that the gross alpha activity concentration is a little higher in Upper Austria than in Canada.

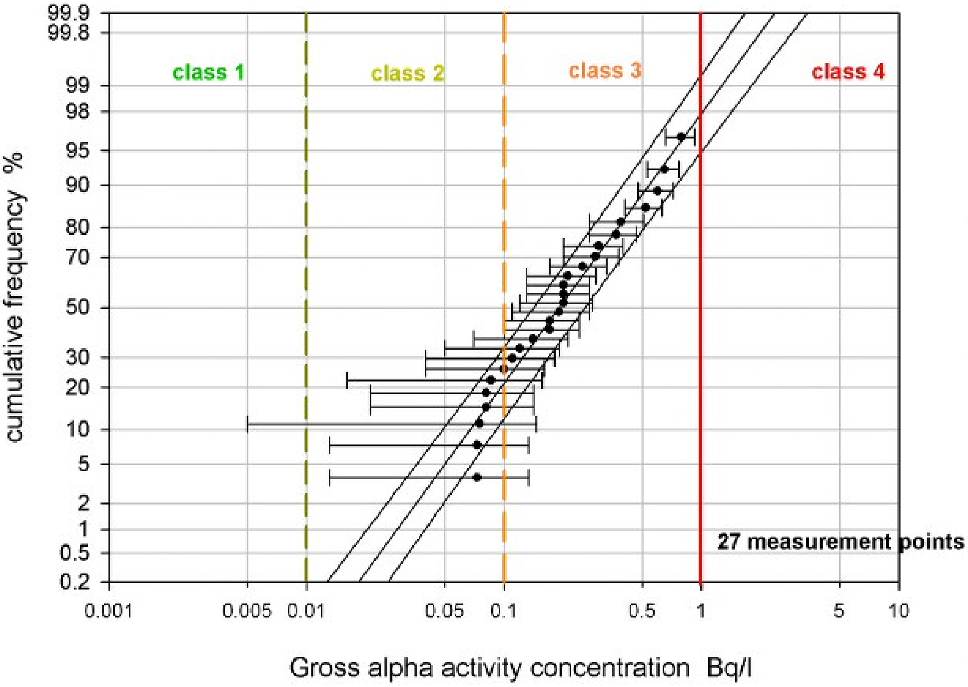

Figure 8.10: *Cumulative frequency distribution of gross alpha activity concentrations of the detailed sampling*

There are several studies about gross alpha activity concentration in drinking water in different countries (e.g. Portugal (Lopes et al., 2006), Italy (Forte et al., 2007)) which shows mean values in the same order like Canada (see Zikovsky, 2006). Higher gross alpha activity concentration were found in drilled wells in Finland (mean 0.36 Bq/l), although in dug wells and springs in the same regions the activity concentrations are much lower (mean 0.06 Bq/l) (Salonen, 1988). This fact also seems to be reflected in the results of Upper Austrian drinking waters, because as mentioned above the point with the highest gross alpha activity concentration is a drilled deep well. This topic will be discussed more detailed in Chapter 15.11.

8.3 Gross Beta

129 samples were analyzed for gross beta activity concentration in the laboratory of the AGES Vienna. **30** of these samples are **above decision limit** of 0.54 to 0.91 Bq/l. As discussed before for gross alpha, a sample above decision limit is already related to class 2. No measurement point was detected with a gross beta activity concentration above 10 Bq/l (class 4), 16 samples are in class 3 (above 1 Bq/l) and 14 in class 2 (Figure 8.11). The measurement point with the highest gross beta activity concentration (3.50 ± 0.56 Bq/l) is again the deep well in the lower Mühlviertel (WVA70, district Freistadt) with the highest radon and gross alpha activity too. Almost all other measurement points in class 3 are also located in the lower Mühlviertel (district Freistadt), one in the upper Mühlviertel (district Rohrbach) and one public well ("sacred well" – see Chapter 9) in the district Steyr-Land in the foothills of the Alps (Figure 8.12).

10 samples of the 30 samples above decision limit were taken **directly at consumers' homes**, 7 are in class 3, 3 in class 2. These samples are all private wells (see Figure 8.11 – identified with H) in the lower and middle Mühlviertel (Figure 8.12 – identified by a square). Only one sample above decision limit taken directly at consumers' homes comes from a small water supply, located in the upper Mühlviertel (WVA34) (Figure 8.11 and 8.12).

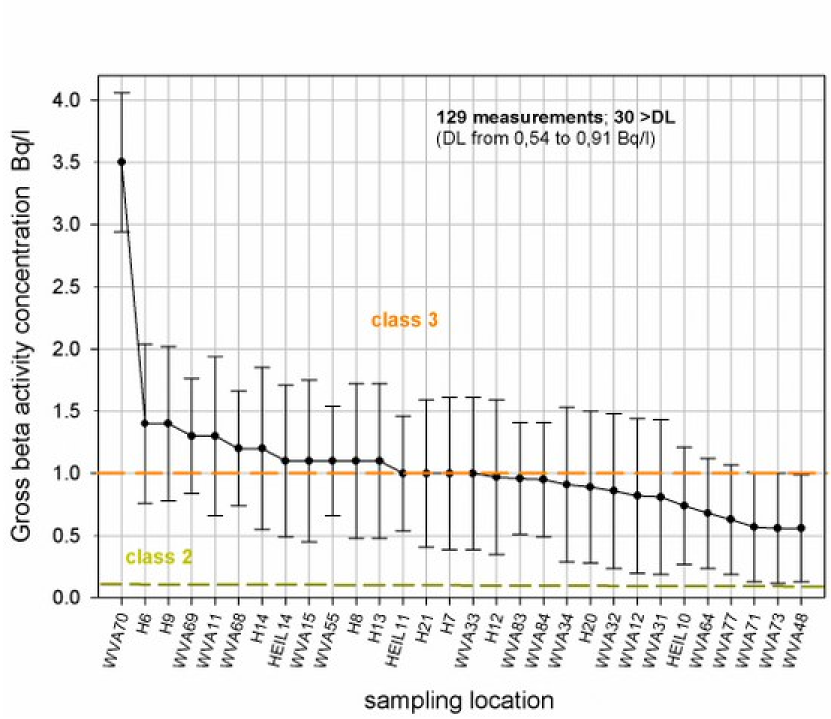

Figure 8.11: *Sorted gross beta activity concentrations above decision limit of the detailed sampling*

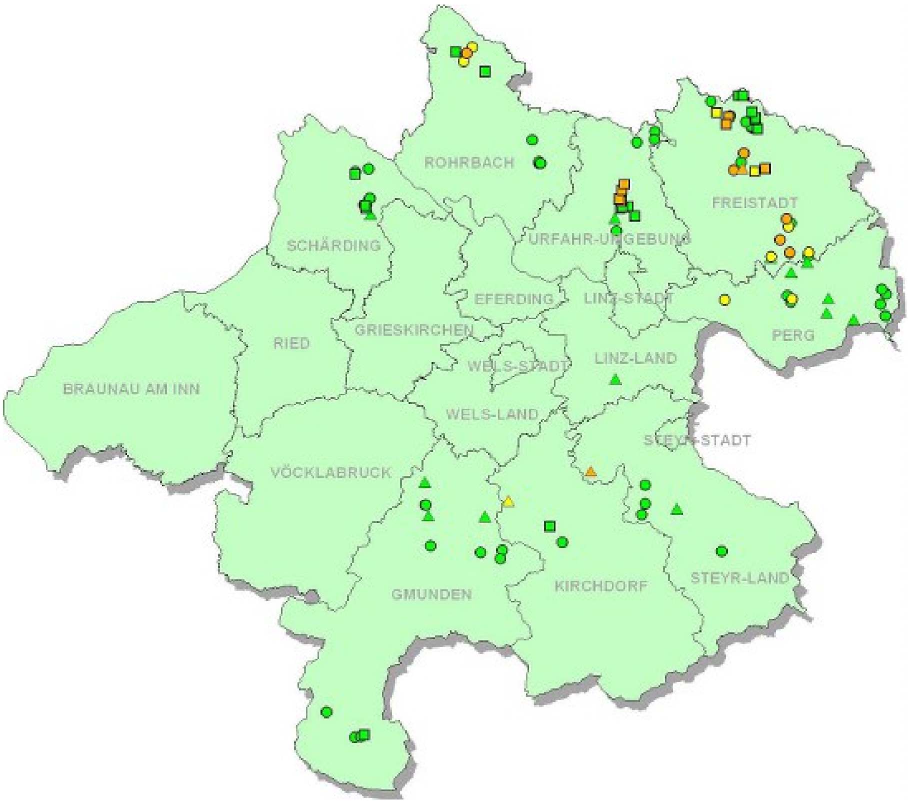

Figure 8.12: *All classified gross beta results of the detailed sampling demonstrated in a map of Upper Austria, legend see Annex A1*

The measured gross beta activity concentrations in Upper Austria are **log-normal distributed** (Figure 8.13) with a **median** of about 1 Bq/l. 50% of all measurements above decision limit are **above the reference limit** of 1 Bq/l and for about 0.5% of all Upper Austrian drinking waters gross beta activity concentrations of more than 2 Bq/l are expected. The median calculated with all gross beta activity concentrations (including samples below decision limit) is only a bit lower with 0.78 Bq/l.

Lopes et al. (2006) detected gross beta in 50 Portuguese drinking waters and did not detect gross beta activity concentrations above the reference level 1 Bq/l. Forte et al. (2007) surveyed 34 drinking water samples in Lombardia, Italy and also did not detect gross beta activity concentrations above the reference level 1 Bq/l. It seems that the gross beta activity concentration in drinking water in Upper Austria is slightly enhanced compared to the average.

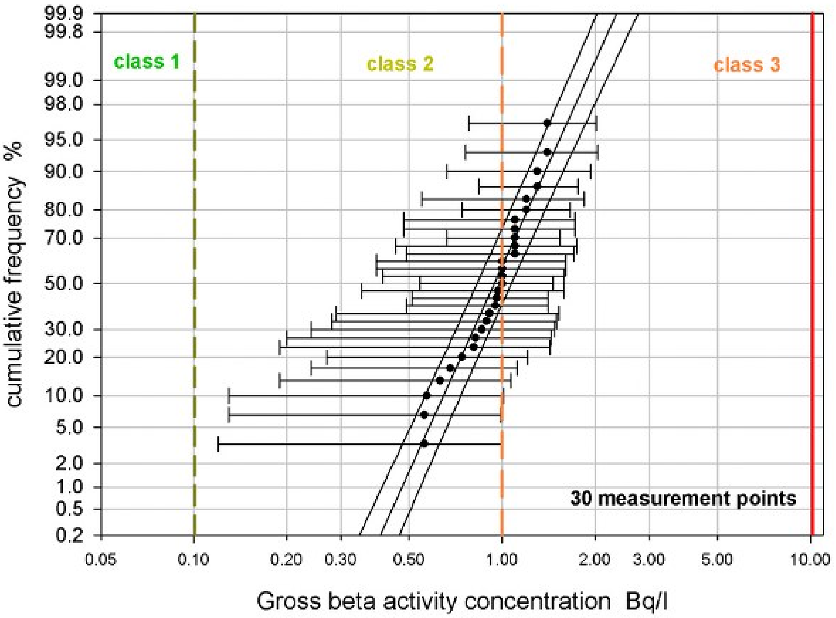

Figure 8.13: *Cumulative frequency distribution of gross beta activity concentrations of the detailed sampling*

8.4 Tritium

129 samples were analyzed for ^{3}H activity concentration in the laboratory of the AGES Vienna. **59** of these samples are **above decision limit** of 0.88 to 1.4 Bq/l. All samples above decision limit are in class 1 (<10 Bq/l) and range between 1.2 and 3.7 Bq/l. Most of the samples above decision limit are located in the lower Mühlviertel, none in the southern region of the Alps. But here again the decision limit and the measured activity concentrations are close together. So no generaliz-

ing regional correlations can be made, because the decision limits are not equal in the different sampling phases and were higher in the measurement session of the southern samples. The highest tritium activity concentration (3.7 ± 1.1 Bq/l) was detected in a deep well in the lower Mühlviertel. 23 of the 59 samples with tritium activity concentrations above decision limit were taken **directly at consumers' homes** (private wells or tap water from water supplies). It seems that there is no big difference between tritium at wells and tritium at water taps as it was observed for radon.

The measured tritium activity concentrations are **log-normal distributed** and show a **median** of about 2 Bq/l (Figure 8.14). The cumulative frequency distribution of the ^{3}H activity concentrations of the survey sampling showed a median of about 1 Bq/l (see Figure 7.5). So within the mentioned different decision limits and measurement uncertainties for ^{3}H there is no real difference between the survey and the detailed sampling. Regarding tritium the selection of measurement points in interesting regions had no affect as it was observed for ^{222}Rn.

It seems that the tritium activity concentrations in Upper Austrian drinking waters are expected to be low according to the applied standards (European Commission, 1998, Republik Österreich, 2001) and **no radiation protection problems are expected** for the Upper Austrian population currently.

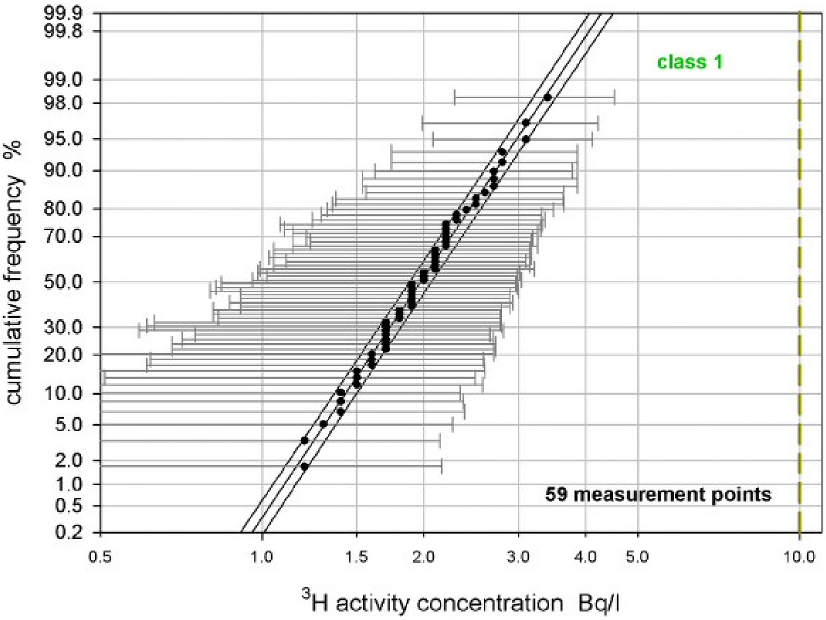

Figure 8.14: *Cumulative frequency distribution of tritium activity concentrations of the detailed sampling*

8.5 Radium-226

129 samples were analyzed for ^{226}Ra activity concentration in the laboratory of the AGES Vienna. **7** of these samples are **above decision limit** of 0.037 to 0.055 Bq/l. Again the decision limit is quite high, so a sample above decision limit can be related already to class 2. 5 samples above decision limit are in class 2 (0.05–0.5 Bq/l), no sample in higher classes (Figure 8.15). The 5 samples with ^{226}Ra activity concentration in class 2 are distributed among Upper Austria. The highest ^{226}Ra activity concentration (0.15 ± 0.04 Bq/l) was detected in a deep well in lower Mühlviertel. The second highest is the private well (H1) in central Mühlviertel which had the highest radon activity concentration and the second highest ^{226}Ra activity concentration in the survey sampling (Figure 8.15). The other 3 samples in class 2 are located in southern Upper Austria. But these samples in southern Upper Austria were all measured within the same sampling phase and the decision limits were a little lower in this session because of counting statistics. So the conclusion that high Radium-226 activity concentrations are expected in southern Upper Austria is not approvable. But it seems that samples with ^{226}Ra are more equally distributed among Upper Austria than radon, and radon and ^{226}Ra is not clearly connected. This theory will be discussed in Chapter 15 and 11.

Because of only 7 ^{226}Ra results above decision limit, no reasonable cumulative frequency distribution can be graphed.

In former studies ^{226}Ra concentrations were measured in drinking water samples for example by Irlweck et al. (1997) in 40 water samples of Austria. The analyzed activity concentrations were all below 5 mBq/l, which corresponds with the rather low activity concentrations in this study. Only the few measurement points with activity concentrations above decision limit are clearly higher, especially in the deep well in the lower Mühlviertel.

Wallner & Steininger (2002) also analyzed ^{226}Ra in 65 drinking water samples of different regions of Austria. The highest ^{226}Ra activity concentration was detected in lower Austria with 111 mBq/l, all others are in the range $<$ 0.3 to 48 mBq/l. These **results** also **correspond very well** with this study, which also showed single samples with clearly higher ^{226}Ra activity concentrations, but apart from these rather low concentrations.

Rühle (1996) reported a ^{226}Ra median of 1665 samples of 4.4mBq/l in Germany, with highest detected activity concentrations of 180 and 260 mBq/l in the Erzgebirge, Germany. The results are very similar with the ones detected in Austria and especially in this thesis.

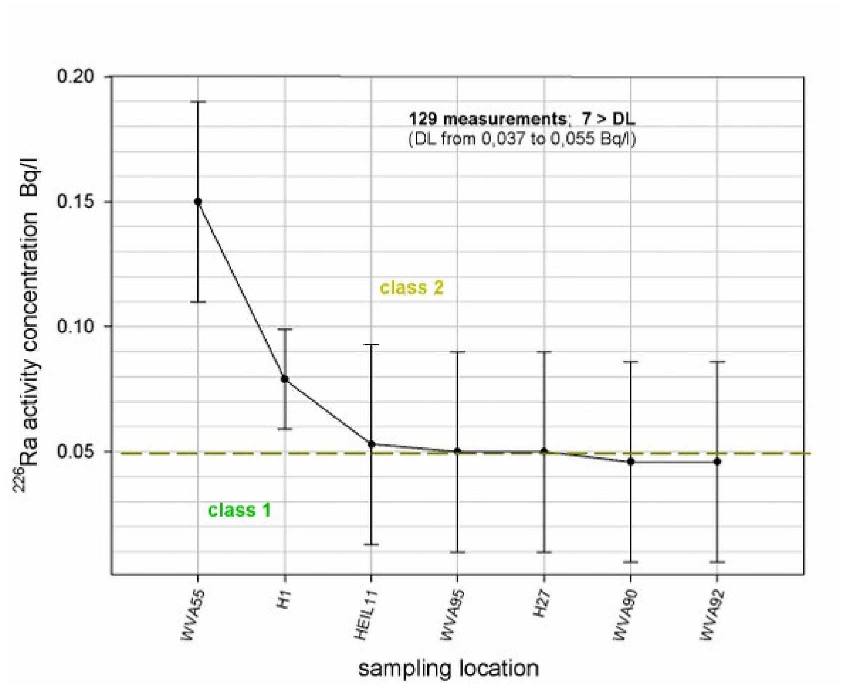

Figure 8.15: *Sorted ^{226}Ra activity concentrations above decision limit of the detailed sampling*

8.6 Radium-228

83 samples were analyzed for ^{228}Ra activity concentration in the laboratory of the AGES Vienna. **23** of these samples are **above decision limit** of 0.005 to 0.007 Bq/l. Only one sample is in class 2, a private well in lower Mühlviertel (district Freistadt) with a ^{228}Ra activity concentration of 0.022 ± 0.006 Bq/l.

The samples with ^{228}Ra activity concentrations above decision limit are distributed among Upper Austria and no direct correlation to the ^{226}Ra results is detectable (see Chapter 11). 7 of the 23 samples with ^{228}Ra activity concentrations above decision limit were taken **directly at consumers' homes**. 6 are private wells (H) in the different regions of the Mühlviertel, one from a water supply in the lower Mühlviertel (WVA3).

The measured ^{228}Ra activity concentrations are **log-normal distributed** and show a **median** of about 0.009 Bq/l (9 mBq/l) (Figure 8.16). According to the cumulative frequency distribution for 99% of Upper Austrian drinking waters ^{228}Ra activity concentrations below 0.02 Bq/l (class 1) are expected.

Wallner & Steininger (2002) analyzed ^{228}Ra in 24 drinking waters in Austria and detected no activity concentration above decision limit (up to 40 mBq/l), which corresponds well with the results of this study with a highest detected ^{228}Ra activity concentration of 22 mBq/l.

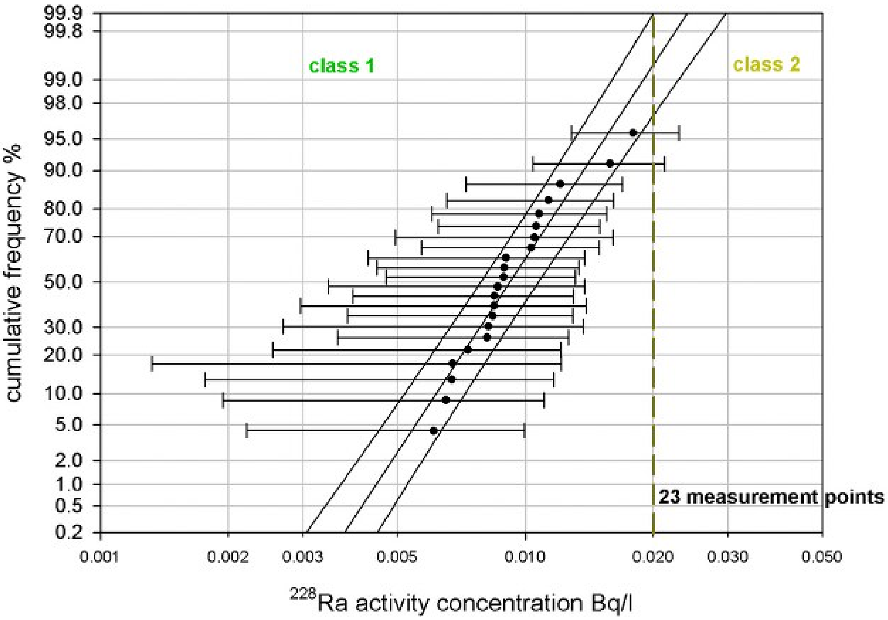

Figure 8.16: *Cumulative frequency distribution of* ^{228}*Ra activity concentrations of the detailed sampling*

8.7 Lead-210 and Polonium-210

As discussed in Chapter 5.2.2 ^{210}Pb and ^{210}Po in the drinking water samples were analyzed in the laboratory of the AGES Vienna by LSC and correction calculations. In the sampling container ^{222}Rn is decaying with a half life of 3.82d and ^{210}Pb is built in the drinking water sample. To determine ^{210}Pb and ^{210}Po this effect has to be corrected. For this correction ^{222}Rn was analyzed in every sample, and although – as described above – these measured radon activity concentrations are much lower than the on-site determined radon activities by Triathler, these values are used for the correction calculations. The measured radon activity concentrations in the same water sample correlate better with the new built ^{210}Pb in it. It was not always possible to analyze the samples immediately, so the correction calculations sometimes results in negative values. These negative values are set to below decision limit for further determinations.

The following calculated ^{210}Pb and ^{210}Po activity concentrations are afflicted with a generally high uncertainty because of the corrections and problems mentioned. The results are possibly too high and maybe overestimate the true values because of the corrections and they should act as estimation. Further measurements should verify these results. For direct determination of ^{210}Pb and ^{210}Po without correction calculations the drinking water samples should be evaporated immediately after sampling to avoid ^{210}Pb building up out of ^{222}Rn. With this method no correction calculations are necessary and the uncertainties would be reduced. Such measurements would be fundamental for exact determination of ^{210}Pb and ^{210}Po and for verification. But with this method, not all radionuclides can be analyzed out of one sample as it was carried out in this thesis. So for this thesis the used method was adequate, because of costs and time management. Nevertheless it would be interesting in the future to carry out ^{210}Pb and ^{210}Po measurements with immediate evaporated samples, because the results in this chapter and in Chapter 16 show, that these nuclides should be considered.

107 samples were analyzed for ^{210}**Pb** activity concentration in the laboratory of the AGES Vienna. **52** of these samples are **above decision limit** of 0.003 to 0.008 Bq/l. 20 samples above decision limit are in class 2, one in class 3. The sample in class 3 with a ^{210}Pb activity concentration of 0.46 ± 0.05 Bq/l is the deep well in lower Mühlviertel (WVA55), which also showed the highest ^{226}Ra activity concentration. The second highest sample (0.10 ± 0.02 Bq/l) is the private well in central Mühlviertel (H1) which showed the highest radon concentration and the second highest ^{226}Ra activity concentration in the survey sampling and the second highest ^{226}Ra activity concentration in the detailed sampling. One sample of the 20 samples in class 2 is located in the Innviertel (Danube fault), all others are located in the different regions of the Mühlviertel (Figure 8.17). 10 of the 20 samples in class 2 were taken **directly at consumers' homes** – all private wells (H, squares in Figure 8.17).

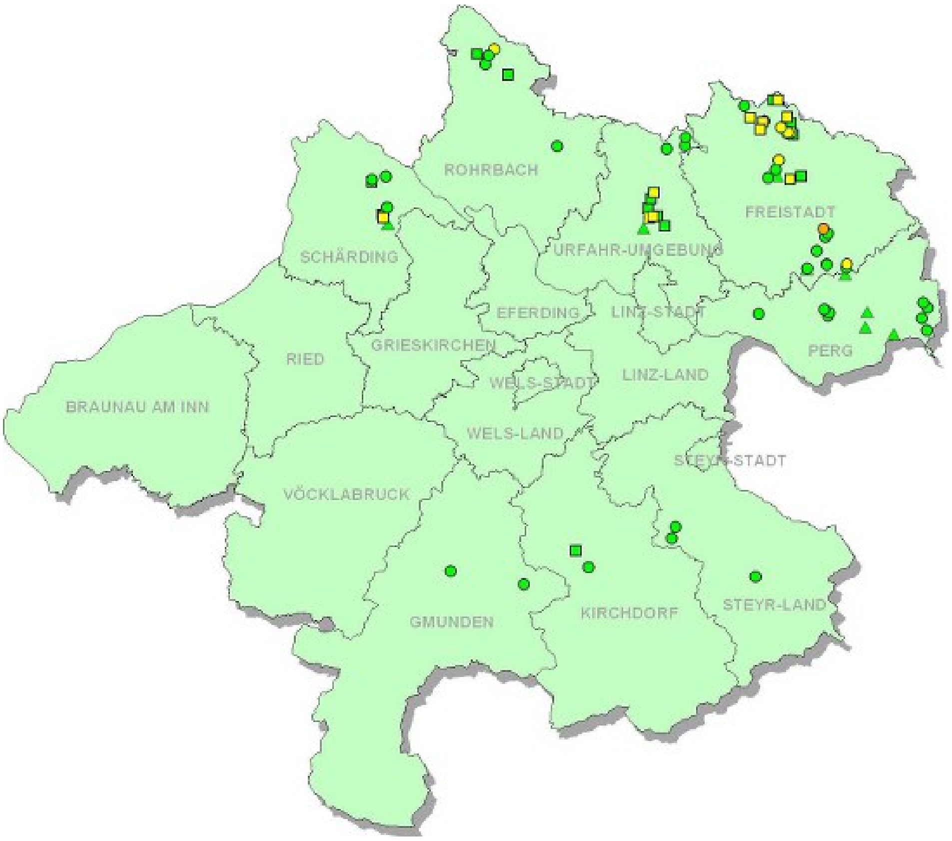

Figure 8.17: *All classified ^{210}Pb results of the detailed sampling demonstrated in a map of Upper Austria, legend see Annex A1*

The ^{210}Pb activity concentrations are **log-normal distributed** and show a **median** of about 0.010 Bq/l (10 mBq/l) (Figure 8.18). According to the cumulative frequency distribution for 99.8% of Upper Austrian drinking waters ^{210}Pb activity concentrations below 0.2 Bq/l are expected, which corresponds with an effective dose below 0.1 mSv/a.

^{210}Pb activity concentrations in drinking waters in Austria were analyzed by Feigl-Heihs (1998) in 40 water samples (mainly tap water) from different regions of Austria. The detected ^{210}Pb activity concentrations range from 1.5 to 102 mBq/l, which corresponds well with the detected ^{210}Pb activity concentrations in this study. Only the deep well in the lower Mühlviertel shows four times higher ^{210}Pb activity concentration.

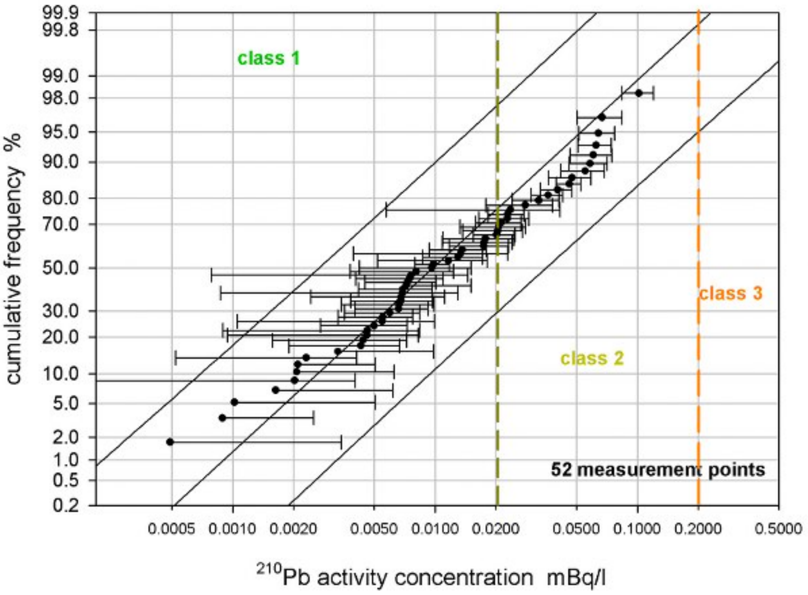

Figure 8.18: *Cumulative frequency distribution of ^{210}Pb activity concentrations of the detailed sampling*

104 samples were analyzed for ^{210}**Po** activity concentration in the laboratory of the AGES Vienna. **92** of these samples are **above decision limit** of 0.003 to 0.008 Bq/l. 51 samples above decision limits are in class 2, 8 in class 3. 7 of the samples in class 3 are located in the lower Mühlviertel, one in the upper Mühlviertel (Figure 8.19). The measurement point with the highest ^{210}Po activity concentration (0.28 ± 0.13 Bq/l) is the deep well in the lower Mühlviertel (WVA55), which also showed the highest ^{210}Pb and ^{226}Ra activity concentration. But the uncertainty of the measured ^{210}Po activity concentration is nearly 50%. The other measurement points in class 3 are also deep wells, one private well and the sample in the upper Mühlviertel were taken at a spring. 3 of these deep wells are the ones with the highest ^{222}Rn activity concentrations in the same village in lower Mühlviertel (WVA70, WVA71, WVA73). The other samples with ^{210}Po activity concentrations in class 2 are located in the Mühlviertel and Innviertel (Danube fault) (Figure 8.19).

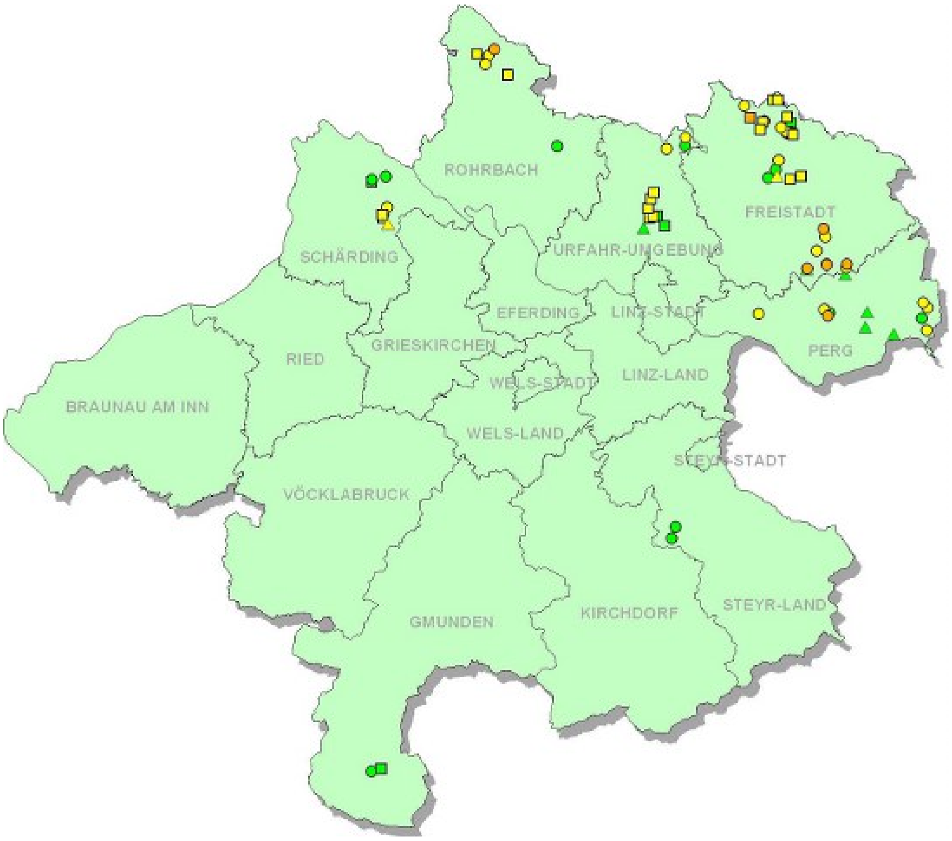

Figure 8.19: *All classified ^{210}Po results of the detailed sampling demonstrated in a map of Upper Austria, legend see Annex A1*

27 of the 104 samples were taken **directly at consumers' homes**, 26 are above decision limit (0.0008–0.0036 Bq/l) (Figure 8.20). Only one of these points has a ^{210}Po activity concentration in class 3, the above mentioned private well in the lower Mühlviertel (H12) with 0.12 ± 0.05 Bq/l. The second and third highest ^{210}Po activity concentrations were detected in different private wells (H13, H14) in the same village in the north of the lower Mühlviertel like H12 (Figure 8.19 and 8.20).

The **median** of the ^{210}Po activity concentrations is about 0.016 Bq/l (16 mBq/l) (Figure 8.21). According to the **cumulative frequency distribution** for 96% of Upper Austrian drinking waters ^{210}Po activity concentrations below 0.1 Bq/l are expected, which corresponds with an effective dose below 0.1 mSv/a. But this also means, that 4% of the Upper Austrian population will be exposed to a effective dose above the applied indicative dose limit of 0.1 mSv/a and so it seems that ^{210}Po is the nuclide with the greatest relevance in view of radiation protection, which is also discussed in Chapter 16.

^{210}Po activity concentrations in drinking waters in Austria were analyzed by Feigl-Heihs (1998) in 40 water samples from different regions of Austria. The detected ^{210}Po activity concentrations range from 0.8 to 63.8 mBq/l, which is a little bit lower than the detected ^{210}Po activity concentrations in this study. But Feigl-Heihs (1998) mainly analyzed tap water directly from consumers' homes. The highest detected

^{210}Po activity concentration directly at a consumer's house in this study is about 120 mBq/l, which is twice as high as the highest activity concentration found in the older study.

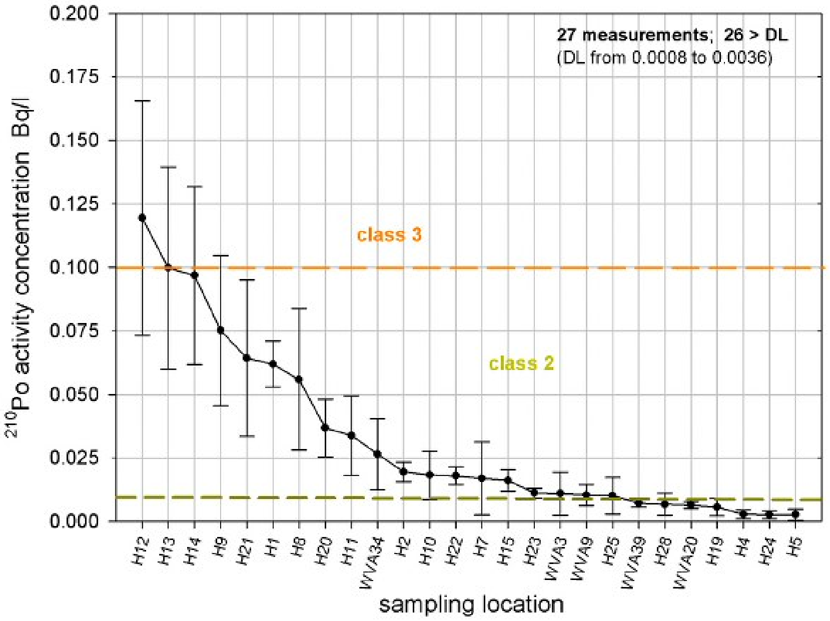

Figure 8.20: *Sorted ^{210}Po activity concentrations directly at consumers' homes*

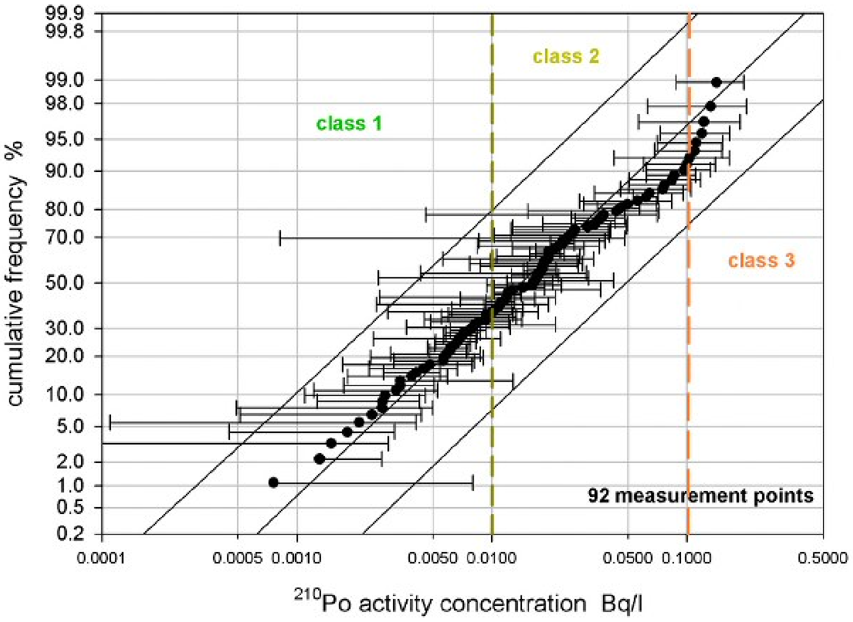

Figure 8.21: *Cumulative frequency distribution of ^{210}Po activity concentrations of the detailed sampling*

8.8 Uranium-238

128 samples were analyzed for ^{238}U activity concentration in the laboratory of the AGES Vienna. **102** of these samples are **above decision limit** of about 0.1 mBq/l. 10 samples above detection limit are in class 2, three in class 3. These three samples are water of a deep well, and water from a spring before and after neutralising water treatment supply in the same village in lower Mühlviertel (district Freistadt) (Figure 8.22). This deep well (WVA70) with the highest ^{238}U activity concentration (1.02 ± 0.05 Bq/l) is the measurement point which already showed the highest radon, gross alpha and gross beta activity concentration. The second and third highest ^{238}U activity concentrations are only less than half the value of the highest measured concentration (0.49 ± 0.02 Bq/l and 0.45 ± 0.02 Bq/l). 9 measurement points with ^{238}U activity concentrations in class 2 are located in lower Mühlviertel (Figure 8.22), one is the private well in central Mühlviertel (H1) with the highest radon concentration in the survey sampling. Two of these samples were taken at the deep wells in the district of Perg, which showed the highest ^{226}Ra activity concentration in the survey sampling and one of the samples in class 2 is the deep well (WVA55), which already showed the highest ^{226}Ra, ^{210}Pb and ^{210}Po activity concentrations in the detailed sampling.

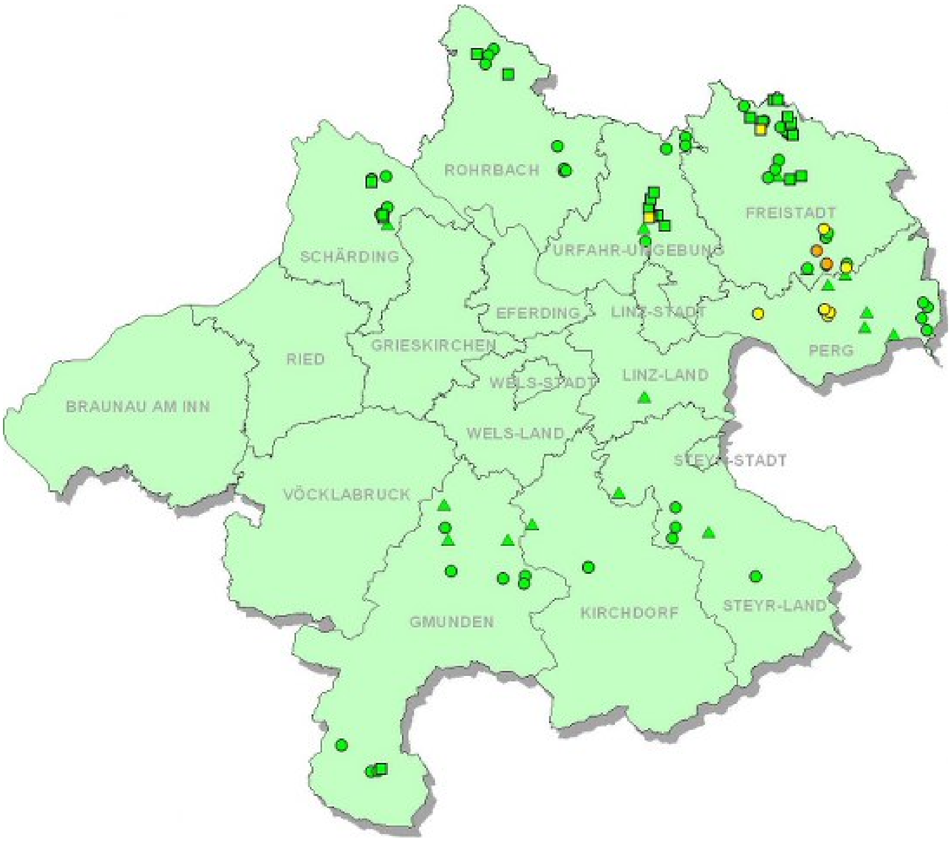

Figure 8.22: *All classified ^{238}U results of the detailed sampling demonstrated in a map of Upper Austria, legend see Annex A1*

Only two samples with ^{238}U activities in class 2 were taken **directly at consumers' homes** – above mentioned H1 and H14, one of the private wells in the village in the north of lower Mühlviertel (Figure 8.22), which showed highest ^{210}Po activity concentrations at consumers' houses.

In line with Chapter 7, the ^{238}U results of the detailed sampling should also be determined according a classification taking into account the new WHO guidance level for uranium (0.19 Bq/l). This classification for the results of the detailed sampling yields only slightly different results. 14 samples are related to class 2, 5 to class 3. The most interesting difference is that with this classification 3 samples of class 2 are located in the south of Upper Austria, which corresponds to the evenly distributed ^{238}U results among Upper Austria of the survey sampling.

The ^{238}U activity concentrations are **log-normal distributed** and yield a **median** of about 0.0025 Bq/l (2.5 mBq/l) (Figure 8.23). The measured ^{238}U activity concentrations of the survey sampling showed a slightly higher median of about 0.006 Bq/l (6 mBq/l). In contrast to the different radon results of the survey and detailed sampling, the sampling in preselected regions according to the selection criteria within the detailed sampling do not impact the median of the ^{238}U activity concentration distribution. Although some higher ^{238}U activity concentrations were detected in some regions of the Mühlviertel, it seems that **in general no significant higher uranium concentrations are expected** within these selected regions. That theory is confirmed by the evenly distributed measurement points with slightly elevated ^{238}U activity concentrations among Upper Austria within the survey sampling.

Figure 8.23 shows, like in the survey sampling, that for 99% of all Upper Austrian drinking waters ^{238}U activity concentrations below 0.37 Bq/l (class 3) are expected.

At least for about 98% of all drinking water samples in Upper Austria ^{238}U activity concentrations below 0.19 Bq/l are expected, which is the new recommended WHO guideline value in the "Guidelines for Drinking-water Quality, 3rd edition" (WHO, 2004).

^{238}U activity concentrations in 37 drinking waters of Austria were also anlayzed by Gegner (2002). The detected ^{238}U activity concentrations range from 1 to 148 mBq/l, with one higher activity concentration (975 mBq/l) in a spa in Salzburg. These are lower results than the highest ^{238}U activity concentrations of this study, but most drinking waters analyzed in this study also showed a much lower activity concentration. Gegner (2002) discussed that the uranium contents in the analyzed drinking waters have a high fluctuation also in wells with only some kilometers distance. This effect was also surveyed in this study, where no clearly higher ^{238}U activity concentrations in drinking waters of the Bohemian massif were detected, as it would be expected.

Some studies in the field of uranium in ground and drinking waters were carried out in Germany. Gellermann & Stolz (1997) analyzed 172 ground waters for ^{238}U and ^{234}U in the eastern part of Germany. The uranium concentration in ground water ranges from below 0.1 mBq/l to more than 1000 mBq/l with a mean of 12 mBq/l.

So these results are in the same order as the measured ^{238}U activity concentrations in Upper Austria, although the median is twice as high as the one detected within this thesis. Other studies in Germany reports similar results of ^{238}U in ground waters, Aurand & Gans (1991) reported a median ^{238}U activity concentration of the former western German federal countries of 2 mBq/l. A high range of fluctuation of ^{238}U activity concentrations was also reported by Gans (1992) (^{238}U activity concentrations ranges from 1 to 500 mBq/l) and also by Bünger & Rühle (1993) and Rühle (1997). The latter analyzed 106 samples of Saxony and Thuringia with a rather high median of 19 mBq/l and a range from 0.1 to 480 mBq/l.

Herranz et al. (1997) analyzed ^{238}U in drinking waters in northern Spain from 4 water treatment plants and detected activity concentration from 0.04 to 2.9 mBq/l with a mean value of 0.8 mBq/l, which is clearly below the detected uranium concentrations in Austria and Germany.

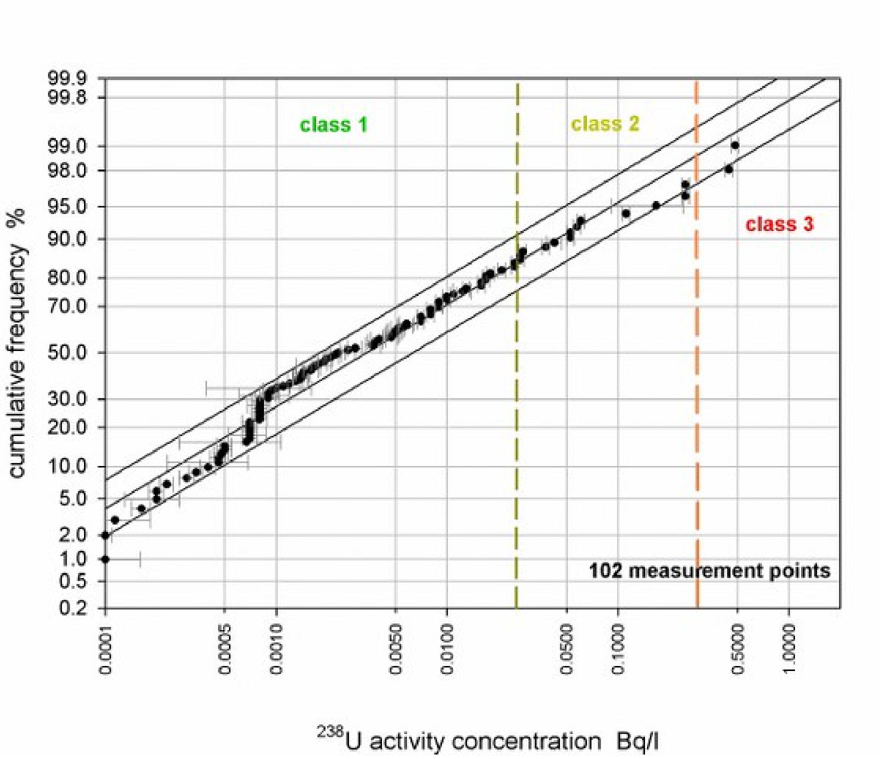

Figure 8.23: *Cumulative frequency distribution of ^{238}U activity concentrations of the detailed sampling*

In Figure 8.24 the cumulative frequencies of ^{238}U activity concentrations **directly at consumers' homes** (green) and at **wells or springs** (blue) are demonstrated. The median of the activity concentrations at consumers' homes is about 0.0015 Bq/l, the median at wells/springs is about 0.004 Bq/l. So the median at wells and springs corresponds better to the median of 0.006 Bq/l of the survey sampling, maybe because the samples in the survey sampling were also taken predominantly directly at wells and springs from the water supplies. The rather huge difference between ^{238}U activity concentrations at wells and at consumers' homes was not detected for the other radionuclides (except radon) and may indicate that uranium losses occur within the water flow from the well to the consumers' houses. This will be surveyed more detailed in Chapter 12.

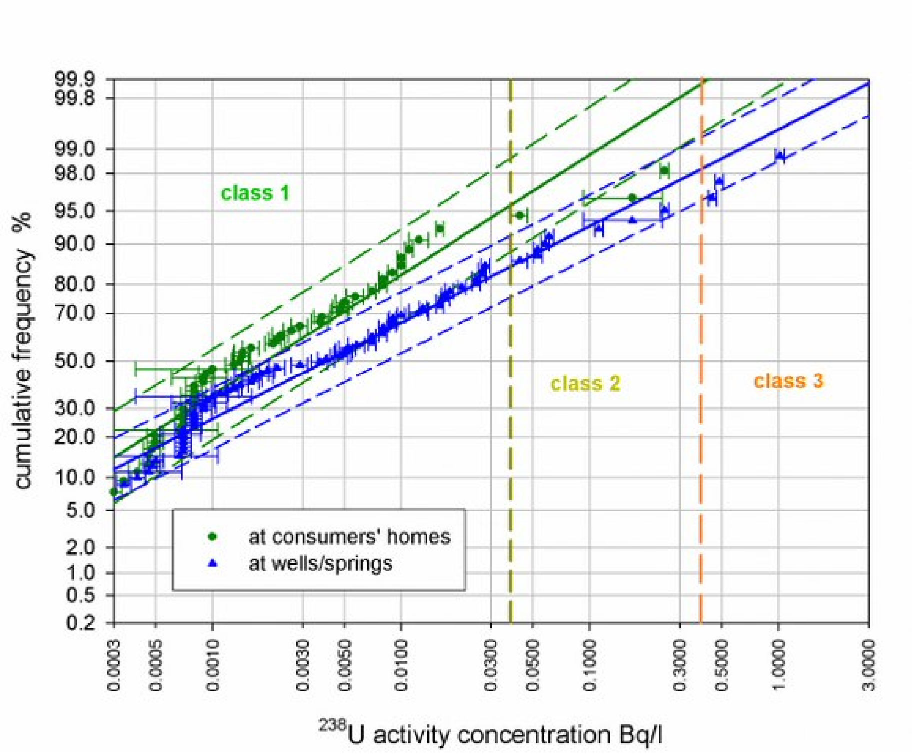

Figure 8.24: *Cumulative frequency distribution of ^{238}U activity concentrations at consumers' homes and wells and springs*

Chapter 9

Results of the ”Sacred Wells”

As mentioned in Chapter 5.1.2 additionally to the sampling in private wells and water supplies 16 samples were taken at **public wells and springs** in the investigated regions within the detailed sampling. These wells were selected from the book ”Sacred wells in Upper Austria” by Hirsch & Ruzicka (2002). These wells and springs are commonly regarded as being **pleasant** or **curative**. So these wells and springs are often used as drinking water by the public. During the sampling people could be watched bottling water from these wells in huge quantities for their own use at home and sometimes coming to these wells even from other districts or from larger towns. These wells or springs are mostly accessible free and sometimes integrated in little chapels.

Sampling at these wells is rather unproblematic because of free access, so samples were taken at some of these so called ”sacred wells” in the surveyed regions, because as described above these wells are used for drinking water on the one hand, and can also provide information about geological correlations on the other.

Although the results of these 16 ”sacred wells” are taken into account and used in the other chapters (e.g. Chapter 8 and 15 – triangles in the maps), in this chapter a brief look is taken at the specific results of the ”sacred wells”.

All **16** samples have ^{222}**Rn** activity concentrations **above decision limit**, 5 are in class 3 with activity concentrations above 100 Bq/l, 6 in class 2, 5 in class 1 (Figure 9.2). The 5 samples in class 1 are all located in the southern region of Upper Austria. The samples with the two lowest activity concentrations in class 2 are located in the central region of Upper Austria (district Linz Land and Steyr). Except for one measurement point (HEIL15) all samples with ^{222}Rn activity concentrations above 100 Bq/l (class 3) are located in the lower Mühlviertel. HEIL 15 is located in the Innviertel (Danube fault). This is mapped in Figure 8.2 in Chapter 8 and illustrated by triangles.

The two samples of the wells with the **highest radon activity concentrations** are declared to be **radon containing** or to be **curative radon wells**. The well with the highest radon activity concentration of 723 ± 61 Bq/l is the *Hedwigsbründl*[1] (HEIL5), a well in the village with a radon spa in the lower Mühviertel, in which also the highest radon activity concentrations in drinking water were detected. This ”sacred well” is integrated in a chapel and is a popular local ”pilgrim” location and sanctuary, because the water is suggested to be curative and therefore many people come to bottle it and use it as drinking water. Fuchs & Thiele (1987) discussed the mineral springs in the lower Mühlviertel and also described the mineral springs used for the spa (*Höllgrafenquelle*, *Krinnerquellen*) and the *Hedwigsbründl* with radon activity concentrations from 5.5 to 36 nCi/l due to strong seasonal variations. Nevertheless a radon concentration above the legal limit of a minimum of 10 nCi/kg for a radon spa is assured (Fuchs & Thiele, 1987, Land Oberösterreich, 1961). In Bequerel the springs varies from 204 to 1332 Bq/l (1 Ci/l = 3.7×1010 Bq/l), which corresponds with the measured radon activity concentrations in this village at the springs with about 380 Bq/l (WVA68+69). This also slightly exceeds the above mentioned legal limit of a minimum of 370 Bq/kg for radon spa water (according 10 nCi/kg, Land Oberösterreich (1961)).

The ”sacred well” with the second highest radon activity concentration (281 ± 27 Bq/l) is located in the north of the ”lower Mühlviertel” and is called *Maria Bründl* (HEIL14). Again the well is integrated in a small pilgrim chapel and a stone tablet nearby specifies the well (Figure 9.1). According to this tablet, the water of the well has a temperature of 7°C, comes from huge depth, never runs dry and contains sulphur, iron and radioactive elements with 20 **Mache units**. Mache unit is an old unit for radon concentration in spring waters named after the Austrian physicist *Heinrich Mache* (1876–1954). (Rühle, 1997, Wikimedia Foundation Inc., 2008a)

$$1\,\mathrm{ME} = 3.64 \times 10^{-10}\,\mathrm{Ci/l} = 13.4545\,\mathrm{Bq/l} \tag{9.1}$$

So, 20 Mache units result in 269 Bq/l, which corresponds very well with the radon activity concentration measured within this thesis (281 ± 27 Bq/l). Fuchs & Thiele (1987) also refer to this *Maria Bründl* as a slightly radioactive spring with 17.5 ME.

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[1] Bründl: vernacularly German for a small well, especially if the water is regarded to be pleasant or curative

Die " Maria Bründl Heilwasserquelle"
wurde durch Fleiß vieler Menschen,durch Geldspenden
sowie Unterstützungen einheimischer Firmen errichtet.
Bedenken Sie auch, dass hier ein Gnadenort entstanden ist,
wo schon viele Menschen Kraft, Heilung und Hilfe bis in
die Gegenwart gefunden haben.
Deshalb laden wir Sie auch ein auf einen Besuch in der
Gnadenkapelle.
Denn ER ist es, der uns dieses Geschenk bereitet .

Beschreibung der Heilwasserquelle:

Die Quelle entspringt rechts neben dem Altar und kommt aus
großer Tiefe. Sie versiegt nie und hat eine Temperatur von ca 7°C.
Das Wasser enthält radioaktive Elemente mit 20 Mache Einheiten
und ist schwefel-und eisenhältig.
Nach wissenschaftlichen Untersuchungen wurde deshalb das
Wasser als akratische Heilquelle anerkannt.

Maria Bründl,2003
Josef Friesenecker
Pfarrer

Figure 9.1: *Stone plate at Maria Bründl in the lower Mühlviertel*

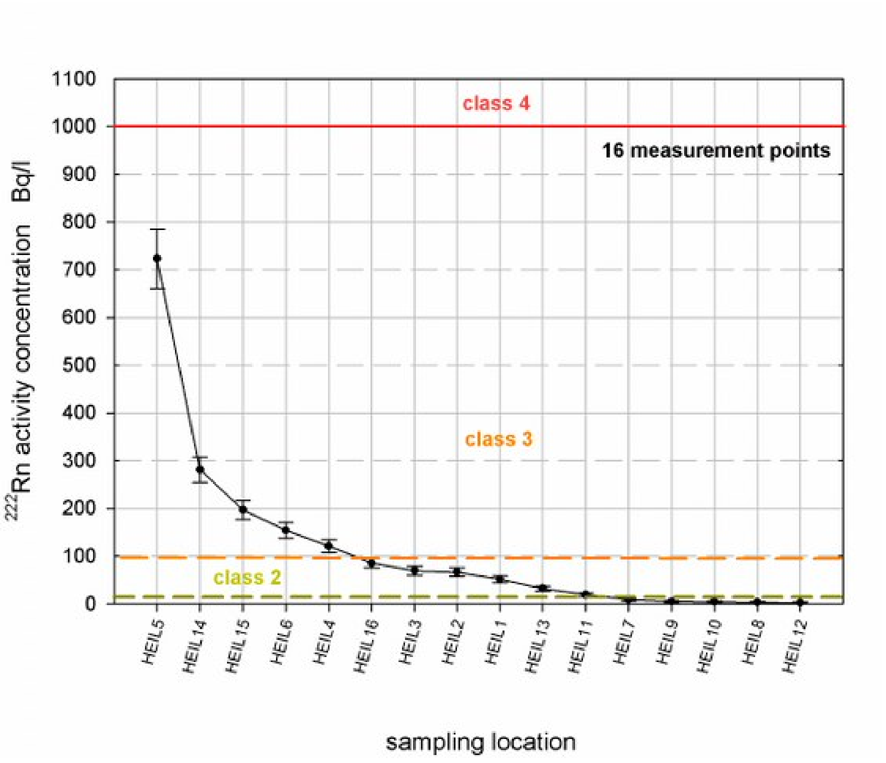


Figure 9.2: *Sorted Radon-222 activity concentration at "sacred wells"*

9 of the measured 16 "sacred wells" have a ^{238}**U** activity concentration **above decision limit**; all are in class 1, clearly below 0.037 Bq/l (figure 9.3). The 9 measurement points are distributed among Upper Austria, the highest, HEIL13 (0.016 ± 0.0008 Bq/l), is located in the central region of Upper Austria (district Linz Land). The second highest is HEIL5, the well with the highest ^{222}Rn activity concentration in lower Mühlviertel. Nevertheless correlation between ^{222}Rn and ^{238}U activity concentration can not be generalized. The samples with the second and third highest radon activity concentrations have low uranium activity concentrations and HEIL13, the sample with highest ^{238}U concentration has a low radon concentration (see Chapter 11).

No ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po activity concentrations above decision limit were measured at the "sacred wells". 4 samples have ^{3}H activity concentrations above decision limit, but all clearly in class 1.

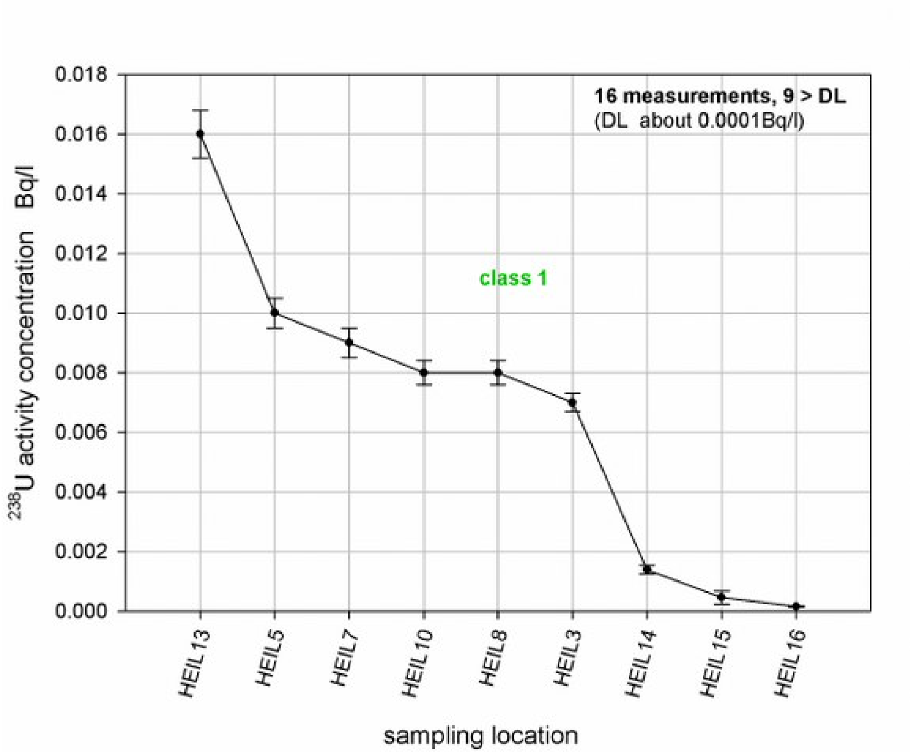


Figure 9.3: *Sorted ^{238}U activity concentration at "sacred wells"*

As discussed in Chapter 8.2 and 8.3 for gross alpha and gross beta the decision limits are rather high and a measurement point with a gross alpha or gross beta activity concentration above decision limit is related already to class 2 or class 3. Four samples have **gross alpha** activity concentrations above decision limit, three of them are in class 3. These three measurement points are all located in the lower Mühlviertel (HEIL6, HEIL3 and HEIL5) with gross alpha activity concentrations of 0.30 ± 0.10 Bq/l, 0.29 ± 0.09 Bq/l and 0.19 ± 0.08 Bq/l. Three samples have **gross beta** activity concentrations above decision limit, two of them are in class 2. The highest gross beta activity concentration (1.10 ± 0.61 Bq/l) was detected at HEIL14, the above discussed "sacred well" in the northern lower Mühlviertel with the second highest radon activity concentration. The measurement points with the second (HEIL11, 1.10 ± 0.61 Bq/l) and third (HEIL10, 0.74 ± 0.47 Bq/l, class 2) highest gross beta activity concentrations are located in the southern region of Upper Austria.

The results of the "sacred wells" for the different radionuclides correspond very well with the results of the drinking water samples in the surveyed regions, which again verify the importance of geology to radioactivity in drinking water (see Chapter 15).

Chapter 10

Comparison and Verification of Measurement Methods

10.1 Introduction

In the framework of this thesis different measurement methods were applied for determining radionuclide activity concentrations in drinking water. As described in Chapter 5 the samples were analyzed for different nuclides in **two different laboratories by different methods** (ICP-MS, LSC, gammaspectrometry). An assortment of the samples was analyzed for the same nuclides by different methods to compare them, especially for ^{222}Rn. ^{222}Rn measurements were also carried out on-site with the **mobile liquid scintillation counter Triathler**. This technique should be established in Upper Austria as a (quick and easy) measurement **method for drinking water monitoring of radon** and was therefore tested within this thesis (two different Triathlers, different cocktails, verification with other methods). The results of these testing measurements are discussed in this chapter and compared with the ^{222}Rn measurements carried out in the laboratories by gammaspectrometry and LSC. The results of the **comparison measurements** of the assorted samples in the two laboratories are discussed too – for a few samples also for other nuclides than radon (^{210}Pb, ^{226}Ra, ^{228}Ra, ^{238}U).

In this chapter also the **impact of the sampling process, sampling container, decanting and transport** on radon activity concentration in drinking water is surveyed with various test and comparison measurements at two sampling points.

The comparison measurements should verify the measurement results for the thesis and should also demonstrate the reasonability and practicability of different measurement methods for the question of surveying natural radioactivity in drinking water. The advantages and problems of the applied measurement methods are discussed in this chapter, and recommendations for correct sampling and measuring are given.

A part of these comparison and verification measurement results was also processed for a paper which is published in the Journal of Applied Radiation and Isotopes (Gruber et al., 2009).

10.2 Comparison of ^{222}Rn Measurements of AGES and LLC Laboratories with Samples of the Survey Sampling

In the **survey sampling phases** samples were taken in water supplies in 1 liter AFNORM glass bottles (see Chapter 5.1.1). At every sampling point a water sample was filled in two of this AFNORM bottles. All samples were analyzed for radon and other nuclides in the laboratory of the AGES by LSC, but 48 of the 79 water samples taken this way were also analyzed for ^{222}Rn by gammaspectrometry in the Low Level Counting Laboratory Arsenal. In these cases one of the two AFNORM bottles of every sampling point was analyzed in the LLC-laboratory, the other one in the AGES-laboratory.

As discussed in Chapter 5.2.3, radon can be analyzed in the LLC-laboratory by two different gamma detectors. Two of the samples were measured with both detectors for comparison, and the results correspond well within the uncertainties (about 15%), but the measured activity concentrations are generally very low, which leads to higher uncertainties due to counting statistics impact.

Figure 10.1 demonstrates the **correlation of the ^{222}Rn activity concentrations** of the 48 water samples **analyzed in both laboratories**. The axes of the figure are in a logarithmic scale because of generally rather low activity concentrations except for the point with the highest activity concentration of 166 ± 20 Bq/l (LLC) and 140 ± 35 Bq/l (AGES) respectively. The results of the radon measurements of the two laboratories do not correspond very well (r^2=0.62, r^2 is defined in Chapter 11.3). In most cases the radon activity concentrations measured at the AGES laboratory are higher than those measured in the LLC laboratory, especially in the region of very low activity concentrations. But the uncertainties of the radon activity concentration measurement results of the AGES laboratory are very high (up to 80%), especially for low activity concentrations. The uncertainties of the LLC laboratory range up to 30% for low concentrations.

The **not clear correlation** of the measurement results of the radon activity concentrations of the two laboratories may result from a not particularly cautious sampling for radon analysis. The samples were taken in the course of the second part of the Upper Austrian water project (see Chapter 5.1.1), and the sampler was maybe not sensitized well enough for the topic radon in drinking water and therfore was not as cautious as required for radon sampling. Besides, the measured radon activity concentrations are very low, and the uncertainties are rather high in this range of activity concentrations. So the correlation would be much better with activity concentrations in a higher range. But nevertheless, for a first estimation of radon activities the correlation of the measured activity concentrations is adequate.

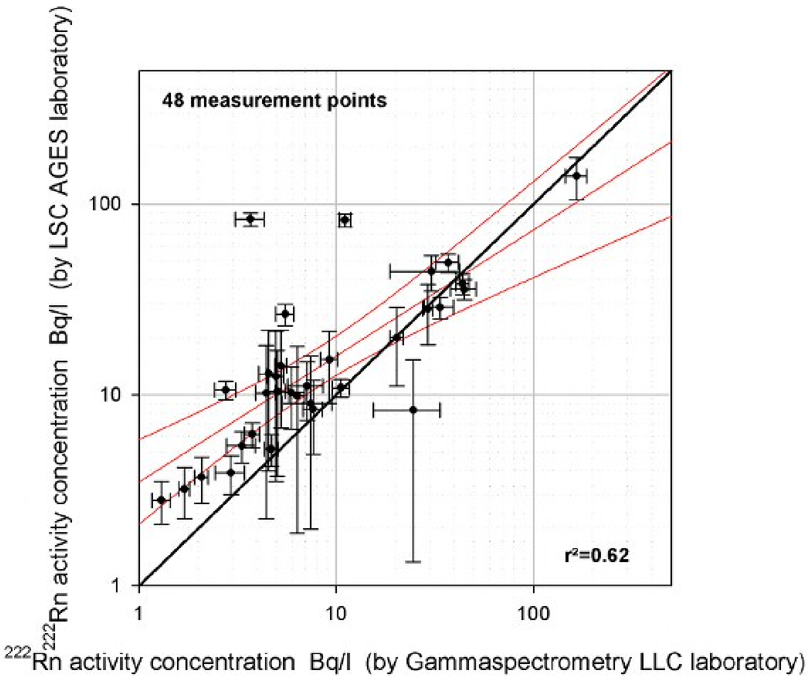


Figure 10.1: *Correlation of ^{222}Rn activity concentrations results from the survey sampling analyzed in the LLC and AGES laboratories*

10.3 Triathler Test and Comparison Measurements

As discussed above and in Chapter 5 the method of analyzing radon by the mobile liquid scintillation counter **Triathler** is **tested and verified** in the framework of this thesis to **establish** it as a **standard procedure for drinking water monitoring**. At every sampling point of the detailed sampling, samples were taken with two different cocktails – the water soluble cocktail Aqualight for immediately measurement, and the extraction cocktail Maxilight (see Chapter 5.2.2). At some sampling points additional samples were taken with the water soluble cocktail Ultima Gold for comparison measures. Some samples besides were also taken in AFNORM glass bottles for verification measurements by gammaspectrometry in the LLC laboratory. Measurements were carried out with two different Triathlers (different versions – the older one (AI) and newer one (OOE)) for comparison and further verification reasons.

Figure 10.2 shows the distribution of the radon activity concentrations measured by the **newer Triathler** (OOE) with **cocktail Aqualight** (black) **and cocktail Maxilight** (red) at the measurement locations. 96 samples were measured with the newer Triathler and both cocktails. The figure demonstrates that the radon activity concentration in water measured with the Aqualight cocktail is almost always slightly higher than the one measured with the Maxilight cocktail. Figure 10.3 illustrates the **correlation of the radon measurements with the cocktails Aqualight and Maxilight by the OOE Triathler** in another way. The linear correlation is very good with an r^2 value of 0.96, but the line of the best benefit (red line) is displaced slightly downwards for lower activity concentrations and slightly upwards for higher activity concentrations. The activity concentrations measured by Aqualight are mostly slightly higher, (mostly 10 to 20%), but at more than 20 measurement points also the radon activity concentration measured by Maxilight cocktail is higher (especially at higher activity concentrations). The black line demonstrates the 45° line, the reference line for exact correlation. For clearness the measurement results are demonstrated without uncertainties, which are always about 8% (see Chapter 5.2.2).

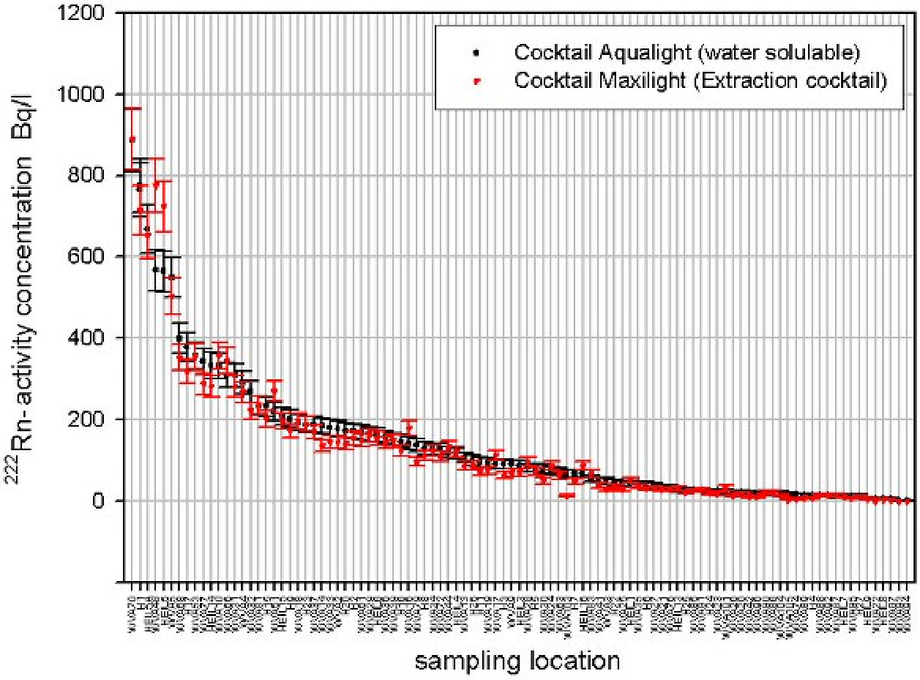


Figure 10.2: *Distribution of the radon activity concentrations analyzed by newer Triathler (OOE) with cocktails Aqualight and Maxilight related to measurement locations*

Figure 10.4 demonstrates a similar situation for the **older Triathler AI**. The linear regression yields an r^2 value of 0.97, but the line of best benefit is displaced downwards more clearly than for the Triathler OOE, because only three radon activity concentrations measured by Maxilight cocktails are above Aqualight. The radon activity concentrations measured by Aqualight cocktail and Triathler AI are in average 30% above the ones measured with Maxilight cocktail, but for the most measurements the difference is less than 30%. In this figure again the uncertainties are disregarded for clearness.

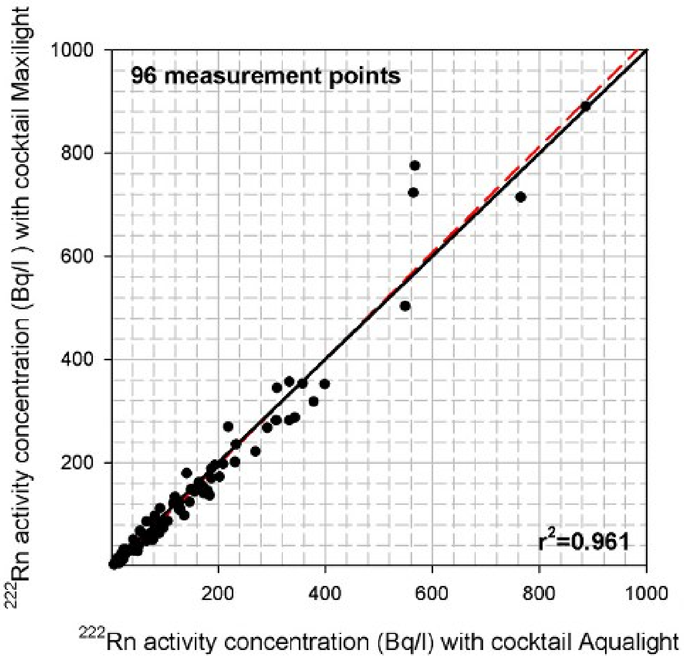


Figure 10.3: *Correlation of radon activity concentration analyzed by newer Triathler OOE with cocktails Aqualight and Maxilight*

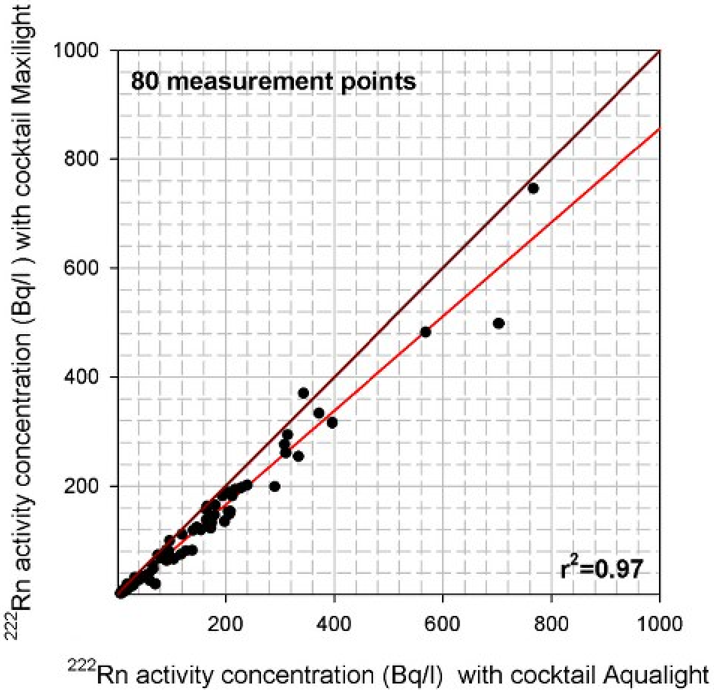


Figure 10.4: *Correlation of radon activity concentration analyzed by older Triathler AI with cocktails Aqualight and Maxilight*

The radon activity concentrations in waters analyzed by Triathler and Maxilight cocktail correspond rather well with the measurements by gammaspectrometry (see below), which is a verified measurement method, tested in the course of various comparison measurements. The radon activity concentrations measured with Triathler and the Aqualight cocktail are clearly higher. Most of the measurements with the Aqualight cocktails were carried out **on-site immediately after sampling**. So it might be that not only ^{222}Rn is measured, but also **^{220}Rn (thoron)**. As defined in Chapter 2.5.1, thoron is a decay product of the ^{232}Th series with a half life of 55.6 s. ^{222}Rn has a natural occurrence of the radon isotopes of 90%, ^{220}Rn of 9%. As an example – the highest measured radon activity concentrations were about 800 Bq/l. If this measurement result also includes ^{220}Rn, this might be a concentration, according to the above mentioned natural occurrence, of about 70 Bq/l. With a half life of 55.6 s, after about 5 minutes the thoron contribution can not be detected anymore with a decision limit of about 1 Bq/l. But the difference between radon activity concentrations measured by Maxilight and Aqualight also occured if the Aqualight measurements were carried out at a later time. So the thoron contribution can not be the main reason for measuring higher radon activity concentrations with the Aqualight cocktail. Though, it can influence the radon activity concentration measurement results by measuring immediately after sampling. To avoid this impact the measurements should be carried out not before waiting 10 minutes (about 10 thoron half lifes), when definitely all of the ^{220}Rn has decayed.

Another explanation for the higher radon activity concentration in water measured by Aqualight cocktail may be a **higher light sensibility**, because sampling and measuring on-site always was carried out in bright circumstances and not in shaded laboratory environments. Analyses with Aqualight cocktail operated in the laboratory mostly also show slightly higher Aqualight activity concentrations, but usually not more than 10%. So it seems that measurements with **Aqualight cocktail** usually yield **slightly higher radon activity concentration** results, but it should be kept down to a minimum by **waiting ten minutes before measuring** to exclude an impact by thoron and shield the vials with the cocktail and the sample in it as good as possible from light.

In Figure 10.5 and 10.6 the **correlations between the two Triathlers** (AI and OOE) are demonstrated differentiated between cocktail Aqualight (Figure 10.5) and Maxilight (Figure 10.6). The **activity concentrations measured by both Triathlers correspond well**, especially with the Maxilight cocktail (r^2=0.997). The line of best benefit is nearly congruent with the 45° line. The correlation regression for the radon activity concentration results of the two Triathlers at Aqualight cocktail yields an r^2 value of 0.987, but the line of best benefit is displaced slightly upwards, because the radon activity concentrations measured with the Triathler AI and Aqualight cocktail are mostly slightly (average 11%) higher than the ones measured by Triathler OOE. In these figures again the uncertainties are disregarded for clearness.

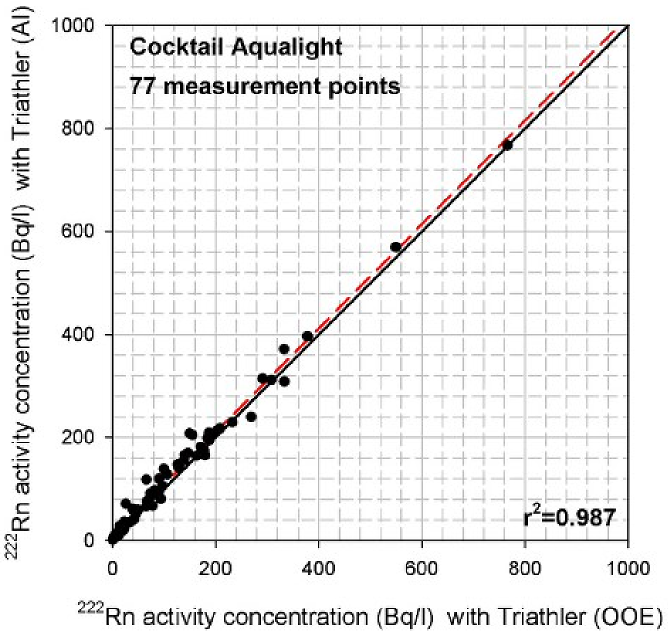


Figure 10.5: *Correlation of radon activity concentration analyzed by cocktail Aqualight with Triathler AI and OOE*

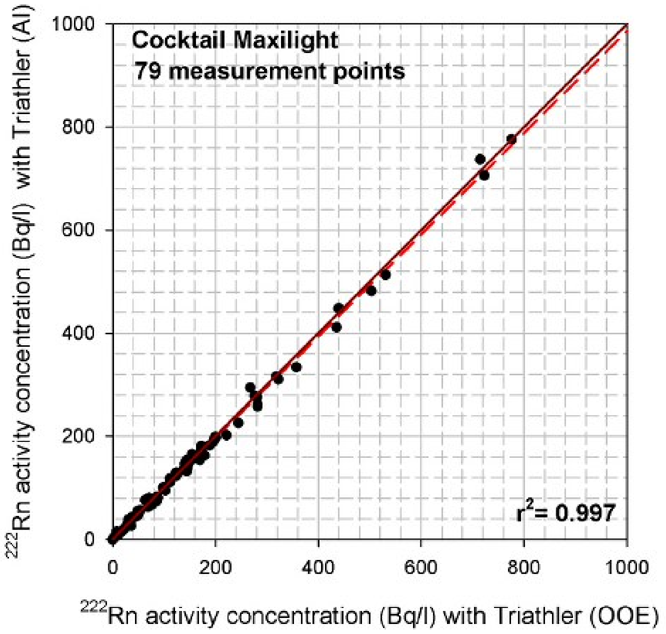


Figure 10.6: *Correlation of radon activity concentration analyzed by cocktail Maxilight with Triathler AI and OOE*

At 10 sampling points samples were taken additionally with the **water soluble cocktail Ultima Gold** (Perkin Elmer, see Chapter 5.2.2) and measured with both Triathlers (AI and OOE). The linear regression of the measured radon activity concentrations by Ultima Gold and the two Triathlers yields an r^2 value of 0.998, but the line of best benefit is displaced upwards, because all radon activity concentrations measured by the older Triathler AI are slightly higher (1 to 30%) (Figure 10.7).

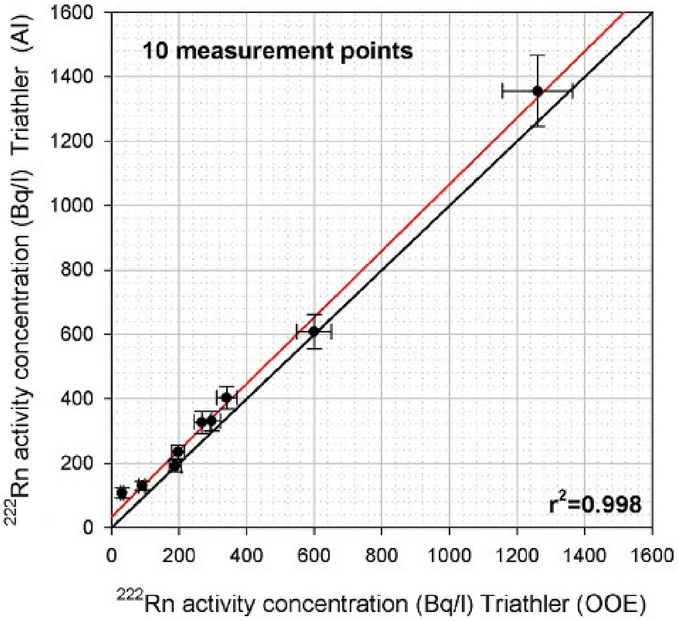


Figure 10.7: *Correlation of radon activity concentration analyzed by cocktail Ultima Gold with Triathler AI and OOE*

Figure 10.8 shows the radon activity concentrations measured by the newer **Triathler OOE and the cocktails Aqualight** (black), **Maxilight** (red) and **Ultima Gold** (green) at the 10 sampling points with a logarithmic scale. At three sampling points, the radon activity concentration measured by Ultima Gold cocktail is clearly higher than the ones measured by Aqualight and Maxilight (up to 60% higher than Maxilight and 56% higher than Aqualight). At the other sampling points the activity concentrations are equal within uncertainties (sometimes Ultima Gold is also lower than Aqualight or Maxilight).

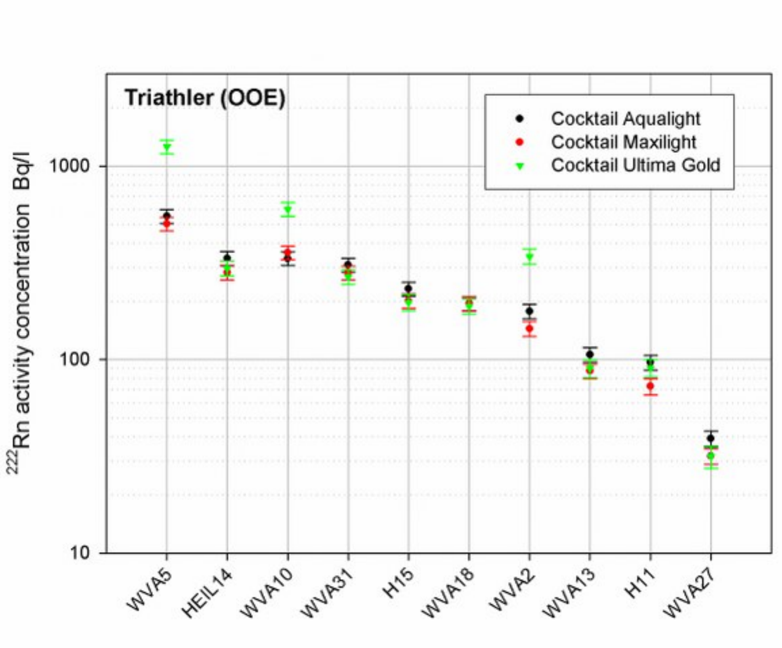


Figure 10.8: *Distribution of the radon activity concentrations analyzed by newer Triathler (OOE) with cocktails Aqualight, Maxilight and Ultima Gold related to measurement locations*

A similar situation results from the measurements by the older Triathler AI, with one more clearly higher radon activity concentration by Ultima Gold measurement (up to 71% higher than Maxilight and 64% higher than Maxilight).

Linear regression estimations of the correlations between radon activity concentrations measured by **Ultima Gold and other cocktails with the 2 Triathlers** reflect the above discussed results. The r^2 value of the linear regression for the Triathler OÖ for the comparison of Ultima Gold and Aqualight is 0.80, and for Ultima Gold and Maxilight 0.90. For the Triathler AI the linear regression of the correlation between Ultima Gold and Aqualight yields an r^2 value of 0.91, and for the correlation between Ultima Gold and Maxilight to 0.89. As an example the correlation between radon activity concentrations analyzed by Triathler AI and cocktails Ultima Gold and Maxilight is shown in Figure 10.9. The figure demonstrates that the r^2 values show a rather good linear correlation, but the line of best benefit is clearly shifted upwards, because of some clearly higher radon activity concentrations measured by Ultima Gold.

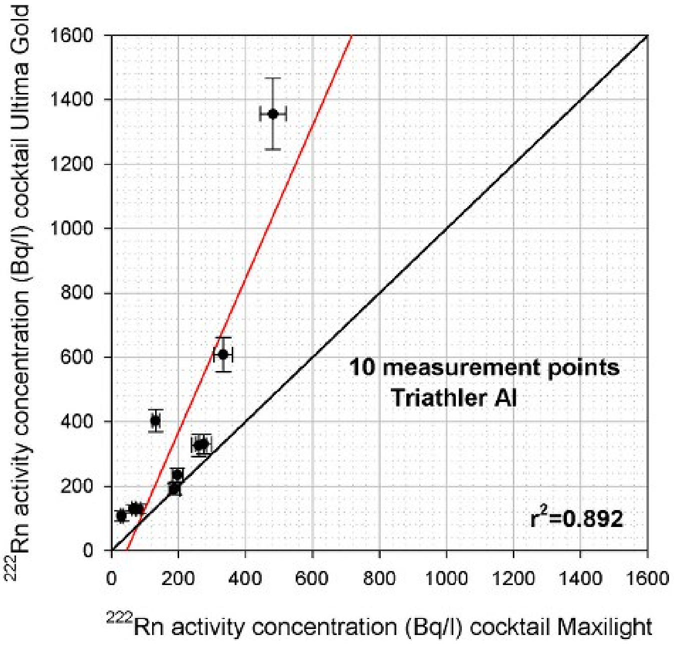


Figure 10.9: *Correlation of radon activity concentration analyzed by Triathler AI with cocktail Ultima Gold and Maxilight*

The single points with clearly higher radon activity concentrations measured by Ultima Gold cocktail may be explained by incautious sampling concerning light. It was a very bright, sunny day when these samples were taken, and the Ultima Gold cocktail is extremely light sensitive. The measurements with the Ultima Gold cocktail were carried out just out of curiosity. The Atomic Institute of the University of Technology Vienna, to whom the Triathler AI belongs, uses this cocktail for their LSC measurements in the laboratory. So a little of this cocktail was taken to the on-site Triathler measurements and some measurements were carried out. No further measurements were operated with this cocktail to get results for this thesis, but only for comparison and test measurements. For testing this cocktail as a routine measurement method, more test measurements should be carried out. Maybe the Ultima Gold cocktail is not applicable for on-site measurements because of its high light sensitivity, or it has to be treated with extreme cautiousness.

10.4 Comparison of Triathler Measurements with Other Methods

As mentioned above at **8 sampling points** additional water samples were taken in AFNORM glass bottles and analyzed by **gammaspectrometry** to verify and compare with the Triathler measurement results. The radon activity concentrations analyzed by Triathler are usually higher than the ones analyzed by gammaspectrometry, but dependent on the used Triathler and cocktail.

The best correlation occurs between gammaspectrometry and measurements with the Triathler OOE and **cocktail Maxilight** (Figure 10.10). The linear regression yields an r^2 value of 0.987 and the line of best benefit is displaced only slightly upwards. The highest deviation is 30%, but the others range between 1 and 15%, at one sampling point the radon activity concentration measured by gammaspectrometry is even 4% above the one measured by Triathler. So at most of the 8 analyzed sampling points the results of the two methods are equal within uncertainties. A similar situation is detected for the comparison of the measurements by Triathler AI and Maxilight cocktail, with an r^2 value of 0.987 and a slightly upwards displaced line of best benefit. The highest deviation is a little bit higher with 36%, and the other deviations range between 5 and 20%. At the same sampling point as above the radon activity concentration measured by gammaspectrometry is 6% above the one measured by Triathler AI and cocktail Maxilight.

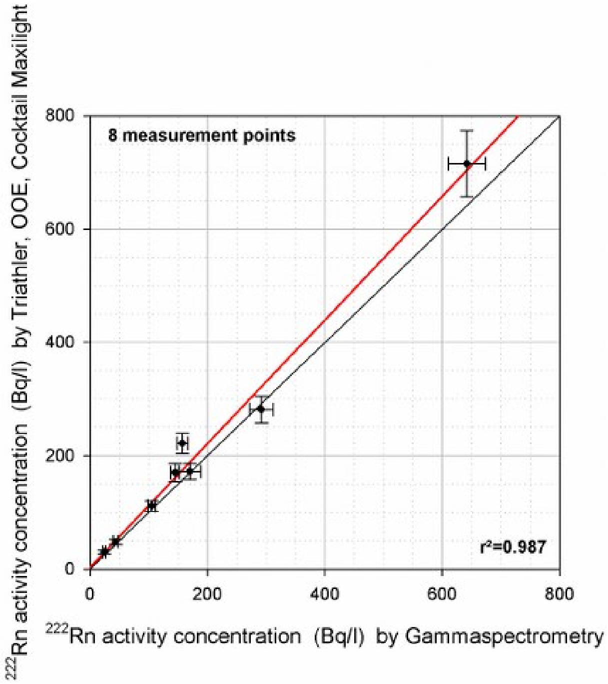


Figure 10.10: *Correlation of radon activity concentrations analyzed by Triathler OOE with cocktail Maxilight and gammaspectrometry*

The radon activity concentrations measured by **Aqualight cocktail** and the two Triathlers are clearly above the ones analyzed by **gammaspectrometry**. Figure 10.11 illustrates the correlation between the radon activity concentration measured by gammaspectrometry and Triathler AI with cocktail Aqualight. The linear regression yields an r^2 value of 0.988 and the line of best benefit is clearly displaced upwards. The highest deviation is about 60% and the others range between 5 and 30%. With this measurement method no radon activity concentration measured by gammaspectrometry is above the ones measured by Triathler AI and cocktail Aqualight. For the measurements with the Traithler OOE and cocktail Aqualight, a similar situation arises, with an r^2 value of 0.985 and deviations in a range between 12 to 42%. In most cases the results of the measurements with these different methods are not equal within uncertainties.

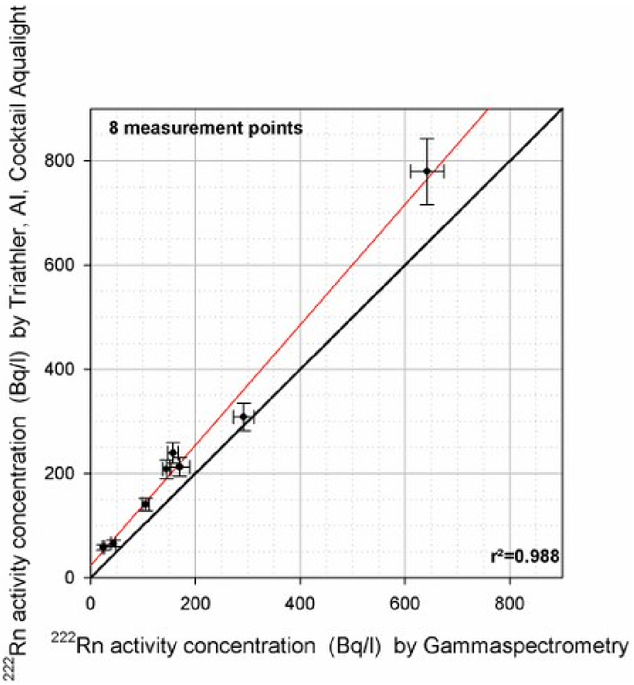


Figure 10.11: *Correlation of radon activity concentrations analyzed by Triathler AI with cocktail Aqualight and gammaspectrometry*

The radon activity concentrations measured by **gammaspectrometry** are slightly lower than the ones measured by **Triathler and Maxilight cocktail**, which nevertheless **correspond rather well**. The gammaspectrometry measurements have been verified and tested for a long time and therefore are very reliable. But the slightly discrepancy with the measurements with the Triathler measurements may be explained by marginal radon losses while filling the water in the AFNORM glass bottle, although this was carried out cautiously. This will be discussed below.

The samples for the nuclide analysis in the AGES laboratory were taken in 2.5 l plastic containers without cautiousness regarding radon losses, because radon analysis was not scheduled for these samples (see Chapter 5.1.2). Nevertheless these samples were analyzed for radon within the routine measuring program in the AGES laboratory, and used to correct the ^{210}Pb measurements (see Chapter 5.2.2).

As a next step the radon activity concentrations measured by **LSC in the AGES laboratory** were compared with the Triathler measurement results. As expected, the **radon activity concentrations measured in the AGES laboratory are clearly below the ones analyzed on-site by Triathler** (average above 70%). Only two samples with a radon activity concentration measured by AGES are slightly above the ones measured by Triathler. As an example, in Figure 10.12 the correlation between radon activity concentrations analyzed by LSC in the AGES laboratory and Triathler OOE with cocktail Aqualight is demonstrated. The linear regression of the 80 measurement points yields an r^2 value of 0.798 and the line of best benefit is displaced clearly upwards. The average deviation is 75.6%. For the measurements with the Aqualight cocktail and the Triathler AI a similar situation is detected (r^2 = 0.811, average deviation = 78.8%). For the measurements with the Maxilight cocktails the deviations are slightly lower (Triathler AI: 71.4%, Triathler OOE: 72.8%), but the line of best benefit is also clearly displaced upwards.

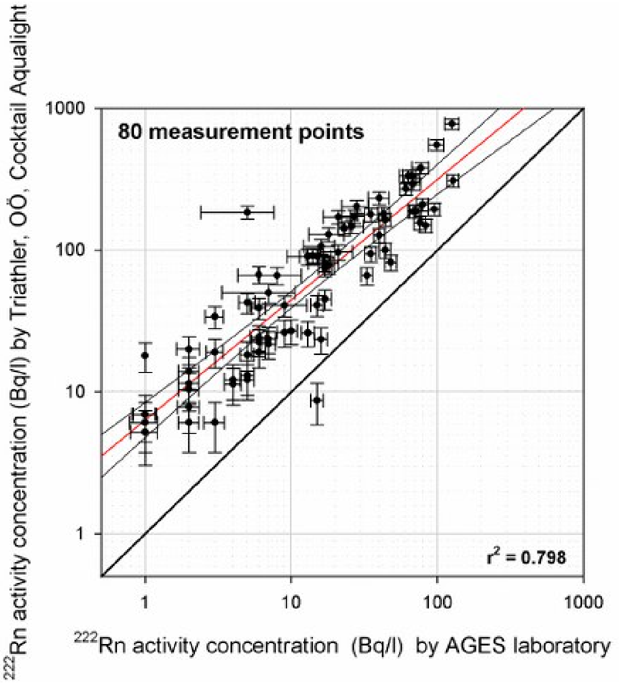


Figure 10.12: *Correlation of radon activity concentrations analyzed by LSC in the AGES laboratory and Triathler OOE with cocktail Aqualight*

The above discussed results confirm the fundamental impact of sampling, storage and transport on the radon activity concentrations. This issue is surveyed below 10.6. In this chapter also radon comparison measurements with the AGES laboratory and Triathler with radon capable sampling are presented.

10.5 Comparison of ^{222}Rn Measurements of AGES and LLC Laboratories with Samples of the Detailed Sampling

In Figure 10.13 the correlation between the radon activity concentrations of the 8 samples analyzed by **gammaspectrometry** in the **LLC laboratory** for comparison reasons and the respective activity concentration measured in the **AGES laboratory by LSC** is presented. The linear regression yields an r^2 value of 0.82

and the line of best benefit is displaced clearly downwards. The average deviation is 72.6% and deviations range between 51 and 83%. As discussed above the low radon activity concentrations of the AGES measurements can be explained with the not radon capable sampling. In Chapter 10.2 the comparison of these two radon measurement techniques showed total different results, because of the same sampling conditions for both methods. In that case the activity concentrations were equal within uncertainties for higher activity concentrations and otherwise the activity concentrations analyzed by the AGES laboratory were above the ones analyzed by gammaspectrometry.

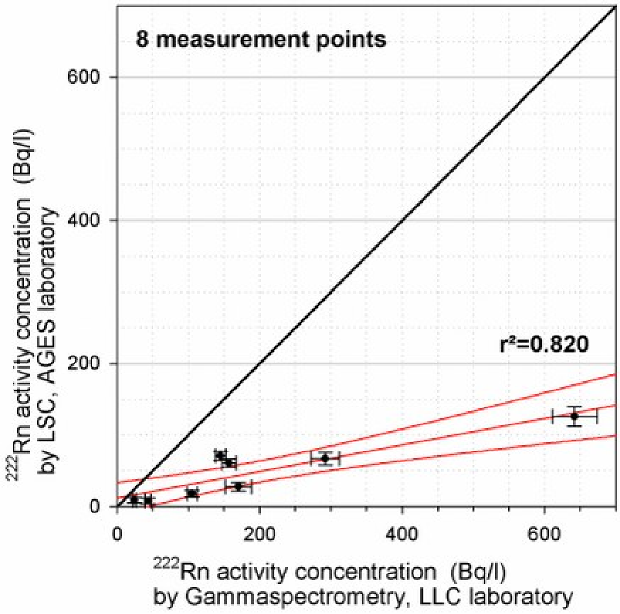


Figure 10.13: *Correlation of radon activity concentrations analyzed by LSC in the AGES laboratory and gammaspectrometry in the LLC laboratory*

In the following chapter the impact of sampling, storage and transport to the radon activity concentrations is discussed and results of measurements with both methods and comparable sampling are shown.

10.6 ^{222}Rn Analysis at two Selected Sampling Points with Different Measurement and Sampling Methods

Different sampling methods, the sampling container, decanting and the transport impact the measured radon activity concentrations. To characterize these impacts various samples were taken by different methods and containers at the **"sacred**

well" *Hedwigsbründl* (HEIL5) in the lower Mühlviertel. This well is easily reachable and open to the public, so sampling is totally unproblematic. Besides, the radon activity concentration in this water is rather high (about 700 Bq/l), so the measurements can be carried out with short measurement times and a radon activity concentration can still be measured after a couple of days' storage of the samples and after losses by transport and decanting.

The samples were taken at HEIL5 on April 4, 2007 in different sampling containers (1 liter glass bottles – AFNORM, 2.5 liter plastic containers) and also pipetted directly in some vials for Triathler measurement with both Aqualight and Maxilight cocktails. The samples were taken carefully (without bubbling) to avoid radon losses in the AFNORM bottles and in the 2.5 liter containers. These containers have a wide opening on the top, which may advantage radon outgassing.

All results of the radon activity concentration of the water from the *Hedwigsbründl* HEIL5 analyzed with different methods and sampling bins are **listed and summarized in Table 10.1**.

One of the water sample taken in a 1 litre AFNORM bottle was sent to the AGES laboratory and was analyzed there for radon and some other nuclides by liquid scintillation counting. Two samples in two other AFNORM glass bottles were directly measured for radon in the **Low Level Counting Laboratory** Arsenal by **gammaspectrometry**. The gammaspectrometry measurements of the two samples correspond well. The radon activity concentration measured by **LSC** in the **AGES laboratory** is slightly higher, but also within uncertainties equal (see table 10.1) . So, unlike in the survey sampling and detailed sampling, the radon measurement results of the AGES and LLC laboratories correspond well. because the samples were taken equally and in are in a range of a higher activity concentration. For generalization more samples should be measured, but it seems that the differences between the AGES and LLC measurements results arise from different sampling and containers, but not from the different measurement method.

On-site water was directly pipetted in four vials, two with **Aqualight cocktail** and two with **Maxilight cocktail** and transported to the laboratory. The samples were measured there by **Triathler**, each sample twice. The Triathler measurements were carried out in the laboratory, because there was no possibility to provide the Triathler on-site. The 4 measurements per cocktail are equal within uncertainties (maximum deviation of 3%). The mean value of the radon activity concentration by Aqualight cocktail and by Maxilight cocktail are given in Table 10.1 with the combined measurement uncertainties of the mean values (Type A, coverage factor k=1 according to ISO (1995)). So as discussed above, again radon activity concentration measured by Aqualight cocktail is about 10% above the one measured by Maxilight. The measurements with both cocktails were carried out some days after sampling, so impact by immediate on-site measurements with thoron and light effects can not be decisive.

The radon activity concentration measured by **Triathler** and pipetted in vials directly on-site is about 13% (for Maxilight cocktail) and 22% (for Aqualight cocktail) higher than the radon activity concentration measured by **gammaspectrometry and LSC in the laboratories** with samples filled in AFNORM glass bottles. It seems that radon losses happen while filling the water in the bottle, in spite of careful sampling without bubbling. To verify this theory and to exclude the possibility that the discrepancy occurs because of different measurement methods, water was pipetted out of the AFNORM bottles in vials with both Maxilight and Aqualight cocktail and analyzed by Triathler. The mean radon activity concentrations of the samples of the two different AFNORM bottles differ for Aqualight cocktail about 5% and are equal for Maxilight cocktail (see Table 10.1). The radon activity concentration in the two bottles measured by Triathler and Maxilight cocktail correspond well with the results measured by gammaspectrometry (3–5% aberration). The radon activity concentrations measured by Triathler and Aqualight cocktail are again about 10–20% higher. So, the well corresponding Maxilight and gammaspectrometry results in the two bottles verify the theory that radon losses of about 10% occur while sampling into the AFNORM bottles.

As mentioned above water samples were additionally taken in two **2.5 l containers**, which were also used in the detailed sampling for the AGES laboratory analyses. These samples were primarily taken for evaporation and analyzing of other nuclides by gammaspectrometry. But also the **radon losses** by filling water in these containers and storing and transporting them are tested, because of the detected and above discussed high radon activity concentration differences in the laboratories. So water was pipetted out of the 2.5 liter containers in the laboratory in vials with Aqualight and Maxilight cocktails and measured by Triathler. The radon activity concentrations of the samples of the two different containers differ for Aqualight cocktail about 4% and for Maxilight cocktail about 10% (see Table 10.1). In both cases the radon activity concentration of container 1 is above the one of container 2. The difference between the radon activity concentration measured by Aqualight and Maxilight cocktail in the containers is again about 5 to 10%. The radon activity concentration of the water samples in the containers measured with Triathler and the two cocktails are all about 30% below the radon concentration measured in vials with water pipetted directly on-site. This **radon activity concentration decrease of 30%** is detected by Aqualight and Maxilight measurements, and seems to represent the radon losses by filling water in the containers and the not existing radon tightness of the containers, although the sampling was carried out carefully without bubbling.

For one more comparison measurement of the methods and survey of **losses by decanting**, part of the water from the container 2 was filled with the help of a funnel in a 1 liter AFNORM glass bottle and measured for radon by gammaspectrometry. Afterwards again water is pipetted out of the AFNORM bottle in vials with Aqualight and Maxilight cocktails and measured by Triathler. The radon activity concentration of the water in the AFNORM bottle analyzed by gammaspectrometry and the results of the Triathler measurements are listed in last column of Table 10.1. The radon activity concentrations measured by gammaspectrometry and Triathler with Maxilight cocktail correspond and are equal within uncertainties. The radon activity concentration measured by Triathler and Aqualight cocktail in this case is

19% above the one measured by Maxilight cocktail. So the results show that **decanting water** from the container in an AFNORM bottle by funnel causes again **25% radon losses**.

Table 10.1: *Mean radon activity concentrations of HEIL5 with different measurement methods and different sampling*

Measurement Method	Measured radon activity concentration (Bq/l)						
	On-site in Vials	**AFNORM AGES**	**AFNORM Bottle 1**	**AFNORM Bottle 2**	**Container 1**	**Container 2**	**AFNORM decanted from Container 2**
Triathler and Aqualight cocktail	708 ± 29	–	670 ± 39	633 ± 37	490 ± 28	469 ± 27	391 ± 25
Triathler and Maxilight cocktail	635 ± 12	–	574 ± 33	576 ± 35	470 ± 27	424 ± 25	317 ± 21
Gamma-spectrometry	–	–	542 ± 28	558 ± 33	–	–	288 ± 16
AGES LSC	–	573 ± 34	–	–	–	–	–

In Chapter 10.8 the results of these comparison measurements are summarized and an instruction for radon water sampling is given.

Similar **comparison measurements** were carried out with samples from two other sampling points (one private well and one tap water of the local water supply) in a village in the Innviertel. These samples were analyzed because no samples were taken in this region in the survey and detailed sampling, but the possibility was offered to get samples from this village later. So these samples were analyzed and should also be used for radon comparison measurements by Triathler and gammapectrometry in the laboratory, but the radon activity concentrations in these waters are very low and therefore comparison measurements do not show as clear results as the one at HEIL5. Nevertheless the results are discussed here.

All results of the radon activity concentration of the water from the two sampling points in the Innviertel analyzed with different methods and sampling bins are **listed and summarized in Table 10.2**.

The **two samples** were not taken directly into 1 liter AFNORM glass bottles or rather pipetted on-site in vials for Triathler measurements, but were filled in a 1.5 liter mineral water PET bottle (private well sample) and a 2 liter **PET bottle** (tap water). Two days after sampling the samples were **decanted** into two AFNORM glass bottles by a funnel in the laboratory and measured by gammaspectrometry for some hours. Afterwards for both samples water was pipetted out of the AFNORM bottles in vials for Triathler measurements with Aqualight and Maxilight cocktails. Additionally water was pipetted in vials from the remaining water in the PET bottles one day later. Because the remaining water level in these bottles was low, pipetting was not possible directly, so the water was poured out in a small beaker and then pipetted in the vials with Aqualight und Maxilight cocktail and measured by Triathler.

The radon activity concentration in the two samples measured by Triathler and Maxilight cocktail correspond with the results measured by gammaspectrometry (equal within uncertainties). The radon activity concentrations measured by Triathler and Aqualight cocktail is again about 10% higher for the sample of the private well, and 70% higher for the tap water sample. But the radon activity concentrations measured in the tap water samples are very low (< 10 Bq/l) and should not be used for comparison purposes, because the decision limit of the Triathler measurements is in the range of some Bq/l and in a scale below 10 Bq/l, the results should be used for estimation only and not overstated as exact values. This limitation causes no problem, because for radiation protection purposes radon activity concentrations in this low range are of no importance.

The radon activity concentration results of the Triathler measurements of the private well sample, which was pipetted from the PET bottle after pouring out, are clearly below those from the AFNORM bottle (81% losses for Maxilight, 63% losses for Aqualight). These radon losses may be caused by non radon tight PET bottles, decanting and also by radon outgassing from the water in the air in the not fully filled bottle. For the tap water sample this effect is not detectable that clearly, but as discussed above, the radon activity concentrations are too low for comparison measurement purposes in this sample.

Table 10.2: *Mean radon activity concentrations of two samples with different measurement methods and different sampling*

Measurement Method	**Measured radon activity concentration (Bq/l)**			
	AFNORM bottle private well	**PET bottle private well**	**AFNORM bottle tap water**	**PET bottle tap water**
Triathler with Aqualight cocktail	30 ± 1	11 ± 1	10 ± 1	9 ± 1
Triathler and Maxilight cocktail	22 ± 1	4 ± 0	2 ± 0	1 ± 0
Gamma-spectrmetry	25 ± 2	–	3 ± 1	–

This example verifies the results discussed above, that decanting, wrong containers, storage time and sampling clearly impact the radon activity concentration in the water sample.

10.7 Measurement of other Radionuclides at Selected Sampling Points

About two liters of the above described water sample HEIL5 (*Hedwigsbründl*) (container 1) were **evaporated** and measured by **gammaspectrometry** for **different radionuclides**. One AFNORM glass bottle was given to the **AGES laboratory**, where it was analyzed not only for radon, but also for some other nuclides according to the detailed sampling analyses by **LSC**. The measurements should serve as comparison measurements, but due to capacity problems in the AGES laboratory at this time the sample could not be analyzed for all nuclides (no ^{210}Pb, ^{210}Po and ^{238}U).

So only the activity concentrations of ^{226}Ra and ^{228}Ra can be compared, but samples from this sampling location (HEIL5) were analyzed within the detailed sampling phase and in Table 10.3, these results also are listed. The comparable activity concentrations correspond, both the results of the LLC-laboratory and the AGES laboratory and the AGES laboratory results at different times. But most of the activity concentrations are rather low (all in class 1) and below decision limits.

Table 10.3: *Measured nuclide activity concentrations at sampling location HEIL5 with different methods and different laboratories and time*

	Measured activity concentrations (Bq/l)		
Nuclide	**LLC laboratory gammaspectrmetry**	**AGES laboratory (comparison measurement)**	**AGES laboratory (detailed sampling phase)**
Gross alpha	–	0.17 ± 0.06	0.19 ± 0.08
Gross beta	–	< 0.56	< 0.56
^{3}H	–	1.4 ± 0.5	< 0.93
^{226}Ra	0.0228 ± 0.0023	< 0.05	< 0.055
^{228}Ra	< 0.0073	< 0.006	< 0.0058
^{210}Pb	< 0.0295	–	< 0.0037
^{210}Po	–	–	< 0.0022
^{238}U	< 0.0357	–	0.0100 ± 0.0005

Additionally, samples of **three measurement points of the detailed sampling phase**, which were analyzed in the AGES laboratory for different radionuclides, were also evaporated and analyzed by **gammaspectrometry** in the LLC laboratory. Only one liter of each sample was available for evaporating, so long measurement time was required for gammaspectrometry and the measured activity concentrations have rather high uncertainties.

In Table 10.4–10.6 the measured nuclide activity concentrations in the two laboratories are listed. Again also the ^{222}Rn activities are shown, in comparison to the

Triathler ^{222}Rn results. The radon activity concentration results of the Triathler measurement and the one measured by gammaspectrometry correspond well, the problems with the AGES ^{222}Rn measurement are discussed above.

The three measurement points are all located in the Mühlviertel, **H1** and **H9** in the **central Mühlviertel**, **WVA32** in the **upper Mühlviertel**. For H1 the measured ^{226}Ra activity concentrations of the two laboratories are equal within uncertainties, the ^{210}Pb concentration of the AGES laboratory are clearly above the one measured in the LLC-laboratory. The ^{238}U activity concentration measured in the AGES laboratory is also slightly above the one measured by gammaspectrometry. A similar situation is shown in Table 10.5, at location H9. The ^{210}Pb activity concentration analyzed by the AGES laboratory is clearly above the one measured in the LLC laboratory. All other nuclides are within uncertainties equal.

For the samples from measurement point WVA32 all measurement results correspond, but are very low, all below decision limit in the LLC analyses.

In general the activity concentrations measurement results for the different nuclides of the two laboratories correspond satisfyingly. Only the results of ^{210}Pb diverge, but the measured activity concentrations in the LLC laboratory are all above decision limit and have a high uncertainty because of evaporating only one liter of water. Usually at least 2 liters of water are required for evaporating water samples for gammaspectrometry analysis for appropriate uncertainties and measurement times.

Table 10.4: *Measured nuclide activity concentrations at sampling location H1 with different methods and different laboratories*

	Measured activity concentrations (Bq/l)		
Nuclide	**LLC laboratory gammaspectrometry**	**AGES laboratory**	**Triathler with Maxilight Cocktail**
Gross alpha	–	< 0.17	–
Gross beta	–	< 0.56	–
^{3}H	–	2.20 ± 1.06	–
^{226}Ra	0.055 ± 0.019	0.079 ± 0.020	–
^{228}Ra	< 0.0173	< 0.006	–
^{210}Pb	< 0.034	0.102 ± 0.018	–
^{210}Po	–	0.062 ± 0.009	–
^{238}U	< 0.038	0.043 ± 0.004	–
^{222}Rn	642 ± 32	126 ± 14	745 ± 63

Table 10.5: *Measured nuclide activity concentrations at sampling location H9 with different methods and different laboratories*

	Measured activity concentrations (Bq/l)		
Nuclide	**LLC laboratory gammaspectrometry**	**AGES laboratory**	**Triathler with Maxilight Cocktail**
Gross alpha	–	< 0.16	–
Gross beta	–	1.40 ± 0.62	–
^{3}H	–	< 1.20	–
^{226}Ra	0.035 ± 0.018	< 0.040	–
^{228}Ra	< 0.0175	< 0.005	–
^{210}Pb	< 0.035	0.058 ± 0.012	–
^{210}Po	–	0.075 ± 0.030	–
^{238}U	< 0.039	0.0022 ± 0.0002	–
^{222}Rn	170 ± 19	28 ± 6	182 ± 18

Table 10.6: *Measured nuclide activity concentrations at sampling location WVA32 with different methods and different laboratories*

	Measured activity concentrations (Bq/l)		
Nuclide	**LLC laboratory gammaspectrometry**	**AGES laboratory**	**Triathler with Maxilight Cocktail**
Gross alpha	–	< 0.16	–
Gross beta	–	0.86 ± 0.62	–
^{3}H	–	1.20 ± 0.94	–
^{226}Ra	< 0.0102	< 0.037	–
^{228}Ra	< 0.0185	0.009 ± 0.005	–
^{210}Pb	< 0.035	< 0.007	–
^{210}Po	–	0.008 ± 0.013	–
^{238}U	< 0.039	0.00080 ± 0.00008	–
^{222}Rn	157 ± 10	61 ± 6	222 ± 22

10.8 Conclusions

The measured radon activity concentrations of the **comparison and testing measurements with the different Triathlers and different LSC-cocktails correspond rather well**, although the measurements with the water solulble cocktail Aqualight is always slightly above the ones measured by Maxilight cocktail, which correspond well with the gammaspectrometry measurements. Nevertheless the immediate on-site measurements with Triathler and the Aqualight cocktail are a good method for a radon survey and monitoring program.

The results of the other methods of radon measurements (LSC by AGES laboratory, gammaspectrometry by LLC laboratory) correspond satisfying, assumed that equal and right sampling was carried out. The results of the other nuclides by LSC and gammaspectrometry correspond also satisfying, although the comparison measurements were carried out with samples with very low activity concentrations, often below decision limit.

The comparison measurements and survey of different sampling bins and methods show that **decanting, wrong containers and sampling strongly impact the radon activity concentration in the water samples**. The used 2.5 liter plastic container causes radon losses of 30% by filling and not radon tightness. Decanting from containers into an AFNORM bottle for gammaspectrometry also causes radon losses of about 25%. Direct sampling in an AFNORM bottle nevertheless causes radon losses of about 10%.

So as a conclusion, **instructions for radon water sampling** are demonstrated. It seems that the best sampling method for radon measurements (without losses) is **direct pipetting of water in vials on-site**. For gammaspectrometry the water should be **directly filled** in AFNORM glass bottles carefully **without bubbling** on-site, because later decanting causes high radon losses. Some losses are nevertheless possible and should be verified. The 2.5 liter plastic containers should not be used for radon analysis, because of high radon losses by sampling into the wide opening and storage and transport in the **not radon tight containers**. **To avoid radon losses by decanting water sampling should always be carried out directly into the measurement bins.**

Therefore it is fundamental to standardize and improve water sampling for radon analyses. More measurements with different sampling containers and bottles and sampling methods should be carried out with the objective to work out **instructions for standardized and correct sampling** (e.g. within a standard – ÖNORM) for analyzing radon in drinking water, which will be an important procedure in the future for radiation protection reasons.

Chapter 11

Correlations of Radionuclides

11.1 General Aspects

In Chapter 8 results of the detailed drinking water sampling were discussed regarding nuclides. It was demonstrated in this chapter that some measurement points have noticeably higher activity concentrations of different nuclides. Others have a higher activity concentration of one nuclide, but low activities of all others. So it is interesting to take a closer look at some measurement points and demonstrate all measured radionuclide activity concentration results at this point to figure out **radionuclide correlations or single occurrences** there. This happens in the **first part** of this chapter.

Furthermore it is of major interest to search for **general nuclide correlations** for scientific reasons on the one hand, and for radiation protection reasons on the other. Knowledge about correlations of different radionuclides can help within dose estimations and risk assessments, if not all nuclides in a drinking water sample have been analyzed but fundamental coherences between the individual nuclides are known. Besides, if well-founded nuclide relations are established, sampling and analyzing would be strongly simplified and time as well as costs could be saved, because samples have to be analyzed for some radionuclides only and then estimated for a total dose. So in the **second part** of this chapter correlations between radionuclides are surveyed, demonstrated and tested. For the radionuclides, analyzed within the detailed sampling, **different correlations** may be **expected**. ^{222}Rn, ^{226}Ra, ^{238}U and ^{210}Po are α-emitter, so correlations with gross alpha activity may be expected; ^{228}Ra, ^{3}H and ^{210}Pb are β-emitter, so correlations with gross beta activity may be expected. On the other hand, ^{226}Ra, ^{222}Rn, ^{210}Pb and ^{210}Po are all generated within the ^{238}U decay series, so correlations are likely. ^{226}Ra and ^{228}Ra are both radium isotopes, which often occur together. So these possible correlations will be surveyed and tested in this chapter.

11.2 Overview of Nuclide Activity Concentrations at Selected Sampling Points

In the following figures (page 147 – 149) **all measured activity concentrations at single measurement points** are demonstrated. The demonstrated measurement points were selected because they attracted attention in Chapter 8 with some higher nuclide activity concentrations (like WVA70, WVA55, H1) at one point or one clearly higher nuclide activity concentration. At least the measurement points with the **two highest activity concentrations from each nuclide** are graphed and especially some **private well** measurement points are taken into account, which will be also interesting for dose calculations. For differentiation some measurement point with lower nuclide activity concentrations are displayed. The demonstrated sampling points should take into account as many different existing cases as possible (higher nuclide activity concentrations, geology regions, private wells, sacred wells, public water supplies). In the figures the measured activity concentration of each nuclide is demonstrated in a bar chart with different colors. All x-axis have the same logarithmic scale, to allow to graph all nuclide activity concentrations in Bq/l and to compare with other measurement points. If the measurement results are above decision limit, *"<DL"* is marked in the graph. At a couple of measurement points some nuclides were not analyzed, displayed in the graph with *"not analysed"*.

As described in Chapter 8 **WVA70** is a deep well in the lower Mühlviertel and has the highest ^{222}Rn, gross alpha, gross beta and ^{238}U activity concentration and the third highest ^{210}Po activity concentration. **WVA73** and **WVA71** are a reservoir and another deep well in the same village and show the second and highest ^{222}Rn activity concentration, but no "leading positions" concerning other nuclides. WVA73 and WVA71 have similar high ^{222}Rn activity concentration as WVA70, but much lower gross alpha, gross beta and ^{238}U activities. On the other hand, WVA71 and WVA73 show similar activity concentration for all nuclides, maybe more water in the reservoir is from the deep well WVA71. Already these first cases illustrate that correlations between different nuclide activity concentrations are a complex problem, if also wells with related conditions (deep well in the same village) show similar radon activities but differ in all others. **WVA48** is the measurement point with the fourth highest radon activity concentration, a well in the lower Mühlviertel with similar activity concentrations like WVA71. The measurement point with the fifth highest radon activity concentration is the private well **H1** in central Mühlviertel, which also shows the second highest ^{210}Pb and ^{226}Ra activity concentration. This figure differs from the results in the lower Mühlviertel discussed above. Gross alpha and gross beta are below decision limit, but higher ^{3}H, ^{226}Ra and ^{210}Pb were detected. Geological dependencies will be discussed in Chapter 15. **HEIL5** is the "sacred well" in the same village as WVA70, WVA71 and WVA73 and has the sixth highest radon activity concentration. Although it is located in the same geological region it shows slightly different results, maybe because it is a surface spring and not a drilled deep well (see Chapter 15.11). In the context of detected higher radon activity concentrations **H12** is illustrated, because private wells are of interest because of dose assessment (Chapter 16). The private well is also located in the lower Mühlviertel, but no other high nuclide activity concentrations were detected.

One of the above mentioned measurement points which attracted attention in Chapter 8 is **WVA55**. This deep well located in the neighbouring village of WVA70 in the lower Mühlviertel shows no particularly high radon activity concentration but the highest ^{210}Pb, ^{210}Po and ^{226}Ra activities and the second highest gross alpha activity.

The third highest ^{226}Ra activity concentration after WVA55 and H1 was detected at a ”sacred well” in southern Upper Austria (**HEIL11**), the radon activity is very low at this point and some nuclides were not analyzed. Two other measurement points in southern Upper Austria have ^{226}Ra activity concentrations in class 2 (**WVA95** and **H27**). Both have very low radon activity concentrations, WVA95 has ^{238}U activity concentration above decision limit, H27 ^{210}Po and ^{210}Pb activity concentrations above decsions limit. As mentioned in Chapter 8, detected higher ^{226}Ra activity concentrations may origin from a better decision limit in this sampling phase and should not be interpreted as higher ^{226}Ra potential in southern Upper Austria. More measurements are necessary to verify the results.

WVA31, a spring in upper Mühlviertel shows the third highest ^{210}Pb activity concentration after WVA55 and H1, but the activity concentration already is one magnitude lower. But this sampling point has ^{222}Rn and ^{210}Po activity concentrations in class 3 and ^{210}Pb and gross beta concentrations in class 2. Measurement points with medium to high result like these should be tested for their total dose (see Chapter 16.3).

The measurement point with the second highest ^{210}Po activity concentration after WVA55 is **WVA47**, a reservoir with water from different wells in the lower Mühlviertel. WVA47 shows a ^{210}Pb activity concentration below decision limit, although it has a high ^{210}Po activity concentration. It also shows a rather high radon activity concentration and ^{238}U in class 2. ^{210}Po, ^{238}U and ^{222}Rn are α-emitters, which correlates to the class 3 gross alpha activity concentration at this point. The activity concentrations of the β-emitters ^{228}Ra, ^{210}Pb and ^{3}H are below decision limit, as the gross beta activity at WVA47 is.

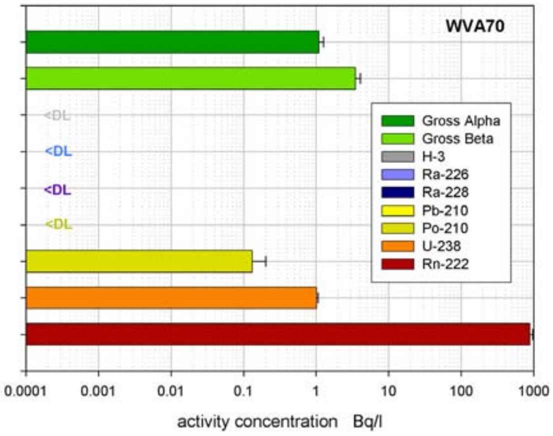
The three measurement points with the highest ^{228}Ra activity concentrations are totally different ones than from other nuclides. It seems that ^{228}Ra has no direct correlation to other nuclides. But a closer look at **H14** – a private well in the north of lower Mühlviertel with the highest ^{228}Ra activity concentration – shows that except for ^{226}Ra all nuclides have also rather high activity concentrations (class 2 or 3). But the measurement points with the second and third highest ^{228}Ra activity concentration (**WVA86** and **WVA97**) – a well and a spring in southern Upper Austria – have low activity concentrations, especially for radon, due to geology (Chapter 15).

Except WVA55 the same four measurement points have the highest gross alpha and ^{238}U activity concentration (**WVA70**, **WVA68**, **WVA69**, **WVA83**). WVA55 has the second highest gross alpha activity concentration and also the seventh highest

^{238}U activity concentration. So it seems that there is a clear correlation between ^{238}U and gross alpha (see below). **WVA68** and **WVA69** are samples before and after deacidification from mixed water from two wells located in the same village in the lower Mühlviertel as WVA70. In Chapter 12 the impact of water treatment on radionuclides in drinking water will be surveyed. The activity concentrations of WVA68 and WVA69 are nearly the same and show the same figure like WVA70, only the single nuclide activity concentrations are a factor 2 to 3 lower. It seems that the similar nuclide activity concentration distribution is dependent on geology (Chapter 15). At **WVA83**, a deep well also in the lower Mühlviertel, similar activity concentrations like WVA68 and WVA69 were detected, except ^{222}Rn, which is about 6 times lower. This demonstrates that there is seemingly no clear correlation between ^{222}Rn and ^{238}U (see below).

H6 and **H9**, two private wells in the same village of H1 in the central Mühlviertel show the second and third highest gross beta activity concentration after WVA70. At H6 ^{228}Ra, ^{210}Po and ^{210}Pb were not analyzed and ^{3}H activity concentration is below decision limit, so no direct correlations with gross beta are detectable. At H9 all nuclides were analyzed, but ^{3}H and ^{228}Ra are below decision limit and ^{210}Pb is in class 2. So also no direct correlations to gross beta are detectable. One might notice the totally different figure of the wells H6 and H9 and the above discussed H1, which shows very high radon activity concentration but a gross beta activity concentration below decision limit. This may be explained by the very heterogeneous geology in this region, which will be surveyed in Chapter 15.

The measurement points with the highest ^{3}H activity concentrations are totally different ones from the discussed above – **WVA13**, a drilled well in the north of lower Mühlviertel and **WVA23**, a reservoir in the Innviertel (Danube fault). In both cases the activity concentration of all other nuclides is low. The measurement point with the third highest ^{3}H activity concentration is a spring in the north of ”lower Mühlviertel” (**WVA5**). Noticeable – another four springs and wells located in the same village as WVA55 are among the ten measurement points with highest ^{3}H activity concentration, which all show similar nuclide activity concentrations. But the detected highest ^{3}H activity concentrations are in the order of some Bq/l.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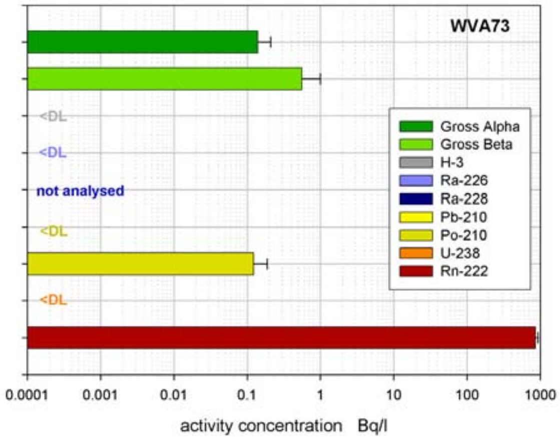
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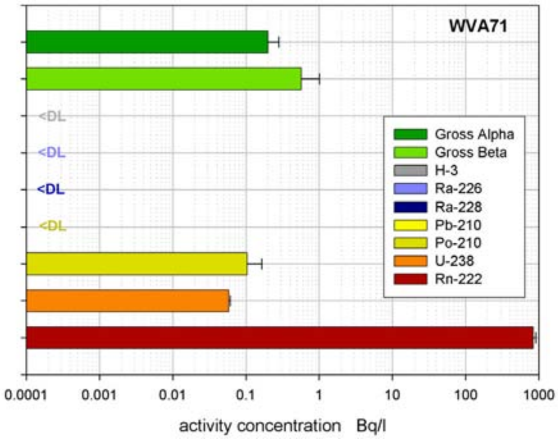
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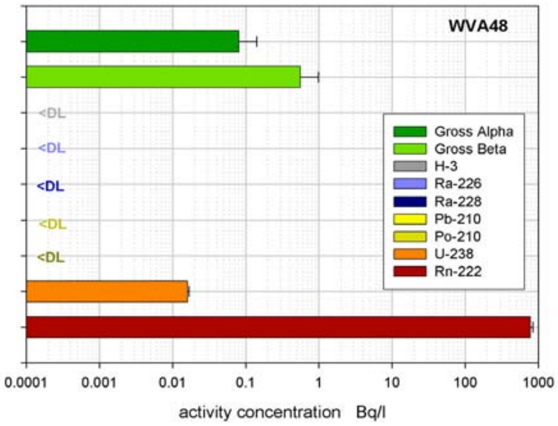
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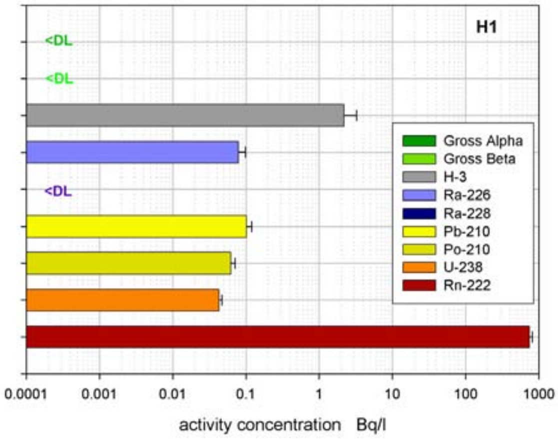
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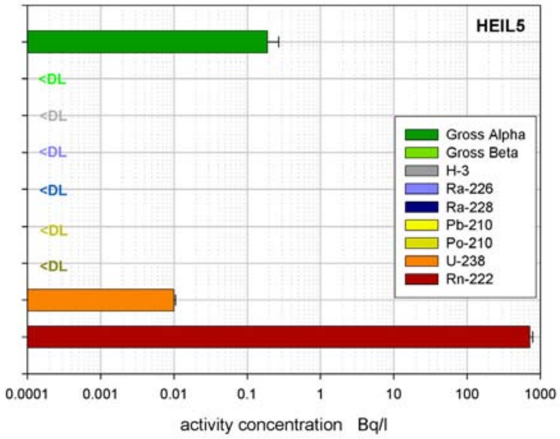
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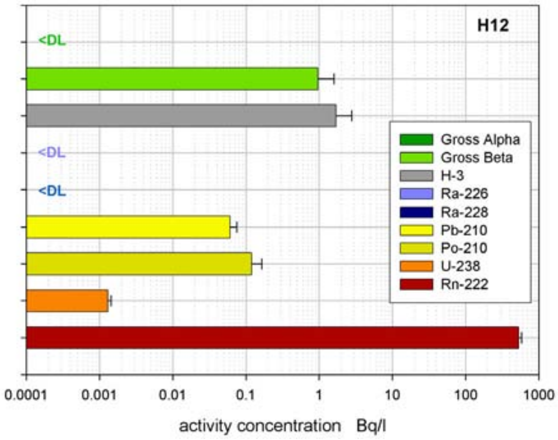
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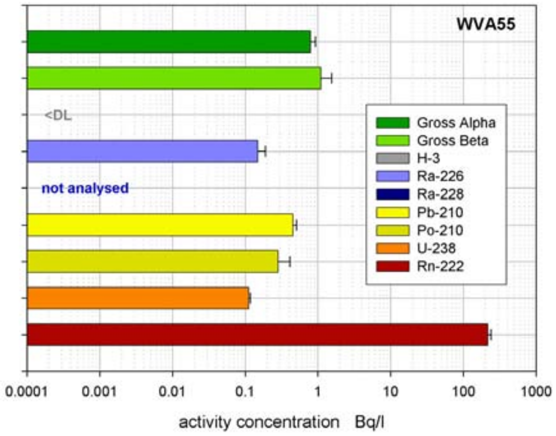
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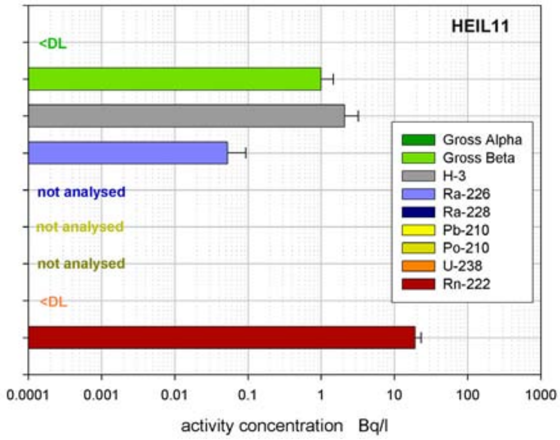
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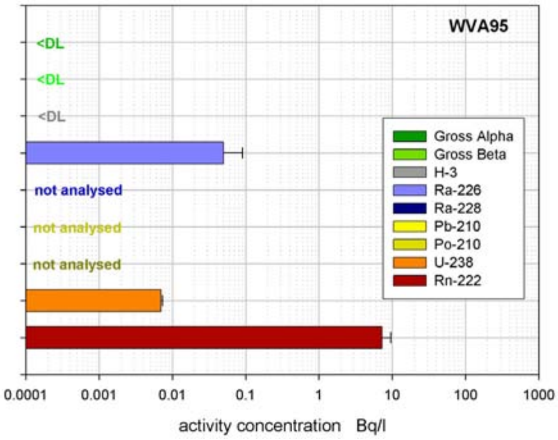
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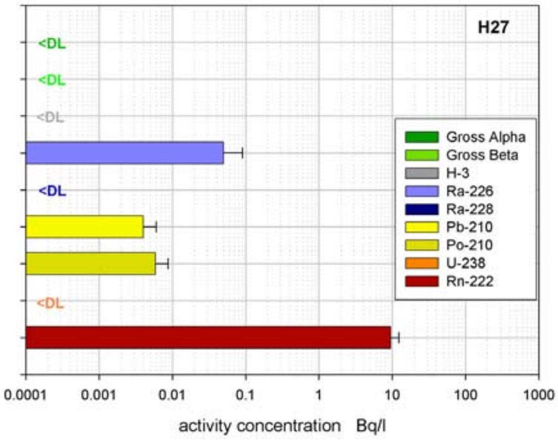
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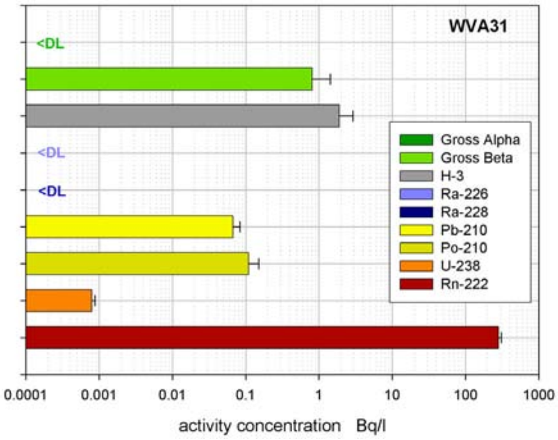
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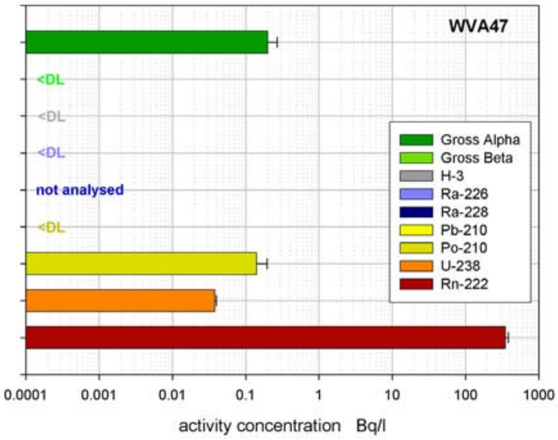
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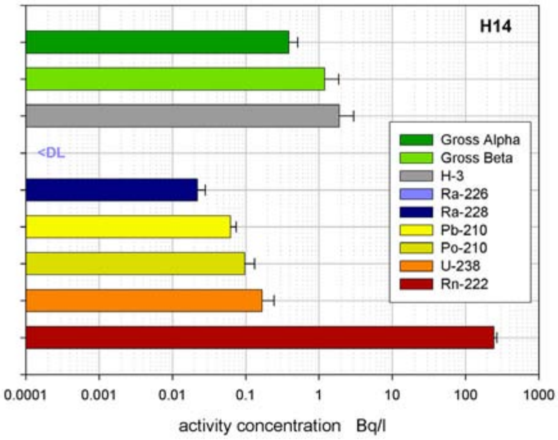
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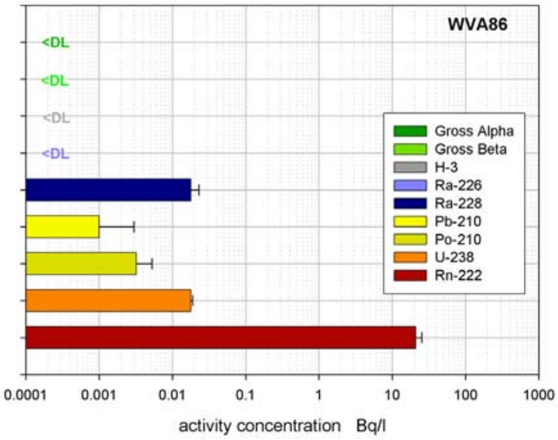
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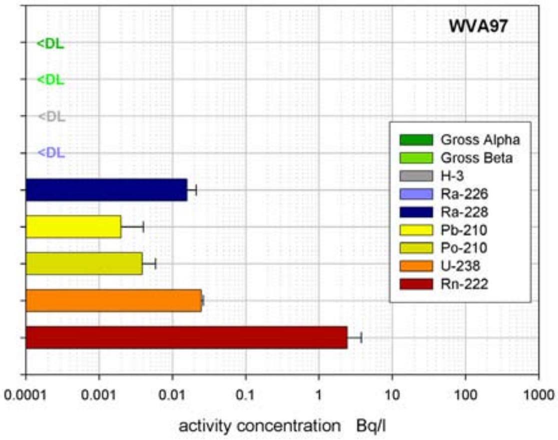
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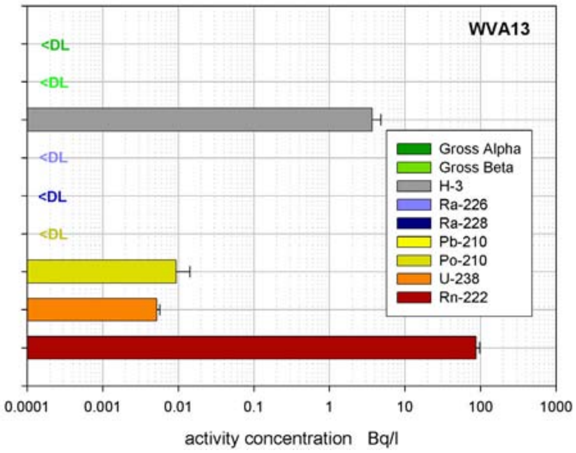
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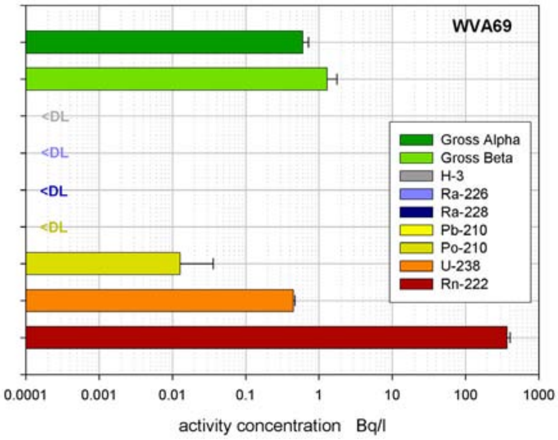
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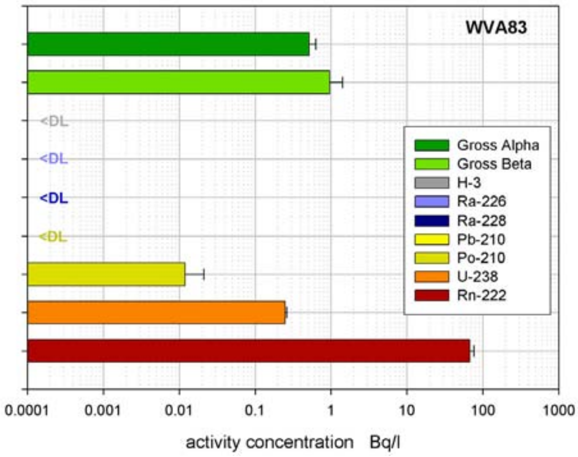
activity concentration Bq/l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Gross Beta
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Ra-226
Ra-228
Pb-210
Po-210
U-238
Rn-222
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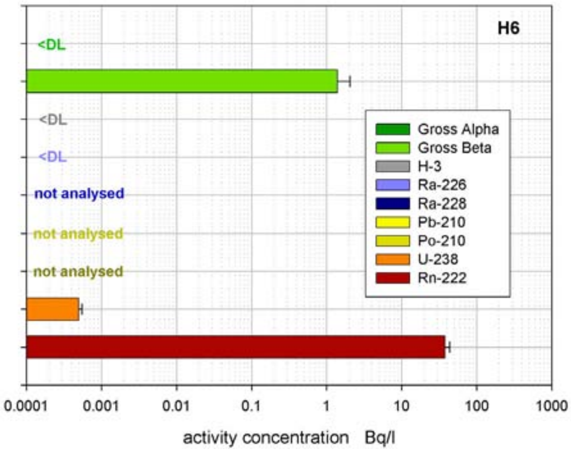
0.001
0.01
0.1
1
10
100
1000
activity concentration Bq/l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not analysed
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Gross Beta
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10
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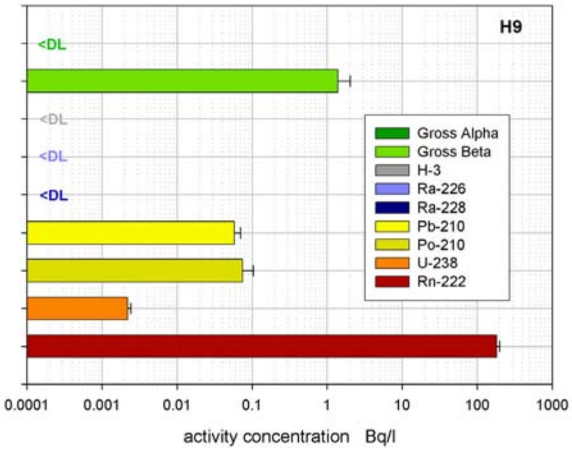
activity concentration Bq/l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Gross Beta
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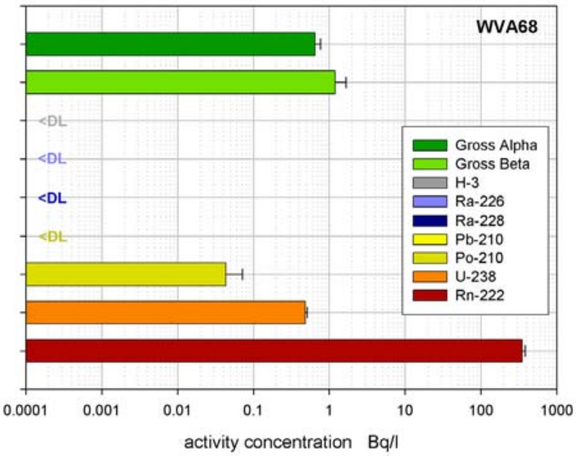
0.01
0.1
1
10
100
1000
activity concentration Bq/l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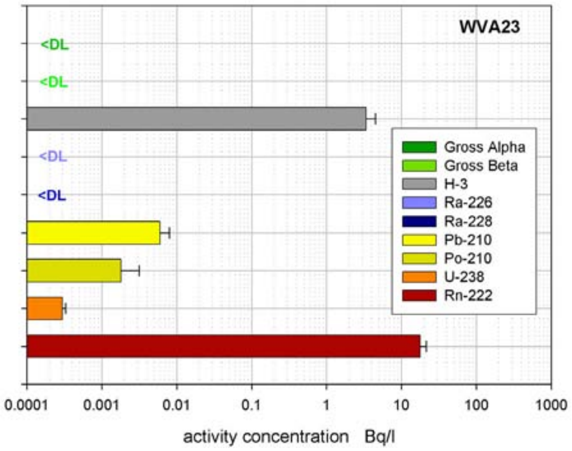
0.01
0.1
1
10
100
1000
activity concentration Bq/l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Po-210
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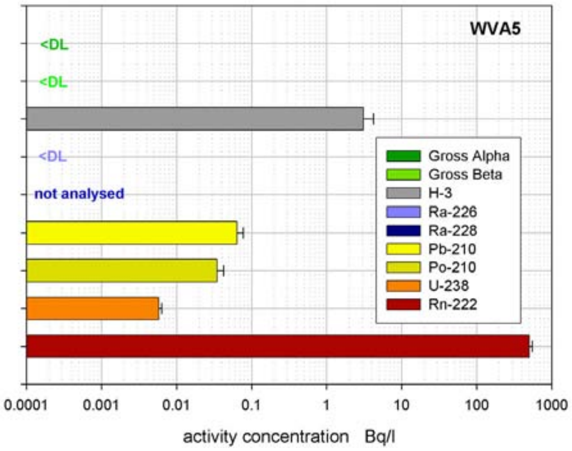
0.001
0.01
0.1
1
10
100
1000
activity concentration Bq/l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Ra-228
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Po-210
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Rn-222
0.0001
0.001
0.01
0.1
1
10
100
1000
activity concentration Bq/l

11.3 Survey of General Nuclide Correlations

The nuclide activity concentration results at single measurement points demonstrate **some nuclide correlations** and **some non-correlations**. As described above it is of major interest to search for general nuclide correlations. In the following figures these possible correlations are demonstrated and tested. As mentioned in previous chapters not all nuclides were analyzed at every measurement point. So the number of comparable measurement points is often limited and additionally many measured activity concentrations are below decision limit. In the figures these activity concentrations above decision limit are set to zero. The **correlations** between two nuclides are tested with a **linear regression** and the **method of sum of squares**. In the figures the linear regression with a 95% confidence interval is graphed and the r^2 value is given. r^2 is the **coefficient of determination**, the most common measure of how well a regression model describes the data. The coefficient of determination has a value between zero and one without unit. The closer r^2 is to one, the better the independent variables predict the dependent variable. r^2 equals zero when the values of the independent variables do not allow any prediction of the dependent variables, and equals one when the dependent variables can be perfectly predicted by the independent ones. So an r^2 value of zero means that knowing X does not allow to predict Y. There is no linear relationship between X and Y, and the best-fit line is a horizontal line going through the mean of all Y values. An r^2 value of 1 means that all points are located exactly on a straight line with no scatter, and knowing X allows to predict Y perfectly. (SPSS Inc., 1997a,b)

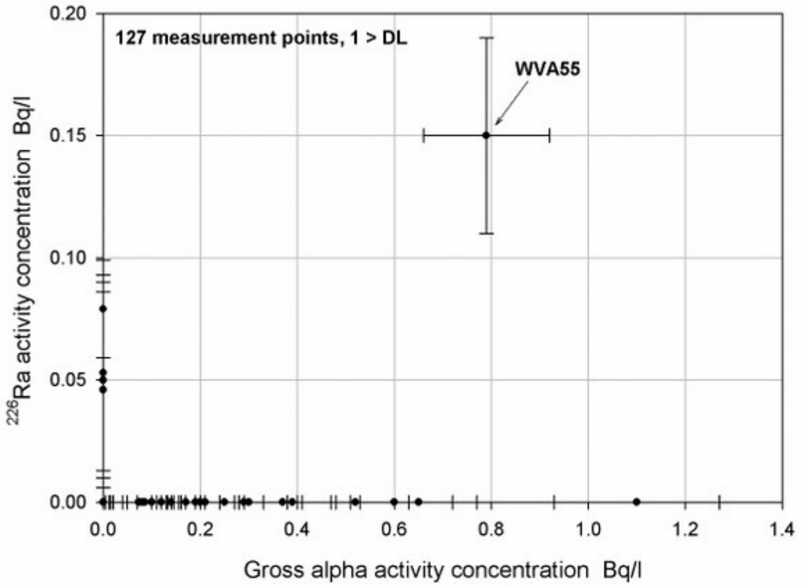

Figure 11.4: *Correlation between gross alpha and* ^{226}Ra

First, for each **nuclide correlation survey** all measurement points at which both nuclides were analyzed are determined and graphed including values below decision limit. In the cases where enough measurement points of both nuclides are above decision limit, also a linear regression is determined only with these points. Sometimes single measurement points have clearly higher activity concentrations, in these cases the linear regressions are also tested without these values. These single measurement points are indicated with their sampling number, to relate with the figures in the first part of this chapter and with other results (geology, heavy metals, . . .). In each figure the numbers of measurement points at which both nuclides were analyzed and the numbers of measurement points at which both nuclides have activity concentrations above decision limit are indicated.

First for **correlations between gross alpha and α-emitting nuclides** (^{222}Rn, ^{226}Ra, ^{238}U, ^{210}Po) and **gross beta and β-emitting nuclides** (^{228}Ra, ^{210}Pb, ^{3}H) are searched.

127 samples were analyzed for **^{226}Ra and gross alpha** and only one of these measurement points is above decision limit both ^{226}Ra and gross alpha (Figure 11.4). This point is WVA55 (see above). No correlation can be detected, because too few measurement points are above decision limit.

128 samples were analyzed for **^{222}Rn and gross alpha**, 27 are above decision limit of both ^{222}Rn and gross alpha (Figure 11.5). The activity concentrations of the nuclides show an r^2 value of 0.17, which cannot be interpreted as a real significant linear correlation, but as a recognizable tendency. Noticeable is measurement point WVA70 (see above) with a higher ^{222}Rn and a clearly higher gross alpha activity concentration. A linear regression evaluation without taking into account WVA70 reduces the r^2 value clearly to 0.083.

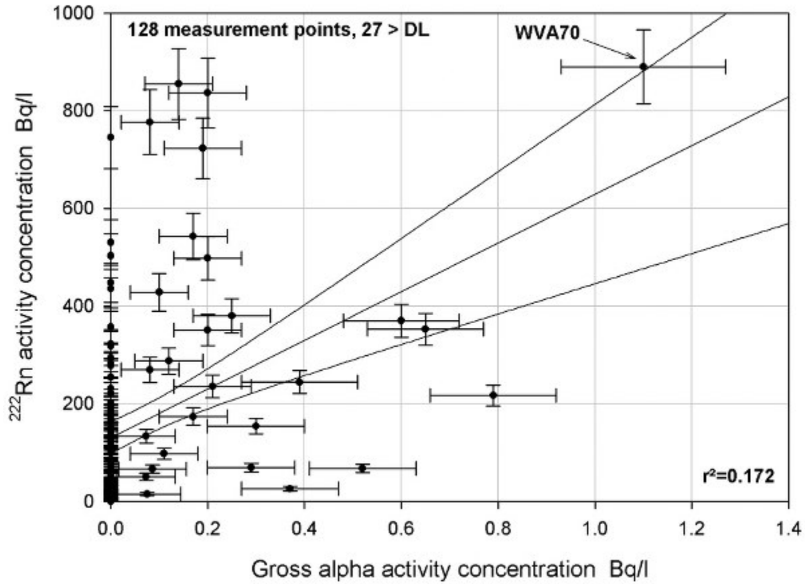

Figure 11.5: *Correlation between gross alpha and ^{222}Rn*

In Figure 11.6 only the ^{222}Rn and gross alpha activity concentrations above decision limit are graphed, which leads to a totally different linear regression. No correlation or tendency can be detected in this case (r^2=0.031). This big discrepancy may be due to the fact that no ^{222}Rn activity concentrations below decision limit exist, but many higher activity concentrations, in contrast to generally rather low gross alpha activity concentrations.

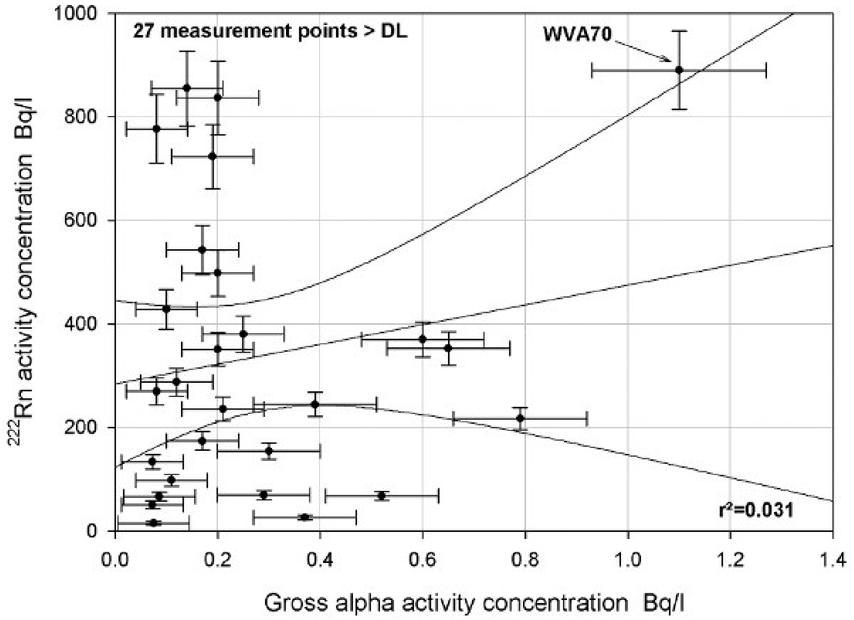

Figure 11.6: *Correlation between gross alpha and ^{222}Rn taking into account only activity concentrations above decision limit*

129 samples were detected for ^{238}**U and gross alpha**, 20 are above decision limit of both ^{238}U and gross alpha (Figure 11.7). The activity concentrations of the nuclides show a significant linear correlation (r^2=0.728). Again WVA70 is noticeably higher and a linear regression calculation without this point reduces the r^2 value to 0.638. But also with this reduced r^2 value the ^{238}U and gross alpha activity concentrations can be interpreted to have a **significant linear correlation**. Evaluation of a linear regression with activity concentrations above decision limit leads to similar results (r^2=0.757). Without taking into account WVA70 the r^2 value reduces to 0.581. The significant correlation between ^{238}U and gross alpha was already supposed in the first part of this chapter, because the same measurement points shows the highest ^{238}U and gross alpha activities. It seems that ^{238}U is the major impact to an enhanced gross alpha activity concentration. But the correlation between gross alpha and ^{238}U cannot be generalized yet, because only 20 measurement points were above decision limit and more measurements are necessary.

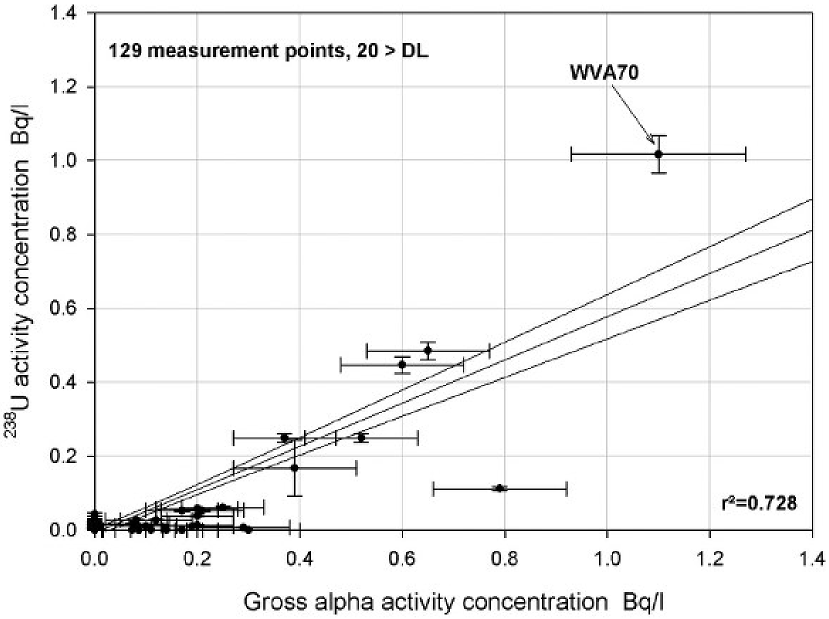

Figure 11.7: *Correlation between gross alpha and* ^{238}U

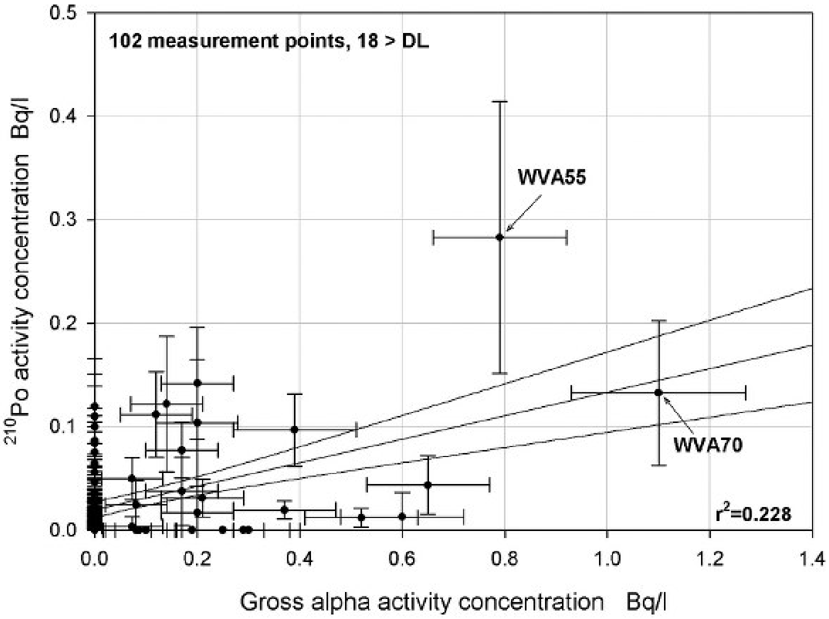

Figure 11.8: *Correlation between gross alpha and* ^{210}Po

102 samples were analyzed for ^{210}**Po and gross alpha**, 18 are above decision limit of both ^{210}Po and gross alpha (Figure 11.8). The activity concentrations of the nuclides show a slight linear correlation with an r^2 value of 0.23. This value decreases to 0.139 if only the 18 measurement points with activity concentrations above deci-

sion limit are considered. Again the measurement points WVA70 and WVA55 have noticeably higher activity concentrations and a calculated linear regression disregarding these two points result in an r^2 value of 0.068 and no linear correlation exists.

79 samples were analyzed for ^{228}**Ra and gross beta**, 5 are above decision limit of both ^{228}Ra and gross beta (Figure 11.9). The calculated r^2 value of the linear regression is 0.005 and no correlation exists. WVA70 has a clearly higher gross beta activity concentration, but a ^{228}Ra activity concentration above decision limit. Estimating a linear regression without taking into account this measurement point decreases the r^2 value. Taking into account only the 5 activity concentrations above decision limit also reduces the r^2 to 0.0002. But there are again far too few measurement results above decision limit to generalize statements about correlations.

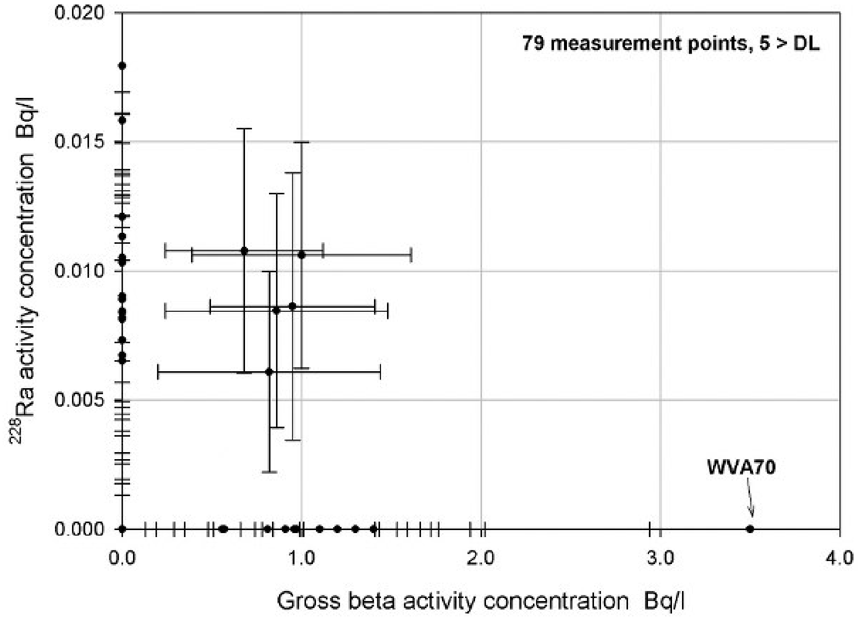

Figure 11.9: *Correlation between gross beta and ^{228}Ra*

107 samples were analyzed for ^{210}**Pb and gross beta**, 14 are above decision limit of both ^{210}Pb and gross beta (Figure 11.10). The calculated r^2 value of the linear regression is 0.043 and no correlation exists. Again WVA70 has a clearly higher gross beta activity concentration, but a ^{210}Pb activity concentration above decision limit and WVA55 has a noticeable higher activity concentration of ^{210}Pb and gross beta. Estimating a linear regression, only taking into account activity concentrations above decision limit, results in a similar r^2 value of 0.048. This value enhances to 0.11 by disregarding the measurement point WVA55, but nevertheless no significant correlation exist.

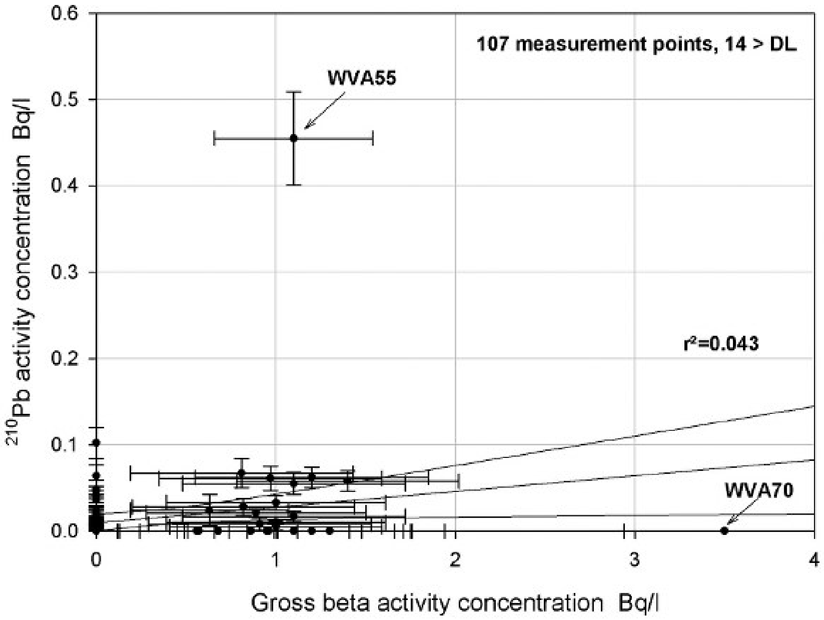

Figure 11.10: *Correlation between gross beta and ^{210}Pb*

129 samples were analyzed for **^{3}H and gross beta**, 15 are above decision limit of both ^{3}H and gross beta (Figure 11.11). The calculated r^2 value of the linear regression is 0.002 and no correlation exists. Again WVA70 has a clearly higher gross beta activity concentration, but an ^{3}H activity concentration above decision limit. Estimating a linear regression, only taking into account activity concentrations above decision limit, results in a much higher r^2 value of 0.30, which means a slightly correlation between ^{3}H and gross beta (Figure 11.12). This big discrepancy between the r^2 values may be due to the fact that all measurement points with higher Tritium values have gross beta activity concentrations below decision limit and vice versa. So this indicates that no correlation between ^{3}H and gross beta exists. If only the measurement points with activity concentrations above decision limit are considered, this fact is disregarded and only measurement points with "medium" activity concentrations are used for calculation, and no extreme values. Besides, the measured ^{3}H activity concentrations are very low and for generalisations more measurements are necessary.

But a correlation between ^{3}H and other surveyed nuclides is nevertheless not expected, because ^{3}H is built in the atmosphere and enters the ground water e.g. by precipitation and is not built in the soil and rocks as all other surveyed nuclides. The ^{3}H activity concentration can be a indicator of origin and age of the water as discussed in Chapter 2.5.4 and 7.

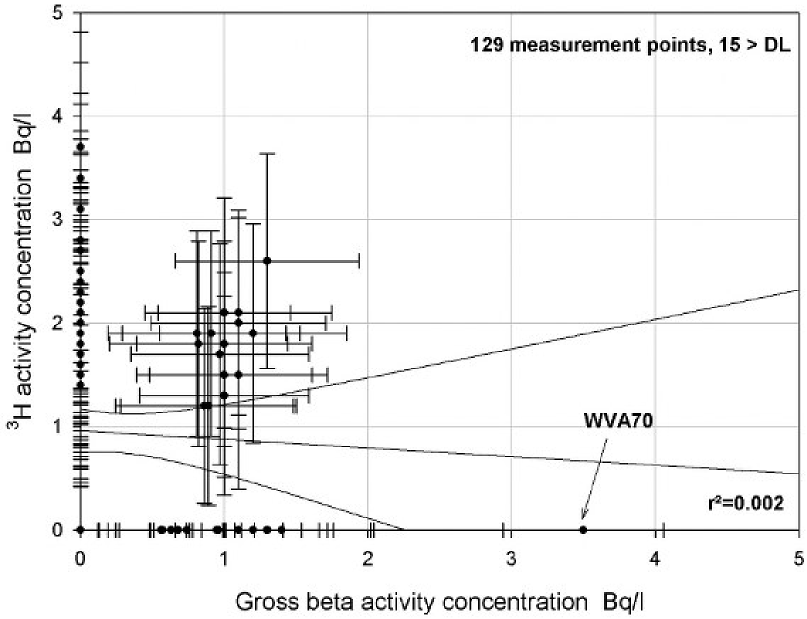

Figure 11.11: *Correlation between gross beta and* ^3H

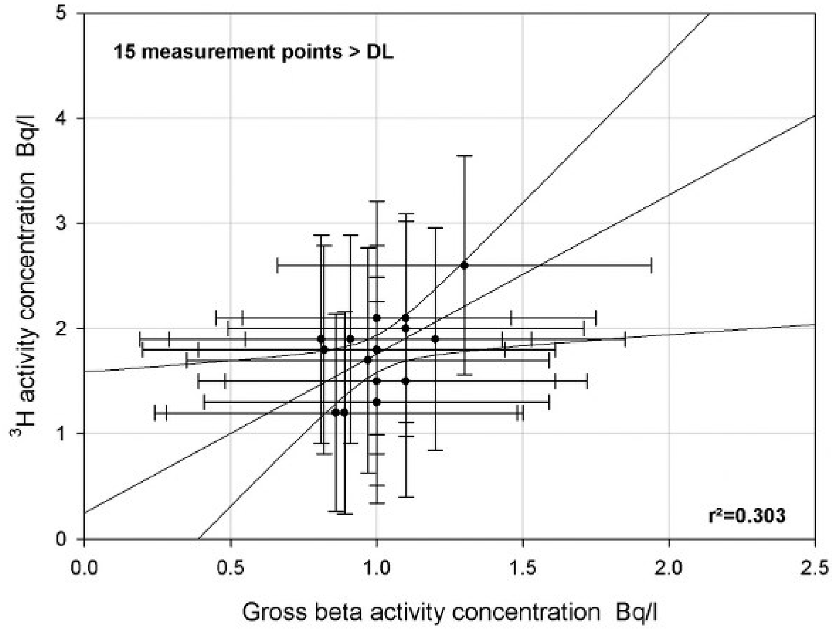

Figure 11.12: *Correlation between gross beta and* ^3H *taking into account only activity concentrations above decision limit*

In Figure 11.13 the correlation between **gross alpha and gross beta** activity concentration is demonstrated. 127 samples were analyzed for gross alpha and gross beta activity concentration, 12 are above decision limit of both gross alpha and gross beta. The calculated r^2 value of the linear regression is 0.234 and a slightly correlation exists. Considering only activity concentrations above decision limit for linear regression, the calculated r^2 value is 0.73, which means a good significant correlation between gross alpha and gross beta (Figure 11.14). The 95% confidence interval is a little broader in this case because of a lower number of measurement points. Disregarding WVA70, which has again a noticeable higher activity concentrations r^2 slightly enhances to 0.76. It seems that gross alpha and gross beta have a relation in drinking water, although for generalisation more results are required.

A red square in Figure 11.13 and 11.14 marks an alpha activity concentration of 0.1 Bq/l and a beta activity concentration of 1 Bq/l (class 3), which are monitoring values given in the draft of the European Commission (EC, 2005). According to the draft with gross alpha and gross beta activity concentrations below this values it can be assumed that the TID is less than 0.1 mSv/a. These values should be used for **screening measurements** to assess the situation and to decide whether nuclide specific measurements are necessary. Only one measurement point with activity concentrations above decision limits lies within this square, 11 are outside of it because of both activity concentrations. 15 measurement points are outside this square at least because of the activity concentration of gross alpha or gross beta. **So nuclide specific measurements are necessary for radiation protection reasons too, not only for scientific surveys.**

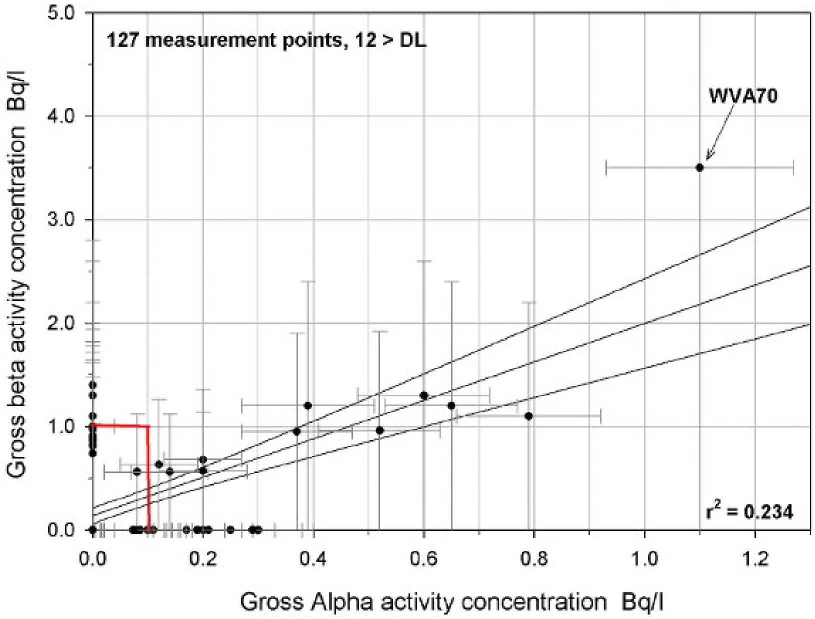

Figure 11.13: *Correlation between gross alpha and gross beta*

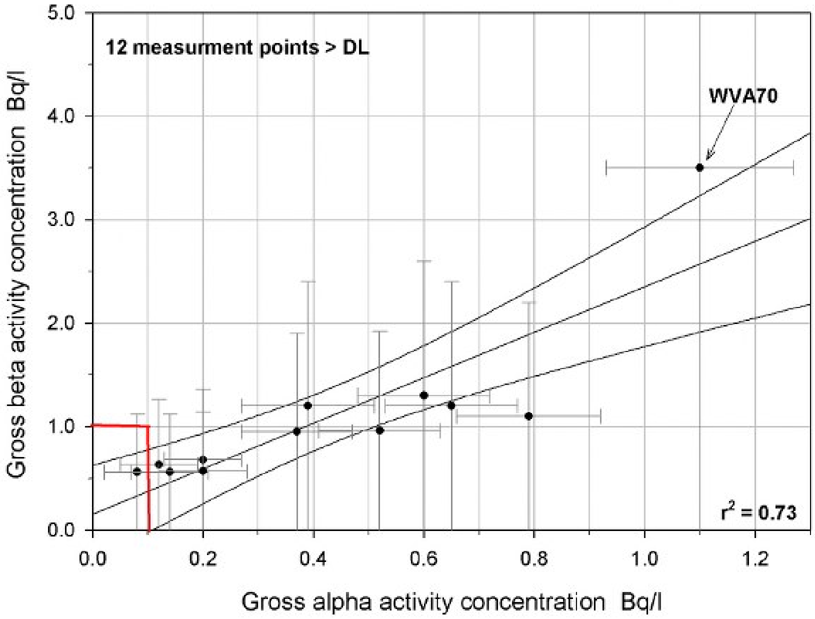

Figure 11.14: *Correlation between gross alpha and gross beta taking into account only activity concentrations above decision limit*

As mentioned above ^{226}Ra, ^{222}Rn, ^{210}Pb and ^{210}Po are all built within the ^{238}**U decay series**, so correlations are considered. In the following figures coherences between these nuclides are surveyed. First it is searched for possible relations between radon and the above mentioned nuclides.

127 samples were analyzed for ^{226}**Ra and** ^{222}**Rn**, 7 are above decision limit of both ^{226}Ra and ^{222}Rn (Figure 11.15). There are two few measurement points to carry out a good linear regression estimation, but nevertheless it was tried and results in an r^2 value of 0.002, so no correlation exists. Taking into account only the 7 measurement points with activity concentrations above decision limit yields a clearly higher r^2 value (r^2=0.16). This is a different effect than in the case of radon and gross alpha (see above). But concerning ^{222}Rn and ^{226}Ra far too few activity concentrations are above decision limit to make predications. Even a closer look at the two outstanding measurement points (H1 and WVA55) shows no direct correlation. WVA55 has by far the highest ^{226}Ra activity concentration, but a medium ^{222}Rn activity concentration. H1 has a rather high ^{222}Rn activity concentration, but not a very high ^{226}Ra concentration either. Regarding to this measurements, it seems that no correlation exists between radon and ^{226}Ra in drinking water, although this correlation would be expected because ^{226}Ra is the mother nuclide of ^{222}Rn.

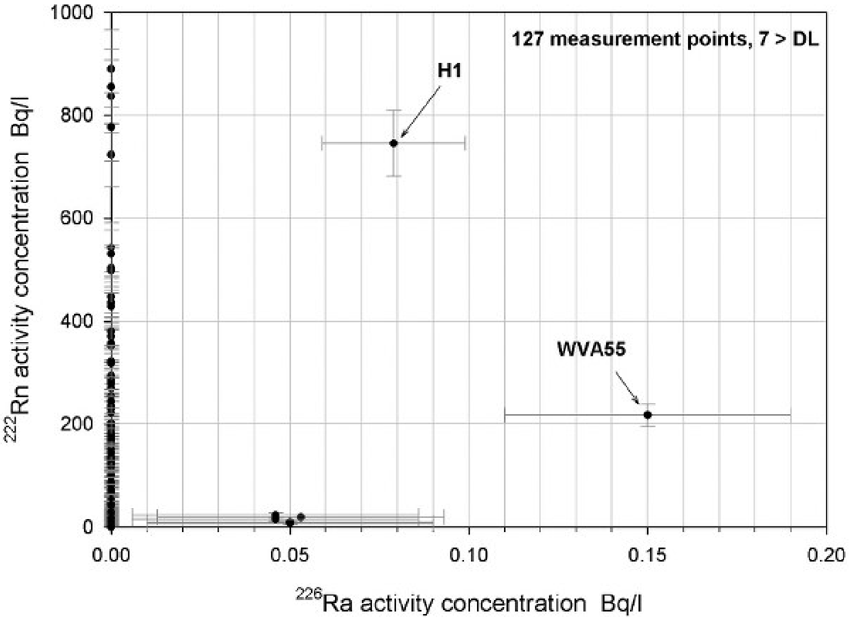

Figure 11.15: *Correlation between ^{222}Rn and ^{226}Ra*

107 samples were analyzed for ^{210}**Pb and** ^{222}**Rn**, 50 are above decision limit of both ^{210}Pb and ^{222}Rn (Figure 11.16). The calculated r^2 value of the linear regression is 0.009, which means definitely no correlation. WVA55 has a clearly higher ^{210}Pb activity concentration and is marked in the figure. Taking into account only measurement points with activity concentrations above decision limit for linear regression, the r^2 value rises to 0.107. Disregarding the "outlier" measurement point WVA55 in the linear regression calculation result in an r^2 value of 0.592 (Figure 11.17). This would point to a slight correlation between ^{210}Pb and ^{222}Rn, which would be suggesting because ^{210}Pb is a radon progeny. But for generalization more measurements are necessary, because no correlation was detected in this case without modification of the values.

Lehmann et al. (1997) and Reichelt et al. (1994) reported that in a drinking water with an ^{222}Rn activity concentration above 100 Bq/l a radon induced ^{210}Pb, ^{210}Bi and ^{210}Po activity concentration of 5 mBq/l can be assumed. In the analyses of the AGES the radon influence is corrected (see Chapter 5.2.2 but other sources of ^{210}Pb and ^{210}Po must exist, which do not result in a clear correlation between radon and its progenies.

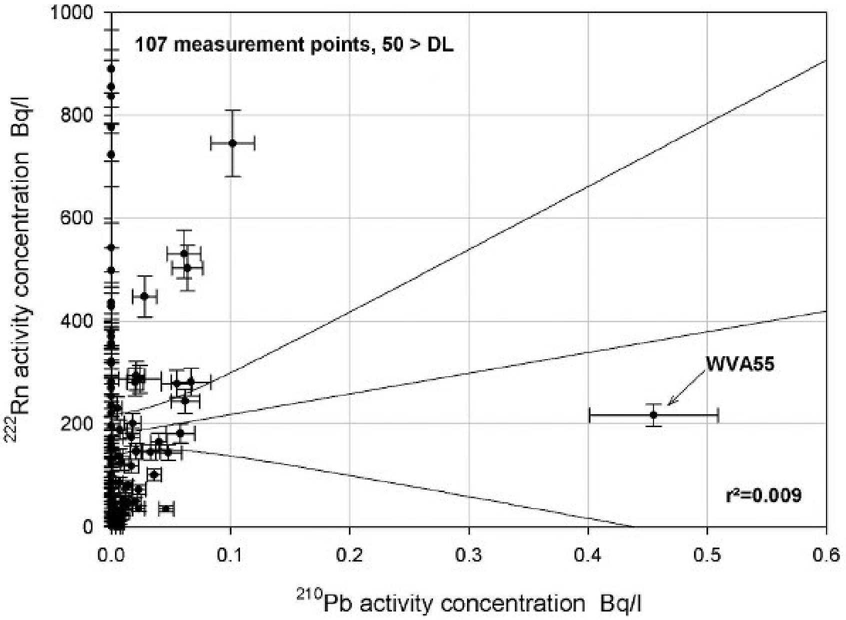

Figure 11.16: *Correlation between ^{222}Rn and ^{210}Pb*

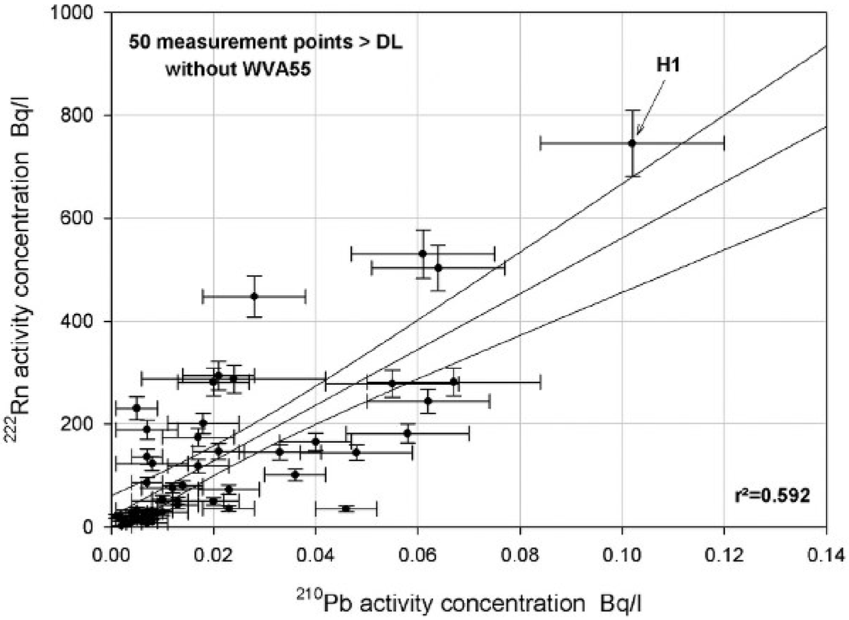

Figure 11.17: *Correlation between ^{222}Rn and ^{210}Pb taking into account only activity concentrations above decision limit and disregarding WVA55*

104 samples were analyzed for **^{210}Po and ^{222}Rn**, 90 are above decision limit of both ^{210}Po and ^{222}Rn (Figure 11.18). The calculated r^2 value of the linear regression is 0.184, which means no correlation, but maybe again a recognizable tendency. Taking into account only measurement points with activity concentrations above decision limit for linear regression, the r^2 value rises to 0.284. In this case many measurement points are above decision limit, but no direct correlations, but also no real exceptions within the measurement points are detectable.

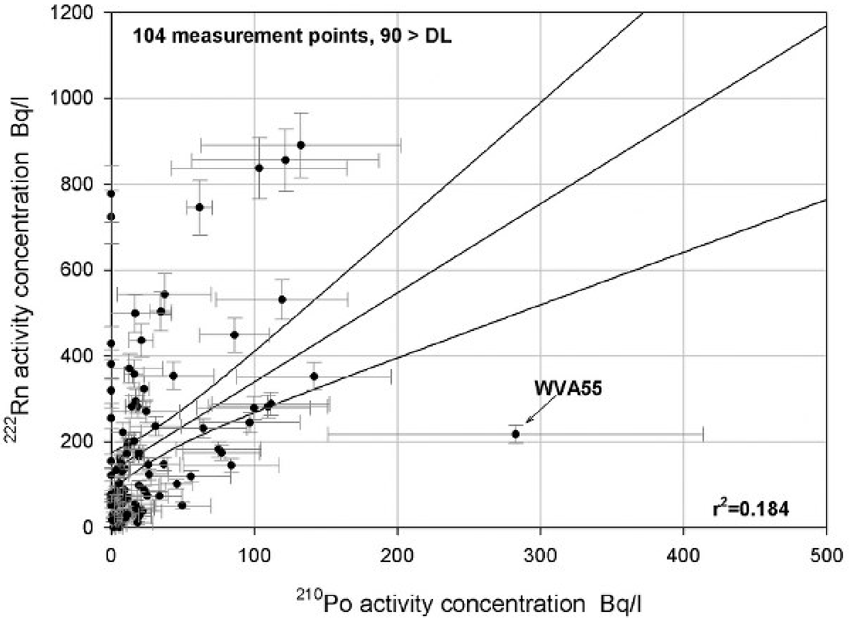

Figure 11.18: *Correlation between ^{222}Rn and ^{210}Po*

128 samples were analyzed for **^{238}U and ^{222}Rn**, 101 are above decision limit of both ^{238}U and ^{222}Rn (Figure 11.19). The calculated r^2 value of the linear regression is 0.133, so no significant correlation exists. Taking into account only measurement points with activity concentrations above decision limit for linear regression estimation results in nearly the same r^2 value (r^2=0.144). WVA70 has a noticeable higher radon and ^{238}U activity concentration – disregarding this measurement point within linear regression estimation yields a much lower r^2 value of 0.026. So it seems that a correlation tendency between radon and ^{238}U origins only from this measurement point WVA70. Generally the measured uranium activity concentrations are rather low, not dependent on the radon activity concentrations. The few measurement points with a little higher ^{238}U activity concentrations show rather low radon activities. So no correlation between ^{238}U and ^{222}Rn is detected within this analysis – for generalization more measurements will be necessary.

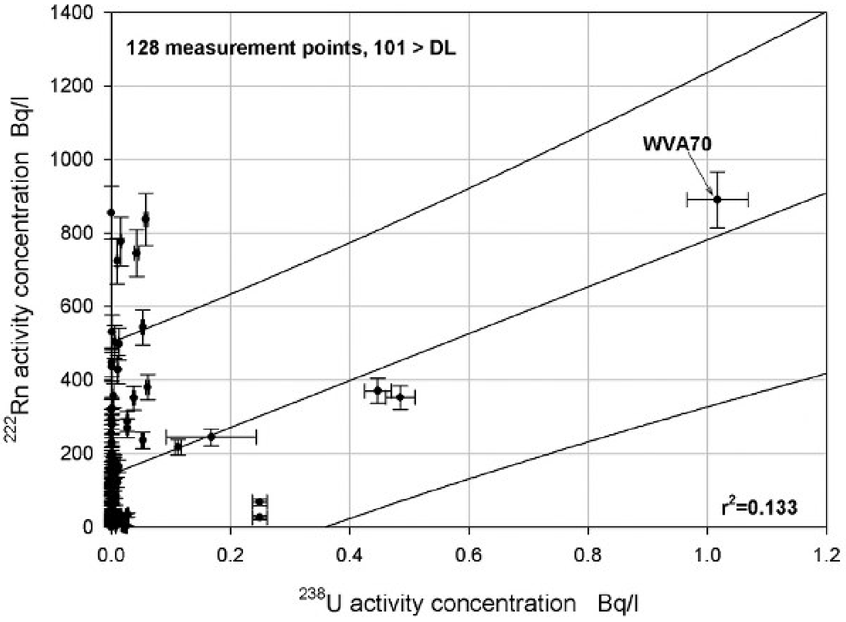

Figure 11.19: *Correlation between ^{222}Rn and ^{238}U*

These figures and regression estimations show that ^{222}**Rn in drinking water does not correlate significantly to other nuclides of the** ^{238}**U decay series**. Only a slight correlation between ^{210}Pb and ^{222}Rn could be observed. According to the results of this thesis, the fast and easy **determination of radon activity concentration is not appropriate to completely evaluate the ”risk” in drinking water originates from other nuclides**. Therefore, also analyses of other nuclides are necessary, because they are dose relevant (especially ^{210}Po and ^{210}Pb – see Chapter 16).

As discussed in other chapters ^{210}Po and ^{210}Pb are dose relevant and therefore they should be analyzed in drinking water. As described in Chapter 5.2.2, these analyses are rather complicated and time-consuming. Therefore it would be a great advantage if only one of these nuclides had to be analyzed directly, because a significant correlation between the nuclides is established. So in Figure 11.20 correlation between ^{210}Po and ^{210}Pb is tested.

104 samples were analyzed for ^{210}**Pb and** ^{210}**Po**, 51 are above decision limit of both ^{210}Pb and ^{210}Po. The linear regression calculation yields an r^2 value of 0.49, which is a slightly significant correlation. Again WVA55 is noticeably higher regarding ^{210}Po and ^{210}Pb activity concentrations. Disregarding this ”outlier” measurement point the r^2 value of the linear regression results in a more than halved r^2 value (r^2=0.23). So again it seems that the correlation tendency originates from the related higher ^{210}Po and ^{210}Pb activity concentrations of WVA55. Considering only measurement points with activity concentrations above decision limit for linear regression estimation the r^2 value enhances to 0.622, and even disregarding WVA55 yields an r^2 value of 0.49 (Figure 11.21). So, although WVA55 intensifies the de-

tected correlation between ^{210}Po and ^{210}Pb, it seems that at least a **correlation tendency** exists between ^{210}Po and ^{210}Pb according to these measurements. But for generalization and above mentioned relation for evaluating the activity of one nuclide out of the other, more measurements are necessary and of interest.

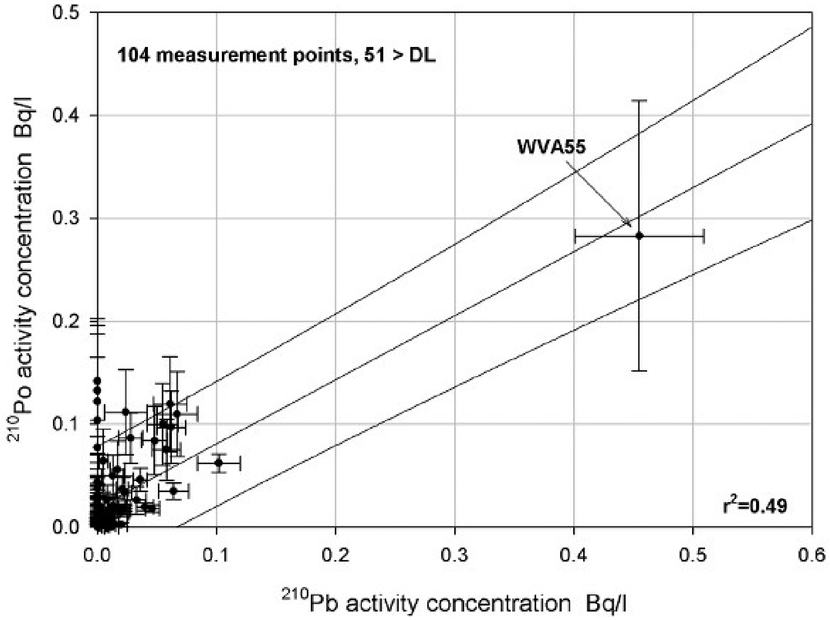

Figure 11.20: *Correlation between ^{210}Pb and ^{210}Po*

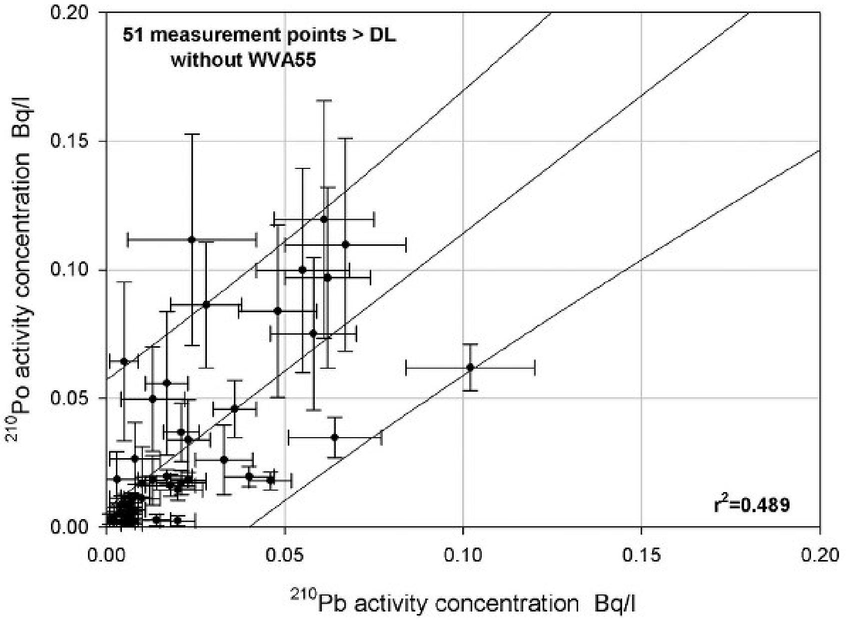

Figure 11.21: *Correlation between ^{210}Pb and ^{210}Po taking into account only activity concentrations above decision limit and disregarding WVA55*

For ^{226}Ra only 7 measurement points are above decision limit. Therefore it is difficult to correlate other nuclides with ^{226}Ra, because too few measurement points are above decision limit. Nevertheless in Figures 11.22 – 11.24 ^{226}Ra is graphed with ^{210}Po, ^{210}Pb and ^{238}U to see possible tendencies and exemptions.

107 samples were analyzed for **^{210}Pb and ^{226}Ra**, 3 are above decision limit of both ^{210}Pb and ^{226}Ra (Figure 11.22). The calculated r^2 value of the linear regression (r^2=0.685) could be interpreted as a significant correlation between ^{210}Pb and ^{226}Ra, but it represents only the related activity concentrations of ^{226}Ra and ^{210}Pb at the measurement points WVA55 and H1. For generalization and further results more measurements are necessary and of major interest.

104 samples were analyzed for **^{210}Po and ^{226}Ra**, 4 are above decision limit of both ^{210}Po and ^{226}Ra (Figure 11.23). The linear regression calculation yields an r^2 value of 0.255, which may be interpreted as a slightly correlation tendency, but not as clearly as with ^{210}Pb and ^{226}Ra. This is because measurement points with higher ^{210}Po activities exist, which have ^{226}Ra activity concentrations below decision limit. Again WVA55 has noticeable higher activity concentrations, and disregarding this measurement point, the r^2 value decreases to 0.0007. But again far too few measurement points are above decision limit to evaluate the correlation between ^{210}Po and ^{226}Ra.

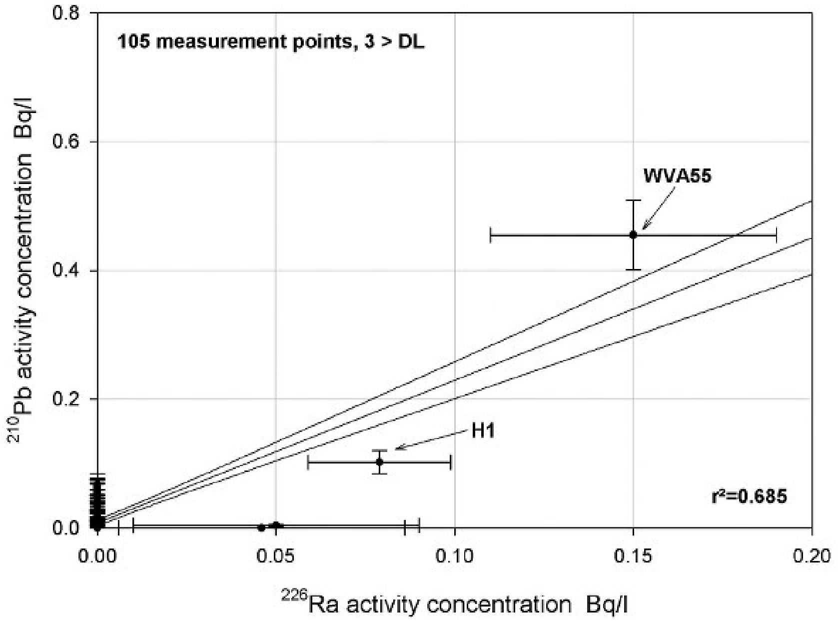

Figure 11.22: *Correlation between ^{210}Pb and ^{226}Ra*

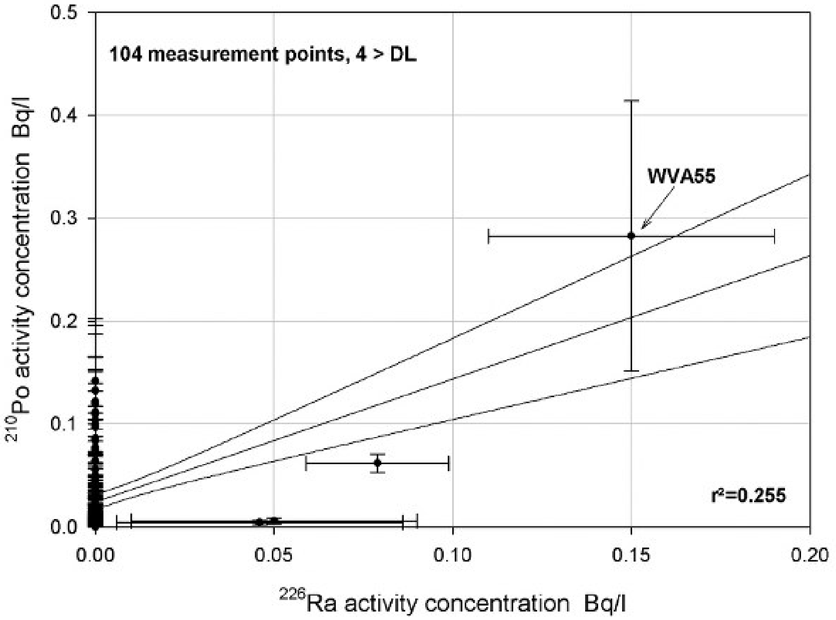

Figure 11.23: *Correlation between ^{210}Po and ^{226}Ra*

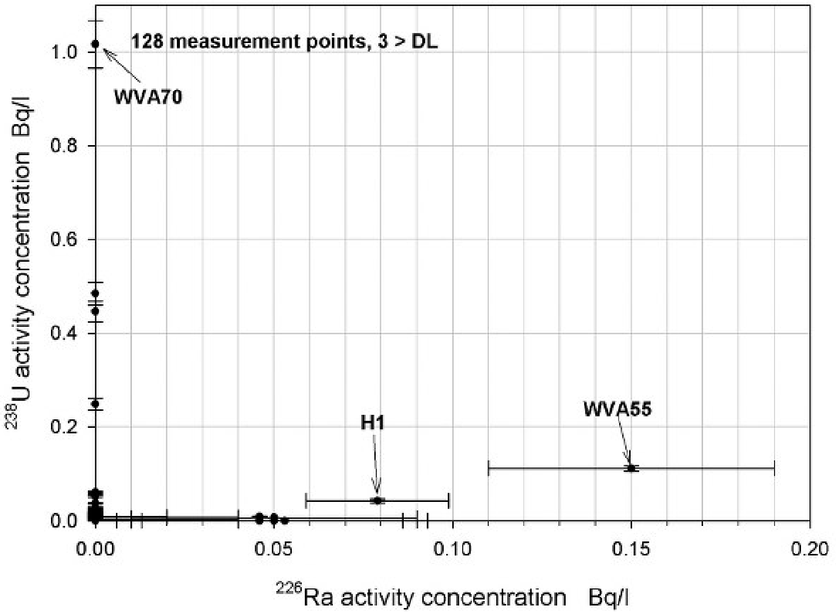

Figure 11.24: *Correlation between ^{238}U and ^{226}Ra*

128 samples were analyzed for ^{238}**U and** ^{226}**Ra**, 3 are above decision limit of both ^{238}U and ^{226}Ra (Figure 11.24). The calculated r^2 value of the linear regression (r^2=0.001) shows clearly that there is no correlation between ^{238}U and ^{226}Ra. Although also in this case WVA55 and H1 are noticeable in the figure, no correlation

is detected between ^{238}U and ^{226}Ra, because the measurement points with the by far higher ^{238}U activity concentrations (e.g. WVA70) have ^{226}Ra activities below decision limit. So no correlation exists between ^{238}U and ^{226}Ra according to the measurements in this thesis, but for generalizations more measurements are of interest. This corresponds with the results of (Jia & Torri, 2007) who also found no statistically significant correlation between the two nuclides and explained it with **different behaviour of radium and uranium in water**.

A correlation between the two radium isotopes ^{226}**Ra and** ^{228}**Ra** can not be tested, because no sample of the 81 measurement points, which were analyzed for both nuclides, is above decision limit of both ^{226}Ra and ^{228}Ra. So, further measurements are necessary for correlations interpretations.

In Figure 11.25 and 11.26 the last two possible correlations between nuclides are graphed – ^{238}U with ^{210}Po and ^{210}Pb.

106 samples were analyzed for ^{238}**U and** ^{210}**Pb**, 46 are above decision limit of both ^{238}U and ^{210}Pb (Figure 11.25). The calculated r^2 value of the linear regression (r^2=0.001) shows no correlation between ^{238}U and ^{210}Pb, because the clearly highest ^{238}U measurement point WVA70 has a ^{210}Pb activity concentration above decision limit and so are the points with the three next highest ^{238}U activity concentrations. Considering only measurement points with activity concentrations above decision limit for linear regression estimation the r^2 value increases clearly to 0.323, but this represents mainly the related slightly higher activity concentrations of ^{238}U and ^{210}Pb at the measurement point WVA55. Disregarding this point, the r^2 value decreases to 0.097. The above mentioned four highest ^{238}U activity concentrations at measurement points with ^{210}Pb activity concentrations above decision limit are an evidence for **no correlation between** ^{210}**Pb and** ^{238}**U**.

103 samples were analyzed for ^{238}**U and** ^{210}**Po**, 78 are abovedecision limit both ^{238}U and ^{210}Pb (Figure 11.26). No linear correlation is detectable between these two nuclides (r^2=0.077), even by taking into account only measurement points with activity concentrations above decision limit (r^2=0.079) and disregarding the highest ^{238}U measurement point WVA70 decreases the r^2 to 0.018.

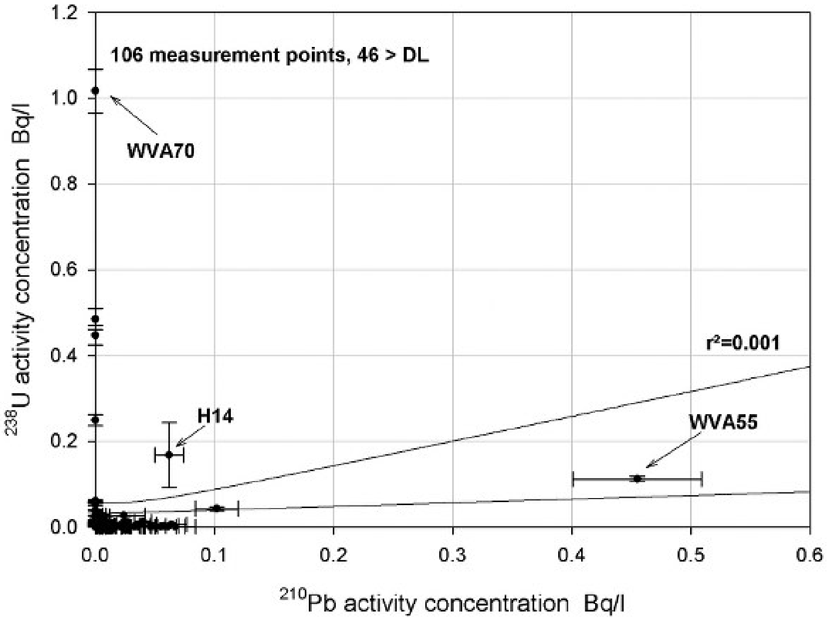

Figure 11.25: *Correlation between ^{238}U and ^{210}Pb*

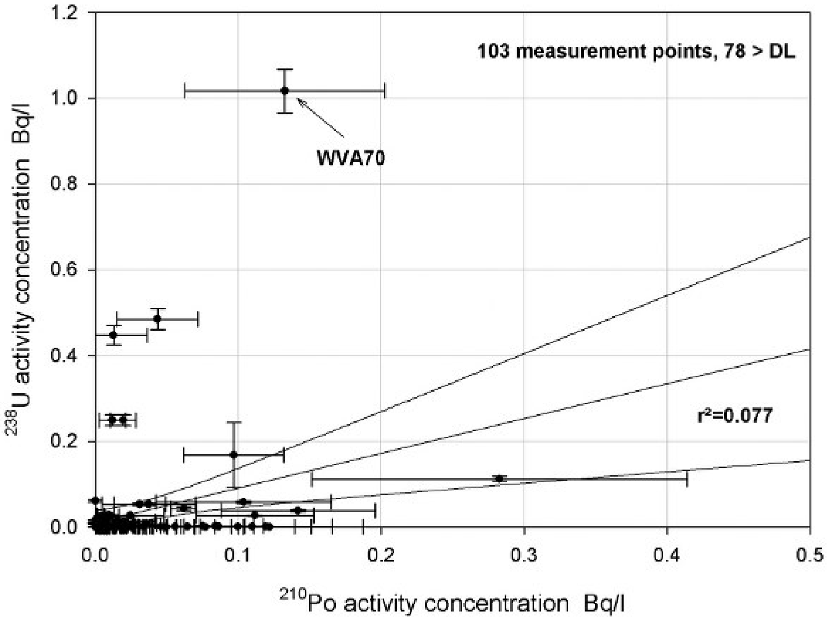

Figure 11.26: *Correlation between ^{238}U and ^{210}Po*

11.4 Survey of Nuclide Correlations in Defined Geological Regions

Within the nuclide correlation evaluations, questions arose concerning nuclide correlations in defined regions. It is interesting to analyze whether the correlations or non-correlations between two nuclides are the same in **different geological or geographic regions** and in **total Upper Austria**. Several times in this thesis a 5 regions allocation was used – 3 parts of the **Mühlviertel (lower, central, upper)**, the **Innviertel (Danube fault)** and **southern Upper Austria**. So this classification was also applied to the nuclide correlations carried out below. For most correlations between nuclides, too few measurement points are above decision limit in each region – so regression calculations are not reasonable in these cases (gross alpha, gross beta, ^{226}Ra, ^{228}Ra and ^{3}H). For ^{222}Rn, ^{238}U, ^{210}Po and ^{210}Pb nuclide correlation in different regions can be analyzed. Three correlations between these nuclides in different regions are discussed here – **^{238}U and ^{222}Rn**, because enough measurement points above decision limit exist, **^{222}Rn and ^{210}Po** also have enough measurement points and a slightly better correlation (see above) and **^{210}Po and ^{210}Pb**, which have rather few measurement points above decision limit, but are interesting because of one of the best detected linear correlations (see above).

The correlations (r^2 value) of the three pairs of nuclides in the 5 regions are listed in tables (Table 11.1 – 11.3).

Table 11.1 shows that the r^2 value of the linear regression estimation of **^{238}U and ^{222}Rn** in the region **lower Mühlviertel** is nearly the same as the one of total Upper Austria, and no significant correlation exists. The similar r^2 value is caused by the largest number of measurement points in this region and mainly by the higher activity concentration of WVA70, which is located in lower Mühlviertel. Disregarding this point reduces the r^2 value to 0.006, which also corresponds to the result of total Upper Austria. The calculated r^2 value of the linear regression in **central Mühlviertel** is very high (r^2=0.901), which means a good linear correlation between ^{222}Rn and ^{238}U in this region. This good correlation mainly depends on the measurement point H1, with a clearly higher ^{222}Rn and ^{238}U activity concentration (Figure 11.27). Disregarding this measurement point, the r^2 value reduces to 0.219, which nevertheless is clearly higher than the r^2 value of total Upper Austria measurement points. So the tendency of a linear regression between ^{238}U and ^{222}Rn in central Mühlviertel exists. In **upper Mühlviertel** and **Innviertel** no correlation can be detected, the r^2 values are very low. In southern Upper Austria the r^2 value is similar to lower Mühlviertel and total Upper Austria, but here no measurement point has noticeably higher activity concentrations, which influences the linear regression singularly. So it seems that in southern Upper Austria a generally better tendency of correlation exists than in lower Mühlviertel. This may be explained by the very low radon activity concentrations in this region, which correspond to the general rather low ^{238}U activity concentrations in total Upper Austria.

Table 11.1: *Correlation between ^{238}U and ^{222}Rn in different regions*

Region	No. of measurement points	No. of measurement points $>$ DL	Coefficient of determination (r^2)
Lower Mühlviertel	62	47	0.115
Central Mühlviertel	15	14	0.901
Upper Mühlviertel	11	11	0.003
Innviertel	14	13	0.005
Southern Upper Austria	26	16	0.148
Total	**128**	**101**	

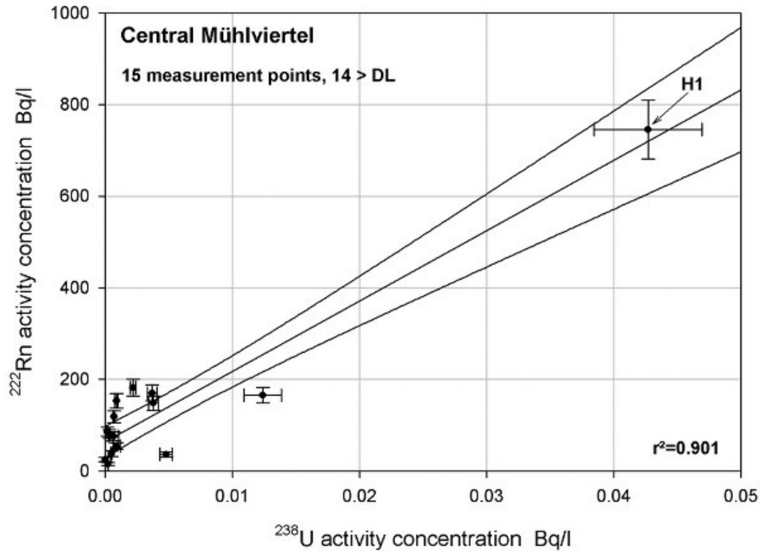

Figure 11.27: *Correlation between ^{238}U and ^{222}Rn in the region central Mühlviertel*

The linear regression estimation of ^{210}**Po and** ^{222}**Rn** in total Upper Austria results in an r^2 value of 0.184 (see above). This reasonably corresponds to the calculated r^2 value in **lower Mühlviertel** (Table 11.2), which is little lower with 0.107. In the other regions, the estimated r^2 values are clearly higher and show recognizable correlation tendencies. This may be because of a lower number of measurement points in each region. The rather high r^2 value in **upper Mühlviertel** mainly depends on the noticeably higher activity concentrations of ^{210}Po and ^{222}Rn at WVA31. Disregarding this measurement point, the r^2 value is clearly reduced to 0.006 and no correlation tendency exists. Disregarding the measurement point H1 in **central Mühlviertel** also reduces the r^2 value of this region to 0.175. Figure 11.28 shows the correlation between ^{222}Rn and ^{210}Po in **southern Upper Austria**, with an r^2 value of 0.323 and no measurement points with ”outlier” activity concentrations exist in this region.

Table 11.2: *Correlation between ^{210}Po and ^{222}Rn in different regions*

Region	**No. of measurement points**	**No. of measurement points > DL**	**Coefficient of determination (r^2)**
Lower Mühlviertel	60	49	0.107
Central Mühlviertel	13	12	0.317
Upper Mühlviertel	8	8	0.449
Innviertel	12	11	0.320
Southern Upper Austria	11	10	0.323
Total	**104**	**90**	

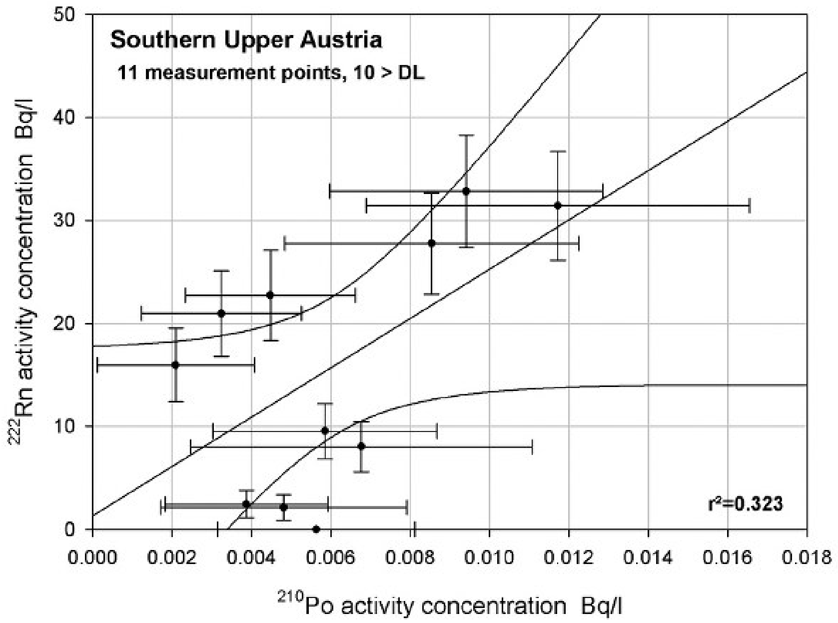

Figure 11.28: *Correlation between ^{210}Po and ^{222}Rn in the region of southern Upper Austria*

The linear regression between ^{210}**Pb and** ^{210}**Po** in the **lower Mühlviertel** yields nearly the same figure than in total Upper Austria and to an r^2 value of 0.507 (Table 11.3), which corresponds well with the one calculated for total Upper Austria (r^2=0.49). Again this slight correlation is mainly caused by WVA55, the measurement point with clearly higher ^{210}Pb and ^{210}Po activity concentrations. Disregarding this "outlier" results in an r^2 value of 0.202, which again corresponds to the one for total Upper Austria (r^2=0.23). For the other regions only less measurement points are above decision limit, nevertheless linear regression estimations were carried out and the estimated r^2 values are rather high in the regions. The highest r^2 value was detected in **upper Mühlviertel** (r^2=0.756), but only 4 measurement points are

Table 11.3: *Correlation between ^{210}Pb and ^{210}Po in different regions*

Region	No. of measurement points	No. of measurement points $>$ DL	Coefficient of determination (r^2)
Lower Mühlviertel	60	22	0.507
Central Mühlviertel	13	8	0.597
Upper Mühlviertel	8	4	0.756
Innviertel	12	9	0.270
Southern Upper Austria	11	8	0.577
Total	**104**	**51**	

above decision limit of both ^{210}Po and ^{210}Pb. Again WVA31 is an "outlier" in this region according to higher activity concentration, and disregarding this measurement point again reduces the r^2 value clearly to 0.073. Figure 11.29 shows the correlation between ^{210}Po and ^{210}Pb in the **Innviertel** (r^2=0.27). The measurement point H22 has noticeable higher activity concentrations, but disregarding this point enhances the r^2 value slightly to 0.362 in opposite to the above mentioned cases. Nevertheless a significant correlation does not exist. In **southern Upper Austria** a slight correlation is detected (r^2=0.577), again without any "outlier" measurement points and generally rather low ^{210}Po and ^{210}Pb activity concentrations.

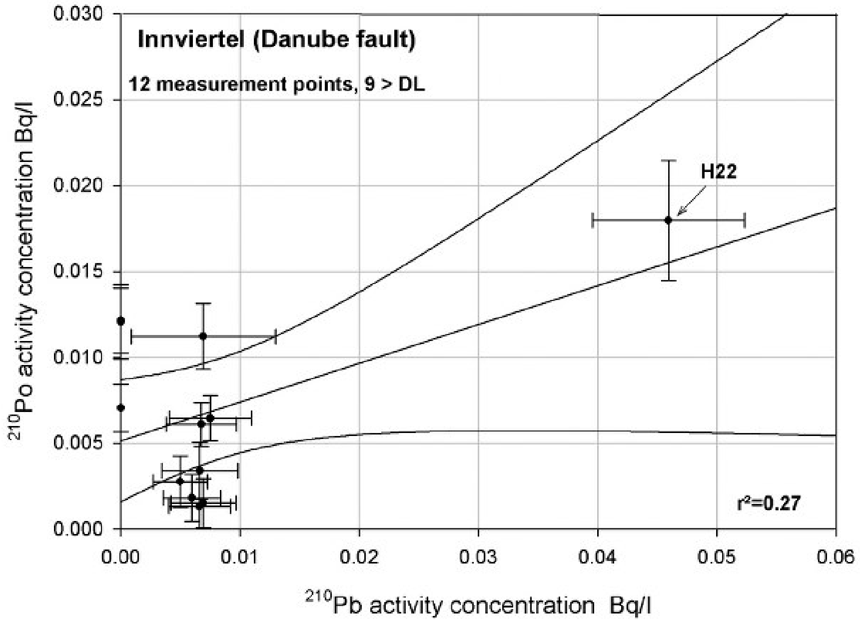

Figure 11.29: *Correlation between ^{210}Po and ^{210}Pb in the region Innviertel*

11.5 Conclusions

In summary, some radionuclide correlations were detected, but **not significant enough** to use them as **prediction for other nuclides**, in case only a different nuclide was analyzed. With the results of this thesis it seems that expected radionuclide **correlations between products of the ^{238}U decay chain are not that clear or well-defined**. So, as mentioned above, it is not possible to measure only the easy analyzable ^{222}Rn activity concentration and estimate the activity concentration of e.g. ^{226}Ra or the decay products ^{210}Po and ^{210}Pb, which are also dose relevant. The analysis showed that at single measurement points clearly higher activity concentrations were detected, which influence the linear regression calculations, but which are often not representative for the entire measurement results of the nuclides. It was shown that **correlations between nuclides are dependent on geographic regions (geology)**. In some regions stronger correlations between nuclides were detected than in others, which may depend on geology and the magnitude of the measured activity concentrations. So in the lower Mühlviertel, where clearly higher ^{222}Rn activity concentrations were measured, but average activity concentrations of other nuclides, low correlations were detected. In Chapter 15 a closer look is taken at the geology of these regions.

All in all the results of this nuclide correlation survey show interesting aspects and tendencies, but cannot be generalized yet, because of too few measurements and especially measurement points with activity concentrations above decision limits. But this topic is of major interest and should be carried on with more measurements and surveys, because these results can fundamentally affect sampling and measurement methods for **drinking water monitoring**.

Chapter 12

Activity Concentration Distributions in Water Units

12.1 General Background

As discussed in Chapter 5.1.2 the detailed sampling was slightly modified while being carried out, because of appearing problems and extended arising questions. As mentioned sometimes in this thesis samples were taken **directly at wells** and **springs** to research hydrogeological radiometric basic data. For the objective of radiation protection the public exposure of radioactivity from **tap water** at the **consumers' homes** is relevant. So it is interesting to survey the variation of the activity concentration of water on the **way from the wells to the consumers** and examine the influences like **water treatment**, **elevated tanks** and **pipes**. Samples were taken at several points within one water unit in the later sampling phases for surveying the activity concentration gradient within the water flow from the well to the consumer.

Because of time and financial reasons not all samples could be analyzed for all radionuclides of interest. So a survey of activity concentration gradients can be carried out mainly for ^{222}Rn. Such a survey was also tried with other nuclides at some water units, but with fewer sampling points within the water flow. Besides, no samples within the water flow in water units were taken in southern Upper Austria. The samples – taken at different wells or springs in the water units – showed already very low ^{222}Rn activity concentrations, so detailed sampling at other points within the water units or at the consumers houses was renounced. Therefore activity concentration distributions within some water units are discussed and illustrated only for the region Mühlviertel and one location in Innviertel (Danube fault).

Within the study of activity concentration variation of water on its way through a water unit, our attention was also turned to the influence of activity concentration by **water treatment techniques**. There are several studies about water treatment techniques and their ability to remove radionuclides from water (e.g. TENAWA – Annanmäki (1999), Schaffer (2005), Staubmann (2002), Wisser (2003)).

In these studies different techniques are tested for their efficiency to reduce or remove different radionuclides from water, which is dependent on the physical and chemical characteristics of the respective radionuclide. According to Wisser (2003), **aeration**, which is the common treatment method used for the physical de-acidification of raw water, is also the primary treatment method to eliminate radon from water. The most common water treatment process is **filtration**, which separates solid particles by directing water through a porous medium. It can effect the removal of solid particles containing U, Ra, Po, Pb. Additionally – in an open filtration process – Rn can be degassed, which can enhance the indoor air of water treatment buildings (see Ringer et al., 2006a,b). Another common removal process is **ion exchange**, which is used to remove toxic ions from polluted water or to eliminate water hardness. Ion Exchange can remove dissolved U, Ra, Po and Pb. The classical method of removing water hardness is **lime softening**, which can remove dissolved Ra. There are some more sophisticated techniques like reverse osmosis or nanofiltration, which are highly suitable for radionuclide removal, but very costly (Wisser, 2003). No more attention is spent on these further methods, because no surveyed water supplies of this thesis use them.

In the water supplies under research within this thesis only few water treatment techniques were existent, mainly because only small water supplies and units were analyzed and the Austrian water is of **high quality** (spring and well water) (Lebensministerium, 2008b).

In the surveyed water supplies in southern Upper Austria water is not treated generally. In this region the water mainly is hard spring water from the limestone Alps (high water hardness). In the analyzed water units in the Mühlviertel and Innviertel the major treatment technique is **filtration by marble gravel** (*Marmorkies*), which consists of calcium-carbonate. This treatment is used fo**r de-acidification** of soft water (low water hardness), which is a result of granite and gneiss (crystalline) bedrock. The aggressive carbonic acid, which is existent in soft water, especially in crystalline Mühlviertel, corrodes water pipes, wherefore de-acidification is necessary. (Land Salzburg, 2008, Landessanitätsdirektion, Land Oberösterreich, 2008, OÖ Wasser Genossenschaftsverband, 2008, Österreichische Vereinigung für das Gas- und Wasserfach (ÖVGW), 2008)

In a few supplies drinking water is exposed to UV radiation for disinfection and sterilization. In one water supply aeration is used to eliminate radon from water (Figure 12.1).

^{222}**Rn activity concentration distributions** in some **water units** are illustrated and discussed. Additionally if enough data exist, also distributions of other nuclides are surveyed. The Radon-222 measurements in the water supplies with open filtration by marble gravel (*Marmorkies*) in the Mühlviertel (open filtration basins – Figure 12.2) show, that this water treatment does not appreciably reduce the ^{222}Rn activity concentration in the water by degassing as mentioned above. In some cases even a slight increase of the radon activity concentration was detected after **de-acidification** by marble gravel. This effect is verified by Bünger (1997), who surveyed the radon concentration in drinking water in water supplies in Berlin.

In this study the raw water had a very low radon activity concentration and was vented before the de-acidification. Therefore the water is poor on radon and on the way through the gravel bed filtration it is capable of reloading with radon. Bünger (1997) explains the effect with ochre accumulations on the filter grains of the gravel bed filtration. These accumulations mainly consist of ferric-manganese oxide hydrates, which are likely to concentrate heavy metals like radium by co-precipitation effects. Variable radium concentrations are expected depending on the age of the filter gravel and thickness of the overlay.

Figure 12.1: *Aeration in a water supply to remove radon from water*

Figure 12.2: *De-acidification of soft water with open filtration by marble gravel (Marmorkies) – open filtration basin*

In spite of low effectiveness of radon activity concentration reduction by filtration with marble gravel, in all surveyed water units the **radon activity concentration decreases** from the well to the tap water at the consumer. It seems that the most effective measure to reduce radon activity in water is **to mix with low radon activity concentration waters** and time. The **duration of dwell** of drinking water in reservoirs (e.g. elevated tank) and in piping network reduces the radon activity concentration because of radioactive decay, and maybe also by adsorptions on accumulations in pipes. These facts are also verified by Bünger (1997).

Usually, water units consist of several wells and springs. As mentioned in Chapter 5.1.2, it was not possible to take samples from all wells and springs because of costs and lack of time, and it was tried to select them representatively. Therefore mainly samples of the different deep wells were taken and sometimes only one selected spring or mixed water of more springs from the same catchment area was taken within one water unit. This is cognizable in the following figures, when samples of mixed water from more wells and springs in reservoirs or water treatment systems were analyzed. Because of our interest to survey the influence of **water treatment techniques** to activity concentration it was tried to take some samples before and after different water treatment techniques.

12.2 Survey of Selected Water Supplies

Figures 12.3 and 12.4 illustrate the two parts of a water unit in the village in the **lower Mühlviertel**, in which the highest Radon-222 activity concentration was measured (WVA70). The water unit consists of **two deep wells** (WVA70 and WVA71) and **some springs**. WVA68 is a mixed water of two springs before on-site **de-acidification** by marble gravel (*Marmorkies*), WVA69 is the same water after treatment. As discussed above, no ^{222}Rn reduction is noticeable by de-acidification, even a slight radon activity concentration elevation is detected. The deep wells show radon activity concentrations of above 800 Bq/l, whereas the clearly deeper one (220 m, WVA71) has a slightly lower concentration than the less deep one (100 m, WVA70). The water of the two deep wells and the springs are mixed in an **elevated tank**. WVA72 is a sample at one of the **first consumers** after the elevated tank. The ^{222}Rn activity concentration at this point has reduced to 100 Bq/l. At WVA73, a subsequent consumer in the water unit, the radon activity concentration has again reduced to 30 Bq/l. So this example demonstrates the above discussed reduction of radon activity concentration by mixing water and duration of dwell in reservoirs and pipes, maybe also adsorptions on accumulations in pipes.

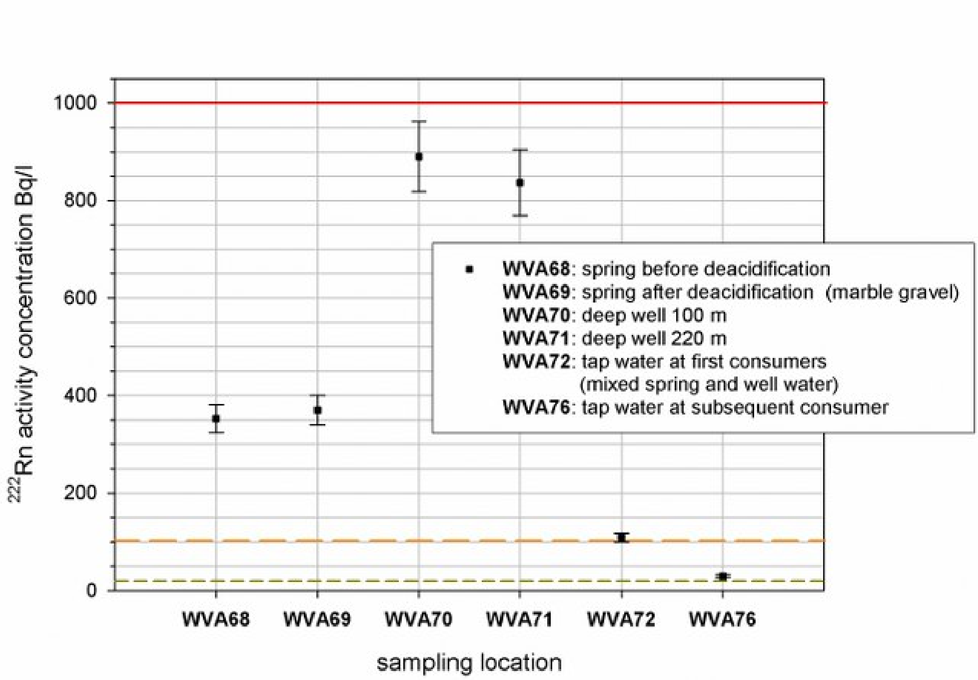

Figure 12.3: ^{222}*Rn activity concentration at different points within a water unit in the lower Mühlviertel*

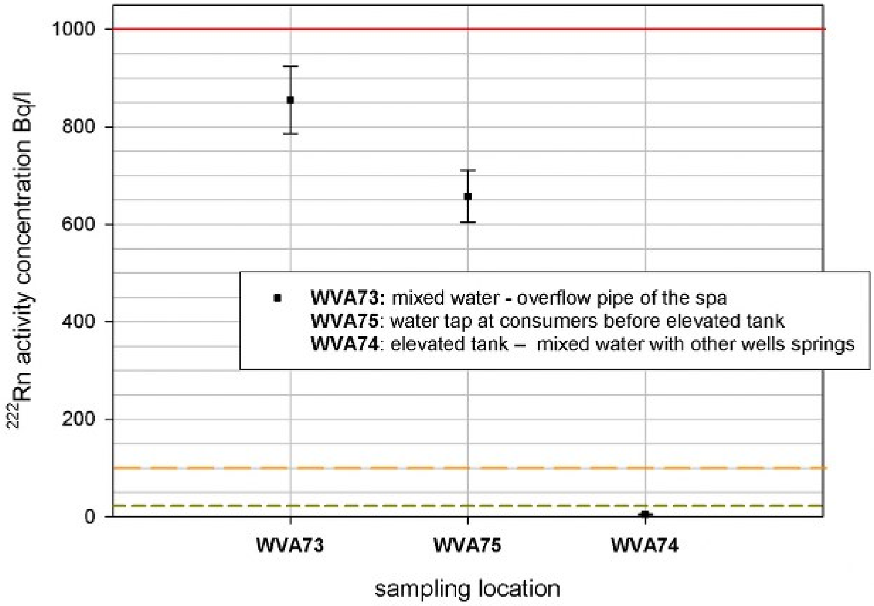

Figure 12.4: ^{222}*Rn activity concentration at different points within a part of a water unit in the lower Mühlviertel*

Figure 12.4 shows the second part of the same water unit in the lower Mühlviertel. Mixed water from **different springs** (WVA73), which are the overflow of springs which are used for the **radon spa** in this village, run to an **elevated tank**, where they are mixed with water from other wells and springs (WVA74). But in between some consumers use the water, which comes directly from the springs (WVA75), before it runs into the elevated tank. The mixed water from the springs has a radon activity concentration of above 800 Bq/l, which reduces to less than 10 Bq/l by mixing and dwell period in the elevated tank. But the radon activity concentration, directly taken at the **consumers tap** in between (WVA75), shows a radon activity concentration of 650 Bq/l, which is the highest measured activity concentration at a consumer's home, fed by water from water supplies. **For these few consumers radiation protection measures may be reasonable.**

Only samples WVA68 to WVA71 of this water unit were analyzed for **other radionuclides**, so no activity concentration distribution from wells and springs to the consumers can be illustrated. The activity concentrations of ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{3}H of these four samples are all below decision limit. For gross alpha, gross beta and ^{238}U the activity concentration analysis of the samples WVA68 and WVA69 shows no impact of the de-acidification. The activity concentrations before and after treatment are equal within uncertainties. An activity concentration decrease is detectable for ^{210}Po after de-acidification from 44 mBq/l to 13 mBq/l, but the uncertainties from these measurements are very high, and so it may as well be equal within uncertainties.

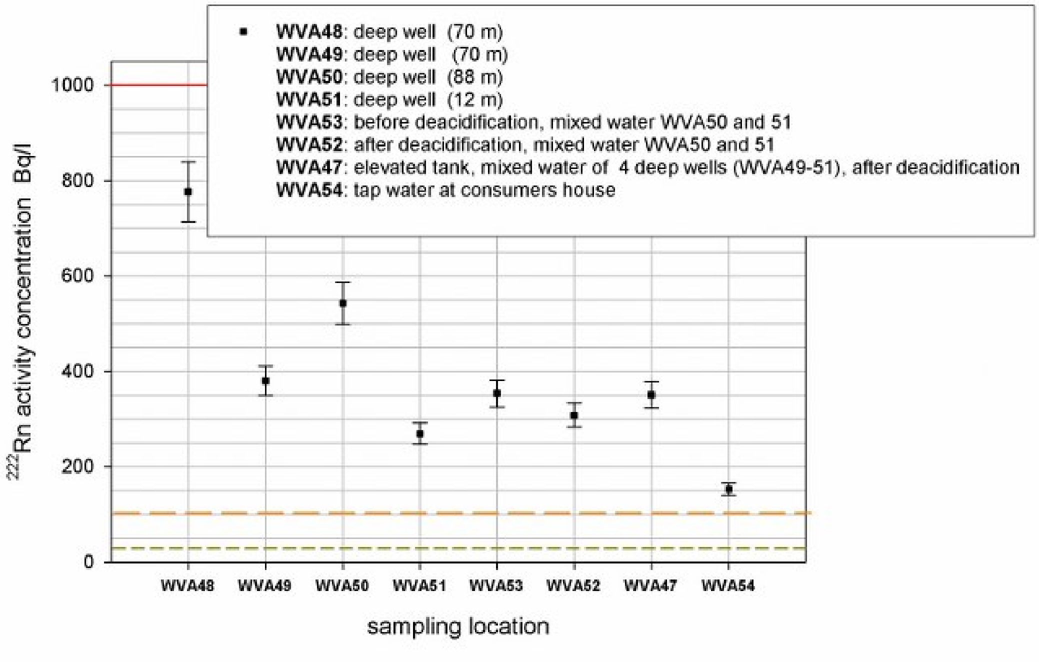

Figure 12.5: *^{222}Rn activity concentration at different points within a water unit in the lower Mühlviertel*

In Figure 12.5 a similar situation is illustrated also in an **other water unit in the lower Mühlviertel**. The unit consists of **4 deep wells**. WVA48 and WVA49 both have the same depth of 70 m and are located close together. Nevertheless the

Table 12.1: *^{238}U activity concentrations in water of 4 deep wells (WVA48–WVA51) and in mixed water of this wells (WVA47)*

Sampling location	**^{238}U activity concentration (Bq/l)**	**Uncertainty ^{238}U activity concentration (Bq/l)**
WVA48	0.0160	0.0008
WVA49	0.0610	0.0031
WVA50	0.0530	0.0027
WVA51	0.0270	0.0013
WVA47	0.0380	0.0019

^{222}Rn activity concentration of WVA49 is half of WVA48. WVA50 and WVA51 are also deep wells located close together with different depths (88 and 12 m). Sample WVA53 is mixed water from the two deep wells WVA50 and WVA51 before **de-acidification**, and WVA52 the same water after de-acidification by marble gravel. Finally WVA54 is a **tap water** taken directly at a consumer's house. As discussed above, de-acidification by marble gravel again does not clearly reduce the radon activity concentration in the water. Mixing higher radon activity concentration water (e.g. WVA50 – 550 Bq/l) with lower radon activity concentration water (e.g. WVA51 – 270 Bq/l) yields a radon activity concentration in the mixed water of 350 Bq/l. Mixing the water of all 4 deep wells and taking into account the duration of dwell and adsorption in reservoirs and pipes, the radon activity concentration at the consumers has halved once more to 150 Bq/l.

Only two samples of this water unit were measured for ^{228}Ra, for ^{226}Ra, ^{3}H and gross beta five samples were analyzed, but all have activity concentrations below decision limits. For ^{210}Po and ^{210}Pb five samples were analyzed, but only a few are above decision limit, so no distribution can be demonstrated. For gross alpha and ^{238}U activity concentration measurement results exist from the 4 deep wells (WVA48-51) and WVA47, the mixed water of all 4 deep wells after de-acidification by marble gravel. The **^{238}U activity concentration** of the **mixed water** is exactly the **product** of the activity concentrations of the **deep wells**. The calculated average activity concentration of the 4 deep wells is 0.039 Bq/l, the measured activity concentration of the mixed water 0.038 Bq/l (Table 12.1). The de-acidification seems to have no clear effect on the ^{238}U activity concentration in the water. For **gross alpha** the mixed water has a higher activity concentration than expected from the mixing process. It may be that the **de-acidification** has **concentrated** the **gross alpha activity** in the water. (Figure 12.6)

No samples were taken directly at a consumer's house for analyzing the ^{238}U and gross alpha activity concentration, so the impact of duration of dwell and adsorption in the pipe net can not be observed.

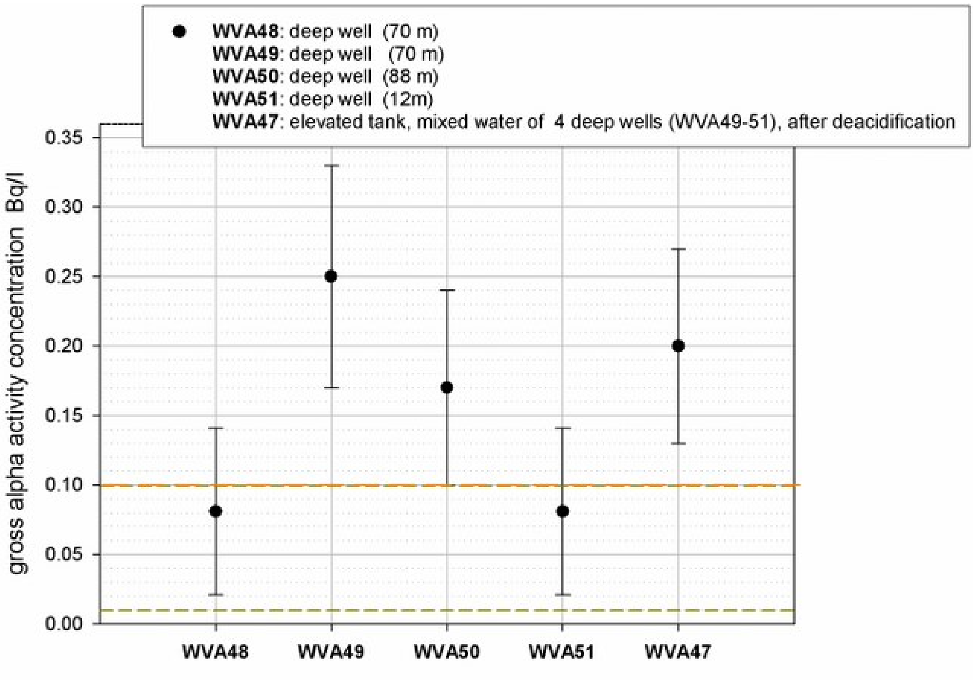

Figure 12.6: *Gross alpha activity concentration at different points within a water unit in the lower Mühlviertel*

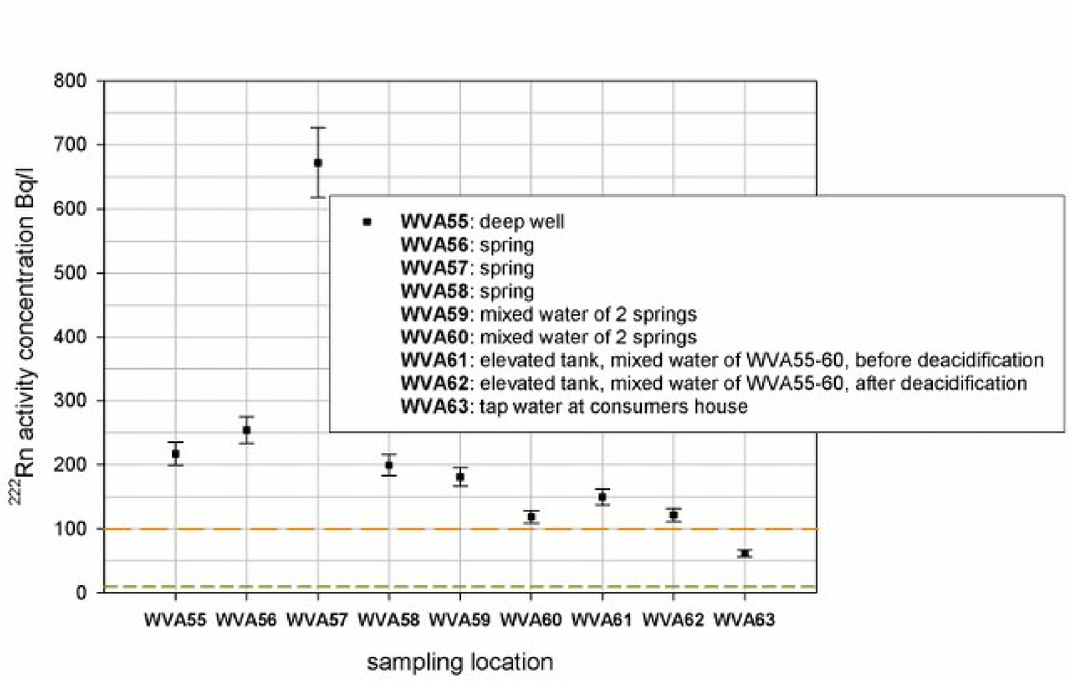

Figure 12.7: ^{222}*Rn activity concentration at different points within a water unit in the lower Mühlviertel*

Figure 12.7 shows **another example** of a **water unit in the lower Mühlviertel**. The water unit consists of **one deep well** (WVA55) and **several springs** (WVA56-60). The waters of the well and the springs are mixed in the **elevated tank** and treated (**de-acidification** by marble gravel). WVA61 is a sample of the mixed water before de-acidification, WVA62 after de-acidification. WVA63 was taken directly from the **tap at a consumer's house**. The well and springs have varying radon activity concentrations of about 120 to 680 Bq/l, the mixed water has a radon activity concentration of about 150 Bq/l, which again is only little reduced by de-acidification to 120 Bq/l. The radon activity concentration of the water is again halved at the way from the elevated tank (after de-acidification) to the consumer (see above).

Only a maximum of three samples of this water unit were analyzed for other radionuclides. For ^{228}Ra and ^{3}H all samples have activity concentrations below decision limit. The deep well (WVA55) is the sampling point with the highest measured ^{226}Ra, ^{210}Po and ^{210}Pb activity concentrations within this thesis. Also activity concentrations above decision limit were measured for ^{238}U, gross alpha and gross beta in this sample. The other two analyzed samples in this unit are one spring (WVA56) and the mixed water after de-acidification (WVA62). The activity concentrations of these two samples are below decision limit for all nuclides. Only ^{238}U shows an activity concentration above decision limit at WVA62, but still one tenth of the activity concentration of WVA55. The low activity concentrations of the mixed water after de-acidification in contrast to the high results at the deep well confirm the above discussed **effective reduction of activity concentration in drinking water** by **mixing waters** with high and low activity concentrations.

In another **water unit** in the **north of the lower Mühlviertel** samples of **5 different springs** were taken, and one after mixing them at a consumer. The radon activity concentrations at the different springs are between 170 and 500 Bq/l, the radon activity concentration at the consumer is clearly lower with 70 Bq/l. So again the dwell duration in the elevated tanks and in the pipes reduces the radon activity concentration strongly.

The six samples were also analyzed for all other nuclides. The ^{226}Ra, gross alpha and gross beta activity concentrations of all samples were below decision limit. The measured ^{3}H activity concentrations of the six samples were equal within uncertainties. The ^{228}Ra activity concentrations at the four analyzed springs were generally rather low and equal within uncertainties (7–12 ± 5–6 mBq/l). The ^{228}Ra activity concentration at the consumer is below decision limit. A similar situation is indicated for ^{210}Pb and ^{210}Po. Different activity concentrations were detected at the springs, but a ^{210}Pb activity concentration below decision limit and a reduced ^{210}Po activity concentration were measured at the consumer. Figure 12.8 shows a similar **^{238}U activity concentration distribution** like in the water unit above. In contrast to the results of the other nuclides, the ^{238}U activity concentration at the consumer is not clearly lower than at the springs. The ^{238}U activity concentration at the **consumer tap water** (0.0023 Bq/l) is again the **result of mixing the waters of the five springs** (average value about 0.0026 Bq/l). For ^{238}U duration of dwell in elevated tanks and in pipes has no effect because of its long half life and it seems that no ^{238}U is lost by adsorption or other processes in the pipes.

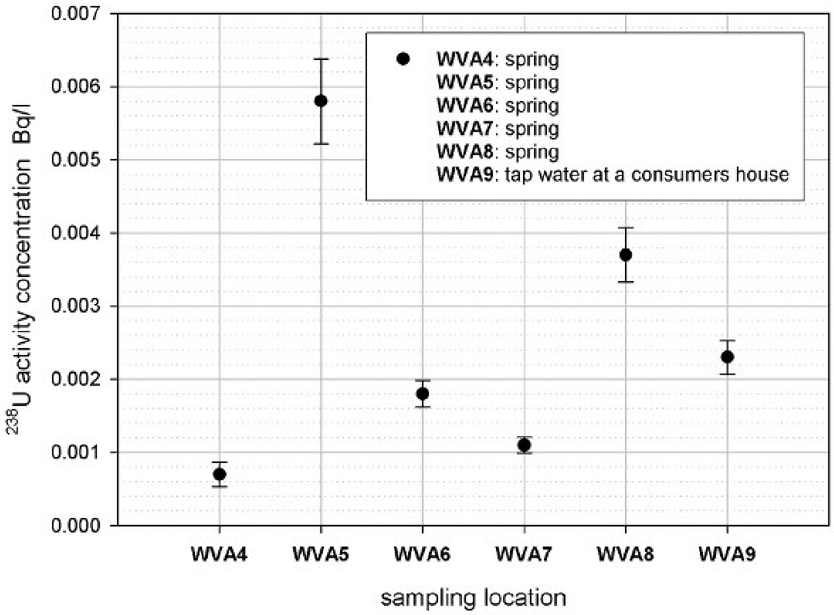

Figure 12.8: *^{238}U activity concentration at different points within a water unit in the lower Mühlviertel*

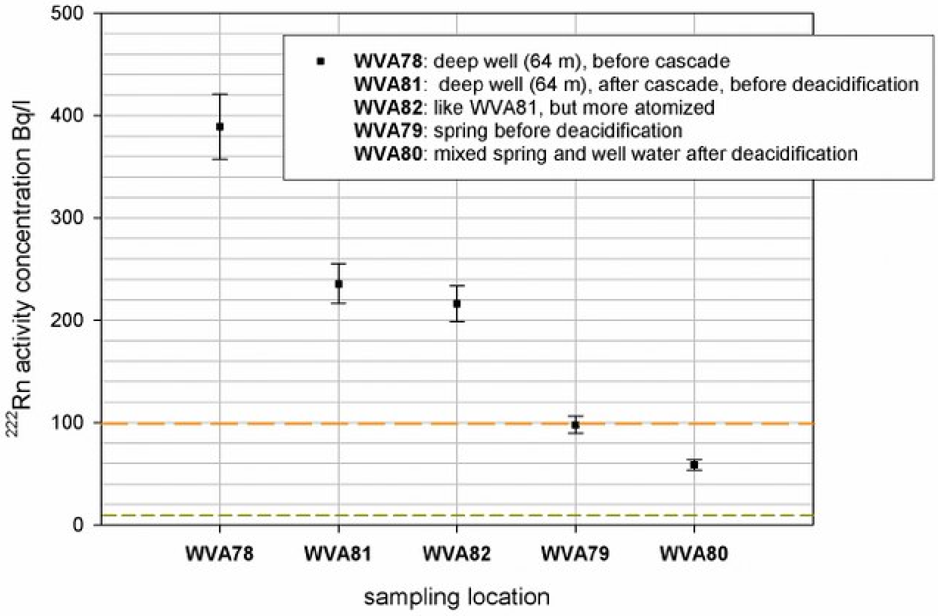

Figure 12.9: *^{222}Rn activity concentration at different points within a water unit in the lower Mühlviertel*

Figure 12.9 is the **last example** of a **water unit in the lower Mühlviertel** and consists of **one deep well** (WVA78) and **one spring** (WVA79). In this water unit structural measures were carried out to reduce radon concentration in the well water because of an official notification, in which a limit of 100 Bq/l is given. Radon should be eliminated in the water by **aeration with a cascade**, which nebulizes the water and is supposed to force radon to degas. WVA78 is the sample of the well water before aeration, WVA81 after aeration. This measure to eliminate radon from the water reduces the radon activity concentration from 389 ± 32 Bq/l to 236 ± 19 Bq/l. The sample WVA82 was also taken at the same location after aeration, but with a stronger nebulization. The stronger nebulization does not have a clearly stronger impact to reduce radon in the water. The radon activity concentration in the drinking water is clearly reduced after the aeration, but does not comply with the limit of 100 Bq/l. The radon activity concentration is reduced about 40–45%, which is clearly below the maximum removal efficiency of 99% given in literature (Wisser, 2003).

WVA79 is the **spring** which belongs to the same water unit, with a radon activity concentration of 98 ± 9 Bq/l. After mixing the well and spring water and after de-acidification the sample at the outlet of the elevated tank (on the way to the consumers) shows a radon activity concentration clearly below 100 Bq/l (WVA80). Again **mixing wells** with different activity concentrations strongly effects the concentration. In this case it seems that additionally the de-acidification in the open, wide basins also reduced the radon activity concentration in the water. To verify this, a sample of the mixed water directly before and after de-acidification should be taken.

No further discussion of an activity concentration distribution in this water unit is reasonable for all other nuclides, because samples were taken only after the aeration (WVA81) and directly at the spring (WVA79). So no effect of aeration and mixing can be surveyed for the other nuclides.

Figure 12.10 demonstrates the radon activity concentration distribution in a **small water unit** in the **upper Mühlviertel**. The small water unit consist of **some springs**, (only one of them is analyzed – WVA31), which are mixed and piped to the **elevated tank**. WVA32 was taken before de-acidification by marble gravel, WVA33 after treatment. WVA34 is **tap water** directly at a consumer's house. The water supply shows a clear radon activity concentration reduction after the de-acidification (from 222 ± 22 to 146 ± 15), in contrast to the above mentioned examples. This may be an effect caused by sampling, because the water of WVA33 was taken after the elevated tank, as access to the water was not possible directly after de-acidification process. So the water could already have had a longer duration of dwell in the reservoir. The reduction from WVA33 (after elevated tank) to the consumer is therefore small, because water does not remain long in the pipes in between.

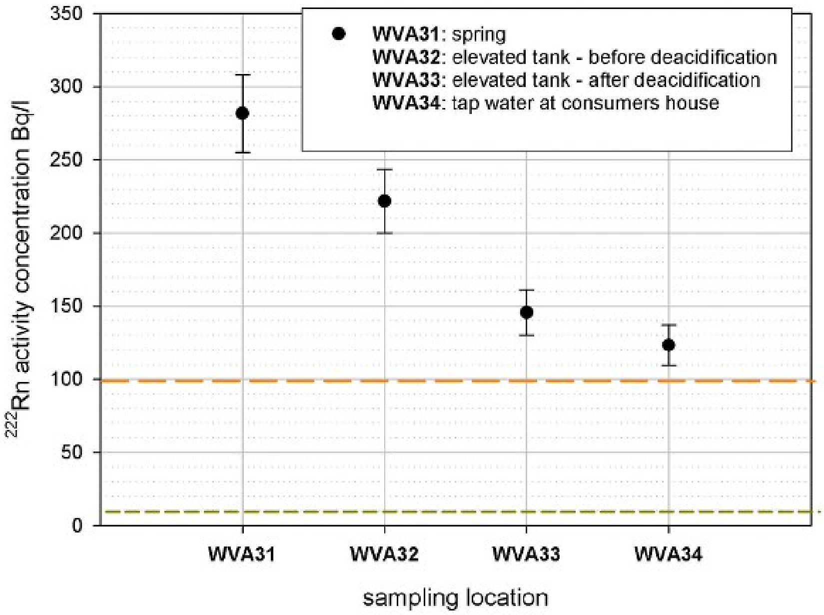

Figure 12.10: *^{222}Rn activity concentration at different points within a water unit in the Upper Mühlviertel*

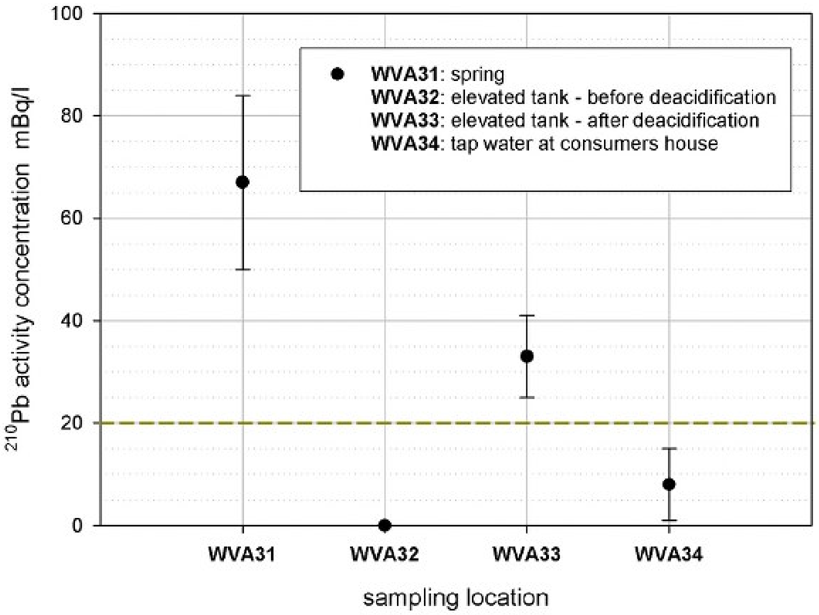

Figure 12.11: *^{210}Pb activity concentration at different points within a water unit in the Upper Mühlviertel*

All four samples of this water unit were analyzed for all **other nuclides**. All ^{226}Ra and gross alpha activity concentrations were below decision limit. The ^{3}H, ^{238}U and gross beta activity concentrations of the four samples are all equal within uncertainties. So it seems that for these nuclides and parameters de-acidification by marble gravel and duration of dwell in the reservoir and pipes have no impact, which was at least discussed above for ^{238}U. The ^{228}Ra activity concentrations at the spring and the consumer are below decision limit, and the sample before the de-acidification is slightly lower than afterwards. It might be an indication of ^{228}Ra enrichment by de-acidification, but the measured activity concentrations are very low and equal within uncertainties, so this indication is very weak.

For ^{210}**Po** and ^{210}**Pb** there is a great difference between the water activity concentration at the spring (WVA31) and before de-acidification (WVA32). The ^{210}Po activity concentration at WVA31 is 110 ± 41 mBq/l, at WVA32 only 8 ± 13 mBq/l. The ^{210}Po activity concentration after de-acidification and at the consumer is equal to WVA32 within uncertainties. The ^{210}Pb activity concentration at WVA31 is 67 ± 17 mBq/l, at WVA32 below decision limit. After de-acidification (WVA33) the ^{210}Pb activity concentration increases to 33 ± 8 mBq/l, and then reduces to 8 ± 7 mBq/l at the consumer (WVA34) (Figure 12.11). The great difference between the ^{210}Pb and ^{210}Po activity concentrations at the spring and before de-acidification can not be explained by now and should be verified by repeated sampling and analyzing. The increase of ^{210}Pb activity concentration after de-acidification might be an indication of enrichment of ^{210}Pb by de-acidification by marble gravel, but this has to be verified by further measurements.

Figure 12.12 illustrates the radon activity concentrations contribution in a **water unit** in the **Innviertel (Danube fault)**. The water unit consists of **two deep wells** (WVA16, WVA17) and **one spring** (WVA18). The water of the wells and the spring is mixed in the **elevated tank** and treated. The treatment consists of a **de-acidification** and a **de-ferrication** filtration tank. The sample WVA19 was taken at the outlet of the elevated tank after treatment on the way to the consumer. WVA20 is **tap water** directly at a consumer's house. The highest radon activity concentration was measured in the spring water (196 ± 20 Bq/l), one well has a slightly lower, the other one a clearly lower radon activity concentration. After mixing, duration of dwell in the reservoir of the elevated tank and treatment (WVA19), the radon activity concentration has clearly reduced to 76 ± 9 Bq/l. On the way to the consumer the radon activity concentration is again strongly reduced to one fourth. This strong radon activity concentration reduction on the way from the elevated tank to the consumer may caused again by duration of dwell in a long pipe net and adsorption at accumulations in the pipes (see above).

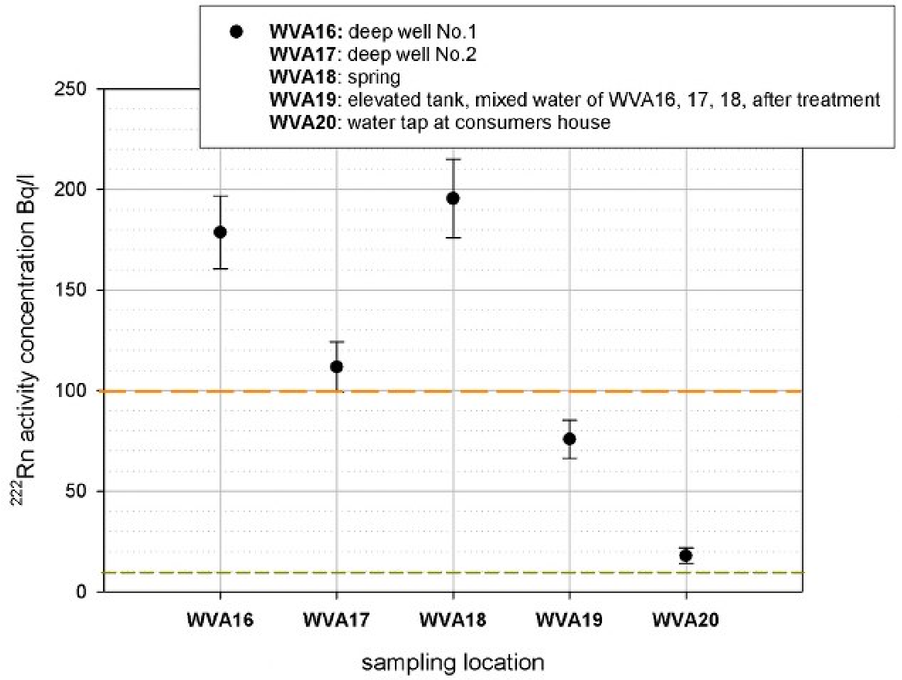

Figure 12.12: *^{222}Rn activity concentration at different points within a water unit in the Innviertel (Danube fault)*

All samples of this water unit were analyzed for ^{226}Ra, gross alpha and gross beta, but the activity concentrations are all below decision limit. The ^{3}H activity concentrations are all equal within uncertainties. ^{228}Ra, ^{210}Po and ^{210}Pb were analyzed only at WVA18-20. All ^{228}Ra and ^{210}Pb activity concentrations are below decision limit. The **^{210}Po activity concentrations** are generally very low, but the concentration at the spring is almost twice as high as the mixed and treated water in the elevated tank and at the consumer. It seems that duration of dwell or treatment has an effect to the ^{210}Po activity concentration, but it could also be caused by very low ^{210}Po activity concentrations at the well waters, which were not analyzed. Figure 12.13 shows the **^{238}U activity concentration** distribution at this water unit. The measured ^{238}U activities are generally rather low, but in contrast to radon the highest ^{238}U activity concentration was detected at one of the wells (WVA16). The second well and the spring have lower activity concentrations. The mixed and treated water in the elevated tank (WVA19) has a similar ^{238}U activity concentration as the waters from the spring and second well. In two examples of water units above, the ^{238}U activity concentration in water after mixing and treatment was exactly the average activity concentration of the mixed waters. In this case the activity concentration of WVA19 is lower than the calculated average activity concentration of the two wells and the spring water. It seems that in this case the treatment procedure or duration of dwell in the reservoir or pipes impacts the ^{238}U activity concentration. Another explanation is that the waters of the different sources are not mixed in equal shares. The measured ^{238}U activity concentration at the consumer (WVA20) (2.63 ± 0.26 mBq/l) is clearly higher than at the elevated tank, and corresponds again exactly to the calculated average activity concentration of the wells and the spring (2.64 mBq/l). So, this seems to be an indication that also in this case duration of dwell and treatments have no impact to the ^{238}U activity concentration, but the water in this water unit is not mixed in equal shares.

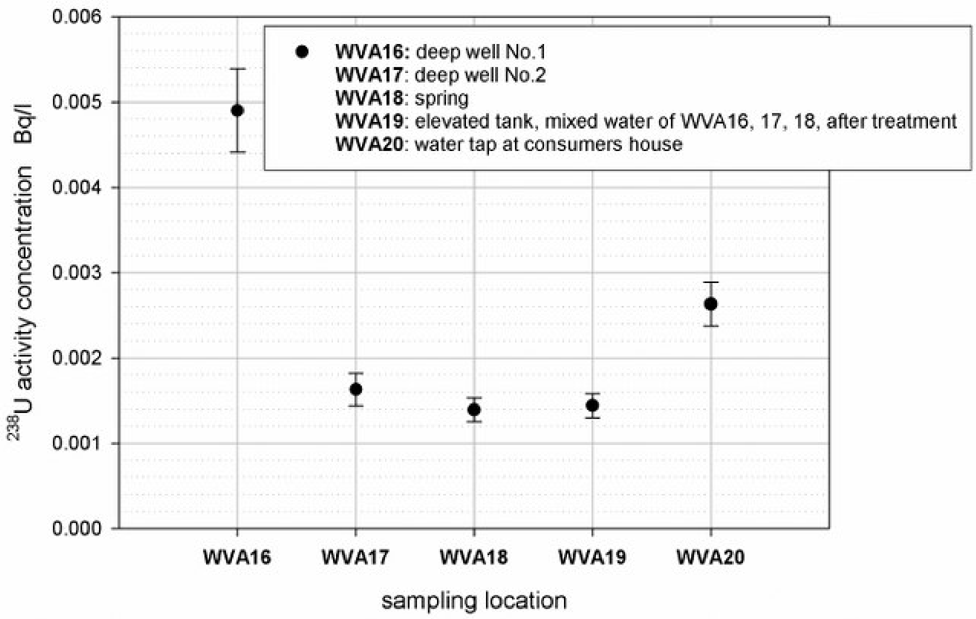

Figure 12.13: *^{238}U activity concentration at different points within a water unit in the Innviertel (Danube fault)*

12.3 Conclusions

In general all these measured activities are **snap-shots** and **cannot be generalized** without huge uncertainties, but they show **tendencies** and **trends**. For more fundamental conclusions more measurements at more points within a water unit have to be carried out.

Nevertheless in the following the trends of these examples are summed up for all nuclides.

The radon activity concentration at wells and spring is rather high, especially in the lower Mühlviertel, nevertheless the measured radon activity concentration at the consumers is typically at least one magnitude lower and below all limits. The main reason for the **reduction of radon activity concentration** within the run of the water from the well to the consumer is **duration of dwell**, because of the short half life of radon and degassing. Lehmann et al. (1997) reported no significant reduction of ^{222}Rn on the way from the wells to the consumer, because of mostly rather short ways and durations. But they did not report influences by losses and outgassing in the tanks and pipes. **Effective reduction** of radon activity concentration by **de-acidification** by marble gravel **could not be detected**. **Aeration** decreases radon activity, but not as effectively as expected, and **mixing waters** with high radon activity concentrations with waters with low radon activity concentration is more effective. Lehmann et al. (1997) also reported losses of radon activity of a maximum of 50% caused by different **water treatment techniques**. This was verified by Bünger & Rühle (1997), who also reported rather low decontamination effect of natural radionuclides in drinking water by water treatment. The highest effect was detected for ^{210}Pb according to Bünger & Rühle (1997), which should also be surveyed in further studies for Upper Austria (see above). The losses of radon activity concentration from the well to the consumer show, that in general for consumers of water from water supplies no radon exposure problems are expected, except in some special cases like the consumers in the water unit, who directly drink the water of the wells from the radon spa. So again **radiation protection attention** has to be given to **private wells**, where the water has only a short way and duration of dwell between the well and the consumer.

For other radionuclides generalization is less acceptable yet, but nevertheless a conclusion is made of our results.

For ^{226}**Ra** nearly all activity concentration measurements are below decision limit and no statements can be made. For ^{228}**Ra**, **gross alpha** and **gross beta** less results exist, and the measured activity concentration often was below decision limit. From the few results above decision limit it seems, that de-acidification and duration of time has no big impact on these nuclides, because the measurement results from different points in one water unit are mostly equal within uncertainties. A similar situation is detected for ^{3}**H**, because the measured activity concentrations are either below decision limit or equal within uncertainties. So no effects by de-acidification, duration of time or losses in pipes or reservoirs are noticed. It also seems that **de-acidification** and **duration of time** have **no impact on the** ^{238}**U activity concentration**, mainly because of its long half life. In our samples the best method to reduce ^{238}U activity concentration in the water is to mix it with water with lower ^{238}U activity concentration. So, for the radiation protection point of view, nuclides like ^{238}U should not be disregarded because their activity concentrations in water cannot be reduced by **duration of dwell**.

For ^{210}**Pb** and ^{210}**Po** too few measurement results exist to make good conclusions. On the one hand it seems that the activity concentrations are reduced by duration of dwell, on the other hand it is also possible, that ^{210}Pb is enriched in de-acidification. It also might be that the ^{210}Po and ^{210}Pb activity concentrations are strongly effected by interactions in the pipes. As shown in Chapter 16, the dose contribution of ^{210}Po and ^{210}Pb is significant, so knowledge of these effects and possibilities to reduce ^{210}Po and ^{210}Pb activity concentration in the water within the water unit are fundamental and should be investigated in further surveys.

Chapter 13

Impact of Physical Drinking Water Characteristics on Radioactivity

As discussed in Chapter 5, **temperature**, **electric conductivity** and **pH value** of the water was determined at the sampling points of the detailed sampling phase, provided that the measurement instrument was serviceable.

13.1 Water Temperature

Water temperature should be low in drinking water, because higher temperatures enhance the solubility of (possibly harmful) water ingredients and expedite the increase of microorganisms, while decreasing the oxygen content (von Löw, 2008). As mentioned in Chapter 2 the water temperature highly influences the solubility of radon in water. An **increasing water temperature decreases** the **solubility of radon in water** (Cothern & Smith, 1987). The Austrian drinking water directive defines an indicator parameter of 25°C maximum drinking water temperature (Republik Österreich, 2001).

Water temperatures of **132 water samples** were measured, and the determined temperatures range from 5 to 18°C. At no sampling point in the Innviertel (Danube fault) water temperature was measured, because no measurement instrument was available in this sampling phase. Figure 13.1 demonstrates the distribution of drinking water temperatures in **different geological regions**. The median of the 14 measurement points in the central Mühlviertel is about 13°C, which is clearly higher than in the other regions, because all sampling points in this region are private wells, with only some meters depth. No spring waters were analyzed in the lower Mühlviertel. In contrast, in southern Upper Austria only springs within the catchment area of the Alps were analyzed, which results in a much lower water temperature (median of the 29 measurement points: 9°C).

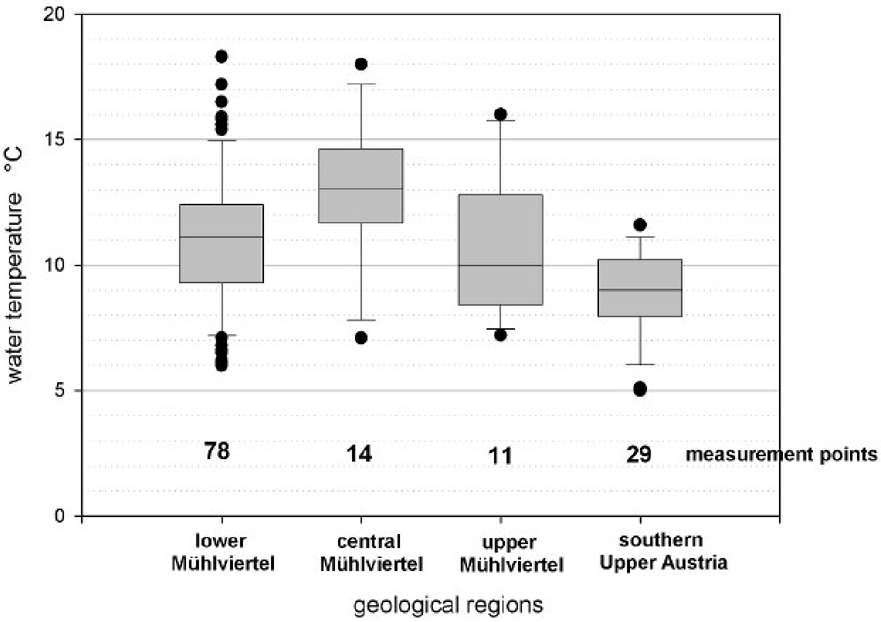

Figure 13.1: *Drinking water temperature in the different geological regions*

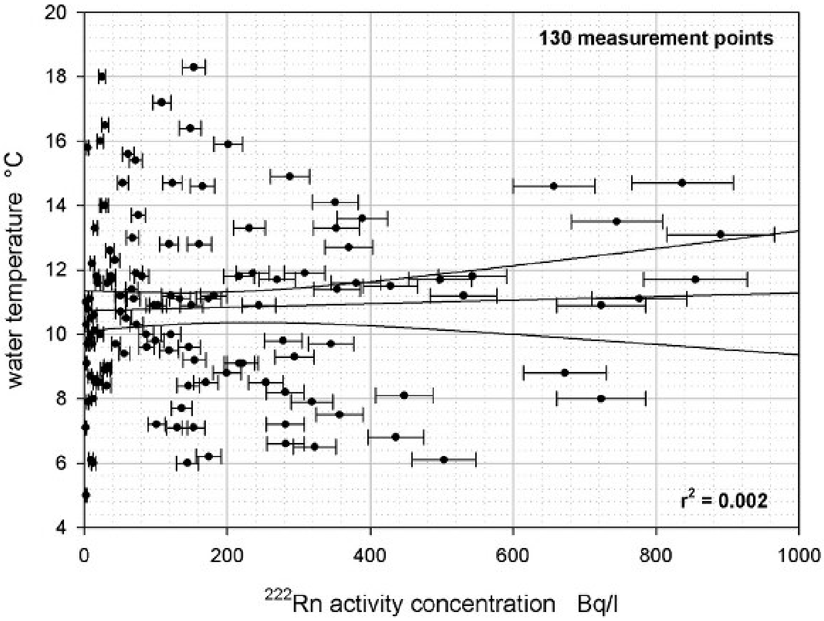

Figure 13.2: *Correlation between drinking water temperature and ^{222}Rn activity concentration*

No correlation between drinking water temperature and radionuclide concentration was detected. Figure 13.2 shows – as an example – the correlation between radon activity concentration and water temperature in the 130 measurement points above decision limit with an r^2 value (defined in Chapter 11) of 0.002. So no impact of the water temperature on the radon solubility could be detected. All other radionuclide concentrations do not show a better correlation either (all r^2 values are below 0.05).

A good correlation between water temperature and radionuclide activity concentration does not exist in the **different geological regions** either. Only in the Upper Mühlviertel a slightly indirect correlation was detected between the water temperature and the ^{222}Rn, ^{210}Pb and ^{210}Po activity concentrations. But only very few measurement points with activity concentrations above decision limits exist in this region (5 to 11). For the three nuclides the activity concentration increases at lower water temperatures as was expected at least for the gas ^{222}Rn. As an example the correlation between ^{210}Po activity concentration and water temperature is illustrated in Figure 13.3 (8 measurement points, r^2=0.278). For ^{222}Rn (11 measurement points) the same trend like ^{210}Po with an r^2 value of 0.174 and for ^{210}Pb (5 measurement points) a good correlation with an r^2 value of 0.797 was detected.

The **water temperature** is mostly **dependent on its origin** (drilled well, spring) and its depth. So the correlation in the Upper Mühlviertel can be explained by some samples of different origins – deep wells with higher activity concentrations and low water temperatures and near surface wells with higher water temperatures and lower activity concentrations. But not all samples follow this easy scheme, as the non correlations showed in all other regions. And no expected clear correlation between higher radon activity concentration because of radon solubility and low water temperature was detected.

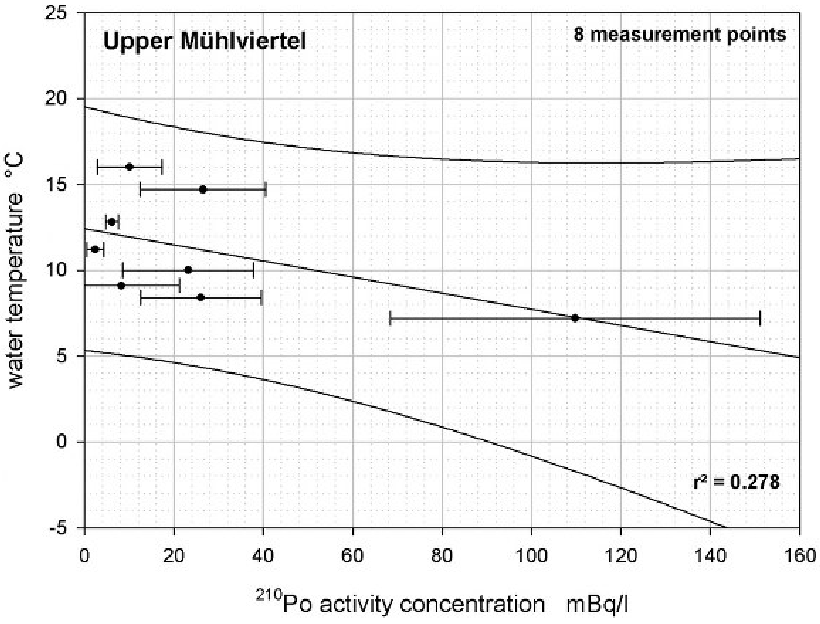

Figure 13.3: *Correlation between drinking water temperature and ^{210}Po activity concentration in the Upper Mühlviertel*

13.2 pH Value

The pH value is a degree of concentration of hydrogen ions H^+ (pH – *potential hydrogenii*) or rather oxoniumions (H_3O^+) and indicates if water is acid, neutral or alkaline. Usually pure water has a neutral to slightly alkaline pH value (pH 7.0–7.5) (Lebensministerium, 2008c, von Löw, 2008). The Austrian drinking water directive defines indicator parameters for pH values of drinking waters from >6.5 and <9.5 (Republik Österreich, 2001). Water with high content of free CO_2 has a pH value in the acid range, and if the pH value lies clearly below the calcium carbonate-carbonic acid balance (calcium carbonate saturation), the calcium carbonate will be suspended and aggressive carbonic acid will be attendant. Aggressive carbonic acid expedites damages and corrosion in pipes, installations and tank materials. With a pH value above the calcium carbonate saturation calcium carbonate precipitation occurs. (von Löw, 2008)

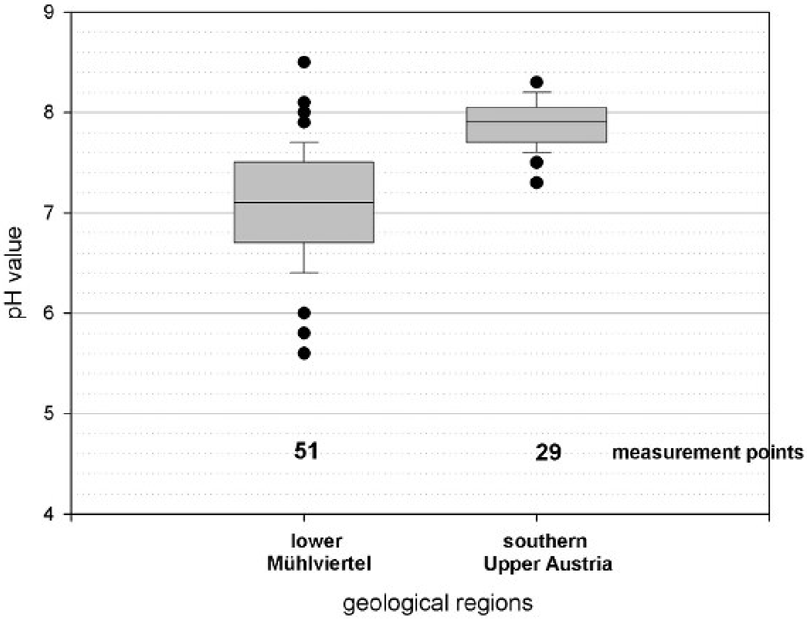

Figure 13.4: *pH values of drinking waters in the lower Mühlviertel and southern Upper Austria*

In the framework of this thesis pH values of **81 water samples** were determined within a range from 5.6 to 8.5. Again the measurement instrument was not available or ready for operation in all sampling phases, so pH value results exist in the geological regions of the lower Mühlviertel and southern Upper Austria only. One single result exists in the central Mühlviertel (pH=7.9). Generally speaking, the pH values in southern Upper Austria are higher than in the lower Mühlviertel (Figure 13.4), with a median of about 7.9 in southern Upper Austria and 7.1 in the lower Mühlviertel, for **geological reasons**. In southern Upper Austria mainly karst groundwater exists, in the lower Mühlviertel crevice ground water. The crevice ground waters have low pH values because of high carbonic acid contents. High carbonic acid contents are typical for regions with crystalline bedrock, as because of missing calcium carbonate fractions no neutralization of the carbonic acid occurs. According to this, ground waters in the northern limestone Alps (karst) have high pH values because of the calcium carbonate containing limestone bedrock. (Amt der Oberösterreichischen Landesregierung, 2008)

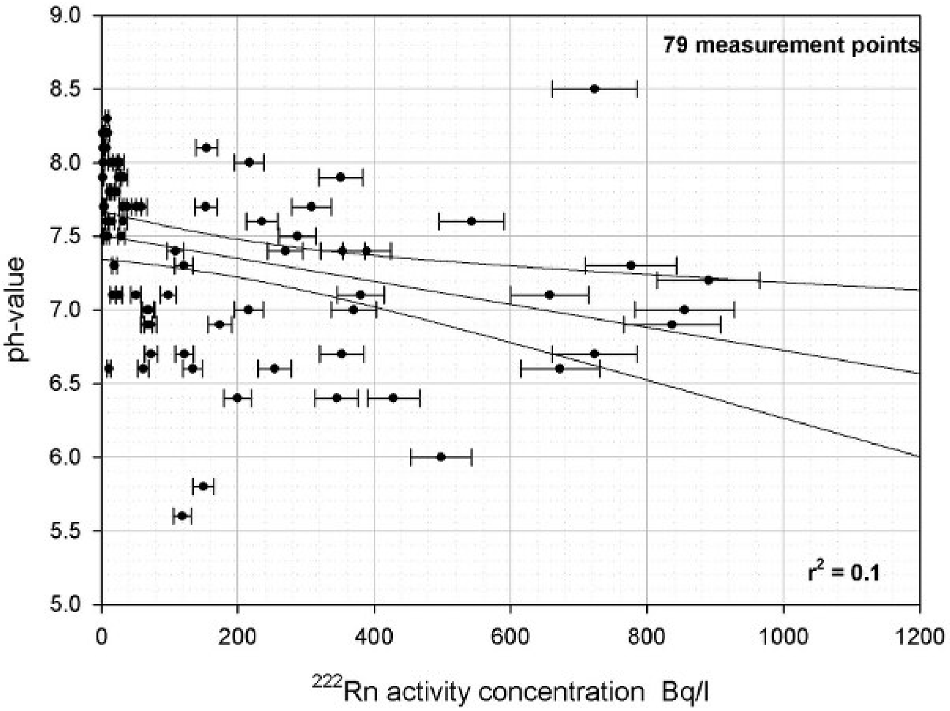

Figure 13.5: *Correlation between pH value and ^{222}Rn activity concentration in drinking water*

No clear correlation was detected between pH values and radionuclide activity concentrations. For Gross beta and ^{210}Pb the number of pH measurement results and activity concentrations above decision limit is rather low (13 and 14), for the others about 30 and all estimated r^2 values are very low ($<$0.07). For radon 79 measurement points were analyzed for pH values and have radon activity concentrations above decision limit. No real correlation can be detected between pH value and radon activity concentration (r^2=0.1), but a slight trend with higher radon activity concentrations at lower pH values may be interpreted (Figure 13.5). In the two different **geologic regions** also no real correlations were detected between pH value and radionuclide activity concentration. For ^{222}Rn no correlation was detected (r^2=0.005 in the lower Mühlviertel and r^2=0.063 in south Upper Austria). The highest r^2 values are 0.292 for ^{210}Pb in southern Upper Austria and 0.267 for ^{210}Po in the lower Mühlviertel (Figure 13.6). For ^{210}Po the activity concentration enhances at higher pH values, for ^{210}Pb decreases at higher pH values. So no clear trend is detectable and no generalization is possible, because too few measurement results are available in each geological region and for all nuclides (always below 20 measurement points).

Almeida et al. (2004) detected a high correlation between low pH value and high radium concentration in Brazil. Too few ^{226}Ra activity concentrations above decision limit exist in this thesis to verify this theory.

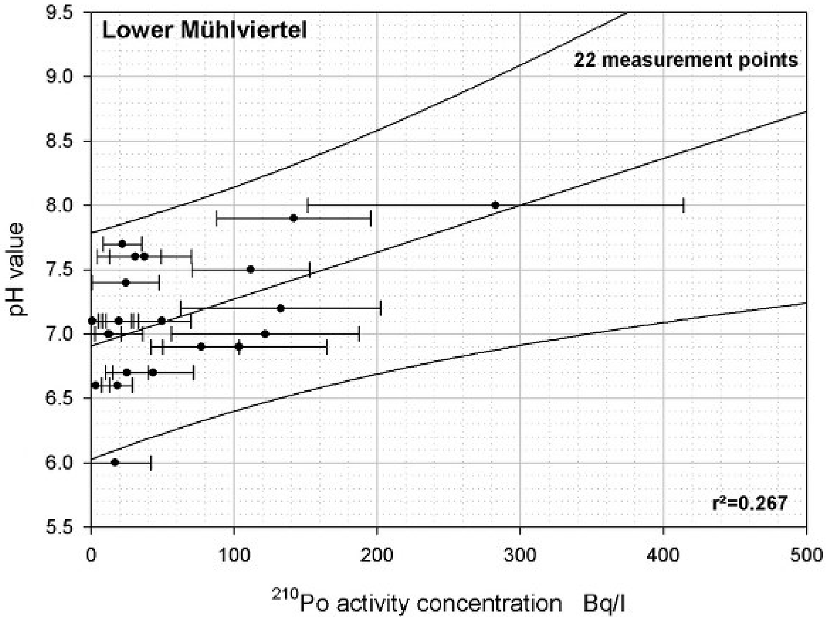

Figure 13.6: *Correlation between pH value and ^{210}Po activity concentration in drinking water in the lower Mühlviertel*

13.3 Electric Conductivity

The electric conductivity is a degree of **concentration of solved salts** (ions, electrolytes) in drinking water with the unit Microsiemens pro cm (µS/cm). The more solved substances are contained in the water, the higher the electric conductivity is. Variations of the – in undisturbed water usually stable – electric conductivity is an indicator for pollution of the water by salts (increase of electric conductivity) or decrease of electric conductivity by surface near impact by precipitation water. (Lebensministerium, 2008c, von Löw, 2008)

The Austrian drinking water directive defines an indicator parameter for electric conductivity of drinking water of 2500 µS/cm at 20°C (Republik Österreich, 2001).

In this thesis **112 drinking water samples** were surveyed for electric conductivity within the detailed sampling phases. As mentioned before, the measurement instrument was not available or ready for operation in all sampling phases, so no electric conductivity results exist in the Innviertel and in the Upper Mühlviertel.

The electric conductivity values in the analyzed drinking waters range from 51 to 888 µS/cm and are generally clearly higher in southern Upper Austria with a median of about 500 µS/cm than in the lower and central Mühlviertel with a median of about 150 µS/cm (Figure 13.7). As discussed above, this is because of **geological differences of karst and crevice ground waters** in the limestones of southern Upper Austria and the crystalline bedrock region Mühlviertel. Water of the limestone region (calcium carbonate) contains more solved substances and therefore the electric conductivity is higher (Amt der Oberösterreichischen Landesregierung, 2008).

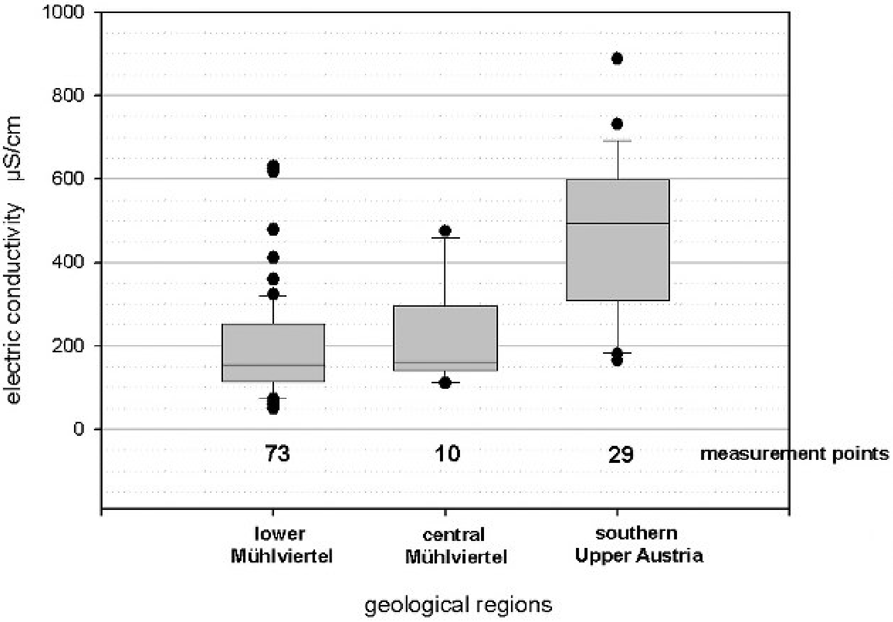

Figure 13.7: *Electric conductivity of drinking waters in the lower and central Mühlviertel and southern Upper Austria*

Measuring the pH value and the electric conductivity of drinking waters in Upper Austria, an easy groundwater classification (three big groundwater types: karst, crevice, pore) is possible (Amt der Oberösterreichischen Landesregierung, 2008).

No correlation was detected between electric conductivity and radionuclide activity concentrations in drinking waters. The r^2 values of all nuclides are very low (<0.07). Figure 13.8 shows for example the correlation of electric conductivity and radon activity concentration with an r^2 value of 0.07, which is the highest r^2 value of all nuclides. Also no correlation was detected between electric conductivity and radionuclide concentration in the three different **geologic regions** (lower and central Mühlviertel and southern Upper Austria).

It seems that the concentration of solved salts in drinking water has no impact to radionuclide concentration at all. But although 112 samples were measured for electric conductivity, not all have radionuclide activity concentrations above decision limit, so only about 20 to 65 measurements points were taken into account for surveying the correlation. For generalization of these results more measurements should be carried out, but a clear trend of no correlation between electric conductivity and radionuclide activity concentration is noticeable. In opposite Almeida et al. (2004) detected a significant correlation between uranium concentration and electrical conductivity in Brazil, but there might be a sea water impact on the ground water composition.

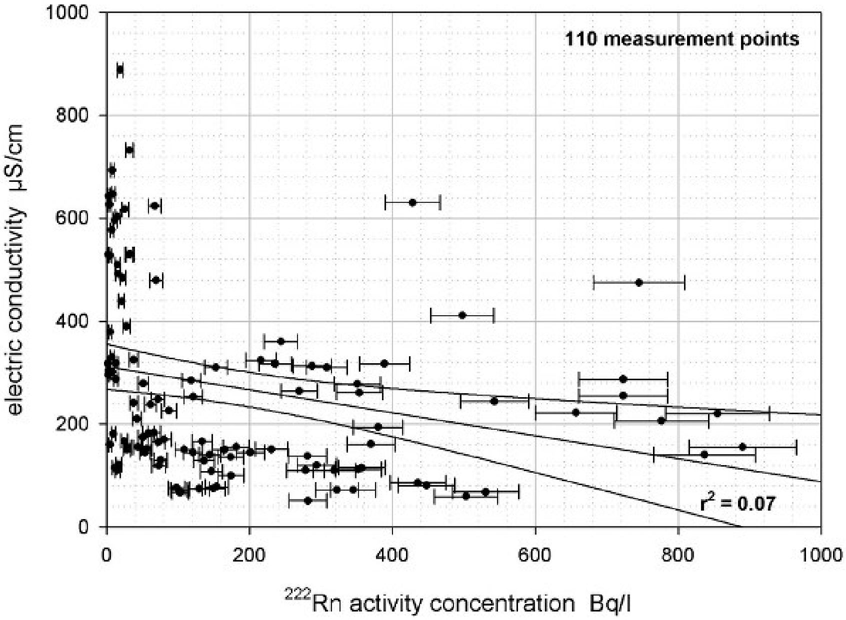

Figure 13.8: *Correlation between electric conductivity and ^{222}Rn activity concentration in drinking water*

Chapter 14

Relation of Heavy Metals and Some Other Elements to the Radioactivity of Drinking Water

14.1 General Background

The principal **natural sources of heavy metals** in the environment are rocks and soils. There are three main rock types. First the magmatic rocks, which are crystallized from magma from the earth mantle with a large variety of different chemical elements. Heavy metals are incorporated as trace elements into the crystal lattice of the magmatic minerals. Secondary the **sedimentary rocks**, which originate from particles of weathered rocks over geological periods of times and can be classified by their grain size (e.g. gravel, sand, silt, clay). Their porous structure enables them to hold fluids and their permeability enables them to transport fluids. They may contain ore deposits of many heavy metals if they are penetrated by ore-bearing hydrothermal fluids. The third rock types are the **metamorphic rocks** generated by metamorphosis. The chemistry of surface waters (rivers, springs, ponds, lakes) is greatly influenced by the kind of soil and rock the water flows on or flows through (Bradl, 2005). These **chemical, physical and biological parameters** affect the **solubility** and **mobility** of **heavy metals**. There are various possibilities for the fate and transport of heavy metals in soil and groundwater.

The main **anthropogenic sources of heavy metals** in groundwater, which is a very direct source of drinking water, are agricultural and industrial activities, landfilling, mining and transportation.

Heavy metals have different anthropogenic main sources and uses through which they can be introduced into the environment. Bradl (2005) listed them in his book.

The **mobility** of heavy metals in environmental media is influenced by chemical driving factors such as pH, redox potential and chemical speciation of an element. Each heavy metal has an individual behaviour, chemical and physical characteristics and individual sources and applications and ecotoxicological effects.

It is not possible to describe all these parameters in this context, but for this work the heavy metals in drinking water and their correlation to radionuclides are studied. Different radionuclides and heavy metals and some other elements were determined with regard to their possible correlations.

14.2 Results

In the framework of this thesis up to **145 water samples** of the detailed sampling phases were analyzed in the laboratory of the AGES Vienna for **different (heavy) metals** (^{107}Ag, ^{75}As, ^{11}B, ^{138}Ba, ^{9}Be, ^{114}Cd, ^{59}Co, ^{52}Cr, ^{63}Cu, ^{54}Fe, ^{57}Fe, ^{69}Ga, ^{7}Li, ^{24}Mg, ^{55}Mn, ^{98}Mo, ^{58}Ni, ^{208}Pb, ^{85}Rb, ^{82}Se, ^{88}Sr, ^{128}Te, ^{205}Tl, ^{51}V, ^{64}Zn, ^{66}Zn) by ICP-MS or ICP-OES. The concentration of the analyzed elements in drinking water are all given in **part per billion (ppb)**. Not all of these elements are heavy metals, like ^{7}Li, ^{9}Be, ^{11}B, others are not even clear metals like ^{75}As. But all these elements were analyzed together in the AGES laboratory and processed for further investigations as ”heavy metals”. These analyses were principally not planned to be a major part of this thesis and the drinking water project in Upper Austria, but were carried out in this framework because of the existence of the well collected water samples to get data for further interpretations and studies. The huge amount of collected heavy metals data needs time to analyze and process them in detail. Some **data processing** was started in the framework of this study, but mainly with regard to possible **correlations between (heavy) metals and radionuclides in drinking water**. The provided data by the AGES were additionally not very well edited and it was several days work to filter the huge amount of data for different questions and convert them into a data base. But now the data base for heavy metal in these 145 drinking water samples is prepared and can be used for further studies, analyses and interpretations.

For some analyzed heavy metals the concentration results are nearly all below decision limit (e.g. ^{52}Cr, ^{89}Mo, ^{58}Ni, ^{128}Te) and for some not all samples were analyzed (e.g. ^{11}B, ^{9}Be). But as a main result, **all analyzed drinking water samples have (heavy) metal concentrations below the parameter limits** defined in the **Austrian drinking water directive** (Republik Österreich, 2001) and the ÖNORM M 6250 (Austrian Standards Institute, 1986). So it seems that the analyzed drinking water samples of Upper Austria are not polluted by heavy metals.

In the framework of this thesis some possible correlations between heavy metals and radionuclides are surveyed and discussed in this chapter.

As a first step it was searched for **possible correlations** between **uranium** and all **other analyzed heavy metals**, but **no clear correlations were found**. The IAEA (1988) also reports, that a correlation of uranium with other constituents and elements in natural waters is commonly erratic or poor in most regions. The most frequent positive correlation are those with SiO_2, HCO_3^-, PO_4^{3-}, SO_4^{2-}, F, Cl, Na, K, Mg, Ca, Sr, V, Se, Mn, Rn, humic matter, pH, hardness, specific conductivity and total dissolved solids. Less frequent positive correlation includes those with Li, Cu, Ba, Ra, Zn, B, Y, rare earth, Ti, Zr, As, Fe, Co and Ni.

In many cases the r^2 value (defined in Chapter 11) of the correlation between ^{238}**U** and **other heavy metals** is clearly below 0.1 and no correlation is detectable at all (Figure 14.3). It is also a problem for correlation surveys, that the ^{238}U activity concentrations as well as most of the other heavy metals concentrations are usually rather low. In some cases single samples show a higher ^{238}U as well as another heavy metal concentration, but no general correlation or trend can be identified. Figure 14.1 and 14.2 illustrate an example for this situation. Higher ^{238}**U and** ^{11}**B** concentrations were detected at one sample (H1 – the private well in central Mühlviertel with a high radon and the second highest ^{226}Ra and ^{210}Pb activity concentration) and a rather good correlation exists (r^2=0.71) (Figure 14.1). But by removing the measurement point H1 in the correlation consideration, correlation can not be detected any more (r^2=0.08) (Figure 14.2).

For some heavy metals a slight correlation trend with ^{238}U was detected, but in the range of r^2 values of 0.1 to 0.2 (e.g. ^{51}V, ^{69}Ga) – but in Figure 14.3 an example for no correlation is illustrated, which stands for the most surveyed cases. The figure demonstrates the correlation between ^{238}**U and** ^{55}**Mn**, with an r^2 value of 0.001. The axes are in a logarithmic scale for more clearness in the lower ranges.

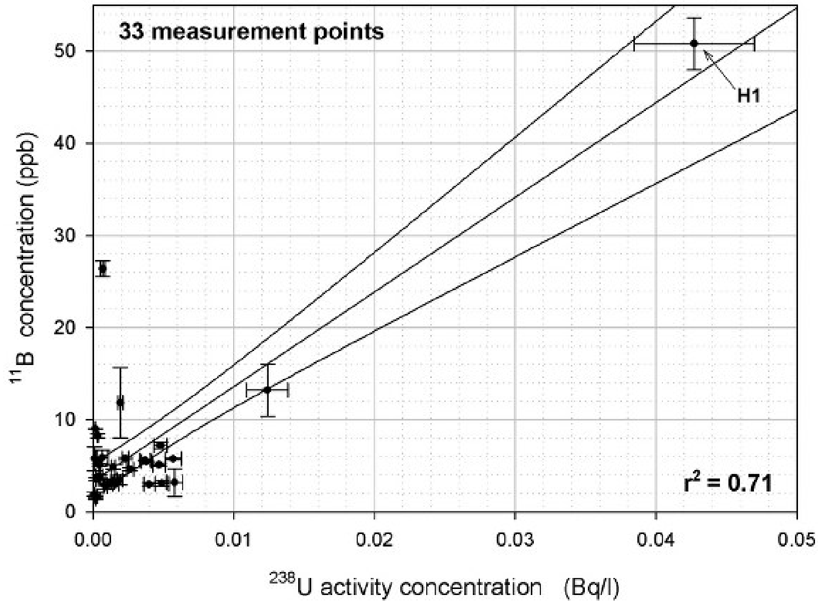

Figure 14.1: *Correlation between* ^{238}U *and* ^{11}B

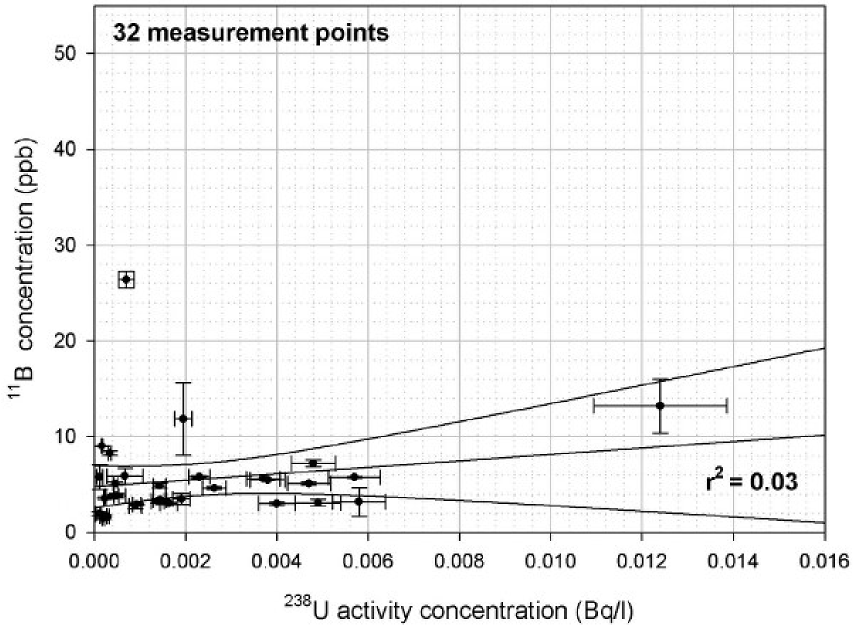

Figure 14.2: *Correlation between ^{238}U and ^{11}B without taking into account the measurement point H1*

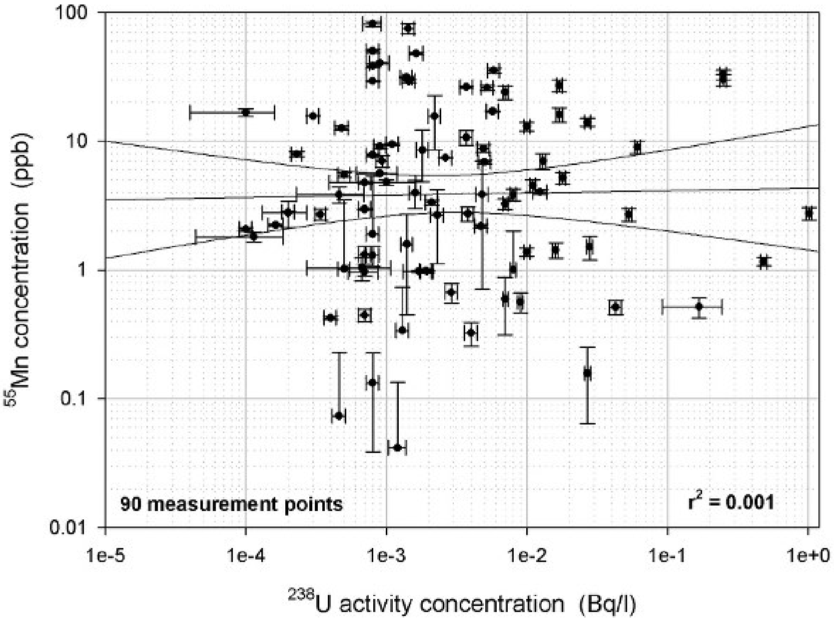

Figure 14.3: *Correlation between ^{238}U and ^{55}Mn*

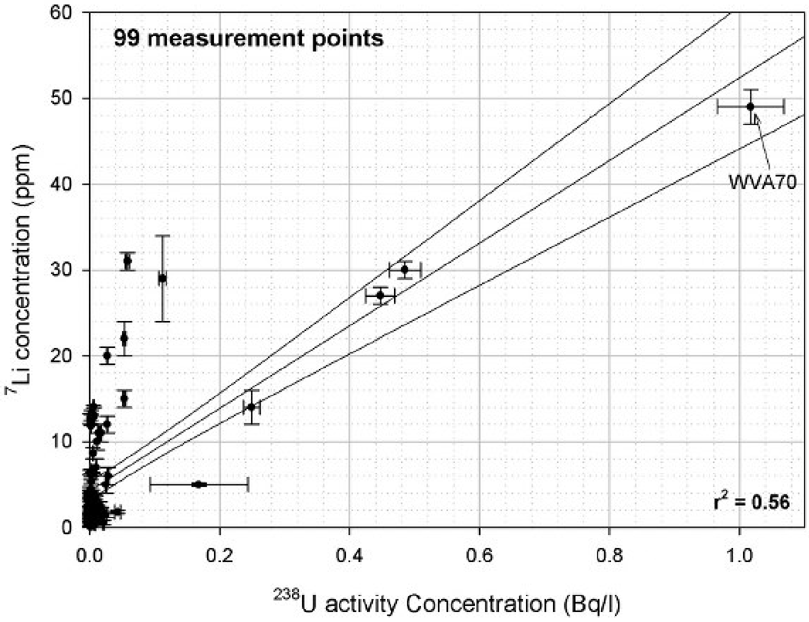

Figure 14.4: *Correlation between ^{238}U and ^{7}Li*

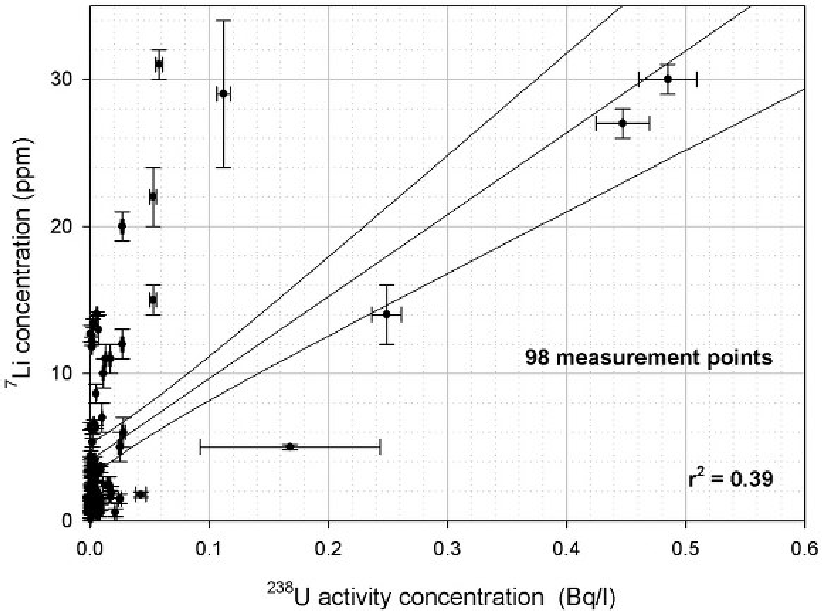

Figure 14.5: *Correlation between ^{238}U and ^{7}Li without taking into account the measurement point WVA70*

A second rather clear correlation was detected between ^{238}**U and** ^{7}**Li**, which is actually not a heavy metal at all. 99 samples are above decision limit for both ^{238}U and ^{7}Li and a correlation of r^2=0.52 was detected (Figure 14.4). The measurement point with both the highest ^{238}U activity and ^{7}Li concentration in the water is WVA70, the deep well in the lower Mühlviertel, which generally showed the highest ^{238}U activity concentration. By removing this point in the correlation consideration again the r^2 value is reduced to 0.39, which is nevertheless a trend of correlation (Figure 14.5).

Due to the correlation between ^{238}U and ^{7}Li, and the clearly higher ^{238}U and ^{7}Li concentration at the measurement point WVA70 in the lower Mühlviertel, more survey is done with regard to **geological or geographic dependencies**. The measured ^{7}Li concentrations in drinking waters were classified to their geologic and geographic origin (Figure 14.6). Higher ^{7}Li concentrations were detected in the drinking waters in the region of the Danube fault (median about 7 ppb) and slightly higher in the lower Mühlviertel (median about 3 ppb), in the other regions the detected median is about 2 ppb. So for further considerations of the correlation between ^{238}U and ^{7}Li a ratio between these two measurement values was calculated and also classified and displayed for different geological and geographic regions (Figure 14.7).

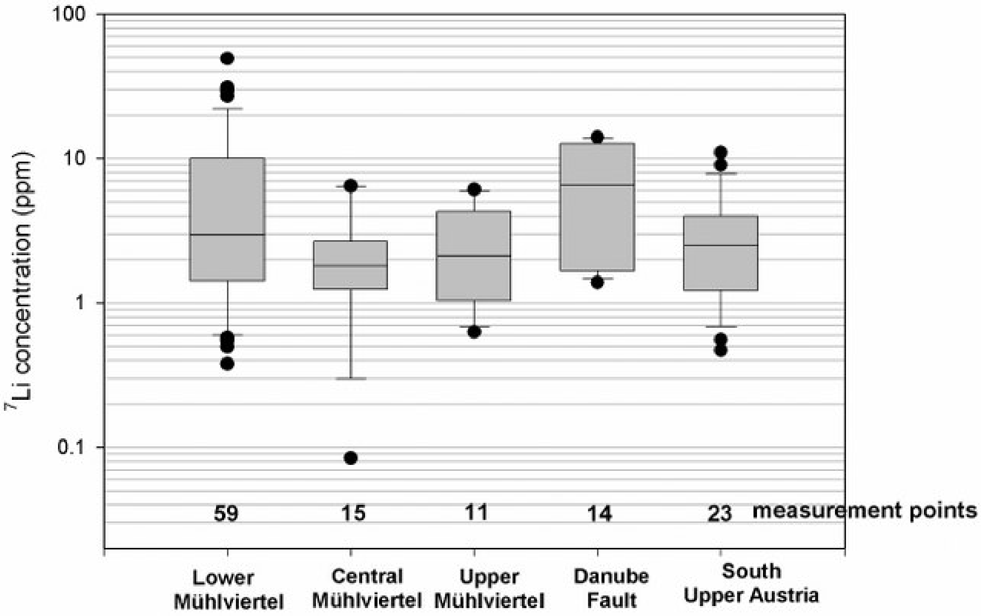

Figure 14.6: *^{7}Li concentration in drinking water in different regions of Upper Austria*

In the **lower Mühlviertel**, the **ratio** between ^{7}**Li (ppb)** and ^{238}**U (mBq/l)** is nearly one, so this could imply a **good correlation**. In the Danube fault the ratio is about 7, because of the highest ^{7}Li concentrations in the drinking water there and rather low ^{238}U activity concentrations. In opposite the ratio in southern Upper Austria is very low with about 0.15, because of rather low ^{7}Li concentrations and medium ^{238}U activity concentrations there. The ratios in the different geological regions seem to indicate better or less correlations between ^{238}U activity concentration and ^{7}Li concentration. Thus correlations were surveyed again in the classified regions in a following step. The correlation between ^{238}U and ^{7}Li in drinking water samples in the lower Mühlviertel is actually better than in entire Upper Austria with an r^2 value of 0.62 (Figure 14.8), as expected because of the ratio in Figure 14.7. Disregarding the highest measurement point WVA70, the r^2 value is reduced to 0.44, but this is nevertheless the highest detected correlation.

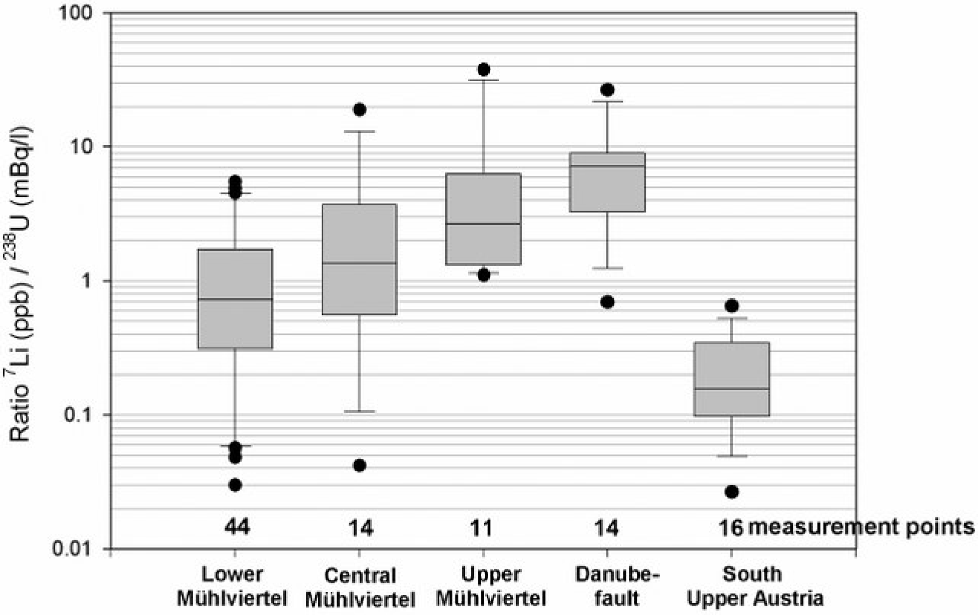

Figure 14.7: *Ratio between ^{7}Li and ^{238}U concentrations in drinking waters in different regions of Upper Austria*

For the **other regions** the **ratios and correlations do not fit well**. The ^{7}Li/^{238}U ratio of the lower Mühlviertel indicates a rather good correlation, but calculations results in an r^2 value of 0.003 (Figure 14.9). On the other hand in the Upper Mühlvierel an r^2 value of 0.38 was determined for the ^{7}Li concentration and ^{238}U activity concentration, although the ratio is higher than in the lower Mühlviertel. For southern Upper Austria the r^2 value for the correlation is 0.1 and for the Danube fault 0.18, which can not really be interpreted as a significant correlation.

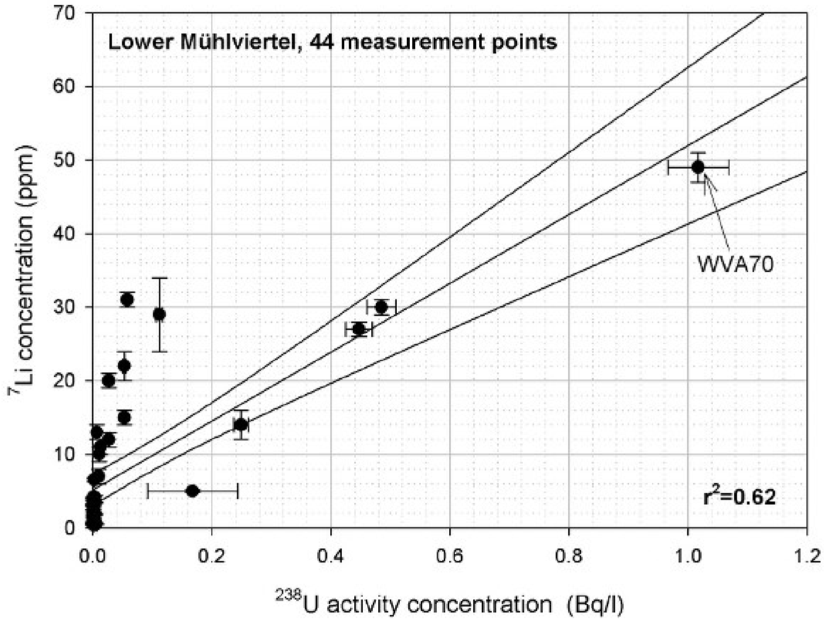

Figure 14.8: *Correlation between ^{238}U and ^{7}Li in drinking water in the lower Mühlviertel*

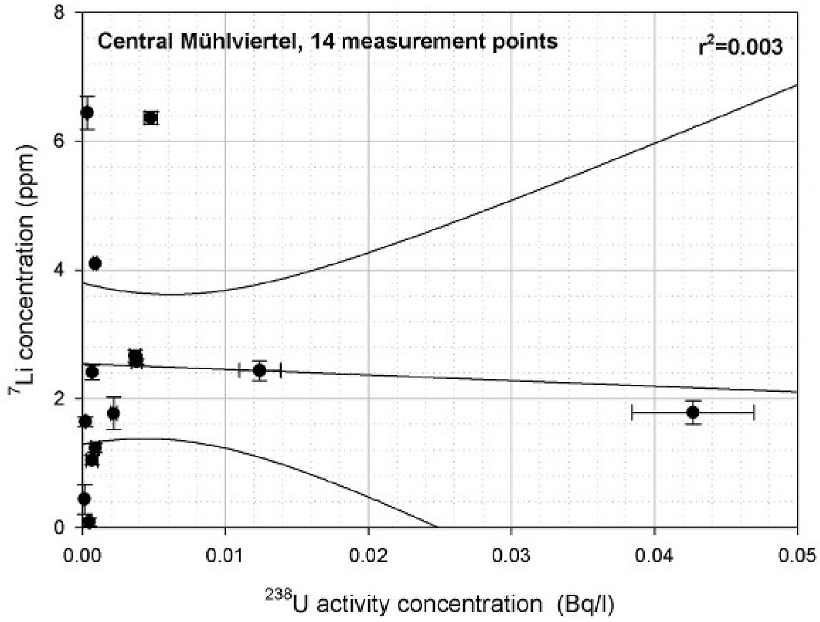

Figure 14.9: *Correlation between ^{238}U and ^{7}Li in drinking water in the central Mühlviertel*

Of the above discussed most frequent positive correlations reported by the IAEA (1988) only a **slight V** and a **rather clear Li correlation was verified** within these measurements.

It was also searched for **correlations between other radionuclides** and **heavy metals**. As discussed before only 7 samples have a ^{226}Ra activity concentration above decision limit. So correlations studies can not be carried out. For ^{3}H all measured activity concentrations are very low and within a close range, so correlation studies with other nuclides are not reasonable. But a survey for ^{228}Ra, ^{210}Pb and ^{210}Po and correlations with (heavy) metals were done. Not all analyzed (heavy) metals were analyzed for possible correlations with these radionuclides but only some mainly selected based on literature. A first obvious possible correlation seems to be existing between the radioactive lead isotope ^{210}**Pb** and the analyzed **stable heavy metal isotope** ^{208}**Pb**. 52 samples have as well a ^{210}Pb and ^{208}Pb concentration above decision limit, but no correlation can be detected (r^2=0.0004). Bradl (2005) reports that Pb has a binding to Fe oxides and a very strong binding to Mn oxides. So possible correlation with the measured Mn and Fe isotopes (^{55}Mn, ^{54}Fe, ^{57}Fe) were analyzed, but no correlations could be detected. The r^2 values of the correlation between ^{210}**Pb and** ^{54}**Fe or** ^{57}**Fe** are in the range of below 0.001, of ^{210}Pb and ^{55}Mn 0.03 (Figure 14.10). The best detected correlation is again between ^{7}**Li and** ^{210}**Pb** with an r^2 value of 0.36 (Figure 14.11). This correlation is mainly influenced by the measurement point WVA55, the deep well in the lower Mühlviertel, in which the clearly highest ^{210}Pb activity concentration was detected. In this sample also the clearly highest ^{7}Li concentration was detected. Disregarding this highest measurement point WVA55, the r^2 value is reduced to 0.02 and no significant correlation exists any more.

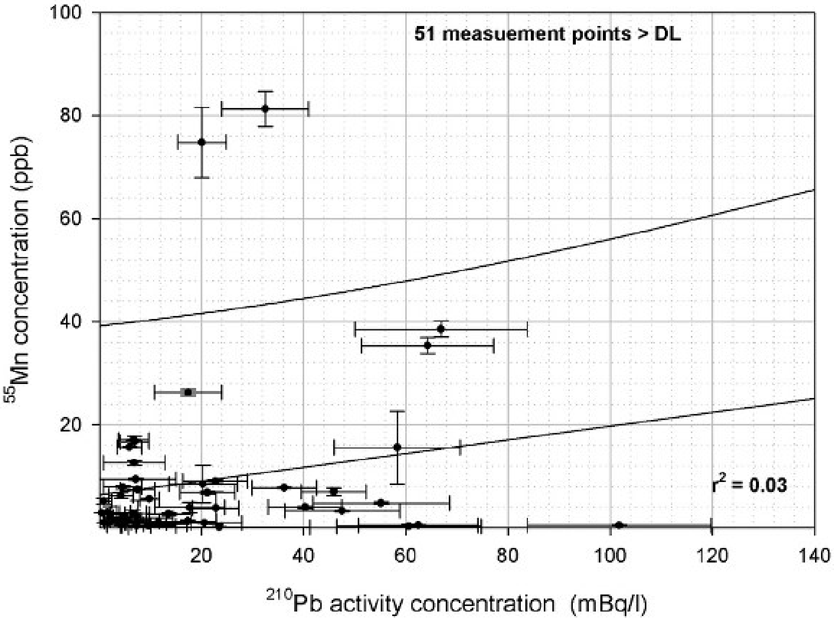

Figure 14.10: *Correlation between* ^{55}*Mn and* ^{210}*Pb*

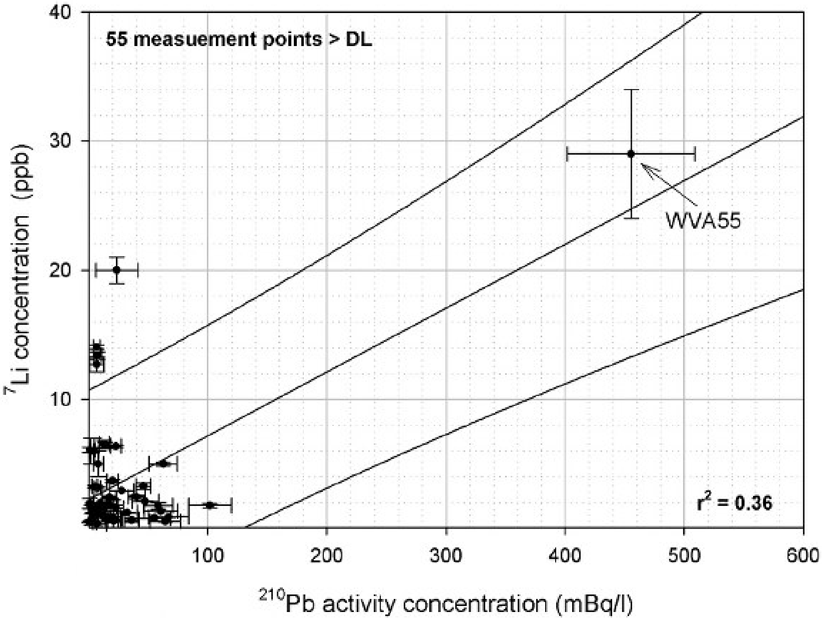

Figure 14.11: *Correlation between ^{7}Li and ^{210}Pb*

Also no correlation exists between ^{210}**Po and** ^{208}**Pb** (r^2=0.001) and a slight correlation trend can be interpreted between ^{210}**Po and** ^{7}**Li** (r^2=0.19). For ^{228}**Ra and** ^{7}**Li** no correlation was found (r^2=0.016), but a rather high r^2 value was detected for the correlation between ^{228}**Ra and** ^{208}**Pb** with 0.34, but only 21 measurement points have ^{228}Ra and ^{208}Pb concentrations above decision limit (Figure 14.12).

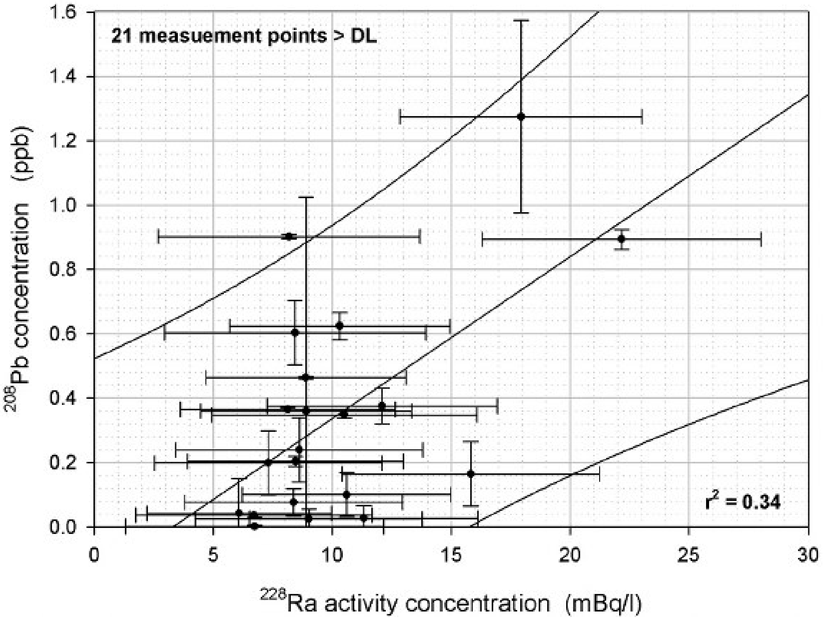

Figure 14.12: *Correlation between ^{208}Pb and ^{228}Ra*

The studies and surveys in this chapter shows, that **no clear correlations between radionuclides and heavy metals or other metals exist** and could be detected in the analyzed water samples. The correlation between ^{7}Li and some radionuclides mainly was caused because of the high ^{7}Li concentration in the single samples with higher ^{238}U activity concentration (WVA70) and ^{210}Pb (WVA55). But it is also obvious, because the alkaline metal Lithium occurs natural in some rocks and stones and in form of its salt it is contained in several mineral waters, and a possible correlation with uranium was reported by the IAEA (1988).

The data of the analyzed (heavy) metals in drinking waters of Upper Austria were collected and processed in a data base and are available for further studies for different thematic questions, not only for radionuclides correlation surveys which were carried out in the framework of this thesis, because of its main theme and interest of natural radionuclides in drinking water.

Chapter 15

Natural Radioactivity in Water Regarding Geology and Hydrogeology

In Chapter 7 and 8 the measurement results were analyzed for different radionuclides and related to their geographic region within Upper Austria. Major attention was spent on the results from tap water samples taken directly at consumers' houses because of radiation protection reasons. In this chapter more basically water sample results are analyzed for their **geological attributes** and therefore mainly samples taken **directly at wells or springs** are taken into account. These studies are rather scientific and not immediately applicable for radiation protection of the population, but a fundamental knowledge of natural radioactivity in drinking and ground water regarding their geology will be a good basis for **future dose assessments for the population** living in certain regions by natural radioactivity in drinking water.

In Chapter 4.3 the **basic geology and hydrogeology situation in Austria** and Upper Austria is discussed. According to the classifications and geology characterisations in this chapter studies will be done here.

The radionuclide activity concentration results of the drinking water (ground water) analyses are surveyed for geological correlations and therefore illustrated in the **hydrogeological map of Upper Austria** (Vohryzka (1973b), for the legend of the map see Annex A1) and with other geological characteristics like **geological faults**. Diagrams to characterize correlations of radionuclide concentrations and geological areas are shown and some sampling points with noticeable results are surveyed regarding geology. Results of all measurement points from the survey sampling are used for the geological analysis, but only the samples taken directly at wells or springs before treatment (private wells, water supplies, "sacred wells") from the detailed sampling phase are taken into account in the geological survey.

Based on the hydrogeological map by (Vohryzka, 1973b) a simplified characterization of the hydrogeological zones were defined in Chapter 4.3. In the following the areas of this characterization are labeled with the corresponding number in brackets for an easier attribution – e.g. (1) for the area *Weinsberger Granit*.

15.1 Radon-222 – Survey Sampling

Figure 15.1 illustrates the classified radon activity concentrations from the survey sampling in the **hydrogeological map of Upper Austria** (Vohryzka, 1973b). Measurement results in class 3 were only detected in the Bohemian Massif, as well as in the *Weinsberger Granit* (1) and in the Gneiss and Granite of the Bohemian Massif (2). Radon activity concentrations in class 2 were detected in all geological areas.

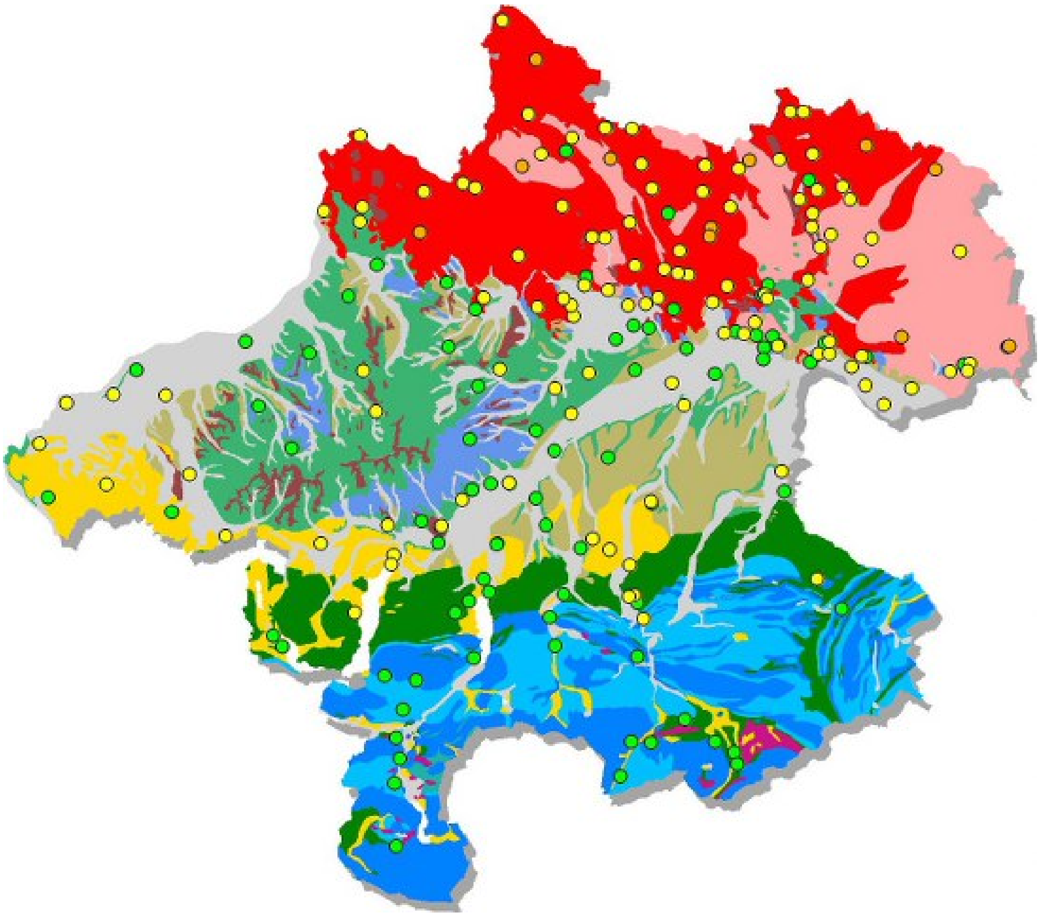

Figure 15.1: *Classified radon activity concentrations of the survey sampling illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b), legend see Annex A1*

In Figure 15.2 the radon results are differed into the 10 geological areas according to the hydrogeological map of Vohryzka (1973b) (see Chapter 4.3). For clarity the y axis is in a logarithmic scale. As expected, the radon activity concentrations in the Bohemian Massif are clearly higher than in the other geological areas. The results in the *Weinsberger Granit* (1) are also higher (median about 50 Bq/l) than in the Gneiss and Granite of the Bohemian Massif (2) (median about 30 Bq/l), although the sampling points with the highest measured radon activity concentration belong to geological area (2). But in the Gneiss and Granite of the Bohemian massif area many different gneiss and granite types with different uranium and thorium concentrations are combined, so more detailed geological maps are necessary to distinguish between them. A higher radon activity concentration in the *Weinsberger Granit* corresponds with the results found by Schubert et al. (2003) – see Chapter 4.3. In this study the highest radon activity concentrations were also found in a deep well in the *Weinsberger Granit*, although the uranium concentration in this granite is rather low.

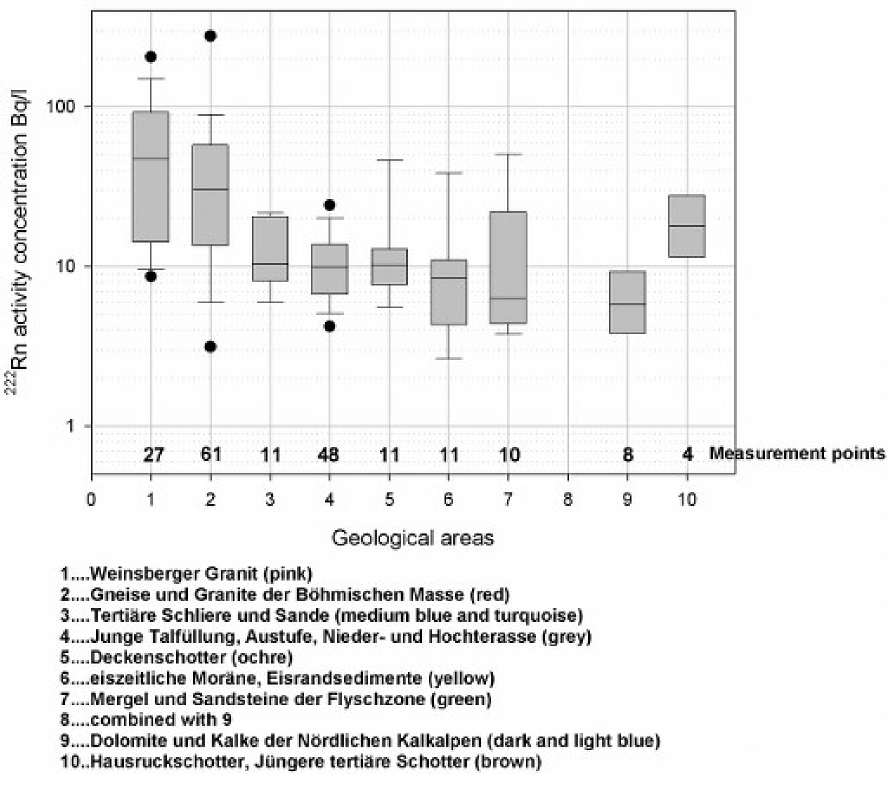

Figure 15.2: *Radon activity concentrations of the survey sampling in the different geological areas of Upper Austria*

Figure 15.3 shows the radon activity concentrations in the different **ground water bodies** according to Amt der Oberösterreichischen Landesregierung (2008) – see Chapter 4.3. As expected the median radon activity concentration in the ground water body Bohemian Massif is clearly above all others with about 40 Bq/l. The median radon activity concentrations in all other ground water bodies are similar with about 8 to 15 Bq/l, with little higher concentrations in the *Salzach-Inn-Mattig* and *Traun-Enns-Platte* – which are both mainly out of *Deckenschotter* and *junge Talfüllungen* according to Vohryzka (1973b). The lowest median radon activity concentration was detected in the ground water body *Flysch* (below 5 Bq/l) corresponding with the median radon activity concentration of the geological area *Flysch* (7) according to Vohryzka (1973b) (Figure 15.2).

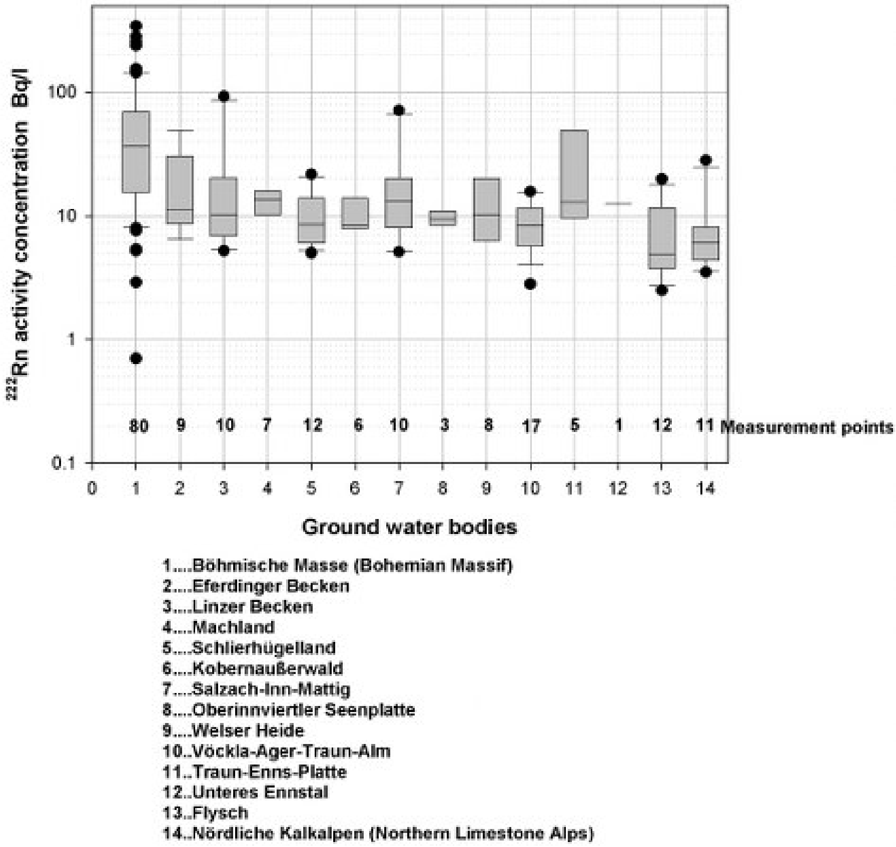

Figure 15.3: *Radon activity concentrations of the survey sampling in the different ground water bodies of Upper Austria*

Geological faults are an important geological characteristic. Figure 15.4 shows a map of Upper Austria with main geological faults and the classified radon activity concentrations of the survey sampling. In geological faults radon from the deep can exhaust to the surface and may influence the radon concentration in the ground water. The figure illustrates the main geological faults in Upper Austria – Danube fault, *Bayrischer Pfahl*, *Haselgraben* and some other faults (especially in the northern limestone Alps). Radon activity concentrations in class 3 were detected along the *Bayrischer Pfahl* fault, and close by the *Haselgraben* and the Danube fault. It may be that the waters from these sampling points close to the faults are influenced by these faults, because the coordinates are from the sampling point and not from the actual origin of the water. Additionally some radon activity concentrations in class 3 were detected at smaller faults in the district of Perg and Freistadt. It seems that a correlation between faults and radon activity concentration in the ground water exists in the Bohemian Massif. No correlations with faults were noticeable in the northern limestone Alps, because less uranium is in the deep bedrock and only not abundant faults. In the detailed sampling more samples were taken at the main faults in the Bohemian Massif to verify this theory (Chapter 8).

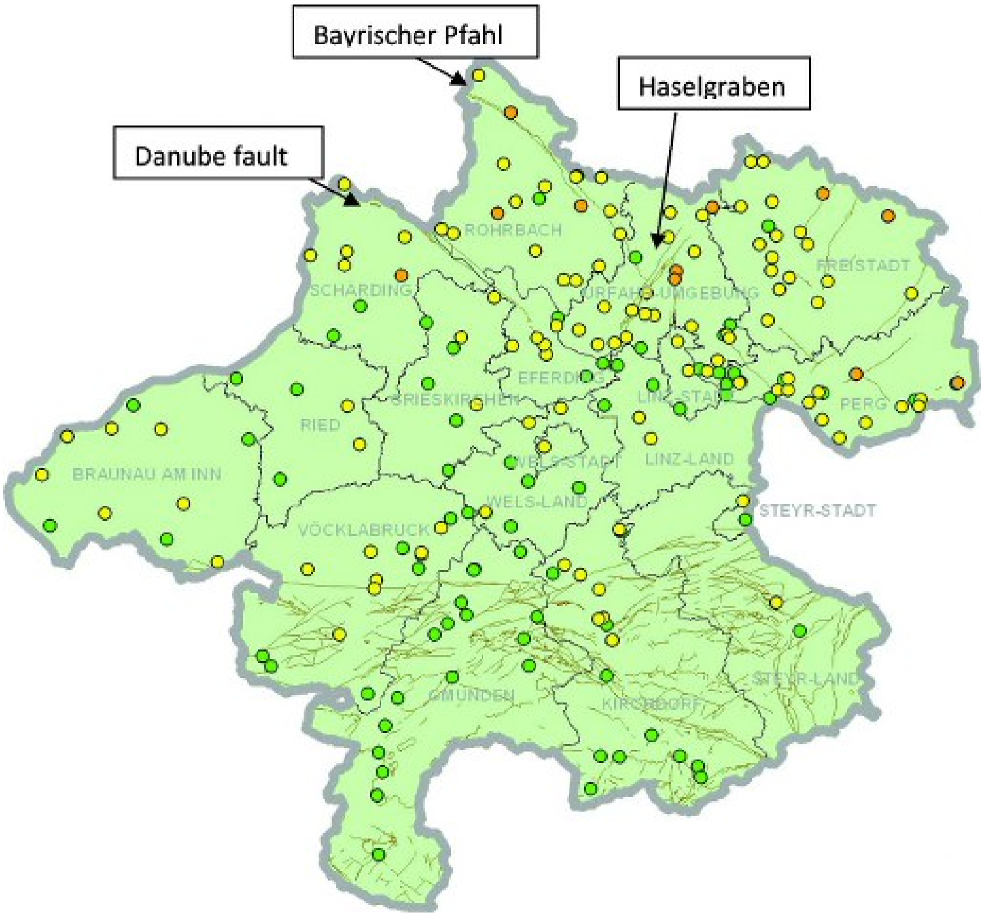

Figure 15.4: *Classified radon activity concentrations of the survey sampling illustrated in a map of Upper Austria with geological faults, legend see Annex A1*

15.2 Radon-222 – Detailed Sampling

In the detailed sampling phases samples were taken only at well selected sampling points according to the criteria in Chapter 8. These samples were taken directly from wells and springs as well as a at consumers' houses. As mentioned above for geological considerations only the **samples directly at wells and springs** and **before treatment** or **without longer duration of dwell** in pipes or tanks are of interest. So **113 samples** from the **detailed sampling phases** are related to these criteria and are used for the geological survey.

In Figure 15.5 the classified radon activity concentrations of the detailed sampling are illustrated in the **hydrogeological map** of Upper Austria (Vohryzka, 1973b).

The detailed sampling results show a similar situation as the ones from the survey sampling. Again radon activity concentrations in class 3 were only detected in the Bohemian Massif, as well in the *Weinsberger Granit* (1) as in the Gneiss and Granite of the Bohemian Massif (2). Only a few measurement points in the Bohemian Massif show radon activity concentrations in class 1. Outside the Bohemian Massif samples were taken mainly in the valleys of the foothills of the Alps – geological young valleys (*junge Talfüllungen* (4) according to Vohryzka (1973b)) and in the region of the northern limestone Alps. Some radon activity concentrations in class 2 were detected in these areas. The detailed sampling along the main **geological faults** verify the results of the survey sampling with radon activity concentrations in class 3, but no clearly higher radon activity concentrations were measured in these points than at other measurement points in the Bohemian massif. For fundamental statements on the impact of geological faults in the Bohemian massif on the radon activity concentration in the ground water more studies should be done, but a basic trend of correlation is noticeable.

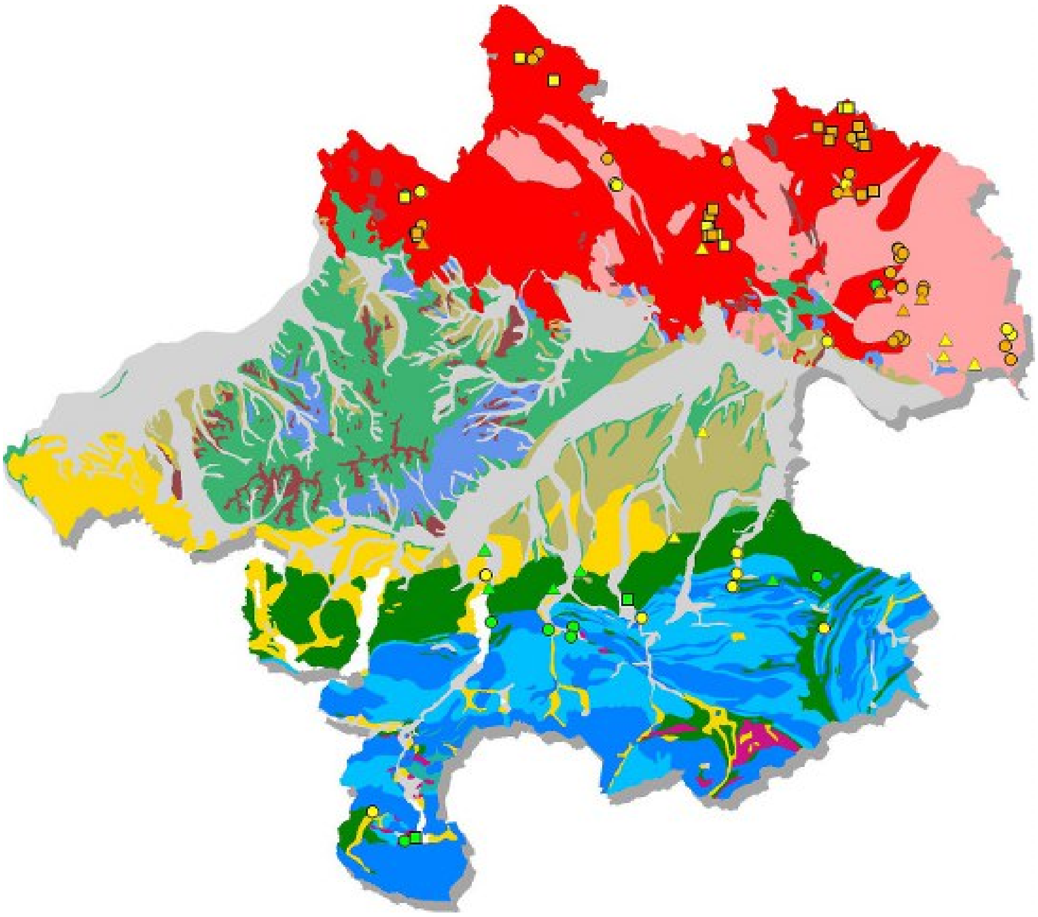

Figure 15.5: *Classified radon activity concentrations of the detailed sampling illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b), legend see Annex A1*

In Figure 15.6 the radon results are differed into the 10 geological areas according to the **hydrogeological map** of (Vohryzka, 1973b) (see Chapter 4.3). But samples were taken not in all of these areas, so only 8 areas are listed, and also in some of these 8 areas too few measurement results exist. For clarity the y axis is again in a logarithmic scale. As discussed in Chapter 8.1 – detailed sampling the radon activity concentrations analyzed in the detailed sampling are above the ones from the survey sampling, especially in the range of higher activity concentration. So the median of the detailed sampling in the *Weinsberger Granit* (1) is about 150 Bq/l, in the survey sampling below 50 Bq/l. In the Granite and Gneiss of the Bohemian Massif area (2) the radon activity concentration is about 200 Bq/l in the detailed sampling, about 30 Bq/l in the survey sampling. This is caused (as discussed in Chapter 8.1) by different sampling and on-site measurement on the one hand, and selected measurement points on the other. In the figure of the detailed sampling only the samples directly at wells and springs without treatment are taken into account. Noticeable is the higher median radon activity concentration in the Gneiss and Granite of the Bohemian massif area (2) than in the *Weinsberger Granit* (1) area in the detailed sampling, in contrast to the survey sampling. This would be expectable, because of the lower uranium content in the *Weinsberger Granit*, but may result from sampling. The sampling points of the detailed sampling were selected because of higher activity concentrations of the survey sampling and in some interesting regions with higher activity concentrations (and more available wells and springs) more wells and springs were surveyed, and this disequilibrium influences the median of the areas. So it would be very interesting and fundamental to study **different granite types** (uranium and thorium concentration) and the different radionuclide activity concentrations in the groundwater originating in this bedrock. These studies would go beyond the scope of this thesis but should be done in the future for a basic data net with respect to evaluations of areas for their **potential risk to the population or workers in water supplies** (according to the ”Ordinance on exposure due to natural radiation sources (Republik Österreich, 2008)) by radioactivity in drinking water.

For the other areas the difference between the survey sampling and the detailed sampling does not exist. For example the median of the radon activity concentration in the *junge Talfüllungen* (4) is about 8 Bq/l in the detailed sampling, but even slightly higher in the survey sampling with 10 Bq/l. A similar situation exists in the northern limestone Alps (9) with a radon activity concentration about 2.5 Bq/l in the detailed sampling and about 5.5 Bq/l in the survey sampling.

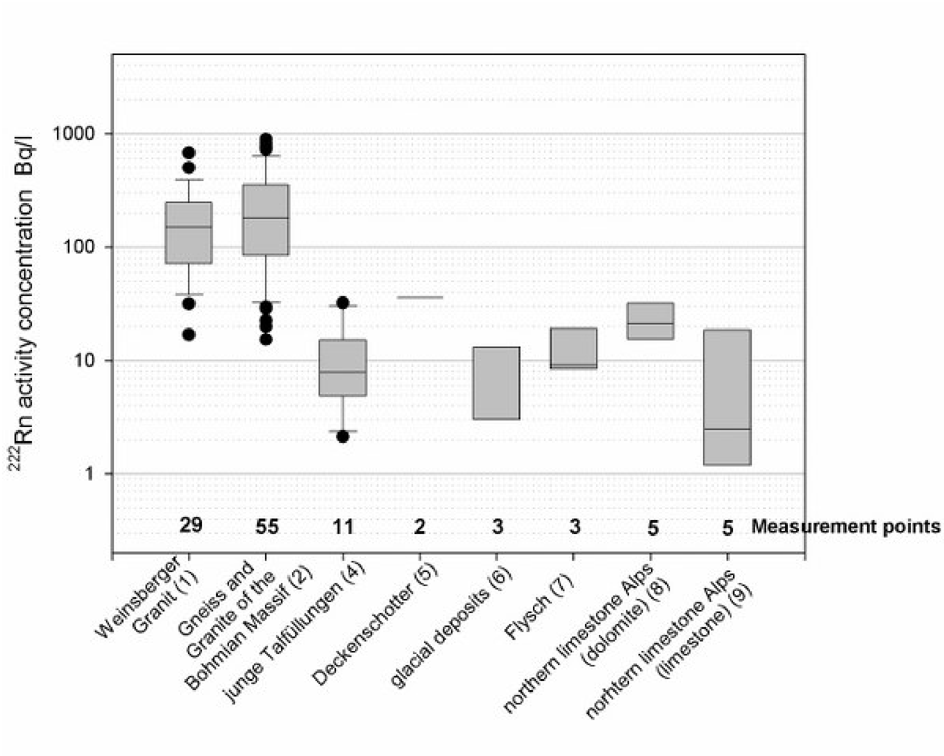

Figure 15.6: *Radon activity concentrations of the detailed sampling in the different geological areas of Upper Austria*

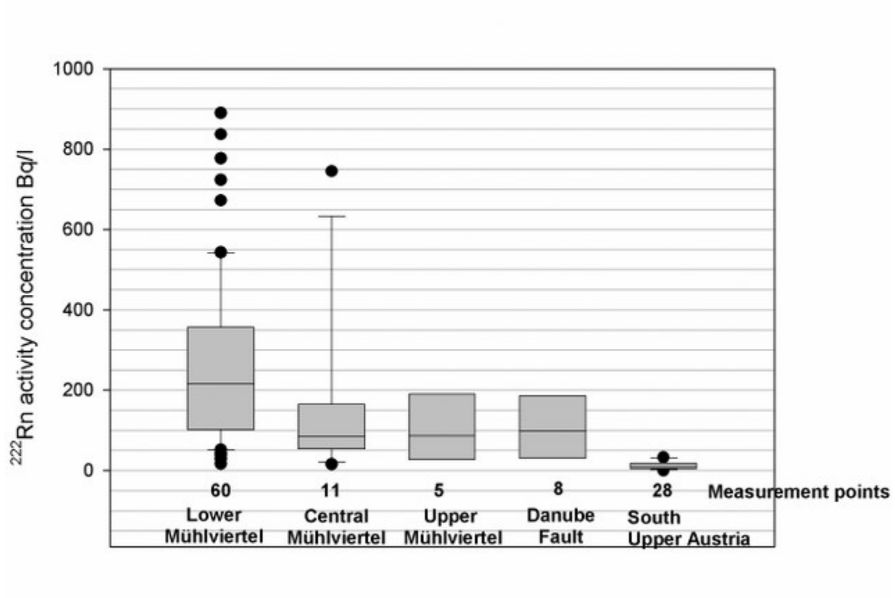

Figure 15.7: *Radon activity concentrations of the detailed sampling in a simplified geographic and geologic classification*

Figure 15.7 shows a **simplified classification** (Chapter 4.3) which is not fundamentally based on geology but divided the Bohemian massif more geographic in

Lower, Central, Upper Mühlviertel and Danube fault and the samples outside of the Bohemian massif as ”southern Upper Austria”. The figure shows a high median radon activity concentration in the lower Mühlviertel (more than 200 Bq/l) and much lower median radon activity concentrations in the central and upper Mühlviertel (less than 100 Bq/l). This fact may again be the result of the above mentioned different frequent sampling in different areas, but it also shows that it would be interesting to deepen studies in the field of the impact of granite and gneiss types to natural radioactivity in groundwater, to explain the difference between radon activity concentrations in different parts of the Bohemian massif.

For the results of the detailed sampling the classification into **ground water bodies** according to (Amt der Oberösterreichischen Landesregierung, 2008) – see Chapter 4.3 is not very reasonable because at the most only a few points are situated in each body except for the Bohemian massif (85) and the northern limestone Alps (14). The median radon activity concentration in the Bohemian massif is about 180 Bq/l, in the northern limestone Alps about 12 Bq/l.

15.3 Uranium-238 – Survey Sampling

Figure 15.8 shows the classified ^{238}U activity concentrations from the survey sampling in the **hydrogeological map of Upper Austria** (Vohryzka, 1973b). No measurement points exist in class 3, some in class 2, distributed in different geological areas. Some class 2 ^{238}U activity concentrations were detected in the *junge Talfüllungen* (4, grey). In Figure 15.9 the ^{238}U results are differed into the 10 geological areas according to the hydrogeological map of Vohryzka (1973b) (see Chapter 4.3). For clarity the y axis is in a logarithmic scale. It is considerable that no clear difference concerning the median uranium activity concentration is detectable in the 10 geological areas, not even in the *Weinsberger Granit* (1) and the granite and gneiss area of the Bohemian massif (2), as it would be expected. The highest median uranium activity concentrations were detected in the glacial deposits (6) and *Flysch* (7) with 0.01 Bq/l, which are both located at the foothill of the Alps. Further survey of uranium contents in different bedrocks will be reasonable and should afford explanations and generalizations in this topic.

The above discussed **geological faults** have no detectable impact on the ^{238}U activity concentration in the ground waters.

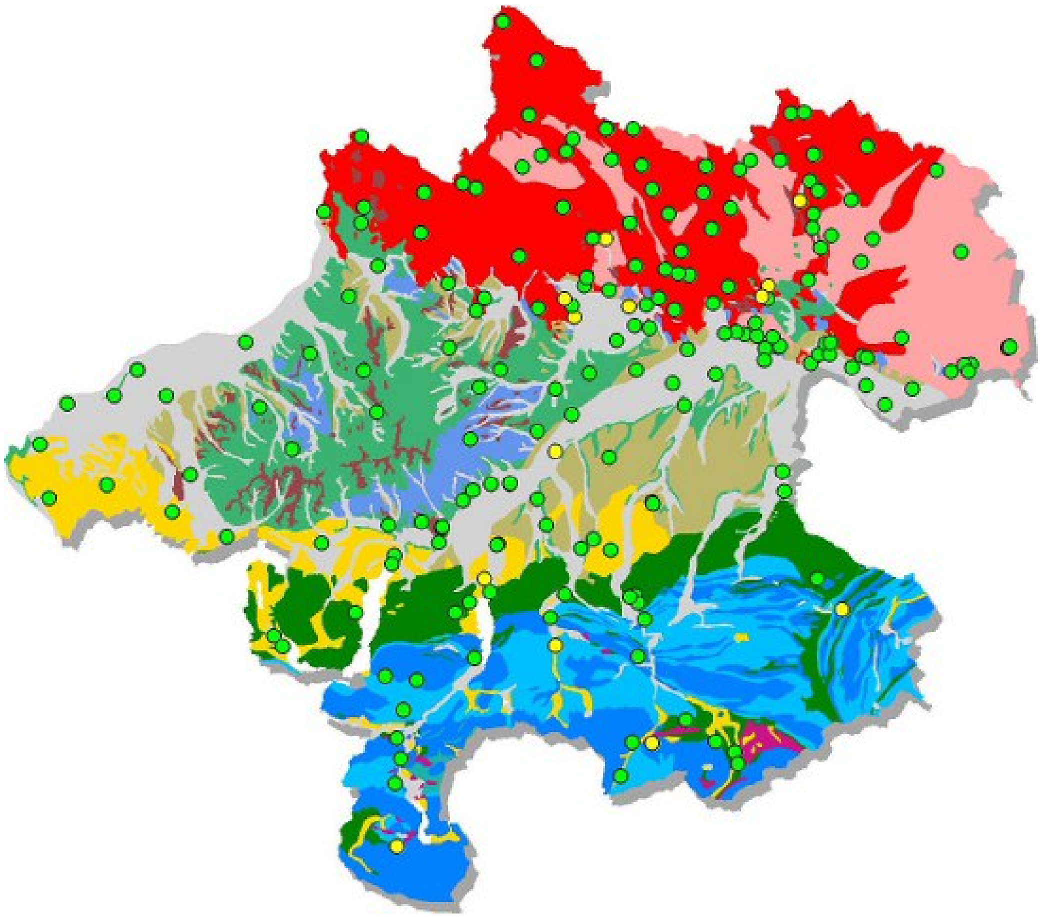

Figure 15.8: *Classified ^{238}U activity concentrations of the survey sampling illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b), legend see Annex A1*

The ^{238}U activity concentrations of the survey sampling are also classified according to the different **ground water bodies** (Amt der Oberösterreichischen Landesregierung, 2008) – see Chapter 4.3, Figure 15.10. The graph shows a similar situation to Figure 15.9 with no clear difference in the median ^{238}U activity concentrations in the different ground water bodies. Slightly higher median ^{238}U activity concentrations ($>$ 0.01 Bq/l) were detected in ground water bodies (3) and (10) – *Linzer Becken* and *Welser Heide*. These two ground water bodies are again both out of *junge Talfüllungen* according to Vohryzka (1973b) similar to the radon activity concentration results discussed above. It seems that the **young sediments of the valleys** in the **foothills of the Alps** are **potential bedrocks** with **higher natural radioactivity concentration** and influence the ground waters. This should also be surveyed in a further study about uranium and thorium content in different bedrocks and their impact on ground water.

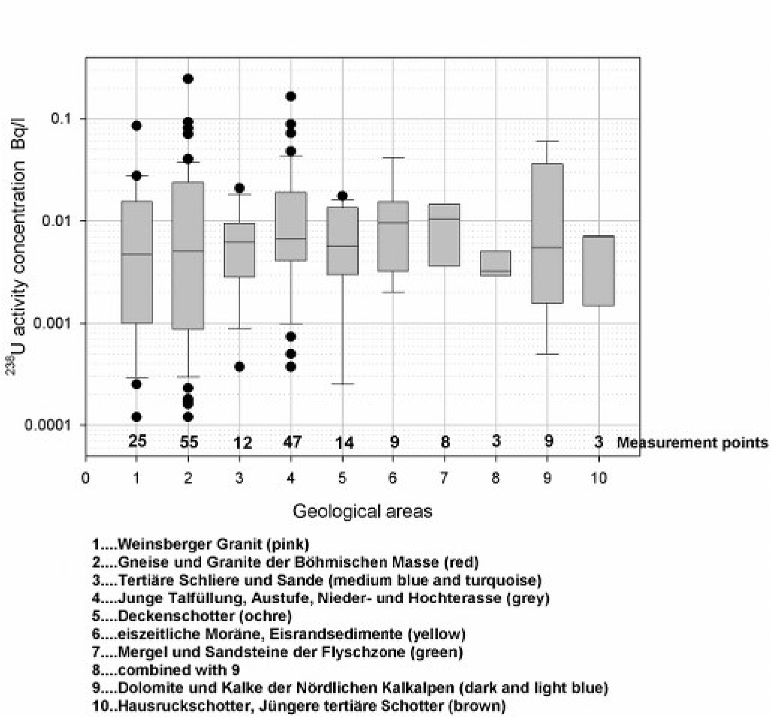

Figure 15.9: ^{238}U *activity concentrations of the survey sampling in the different geological areas of Upper Austria*

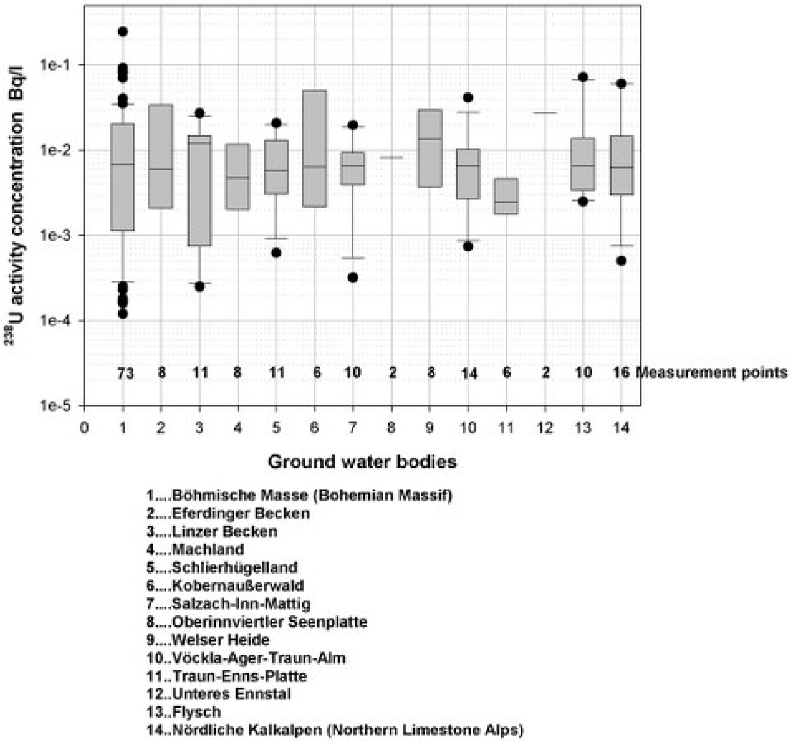

Figure 15.10: ^{238}U *activity concentrations of the survey sampling in the different ground water bodies of Upper Austria*

15.4 Uranium-238 – Detailed Sampling

80 of the 101 samples from the detailed sampling phase used for the geological survey are above decision limit for ^{238}U. ^{238}U activity concentrations were only detected in the Bohemian massif, more exactly in the *Weinsberger Granit* (1) and Granite and Gneiss area of the lower Mühlviertel (2). Figure 15.11 illustrates the ^{238}U activity concentrations in the **geological areas** according to Vohryzka (1973b), in Figure 15.12 the results are summarized in 5 areas – lower, central and upper Mühlviertel, Danube fault and southern Upper Austria. Although the highest ^{238}U activity concentrations (class 2 and 3) were detected in the lower Mühlviertel, the highest median activity concentration was detected in southern Upper Austria, especially in the *junge Talfüllungen* (4) and the dolomites of the Northern limestone Alps (8) (Figure 15.11). In these regions the ^{238}U activity concentrations in the samples have a smaller variation than in the Bohemian massif, but there are also less measurement points.

The **higher ^{238}U activity** concentrations in the **limestone Alps** correspond with reports stating that in geological uranium-rich areas the high uranium concentration in surface and underground water is often associated with a high carbonate concentration as uranium has a higher solubility due to the complex reaction with carbonates (Jia & Torri, 2007).

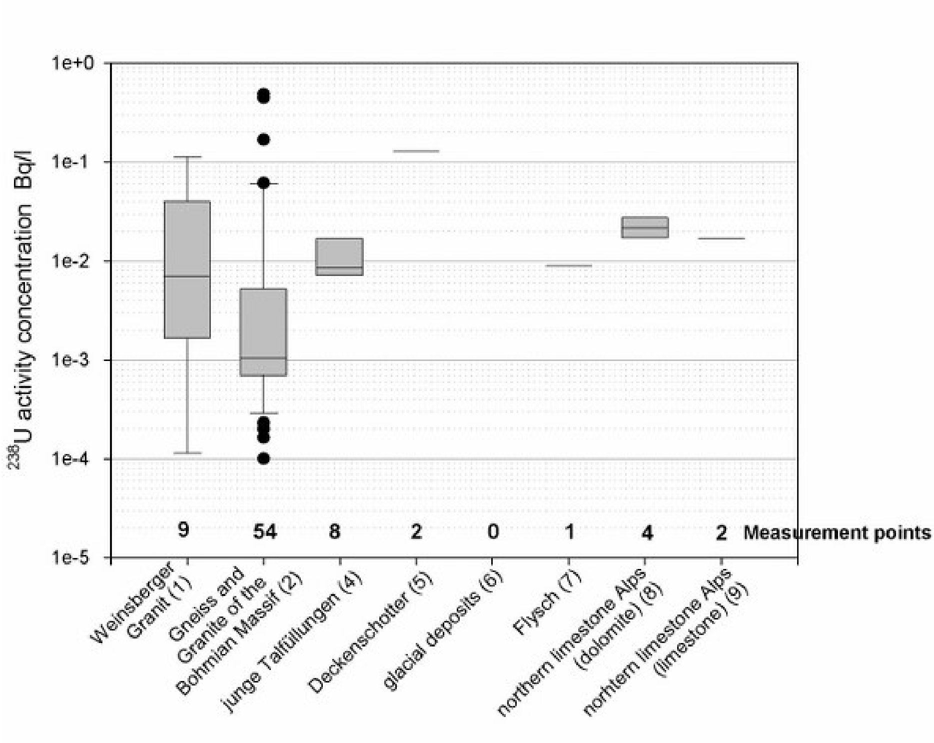

Figure 15.11: *^{238}U activity concentrations of the detailed sampling in the different geological areas of Upper Austria*

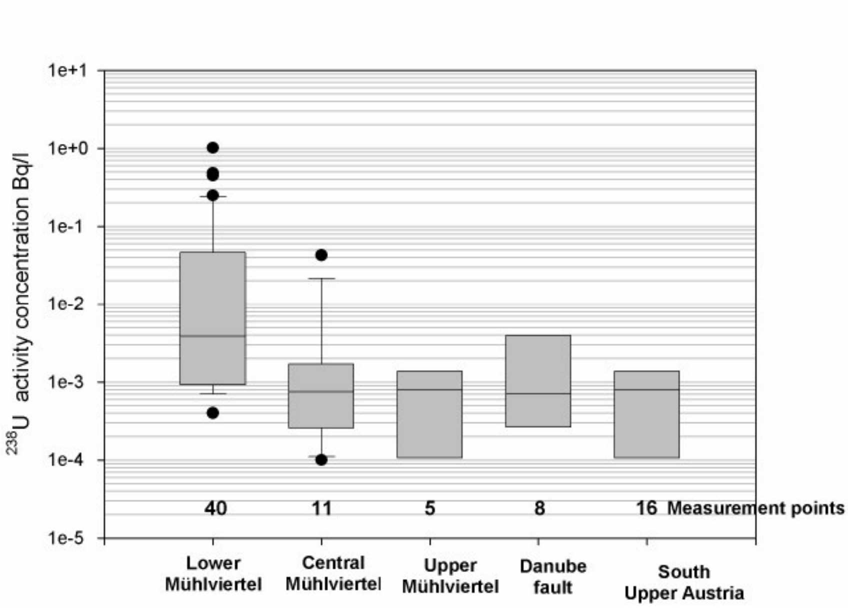

Figure 15.12: *^{238}U activity concentrations of the detailed sampling in a simplified geographic and geologic classification*

Again the classification into **ground water bodies** according to Amt der Oberösterreichischen Landesregierung (2008) is not very reasonable because very few points are situated in each body, except for the Bohemian massif (65). 7 measurement points with ^{238}U activity concentration above decision limit are situated in the northern limestone Alps, 10 in the ground water body *Vöckla-Ager-Traun-Alm*. The median ^{238}U activity concentration in the Bohemian massif is about 0.0015 Bq/l, in the *Vöckla-Ager-Traun-Alm* body 0.008 Bq/l and in the northern limestone Alps about 0.015 Bq/l. So this verifies the result of the other classifications that the median ^{238}U activity concentration in southern Upper Austria (northern limestone Alps) is one order of magnitude above the one in the Bohemian massif, although the highest single measured ^{238}U activity concentrations are located in the lower Mühlviertel (Bohemian massif). So it seems that the ^{238}U activity concentration in water is highly dependent on the different granite and gneiss types and therefore the fluctuation in this region is rather high. In southern Upper Austria the ^{238}U activity concentration is slightly higher in all water samples, but no single waters with clearly higher ^{238}U activity concentrations were detected. So these results again show that it would be interesting to deepen studies in the field of the **impact of different bedrocks** (e.g. granite and gneiss types, young sediments) on natural radioactivity in groundwater, to explain the difference between radon activity concentrations in different parts of the Bohemian massif and the foothill of the Alps.

15.5 Tritium

All measured ^{3}H activity concentrations of the survey sampling are in class 1, the detailed ^{3}H activity concentrations in **different geological areas according to Vohryzka (1973b)** are illustrated in Figure 15.13. No big differences are identifiable within the different geological areas. Only 44 of the 102 samples from the detailed sampling phase for the geological survey have an ^{3}H activity concentration above decision limit. According to the classification of Vohryzka (1973b), 40 of these samples are located in the granite and gneiss of the Bohemian massif area, 3 in the *Weinsberger Granit* and one in the *Flysch*. This sampling point is a "sacred well" and the only sampling point with an ^{3}H activity concentration above decision limit outside the ground water body Bohemian massif.

The ^{3}H activity concentrations of the survey sampling in the different ground water bodies according to Amt der Oberösterreichischen Landesregierung (2008) show a similar situation to Figure 15.13 with no clear differences in the median ^{3}H activity concentrations (about 1 Bq/l) in the different geological areas or ground water bodies (Figure 15.14).

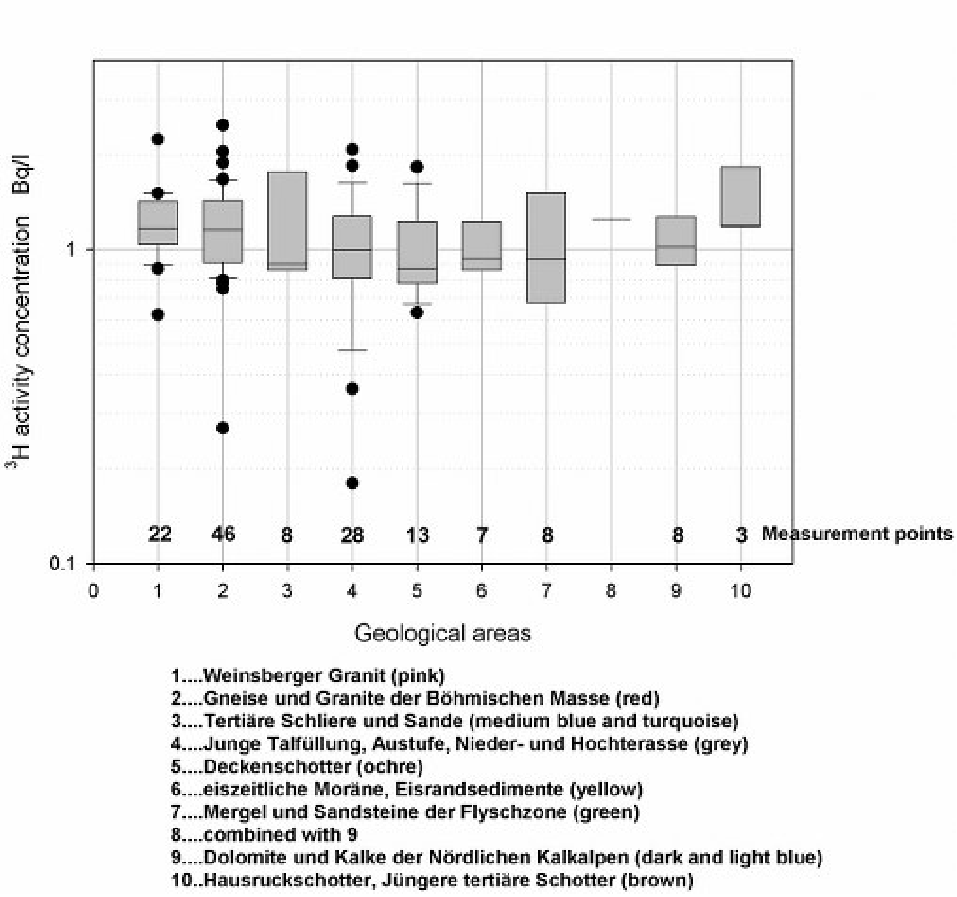

Figure 15.13: *^{3}H activity concentrations of the survey sampling in the different geological areas of Upper Austria*

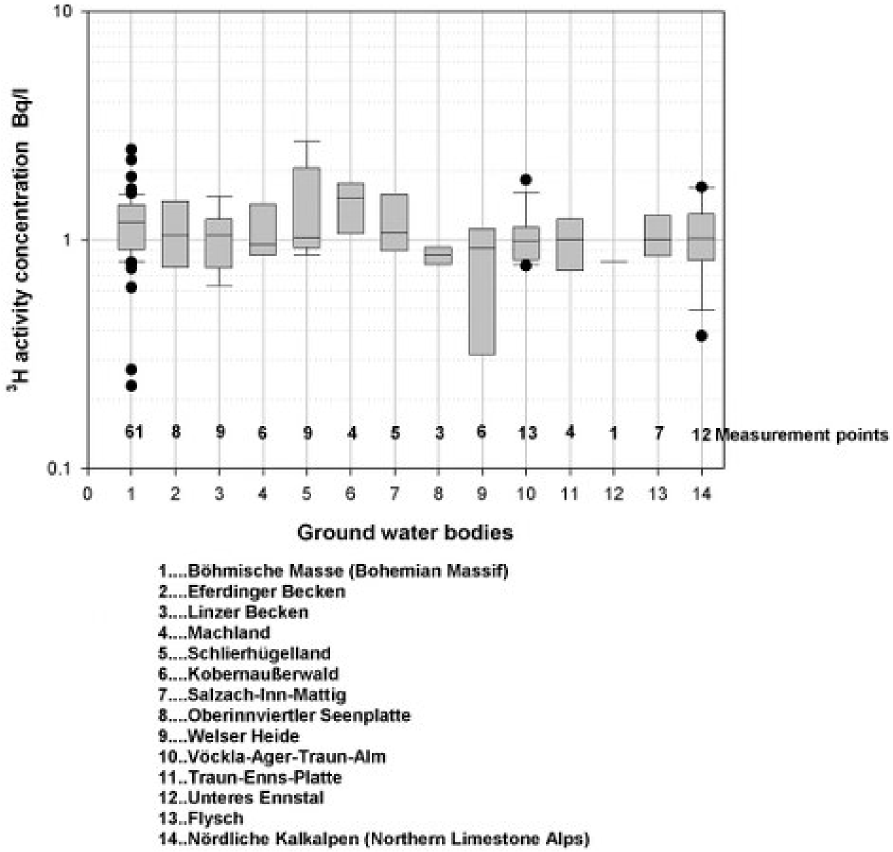

Figure 15.14: *^{3}H activity concentrations of the survey sampling in the different ground water bodies of Upper Austria*

15.6 Gross Alpha and Gross Beta

In the **survey sampling gross alpha-beta activity concentration** was analyzed in 203 samples, 18 are above decision limit. Only one sample shows a gross alpha-beta activity concentration in class 2, situated in the geological area *Deckenschotter* (5) according to Vohryzka (1973b). 5 sampling points with a gross alpha-beta activity concentration above decision limit are located in the geological area *Weinsberger Granit* (1), 4 in glacial deposits (6), 3 in the granite and gneiss areas of the Bohemian massif (2) and in the *Deckenschotter* (5), 2 in *Flysch* (7) and 1 in *junge Talfüllungen* (4). No sampling points with gross alpha-beta activity concentration above decision limit are situated in the northern limestone Alps. According to the classification in **ground water bodies** (Amt der Oberösterreichischen Landesregierung, 2008) a similar situation exists – the one sampling point in class 2 is situated in the ground water body Bohemian massif together with 7 others above decision limit. 5 samples with gross alpha-beta activity concentrations above decision limit are located in the ground water body *Flysch*. This is a rather high percentage, because only a total of 12 samples were taken in this ground water body. For comparison – 78 samples were taken in the Bohemian massif, but only 8 of them are above decision limit.

In the **detailed sampling gross alpha activity concentration** and **gross beta activity concentration** were analyzed separately. In Figure 15.15 the classified **gross alpha** activity concentrations are illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b). 23 of 102 samples have gross alpha activity concentrations above decision limit. One sampling point has a gross alpha activity concentration in class 4, located in the geological area (2) – granite and gneiss of the Bohemian massif. Measurement results in class 3 were also only detected in the Bohemian Massif, in the *Weinsberger Granit* (1) as well as in the gneiss and granite of the Bohemian Massif (2). Only one sample outside of the Bohemian massif has a gross alpha activity concentration in class 2 – in the dolomites of the northern limestone Alps (8).

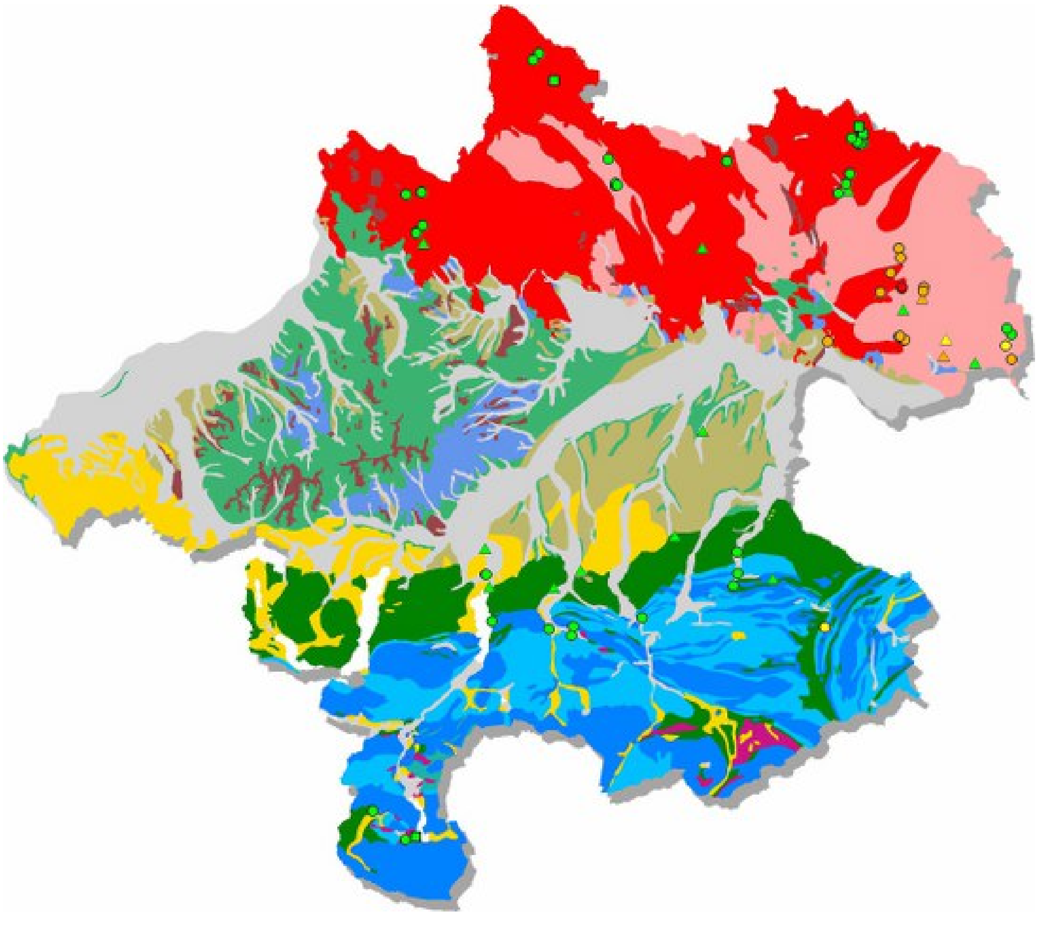

Figure 15.15: *Classified gross alpha activity concentrations of the detailed sampling illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b), legend see Annex A1*

Figure 15.16 demonstrates a higher median gross alpha activity concentration in the granites and gneiss of the Bohemian massif (2) than in the *Weinsberger Granite* (1), similar to the radon results in the detailed sampling. But there is also a higher variation between the samples in area (2) than in the *Weinsberger Granit* (1). It is noticeable that gross alpha activity concentrations above class 1 were only detected within the Bohemian massif in the lower Mühlviertel. This may result from a granite type with high gross alpha activity concentration in this area and as mentioned above it would be very interesting and fundamental to study different granite types (uranium and thorium concentration) and the different radionuclide activity concentrations in the groundwater originating in this bedrock.

Only one sample with gross alpha activity concentration above decision limit is located in the **ground water body** (14) northern limestone Alps, all others are located in the ground water body (1) Bohemian massif according to Amt der Oberösterreichischen Landesregierung (2008).

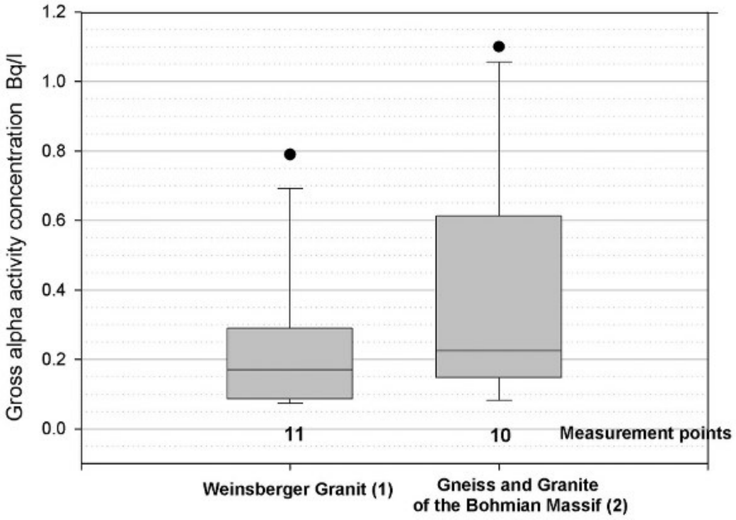

Figure 15.16: *Gross alpha activity concentration of the detailed sampling in the different granite types of the Bohemian massif*

25 of 102 samples of the **detailed sampling** are above decision limit according to their **gross beta activity concentration**. Samples with gross beta activity concentrations in class 3 are located in the *Weinsberger Granit* (1) and in the granites and gneiss of the Bohemian massif (2), one also in the *Flysch* (7). In difference to the gross alpha activities gross beta activity concentrations above class 1 are detected in the entire Bohemian massif, not only in the lower Mühlviertel (Figure 15.17). So it seems that higher gross alpha and gross beta activity concentrations are not obligatorily expectable in the same bedrock. Nevertheless Figure 15.18 shows a similar situation as Figure 15.16 with a higher median gross beta activity concentration in the granites and gneiss of the Bohemian massif (2) than in the *Weinsberger Granit* (1).

One sample with gross beta activity concentration above decision limit is located in the ground water body (10) *Vöckla-Ager-Traun-Alm* and one in (13) *Flysch*, all others are located in the ground water body (1) Bohemian massif according to Amt der Oberösterreichischen Landesregierung (2008).

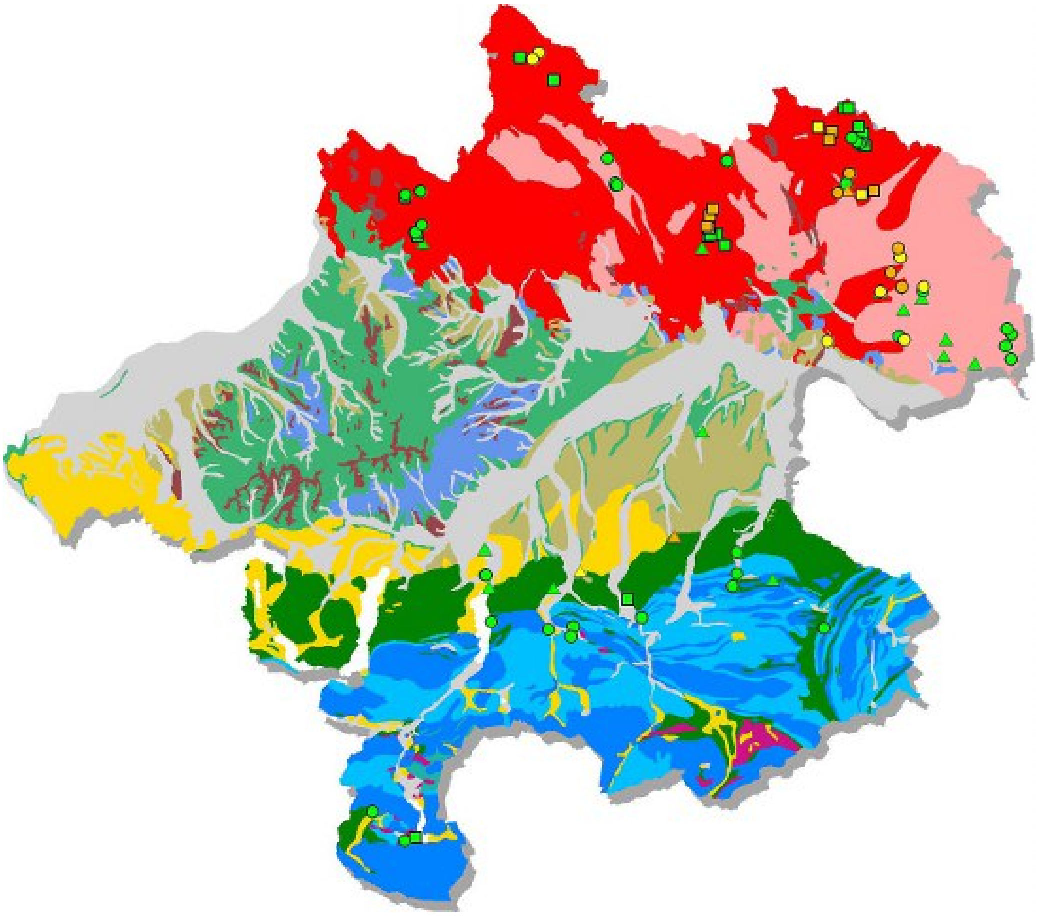

Figure 15.17: *Classified gross beta activity concentrations of the detailed sampling illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b), legend see Annex A1*

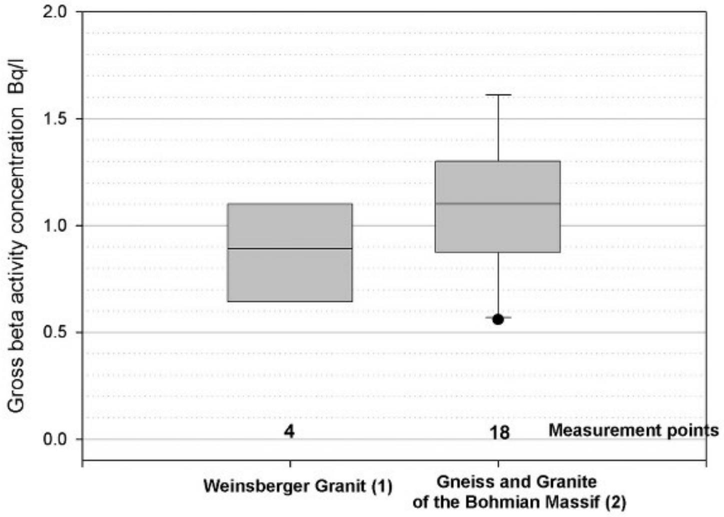

Figure 15.18: *Gross beta activity concentration of the detailed sampling in the different granite types of the Bohemian massif*

15.7 Radium-226

In the **survey sampling** 203 samples were analyzed for ^{226}Ra, only two of them were above decision limit, one in the geological area granites and gneiss of the Bohemian massif (2), one in the geological area *Deckenschotter* (5), which is the same point with the gross alpha-beta activity concentration in class 2. Both measurement points are located in the lower and central Mühlviertel and thus in the ground water body Bohemian massif.

7 of 102 samples of the **detailed sampling** have ^{226}Ra activity concentrations above decision limit, 5 of them in class 2. Figure 15.19 shows that the 5 sampling points with ^{226}Ra activity concentrations in class 2 are distributed among 5 geological areas according to Vohryzka (1973b) – *Weinsberger Granit* (1), granites and gneiss of the Bohemian massif (2), *junge Talfüllungen* (4), *Flysch* (7) and the northern limestone alps (9). So for ^{226}Ra no conclusion can be made regarding geology impacts to ground water. More measurements have to be carried out for further propositions.

For classification concerning **ground water bodies** (Amt der Oberösterreichischen Landesregierung, 2008) a similar situation exists – the 7 samples with ^{226}Ra activity concentrations above decision limit are distributed in 5 ground water bodies – two in Bohemian massif (1), one in *Vöckla-Ager-Traun-Enns* (10), two in *unteres Ennstal* (12), one in *Flysch* (13) and one in the northern limestone Alps (14).

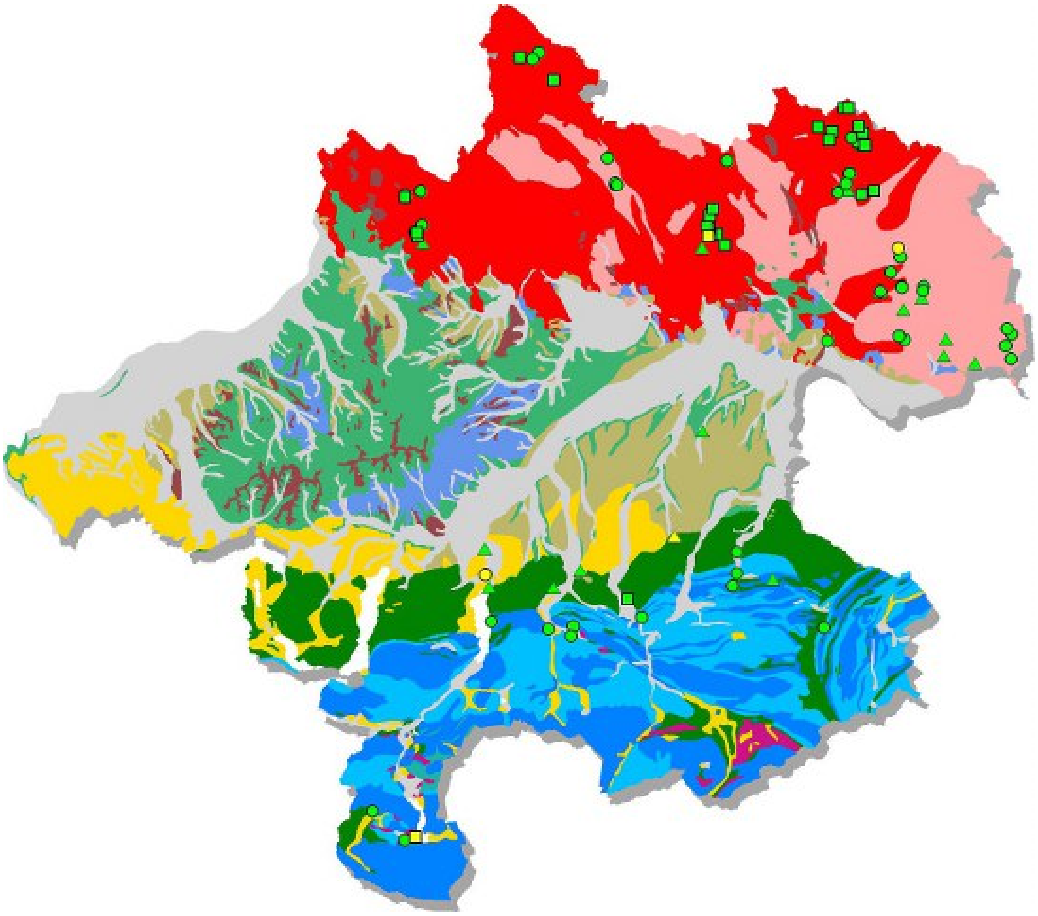

Figure 15.19: *Classified* ^{226}*Ra activity concentrations of the detailed sampling illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b), legend see Annex A1*

15.8 Radium-228

66 samples were analyzed for ^{228}Ra in the **detailed sampling** regarding the geological survey. 17 of these samples have a ^{228}Ra activity concentration above decision limit, but all are in class 1. Most of the samples (13) with ^{228}Ra activity concentration above decision limit are located in the geological area of the granites and gneiss of the Bohemian massif (2) according to (Vohryzka, 1973b). Only one of these samples is located in the *Weinsberger Granit* (1) and three samples are located in the Northern limestone Alps (geological area 8+9). It seems that the ^{228}Ra activity concentration in water is higher in the granite and gneiss of the Bohemian massif than in the *Weinsberger Granit*, but the measured activity concentrations are very low and no conclusions are approvable. 14 samples with ^{228}Ra activity concentrations above decision limits are detected in the ground water body Bohemian massif, two in the northern limestone Alps (14) and one in *Flysch* (13).

15.9 Lead-210

83 samples were analyzed for ^{210}Pb in the **detailed sampling** regarding the geological survey. 46 of these samples have a ^{210}Pb activity concentration above decision limit, one of them is in class 3. This measurement point is located in the geological area *Weinsberger Granite* (1) and has a clearly higher activity concentration than all others (455 ± 54 mBq/l). The point with the second highest ^{210}Pb activity concentration (102 ± 18 Bq/l) was detected in the area of granites and gneiss of the Bohemian massif, where altogether 33 of the 46 measurement points with ^{210}Pb activity concentration above decision limit are located (Figure 15.21). Figure 15.20 demonstrates the ^{210}Pb activity concentrations in **5 geological areas according to Vohryzka (1973b)**. It shows that the median ^{210}Pb activity concentration is slightly higher in the *Weinsberger Granit*, but only 3 measurement points exists in this area including the one with the very high activity concentration mentioned above. But also in the geological area of granite and gneiss of the Bohemian massif the ^{210}Pb results varies rather strong. So it seems that the ^{210}Pb activity concentration in water is highly dependent on the granite type but probably also on other effects. On the other hand the ^{210}Pb activity concentrations in the different bedrocks from the northern limestone Alps (dolomite, limestone) correspond rather well. So it would be very interesting to survey bedrocks and their impact on ground water not only for uranium and thorium but also for the progenies like ^{210}Pb.

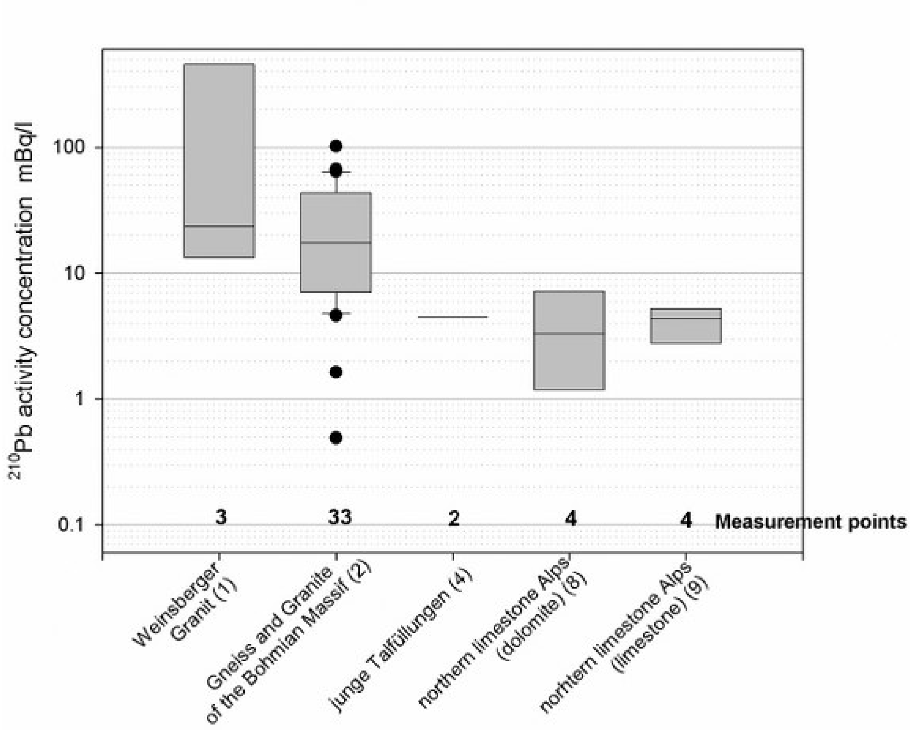

Figure 15.20: ^{210}*Pb activity concentrations of the detailed sampling in the different geological areas of Upper Austria*

36 of the samples with ^{210}Pb activity concentration above decision limit are located in the **ground water body** Bohemian massif (1), 8 in the northern limestone Alps (14), one in the *Traun-Enns-Platte* (11) and in *Flysch* (13) respectively.

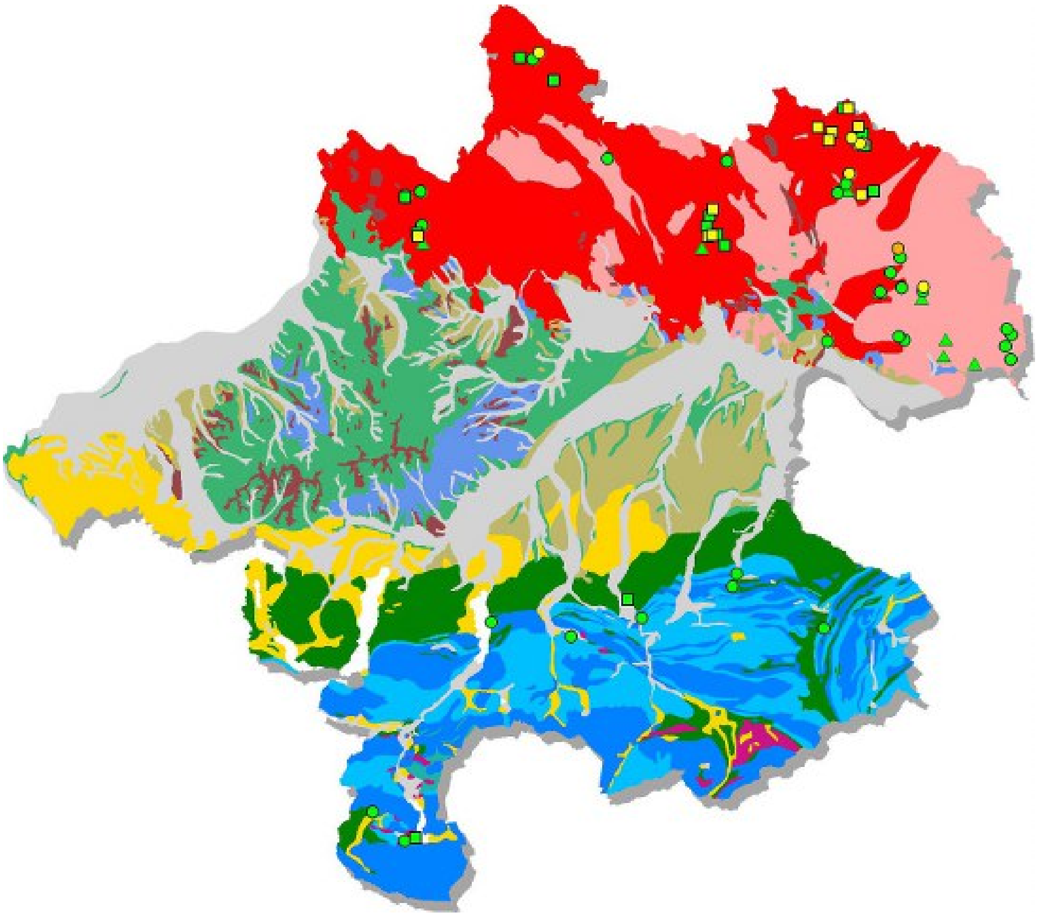

Figure 15.21: *Classified ^{210}Pb activity concentrations of the detailed sampling illustrated in the hydrogeological map of Upper Austria (Vohryzka, 1973b), legend see Annex A1*

15.10 Polonium-210

71 of 81 analyzed samples have a ^{210}Po activity concentration above decision limit. 6 of them are in class 3, all in the Bohemian massif, in the Weinsberger Granite (1) as well as in the gneiss and granites of the Bohemian massif (2). Figure 15.22 demonstrates the ^{210}Po activity concentrations in **5 geological areas** according to Vohryzka (1973b). It shows that the median ^{210}Po activity concentration in the *Weinsberger Granit* (1) and the gneiss and granites of the Bohemian massif (2) are nearly equal, in the other areas clearly below them. The geological areas grouped in more **geographic classification** shows again a slightly higher median ^{210}Po activity concentration in the lower Mühlviertel than in the central and upper Mühlviertel, which was also found for ^{222}Rn (see above). In the other areas the ^{210}Po

activity concentration is clearly lower (Figure 15.23). Further studies to survey the different granite types and bedrocks especially in the Bohemian massif will be reasonable in the future to get a data base for interpretations and generalization of these results.

60 of the 71 samples with ^{210}Po activity concentration above decision limit are located in the **ground water body** Bohemian massif (1), 8 in the northern limestone Alps (14), one in *Traun-Enns-Platte* (11), in *unteres Ennstal* (12) and in *Flysch* (13) respectively.

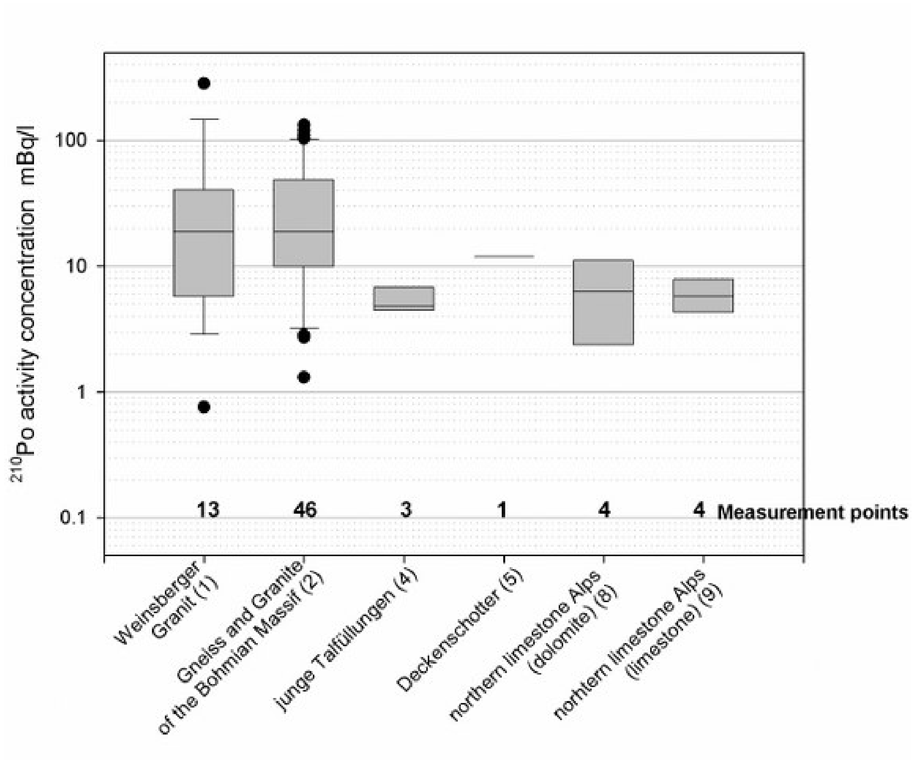

Figure 15.22: ^{210}*Po activity concentrations of the detailed sampling in the different geological areas of Upper Austria (Vohryzka, 1973b)*

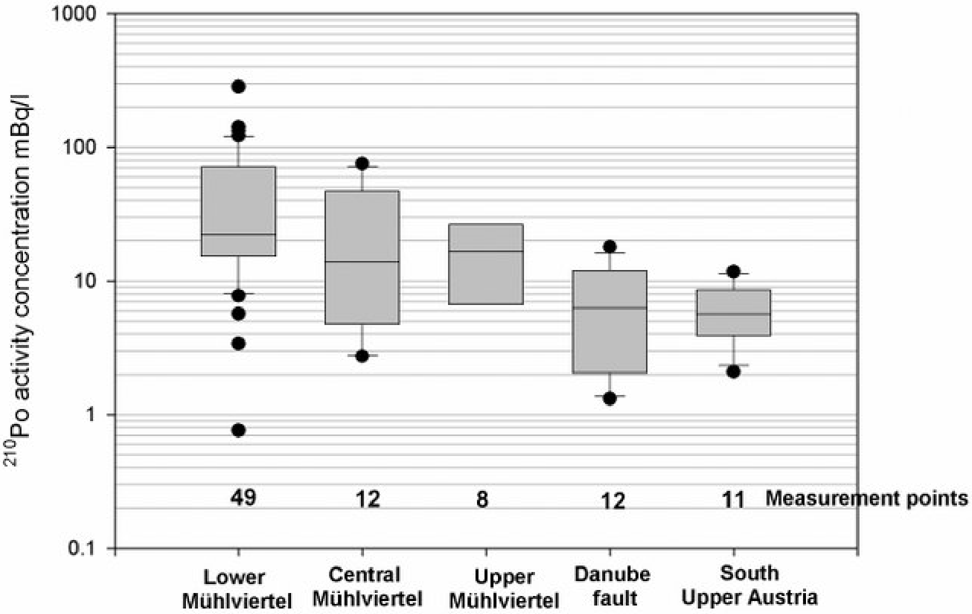

Figure 15.23: *^{210}Po activity concentrations of the detailed sampling in a simplified geographic and geologic classification*

15.11 Drilled Wells and Springs

The water samples from the detailed sampling examined in this chapter were taken directly at wells and springs. So from a geological point of view it is also of interest to distinguish between activity concentrations in waters from **drilled wells** and waters from **springs**. Without taking into account geological areas, the median **radon activity concentration** in drilled wells (about 120 Bq/l) is slightly higher than in springs (about 95 Bq/l) as expected (Figure 15.24). But within the measurements it was noticeable, that often higher activity concentrations were detected in springs than in deep wells. So it seems that in some bedrocks the water of a drilled well originates from compact bedrock and does not absorb the radioactivity concentration of the material. In contrast, in some surfaces near springs in weathered bedrock the water has more contact with the material and absorbs more radioactivity concentration from it and therefore has higher activity concentrations than some waters from deep wells in the same region. An example are the wells and springs in the Danube fault – the median of the drilled wells is about 75 Bq/l, whereas the median radon activity concentration in springs is about 140 Bq/l (Figure 15.25).

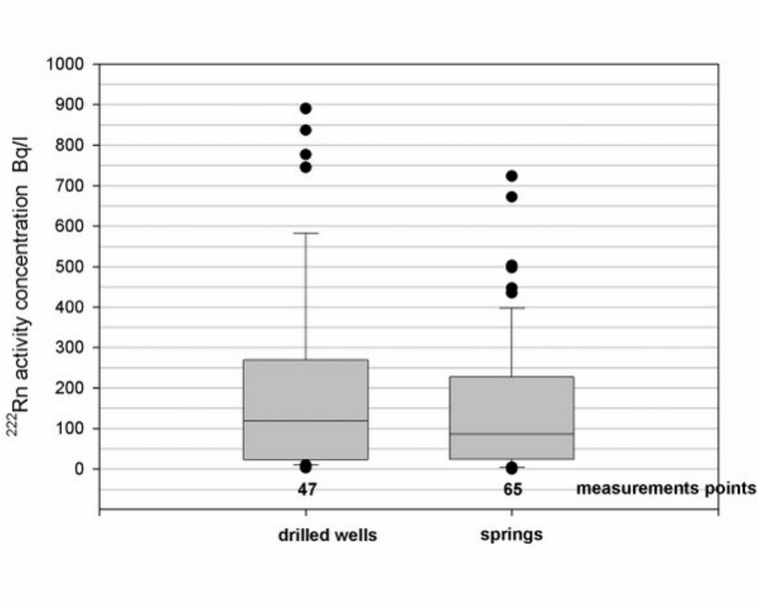

Figure 15.24: *^{222}Rn activity concentrations in wells and springs in Upper Austria*

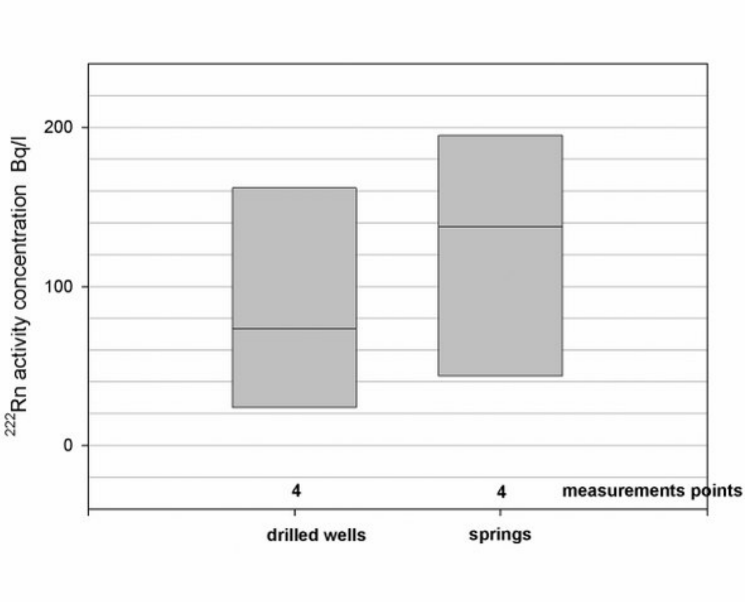

Figure 15.25: *^{222}Rn activity concentrations in wells and springs in the Danube fault*

For ^{226}Ra and ^{228}Ra too few measurement results above decision limit exist to reasonably distinguish between springs and wells. For gross alpha, gross beta and ^{3}H no significant difference between the median activity concentrations in spring and well water is detectable. For ^{238}**U** and ^{210}**Po** the median activity concentration in total Upper Austria is negligible, but taken into account only the sampling points in southern Upper Austria the **median activity concentrations in springs are above the ones in drilled wells**. For ^{210}**Pb** the median activity concentration in well waters are slightly above the one in spring water in total Austria, but again the median activity concentration in spring water is slightly higher in south Upper Austria. This might be because of bedrock with low uranium and thorium concentration in this region, so a deep well does not enhance the activity concentration in the water because of less radioactivity concentration in the solid and compact bedrock of water origin. In contrast in the springs originating in the more weathered surface near bedrock more radioactivity is emitted to the water due to more contact from the water whith the rocks because of finer grained bedrock.

15.12 Geological Survey of Some Noticeable Sampling Points

As discussed above and in some chapters before, it would be very interesting and fundamental to study **different granite types** (uranium and thorium concentration) and the different radionuclide activity concentrations in the **groundwater originating from this bedrock**. As a first step in this chapter the sampling points of some communities are illustrated in sections of a **detailed geological map of Upper Austria** (Geologische Bundesanstalt, 2006). The main trigger for carrying out these illustrations was the detailed sampling at one sampling point in the central Mühlviertel. As discussed in Chapter 8.1 the owner prohibited another sampling at the sampling point with the second highest radon activity concentration within the survey sampling in the detailed sampling phase. So a sample was taken at a private well located next to this one, and the measured radon activity concentration there was very low. A look at the geological map showed, that the geology in this region is very heterogeneous. So this was surveyed more closely.

The used geological map is an online-version of the **geological map of Upper Austria 1:200 000** (Geologische Bundesanstalt, 2006), which can be downloaded from the homepage of the Geological Survey of Austria for free[1]. To buy these data (for a geo-information system program) is very expensive and could not be realized within this thesis, and so these surveys are carried out with the free online version. So the quality of the maps is not very high, but nevertheless it gives a good insight for this topic and that it is also reasonable to do such illustrations in the future with more data. In Figure 15.26 an extract of the legends of the used geological map (Geologische Bundesanstalt, 2006) is illustrated with the **different geologic zones** and **different granite types**.

Figure 15.27 demonstrates the heterogenic geological situation in the community in the **central Mühlviertel (Hellmonsödt)** with the second highest radon activity concentration in the survey sampling. In the detailed sampling phase several samples were taken within these communities. In the figure only samples at private wells or directly at springs or wells were taken into account for the geological survey. In Table 15.1 the radon activity concentrations at the sampling points are listed with their geologic zone or granite type.

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[1] http://www.geologie.ac.at/

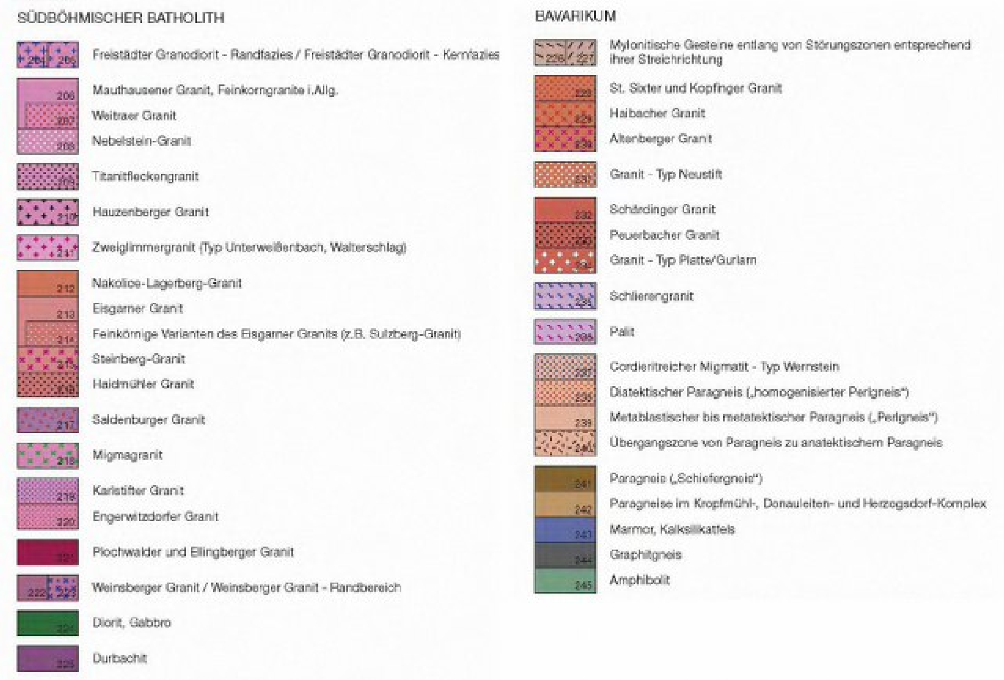

Figure 15.26: *Legend of the geological maps (Geologische Bundesanstalt, 2006)*

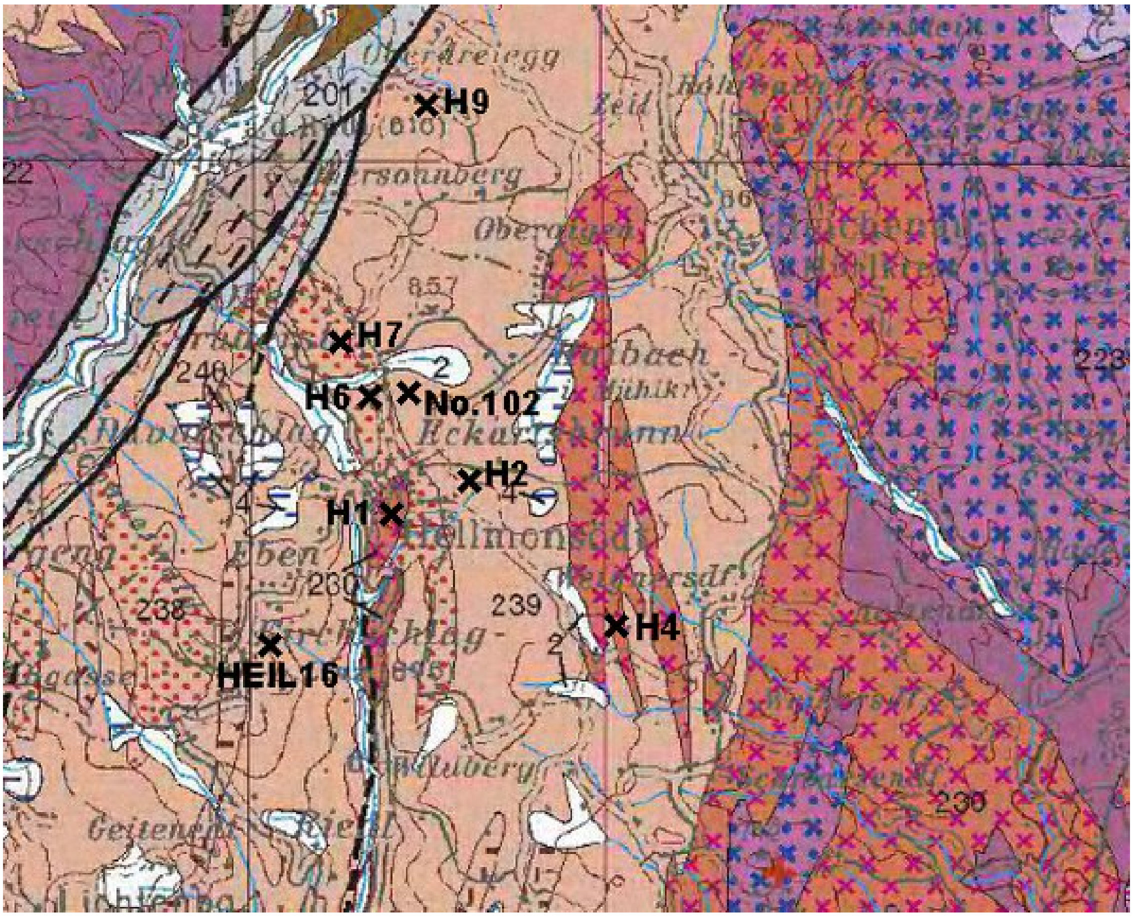

Figure 15.27: *Sampling points in different bedrocks in the community Hellmonsödt in the central Mühlviertel (Geologische Bundesanstalt, 2006)*

Table 15.1 shows, that the radon activity concentrations in the communities vary strongly from 745 Bq/l to 15 Bq/l.

Table 15.1: ^{222}Rn *activity concentration at sampling points in different geologic zones or granite types in the community Hellmonsödt*

Sampling point	**Radon-222 activity concentration (Bq/l)**	**Geological zone/ Granite type**
Survey sampling point (No. 102)	342 ± 11	Metablastischer bis Metatektischer Paragneis ("Perlgneis") (239)
H1	745 ± 64	Altenberger Granit (230)
H2	165 ± 17	Metablastischer bis Metatektischer Paragneis ("Perlgneis") (239)
H4	15 ± 4	Altenberger Granit (230)
H6	38 ± 6	Diatektischer Paragneis (238)
H7	54 ± 8	Diatektischer Paragneis (238)
H9	182 ± 18	Metablastischer bis Metatektischer Paragneis ("Perlgneis") (239)
HEIL16	86 ± 10	Metablastischer bis Metatektischer Paragneis ("Perlgneis") (239)

The sampling point with the second highest Radon activity concentration in the survey sampling (**no. 102**) is located in a zone with bedrock *Perlgneiss* (239, light rose colour in the geological map (Geologische Bundesanstalt, 2006), Figure 15.26). The sampling point next to this point in the detailed sampling phase was the point **H6**, which is already located in another gneiss-type called *Diatektischer Paragneis* (238, light rose colour with red dots, Figure 15.26). The radon activity concentration is much lower at this point. The sampling points **H2, H9 and HEIL16** are also located in the *Perlgneis* as the sampling point from the survey sampling. The radon activity concentrations vary from 342 Bq/l to 86 Bq/l, but are all in a medium radon activity concentration range. The radon activity concentrations at the two sampling points **H6 and H7** in the *Diatektischer Paragneis* are clearly lower. The highest radon activity concentration in this community were detected at sampling point **H1**, which is located in a granite bedrock called *Altenberger Granit* (230, dark rose colour with pink crosses, Fig. 15.26). This would correspond to Schubert et al. (2003) who reported rather high Uranium contents in this granite type. But at the second sampling point in the *Altenberger Granit* (**H4**) only a radon activity concentration of 15 Bq/l was detected. But the area where this point is located is very close to and splitted with the *Perlgneiss*, so the bedrock from which the ground water comes from may be not only the *Altenberger Granit*.

The results demonstrate the great **impact of different geologic bedrocks** and **different granite** and **gneiss types** to the radon activity concentration in the ground and drinking water. So further studies and surveys are very reasonable to characterize and generalize this impact.

Figure 15.28 demonstrates the sampling points in the community in the **lower Mühlviertel** with the highest detected radon activity concentrations in the detailed sampling phase (**Bad Zell**). In this community high radon concentrations were detected in two deep wells, but also in some springs and a ”sacred well”. In the community also a **radon spa** exists. The geology in this area is not as heterogeneous as in the region of Hellmonsödt in the central Mühlviertel. Mainly two different granite types are predominant. The *Weinsberger Granit* (222, dark rose, Fig. 15.26), which was also discussed as a separate zone above, and the *Mauthausner Granit* (206, light rose, Fig. 15.26). The deep wells **WVA70** and **WVA71** with the highest ^{222}Rn activity concentrations are both located in the *Weinsberger Granit*, also the sampling point **WVA68**, which is a collection point of some different springs (Table 15.2). The ”sacred well” (**HEIL5**), which is a surface near spring, with the highest radon activity concentration is located in the *Mauthausener Granit*. Schubert et al. (2003) also found the highest radon activity concentration in a drilled well in the *Weinsberger Granit*, also in the same community. The *Weinsberger Granit* has a rather low uranium and thorium content nevertheless it shows very high radon activity concentrations in drinking water originating in it. In the discussions above the radon activity concentrations detected in the survey sampling were higher in the *Weinsberger Granit* than in the granite and gneiss of the Bohemian massif. But in the detailed sampling the median of the radon activity concentrations of the granite and gneiss of the Bohemian massif were higher, but may results because of sampling (see above). But the radon activity concentration at the sacred well HEIL5 is nevertheless rather high and one of the highest detected radon activity concentrations in Upper Austria, and in the same range of the activity concentration detected at H1 in the *Altenberger Granit* in Hellmonsödt. So it would be very interesting to do further surveys to characterize these granite types and distinguish them by their uranium, thorium or radium contents. But it seems that *Weinsberger Granit*, *Altenberger Granit* and *Mauthausener Granit* have potential for high radon activity concentrations in drinking water originating in them.

Table 15.2: ^{222}*Rn activity concentration at sampling points in different geologic zones and granite types in the community Bad Zell*

Sampling point	Radon-222 activity concentration (Bq/l)	Geological zone/ Granite type
WVA68	353 ± 32	Weinsberger Granit (222)
WVA70	890 ± 75	Weinsberger Granit (222)
WVA71	837 ± 71	Weinsberger Granit (222)
HEIL5	723 ± 62	Mauthausener Granit (206)

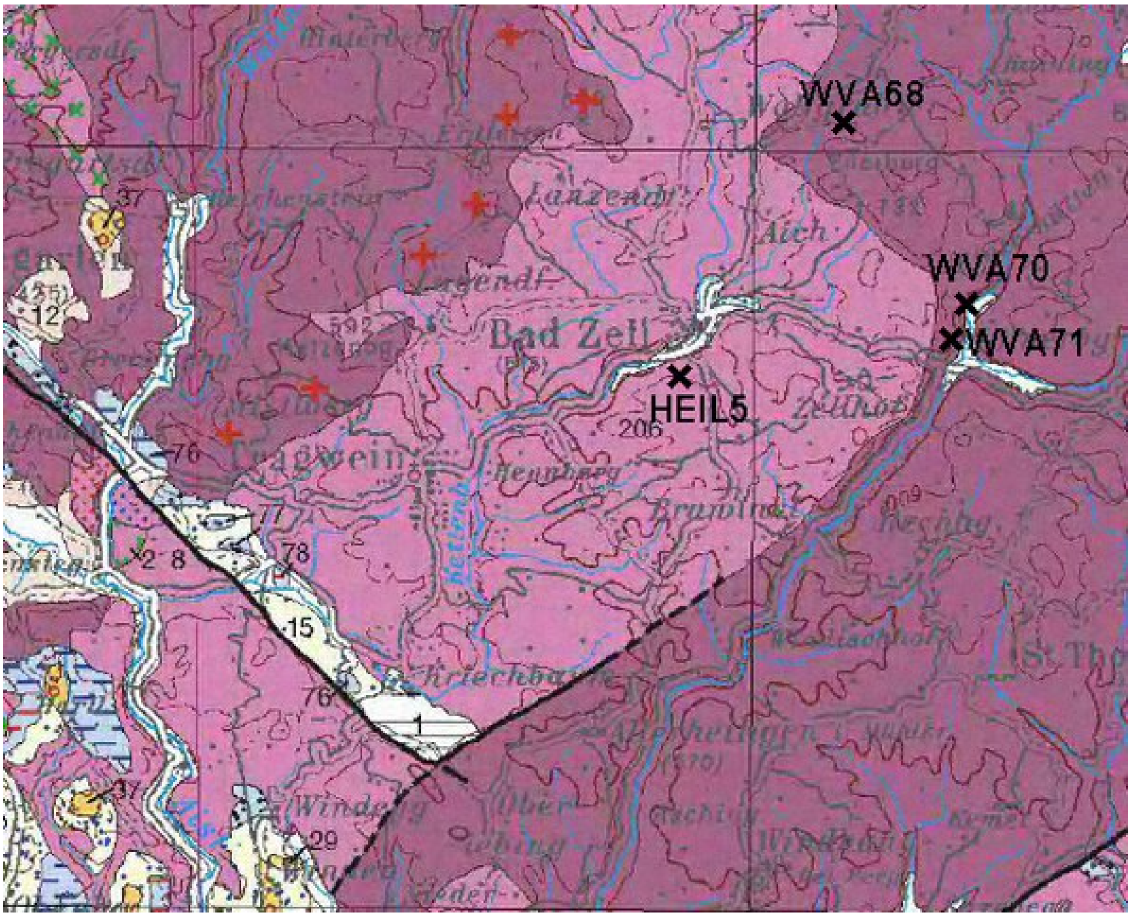

Figure 15.28: *Sampling points in different bedrocks in the community Bad Zell in the lower Mühlviertel (Geologische Bundesanstalt, 2006)*

As another example of varying radon activity concentration results and a rather heterogeneous geological situation the community **St. Oswald** in the **lower Mühlviertel** is surveyed. Figure 15.29 illustrates the different geological zones and granite types in this area and the sampling points, which are characterized in Table 15.3. The highest radon activity concentration in the community were detected at collection point of some different springs (**WVA11**), which is located again in the *Mauthausener Granit* (206, light rose, Fig. 15.26) as the sacred well in Bad Zell. The activity concentration is a little lower than the one at the sacred well in Bad Zell (HEIL5), but nevertheless rather high and in the same range. The sampling point **H21** is again located in *Weinsberger Granit* (222, dark rose, Fig. 15.26) and show a clearly lower radon activity concentration than the deep wells in Bad Zell. But the sampling point is a private well and no drilled deep well. The sampling points **HEIL14** – a "sacred well" – and **H20** – a private well – are located in a geologic zone called "Transitions region from *Paragneis* to *anatektischer Paragneis*" (240, light rose with red lines, Fig. 15.26) and show a medium radon activity. The sampling point H20 has a radon activity concentration clearly below the "sacred well" HEIL14, but this may also be, because the water from the private well may be influenced by the nearby zone of "aggradational deposit" (2, white). In this bedrock also the deep well **WVA13** is located, which shows a low radon activity concentration of 87 Bq/l. For a deep well in the Bohemian massif this is a very low radon activity concentration.

The results in this community approve the statement from above that both the *Mauthausener Granit* and *Weinsberger Granit* have **potential for high radon activity concentrations in drinking water** originating in them. Also other different granite and gneiss zones could yield medium and higher radon activity concentrations and should be surveyed in the future. The result of the deep well (WVA13) in the "aggradational deposit" shows the clear difference of ground and drinking water originating from granite or gneiss or from other bedrocks, also within the same geologic area like the Bohemian massif.

Table 15.3: *^{222}Rn activity concentration at sampling points in different geologic zones and granite types in the community St. Oswald*

Sampling point	Radon-222 activity concentration (Bq/l)	Geological zone/ Granite type
WVA11	436 ± 39	Mauthausener Granit (206)
WVA13	87 ± 10	Flussablagerung/ Aggradational deposit (2)
HEIL14	281 ± 27	Transitions region from Paragneis to anatektischer Paragneis (240)
H20	147 ± 16	Transitions region from Paragneis to anatektischer Paragneis (240)
H21	231 ± 22	Weinsberger Granit (222)

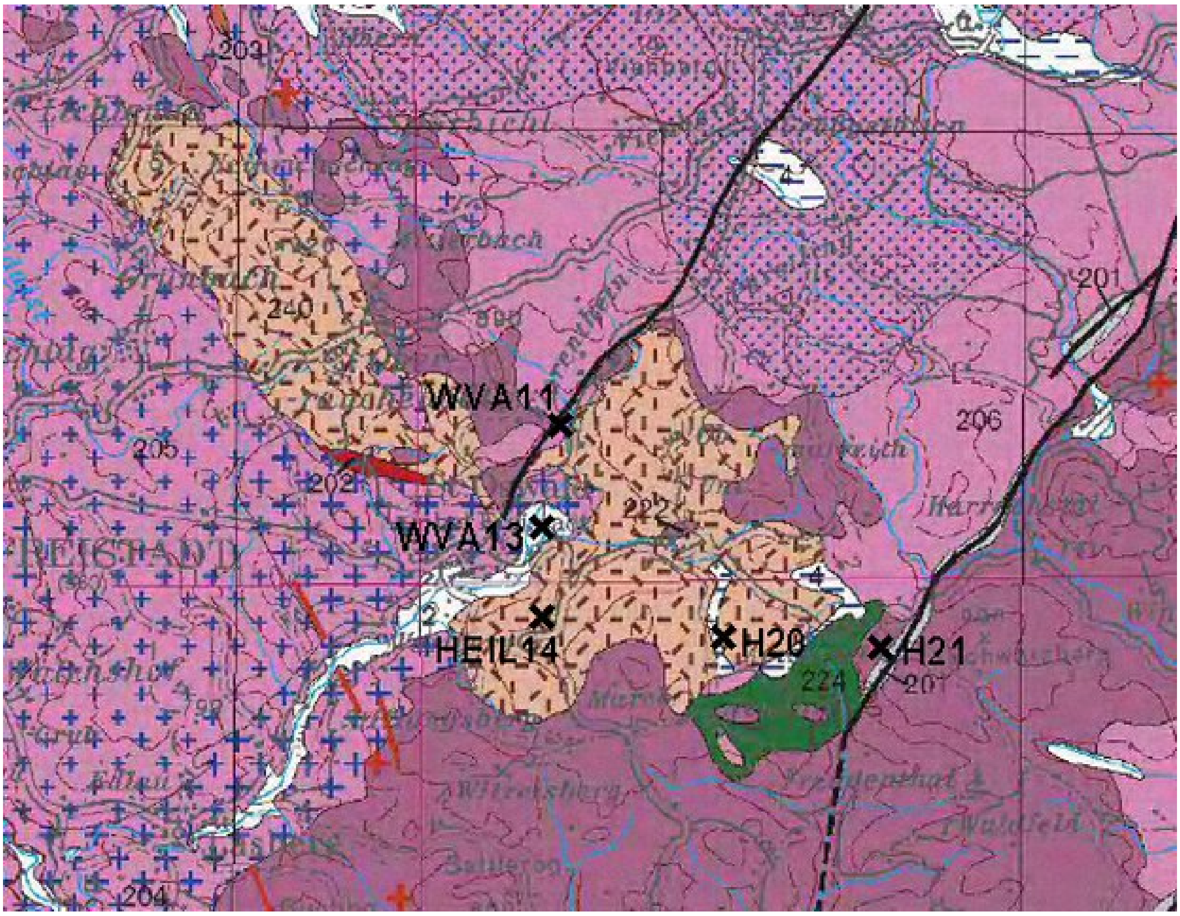

Figure 15.29: *Sampling points in different bedrocks in the community St. Oswald in the lower Mühlviertel (Geologische Bundesanstalt, 2006)*

In the neighbouring communities **Sandl and Windhaag bei Freistadt** in the **lower Mühlviertel** 13 samples were taken at private wells and directly at some deep wells and springs. The radon activity concentrations in these samples vary from 43 to 531 Bq/l (see Table 15.4) and are located in three different granite types, which is illustrated in Figure 15.30. The private wells **H10**, **H11**, **H16**, **H17** and **H19** are again located in the bedrock *Weinsberger Granit* (222, dark rose, Fig. 15.26). The radon activity concentrations vary from 43 to 318 Bq/l in the *Weinsberger Granit*, so the activity concentrations detected here are clearly lower than the ones from the deep wells in Bad Zell. The radon activity concentration at the private well H17 is in a medium range, but the others are clearly lower, especially the private wells H10 and H11. The detected activity concentrations at these sampling points do not concur with the theory that *Weinsberger Granit* causes higher radon activity concentration in drinking water. But it would verify the rather low uranium and thorium content in the Weinsberger Granite reported by Schubert et al. (2003). Again the results demonstrate the need of further studies in this thematic field.

The private wells **H13**, **H14**, **H15** and the springs **WVA5**, **WVA7**, **WVA8** and the sample point of collected springs **WVA4** are located in the *Karlstifter Granit* (219, light rose with blue dots, Fig. 15.26). The detected radon activity concentrations are all in the range of about 200 to 300 Bq/l, only the spring WVA5 has a higher activity concentration of 503 Bq/l. No low radon activity concentrations were detected in waters originating within this granite type. So it seems that the *Karlstifter Granit* has potential to influence ground and drinking water originating in it for a medium to high radon activity concentration.

The highest radon activity concentration in these communities was detected in the *Freistädter Granodiorit* (205, light rose with blue crosses, Fig. 15.26) with 531 Bq/l (**H12**). That rather high radon activity concentration in the private well water is in disagreement with the low Uranium contents (1–3 ppm) in this granite type reported by Schubert et al. (2003). Schubert also reported a better correlation between Uranium concentration in bedrock and radon concentration in water in springs than in drilled wells (see Chapter 4.3).

The results again demonstrate the **diversity of this thematic field of uranium and thorium content** of **different granite types** and the **natural radionuclide activity concentration in the waters** originating from them. The last example also demonstrated that a low uranium content in the granite does not allow to predict a low radon activity concentration. Further studies should be done in this field for a better characterization of the correlation of activity concentrations in waters and geology and bedrock which may yield generalizations and predictions.

Table 15.4: *^{222}Rn activity concentration at sampling points in different geologic zones and granite types in the communities Windhaag bei Freistadt and Sandl*

Sampling point	Radon-222 activity concentration (Bq/l)	Geological zone/ Granite type
H10	43 ± 6	Weinsberger Granit (222)
H11	73 ± 9	Weinsberger Granit (222)
H12	531 ± 47	Freistädter Granodiorit (205)
H13	278 ± 27	Karlstifter Granit (219)
H14	244 ± 24	Karlstifter Granit (219)
H15	201 ± 20	Karlstifter Granit (219)
H16	130 ± 14	Weinsberger Granit (222)
H17	318 ± 30	Weinsberger Granit (222)
H19	103 ± 12	Weinsberger Granit (222)
WVA4	294 ± 28	Karlstifter Granit (219)
WVA5	503 ± 44	Karlstifter Granit (219)
WVA7	322 ± 30	Karlstifter Granit (219)
WVA8	174 ± 18	Karlstifter Granit (219)

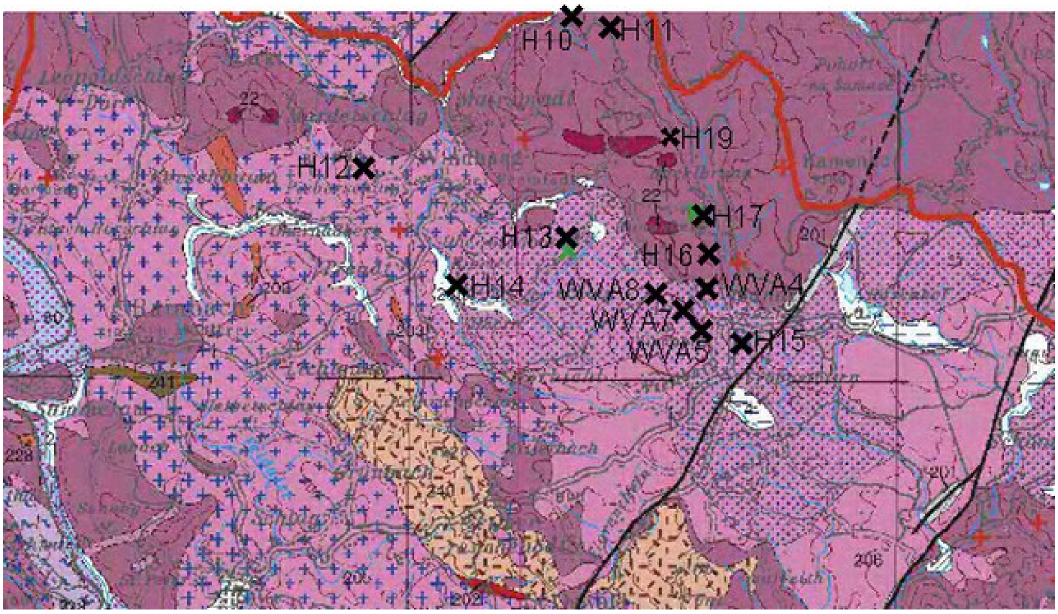

Figure 15.30: *Sampling points in different bedrocks in the community Windhaag bei Freistadt and Sandl in the lower Mühlviertel (Geologische Bundesanstalt, 2006)*

Chapter 16

Dose Calculations

So far, the results of this thesis were discussed regarding the measured activity concentrations. The relevant parameter for radiation protection of the public is the **dose induced by radionuclides in drinking water**. One of the indicator parameters in the Austrian drinking water directive (Republik Österreich, 2001) is the total indicative dose (TID), which is discussed in Chapter 3.1, and the classification of the activity concentrations were calculated according to this existing dose standard of 0.1 mSv/a (see Chapter 6).

16.1 Total Indicative Dose According to ÖNORM S 5251

For Austria the determination of the parameter **total indicative dose** (TID) appointed in the Austrian drinking water directive (Republik Österreich, 2001) is defined in the Austrian standard ÖNORM S 5251 – Determination and evaluation of the total dose due radionuclides in drinking water (Austrian Standards Institute, 2005b). The total indicative dose is defined as the effective dose caused by ingestion of the annual consumption of drinking water, without taking into account the contributions of tritium, potassium-40, radon and radon progenies. As parametric value according to the drinking water directive (Republik Österreich, 2001) a total dose of 0.1 mSv in a reference period of one year is established (= total indicative dose of 0.1 mSv/a). According to the Austrian standard basically only the natural radionuclides ^{226}**Ra and** ^{228}**Ra are taken into account** for dose estimation. Other natural radionuclides (e.g. ^{232}Th, ^{234}Th) are generally neglected because of their geochemical behaviour (e.g. low solubility in water) or their low contribution to the effective dose (in Austria for example ^{238}U and ^{234}U). Artificial and other natural radionuclides (except ^{3}H, ^{40}K, ^{222}Rn, radon progenies) are taken into account for dose estimation only if a **dose relevant concentration is reasonably expected**.

The activity concentrations of the radionuclides taken into account for the surveyed drinking water are determined in one water sample. If the determination is radiometric, the Austrian standards ÖNORM S 5250-1 and ÖNORM S 5250-2 (Austrian Standards Institute, 2002, 2005a) must be obeyed – with defined limits of detection and total measurement uncertainties (Austrian Standards Institute, 2005b).

The **total dose** (GD) is the sum of the dose contributions of the single radionuclides (GD_i) (according to ÖNORM S 5251 basically only ^{226}Ra and ^{228}Ra), which are calculated from the activity concentrations (c_i) with the legal valid dose conversion factors ($h(g)_i$) for adults (age $>$17 a) respectively (Table 16.1) and an annual consumption (KM) of 730 l/a (according to ÖNORM S 5251) (Formula 16.1). Activity concentrations below decision limit are set to zero. Uncertainties are calculated by error propagation without taking into account an uncertainty contribution of consumption and dose conversion factors.

$$GD = \sum_i GD = \sum_i h(g)_i \cdot c_i \cdot KM \qquad (16.1)$$

For the **compliance of a reference value for the total dose** (RGD – e.g. the total indicative dose according to the drinking water directive (Republik Österreich, 2001)) the following requirement has to be proved, whereas ΔGD is the uncertainty of the total dose:

$$RGD \geq GD - \Delta GD \qquad (16.2)$$

If the requirement according to Formula 16.2 is achieved, the reference value for the total dose is complied, otherwise the reference value is exceeded.

In Table 16.1 the **dose conversion factors for different age groups** and nuclides are listed according to the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (IAEA, 1996). The dose conversion factors are the values of the committed effective dose per unit intake via ingestion for members of the public at different age groups according to Table A of Annex III of the Basic Safety Standards (IAEA, 1996). The annual intake values were chosen by the Article 31 working party on radioactivity in drinking water after overview of different intake values by different organisations, including the WHO. There is no defined intake for class 12–17a (Risica & Grande, 2000).

Table 16.1: *Dose conversion factors for different age groups and nuclides according to the Basic Safety Standards (IAEA, 1996) and annual water intakes according to European Commission (1998), Risica & Grande (2000)*

Age group	Dose conversion factors (Sv/Bq)					Annual water intake (l)
	^{228}Ra	^{226}Ra	^{238}U	^{210}Pb	^{210}Po	
<1	3.0 E-5	4.7 E-6	3.4 E-7	8.4 E-6	2.6 E-5	250
1–2	5.7 E-6	9.6 E-7	1.2 E-7	3.6 E-6	8.8 E-6	350
2–7	3.4 E-6	6.2 E-7	8.0 E-8	2.2 E-6	4.4 E-6	350
7–12	3.9 E-6	8.0 E-7	6.8 E-8	1.9 E-6	2.6 E-6	350
(12–17	5.3 E-6	1.5 E-6	6.7 E-8	1.9 E-6	1.6 E-6)	
>17	6.9 E-7	2.8 E-7	4.5 E-8	6.9 E-7	1.2 E-6	730

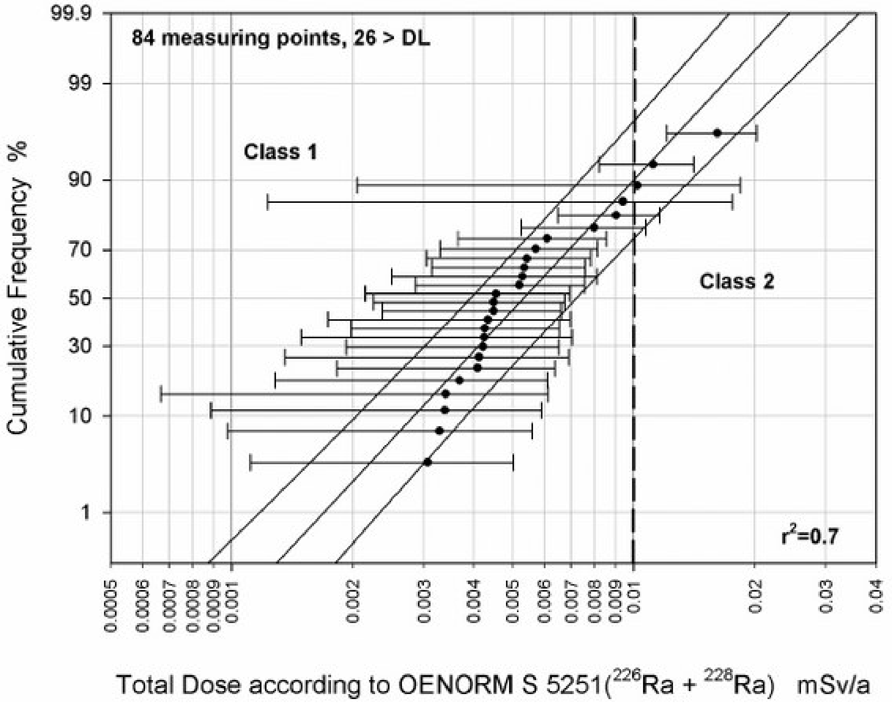

Figure 16.1: *Cumulative frequency of the total dose according to ÖNORM S 5251 (Austrian Standards Institute, 2005b)*

In Figure 16.1 the **total dose according to the Austrian standard ÖNORM S 5251** (Austrian Standards Institute, 2005b) is determined for the measurement results of the detailed sampling of this thesis and illustrated in a cumulative frequency graph. At 84 measurement points both ^{226}Ra and ^{228}Ra were analyzed, in 26 of these at least the activity concentration of one of the nuclides was above decision limit. No calculated total dose is above the parametric value of 0.1 mSv/a (class 3 or 4), only 3 samples have a total dose in class 2 (0.01 mSv/a). According to the cumulative frequency demonstration 90% of all drinking waters have a total dose below 0.1 mSv/a and 99.9% will have a total dose below 0.025 mSv/a with an r_2 value of the cumulative frequency distribution of 0.7. It seems that the results of the surveyed drinking water samples are far away from exceeding the parametric value of the total indicative dose of the Austrian drinking water directive.

16.2 Total Effective Dose Considering Other Nuclides

The calculated dose according to ÖNORM S 5251 (Austrian Standards Institute, 2005b) is rather low, as discussed above. So doses of different other nuclides were calculated to survey their contributions. In Figure 16.2 the **total dose** according to ÖNORM S 5251 (Austrian Standards Institute, 2005b) is **expanded with** the dose contribution of **^{238}U**. Much more samples have a ^{238}U activity concentration above decision limit than for ^{228}Ra and ^{226}Ra, so a **cumulative frequency** of 66 measurement points is illustrated in the graph. No uncertainties are illustrated because of the low measurement values. The cumulative frequency shows a similar situation as in Figure 16.1, with no detected total doses in class 3 and only a few in class 2. Table 16.1 shows, that the dose conversion factor for ^{238}U is one magnitude below the ones of ^{226}Ra and ^{228}Ra, so the dose contribution is not very high, as discussed in the ÖNORM S 5251 (Austrian Standards Institute, 2005b).

Nevertheless, some single points yield a clear dose contribution by ^{238}U, like the measurement point with the highest total dose (0.033 mSv/a) in Figure 16.2. This point in the lower Mühlviertel (WVA70) has a ^{226}Ra and ^{228}Ra activity concentration below decision limit, but the highest measured ^{238}U activity concentration, and therefore yields the highest dose contribution of all analyzed samples. So it seems that disregarding ^{238}U in the total dose calculation may sometimes not be justified, but on the other hand, the dose contribution of this sample is still clearly below the parametric value of the total indicative dose of 0.1 mSv/a. But the cumulative frequency distribution of the total dose calculations with ^{238}U predicts a total dose above 0.1m Sv/a for some percents of the drinking waters in Upper Austria (r^2 of the distribution: 0.58) in contrast to the cumulative frequency distribution of the total dose without ^{238}U (Figure 16.1).

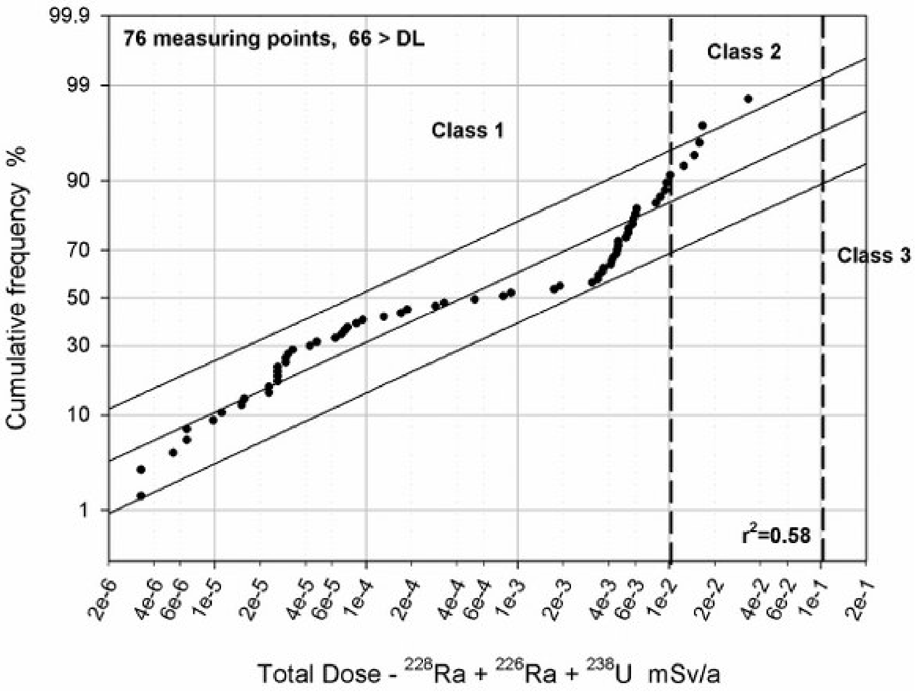

Figure 16.2: *Cumulative frequency of the total dose according to ÖNORM S 5251 (Austrian Standards Institute, 2005b) taking into account the* ^{238}*U dose contribution*

Although **radon progenies** are not taken into account for calculations of total dose in the Austrian drinking water directive and the ÖNORM S 5251, a **total effective dose** determination is done for ^{210}**Po and** ^{210}**Pb** out of interest, because of their **higher dose conversion factors** (as listed in Table 16.1). At 104 measurement points both ^{210}Po and ^{210}Pb were analyzed, at 93 at least one of them has an activity concentration above decision limit (Figure 16.3). The cumulative frequency distribution shows, that the dose contribution of these two nuclides is much higher than the one of the radium isotopes and ^{238}U. According to this distribution (r^2=0.42) for about 10% of the drinking waters a total effective dose of above 0.1 mSv/a is predicted, for 0.1% more than 0.8 mSv/a. Ten measurement points have a ^{210}Po and ^{210}Pb total effective dose in class 3 – above the parametric value of the total indicative dose of 0.1 mSv/a in the Austrian drinking water directive. So these nuclides should not be unconsidered in the drinking water directive and the Austrian standards because of their high dose contribution. Not to disregard the ^{210}Po and ^{210}Pb dose contribution is also discussed by the Strahlenschutzkommission (SSK) (2003), although mainly for babies. It says that for babies effective doses caused by ^{210}Pb and ^{210}Po of 0.28 mSv/a and 0.44 mSv/a respectively are expected and the parametric values for these nuclides in the Recommendation Euratom 2001/928 (European Commission, 2001) are not conform with a parametric value of total dose of 0.1 mSv/a for babies.

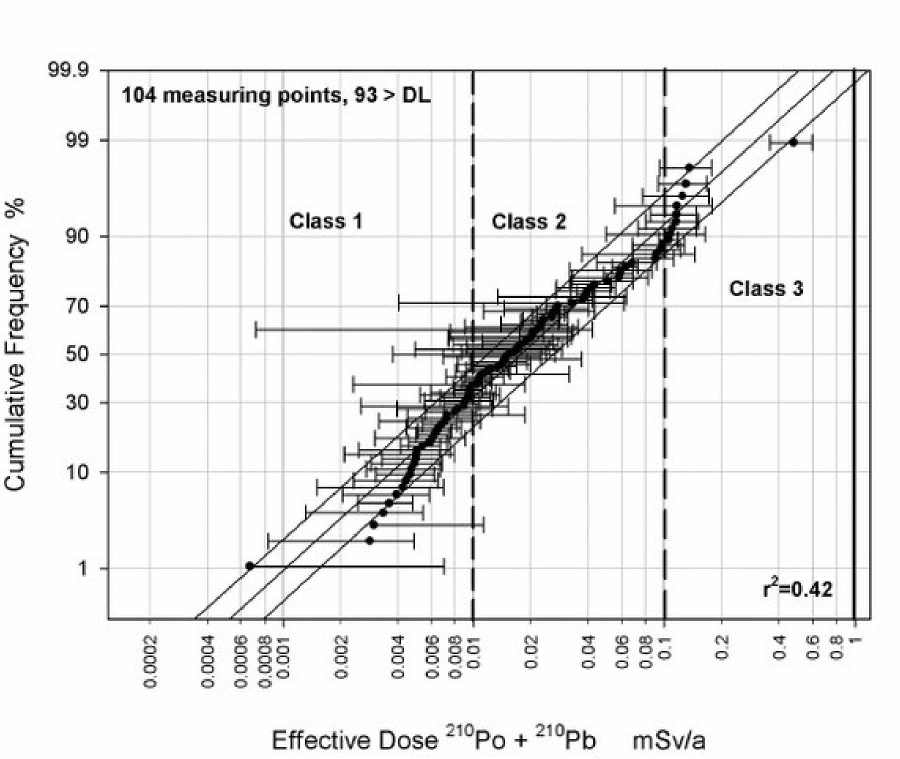

Figure 16.3: *Cumulative frequency of the total effective dose by ^{210}Po and ^{210}Pb calculated for adults*

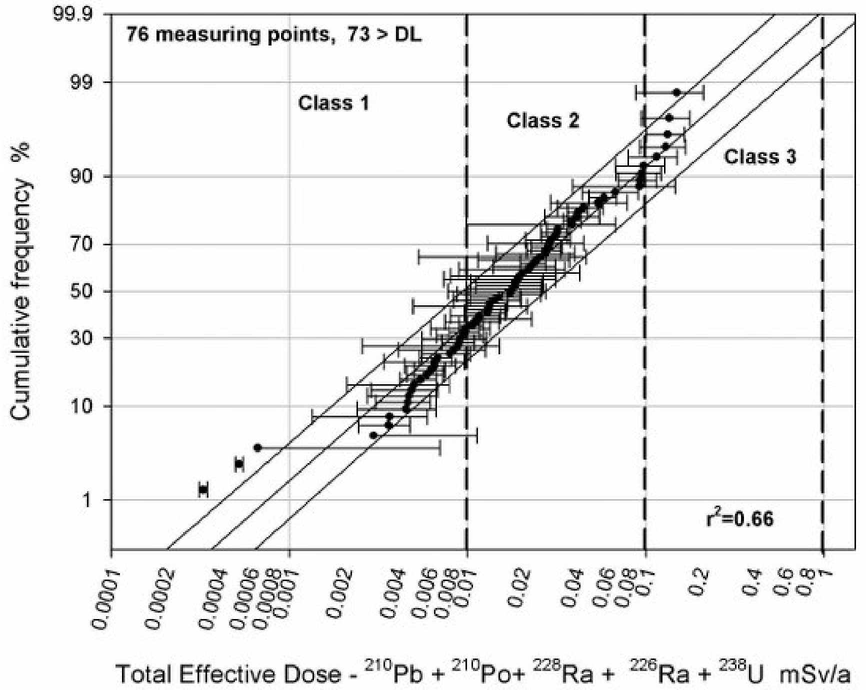

Figure 16.4: *Cumulative frequency of the total effective dose (^{210}Po, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{238}U) calculated for adults*

In Figure 16.4 the **total effective dose of ^{226}Ra, ^{228}Ra, ^{238}U, ^{210}Po and ^{210}Pb** is demonstrated. At 76 measurement points all five nuclides were analyzed, at 73 at least one of them is above decision limit. The cumulative frequency distribution is similar to the one of the ^{210}Po and ^{210}Pb dose (Figure 16.3) because they have the highest dose contribution. Again for about 10% of the drinking water a total effctive dose above 0.1 mSv/a is expected, 5 measurement points have a total dose in class 3. Some of the measurement points with the highest ^{210}Po and ^{210}Pb doses are not included in this distribution, because not all nuclides were analyzed at these points.

For **^{3}H** a reference activity concentration of 100 Bq/l is stated in the drinking water directive in addition to the total indicative dose (European Commission, 1998, Republik Österreich, 2001) and ^{3}H is not taken into account for dose calculations. But for interest also the effective dose of ^{3}H is determined in this thesis, with a **low dose conversion factor** of $1.8 \cdot 10^{-8}$ Sv/Bq according to WHO (2004). Figure 16.5 demonstrates the cumulative frequency distribution of the effective dose in drinking waters caused by ^{3}H. 129 samples were analyzed for ^{3}H, 59 are above decision limit. The determined effective doses are all in class 2 (0.01 to 0.1 mSv/a), and according to the cumulative frequency distribution (r^2=0.9) no effective doses caused by ^{3}H above 0.1 mSv/a are expected for 99.9% of all drinking waters in Upper Austria.

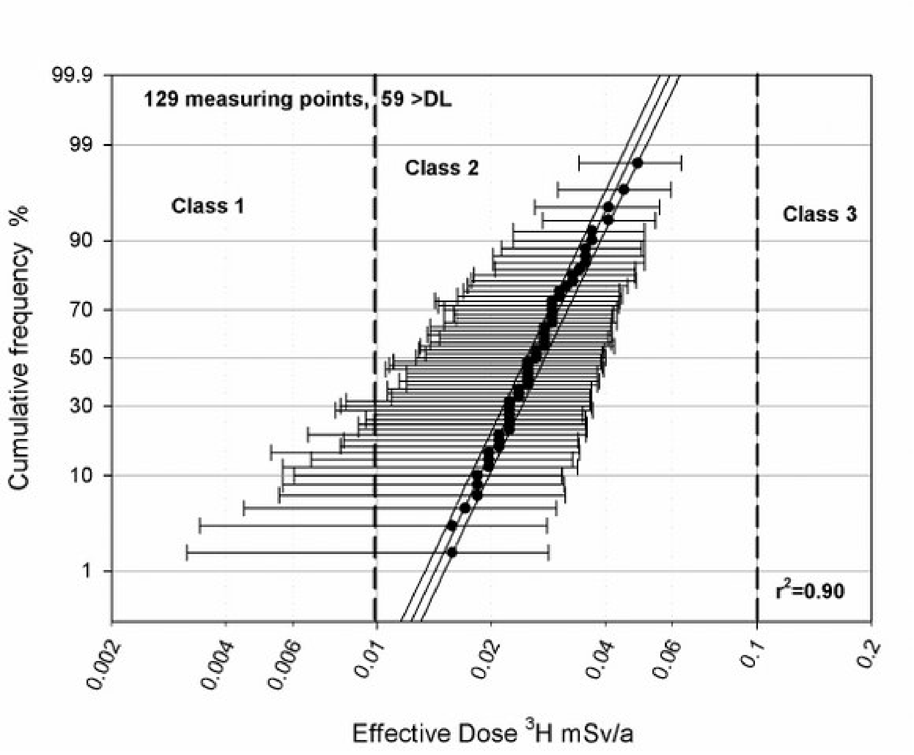

Figure 16.5: *Cumulative frequency distribution of the effective dose caused by ^{3}H for adults*

16.3 Effective Doses for Different Age Groups

All determined and by now discussed doses refer to **adults** (age >17a) with an annual consumption of 730 l/a as stated in the Austrian drinking water directive and the ÖNORM S 5251:2005. Table 16.1 shows the **dose conversion factors** and **annual drinking water consumption** amount of **babies and children of different age groups**. In some countries total doses and standard or reference parameters are related to these age groups or are discussed in different publications (e.g. Aurand & Rühle, 2003, Bundesrepublik Deutschland, 2001, Republik Österreich, 2001). Out of interest and discussion some dose determinations with different age groups and nuclides were done in this thesis, too.

The following figures demonstrate the **cumulative frequency distributions for effective dose caused by different radionuclides in drinking water for different age groups** with the dose conversion factors and annual drinking water consumption listed in Table 16.1. No effective dose was determined for the age group 10 to 17 years because of the lack of a defined intake (Risica & Grande, 2000).

The cumulative frequency distribution of the effective dose caused by ^{228}**Ra** in drinking water shows a effective dose slightly above 0.1 mSv/a for babies ($<$ 1 year) at 3 measurement points, for all other age groups the effective dose at all measurement points are below 0.1 mSv/a (Figure 16.6). For more than 80% of the Upper Austrian drinking waters a ^{228}Ra dose even for age group $<$ 1 year below 0.1 mSv/a are expected, for all other age groups for more than 99.9% a dose below 0.1 mSv/a is predicted. So the existing discussion about the huge dose contribution and hazard by ^{228}Ra in drinking water for babies (e.g. in Germany) cannot be verified within the measurement results of this thesis.

For ^{226}**Ra** only one of 129 measurement points shows a effective dose above 0.1 mSv/a for babies, the determined dose for all other age groups are clearly below 0.1 mSv/a (Figure 16.7). Figure 16.8 illustrates the cumulative frequency of the sum of the effective dose of ^{226}**Ra and** ^{228}**Ra** according to ÖNORM S 5251, but determined for all age groups. The cumulative frequency distributions are similar to the ones of the dose of ^{228}Ra, because only 7 measurement points of ^{226}Ra are above decision limit and the measurement point with the highest measured ^{226}Ra activity concentration (WVA55) is not included in this distribution, because ^{228}Ra was not analyzed at this point. As discussed above, the contribution of the effective doses were only summed at measurement points where all nuclides relevant for the sum were analyzed.

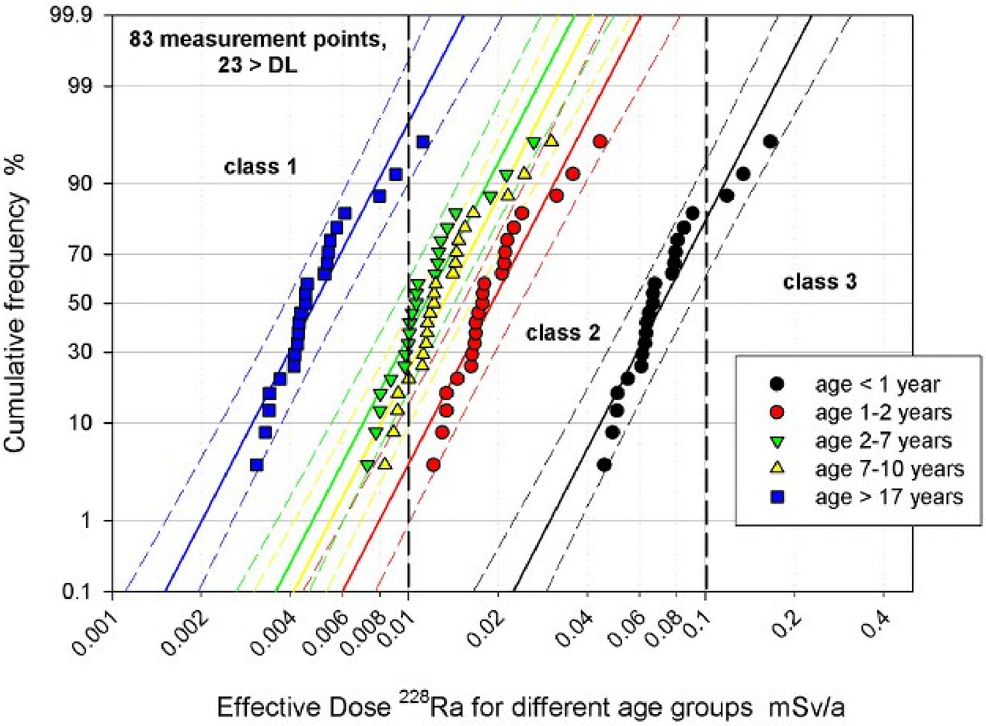

Figure 16.6: *Cumulative frequency distribution of the effective dose in drinking water caused by ^{228}Ra for different age groups*

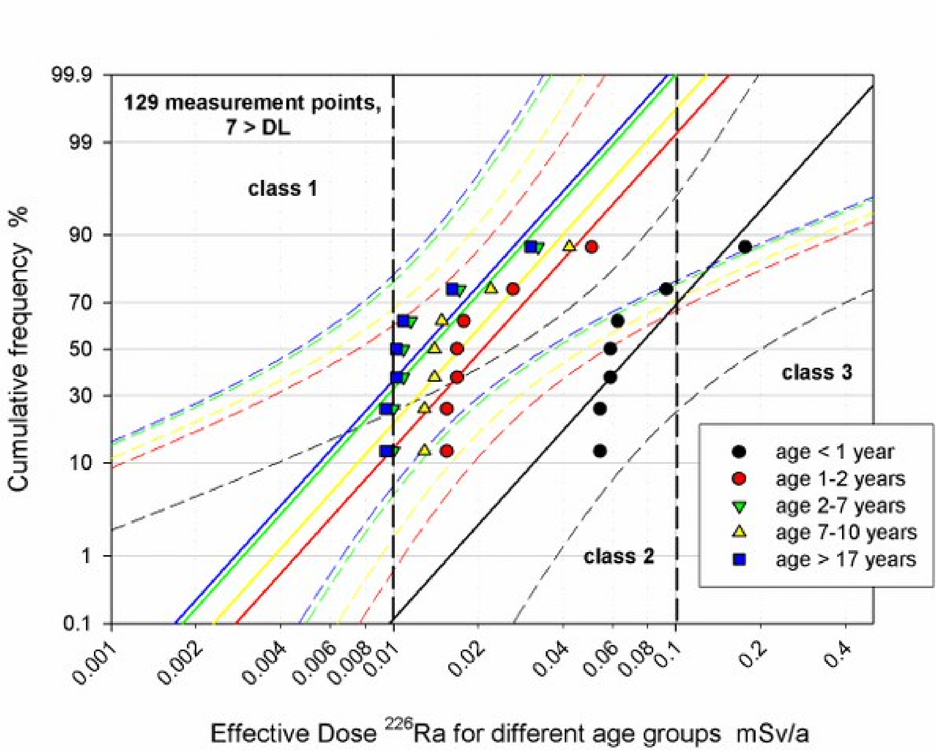

Figure 16.7: *Cumulative frequency distribution of the effective dose in drinking water caused by ^{226}Ra for different age groups*

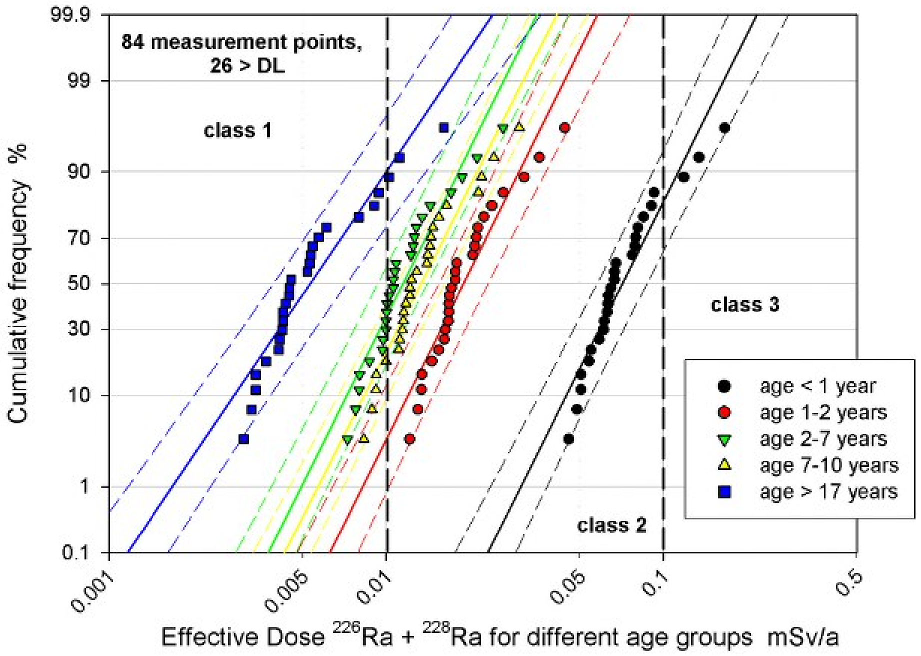

Figure 16.8: *Cumulative frequency distribution of the sum of the effective doses in drinking water caused by ^{226}Ra and ^{228}Ra for different age groups*

The **cumulative frequency distribution** of the effective dose caused by ^{238}**U** in drinking water shows more measurement points at low doses (Figure 16.9) down to 10^{-6} mSv/a, because of the very low decision limit for ^{238}U activity measurements and the **low dose conversion factors**. No measurement points with efective doses above 0.1 mSv/a caused by ^{238}U were detected, not even for babies (age group $<$ 1a). According to the cumulative frequency distribution a ^{238}U effective dose for babies above 0.1 mSv/a in Upper Austrian drinking waters is only expected for less than 1% of the waters. The cumulative frequency distribution of the efective doses caused by ^{238}U for different age groups does not show clear differences, and for example the effective dose for adults ($>$17 years) is higher than for age group 7 to 10. This is because of the similar dose conversion factors of the older age groups but the much higher annual water intake of adults (see Table 16.1).

The **sum of the effective dose contributions of** ^{228}**Ra,** ^{226}**Ra and** ^{238}**U** in drinking water is illustrated in Figure 16.10. The cumulative frequency distribution is divided in two parts – the low dose contributions which are only caused by ^{238}U, and the range between 0.01 and 0.1 mSv/a, where the effective doses caused by ^{226}Ra and ^{228}Ra are predominant because of the low dose conversion factors for ^{238}U. In detail for the higher doses the ^{228}Ra effective dose contribution dominates the distribution, especially with the 3 measurement points with a effective dose for babies above 0.1 mSv/a. So the main dose contribution, by taking into account only the nuclides relevant for TID according to the drinking water directive (European Commission, 1998, Republik Österreich, 2001) is caused by ^{228}Ra in drinking water, which is obvious because of the clearly higher dose conversion factors.

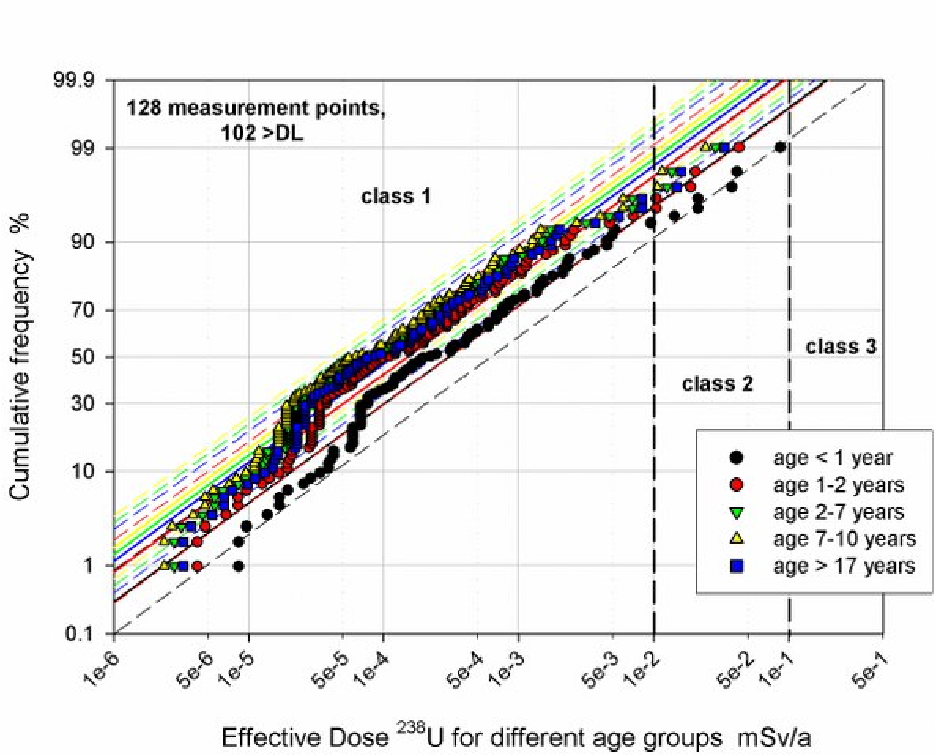

Figure 16.9: *Cumulative frequency distribution of the effective dose in drinking water caused by ^{238}U for different age groups*

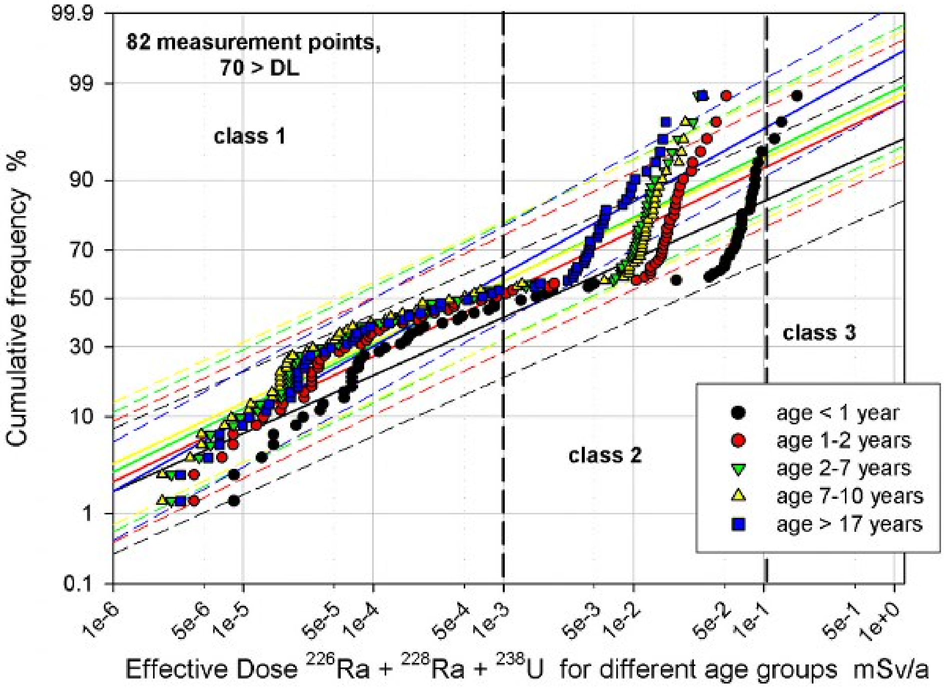

Figure 16.10: *Cumulative frequency distribution of the sum of the effective doses in drinking water caused by ^{226}Ra, ^{228}Ra and ^{238}U for different age groups*

A slightly different situation is observed for dose determinations for ^{210}**Pb and** ^{210}**Po** in drinking water. The calculated effective doses of several measurement points are above 0.1 mSv/a, especially for ^{210}Po. Rather high ^{210}Po activity concentrations were detected in the analyzed water samples and the dose conversion factors for ^{210}Po are in the range of the ones for ^{228}Ra.

Figure 16.11 shows the cumulative frequency distribution of the annual effective dose in drinking waters caused by ^{210}**Pb**. For one sample (WVA55) the determined dose for all age groups is above 0.1 mSv/a because of the clearly higher measured ^{210}Pb activity concentration (455 ± 54 mBq/l) at this point. Besides, the effective dose for **babies** (<1a) caused by ^{210}Pb at 9 measurement points are above 0.1 mSv/a, and for 1% of the drinking waters a effective dose for babies above 0.5 mSv/a is expected.

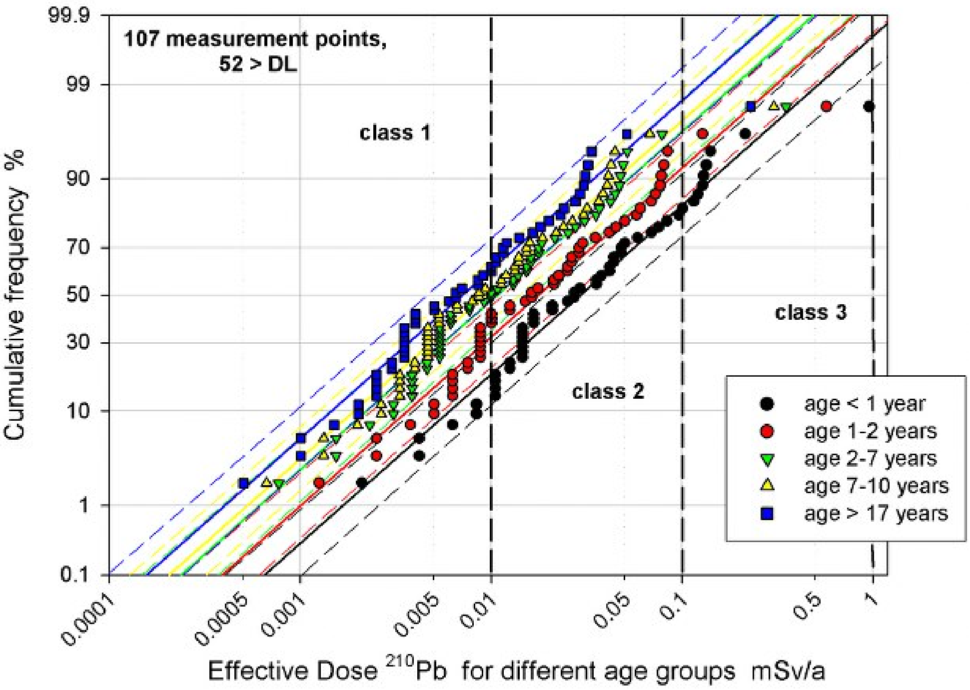

Figure 16.11: *Cumulative frequency distribution of the effective dose in drinking water caused by ^{210}Pb for different age groups*

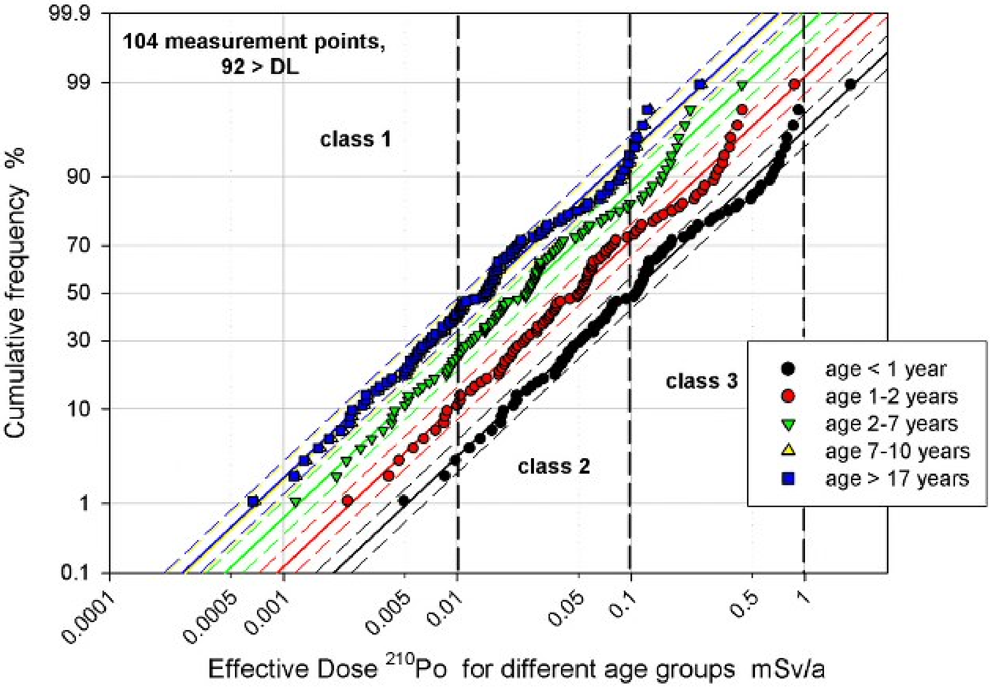

Figure 16.12: *Cumulative frequency distribution of the effective dose in drinking water caused by ^{210}Po for different age groups*

The cumulative frequency distributions of the effective doses caused by ^{210}**Po** for different age groups are demonstrated in Figure 16.12. The cumulative frequency distributions of doses for adults (>17a) and age group 7–10 a are nearly the same, because of the proportion of their dose conversion factors and the water intakes. 48 of the measurement points have a effective dose for babies (<1a) caused by ^{210}Po above 0.1 mSv/a, at least 5 have also a effective dose for adults above 0.1 mSv/a. So the dose contribution caused by ^{210}Po in drinking water is very high compared to other nuclides, which was also discussed by the Strahlenschutzkommission (SSK) (2003) (see above). For about 5% of the drinking waters in Upper Austria a effective dose for babies caused by ^{210}Po above 1 mSv/a is expected according to the cumulative frequency distribution in Figure 16.12.

A **summation of the** ^{210}**Pb and** ^{210}**Po effective doses** shows a similar cumulative frequency distribution, dominated by the higher ^{210}Po doses (Figure 16.13). The effective dose of the measurement point WVA55 with the highest measured ^{210}Pb and ^{210}Po activity concentration is 2.79 mSv/a for babies. According to the cumulative frequency distribution in Figure 16.13 a ^{210}Pb and ^{210}Po effective dose of more than 1 mSv/a can be expected for some percents of the Upper Austrian drinking water.

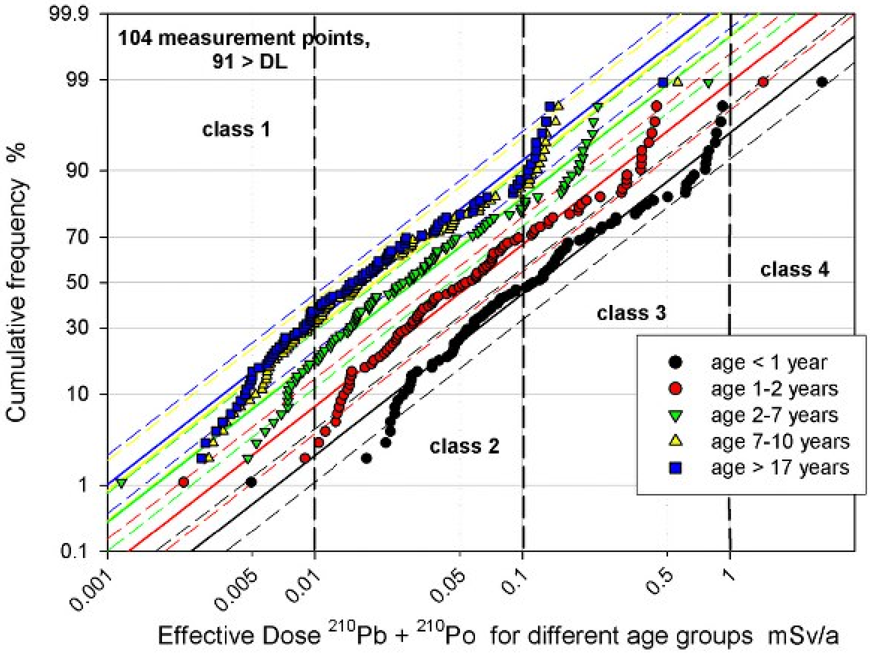

Figure 16.13: *Cumulative frequency distribution of the sum of the effective doses in drinking water caused by ^{210}Pb and ^{210}Po for different age groups*

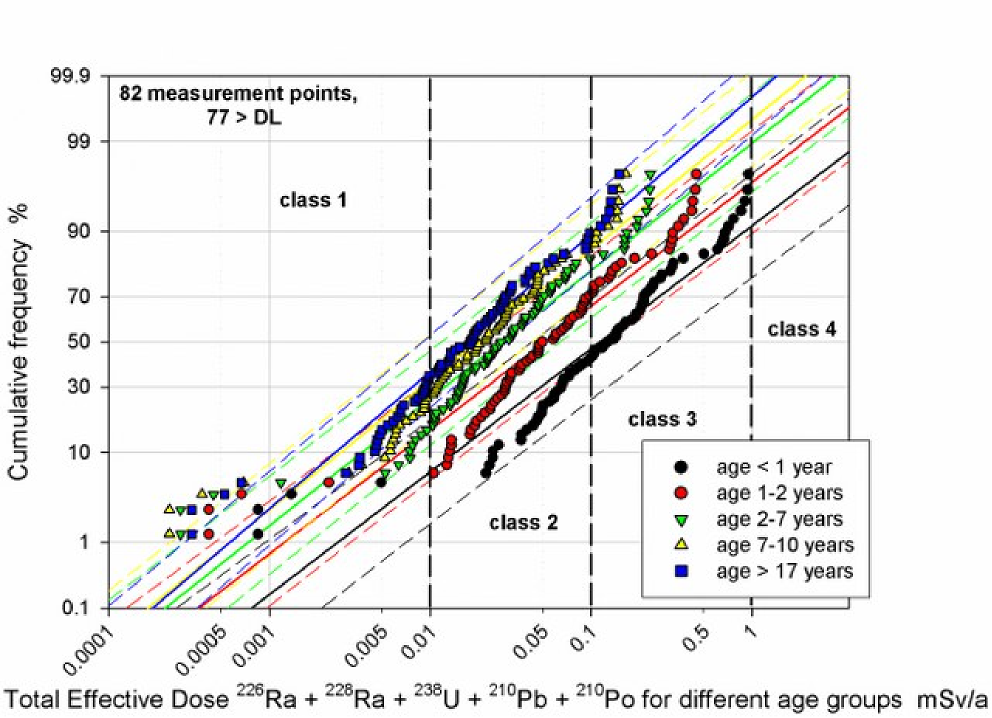

Figure 16.14: *Cumulative frequency distribution of the sum of the effective doses in drinking water caused by ^{226}Ra, ^{228}Ra, ^{238}U, ^{210}Pb and ^{210}Po for different age groups*

Figure 16.14 finally illustrates the **cumulative frequency distribution** of the sums of the **effective doses of ^{226}Ra, ^{228}Ra, ^{238}U, ^{210}Pb and ^{210}Po** for different age groups. As mentioned above ^{228}Ra was not analyzed at the measurement point WVA55, where the highest ^{226}Ra, ^{210}Po and ^{210}Pb activity concentration were measured. So, this point is not included in the sum of the effective doses demonstration of Figure 16.14 and thus no measurement point shows a total effective dose for babies above 1 mSv/a. The **cumulative frequency distribution** is again **subdivided into several regions**. In the range of the very low doses the ^{238}U dose contribution dominates, because the measurements of the other nuclides were above decision limits there and have higher dose conversion factors. In the range between 0.01 and 0.2 mSv/a the total effective dose is a real sum of all nuclides, although the ^{210}Po and ^{210}Pb predominates because regarding ^{228}Ra and ^{226}Ra only a few measurement points are above decision limit. In the range above 0.2 mSv/a only the ^{210}Pb and ^{210}Po dose contributions impact the total effective dose, and especially the ^{210}Po predominates.

This situation is also shown in Figure 16.15 and 16.16, which illustrates the **dose contributions of the nuclides to the total effective dose for adults (>17a) and babies (<1a)**. The dose contribution of ^{210}Po dominates (up to 80–100%), especially in the range of higher total effective doses together with ^{210}Pb (about 20–40%). In the range about 0.1 mSv/a total effective dose for babies and 0.01 mSv/a for adults also clear dose contributions from other nuclides exists (e.g ^{228}Ra with 20 to 40%).

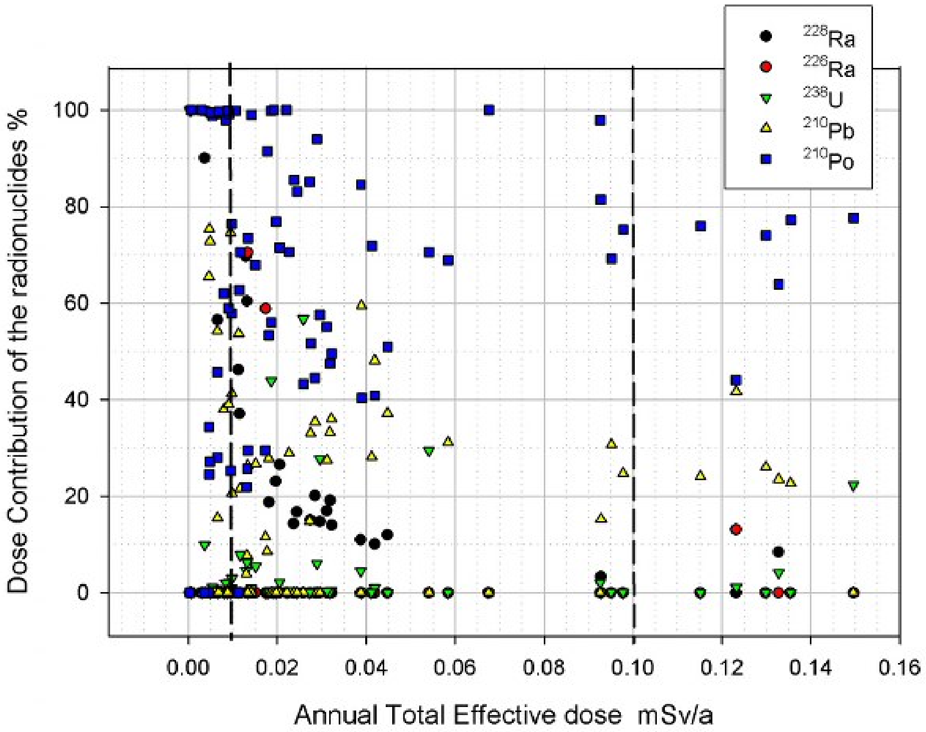

Figure 16.15: *Dose contributions of the nuclides to the annual total effective dose for adults (>17a)*

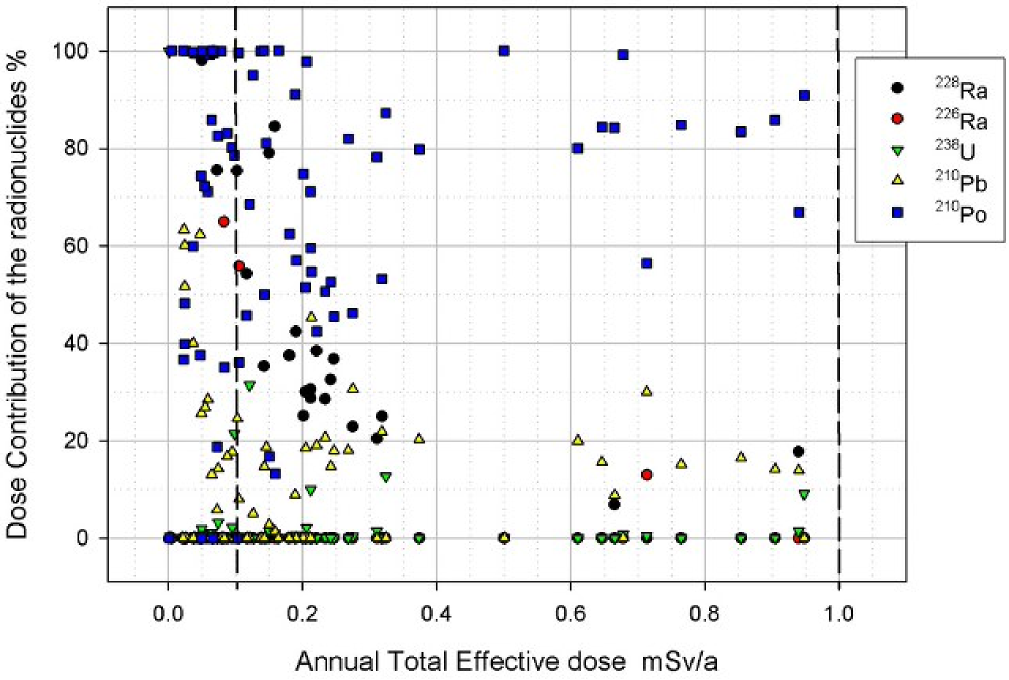

Figure 16.16: *Dose contributions of the nuclides to the annual total effective dose for babies (<1a)*

In Table 16.2 the **sum of the effective doses** of ^{226}Ra, ^{228}Ra, ^{238}U, ^{210}Po and ^{210}Pb for different age groups are listed for some **selected measurement points**. These points were selected because of clearly higher activity concentrations of one or more radionuclides (discussed in Chapter 11). The table illustrates the difference of total effective doses in waters of different measurement points.

16.4 Social and Legal Impact of Results

Dose assessments of radionuclides in drinking water are an **issue of steady discussion** within countries of the European Union and others and also within experts of one country. There are also discussions about the **legislative regulation** and **implementation** of **dose parameters** or of **reference activity concentrations**. As discussed and explained in this chapter, the European drinking water directive only states the total indicative dose and a tritium activity concentration, which was also adopted in various national laws (like Austria and Germany for example). For other radionuclides like the radon progenies ^{210}Po and ^{210}Pb only different (activity concentration related) recommendations exists (see Chapter 3.1). So some institutions for example require standardization for all radionuclides and a definition of dose reference value for the utilization of drinking water related to the activity concentrations of all radionuclides in the water (Strahlenschutzkommission (SSK), 2003).

Table 16.2: *Different total effective doses in waters of different measurement points*

Measurement point	**Total dose (^{226}Ra, ^{228}Ra, ^{238}U, ^{210}Po, ^{210}Pb) (mSv/a)**				
	<1 a	**1–2 a**	**2–7 a**	**7–10 a**	**>17 a**
H1	0.713	0.348	0.192	0.147	0.123
H14	0.940	0.428	0.230	0.164	0.133
HEIL5	0.001	0.0004	0.0003	0.0002	0.0003
WVA31	0.854	0.422	0.221	0.144	0.130
WVA47[1]	0.922	0.437	0.218	0.129	0.124
WVA48	0.0014	0.0007	0.0005	0.0004	0.0005
WVA55[1]	2.793	1.444	0.786	0.560	0.477
WVA70	0.948	0.451	0.233	0.145	0.150
WVA83	0.099	0.047	0.025	0.017	0.019

1) *For the doses only ^{210}Po and ^{210}Pb were taken into account.*

As shown above the dose caused by ^{210}**Po and** ^{210}**Pb** is not negligible compared to the dose contribution of the radium isotopes. So these nuclides should be taken into account in applied regulations and dose calculations. This fact was also surveyed and discussed by other studies (e.g. Jia & Torri, 2007) who surveyed 17 drinking waters in Italy and found the highest dose contribution in drinking waters from a single nuclide is caused by ^{210}Po with about 30%.

Another big issue of discussion are **age groups** – in the Austrian standard ÖNORM S 5251 (Austrian Standards Institute, 2005b) the total indicative dose reference value of 0.1 mSv/a is defined for adults (> 17 years), but there are some countries and experts who recommend to take into account the dose for babies or young children in directives or guidelines. Different studies discuss the health effect for babies and young children by ionisation radiation, but also the effect by drinking water might be small because of the low consumption of water by babies and young children. Some publications discuss that in some life periods the organism is more susceptible to the presence of radionuclides in water. Bronzovic & Marovic (2004) for example discussed that the highest exposure group to ^{226}Ra in drinking water are infants and boys between 13 and 17. These are life periods of intensive production of the sexual hormone testosterone, which aids Ca deposit in the bone and accompanied with rapid growth that could amplify deposition of ^{226}Ra (as Ca homologue) into the bone.

An annual medium radiation exposure by drinking water was estimated in Germany for babies in the magnitude of 20 µSv and for adults of 2 µSv, which is compared to the radiation exposure by intake of natural radionuclides in total food only about 3% for babies and less than 1% for adults of the estimated effective dose (Aurand & Rühle, 2003).

For the results of this thesis a medium total dose (according to ÖNORM S

5251 taking into account only ^{226}Ra and ^{228}Ra) of about 3 µSv/a for adults and about 60 µSv/a for babies was detected. This is also related to the 1% of the annual effective dose by ingestion of natural radionuclides of an adult in Austria (0.29 mSv/a, Mück (2001) – see Chapter 2.3). The medium annual total dose taking also into account ^{210}Po, ^{210}Pb and ^{238}U of the results of this thesis yields 20 µSv for adults, which is about 7% of the annual effective dose by ingestion of natural radionuclides of an adult in Austria (Mück, 2001).

The dose limit of 0.1 mSv/a for drinking water is from **minor impact for radiation protection** and **health care of the population** compared to radiation exposure by other natural radioactivity sources like radon in indoor air or ingestion by food (Aurand & Rühle, 2003). Therefore a reasonable method should be found to comply with reasonable (and well defined, standardized and discussed) limits for radiation protection by radioactivity in drinking water with reasonable costs and efforts for the advantage and protection.

16.5 Alternative Exposure Model

The estimated doses by natural radioactivity in drinking water did not much exceed limits or recommended values, especially not for adults. There are different studies and publications for such dose estimations in different countries. Most of them carry out this total dose estimation for adults according to the recommended method by the WHO and EU (see above) with a consumption of 2 liters per day during one year period of time, which is an annual amount of 730 l for adults and dose conversion factors. The 730 liters annual consumption is treated as if it was ingested at once for dose estimation. Bronzovic et al. (2006) presented a different method of ^{226}Ra effective dose calculation following a **continuous intake of water** (taking into account the intake frequency), which is based on the assessment of cumulated activity including $m(t)$ value, which describes the **fraction of a unit intake retained in the whole body at time t after intake** (IAEA, 2004). The paper says that most of the ^{226}Ra is excreted from the body during the first days after ingestion while the rest of ^{226}Ra follows the metabolic path of calcium which means that it would be lodged principally into the bone. Each next intake follows the same distribution in the body. The great amount of ingested ^{226}Ra will be excreted from the body (by faeces and urine) and the rest will be distributed among the organs according to metabolic parameters. This method shows significant lower values of ^{226}Ra effective dose, which will not exceed the recommended 0.1 mSv/a for most drinking waters.

Some dose surveys were carried out in this thesis based on the dose estimations by Bronzovic et al. (2006) discussed above. For the survey the **continuous water intake** (2 liters a day) and the **excretion of radionuclides from the body** are used. The excretion of radionuclides from the body is described by the $m(t)$ value (IAEA, 2004). In this IAEA publication the body retaining fraction of intake $m(t)$ is given for different radionuclides, time (given in days, on an expanding scale), types of intake (ingestion, inhalation) and for the whole body content or parts of the body. For estimation the daily dose, the $m(t)$ value of every day has to be calculated by logarithmic interpolation between adjacent values as recommended in the IAEA publication (IAEA, 2004). Figures 16.17 to 16.19 show the run of the $m(t)$ **value for ^{228}Ra, ^{226}Ra, ^{210}Po and ^{210}Pb**. The distribution of the $m(t)$ value of ^{228}Ra and ^{226}Ra is the same. The calculation of the $m(t)$ values used were based on the f_1 values for adults (0.1 for ^{210}Po, 0.2 for ^{226}Ra, ^{228}Ra and ^{210}Pb). The f_1 values give the fraction of an element entering the gastrointestinal tract which reaches body fluids and the retention of a radionuclide in the body depends on it. These values are reported by the ICRP for age groups. (IAEA, 2004, ICRP, 1993)

The distributions of the $m(t)$ values (Figures 16.17 – 16.19) illustrate a **very rapid decline in the first days after intake**. After the first day e.g. only about 70% of the ^{226}Ra is remaining in the body, after the second day only about 40% and after the fourth day below 10% are remaining in the body. For the rest of the year some percentages of the radionuclides which were ingested are remaining in the body and followed the metabolic path of calcium (see above).

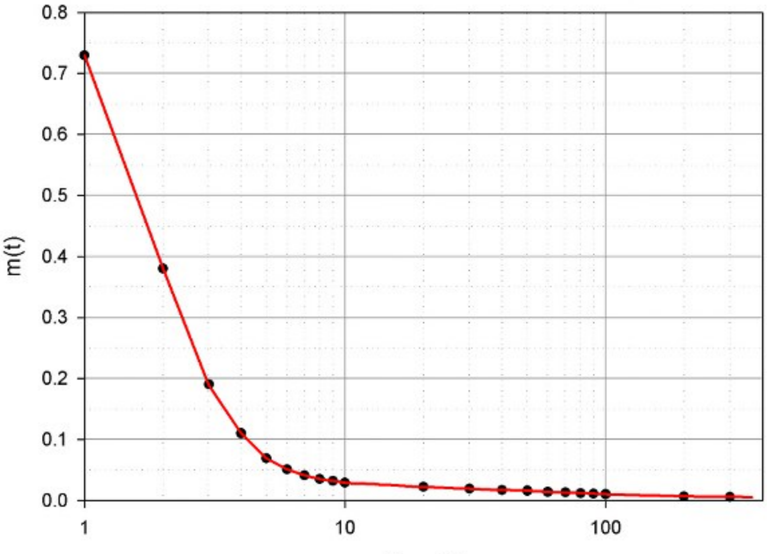

Figure 16.17: *The fraction of a unit intake retained in the whole body at time t after intake ($m(t)$ value, for one year) for the radionuclides ^{226}Ra and ^{228}Ra*

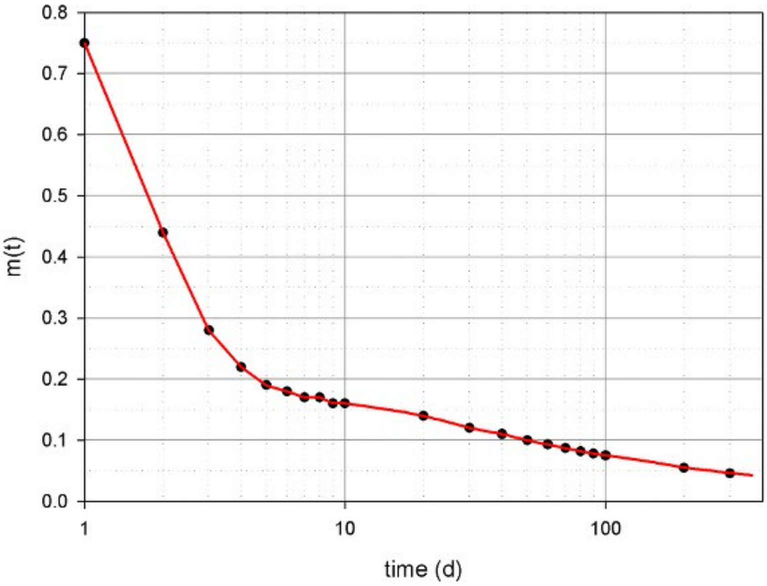

Figure 16.18: *The fraction of a unit intake retained in the whole body at time t after intake ($m(t)$ value, for one year) for the radionuclide ^{210}Pb*

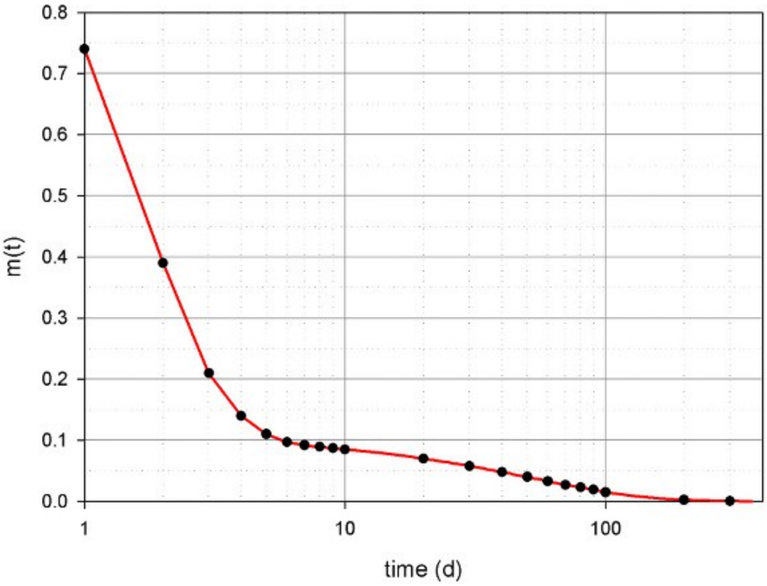

Figure 16.19: *The fraction of a unit intake retained in the whole body at time t after intake ($m(t)$ value, for one year) for the radionuclide ^{210}Po*

The radionuclide activity concentration in the body is therefore determined by the radionuclide activity concentration in the drinking water, the daily intake (2 liters) and the $m(t)$ value (Equation 16.3). So a **daily total radionuclide activity in the body** was calculated by a daily 2 liter drinking water intake with a constant activity concentration and a daily excretion specified by the $m(t)$ value. Because of the high rate of excretion of the radionuclides from the body, the remaining radionuclides in the body trend to constant activity after a few days. This constant activity is slightly above the daily intaken activity concentration by drinking water. Figure 16.20 shows as example this activity distribution for one year for ^{228}Ra with an activity concentration in drinking water of 0.2 Bq/l and a 2 litres per day intake. The activity remains constant at 0.4265 Bq since the fifth day.

$$A_{body} = A_{water} \cdot m(t) \tag{16.3}$$

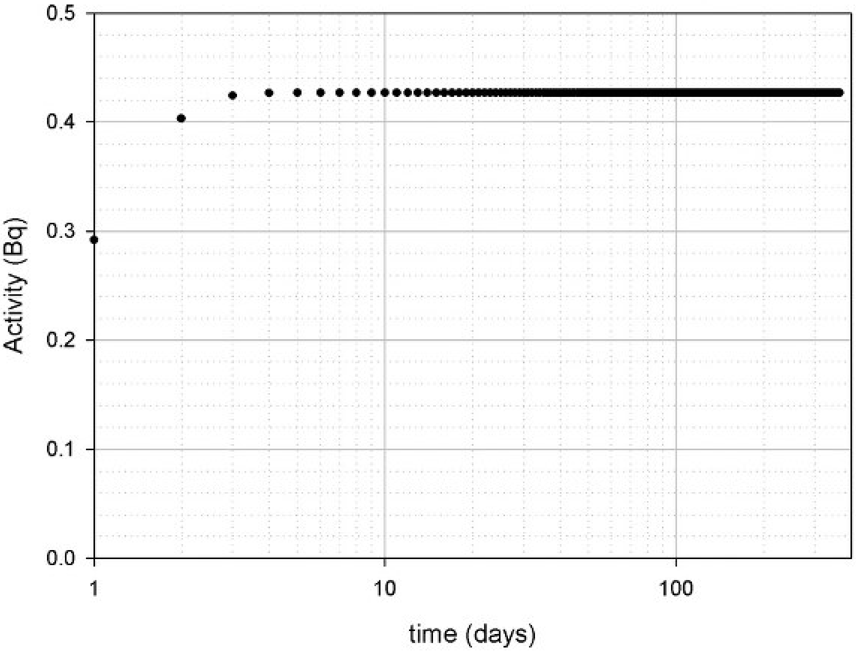

Figure 16.20: *^{228}Ra activity distribution in the body caused by drinking water (0.2 Bq/l) and 2 l daily intake in one year*

For dose estimations the **absorbed dose** caused by radionuclides in drinking water in the body is calculated (see Equation 16.4) and out of it the **equivalent dose** with the **radiation weighting factor** (Equation 16.5). The absorbed dose and equivalent dose are determined for the whole body without distinguishing between tissues and organs and related to a 70 kg reference person. For estimation of the absorbed dose in the body the energy emitted in the body by a radionuclide in drinking water has to be determined. ^{226}Ra and ^{210}Po are mainly α-emitters, ^{228}Ra and ^{210}Pb β-emitters. In Table 16.3 the α and β energies of the nuclides and their branching ratios are listed. The energy is transformed from eV into J by 1eV=$1.602 \cdot 10^{-19}$ J (Bröcker, 1993).

For the **effective dose** (Equation 16.6 and 16.7) it is also necessary to determine the fraction of energy which is absorbed by the target organ, called **absorbed frac-**

tion. For α-emitters it is 1, because of the small reach of the α-particles the emitted energy is believed to be absorbed in the same organ (Bronzovic et al., 2006). For β-particles it is also set 1, because in this estimation only the energy absorbed in the whole body is taken into account. Gamma decay and gamma energies are not taken into account in these estimations. As mentioned above, in this dose estimation the effective dose is determined for the whole body without differentiation of tissues and organs. So no **tissue weighting factors** are taken into account. The radiation weighting factor for α-particles is 20, for β-particles 1.

$$D_d[\mathrm{Gy/d}] = \frac{dE_d}{dm} = \frac{\sum_{\alpha,\beta} E_{\alpha,\beta}[\mathrm{J/Bq}] \cdot A_{body,d}[\mathrm{Bq/d}]}{m[\mathrm{kg}]} \tag{16.4}$$

D_d	... absorbed dose per day
E_d	... absorbed energy in the body per day
$E_{\alpha,\beta}$	... energy per alpha or beta decay
$A_{body,d}$	... activity in the body per day
m	... mass of reference person

$$H_T[\mathrm{mSv/d}] = \sum_{\alpha,\beta} D_d \cdot w_R \tag{16.5}$$

H_T	... equivalent dose per day
w_R	... radiation weighting factor

$$E_{eff,d}[\mathrm{mSv/d}] = H_T \cdot w_T \cdot \Phi \tag{16.6}$$

w_T	... tissue weighting factor
Φ	... absorbed fraction
$E_{eff,d}$	... effective dose per day

$$E_{eff,a}[\mathrm{mSv/a}] = \sum_{d=1}^{365} E_{eff,d} \tag{16.7}$$

$E_{eff,a}$	... effective dose per year

The **effective dose for the different radionuclides according to the discussed method** was determined for the parametric value (intersection between class 2 and 3) of the activity concentration in the drinking water for comparison reasons. As discussed several times within this thesis this activity concentration complies with the total indicative dose of 0.1 mSv/a as stated in the drinking water directive. In Table 16.4 the used activity concentrations are given, together with the effective dose after one year, determined as discussed above, taking into account the daily water intake and drinking water activity concentration, the fraction of a unit intake retained in the whole body at time t after intake $m(t)$, the absorbed dose (by absorbed energy in the body by radioactive decays), the radiation weighted factor and the absorbed fraction (see Equation 16.4 – 16.7). According to Krieger (2002) the **natural decay chains were separated into sections** because of the half life of the progenies. ^{226}Ra is examined separately, for ^{228}Ra the progenies ^{228}Ac, ^{228}Th and ^{224}Ra should be taken into account, because of the short half life of the decay products – they have the same specific activity as the mother nuclide because of radioactive equilibrium. ^{210}Pb and ^{210}Po can be discussed together, but the doses are determined for each one in this purpose, because the nuclides were analyzed separately.

Table 16.3: *The major energies of the nuclides (in MeV) and their branching ratios (in %) (Korea Atomic Energy Research Institute, 2009)*

Radionuclide	**α-energy keV**	**β-energy keV**	**Relative frequency of emission**
^{226}Ra	4601.0		5.6
	4784.3	94.6	
^{228}Ra		12.8	30.0
		25.7	20.0
		39.2	40.0
		39.6	10.0
^{228}Ac		402.7	1.8
		438.6	2.5
		444.2	1.1
		481.0	4.2
		488.7	1.2
		958.6	3.1
		973.5	5.1
		1004.0	5.9
		1104.5	3.1
		1158.0	29.9
		1730.9	11.7
		2069.2	8.0
^{228}Th	5340.4		27.2
	5423.2		72.2
^{224}Ra	5448.6		5.1
	5685.4		94.9
^{210}Pb		16.6	84.0
		63.1	16.0
^{210}Po	5304.3		100.0

The **effective dose** with this method was also estimated for **babies ($<$ 1a)**. The $m(t)$ value is only given for adults in the IAEA publication (IAEA, 2004) and not for babies. For this estimation the $m(t)$ values for adults are also used for babies, which will be discussed and adapted in the future. For the daily water intake for babies 0.7 l are used, which is assumed based on a 250 l water intake per year reported in European Commission (1998) and Risica & Grande (2000). For mass of a reference baby 7 kg are applied for the estimation. The results are listed in Table 16.4.

The results in Table 16.4 show that the **effective doses calculated with this**

method are at least 2 magnitudes lower for adults than the recommended 0.1 mSv/a with the same activity concentration in drinking water because of taking into account a daily excretion of radionuclides of the body and also **at least one magnitude lower even for babies**. The calculated effective dose is especially very low for the β-emitting nuclides ^{228}Ra and ^{210}Pb because of the lower energy dose caused by β decay and the lower radiation weighting factor. But also by taking into account a radioactive equilibrium with the progenies of ^{228}Ra the estimated effective dose is again 2 magnitudes below 0.1 mSv/a for adults and again also one magnitude below 0.1 mSv/a for babies. So it seems that the dose caused by radioactivity in drinking water may be much lower than discussed within the directives.

Table 16.4: *Estimated effective dose for the whole body of adults and babies for selected activity concentrations in drinking water*

Radionuclide	**Activity concentration in drinking water (Bq/l)**	**Effective dose (mSv/a) for the whole body of adults (>17a)**	**Effective dose (mSv/a) for the whole body of babies (<1a)**
^{226}Ra	0.5	0.0074	0.0257
^{228}Ra	0.2	8.80 E-7	3.08 E-6
^{228}Ra and progenies ^{228}Ac, ^{228}Th, ^{224}Ra	0.2 each	0.0066	0.0230
^{210}Pb	0.2	8.63 E-7	3.02 E-6
^{210}Po	0.1	0.0017	0.0059

The method of calculating the total indicative dose according to the Austrian drinking water directive and the ÖNORM is a rather easy and quick method to estimate doses and yields maximum dose values. So it is a **good method of dose estimation for monitoring and surveillance because it yields an upper limit**. But if this method exceeds the recommended limit of total indicative dose of 0.1 mSv/a it is reasonable to not automatically classify this water as disabled for consumption, but taking into account a more detailed survey of the water with for example dose calculations with other methods like the one discussed here.

The **different dose calculations** and extimations yield **varying results** and should be **discussed and extended in the future**. As a particular outlook detailed research will be done based on the dose estimation method of this chapter in a future project. The effective dose should not only be calculated for the whole body but the radionuclide transfer in different organs will be taken into account (tissue weighting factors). Additionally more detailed and specific effective dose calculations will be done based on body modeling and age of the water consumers.

Chapter 17

Conclusions, Interpretation and Prospects

17.1 Conclusions and Interpretation

17.1.1 General Nuclide Activity Concentration Results

354 drinking water samples were analyzed for different radionuclides within this thesis to survey the **population's radiation exposure** caused by drinking water in Upper Austria and to survey a **hydrological-radiometric basis data net** of ground and spring waters used as drinking waters in Upper Austria.

In Table 17.1 the **median and maximum activity concentrations for all analyzed radionuclides** are listed – classified into results of the survey sampling and the detailed sampling. Compared with the activity concentrations measured in Germany reported by Rühle (1996) (Table 3.3, Chapter 3.3) the median and maximum ^{238}U activities are in the same range, although the highest measured ^{238}U activity concentration in Upper Austria is clearly higher. The ^{3}H activity concentrations are in the same range but also higher in Upper Austria. The ^{210}Po and ^{210}Pb median and maximum concentrations are clearly higher in Upper Austria. The maximum ^{222}Rn activity concentration in Germany is clearly higher than in Upper Austria, but the median activity concentration is clearly higher in Upper Austria, even in the survey sampling. For ^{226}Ra the median and maximum activity concentrations in Germany are above the ones in Upper Austria.

In Figure 17.1 the range of the activity concentrations of the surveyed radionuclides are also illustrated in a Box Plot diagram and the classification for each nuclide is displayed for interpretation purposes. Only the sampling points with activity concentrations above decision limits are taken into account and the number is listed below each box plot.

Table 17.1: *Median and maximum activity concentrations for different radionuclides in Upper Austria*

	Survey Sampling			Detailed Sampling		
Radio-nuclide	No. of measure-ments	Activity conc. median (Bq/l)	Activity conc. max (Bq/l)	No. of measure-ments	Activity conc. median (Bq/l)	Activity conc. max (Bq/l)
^{222}Rn	205	13	344	149	100	890
^{3}H	203	1.0	2.70	129	1.8	3.70
^{226}Ra	203	<DL (0.030–0.045)	0.08	129	<DL (0.037–0.055)	0.15
^{238}U	203	0.006	0.25	128	0.0025	1.02
^{228}Ra	n.a.	n.a.	n.a.	83	0.009	0.02
^{210}Po	n.a.	n.a.	n.a.	104	0.016	0.28
^{210}Pb	n.a.	n.a.	n.a.	107	0.010	0.46

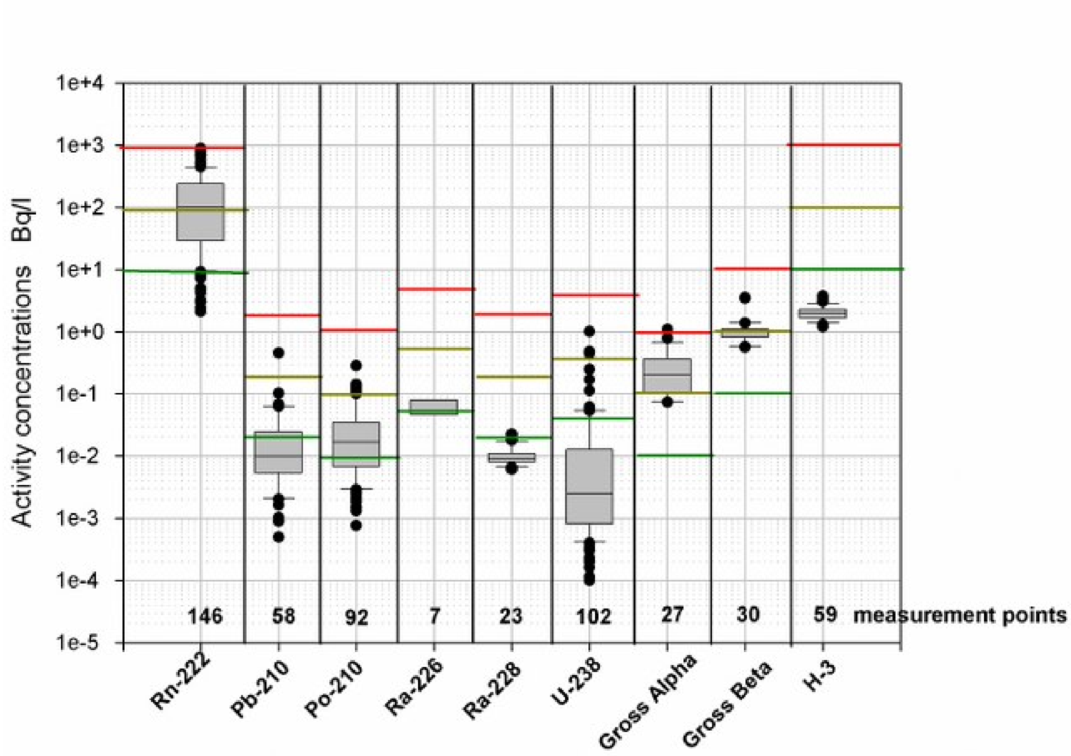

Figure 17.1: *Activity concentrations of analyzed natural radionuclides in drinking waters in Upper Austria (taking into account only sampling points with activity concentrations above decision limit*

17.1.2 Comparison and Testing of Measurement and Sampling Methods

The **testing of different sampling methods** and especially the **testing of the on-site measurement technique for radon (Triathler)** showed that correct analysis results are mainly dependent on **proper sampling**, especially for the gas radon. The best sampling method for radon measurements (without losses) is direct pipetting of water in vials on-site. For gammaspectrometry the water should be directly filled into AFNORM glass bottles carefully without bubbling on-site, because later decanting causes high radon losses. Nevertheless, some losses are possible and should be verified. Plastic containers should not be used for radon analysis, because of high radon losses at sampling and especially due to storage and transport in the not radon tight containers. To avoid radon losses by decanting water sampling should always be carried out directly into the measurement bins carefully without bubbling. With correct sampling the results of the different measurement methods (Triathler, LSC, gammaspectrometry) correspond satisfyingly and can be used as standard radon measurement methods for drinking water. For other natural radionuclides the sampling has no clear impact on the analyzed activity concentrations by different methods, but not enough comparison measurements were carried out in the framework of this thesis to make recommendations and conclusions for measurement methods.

17.1.3 Correlations of Radionuclides

In this thesis the different **radionuclide activity concentrations of the analyzed drinking water samples were surveyed for correlations**, because in the case of clear and funded correlations, predictions about radionuclide activity concentrations could be made within dose assessments without analyzing all nuclides. Some **radionuclide correlations** were detected within this thesis, but not sufficiently significant to use them as prediction for other nuclides, when only one nuclide was analyzed. The results of this thesis show that expected radionuclide correlations between products of the ^{238}**U decay chain** are not that clear or well-defined. So it seems that it is not enough to measure only the easy analyzable ^{222}Rn activity concentration and to subsequently estimate the activity concentration of e.g. ^{226}Ra or the decay products ^{210}Po and ^{210}Pb for dose assessment. It was shown, that correlations between nuclides are **dependent on geographic regions (geology)**. In some regions stronger correlations between nuclides were detected than in others, which is caused by geology and the magnitude of the measured activity concentrations. So in the lower Mühlviertel – where clearly higher ^{222}Rn activity concentrations but average activity concentrations of other nuclides were measured – low correlations were detected. The results of this thesis do not allow generalizations about nuclide correlations in drinking water, because of **small-scaled hydrogeological heterogenity of Upper Austrian regions** and due to too less measurements and especially measurement points with activity concentrations above decision limits. But this topic is of major interest and should be carried on with more measurements and surveys, because these results can affect sampling and measurement methods for drinking water monitoring fundamentally.

17.1.4 Nuclide Activity Distributions Within Water Units

In the framework of this thesis samples were taken at **different locations within water units** to survey the **nuclide activity distributions along the water flow**, because the public exposure of radioactivity from tap water at the consumers' homes is relevant for the objective of radiation protection. So it is interesting to verify the variation of the activity concentration of water on its way from the wells to the consumers and to examine influences like **water treatment**, **elevated tanks** and **pipes**.

The radon activity concentration at wells and spring is rather high, especially in the lower Mühlviertel, nevertheless the measured radon activity concentrations at the consumers are typically at least one magnitude lower and below all limits. The main reason for the **reduction of radon activity concentration** within the run of the water from the well to the consumer is **duration of dwell**, because of the short half life of radon and degassing. Effective reduction of radon activity concentration by **de-acidification** by marble gravel could not be detected. **Aeration** decreased radon activity, but not as effective as expected. **Mixing waters** with high radon activity concentrations with waters with low radon activity concentration is more effective. The losses of radon activity concentration from the well to the consumer show, that in general for consumers of water from water supplies no radon exposure problems are expected, except for some special cases. It seems that the radiation protection attention has to be paid to **private wells**, where the water has only a short way and duration of dwell between the well and the consumer.

For **other radionuclides** generalization is less acceptable yet. For ^{226}Ra, ^{228}Ra, gross alpha, gross beta and ^{3}H at many sampling points the activity concentration is below detecision limit or equal within uncertainities. No clear impact could be detected on duration of dwell or treatment techniques. It also seems that de-acidification and duration of time has no impact on the ^{238}U activity concentration, mainly because of its long half-life. In our samples the best method to reduce ^{238}U activity concentration in the water is to mix it with water with lower ^{238}U activity concentration. So, for the radiation protection point of view, nuclides like ^{238}U, whose activity concentrations in the water are not reduced by duration of dwell should not be disregarded. For ^{210}Pb and ^{210}Po also too few measurement results exist to make good conclusions. On the one hand it seems that the activity concentrations are reduced by duration of dwell, on the other hand it is also possible that ^{210}Pb is enriched in de-acidification. It might also be, that the ^{210}Po and ^{210}Pb activity concentration are strongly effected by interactions in the pipes.

Further studies would be interesting with the purpose of possible activity reduction within water units to make fundamental generalizations and conclusions about activity concentrations of wells and springs in some regions and the dose relevance for the population. For radiation protection reasons and regulations purposes it is reasonable to take samples directly at consumers' tap waters because at least for radon a clear reduction within the water flow was detected. Further studies should deepen these results and a directive or standard should be established to regulate the sampling and analyzes for regulation purposes and dose assessment.

17.1.5 Correlation of Radionuclides and Other Parameters in Drinking Water

The survey of **correlations between radioactivity concentration and other parameters** like **pH**, **water temperature**, **electric conductivity** or various **(heavy) metals** in drinking waters **did not bring up any clear correlations**. Some trends were noticed, but also expected correlations (e.g. between some heavy metals and nuclides) could not be verified clearly. It seems that radionuclide concentrations in drinking water are not correlated with other heavy metals or physical parameters. To generalize these statements further measurements and surveys should be carried out.

17.1.6 Impact of Geology on Radioactivity in Drinking Water

However, the survey of **geology's impact on radioactivity in drinking water** showed **clear dependencies**. As expected the radon activity concentrations in the drinking waters in the Bohemian massif are in general clearly higher than in others, but there are also many differences within the concentrations in the Bohemian massif possibly caused by different bedrocks and granite types. Therefore it would be reasonable to **survey different granite types** and other rocks for their uranium and other natural radionuclide content. The activity concentrations of ^{210}Pb and ^{210}Po in drinking waters are also clearly higher in the Bohemian massif, but for all other nuclides this effect cannot be detected that clearly. The points with higher ^{238}U and ^{226}Ra activity concentrations are distributed among Upper Austria. The differentiation between waters from **deep wells** and from **springs** also did not show higher activity concentrations in deep wells as clearly as expected. A slightly higher radon activity concentration was detected in total Upper Austria in drilled wells than in springs, but not as clearly as expected. And in some geological regions the activity concentrations were higher in springs (e.g. Danube fault). It seems that in some bedrocks the water of drilled well originates from compact bedrock and does not absorb the radioactivity concentration of the material. In contrast in some surface near springs in weathered bedrock the water has more contact with the material and absorbs more radioactivity concentration from it and therefore has higher activity concentrations than some waters from deep wells in the same region. So the results of the geological survey show that not all expected correlations are detected that clearly and cannot be assumed as definite. More measurements and surveys would be reasonable in this field for better generalizations and detailed correlations for the interpretation and classification of drinking water in geological regions.

17.1.7 Map of "Radon in Spring and Ground Waters in Upper Austria"

The map "Radon in Water in Austria" (Friedmann (2006, 2008), Figure 3.1, Chapter 3.2) illustrates the regions in Austria with higher radon risk by the consumption of drinking water. The motivation of the map is an **assessment of the probability of occurrence of higher radon activity concentrations in spring and ground waters**.

For comparison reasons and for interpretation purposes the **radon results of this study** were also illustrated (by ArcGis software) in a map of Upper Austria by communities. The results of the survey and detailed sampling were used for this purpose and related with the communities. A **medium and maximum radon activity concentration in the communities** was mapped. Figures 17.2 and 17.3 demonstrates the communities with the medium and maximum radon activity concentration in drinking water classified according to the classification of this thesis (see Chapter 6). No results above 1000 Bq/l exist, therefore class 4 does not exist in the maps. Figures 17.4 and 17.5 show the communities with the medium and maximum radon activity concentration in drinking water arranged in another classification to view more details in the radon activity concentration region above 100 Bq/l. In the white communities no measurements were carried out within this thesis.

Communities with medium and maximum radon activity concentrations above 300 Bq/l were only detected in the lower Mühlviertel, in southern Upper Austria only communities with medium radon activity concentrations below 30 Bq/l exist.

For other nuclides too few measurement points with activity concentrations above decision limit exist, or the results are that low, that summing up the results for communities and classify them, yields a situation with all communities within the same class.

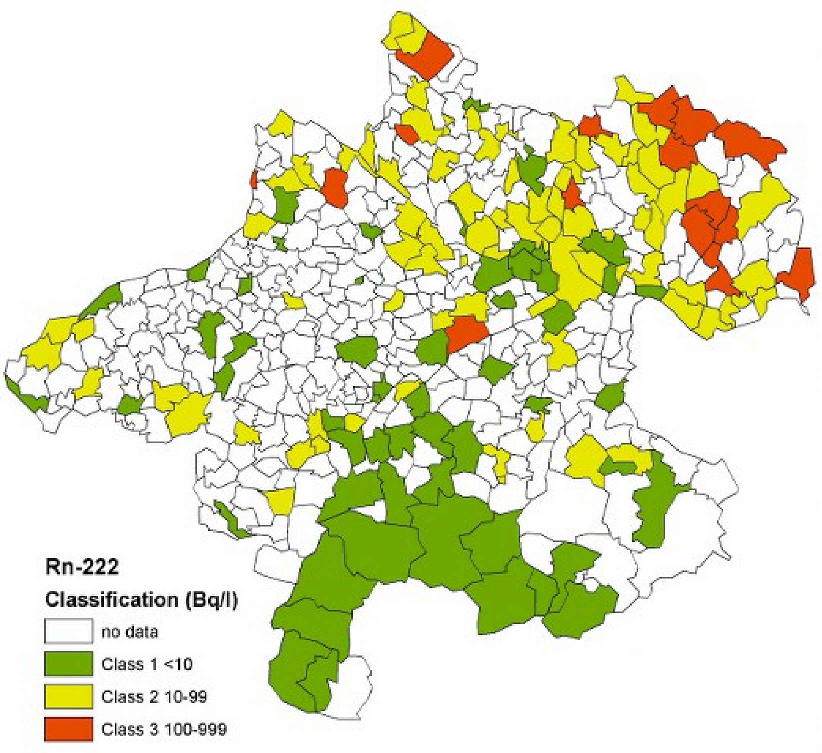

Figure 17.2: *Classified medium radon activity concentration in drinking water in communities, classification according to Chapter 6*

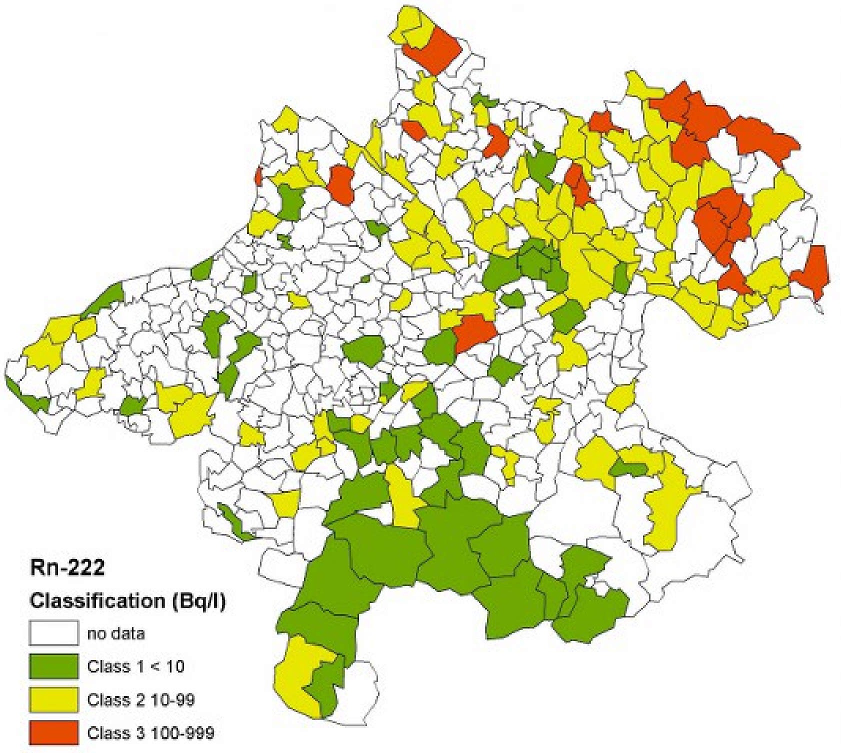

Figure 17.3: *Classified maximum radon activity concentration in drinking water in communities, classification according to Chapter 6*

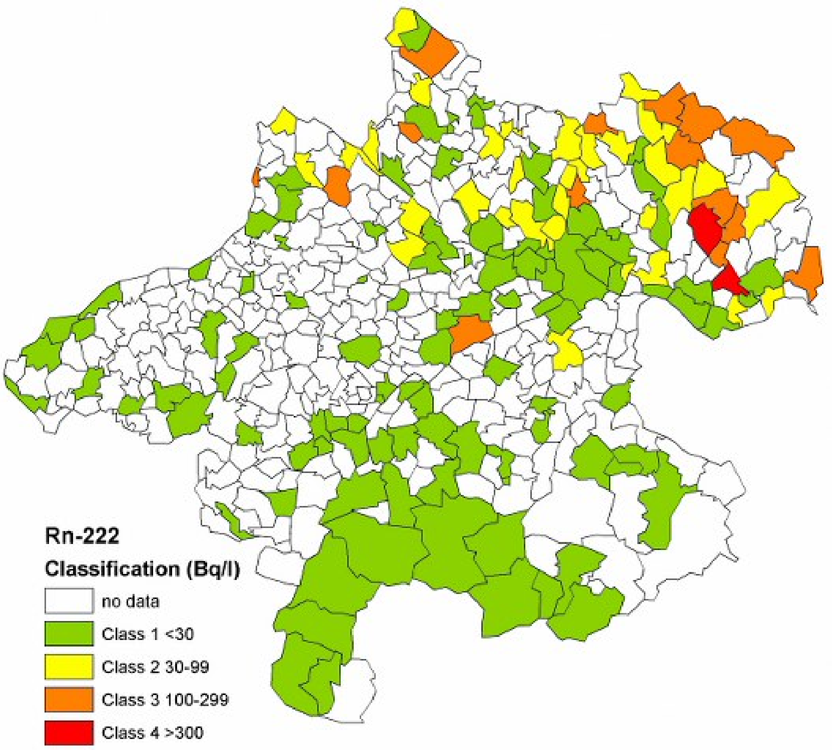

Figure 17.4: *Classified medium radon activity concentration in drinking water in communities, new classification*

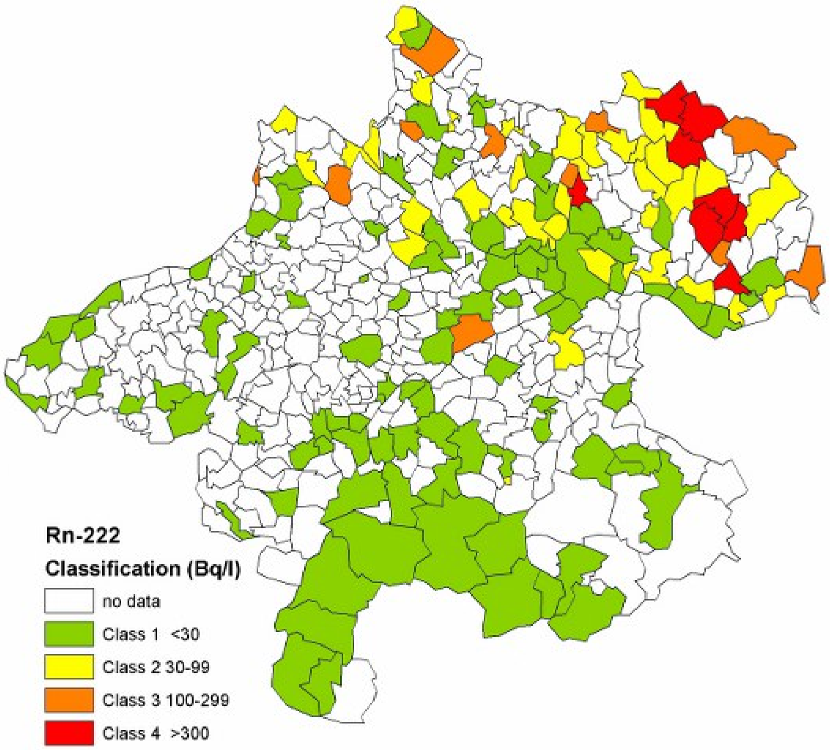

Figure 17.5: *Classified maximum radon activity concentration in drinking water in communities, new classification*

17.1.8 Correlation Between Indoor Radon and Radon in Drinking Water in Upper Austria

In the "**Austrian Radon project**" (Friedmann, 2006, 2008) about 40000 measurements of indoor radon were carried out in Austria and the data were processed and mapped. The medium and maximum indoor radon activity concentrations of communities are given in Friedmann (2006). This data were used to correlate them with the radon drinking water concentration (analyzed within this study) in the Upper Austrian communities.

Figures 17.6 and 17.7 illustrate the **correlation between the medium and maximum radon indoor and drinking water concentration in the communities**. The correlation is very weak with an r^2 value of 0.123 for the medium and an even lower for the maximum concentrations (r^2=0.07). In the figures the communities with the highest concentrations are noted, mainly located in the Mühlviertel. But some communities have very high indoor radon activity concentrations, but very low radon concentrations in drinking waters (e.g. Gmunden, Gosau in southern Upper Austria). So in general higher radon activity concentrations in water were only detected in the Bohemian massif, but higher indoor radon concentrations also occur in other geological regions (e.g. ice age deposits). In Figure 17.8 the correlation between the medium indoor radon and radon in drinking water is illustrated in the Bohemian massif only. But also no clear correlation was detected in this geological region (r^2=0.14).

So **no clear correlation between indoor radon and radon in drinking water in the communities could be detected**. It seems that the dependencies of both indoor radon concentration and radon in drinking water are too complex to get easy correlations between them. Deflorin (2004) surveyed whether radon measurements in ground and well waters could replace indoor radon measurements for the purpose of classification of radon areas in Graubünden. He detected a clearer correlation between indoor radon and drinking water radon, but he also recommend not to replace the method because of high uncertainties and error probability. The geological influences to radon in soil gas (Gruber et al., 2008) and radon in drinking water are too complex to combine them and make predictions, and for radon indoor assessments many other influences have to be taken into account. In a planned future project about radon in soil gas in different geological zones in Austria also radon in ground water will be analyzed and the correlation to radon in soil gas surveyed (Gruber et al., 2008).

Nevertheless the **radon concentration in drinking water influences the indoor radon concentration** by release from tap water (e.g. washing, showering, operating washing machine or dishwasher). Rühle (1997) reports a transfer factor of $5 \cdot 10^{-5}$ to $1 \cdot 10^{-4}$ for the transfer of radon from the water into the indoor air in the living quarters. A radon concentration of 50 Bq/l enhances the indoor radon concentration of maximum 5 Bq/m^3. Higher short-term concentrations can occur in the bathroom while showering.

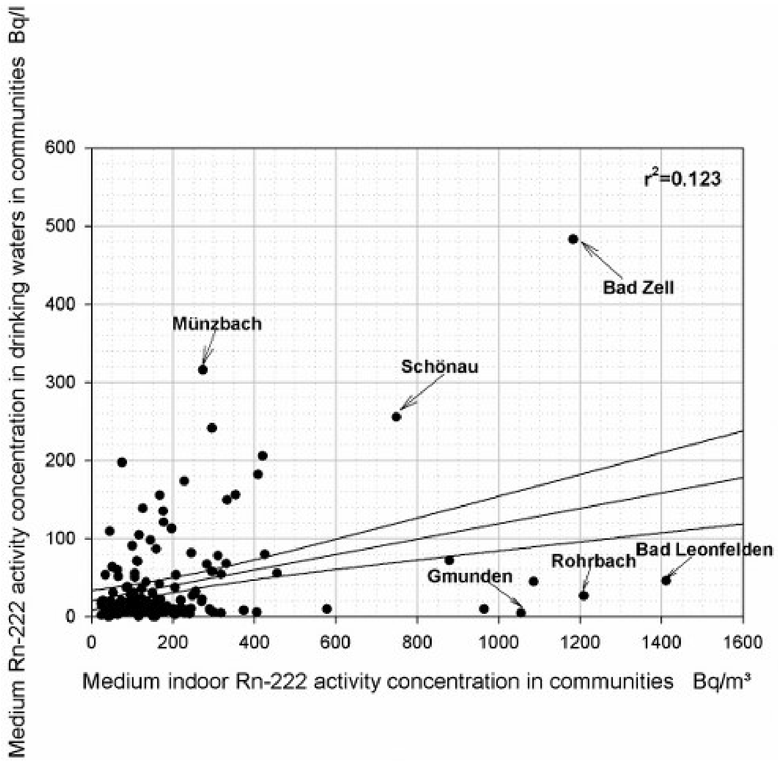

Figure 17.6: *Correlation between the medium radon indoor and drinking water radon concentration in Upper Austrian communities*

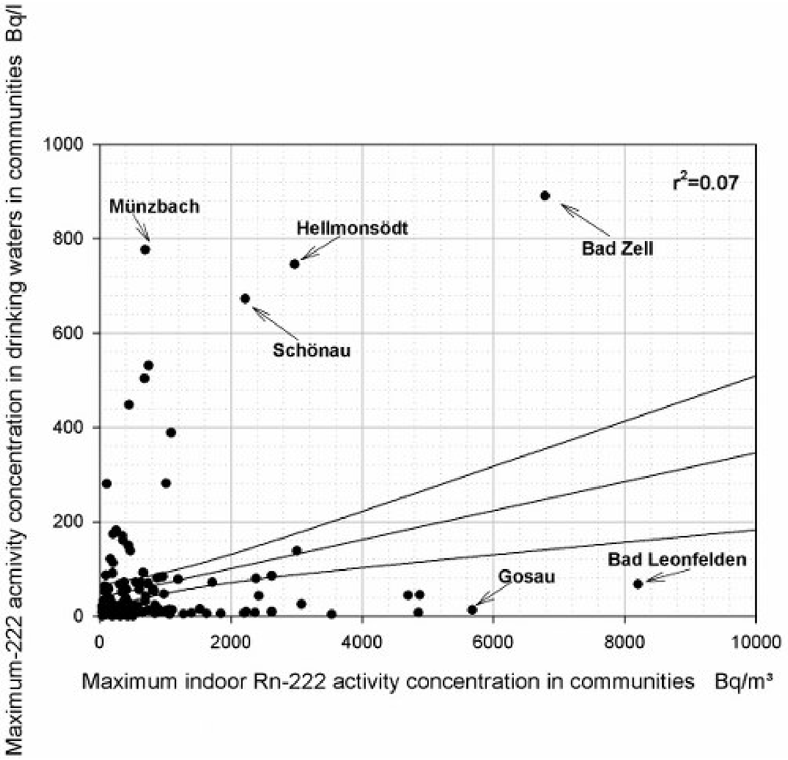

Figure 17.7: *Correlation between the maximum radon indoor and drinking water radon concentration in Upper Austrian communities*

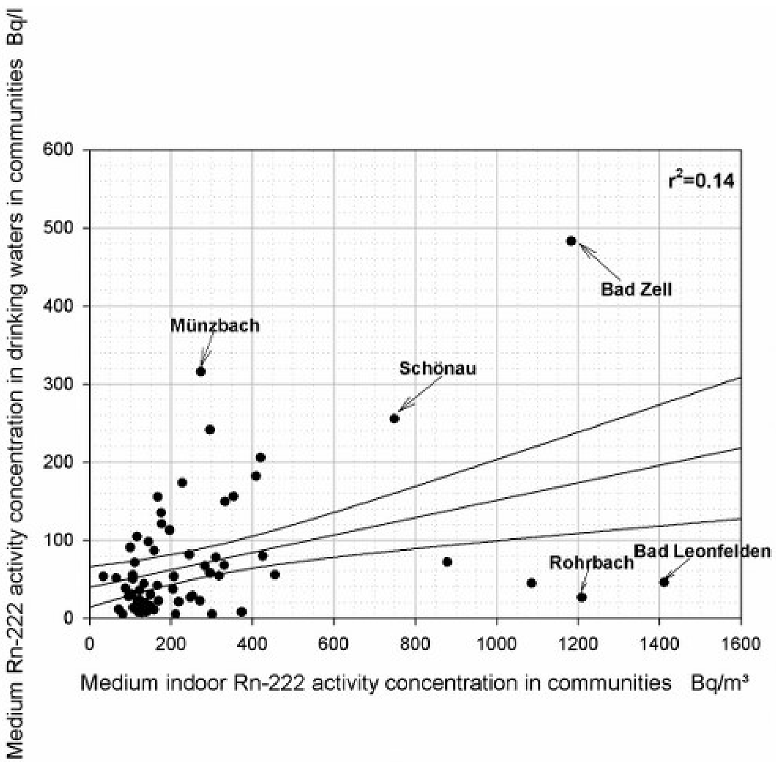

Figure 17.8: *Correlation between the medium indoor radon and drinking water radon concentration in the Bohemian massif*

17.1.9 Dose estimations and recommendation of radionuclide concentrations for drinking water assessment

Dose assessments for radioactivity in drinking water for the Upper Austrian population were carried out in the framework of this thesis. The determination of the parameter **total indicative dose** (TID) appointed in the Austrian drinking water directive (Republik Österreich, 2001) is defined in the Austrian standard ÖNORM S 5251 (Austrian Standards Institute, 2005b). According to this only ^{226}Ra and ^{228}Ra are taken into account and the assessment is carried out for adults. No calculated total dose is above the total indicative dose of 0.1 mSv/a. Taking into account ^{238}U also no total dose above the total indicative dose of 0.1 mSv/a is detected. Taking into account ^{210}Po and ^{210}Pb a clearly higher dose contribution is detected, which demonstrates that these nuclides should not be neglected within regulations and guidelines. For interest also dose assessments for **other age groups** were carried out, which also yields doses above 0.1 mSv/a only in the case of ^{210}Po and ^{210}Pb.

Dose assessments with an alternative model taking into account continuously intake and excreting of radionuclides through drinking water yield much lower total annual doses. These assessments were done based on the dose estimation by Bronzovic et al. (2006) with an daily excretion of radionuclides out of the body described by the $m(t)$ value (IAEA, 2004). According to this a very rapid decline of radionuclides remaining in the body within the first days after intake is reported. With a daily water intake and this countinous excretion a constant activity is reached in the body after a few days. Out of this an **effective dose** is estimated for an entire adult reference body and also for babies for **^{226}Ra, ^{228}Ra, ^{210}Po and ^{210}Pb**. The effective dose results estimated with this method are **at least 2 magnitudes lower for adults than** the recommended **parametric value of 0.1 mSv/a** with the same activity concentration in drinking water, and also at least one magnitude lower for babies. Detailed research will be done based on this dose estimation method in a future project with differentiation for different organs and body modeling.

This survey should illustrate that **risk for the population** caused by natural radioactivity in drinking waters **only occurs in individual cases** and for **most of the Upper Austrian drinking waters no hazards for the population exist**. Nevertheless drinking water should be controlled and surveyed regarding radioactivity, but drinking waters with a little enhanced radioactivity concentrations according to different standards, recommendations and guidelines should not be set disabled for drinking water purposes without further surveys and dose assessments. For this purpose within this thesis guideline values were developed to simplify and standardize experts' activities in drinking water affairs.

Because of **no existing limit values or action values for radioactivity in drinking water** in the regulations, the existing parameter values (which are set very low, because they are meant only for monitoring purposes) often are wrongly used as limit values in the framework of expertises. This sometimes yields prohibition of usage of drinking water, which is totally harmless according to radiation protection point of view. So, for more clarity for experts, operators of water supplies and consumers as a first step an **intervention level of a total dose of 1 mSv/a is recommended**. When this intervention level is exceeded remedial measures in an appropriate time frame should be regulated. The easiest remedial measures are non-use of this drinking water or mixing with waters with clearly lower concentration of radionuclides.

Radon and radon progenies are not taken into account in the total indicative dose. For clarity for experts' works **intervention levels** also for these radionuclides are recommended. For ^{222}Rn an intervention level of 1000 Bq/l is considered to be sufficient, for ^{210}Po and ^{210}Pb intervention levels of 1 Bq/l and 2 Bq/l respectively are recommended. **It is recommended, that in drinking waters, in which a radon activity concentration of above 100 Bq/l is expected, also ^{210}Po and ^{210}Pb should be analyzed**. For drinking waters with low radon activity concentrations but indications of higher ^{210}Po and ^{210}Pb exists, these nuclides should be surveyed too, because of their above discussed high dose contribution.

In Table 17.2 the **recommended radionuclide concentrations for assessment of drinking water** are listed. In this table the directives, guidelines and recommendation of different authorities, which are used as basis for monitoring levels are listed too. **The intervention levels are always set ten times higher**.

These recommendations were worked out in the framework of the drinking water project and this thesis together with the Austrian Ministry of Health, Family and Youth (BMGFJ). The recommendations are already adopted and published in the **"Austrian food and drinking water codex"**, *Codex alimentarius Austriacus*, Chapter B, drinking water, Pkt. 8.4.3 (BMGFJ, 2008b) – an intervention level of a total indicative dose of 1 mSv/a and the recommended radon and radon progeny intervention levels. The "Austrian food and drinking water codex" is not a regulation, but an important implement which proclaims terms and definitions, technical names and research methods and in Austria is edited by the "Ministry of Health, Family and Youth". The codex alimentarius is a collective arrangement of the World Health Organisation (WHO) and the Food and Agriculture Organisation (FAO) of the United Nations (UN) (BMGFJ, 2008a).

The recommendation should also yield an **extension of the Austrian drinking water regulations**.

Table 17.2: *Recommended radionuclide concentrations for drinking water assessment*

	Activity concentration (Bq/l)		
Nuclide	**Monitoring**	**Intervention**	**Monitoring values based on**
$^{226}Ra^{1}$	0.5	5.0	EC 2005, Draft
$^{228}Ra^{1}$	0.2	2.0	EC 2005, Draft
$^{3}H^{2}$	100	1000	EC, Council Directive 98/83/EC; Austrian drinking water regulation BGBl. II 304/2001
$^{222}Rn^{2}$	100	1000	EC, Comission Recommendation 2001/982/Euratom
^{210}Pb	0.2	2.0	EC, Comission Recommendation 2001/982/Euratom
^{210}Po	0.1	1.0	EC, Comission Recommendation 2001/982/Euratom
$^{238}U^{3}$	0.37^{4}	3.7	WHO Guidelines for drinking water quality, 1998

[1] *Listed values of activity concentration are maxima, when only the considered radionuclide is available. If other radionuclides are available, the cumulative dose according to ÖNORM S525:20051 has to be calculated by using formula (1).*

[2] *Listed values are broad below dose relevance.*

[3] *The uranium limit is based on chemo toxic effect; the WHO guidelines for drinking water quality 2004 recommend a value of 15 µg/l natural uranium (accords to 0.19 Bq/l ^{238}U)*

[4] *According to a uranium concentration (natural isotopic alloy) of 30 µg/l*

17.2 Prospects

The results and measurements of this thesis provide a **good basis data net** and afforded many surveys and discussions on different questions and topics. Nevertheless it is reasonable and of great interest to **update these data** to **verify conclusions** and to **provide generalizations** and further insights. So, further measurements of natural radioactivity in drinking should be carried out in the future. The **measurements and sampling** should be **standardized** for radiation protections reasons and also for comparable results in official proceedings and surveys. Therefore it is reasonable to work out a **guideline or standard in Austria** and implement it.

Additionally – as it was mentioned sometimes above – it is reasonable and recommended to **extend the existing drinking water directives or standards** for other radionuclides, especially for ^{210}Po because of its major dose contribution. Besides the effective drinking water directives or standards should also include radionuclide concentration values for drinking water assessments as it was discussed above.

Besides, drinking water analyses, surveys, measurements and analyses of **uranium** and **other natural radionuclides contents** in different rocks and soils with impact on ground and drinking waters should be carried out. As discussed in Chapter 15 drinking water may be essentially influenced by geology and radionuclide contents in the surrounding rocks and soil. Knowledge of this may afford predictions of radionuclide contents in drinking waters and may provide identification of correlations and circumstances.

As discussed above, further **dose estimations with the alternative model** will be carried out in a future poject, taking into account not only the whole body but a radionuclide transfer in different organs and age dependent body modeling.

As a first available step of continuation of the thesis and drinking water project, the data net in Upper Austria will be updated by the **routine measuring program** worked out in the framework of this thesis. On-site radon measurements by the Triathler will be carried out within the framework of the existing water program – **"On the way for our drinking water"** in Upper Austria. In this project more than 1000 water samples of private wells and small water supplies are analyzed every year for different chemical, physical and bacteriologic parameters on-site in a measuring bus (Land Oberösterreich, 2008). In the future also **radon** will be analyzed by Triathler within this project and additionally samples will be collected at selected locations for analyses of **other natural radionuclides** in the laboratory. For georeferencing coordinates are recorded and details of the sampling location are stored. The radon data will be processed and related with the other analyzed parameters and other topics like geology. So the **data net of natural radioactivity in drinking water in Upper Austria will be updated** and tightened enormously and unsettled questions of this thesis may be answered and theories verified.

The measurements, analyses, developments, results and discussion of this thesis and the drinking water project in Upper Austria should be used as basis for further studies in other regions of Austria and should act as an example. As written in the beginning – **drinking water is one of the most important natural resource for human life** and therefore it should be surveyed also for radioactivity for radiation protection of the population as it is required in the drinking water directive in Austria (Republik Österreich, 2001) and also in the European Drinking Water Directive 98/83/EC (European Commission, 1998).

List of Tables

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Bibliography

Ahrens, L. 1965. The Distribution of the Elements in Our Planet (New York: McGraw Hill)

Åkerblom, G. 1999. Radon Legislation and Natural Guidelines (Swedish Radiation Protection Institute, Department of Environmental Monitoring and Dosimetry), SSI rapport 99:18

Åkerblom, G. & Lindgren, J. 1997. Mapping of Groundwater Radon Potential. *European Geologist* pp. 13–22

Alabdula'aly, A. 1999. Occurrence of Radon in the Central Region Groundwater of Saudi Arabia. *Journal of Environmental Radioactivity* 44:85–95

Almeida, R., Lauria, D., Ferreira, A., & Sracek, O. 2004. Groundwater Radon, Radium and Uranium Concentrations in Regiao dos Lagos, Rio de Janeiro State, Brazil. *Journal of Environmental Radioactivity* 73:323–334

Amt der Oberösterreichischen Landesregierung. 1997. Radon in Oberösterreich, Teilergebnisse aus dem Österreichischen Nationalen Radonprojekt (ÖNRAP) (Linz), Broschüre

Amt der Oberösterreichischen Landesregierung. 2007. Hausbrunnen. `http://www.ooewasser.at/sitex/index.php/page.12/`, 18.6.2007

Amt der Oberösterreichischen Landesregierung. 2008. Grundwassergüte in Oberösterreich 1992-2007, ed. Direktion Umwelt und Wasserwirtschaft, Abteilung Grund- und Trinkwasserwirtschaft (Linz)

Annanmäki, M. 1999. Treatment Techniques for Removing Natural Radionuclides from Drinking Water (TENAWA), Final Report, Euratom

Aurand, K. & Gans, I. 1991. Radioaktive Stoffe und die Trinkwasserverordnung, in *Die Trinkwasserverordnung*, eds. K. Aurand et al. (Berlin: Erich Schmidt Verlag)

Aurand, K. & Rühle, H. 2003. Radioaktive Stoffe und die Trinkwasserverordnung, in *Die Trinkwasserverordnung – Einführung und Erläuterungen für Wasserversorgungsunternehmen und Überwachnungsbehörden*, eds. A. Grohmann, U. Hässelbarth, & W. Schwertfeger, 4th edition (Berlin)

Austrian Standards Institute. 1986. ÖNORM M 6250: Public Drinking Water Supply; Requirements for Drinking (Potable) Water Quality (Vienna)

Austrian Standards Institute. 2002. ÖNORM S 5250-1: Counting Statistics in Radioactivity Measurements – Part 1: Uncertainties of Measurements, Decision Threshold and Detection Limit (Vienna)

Austrian Standards Institute. 2003. ÖNORM S 5280-2: Radon – Part 2: Technical Precautionary Measures in the Case of Buildings (Vienna)

Austrian Standards Institute. 2005a. ÖNORM S 5250-2: Counting Statistics in Radioactivity Measurements – Part 2: Spectrometric Measurements (Vienna)

Austrian Standards Institute. 2005b. ÖNORM S 5251: Determination and Evaluation of the Total Dose Due to Radionuclides in Drinking Water (Vienna)

BEIR VI. 1999. Health Effects of Exposure to Radon, ed. Committee on Health Risks of Exposure to Radon (Washington, D.C.: National Academy Press)

Bliefert, C. 1994. Umweltchemie (Weinheim, Deutschland: VCH Verlagsgesellschaft mbH)

BMGFJ. 2007. Österreichischer Trinkwasserbericht 2002-2004, ed. Bundesministerium für Gesundheit, Familie und Jugend, Sektion IV Verbrauchergesundheit und Gesundheitsprävention (Vienna), BMGJJ-75310/0017-IV/B/7/2007

BMGFJ. 2008a. FAO/WHO – Codex Alimentarius. `http://www.bmgfj.gv.at/cms/site/standard.html?channel=CH0943&doc=CMS1056641057751`, 27.11.2008

BMGFJ. 2008b. Österreichisches Lebensmittelbuch, 4th edition, ed. Bundesministerium für Gesundheit, Familie und Jugend

Böhm, C. 2002. Radon im Wasser – Überblick für den Kanton Graubünden. *Jahresbericht Naturforschende Gesellschaft Graubünden* 111:49–79

Bradl, H., ed. 2005. Interfaces Science and Technology, Vol. 6, Heavy Metals in the Environment (London: Elsevier)

Bröcker, B. 1993. Dtv-Atlas zur Atomphysik, 5th edition (München: Deutscher Taschenbuchverlag GmbH & Co. KG)

Bronzovic, M. & Marovic, G. 2004. Water Intake and Age-Dependent Dose Assessment, in IRPA11 Proceedings, Madrid, pp. 23–28

Bronzovic, M., Marovic, G., & Vrtar, M. 2006. Public Exposure to Ra-226 in Drinking Water. *Arh Hig Rada Toksikol* 57:39–44

Bundesamt für Strahlenschutz. 2008. Natürliche Radionuklide in Trinkwässern. `www.bfs.de/ion/nahrungsmittel/trinkwasser.html`, 13.11.2008

Bundesrepublik Deutschland. 2001. Verordnung über die Qualität von Wasser für den menschlichen Gebrauch (Trinkwasserverordnung TrinkwV2001), vom 21. Mai 2001 (BGBl. I S. 959), geändert durch Artikel 363 der Verordnung vom 31. Oktober 2006 (BGBl. I S. 2407) (Bonn)

Bünger, T. 1997. Untersuchungen zum Radongehalt des Trinkwassers in Berliner Wasserwerken, in *Radon und Trinkwasser. Schriftenreihe des Vereins für Wasser-, Boden- und Lufthygiene* (Berlin), pp. 29–37

Bünger, T. & Rühle, H. 1993. Natürliche Stoffe im Trinkwasser ausgewählter Gebiete in Sachsen und Thüringen, in *Umweltradioaktivität, Radioökologie, Strahlenwirkungen*, Vol. I (Köln: Verlag TÜV Rheinland), pp. 85–97

Bünger, T. & Rühle, H. 1997. Determination and Evaluation of Natural Radioactive Nuclides in Drinking Water From Selected Water Supplies of the Erzgebirge and the Vogtland Regions. *Kerntechnik* 62

Campos, M., Penna-Franca, E., Labao, N., Trinidade, H., & Sanchett, J. 1986. Migration of Radium From Ore Deposit of the Moro de Ferro, Pocos de Caldas, Brazil. *Journal of Environmental Radioactivity* 3:145–161

Clavensjö, B. & Åkerblom, G. 1994. The Radon Book – Measurements Against Radon, ed. The Swedish Council for Building Research (Stockholm)

Cothern, C. & Smith, jr, J. 1987. Environmental Science Research, Vol. 35, Environmental Radon (New York: Plenum Publishing Corporation)

Darby, S., Hill, D., Auvinen, A., et al. 2005. Radon in Homes and Risk of Lung Cancer: Collaborative Analysis of Individual Date from 13 European Case-Control Studies. *BMJ 2005* pp. 223–330

Deflorin, O. 2004. Natürliche Radionuklide in Grundwässern des Kantons Graubünden, Dissertation, Universität Neuchâtel (Switzerland)

Diehl, J. 2003. Radioaktivität in Lebensmitteln (WILEY-VCH GmbH & Co)

Ditto, M. 2005. Radioaktivitätsmessungen in Österreich 2003 und 2004. Daten und Bewertung, ed. Bundesministerium für Gesundheit und Frauen, Sektion III und Bundesministerium für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft, Sektion V (Wien)

Ditto, M., Fimml, W., Karg, V., Korner, M., & Weisz, J. 1999. Radon-222 im Grundwasser. Ein österreichweiter Überblick, ed. Bundesanstalt für Lebensmitteluntersuchung und -Forschung Wien, Abteilung Radiochemie und Strahlenschutz (Wien)

Eidgenössisches Department des Inneren (EDI). 1995. Verordnung des EDI über Fremd- und Inhaltstoffe in Lebensmitteln (Fremd- und Inhaltstoffeverordnung FIV) (Bern), 817.021.23 vom 26. Juni 1995, Stand 1. April 2008

Eisenbud, M. & Gesell, T. 1997. Environmental Radioactivity. From Natural, Industrial, and Military Sources, 4th edition (Academic Press)

EPA. 1991. National Primary Drinking Water Regulations, Radionuclides; Proposed Rule, ed. Environmental Protection Agency (Washington, D.C.), Fed. Regis. 56 (No. 138), 33050-33123

EPA. 1993. Radon – A Physician´s Guide: The Health Threat With A Simple Solution. `http://www.epa.gov/radon/pubs/physic.html`, 15.01.2008

EPA. 2000. National Primary Drinking Water Regulations; Radionuclides; Final Rule. Part II. 40 CFR Parts 9, 141 and 142, ed. U.S. Environmental Protection Agency (Washington D.C.)

EPA. 2009. Radioactive Equilibrium. `http://www.epa.gov/rpdweb00/understand/equilibrium.html`, 08.02.2009

Eppendorf. unknown. Eppendorf Multipette 4780, Bedienungsanleitung

Erlandsson, B., Jakobsson, B., & Jönsson, G. 2001. Studies of the Radon Concentration in Drinking Water from the Horst Söderasen in Southern Sweden. *Journal of Environmental Radioactivity* 53:145–154

ETH Zürich. 2007. Nachweis von β-Strahlung mittels Flüssigszintillationsmessung. Radiochemisches Praktikum, P15. `http://www.analytik.ethz.ch/praktika/analytisch/radiochemie/unterlagen/p15.pdf`, 15.11.2007

European Commission. 1990. Commission Recommendation 90/143/Euratom of 21 February 1990 on the Protection of the Public Against Indoor Exposure to Radon, L080/26, 27.03.1990

European Commission. 1996. Council Directive 96/29/Euratom of 13 May 1996 Laying Down Basic Safety Standards for the Protection of the Health of Workers and the General Public Against the Dangers Arising from Ionizing Radiation, L159/1, 29.06.1996

European Commission. 1998. Council Directive 98/83/EC of 3 November 1998 on the Quality of Water Intended for Human Consumption, L330/32, 05.12.1998

European Commission. 2001. Commission Recommendation 2001/982/Euratom of 20 December 2001 on the Protection of the Public Against Exposure to Radon in Drinking Water Supplies, L344/85, 28.12.2001

European Commission. 2005. Commission Directive of Defining Requirements for the Parameters for Radioactivity for Monitoring the Quality of Water for the Council Directive 98/83 of 3 November 1998 on the Quality of Water Intended for Human Consumption, Draft, V 3.0, 29.11.2005

Feigl-Heihs, M. 1998. Bestimmung von Pb-210 und Po-210 in Trinkwasserproben, Diplomarbeit, Universität Wien (Wien)

Fettweis, P. & Schwenn, H. 1998. Semiconductor Detectors, in *Handbook of Radioactivity Analysis*, ed. M. L'Annunziata (Academic Press)

Firestone, R. & Ekström, L. 2008. WWW Table of Radioacitve Isotopes, LBNL Isotopes Project – LUNDS Universitet, Database Version 2/28/99. `http://ie.lbl.gov/toi/index.asp`, 30.11.2008

Forte, M., Rusconi, R., Cazzaniga, M., & Sgorbati, G. 2007. The Measurement of Radioactivity in Italian Drinking Waters. *Microchemical Journal* 85/1:98–102

Frenzel, E. 1999. Die LSC-Messung von Radon (Rn-222) in Wasser mit dem Triathler. Überarbeitete Applikationsschrift, Hidex/Frenzel Consulting & Instruments

Frenzel, E. 2004. Anleitung für den Triathler. Tips & Tricks: No.7, Berechnung der Rn-222 in Wasser Konzentration, Frenzel Consulting & Instruments, Benutzerhandbuch

Frenzel, E. 2005. Dokumentation Triathler TM, FCI, Frenzel Consulting & Instruments, Benutzerhandbuch

Friedmann, H. 1997. Potential Radon Risk from Drinking Water in Austria, EU-Projekt "Treatment Techniques for Removing Natural Radionuclides from Drinking Water" (TENAWA), gefördert von der Europäischen Kommission Vertragsnummer F14PCT960054 (Institut für Radiumforschung und Kernphysik, Universität Wien)

Friedmann, H. 1999. Prognose von Radongehalten im Wasser für potentielle Trinkwasserquellgebiete, ed. Institut für Radiumforschung und Kernphysik, Universität Wien, unpublished

Friedmann, H. 2006. Radonbelastung in Österreich. Endbericht, ed. Bundesministerium für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft

Friedmann, H. 2008. Radon in Wasser. `http://homepage.univie.ac.at/harry.friedmann/Radon/welcome.htm`, 12.11.2008

Fuchs, W. & Thiele, O. 1987. Geologische Karte der Republik Österreich 1:50000, Erläuterungen zu Blatt 34 Perg (Wien: Geologische Bundesanstalt)

Gans, I. 1992. Natürliche radioaktive Stoffe im Trinkwasser. *Bundesgesundheitsblatt 6*. pp. 300–301

Gegner, M. 2002. Urangehalt und Uranisotopenverhältnisse in österreichischen Trinkwasserproben, Diplomarbeit, Universität Wien (Wien)

Gellermann, R. & Stolz, W. 1997. Uran in Wässern – Untersuchungen in ostdeutschen Flüssen und Grundwässern. *Zeitschrift für Umweltchemie und Ökotoxikologie* 9 (2):87–92

Geologische Bundesanstalt. 2006. Geologische Karte von Oberösterreich 1:200.000 (Wien: Geologische Bundesanstalt)

Gerdes, A. 1997. Geochemische und thermische Modelle zur Frage der spätorogenen Granitgenese am Beispiel des Südböhmischen Batholits: Basaltisches Underplating oder Krustenstapelung?, Dissertation, Universität Göttingen

Gerzabek, M. 2005. Radionuklide in terrestrischen Ökosystemen, Universität für Bodenkultur Wien, Institut für Bodenforschung, Skriptum zur Vorlesung

Godoy, J. & Godoy, M. 2006. Natural Radioactivity in Brazilian Groundwater. *Journal of Environmental Radioactivity* 85:71–83

Gruber, V., Baumgartner, A., Seidel, C., & Maringer, F. 2008. Radon Risk in Alpine Regions in Austria: Risk Assessment as a Settlement Planning Strategy. *Radiation Protection Dosimetry* 130/1:88–91

Gruber, V., Maringer, F., & Landstetter, C. 2009. Radon and Other Natural Radionuclides in Drinking Water in Austria: Measurement and Assessment. *Applied Radiation and Isotopes*, doi:10.1016/j.apradiso.2009.01.056, in press

Gruber, V. et al. 2006. Strahlenexposition durch Trinkwasser in Oberösterreich – 2004 bis 2006. Teilprojekt Bevölkerungsexposition. Endbericht, ed. Land Oberösterreich, Abteilung Umwelt- und Anlagentechnik

Gruber, V. et al. 2007. Strahlenexposition durch Trinkwasser in Oberösterreich – 2004 bis 2006. Teilprojekt Bevölkerungsexposition. Zusammenfassender Endbericht, ed. Land Oberösterreich, Abteilung Umwelt- und Anlagentechnik

Haberer, K. 1989. Umweltradioaktivität und Trinkwasserversorgung (München: Oldenbourg Verlag GmbH)

Hakonson-Hayes, A., Fresquez, P., & Whicker, F. 2002. Assessing Potential Risks From Exposure to Natural Uranium in Well Water. *Journal of Environmental Radioactivity* 59:29–40

Herranz, M., Abelairas, A., & Legarda, F. 1997. Uranium Contents and Associated Effective Doses in Drinking Water from Biscay (Spain). *Applied Radiation and Isotopes* 48(6):857–861

Hidex OY. 2005. Triathler Mulitlabel Tester. Owner's Handbook, Hidex OY, http://www.hidex.com

Hirsch, S. & Ruzicka, W. 2002. Heilige Quellen in Oberösterreich (Linz: Freya Verlag)

IAEA. 1984. The Behaviour of Radium in Waterways and Aquifers. Final Report of the Co-ordinated research programme in source, distribution, movement and deposition of radium in inland waterways and aquifers sponsored by the IAEA from 1976 to 1980, ed. International Atomic Energy Agency, IAEA TECDOC Series No. 301 (Vienna)

IAEA. 1988. Geochemical Exploration for Uranium, ed. International Atomic Energy Agency, Technical Reports Series No. 284 (Vienna)

IAEA. 1990. The Environmental Behaviour of Radium, ed. Internation Atomic Energy Agency, Technical Reports Series No. 310, Vol. 1 & 2 (Vienna)

IAEA. 1996. International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources, ed. International Atomic Energy Agency, Safety Reports Series, No. 115 (Vienna)

IAEA. 2004. Methods for Assessing Occupational Radiation Doses Due to Intakes of Radionuclides, ed. International Atomic Energy Agency, Safety Reports Series, No. 37 (Vienna)

ICRP. 1991. ICRP Publication 60: 1990 Recommendations of the International Commission on Radiological Protection, ed. International Commission of Radiological Protection, Annals of the ICRP, Vol 21/1–3 (Elsevier)

ICRP. 1993. ICRP Publication 67: Age dependent doses to members of the public from intake of radionuclides: Part 2, Ingestion dose coefficients, ed. International Commission of Radiological Protection, Annals of the ICRP, Vol. 23/3–4 (Elsevier)

ICRP. 1996. ICRP Publication 72: Ann. ICRP 26: 15-89, 1996, ed. International Commission of Radiological Protection, Annals of the ICRP, Vol. 26 (Elsevier)

ICRP. 2008. ICRP Publication 103: Recommendations of the ICRP, ed. International Commission of Radiological Protection, Annals of the ICRP, Vol. 32/2–4 (Elsevier)

Irlweck, K., Feigl-Heihs, M., Liepold, K., & Wallner, G. 1997. Die Bestimmung von Blei-210 und Polonium-210 sowie Radium-226 in Trinkwasserproben aus verschiedenen Gebieten in Österreich. Endbericht, Werkvertrag Zl.353.019/0-III/9/97, Bundeskanzleramt Sektion VI

ISO. 1995. Guide to the Expression of Uncertainty in Measurement (GUM), ed. International Organization for Standardization (Genf)

Ivanovich, M. & Harmon, R., eds. 1982. Uranium Series Disequilibrium: Applications to Environmental Problems (Oxford University Press)

Jia, G. & Torri, G. 2007. Estimation of Radiation Doses to Members of the Public in Italy From Intakes of Some Important Naturally Occuring Radionuclides (U-238, U-234, U-235, Ra-226, Ra-228, Ra-224, Po-210) in Drinking Water. *Applied Radiation and Isotopes* 65:849–857

Karpinska, M., Wolkowicz, S., Mnich, Z., et al. 2002. Comparative Studies of Health Hazard From Radon (Rn-222) in Two Selected Lithologic Formations in the Suwalki Region (in Poland). *Journal of Environmental Radioactivity* 61:149–158

Katzlberger, C., Wallner, G., & Irlweck, K. 2001. Determination of Pb-210, Bi-210, Po-210 in Drinking Water. *Journal of Radioanalytical and Nuclear Chemistry* 249(1):191–196

Knoll, G. 2000. Radiation Detection and Measurement, 3rd edition (New York: John Wiley & Sons)

Koelzer, W. 2001. Lexikon zur Kernenergie, ed. Forschungszentrum Karlsruhe GmbH (Karlsruhe)

Korea Atomic Energy Research Institute. 2009. Table of Nuclides. `http://atom.kaeri.re.kr/`, 21.1.2009

Kozlowska, B., Hetman, A., & Zipper, W. 1999. Determination of Rn-222 in Natural Water Samples From Health Resorts in the Sudety Mountains by the Liquid Scintillation Technique. *Applied Radiation and Isotopes* 51:475–480

Krakhofer, A. 2002. Radon in Grund- und Quellwasser im nördlich der Donau gelegenen Gebiet Österreichs, Diplomarbeit, Universität für Bodenkultur (Wien)

Krieger, H. 2002. Strahlenphysik, Dosimetrie und Strahlenschutz. Vol. 1 – Grundlagen, 5th edition (Wiesbaden: B. G. Teubner Verlag)

Krieger, H. 2004. Grundlagen der Strahlungsphysik und des Strahlenschutzes (Wiesbaden: B. G. Teubner Verlag)

Land Oberösterreich. 1961. Gesetz über natürliche Heilvorkommen und Kurorte (Oö. Heilvorkommen- und Kurortegesetz – Oö. HKG), Landesgesetzblatt LGBl. Nr. 47/1961. `http://www.ris2.bka.gv.at/GeltendeFassung.wxe?QueryID=LrOO&Gesetzesnummer=10000048&ShowPrintPreview=True`, 12.8.2008

Land Oberösterreich. 2008. Für unser Trinkwasser unterwegs – Informationen zum Laborbus. `http://www.land-oberoesterreich.gv.at/cps/rde/xchg/SID-2C1526E3-035BB70F/ooe/hs.xsl/15077_DEU_HTML.htm`, 18.12.2008

Land Salzburg. 2008. Grundwasserhärte, Wasserhärte, Härtegrade. `http://www.salzburg.gv.at/themen/nuw/wassererangelegenheiten/gewaesserschutz/guete/gw/haerte.htm`, 08.04.2008

Landessanitätsdirektion, Land Oberösterreich. 2008. Information agressive Wässer. `http://www.land-oberoesterreich.gv.at/cps/rde/xbcr/SID-3DCFCFC3-7DCE7576/ooe/San_Infoblatt_aggressive_Waesser_NEU.pdf`, 08.04.2008

Landstetter, C. & Katzlberger, C. 2005. Rapid Method for Determining Natural Radionuclides in Drinking Water, in Proceedings Book of the 2005 International Liquid Scintillation Conference, Radiocarbon, Katowice, pp. 181–190

Lebensministerium. 2008a. Grundwasser. `http://wasser.lebensministerium.at/article/articleview/60324/1/14151/`, 08.04.2008

Lebensministerium. 2008b. Trinkwasser und Wasserversorgung. `http://wasser.lebensministerium.at/article/articleview/60323/1/14151`, 08.04.2008

Lebensministerium. 2008c. Wasserqualität. `http://www.wassernet.at/article/archive/5727`, 27.05.2008

Lehmann, K., Reichelt, A., & Eder, E. 1997. Strahlenexposition durch erhöhte Radonaktivitätskonzentration im Trinkwasser bei Verzehr und Verbrauch, in *Radon und Trinkwasser. Schriftenreihe des Vereins für Wasser-, Boden- und Lufthygiene* (Berlin), pp. 81–89

Leichtfried, W. 2004. Hydrogeologie und Radonpotential in Oberösterreich, persönliche Mitteilung des Autors, 13.07.2004

Limson-Zamora, M., Tracy, B., Zielinski, J., Meyerhof, D., & Moss, M. 1998. Chronic Ingestion of Uranium in Drinking Water: A Study of Kidney Bioeffects in Humans. *Toxicilogical Sciences* 43:68–77

Lopes, I., Madruga, M., Ferrador, G., et al. 2006. Monitoring of Gross Alpha, Gross Beta and Tritium Activities in Portuguese Drinking Waters, in Proceedings of Second European IRPA Congress on Radiation Protection, Paris

Magill, J. & Galy, J. 2005. Radioactivity, Radionuclides, Radiation (Springer-Verlag)

Marsden, E. & Collins, M. 1963. Particle Activity and Free Radicals From Tobacco. *Nature* 198:962–964

McKone, T., Brand, K., & Shan, C. 1997. Environmental Health-Risk Assessment for Tritium Releases at the National Tritium Labeling Facility at Lawrence Berkeley National Laboratory, University of California, Berkeley, California. `http://www.lbl.gov/ehs/esg/tritium/tritium`, 16.11.2007

Möbius-Ramamonjisa, T., Möbius, S., & Frenzel, E. 1998. Becquerel's Stein. Kernstrahlungsmesstechnik im Experiment. *Strahlenschutzpraxis* 4

Mück, K. 2001. Radioaktivität und Strahlung in unserer Umwelt. Die Strahlenexposition des Österreichers, ed. B. G. Austrian Research Center Seibersdorf, ARC-G-0004

Nazaroff, W., Moed, B., & Sextro, R. 1988. Soil as a Source of Indoor Radon: Generation, Migration, and Entry, in *Radon and its Decay Products in Indoor Air*, eds. W. Nazaroff & A. Nero, jr (Canada: John Willey & Sons)

Nero, jr, A. 1988. Radon and its Decay Products in Indoor Air: An Overview, in *Radon and its Decay Products in Indoor Air*, eds. W. Nazaroff & A. Nero, jr (Canada: John Willey & Sons)

NHMRC. 2001. Australian Drinking Water Guidelines 1996 – Update 2001, National water quality management strategy paper No. 6, ed. National Health and Medical Research Council/ Agricultural and Resource Managment Council of Australia and New Zealand (ARMCANZ) (Canberra: Australian Government Publishing Service)

Nonner, J. 2003. Introduction to Hydrogeology (Lisse: Swets & Zeitlinger B.V.)

NRC. 1999. Risk Assessment of Radon in Drinking Water, ed. NRCNational Research Council, Commission on Life Science, Board on Radiation Effects Research; Committee on Risk Assessment of Exposure To Radon in Drinking Water (Washington, D.C.: National Academy Press)

Nuccetelli, C. & Risica, S. 2006. Radionuclides in Drinking Water: The Approach of the European Union, the World Health Organization and the US Environmental Protection Agency, in Proceedings of Second European IRPA Congress on Radiation Protection, Paris

Ontario Ministry of Environment and Energy. 2003. Ontario Drinking-Water Quality Standards Regulation, O.Reg 169/03

OÖ Wasser Genossenschaftsverband. 2008. Wasseraufbereitung. `http://www.ooewasser.at/sitex/index.php/page.129/`, 08.04.2008

Österreichische Vereinigung für das Gas- und Wasserfach (ÖVGW). 2008. Wasserwerk. `http://www.wasserwerk.at/`, 08.04.2008

Otwoma, D. & Mustapha, A. 1998. Measurements of Rn-222 concentration in Kenyan Groundwater. *Health Physics* 74(1):91–95

Passo, jr, C. & Cook, G. 1996. Handbook of Environmental Liquid Scintillation Spectrometry. A Compilation of Theory and Methods (USA: Packard Instrument Company)

Peres, A. & Hiromoto, G. 2002. Evaluation of Pb-210 and Po-210 in Cigarette Tobacco Produced in Brazil. *Journal of Environmental Radioactivity* 62:115–119

Perkin Elmer Instruments. 2001. The 30-Minute Guide to ICP-MS. `http://las.perkinelmer.com/Content/TechnicalInfo/TCH_ICPMSThirtyMinuteGuide.pdf`, 13.11.2008

Reichelt, A., Rauh, H., Riepl, S., & Lehman, K. 1994. Anthropogene Stoffe und Produkte mit natürlichen Radionukliden. Teil 3: Untersuchungen zur Strahlenexposition der Bevölkerung, ed. TÜV Bayern im Auftrag des Bayrischen Staatsministeriums für Landesentwicklung und Umweltfragen (München)

Republik Österreich. 2001. 304. Verordnung des Bundesministers für soziale Sicherheit und Generationen über die Qualität von Wasser für den menschlichen Gebrauch (Trinkwasserverordnung – TWV), BGBl.II, Nr.304, ausgegeben am 21. August 2001, ed. Republik Österreich (Wien)

Republik Österreich. 2006. 191. Verordnung des Bundesministers für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft, des Bundesministers für Verkehr, Innovation und Technologie, der Bundesministerin für Gesundheit und Frauen über allgemeine Maßnahmen zum Schutz von Personen vor Schäden durch ionisierende Strahlung (Allgemeine Strahlenschutzverordnung – AllgStrSchV), BGBl II, 191/2006, ed. Republik Österreich (Wien)

Republik Österreich. 2008. 2. Verordnung des Bundesministers für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft und des Bundesministers für Wirtschaft und Arbeit betreffend Strahlenschutz bei natürlichen terrestrischen Strahlenquellen (Natürliche Strahlenquellen-Verordnung – NatStrV), BGBl II, 2/2008, ed. Republik Österreich (Wien)

Ringer, W., Simader, M., Bernreiter, M., Aspek, W., & Kaineder, H. 2006a. Occupational Radiation Exposure in Upper Austrian Water Supplies and Spas, in Proceedings of Second European IRPA Congress on Radiation Protection, Paris

Ringer, W. et al. 2006b. Strahlenexposition durch Trinkwasser in Oberösterreich – 2004-2006. Teilprojekt Strahlenexposition von Beschäftigten in oberösterreichischen Wasserwerken. Endbericht, ed. Land Oberösterreich, Abteilung Umwelt- und Anlagentechnik

Risica, S. & Grande, S. 2000. Council Directive 98/83/EC on the Quality of Water Intended for Human Consumption: Calculation of Derived Activity Concentrations, ed. I. S. D. Sanita (Roma), Rapporti ISTISAN 00/16

Rühle, H. 1996. Radioaktivität in verschiedenen Wasservorkommen, in *Umweltradioaktivität*, ed. A. Siehl (Berlin: Ernst & Sohn)

Rühle, H. 1997. Radon in unserer Umwelt und die damit verbundene Strahlenexposition der Bevölkerung. *Radon und Trinkwasser, Schriftenreihe des Vereins für Wasser-, Boden- und Lufthygiene* pp. 39–55

Salonen, L. 1988. Natural Radionuclides in Ground Water in Finland. *Radiation Protection Dosimetry* 24:163–166

Salonen, L. 1994. U-238 Series Radionuclides as a Source of Increased Radioactivity in Groundwater Originating from Finnish Bedrock, in Conference Proceedings: Future Groundwater Resources at Risk, Helsinki, pp. 71–84

Salonen, L. & Huikuri, P. 2002. Elevated Levels of Uranium Series Radionuclides in Private Water Supplies in Finland, in Proceedings of the 5th International Conference on High Levels of Natural Radiation and Radon Areas, Vol. 2, Munich, pp. 87–91

Schaffer, C. 2005. Untersuchungen zur Entfernung von natürlicher Radioaktivität aus dem Trinkwasser, Diplomarbeit, Universität für Bodenkultur (Wien)

Scheffer, F. & Schachtschabel, P. 2002. Lehrbuch der Bodenkunde, 15th edition (Heidelberg: Spektrum Akademischer Verlag GmbH)

Schönhofer, F. 1989. Determination of Rn-222 and Ra-226 in Mineral Water and Drinking Water – A Survey in Austria. *Analyst* 114:1345–1347

Schönhofer, F. 1990. Radon-222 und Radium-226 in Trinkwasser in Niederösterreich, Report BALUF-RC-90-1

Schönhofer, F. 1992. Measurements of Ra-226 in Water and Rn-222 in Water and Air by Liquid Scintillation Counting. *Rad. Prot. Dosimetry* 45(1/5):123–125

Schönhofer, F. & Maringer, F. 2005. The EU Drinking Water Directive. The Austrian Standard and an Ultra Low-Level Liquid Scintillation Spectrometry Approach for Assuring Compilance, in Proceedings Book of the 2005 International Liquid Scintillation Conference, Radiocarbon, Katowice

Schönhofer, F. & Pock, K. 1996. Rn-222 and Ra-226 in Drinking Water in a Granitic Area in Austria: Measurements with Liquid Scintillation Spectrometry, in Radiocarbon: Liquid Scintillation Spectrometry, ed. G. Cook, D. Harkness, A. MacKenzie, B. Miller, & E. Scott

Schubert, G., Finger, F., Gasser, V., & Lettner, H. 2003. Radionuklide im Grundwasser des kristallinen Untergrunds im Mühlviertel. Endbericht zum Pilotprojekt, ed. Geologische Bundesanstalt Wien

Segovia, N., Tamez, E., Pena, P., et al. 1999. Groundwater Flow System in the Valley of Toluca, Mexico: An Assay of Natural Radionuclide Specific Activities. *Applied Radiation and Isotopes* 50:589–598

Skeppström, K. & Olofsson, B. 2005. Radon in Ground Water: Analysis of Causes Using GIS and Multivariate Statistics. A Case Study in the Stockholm County. *International Congress Series: High Level of Natural Radiation and Radon Areas: Radiation Dose and Health Effects* 1276:206–207

SPSS Inc. 1997a. Sigma Plot – Transform & Regressions, Reference Manual. Sigma Plot® 4.0 for Windows, SPSS Inc., USA

SPSS Inc. 1997b. Sigma Plot – User's Manual. Sigma Plot® 4.0 for Windows, SPSS Inc., USA

Staubmann, K. 2002. Untersuchungen zur Entfernung natürlicher radioaktiver Stoffe aus Trinkwässern und Überblick über deren Verbreitung in Österreich, ed. Institut für Wasservorsorge, Universität für Bodenkultur, Wiener Mitteilungen, Wasser-Abwasser-Gewässer, Band 179 (Wien)

Stolz, W. 2005. Radioaktivität – Grundlagen, Messungen, Anwendungen, 5th edition (Vieweg und Teubner)

Strahlenschutzkommission (SSK). 2003. Strahlenexposition durch Blei-210 und Polonium-210 im Trinkwasser. Stellungnahme der Strahlenschutzkommission, Verabschiedet in der 188. Sitzung der Strahlenschutzkommission am 2.12.2003

Sullivan, P., Agardy, F., & Clark, J. 2005. The Environmental Science of Drinking Water (USA: Elsevier)

Taylor, D. 2002. Radionuclides in the Environment, in *Heavy Metals in the Environment*, ed. Bibudhendra Sarkar (New York: Marcel Dekker, Inc.)

Tayyeb, Z., Kinsara, A., & Farid, S. 1998. A Study on the Radon Concentrations in Water in Jeddah (Saudi Arabia) and the Associated Health Effects. *Journal of Environmental Radioactivity* 38:97–104

Thalmann, F., Schermann, O., Schroll, E., & Hausberger, G. 1989. Geochemischer Atlas der Republik Österreich 1:1,000.000 Böhmische Masse und Zentralzone der Ostalpen (Bachsedimente $<$ 0,18mm) (Wien: Geologische Bundesanstalt)

Trautmannsheimer, M. 2002. Radonexponierte Arbeitsplätze in Wasserwerken in Bayern, ed. Bayerisches Landesamt für Umweltschutz, Abschlussbericht

Tschurlovits, M. 2000. Technischer Strahlenschutz (Wien), Technische Universität Wien, Skriptum zur Vorlesung, LVA 141.010

Umweltbundesamt Wien. 2007a. Groundwater. `http://www.ubavie.gv.at/en/umweltschutz/wasser/grundwasser/?&wai=1`, 07.11.2007

Umweltbundesamt Wien. 2007b. Trinkwasser. `http://www.umweltbundesamt.at/umweltschutz/wasser/trinkwasser/`, 18.6.2007

University of Wisconsin. 2007. Liquid Scintillation Counting; Radiation Safety Program. `http://www.ehs.psu.edu/radprot/LSC_Theory1.pdf`, 15.11.2007

UNSCEAR. 1966. Report on the United Nations Scientific Committee on the Effects of Atomic radiation, General Assembly, Offical Records, Twenty-First Session, Supplement No. 14 (A/6314), ed. United Nations Scientific Committee on The Effects of Atomic Radiation (New York)

UNSCEAR. 1982. Ionizing Radiation – Sources and Biological Effects, ed. United Nations Scientific Committee on The Effects of Atomic Radiation (New York)

UNSCEAR. 1993. Sources and Effects of Ionizing Radiation, UNSCEAR 1993 Report to the General Assembly, ed. United Nations Scientific Committee on The Effects of Atomic Radiation (New York)

UNSCEAR. 2000. Sources and Effects of Ionizing Radiation, ed. United Nations Scientific Committee on The Effects of Atomic Radiation (New York)

U.S. Department of Health and Human Services. 1999. Toxicological Profile For Uranium, Public Health Service, Agency for Toxic Substances and Disease Registry

Valkovic, V. 2000. Radioactivity in the Environment (Amsterdam: Elsevier Science B.V.)

Villalba, L., Colmenero Sujo, L., Montero Cabrera, M., et al. 2005. Radon Concentration in Ground and Drinking Water in the State of Chihuahua, Mexico. *Journal of Environmental Radioactivity* 80:139–151

Vogelsang, D. 1998. Grundwasser (Berlin-Heidelberg: Springer-Verlag)

Vohryzka, K. 1973a. Hydrogeologie von Oberösterreich, ed. Amt der oberösterreichischen Landesregierung, Abteilung Wasser- und Energierecht (Linz)

Vohryzka, K. 1973b. Hydrogeologie von Oberösterreich. Geologische Spezialkarte, ed. Amt der oberösterreichischen Landesregierung, Abteilung Wasser- und Energierecht (Linz)

von Buttlar, H. & Roth, M. 1990. Radioaktivität. Fakten, Ursachen, Wirkungen (Springer Verlag)

von Löw, E. 2008. Physikalisch chemische Kenngrößen und chemische Stoffe, Wasserlabor. `http://www.med.uni-marburg.de/stpg/ukm/lt/umwelthyg/kapb3.htm`, 19.05.2008

von Philipsborn, H. 1998. Strahlenschutz – Radioaktivität und Strahlungmessung, 6th edition, ed. Bayrisches Staatsministerium für Landesentwicklung und Umweltfragen (München)

Voronov, A. 2004. Radon-Rich Waters in Russia. *Environmental Geology* 46:630–634

Wallner, G. 2002. Determination of Ra-228, Ra-226 and Pb-210 in Drinking Water Using Liquid Scintillation Counting, in Radiocarbon: LSC2001 – Advances in Liquid Scintillation Spectrometry, pp. 269–274

Wallner, G. & Steininger, G. 2002. Bestimmung der Radium-226 und Radium-228 Gehalte in österreichischen Trinkwässern und Mineralwässern. Endbericht, ed. Bundesministerium für soziale Sicherheit und Generationen, Sektion IX (Wien), Werkvertrag GZ 353.019/0-IX/9/01

WHO. 1984. Guidelines for Drinking Water, ed. World Health Organization (Genf)

WHO. 1993. Guidelines for Drinking Water Quality, 2nd edition, ed. World Health Organization (Genf)

WHO. 2000. Air Quality Guidelines for Europe. `http://www.euro.who.int/document/e71922.pdf`, 15.01.2008

WHO. 2004. Guidelines for Drinking Water Quality, 3rd edition, ed. World Health Organization (Genf)

Wikimedia Foundation Inc. 2008a. Heinrich Mache. `http://de.wikipedia.org/wiki/Heinrich_Mache`, 17.11.2008

Wikimedia Foundation Inc. 2008b. Phase Diagram. `http://commons.wikimedia.org/wiki/File:Image_Phase-diag2.svg`, 14.12.2008

Wisser, S. 2003. Balancing Natural Radionuclides in Drinking Water Supply. An Investigation in Germany and Canda With Respect to Geology, Radiometry and Legislation, Dissertation, Johannes Gutenberg-Universität (Mainz)

Xinwei, L. & Xiaolan, Z. 2004. Study of the Radon Concentrations in Drinking Water From Three Main Cities of Shaanxi Province, China. *Environmental Geology* 45:1082–1086

Zalewski, M., Karpinska, M., Mnich, Z., Kapala, J., & Zalewski, P. 2001. Study of Rn-222 Concentrations in Drinking Water in the North-Eastern Hydroregions of Poland. *Journal of Environmental Radioactivity* 53:167–173

Zhuo, W., Iida, T., & Yang, X. 2001. Occurrence of Rn-222, Ra-226, Ra-228 and U in Groundwater in Fujian Province, China. *Journal of Environmental Radioactivity* 53:111–120

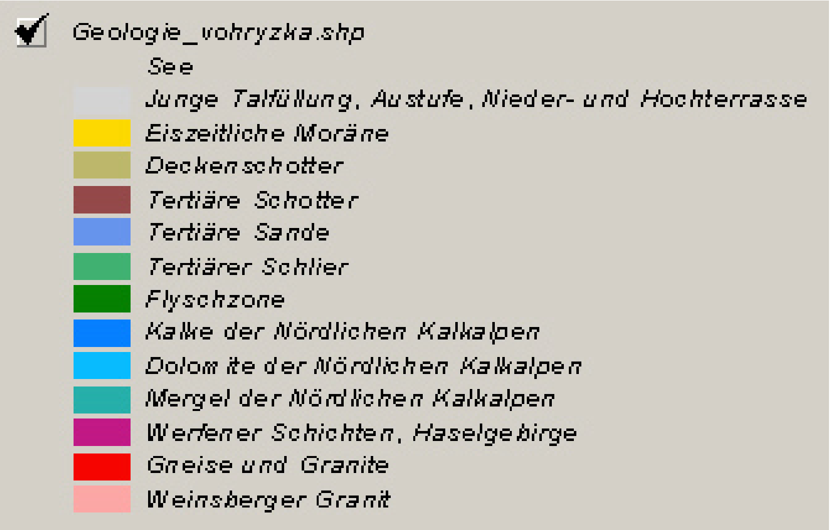
Zikovsky, L. 2006. Alpha Radioactivity in Drinking Water in Quebec, Canada. *Journal of Environmental Radioactivity* 88:306–309

Zikovsky, L. & Chah, B. 1990. The Lognormal Distribution of Radon Concentration in Ground Water. *Ground Water* 28(5):673–676

Annex

A1. Legends

Legend for the hydrogeological map of Upper Austria (Vohryzka, 1973b)



☑ Geologie_vohryzka.shp
- See
- ⬜ Junge Talfüllung, Austufe, Nieder- und Hochterrasse
- 🟨 Eiszeitliche Moräne
- 🟫 Deckenschotter
- 🟫 Tertiäre Schotter
- 🟦 Tertiäre Sande
- 🟩 Tertiärer Schlier
- 🟩 Flyschzone
- 🟦 Kalke der Nördlichen Kalkalpen
- 🟦 Dolomite der Nördlichen Kalkalpen
- 🟩 Mergel der Nördlichen Kalkalpen
- 🟪 Werfener Schichten, Haselgebirge
- 🟥 Gneise und Granite
- 🟥 Weinsberger Granit

Classification of the survey sampling results

- 🟢 Class 1
- 🟡 Class 2
- 🟠 Class 3
- 🔴 Class 4

Classification of the detailed sampling results

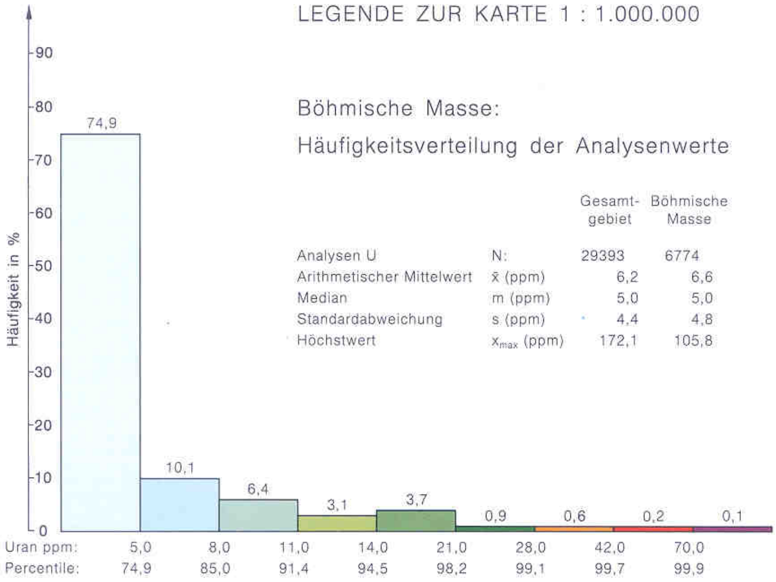
Water Units (WVA)
- 🟢 Class 1
- 🟡 Class 2
- 🟠 Class 3
- 🔴 Class 4

Private Wells (H)
- 🟩 Class 1
- 🟨 Class 2
- 🟧 Class 3
- 🟥 Class 4

“Sacred Wells” (HEIL)
- ▲ Class 1
- ▲ Class 2
- ▲ Class 3
- ▲ Class 4

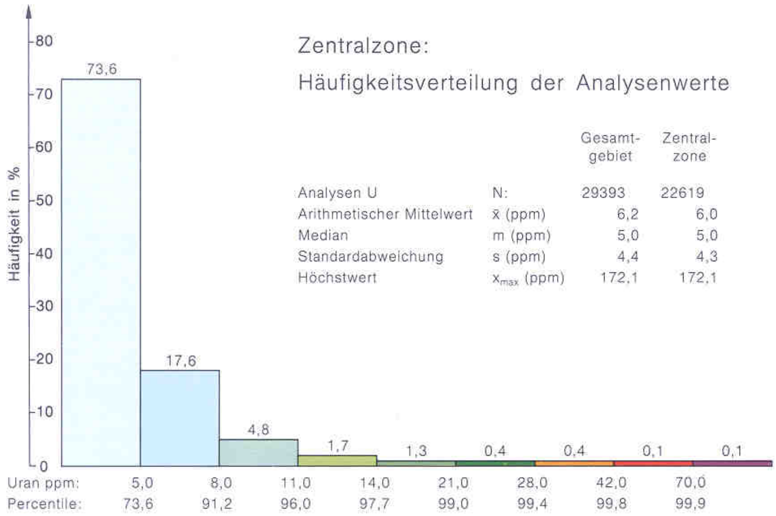
Legend for the geochemical atlas of Austria – uranium (Thalmann et al., 1989)

LEGENDE ZUR KARTE 1 : 1.000.000



Böhmische Masse:
Häufigkeitsverteilung der Analysenwerte

		Gesamt-gebiet	Böhmische Masse
Analysen U	N:	29393	6774
Arithmetischer Mittelwert	$\bar{x}$ (ppm)	6,2	6,6
Median	m (ppm)	5,0	5,0
Standardabweichung	s (ppm)	4,4	4,8
Höchstwert	x_{max} (ppm)	172,1	105,8



Zentralzone:
Häufigkeitsverteilung der Analysenwerte

		Gesamt-gebiet	Zentral-zone
Analysen U	N:	29393	22619
Arithmetischer Mittelwert	$\bar{x}$ (ppm)	6,2	6,0
Median	m (ppm)	5,0	5,0
Standardabweichung	s (ppm)	4,4	4,3
Höchstwert	x_{max} (ppm)	172,1	172,1

Unbeprobte zusammenhängende Gletscherregion ▨

A2. Radiometric results of the survey sampling

n.a.	. . . not analyzed
<DL	. . . below decison limit

Sample taken by (see Chapter 5.1.1)

1 . . . in water supplies within the second part of the Upper Austrian water project
2 . . . in private wells and small water supplies by the AGES, CC Hydroanalytik
3 . . . in fractured aquifers and karstic areas within the WGEV sampling

				Activity concentration Bq/l									
Sample No.	Sample taken by	Sampling date	Sampling community	gross alpha-beta	unc. gross alpha-beta	H-3	unc. H-3	Ra-226	unc. Ra-226	U-238	unc. U-238	Rn-222	unc. Rn-222
1	1	27.10.2004	Hellmonsödt	<DL		1.02	0.74	<DL		0.0051	0.0006	35.8	4.3
2	1	27.10.2004	Hellmonsödt	<DL		0.84	0.73	<DL		0.0050	0.0006	8.9	2.4
3	1	27.10.2004	Hellmonsödt	<DL		0.75	0.72	<DL		0.0048	0.0006	5.4	2.4
4	1	27.10.2004	Bad Leonfelden	0.59	0.37	0.85	0.74	<DL		0.0040	0.0005	38.3	4.6
5	1	27.10.2004	Bad Leonfelden	<DL		<DL		<DL		0.0145	0.0018	49.3	5.4
6	1	27.10.2004	Bad Leonfelden	<DL		1.05	0.74	<DL		0.0162	0.0020	28.8	3.8
7	1	28.10.2004	Helfenberg	<DL		1.47	0.77	<DL		0.0048	0.0006	83.3	7.1
8	1	28.10.2004	Grünbach	0.45	0.37	1.44	0.77	<DL		0.0402	0.0049	82.4	7.0
9	1	28.10.2004	Grünbach	0.45	0.36	1.15	0.75	<DL		0.0309	0.0037	26.5	3.5
10	1	02.11.2004	Ebensee	<DL		<DL		<DL		<DL		3.9	1.0
11	1	02.11.2004	Altmünster	0.54	0.34	0.90	0.86	<DL		0.0058	0.0007	5.2	1.0
12	1	02.11.2004	Altmünster	<DL		<DL		<DL		0.0025	0.0003	3.7	1.0
13	1	02.11.2004	Gmunden	0.49	0.34	<DL		<DL		0.0030	0.0004	3.2	1.0
14	1	02.11.2004	Gmunden	<DL		<DL		<DL		0.0717	0.0087	3.9	0.9
15	1	02.11.2004	Laakirchen	<DL		<DL		<DL		<DL		5.4	1.0
16	1	03.11.2004	Ulrichsberg	<DL		0.87	0.71	<DL		0.0009	0.0001	140.2	35.7
17	1	04.11.2004	Bad Ischl	<DL		<DL		<DL		0.0083	0.0010	5.3	0.8
18	1	04.11.2004	Bad Ischl	<DL		<DL		<DL		0.0055	0.0007	6.2	0.9
19	1	04.11.2004	Nußdorf am Attersee	0.39	0.33	<DL		<DL		0.0035	0.0004	10.9	1.2
20	1	04.11.2004	Lenzing	0.39	0.34	<DL		<DL		<DL		10.6	1.2
21	1	04.11.2004	Vöcklabruck	<DL		1.09	0.87	<DL		0.0010	0.0001	2.8	0.7
22	1	09.11.2004	Kirchdorf a/d Krems	<DL		1.64	0.75	<DL		<DL		28.2	9.9
23	1	09.11.2004	Kirchdorf a/d Krems	0.47	0.37	0.85	0.70	<DL		<DL		19.9	8.7
24	1	09.11.2004	Bad Hall	<DL		1.03	0.71	<DL		0.0032	0.0004	12.9	8.9
25	1	09.11.2004	Bad Hall	<DL		<DL		<DL		0.0027	0.0003	<DL	
26	1	09.11.2004	Steyr	<DL		<DL		<DL		0.0315	0.0038	<DL	
27	1	09.11.2004	Steyr	<DL		0.80	0.70	<DL		0.0222	0.0027	12.5	8.9
28	1	10.11.2004	Steyregg	<DL		1.05	0.71	<DL		0.0007	0.0001	14.2	7.6
29	1	10.11.2004	Steyregg	0.46	0.37	1.10	0.72	<DL		0.0270	0.0033	9.9	8.0
30	1	10.11.2004	Steyregg	<DL		1.32	0.73	<DL		0.0002	0.0000	<DL	
31	1	10.11.2004	Mauthausen	0.42	0.37	0.87	0.71	<DL		0.0002	0.0000	<DL	
32	1	10.11.2004	Mauthausen	<DL		0.87	0.71	<DL		0.0134	0.0016	10.2	7.9
33	1	11.11.2004	Schwanenstadt	<DL		<DL		<DL		0.0102	0.0012	9.0	7.0
34	1	11.11.2004	Schwanenstadt	<DL		0.84	0.70	<DL		0.0031	0.0004	10.4	6.7
35	1	11.11.2004	Stadl-Paura	<DL		0.96	0.85	<DL		0.1662	0.0201	8.3	7.0
36	1	11.11.2004	Stadl-Paura	<DL		1.39	0.88	<DL		0.0060	0.0007	11.6	6.4
37	1	11.11.2004	Attnang-Puchheim	<DL		1.83	0.91	<DL		0.0015	0.0002	15.3	6.3
38	1	11.11.2004	Attnang-Puchheim	<DL		1.18	0.86	<DL		<DL		10.1	6.5
39	1	12.11.2004	Rohrbach in Oberösterreich	<DL		2.24	0.92	<DL		0.0056	0.0007	44.3	9.1
40	1	12.11.2004	Rohrbach in Oberösterreich	<DL		0.99	0.90	<DL		0.0021	0.0003	9.0	5.6
41	1	15.11.2004	Aschach a/d Donau	<DL		1.16	0.86	<DL		<DL		11.1	3.8

				Activity concentration Bq/l									
Sample No.	Sample taken by	Sampling date	Sampling community	gross alpha-beta	unc. gross alpha-beta	H-3	unc. H-3	Ra-226	unc. Ra-226	U-238	unc. U-238	Rn-222	unc. Rn-222
42	1	15.11.2004	Aschach a/d Donau	<DL		1.58	0.88	<DL		<DL		<DL	
43	1	15.11.2004	Eferding	<DL		<DL		<DL		0.0882	0.0107	10.3	3.7
44	1	15.11.2004	Eferding	<DL		<DL		<DL		<DL		13.5	3.9
45	1	15.11.2004	Grieskirchen	<DL		<DL		<DL		0.0043	0.0005	8.4	3.6
46	1	15.11.2004	Grieskirchen	<DL		0.93	0.85	<DL		0.0056	0.0007	13.9	4.1
47	1	15.11.2004	Gunskirchen	<DL		<DL		<DL		0.0479	0.0058	6.1	3.9
48	1	15.11.2004	Gunskirchen	<DL		0.87	0.84	<DL		0.0055	0.0007	5.7	4.5
49	1	16.11.2004	Freistadt	<DL		0.91	0.72	<DL		0.0345	0.0042	0.7	0.5
50	1	16.11.2004	Freistadt	<DL		<DL		<DL		0.0805	0.0098	19.5	2.6
51	1	16.11.2004	Gallneukirchen	<DL		<DL		<DL		0.0311	0.0038	28.3	3.4
52	1	16.11.2004	Gallneukirchen	<DL		<DL		<DL		0.0703	0.0085	2.9	1.5
53	1	16.11.2004	Engerwitzdorf	<DL		<DL		<DL		0.0138	0.0017	15.5	2.3
54	1	16.11.2004	Engerwitzdorf	<DL		<DL		<DL		0.0926	0.0112	0.7	0.5
55	1	17.11.2004	Braunau am Inn	<DL		<DL		<DL		0.0088	0.0011	8.7	1.4
56	1	17.11.2004	Braunau am Inn	<DL		<DL		<DL		0.0026	0.0003	6.1	1.1
57	1	19.11.2004	Pregarten	<DL		<DL		<DL		0.0068	0.0008	8.7	1.3
58	1	19.11.2004	Pregarten	<DL		<DL		<DL		0.0079	0.0010	12.5	1.7
59	3	22.11.2004	Puchenau	<DL		0.82	0.72	<DL		0.0047	0.0006	7.6	2.8
60	3	22.11.2004	Lichtenberg	<DL		0.80	0.71	<DL		0.0001	0.0000	30.3	5.1
61	1	22.11.2004	Waldhausen im Strudengau	<DL		0.81	0.61	<DL		0.0024	0.0003	18.9	2.8
62	3	22.11.2004	Oberneukirchen	<DL		1.25	0.74	<DL		0.0009	0.0001	5.2	3.6
63	1	22.11.2004	Grein	<DL		0.62	0.59	<DL		0.0016	0.0002	12.1	4.1
64	1	22.11.2004	Grein	<DL		0.94	0.87	<DL		0.0013	0.0002	8.6	1.8
65	3	22.11.2004	Bad Leonfelden	<DL		1.66	0.76	<DL		0.0025	0.0003	67.4	9.0
66	1	22.11.2004	Perg	<DL		1.30	0.97	<DL		0.0019	0.0002	38.0	14.8
67	1	23.11.2004	Schlierbach	<DL		0.65	0.59	<DL		0.0022	0.0003	52.7	47.3
68	1	23.11.2004	Schlierbach	<DL		0.97	0.73	<DL		0.0020	0.0002	45.4	19.2
69	1	23.11.2004	Ottensheim	<DL		0.49	0.44	<DL		0.0021	0.0003	49.0	12.5
70	1	23.11.2004	Ottensheim	<DL		0.73	0.55	<DL		0.0021	0.0002	47.2	12.6
71	3	23.11.2004	Großraming	<DL		1.14	0.73	<DL		0.0404	0.0049	<DL	
72	1	23.11.2004	Traun	<DL		0.36	0.36	<DL		0.0212	0.0026	22.9	6.4
73	1	23.11.2004	Traun	<DL		0.18	0.17	<DL		0.0308	0.0037	10.8	7.7
74	1	24.11.2004	Schärding	<DL		0.27	0.20	<DL		0.0260	0.0032	16.9	15.7
75	1	24.11.2004	Schärding	<DL		0.23	0.17	<DL		0.0284	0.0034	13.8	9.2
76	3	25.11.2004	Perg	<DL		1.03	0.73	<DL		0.0066	0.0008	7.0	2.0
77	1	25.11.2004	St.Georgen a/d Gusen	<DL		0.63	0.54	<DL		0.0175	0.0021	10.4	3.4
78	1	25.11.2004	St.Georgen a/d Gusen	<DL		0.83	0.66	<DL		0.0120	0.0015	8.7	3.2
79	1	25.11.2004	Luftenberg a/d Donau	<DL		0.73	0.40	<DL		0.0148	0.0018	5.2	3.5
80	1	25.11.2004	Luftenberg a/d Donau	<DL		0.78	0.46	<DL		0.0134	0.0016	6.9	2.1
81	1	29.11.2004	Spital am Pyhrn	<DL		0.76	0.59	<DL		0.0141	0.0017	6.1	2.3
82	1	29.11.2004	Spital am Pyhrn	<DL		0.38	0.38	<DL		0.0137	0.0017	6.5	2.3

				Activity concentration Bq/l									
Sample No.	**Sample taken by**	**Sampling date**	**Sampling community**	**gross alpha-beta**	**unc. gross alpha-beta**	**H-3**	**unc. H-3**	**Ra-226**	**unc. Ra-226**	**U-238**	**unc. U-238**	**Rn-222**	**unc. Rn-222**
83	2	29.11.2004	Goldwörth	<DL		0.86	0.74	<DL		0.0414	0.0050	11.1	3.9
84	2	29.11.2004	Feldkirchen	<DL		1.09	0.76	<DL		0.0115	0.0014	11.6	3.9
85	1	29.11.2004	Wels	<DL		0.98	0.64	<DL		0.0265	0.0032	11.3	7.0
86	2	30.11.2004	Alkoven	<DL		1.01	0.76	<DL		0.0078	0.0009	7.0	3.2
87	2	30.11.2004	Alkoven	<DL		1.61	0.74	<DL		0.0005	0.0001	6.4	3.2
88	3	01.12.2004	Wilhering	<DL		<DL		<DL		0.0043	0.0005	<DL	
89	2	01.12.2004	Prambachkirchen	<DL		1.89	0.80	<DL		0.0339	0.0041	15.2	3.3
90	1	01.12.2004	Mondsee	<DL		0.56	0.40	<DL		0.0191	0.0023	2.5	1.3
91	1	01.12.2004	Mondsee	<DL		1.23	0.80	<DL		0.0096	0.0012	7.6	4.4
92	2	01.12.2004	Peuerbach	<DL		<DL		<DL		0.0335	0.0041	8.0	2.8
93	3	01.12.2004	Stroheim	<DL		1.23	0.79	<DL		0.2461	0.0298	16.8	11.8
94	2	01.12.2004		<DL		<DL		<DL		0.0088	0.0011	5.8	3.0
95	2	01.12.2004	Kopfing	<DL		0.78	0.73	<DL		0.0134	0.0016	280.2	30.6
96	2	01.12.2004	Rainbach/I.	<DL		2.05	0.82	<DL		<DL		13.7	3.2
97	2	01.12.2004	Hofmarkt	<DL		2.49	0.84	<DL		0.0108	0.0013	37.6	5.5
98	2	02.12.2004	Mayrhof	<DL		0.97	0.75	<DL		0.0006	0.0001	6.6	2.4
99	2	02.12.2004	Senftenbach	<DL		<DL		<DL		0.0058	0.0007	8.6	2.5
100	2	02.12.2004	Peterskirchen	<DL		0.86	0.74	<DL		0.0021	0.0003	18.7	3.4
101	3	07.12.2004	Lichtenau	<DL		1.43	0.79	<DL		0.0011	0.0001	8.0	5.6
102	3	09.12.2004	Hellmonsödt	<DL		0.86	0.76	0.046	0.043	<DL		342.3	37.3
103	3	09.12.2004	Reichenthal	<DL		1.51	0.81	<DL		0.0002	0.0000	143.2	17.4
104	3	09.12.2004	Leopoldschlag	<DL		1.19	0.78	<DL		0.0073	0.0009	45.2	7.7
105	3	09.12.2004	Lasberg	<DL		1.28	0.79	<DL		0.0238	0.0029	42.0	7.1
106	2	09.12.2004	Gaspoltshofen	<DL		1.80	0.81	<DL		0.0117	0.0014	8.1	1.3
107	3	09.12.2004	St. Leonhard b. Freistadt	0.37	0.33	1.10	0.78	<DL		0.0011	0.0001	71.7	10.4
108	2	09.12.2004	St.Marienkirchen	<DL		1.67	0.79	<DL		0.0068	0.0008	20.5	2.5
109	2	09.12.2004	Neumarkt/H.	<DL		2.08	0.86	<DL		0.0169	0.0020	5.8	1.1
110	2	09.12.2004	Taufkirchena.d.Pram	<DL		1.02	0.79	<DL		0.0066	0.0008	5.0	1.0
111	2	09.12.2004	Neustift	<DL		1.06	0.80	<DL		0.0009	0.0001	53.4	5.8
112	2	09.12.2004	St.Agatha	<DL		1.39	0.78	<DL		0.0067	0.0008	59.9	6.5
113	2	13.12.2004	Bad Goisern	<DL		<DL		<DL		0.0051	0.0006	<DL	
114	2	13.12.2004	Hallstatt	<DL		1.35	0.82	<DL		0.0601	0.0073	<DL	
115	2	13.12.2004	Schwand	<DL		0.86	0.74	<DL		0.0061	0.0007	19.9	3.6
116	2	13.12.2004	Lohnsburg	<DL		<DL		<DL		0.0004	0.0000	8.4	2.6
117	2	13.12.2004	Überackern	<DL		1.07	0.75	<DL		0.0068	0.0008	20.5	3.7
118	2	13.12.2004	Aspach	<DL		<DL		<DL		0.0027	0.0003	7.0	2.6
119	2	13.12.2004	Hochburg-Ach	<DL		1.98	0.82	<DL		0.0108	0.0013	10.0	2.8
120	2	13.12.2004	KirchdorfamInn	<DL		0.93	0.74	<DL		0.0058	0.0007	5.1	2.9
121	2	13.12.2004	Eggelsberg	<DL		0.86	0.74	<DL		0.0115	0.0014	10.9	2.9
122	2	14.12.2004	BadWimsbach	<DL		<DL		<DL		0.0102	0.0012	8.4	2.4
123	2	14.12.2004	Vorchdorf	<DL		0.80	0.74	<DL		0.0007	0.0001	7.4	2.3

				Activity concentration Bq/l									
Sample No.	**Sample taken by**	**Sampling date**	**Sampling community**	**gross alpha-beta**	**unc. gross alpha-beta**	**H-3**	**unc. H-3**	**Ra-226**	**unc. Ra-226**	**U-238**	**unc. U-238**	**Rn-222**	**unc. Rn-222**
124	2	14.12.2004	Seewalchen	<DL		0.80	0.74	<DL		0.0061	0.0007	15.7	3.0
125	2	15.12.2004	Burgkirchen	<DL		<DL		<DL		0.0196	0.0024	12.5	2.6
126	3	15.12.2004	Haslach/Mühl, Hörleinsödt	<DL		1.19	0.78	<DL		0.0006	0.0001	13.4	2.3
127	3	15.12.2004	Laussa, Pechgraben	<DL		1.01	0.77	<DL		0.0122	0.0015	11.8	2.2
128	2	15.12.2004	Munderfing	<DL		<DL		<DL		0.0043	0.0005	13.6	2.6
129	3	15.12.2004	Atzesberg	<DL		0.87	0.76	<DL		0.0105	0.0013	112.8	12.3
130	3	15.12.2004	Hofkirchen i. Mühlkreis	<DL		0.98	0.77	<DL		0.0006	0.0001	19.9	2.9
131	2	15.12.2004	Kirchberg	<DL		0.78	0.73	<DL		0.0048	0.0006	9.4	2.3
132	3	15.12.2004	St. Martin i. Mühlkreis	<DL		1.27	0.83	<DL		0.0848	0.0103	16.5	2.6
133	2	15.12.2004	Ostermiething	<DL		0.93	0.74	<DL		<DL		8.4	2.1
134	2	15.12.2004	Lengau	<DL		<DL		<DL		0.0077	0.0009	14.6	2.7
135	2	16.12.2004	Mitterstoder	<DL		<DL		<DL		0.0032	0.0004	<DL	
136	2	16.12.2004	Hinterstoder	<DL		1.03	0.81	<DL		0.0598	0.0072	8.1	6.4
137	2	16.12.2004	Mitterstoder	<DL		1.70	0.91	<DL		0.0032	0.0004	<DL	
138	2	16.12.2004	Klaus	<DL		0.88	0.80	<DL		0.0005	0.0001	9.7	5.7
139	2	16.12.2004	Buchkirchen	<DL		2.69	0.85	<DL		0.0208	0.0025	21.6	3.4
140	2	16.12.2004	Krenglbach	<DL		1.85	0.81	<DL		0.0130	0.0016	13.8	2.5
141	2	16.12.2004	Luftenberg	<DL		<DL		<DL		0.0004	0.0000	6.6	1.7
142	2	16.12.2004	Ansfelden	<DL		1.26	0.75	<DL		0.0031	0.0004	9.5	1.9
143	2	16.12.2004	Leonding	<DL		<DL		<DL		0.0056	0.0007	6.9	1.8
144	2	16.12.2004	Scharnstein	<DL		0.77	0.72	<DL		0.0143	0.0017	7.1	1.7
145	2	16.12.2004	Grünau	<DL		0.99	0.73	<DL		0.0413	0.0050	4.3	3.9
146	2	16.12.2004	Linz	<DL		1.55	0.77	<DL		0.0149	0.0018	18.6	2.9
147	2	16.12.2004	Scharnstein	<DL		<DL		<DL		0.0058	0.0007	6.0	1.8
148	2	16.12.2004	Linz	<DL		<DL		<DL		0.0052	0.0006	25.3	3.4
149	2	16.12.2004	Pettenbach	<DL		<DL		<DL		0.0010	0.0001	7.7	1.9
150	2	20.12.2004	Münzbach	0.40	0.34	1.38	0.75	<DL		0.0047	0.0006	239.0	26.1
151	2	20.12.2004	Baumgartenberg	0.90	0.36	1.83	0.78	<DL		0.0066	0.0008	13.4	3.3
152	2	20.12.2004	Mitterkirchen	0.47	0.34	<DL		<DL		0.0027	0.0003	13.8	3.5
153	2	20.12.2004	Klam	<DL		<DL		<DL		0.0072	0.0009	21.0	4.1
154	3	20.12.2004	Reichenau i. Mühlkreis	<DL		1.16	0.78	<DL		0.0046	0.0006	37.6	4.1
155	3	20.12.2004	Schenkenfelden	<DL		0.94	0.77	<DL		0.0001	0.0000	53.4	5.8
156	3	20.12.2004	St. Oswald b. Freistadt	<DL		1.46	0.80	<DL		0.0248	0.0030	90.9	9.9
157	3	20.12.2004	Kefermarkt	0.35	0.33	1.41	0.80	<DL		0.0133	0.0016	12.3	1.6
158	2	21.12.2004	Ried/Riedmark	1.05	0.40	<DL		0.080	0.039	0.0042	0.0005	54.2	5.9
159	2	21.12.2004	Naarn	<DL		0.83	0.72	<DL		0.0190	0.0023	12.4	1.7
160	2	22.12.2004	Perg und Umgebung	<DL		<DL		<DL		0.0025	0.0003	15.9	1.9
161	2	22.12.2004	Waldhausen	<DL		<DL		<DL		0.0067	0.0008	154.5	16.9
162	2	22.12.2004	Königswiesen	<DL		1.44	0.84	<DL		0.0009	0.0001	79.9	8.7
163	2	22.12.2004	Liebenau	0.36	0.35	1.40	0.83	<DL		0.0232	0.0028	149.1	16.3
164	2	22.12.2004	Sandl	<DL		0.91	0.80	<DL		0.0153	0.0019	254.5	24.7

				Activity concentration Bq/l									
Sample No.	Sample taken by	Sampling date	Sampling community	gross alpha-beta	unc. gross alpha-beta	H-3	unc. H-3	Ra-226	unc. Ra-226	U-238	unc. U-238	Rn-222	unc. Rn-222
165	2	22.12.2004	Freistadt	<DL		<DL		<DL		0.0118	0.0014	31.2	3.4
166	2	22.12.2004	Waldburg	<DL		1.04	0.75	<DL		0.0067	0.0008	90.8	9.9
167	2	22.12.2004	Leopoldschlag	<DL		1.67	0.79	<DL		0.0350	0.0042	57.4	6.3
168	2	22.12.2004	Gutau	<DL		0.99	0.73	<DL		0.0275	0.0033	44.7	4.9
169	2	22.12.2004	Kefermarkt	<DL		0.81	0.73	<DL		0.0048	0.0006	46.6	5.1
170	2	11.01.2005	Regau	<DL		1.07	0.70	<DL		<DL		7.1	4.1
171	2	11.01.2005	Timelkam	<DL		0.92	0.69	<DL		0.0081	0.0010	13.7	4.0
172	2	11.01.2005	Frankenmarkt	<DL		1.29	0.72	<DL		0.0041	0.0005	12.5	4.0
173	3	12.01.2005	Kirchdorf a/d Krems	0.39	0.36	1.77	0.90	<DL		0.0070	0.0009	4.4	2.9
174	3	12.01.2005	Roßleithen	<DL		1.00	0.81	<DL		0.0072	0.0009	4.5	3.7
175	2	12.01.2005	Gramastetten	<DL		1.35	0.72	<DL		0.0002	0.0000	56.6	8.2
176	3	12.01.2005	Vorderstoder	<DL		<DL		<DL		0.0147	0.0018	4.4	3.4
177	2	12.01.2005	St.Gotthard	<DL		1.60	0.74	<DL		0.0003	0.0001	81.2	10.8
178	2	12.01.2005	St.Martin	<DL		1.07	0.70	<DL		0.0021	0.0003	71.4	9.5
179	2	12.01.2005	Altenfelden	<DL		1.49	0.74	<DL		0.0002	0.0000	23.2	4.5
180	3	13.01.2005	Eidenberg	<DL		<DL		<DL		0.0005	0.0001	27.8	4.0
181	3	13.01.2005	Gramastetten	<DL		<DL		<DL		<DL		14.7	2.7
182	2	13.01.2005	Sarleinsbach	<DL		1.51	0.73	<DL		<DL		21.9	4.3
183	2	13.01.2005	Peilstein	<DL		1.01	0.70	<DL		0.0002	0.0001	77.9	9.4
184	2	13.01.2005	Schwarzenberg	<DL		1.40	0.73	<DL		<DL		57.9	7.7
185	3	17.01.2005	Oftering	<DL		0.92	0.81	<DL		0.0030	0.0004	7.7	1.3
186	3	17.01.2005	Waizenkirchen	<DL		1.47	0.84	<DL		0.0007	0.0001	70.6	7.7
187	3	17.01.2005	Haibach b. Schärding	<DL		1.07	0.82	<DL		0.0045	0.0006	63.5	6.9
188	2	18.01.2005	St.Veit	<DL		1.18	0.71	<DL		0.0003	0.0001	71.4	7.8
189	2	18.01.2005	St.Peter	<DL		1.42	0.72	<DL		0.0007	0.0001	101.3	11.0
190	2	18.01.2005	Helfenberg	<DL		1.19	0.79	<DL		<DL		50.5	6.1
191	3	19.01.2005	Steyregg	<DL		1.14	0.82	<DL		0.0013	0.0002	92.4	10.1
192	3	19.01.2005	Grein	<DL		<DL		<DL		0.0176	0.0021	17.7	2.1
193	2	19.01.2005	Ebensee	<DL		0.90	0.78	<DL		0.0009	0.0001	<DL	
194	2	19.01.2005	Ebensee	<DL		1.03	0.80	<DL		0.0023	0.0003	3.5	1.3
195	2	19.01.2005	Kirchdorf	<DL		1.28	0.81	<DL		0.0038	0.0005	12.7	2.0
196	2	19.01.2005	Schlierbach	0.83	0.27	1.08	0.76	<DL		0.0031	0.0004	11.4	1.8
197	2	20.01.2005	Sipbachzell	<DL		1.30	0.80	<DL		0.0087	0.0011	<DL	
198	2	22.01.2005	Schlatt	<DL		0.86	0.82	<DL		0.0076	0.0009	<DL	
199	3	24.01.2005	Altenberg bei Linz	<DL		1.24	0.80	<DL		0.0002	0.0000	15.3	3.7
200	3	24.01.2005	Hellmonsoedt	0.77	0.25	1.85	0.85	<DL		0.0410	0.0049	344.0	37.5
201	3	24.01.2005	St. Oswald bei Freistadt	<DL		2.22	0.64	<DL		0.0120	0.0014	47.3	6.9
202	3	24.01.2005	Kefermarkt	<DL		<DL		<DL		0.0070	0.0008	20.3	4.2
203	3	24.01.2005	Hagenberg im Mühlkreis	<DL		1.19	0.79	<DL		0.0071	0.0009	30.4	5.2
204	3	31.01.2005	Laakirchen	<DL		1.04	0.78	<DL		0.0070	0.0008	4.6	1.2
205	3	31.01.2005	Bad Ischl	<DL		0.79	0.77	<DL		0.0029	0.0004	3.8	1.2

A3. Radiometric results of the detailed sampling

n.a.	. . . not analyzed
<DL	. . . below decison limit

Sample	Sampling date	Activity concentration Bq/l																	
		gross alpha	unc. gross alpha	gross beta	unc. gross beta	H-3	unc. H-3	Ra-226	unc. Ra-226	Ra-228	unc. Ra-228	U-238	unc. U-238	Pb-210	Unc. Pb-210	Po-210	Unc. Po-210	Rn-222	unc. Rn-222
H1	20.06.2005	<DL		<DL		2.20	1.06	0.079	0.020	<DL		0.0427	0.0043	0.102	0.018	0.062	0.009	745.2	63.8
H2	20.06.2005	<DL		<DL		1.70	1.02	<DL		0.008	0.005	0.0124	0.0015	0.040	0.007	0.020	0.004	165.2	17.0
H3	20.06.2005	<DL		<DL		1.60	0.99	<DL		0.010	0.005	0.0007	0.0004	0.012	0.006	<DL		75.6	9.4
H4	20.06.2005	<DL		<DL		1.70	1.02	<DL		n.a.		0.0002	0.0000	0.005	0.004	0.003	0.002	15.2	3.5
H5	20.06.2005	<DL		<DL		1.70	0.99	<DL		<DL		0.0003	0.0000	0.014	0.004	0.003	0.002	80.5	9.9
H6	20.06.2005	<DL		1.40	0.64	<DL		<DL		n.a.		0.0005	0.0001	n.a.		n.a.		37.7	5.9
H7	20.06.2005	<DL		1.00	0.61	1.50	0.99	<DL		n.a.		0.0009	0.0003	0.010	0.002	0.017	0.014	53.8	7.5
H8	20.06.2005	<DL		1.10	0.62	<DL		<DL		n.a.		0.0007	0.0001	0.017	0.006	0.056	0.028	118.5	13.1
H9	20.06.2005	<DL		1.40	0.62	<DL		<DL		<DL		0.0022	0.0002	0.058	0.012	0.075	0.030	181.6	18.4
H10	21.06.2005	<DL		<DL		1.90	1.10	<DL		<DL		0.0029	0.0003	0.013	0.004	0.018	0.010	42.7	6.4
H11	21.06.2005	<DL		<DL		1.90	1.08	<DL		<DL		0.0009	0.0001	0.023	0.006	0.034	0.016	72.7	9.2
H12	21.06.2005	<DL		0.97	0.62	1.70	1.07	<DL		<DL		0.0013	0.0001	0.061	0.014	0.119	0.046	530.6	46.6
H13	21.06.2005	<DL		1.10	0.62	1.50	1.10	<DL		<DL		0.0010	0.0006	0.055	0.013	0.100	0.040	278.2	26.3
H14	21.06.2005	0.39	0.12	1.20	0.65	1.90	1.06	<DL		0.022	0.006	0.1678	0.0755	0.062	0.012	0.097	0.035	244.3	23.5
H15	21.06.2005	<DL		<DL		2.10	1.05	<DL		0.008	0.006	0.0016	0.0002	0.018	0.007	0.016	0.004	201.2	20.0
H16	21.06.2005	<DL		<DL		2.20	1.10	<DL		<DL		0.0007	0.0001	<DL		0.008	0.002	129.8	14.1
H17	21.06.2005	<DL		<DL		2.80	1.06	<DL		0.009	0.004	0.0007	0.0001	<DL		<DL		318.4	29.5
H18	21.06.2005	<DL		<DL		1.90	1.03	<DL		<DL		0.0008	0.0001	0.036	0.006	0.046	0.011	101.2	11.7
H19	21.06.2005	<DL		<DL		2.10	1.07	<DL		<DL		0.0007	0.0001	<DL		0.006	0.003	103.1	11.8
H20	22.06.2005	<DL		0.89	0.61	1.20	0.96	<DL		n.a.		0.0050	0.0005	0.021	0.005	0.037	0.011	146.7	15.5
H21	22.06.2005	<DL		1.00	0.59	1.30	0.96	<DL		n.a.		0.0008	0.0001	0.005	0.004	0.064	0.031	230.8	22.4
H22	05.07.2005	<DL		<DL		2.20	0.99	<DL		<DL		0.0009	0.0001	0.046	0.006	0.018	0.003	35.2	5.7
H23	05.07.2005	<DL		<DL		1.80	0.97	<DL		<DL		0.0005	0.0001	0.007	0.006	0.011	0.002	188.9	19.0
H24	05.07.2005	<DL		<DL		<DL		<DL		n.a.		0.0047	0.0005	0.005	0.002	0.003	0.001	19.8	4.0
H25	06.07.2005	<DL		<DL		1.60	1.10	<DL		<DL		0.0001	0.0000	<DL		0.010	0.007	22.4	4.3
H26	06.07.2005	<DL		<DL		1.70	1.09	<DL		0.008	0.005	0.0008	0.0001	<DL		0.023	0.015	86.6	10.4
H27	08.11.2005	<DL		<DL		<DL		0.050	0.040	<DL		<DL		0.004	0.002	0.006	0.003	9.6	2.7
H28	09.11.2005	<DL		<DL		<DL		<DL		<DL		n.a.		0.007	0.004	0.007	0.004	8.0	2.4

Sample	Sampling date	Activity concentration Bq/l																	
		gross alpha	unc. gross alpha	gross beta	unc. gross beta	H-3	unc. H-3	Ra-226	unc. Ra-226	Ra-228	unc. Ra-228	U-238	unc. U-238	Pb-210	Unc. Pb-210	Po-210	Unc. Po-210	Rn-222	unc. Rn-222
HEIL1	24.08.2005	<DL		<DL		<DL		<DL		<DL		<DL		<DL		<DL		51.3	7.2
HEIL2	24.08.2005	0.09	0.07	<DL		<DL		<DL		<DL		<DL		<DL		<DL		66.6	8.6
HEIL3	24.08.2005	0.29	0.09	<DL		<DL		<DL		n.a.		0.0070	0.0003	<DL		<DL		69.4	8.9
HEIL4	24.08.2005	<DL		<DL		<DL		<DL		n.a.		<DL		n.a.		n.a.		121.0	13.3
HEIL5	24.08.2005	0.19	0.08	<DL		<DL		<DL		<DL		0.0100	0.0005	<DL		<DL		723.2	62.1
HEIL6	26.08.2005	0.30	0.10	<DL		<DL		<DL		<DL		<DL		<DL		<DL		154.1	16.1
HEIL7	08.11.2005	<DL		<DL		<DL		<DL		n.a.		0.0090	0.0005	n.a.		n.a.		9.1	2.6
HEIL8	08.11.2005	<DL		<DL		<DL		<DL		n.a.		0.0080	0.0004	n.a.		n.a.		3.3	1.5
HEIL9	08.11.2005	<DL		<DL		<DL		<DL		n.a.		<DL		n.a.		n.a.		4.9	1.9
HEIL10	09.11.2005	<DL		0.74	0.47	<DL		<DL		n.a.		0.0080	0.0004	n.a.		n.a.		4.3	1.8
HEIL11	09.11.2005	<DL		1.00	0.46	2.10	1.11	0.053	0.040	n.a.		<DL		n.a.		n.a.		19.1	4.0
HEIL12	09.11.2005	<DL		<DL		<DL		<DL		n.a.		<DL		n.a.		n.a.		2.4	1.3
HEIL13	10.11.2005	<DL		<DL		<DL		<DL		n.a.		0.0160	0.0008	n.a.		n.a.		32.2	5.4
HEIL14	22.06.2005	<DL		1.10	0.61	2.00	1.02	<DL		n.a.		0.0014	0.0001	<DL		0.019	0.009	281.3	26.5
HEIL15	05.07.2005	<DL		<DL		1.90	0.99	<DL		<DL		0.0005	0.0002	<DL		0.012	0.002	197.0	19.6
HEIL16	07.07.2005	<DL		<DL		1.40	0.98	<DL		<DL		0.0002	0.0000	<DL		0.004	0.001	85.7	10.3

Sample	Sampling date	Activity concentration Bq/l																	
		gross alpha	unc. gross alpha	gross beta	unc. gross beta	H-3	unc. H-3	Ra-226	unc. Ra-226	Ra-228	unc. Ra-228	U-238	unc. U-238	Pb-210	Unc. Pb-210	Po-210	Unc. Po-210	Rn-222	unc. Rn-222
WVA1	21.06.2005	<DL		<DL		<DL		<DL		0.009	0.005	0.0005	0.0001	<DL		0.017	0.013	44.0	6.6
WVA2	21.06.2005	<DL		<DL		2.20	1.12	<DL		<DL		0.0021	0.0002	0.048	0.011	0.084	0.034	144.3	15.3
WVA3	21.06.2005	<DL		<DL		2.50	1.13	<DL		0.007	0.005	0.0004	0.0000	0.010	0.005	0.011	0.008	28.7	5.0
WVA4	21.06.2005	<DL		<DL		1.50	1.07	<DL		0.012	0.005	0.0007	0.0002	0.021	0.007	0.017	0.005	294.0	27.5
WVA5	21.06.2005	<DL		<DL		3.10	1.12	<DL		n.a.		0.0058	0.0006	0.064	0.013	0.035	0.008	503.2	44.4
WVA6	21.06.2005	<DL		<DL		2.80	1.06	<DL		0.011	0.005	0.0018	0.0002	0.020	0.007	0.014	0.004	281.7	26.5
WVA7	21.06.2005	<DL		<DL		2.70	1.16	<DL		0.007	0.005	0.0011	0.0001	<DL		0.023	0.003	322.5	29.8
WVA8	21.06.2005	<DL		<DL		2.70	1.08	<DL		0.011	0.006	0.0037	0.0004	0.017	0.007	0.020	0.003	174.2	17.8
WVA9	22.06.2005	<DL		<DL		2.10	1.05	<DL		<DL		0.0023	0.0002	<DL		0.010	0.004	68.9	8.8
WVA10	22.06.2005	<DL		<DL		<DL		<DL		<DL		0.0040	0.0004	<DL		0.016	0.007	357.2	32.6
WVA11	22.06.2005	<DL		1.30	0.64	2.60	1.04	<DL		<DL		0.0008	0.0001	n.a.		0.021	0.009	435.8	39.0
WVA12	22.06.2005	<DL		0.82	0.62	1.80	0.99	<DL		0.006	0.004	0.0012	0.0002	0.028	0.010	0.086	0.024	447.8	39.9
WVA13	22.06.2005	<DL		<DL		3.70	1.11	<DL		<DL		0.0052	0.0005	<DL		0.009	0.005	87.2	10.5
WVA14	22.06.2005	<DL		<DL		1.40	0.94	<DL		n.a.		0.0007	0.0001	0.007	0.003	0.009	0.004	136.5	14.6
WVA15	23.06.2005	<DL		1.10	0.65	2.10	0.99	<DL		n.a.		0.0017	0.0004	<DL		0.008	0.004	56.1	7.7
WVA16	05.07.2005	<DL		<DL		2.20	0.97	<DL		n.a.		0.0049	0.0005	n.a.		n.a.		178.7	18.1
WVA17	05.07.2005	<DL		<DL		2.00	1.04	<DL		n.a.		0.0016	0.0002	n.a.		n.a.		111.7	12.6
WVA18	05.07.2005	<DL		<DL		1.70	0.95	<DL		n.a.		0.0014	0.0001	<DL		0.012	0.002	195.5	19.5
WVA19	05.07.2005	<DL		<DL		1.40	0.97	<DL		<DL		0.0014	0.0001	<DL		0.007	0.001	75.8	9.5
WVA20	05.07.2005	<DL		<DL		2.00	0.98	<DL		<DL		0.0026	0.0003	0.008	0.003	0.006	0.001	18.0	3.8
WVA21	05.07.2005	<DL		<DL		<DL		<DL		<DL		0.0057	0.0006	0.007	0.003	0.006	0.001	29.1	5.1
WVA22	05.07.2005	<DL		<DL		2.20	1.06	<DL		<DL		0.0001	0.0001	0.007	0.003	0.002	0.001	<DL	<DL
WVA23	05.07.2005	<DL		<DL		3.40	1.12	<DL		<DL		0.0003	0.0000	0.006	0.002	0.002	0.001	17.7	3.8
WVA24	05.07.2005	<DL		<DL		2.30	1.06	<DL		<DL		0.0002	0.0001	0.007	0.003	0.003	0.002	86.2	10.4
WVA25	05.07.2005	<DL		<DL		2.10	1.05	<DL		<DL		0.0002	0.0000	0.007	0.003	0.001	0.001	29.5	5.1
WVA26	06.07.2005	<DL		<DL		<DL		<DL		n.a.		0.0005	0.0001	n.a.		n.a.		14.0	3.3
WVA27	06.07.2005	<DL		<DL		1.60	0.98	<DL		n.a.		0.0020	0.0002	n.a.		n.a.		31.6	5.3
WVA28	06.07.2005	<DL		<DL		<DL		<DL		n.a.		0.0001	0.0001	n.a.		n.a.		99.6	11.5
WVA29	06.07.2005	<DL		<DL		2.30	1.01	<DL		<DL		0.0019	0.0002	<DL		0.006	0.001	160.9	16.7
WVA30	06.07.2005	<DL		<DL		3.10	1.02	<DL		n.a.		0.0014	0.0001	0.020	0.005	0.002	0.002	50.2	7.1
WVA31	06.07.2005	<DL		0.81	0.62	1.90	0.99	<DL		<DL		0.0008	0.0001	0.067	0.017	0.110	0.041	281.5	26.5
WVA32	06.07.2005	<DL		0.86	0.62	1.20	0.94	<DL		0.008	0.005	0.0008	0.0001	<DL		0.008	0.013	221.7	21.7
WVA33	06.07.2005	<DL		1.00	0.61	1.80	0.99	<DL		0.011	0.004	0.0008	0.0001	0.033	0.008	0.026	0.014	145.6	15.4
WVA34	06.07.2005	<DL		0.91	0.62	1.90	0.99	<DL		<DL		0.0008	0.0001	0.008	0.007	0.027	0.014	123.3	13.5
WVA35	07.07.2005	<DL		<DL		2.70	1.16	<DL		0.009	0.004	0.0048	0.0005	0.023	0.005	0.018	0.003	36.0	5.8
WVA36	07.07.2005	<DL		<DL		1.70	1.11	<DL		<DL		0.0009	0.0001	<DL		0.007	0.002	152.8	16.0
WVA37	07.07.2005	<DL		<DL		2.40	1.08	<DL		n.a.		0.0037	0.0004	<DL		0.011	0.002	170.1	17.4
WVA38	07.07.2005	n.a.		n.a.		n.a.		n.a.		n.a.		n.a.		n.a.		n.a.		n.a.	
WVA39	07.07.2005	<DL		<DL		2.50	1.15	<DL		n.a.		0.0038	0.0004	<DL		0.007	0.001	148.3	15.6
WVA40	24.08.2005	<DL		<DL		<DL		<DL		<DL		<DL		<DL		0.001	0.007	16.8	3.7
WVA41	24.08.2005	<DL		<DL		<DL		<DL		<DL		<DL		<DL		0.022	0.014	38.0	6.0
WVA42	24.08.2005	<DL		<DL		<DL		<DL		<DL		<DL		0.003	0.006	0.018	0.011	11.0	2.9
WVA43	24.08.2005	<DL		<DL		<DL		<DL		<DL		<DL		<DL		0.025	0.015	73.0	9.2
WVA44	24.08.2005	0.07	0.06	<DL		<DL		<DL		<DL		<DL		<DL		0.003	0.009	133.8	14.4
WVA45	24.08.2005	0.17	0.07	<DL		<DL		<DL		<DL		<DL		<DL		0.077	0.027	173.8	17.7
WVA46	24.08.2005	0.07	0.06	<DL		<DL		<DL		n.a.		<DL		0.013	0.009	0.050	0.020	50.8	7.2
WVA47	24.08.2005	0.20	0.07	<DL		<DL		<DL		n.a.		0.0380	0.0019	<DL		0.142	0.054	351.0	32.1
WVA48	24.08.2005	0.08	0.06	0.56	0.43	<DL		<DL		<DL		0.0160	0.0008	<DL		<DL		776.7	66.4
WVA49	24.08.2005	0.25	0.08	<DL		<DL		<DL		n.a.		0.0610	0.0031	<DL		<DL		380.2	34.5
WVA50	24.08.2005	0.17	0.07	<DL		<DL		<DL		0.008	0.005	0.0530	0.0027	<DL		0.037	0.033	543.0	47.6
WVA51	24.08.2005	0.08	0.06	<DL		<DL		<DL		n.a.		0.0270	0.0013	<DL		0.024	0.024	269.6	25.6
WVA52	24.08.2005	n.a.		n.a.		n.a.		n.a.		n.a.		n.a.		n.a.		n.a.		308.1	28.7

Sample	Sampling date	Activity concentration Bq/l																	
		gross alpha	unc. gross alpha	gross beta	unc. gross beta	H-3	unc. H-3	Ra-226	unc. Ra-226	Ra-228	unc. Ra-228	U-238	unc. U-238	Pb-210	Unc. Pb-210	Po-210	Unc. Po-210	Rn-222	unc. Rn-222
H1	20.06.2005	<DL		<DL		2.20	1.06	0.079	0.020	<DL		0.0427	0.0043	0.102	0.018	0.062	0.009	745.2	63.8
H2	20.06.2005	<DL		<DL		1.70	1.02	<DL		0.008	0.005	0.0124	0.0015	0.040	0.007	0.020	0.004	165.2	17.0
H3	20.06.2005	<DL		<DL		1.60	0.99	<DL		0.010	0.005	0.0007	0.0004	0.012	0.006	<DL		75.6	9.4
H4	20.06.2005	<DL		<DL		1.70	1.02	<DL		n.a.		0.0002	0.0000	0.005	0.004	0.003	0.002	15.2	3.5
H5	20.06.2005	<DL		<DL		1.70	0.99	<DL		<DL		0.0003	0.0000	0.014	0.004	0.003	0.002	80.5	9.9
H6	20.06.2005	<DL		1.40	0.64	<DL		<DL		n.a.		0.0005	0.0001	n.a.		n.a.		37.7	5.9
H7	20.06.2005	<DL		1.00	0.61	1.50	0.99	<DL		n.a.		0.0009	0.0003	0.010	0.002	0.017	0.014	53.8	7.5
H8	20.06.2005	<DL		1.10	0.62	<DL		<DL		n.a.		0.0007	0.0001	0.017	0.006	0.056	0.028	118.5	13.1
H9	20.06.2005	<DL		1.40	0.62	<DL		<DL		<DL		0.0022	0.0002	0.058	0.012	0.075	0.030	181.6	18.4
H10	21.06.2005	<DL		<DL		1.90	1.10	<DL		<DL		0.0029	0.0003	0.013	0.004	0.018	0.010	42.7	6.4
H11	21.06.2005	<DL		<DL		1.90	1.08	<DL		<DL		0.0009	0.0001	0.023	0.006	0.034	0.016	72.7	9.2
H12	21.06.2005	<DL		0.97	0.62	1.70	1.07	<DL		<DL		0.0013	0.0001	0.061	0.014	0.119	0.046	530.6	46.6
H13	21.06.2005	<DL		1.10	0.62	1.50	1.10	<DL		<DL		0.0010	0.0006	0.055	0.013	0.100	0.040	278.2	26.3
H14	21.06.2005	0.39	0.12	1.20	0.65	1.90	1.06	<DL		0.022	0.006	0.1678	0.0755	0.062	0.012	0.097	0.035	244.3	23.5
H15	21.06.2005	<DL		<DL		2.10	1.05	<DL		0.008	0.006	0.0016	0.0002	0.018	0.007	0.016	0.004	201.2	20.0
H16	21.06.2005	<DL		<DL		2.20	1.10	<DL		<DL		0.0007	0.0001	<DL		0.008	0.002	129.8	14.1
H17	21.06.2005	<DL		<DL		2.80	1.06	<DL		0.009	0.004	0.0007	0.0001	<DL		<DL		318.4	29.5
H18	21.06.2005	<DL		<DL		1.90	1.03	<DL		<DL		0.0008	0.0001	0.036	0.006	0.046	0.011	101.2	11.7
H19	21.06.2005	<DL		<DL		2.10	1.07	<DL		<DL		0.0007	0.0001	<DL		0.006	0.003	103.1	11.8
H20	22.06.2005	<DL		0.89	0.61	1.20	0.96	<DL		n.a.		0.0050	0.0005	0.021	0.005	0.037	0.011	146.7	15.5
H21	22.06.2005	<DL		1.00	0.59	1.30	0.96	<DL		n.a.		0.0008	0.0001	0.005	0.004	0.064	0.031	230.8	22.4
H22	05.07.2005	<DL		<DL		2.20	0.99	<DL		<DL		0.0009	0.0001	0.046	0.006	0.018	0.003	35.2	5.7
H23	05.07.2005	<DL		<DL		1.80	0.97	<DL		<DL		0.0005	0.0001	0.007	0.006	0.011	0.002	188.9	19.0
H24	05.07.2005	<DL		<DL		<DL		<DL		n.a.		0.0047	0.0005	0.005	0.002	0.003	0.001	19.8	4.0
H25	06.07.2005	<DL		<DL		1.60	1.10	<DL		<DL		0.0001	0.0000	<DL		0.010	0.007	22.4	4.3
H26	06.07.2005	<DL		<DL		1.70	1.09	<DL		0.008	0.005	0.0008	0.0001	<DL		0.023	0.015	86.6	10.4
H27	08.11.2005	<DL		<DL		<DL		0.050	0.040	<DL		<DL		0.004	0.002	0.006	0.003	9.6	2.7
H28	09.11.2005	<DL		<DL		<DL		<DL		<DL		n.a.		0.007	0.004	0.007	0.004	8.0	2.4

A4. pH values, electric conductivities, temperatures

n.a. . . . not analyzed

Sample	Sampling Date	Temperature °C	Electric conductivity µS/cm	pH-value
H1	20.06.2005	13.5	475	n.a.
H2	20.06.2005	14.6	150	n.a.
H3	20.06.2005	13.7	130	n.a.
H4	20.06.2005	13.3	110	n.a.
H5	20.06.2005	11.8	170	n.a.
H6	20.06.2005	11.8	325	n.a.
H7	20.06.2005	14.7	145	n.a.
H8	20.06.2005	12.8	285	n.a.
H9	20.06.2005	11.2	155	n.a.
H10	21.06.2005	12.3	210	n.a.
H11	21.06.2005	11.9	165	n.a.
H12	21.06.2005	11.2	68	n.a.
H13	21.06.2005	9.8	110	n.a.
H14	21.06.2005	10.9	360	n.a.
H15	21.06.2005	15.9	145	n.a.
H16	21.06.2005	7.1	74	n.a.
H17	21.06.2005	7.9	110	n.a.
H18	21.06.2005	7.2	72	n.a.
H19	21.06.2005	10.9	67	n.a.
H20	22.06.2005	9.6	108	n.a.
H21	22.06.2005	13.3	151	n.a.
H22	05.07.2005	n.a.	n.a.	n.a.
H23	05.07.2005	n.a.	n.a.	n.a.
H24	05.07.2005	n.a.	n.a.	n.a.
H25	06.07.2005	16.0	n.a.	n.a.
H26	06.07.2005	10.0	n.a.	n.a.
H27	08.11.2005	6.1	181	8.2
H28	09.11.2005	11.1	693	7.6
HEIL1	24.08.2005	11.2	279	7.7
HEIL2	24.08.2005	11.4	183	6.9
HEIL3	24.08.2005	11.1	479	7.0
HEIL4	24.08.2005	10.0	145	7.3
HEIL5	24.08.2005	10.9	255	6.7
HEIL5	11.01.2006	8.0	287	8.5
HEIL6	26.08.2005	9.2	78	8.1
HEIL7	08.11.2005	10.5	647	7.5
HEIL8	08.11.2005	9.1	643	7.7
HEIL9	08.11.2005	10.8	527	7.7
HEIL10	09.11.2005	9.7	626	7.7
HEIL11	09.11.2005	11.6	888	7.3
HEIL12	09.11.2005	11.0	529	7.9
HEIL13	10.11.2005	11.6	732	7.6
HEIL14	22.06.2005	8.2	138	n.a.
HEIL15	05.07.2005	n.a.	n.a.	n.a.
HEIL16	07.07.2005	n.a.	n.a.	n.a.

Sample	Sampling Date	Temperature °C	Electric conductivity µS/cm	pH-value
WVA1	21.06.2005	9.7	155	n.a.
WVA2	21.06.2005	6.0	140	n.a.
WVA3	21.06.2005	14.0	150	n.a.
WVA4	21.06.2005	9.3	120	n.a.
WVA5	21.06.2005	6.1	59	n.a.
WVA6	21.06.2005	6.6	51	n.a.
WVA7	21.06.2005	6.5	72	n.a.
WVA8	21.06.2005	6.2	100	n.a.
WVA9	22.06.2005	n.a.	n.a.	n.a.
WVA10	22.06.2005	7.5	115	n.a.
WVA11	22.06.2005	6.8	86	n.a.
WVA12	22.06.2005	8.1	81	n.a.
WVA13	22.06.2005	9.6	226	n.a.
WVA14	22.06.2005	7.7	129	n.a.
WVA15	23.06.2005	9.4	152	n.a.
WVA16	05.07.2005	n.a.	n.a.	n.a.
WVA17	05.07.2005	n.a.	n.a.	n.a.
WVA18	05.07.2005	n.a.	n.a.	n.a.
WVA19	05.07.2005	n.a.	n.a.	n.a.
WVA20	05.07.2005	n.a.	n.a.	n.a.
WVA21	05.07.2005	n.a.	n.a.	n.a.
WVA22	05.07.2005	n.a.	n.a.	n.a.
WVA23	05.07.2005	n.a.	n.a.	n.a.
WVA24	05.07.2005	n.a.	n.a.	n.a.
WVA25	05.07.2005	n.a.	n.a.	n.a.
WVA26	06.07.2005	10.6	n.a.	n.a.
WVA27	06.07.2005	8.4	n.a.	n.a.
WVA28	06.07.2005	9.8	n.a.	n.a.
WVA29	06.07.2005	12.8	n.a.	n.a.
WVA30	06.07.2005	11.2	n.a.	n.a.
WVA31	06.07.2005	7.2	n.a.	n.a.
WVA32	06.07.2005	9.1	n.a.	n.a.
WVA33	06.07.2005	8.4	n.a.	n.a.
WVA34	06.07.2005	14.7	n.a.	n.a.
WVA35	07.07.2005	12.6	n.a.	n.a.
WVA36	07.07.2005	7.1	n.a.	n.a.
WVA37	07.07.2005	8.5	n.a.	n.a.
WVA38	07.07.2005	n.a.	n.a.	n.a.
WVA39	07.07.2005	16.4	n.a.	n.a.
WVA40	24.08.2005	11.8	121	7.1
WVA41	24.08.2005	11.7	241	7.7
WVA42	24.08.2005	12.2	115	6.6
WVA43	24.08.2005	10.3	119	6.7
WVA44	24.08.2005	11.1	166	6.6
WVA45	24.08.2005	11.1	136	6.9
WVA46	24.08.2005	10.7	174	7.1
WVA47	24.08.2005	14.1	278	7.9
WVA48	24.08.2005	11.1	206	7.3
WVA49	24.08.2005	11.6	194	7.1
WVA50	24.08.2005	11.8	244	7.6
WVA51	24.08.2005	11.7	264	7.4
WVA52	24.08.2005	11.9	310	7.7

Sample	Sampling Date	Temperature °C	Electric conductivity µS/cm	pH-value
WVA53	24.08.2005	11.4	261	7.4
WVA54	24.08.2005	18.3	310	7.7
WVA55	25.08.2005	9.1	n.a.	8.0
WVA56	25.08.2005	8.5	n.a.	6.6
WVA57	25.08.2005	8.8	n.a.	6.6
WVA58	25.08.2005	8.8	n.a.	6.4
WVA59	25.08.2005	n.a.	n.a.	n.a.
WVA60	25.08.2005	9.5	n.a.	5.6
WVA61	25.08.2005	10.9	75	5.8
WVA62	25.08.2005	11.2	253	6.7
WVA63	25.08.2005	15.6	238	6.6
WVA64	25.08.2005	11.7	411	6.0
WVA65	25.08.2005	11.5	630	6.4
WVA66	25.08.2005	9.7	72	6.4
WVA67	25.08.2005	15.4	248	6.9
WVA68	25.08.2005	13.3	112	6.7
WVA69	25.08.2005	12.7	161	7.0
WVA70	25.08.2005	13.1	155	7.2
WVA71	25.08.2005	14.7	140	6.9
WVA72	25.08.2005	17.2	150	7.4
WVA73	25.08.2005	11.7	220	7.0
WVA74	25.08.2005	15.8	160	7.5
WVA75	25.08.2005	14.6	222	7.1
WVA76	25.08.2005	16.5	155	7.5
WVA77	26.08.2005	14.9	313	7.5
WVA78	26.08.2005	13.6	317	7.4
WVA79	26.08.2005	10.9	76	7.1
WVA80	26.08.2005	10.5	182	7.7
WVA81	26.08.2005	11.9	317	7.6
WVA82	26.08.2005	11.8	324	7.0
WVA83	26.08.2005	13.0	624	7.0
WVA84	26.08.2005	14.0	617	7.1
WVA85	26.08.2005	18.0	167	7.9
WVA86	10.11.2005	8.5	439	8.0
WVA87	10.11.2005	8.6	510	7.8
WVA88	10.11.2005	8.5	492	7.8
WVA89	10.11.2005	10.0	301	8.3
WVA90	10.11.2005	10.0	484	7.8
WVA91	10.11.2005	8.9	390	8.0
WVA92	10.11.2005	10.1	603	7.6
WVA93	08.11.2005	5.1	165	8.1
WVA94	08.11.2005	7.6	184	7.9
WVA95	08.11.2005	9.8	577	7.6
WVA96	08.11.2005	9.7	596	7.8
WVA97	08.11.2005	10.3	296	8.1
WVA98	09.11.2005	7.9	379	8.2
WVA99	09.11.2005	8.7	330	8.1
WVA100	09.11.2005	7.1	317	8.2
WVA101	09.11.2005	9.0	530	7.9
WVA102	09.11.2005	8.9	529	7.7
WVA103	09.03.2006	8.0	287	8.0
WVA104	09.03.2006	6.0	318	8.0
WVA105	09.03.2006	5.0	303	8.0

Curriculum Vitae

Personal Data

Name: Valeria Gruber
Place of birth: Steyr, Austria
Date of birth: 1977-04-23
Citizenship: Austria

Schooling

1983–1987	Primary school
1987–1991	Secondary school
1991–1995	High school for natural sciences

Higher Education

10/2004–03/2009	University of Natural Resources and Applied Life Sciences Vienna Dissertation in the field ”Natural Radioactivity in Drinking Water”
10/1995–06/2004	Diploma study, Technical Physics, University of Technology Vienna Diploma Thesis: ”Untersuchung und Evaluierung der geogenen Radon-Aktivitätskonzentration in eiszeitlich-glazialen Ablagerungen in Oberösterreich”
10/1997–	Diploma study, Astronomy, University of Vienna

Professional Experience

08/2008–	Head of ”Testing Laboratory for Environmental Radioactivity and Radiation Protection”, University of Natural Resources and Applied Life Sciences Vienna
07/2004–	University of Natural Resources and Applied Life Sciences Vienna, Department for Forest- and Soil Sciences, Low-Level Counting Laboratory Arsenal (Scientific staff)
10/2002–06/2004	University of Natural Resources and Applied Life Sciences Vienna, Department for Forest- and Soil Sciences, Low-Level Counting Laboratory Arsenal (part-time employed)
09/2002	Austrian Research Center Seibersdorf, Division for Environmental Research (internship)
08/2000, 09/2001	Office of the Upper Austrian Government, Division for Environmental Protection, Subdivision for Noise- and Radiation Protection (internship)