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Water quality characterization and treatment of stormwater runoff generated from vehicle trafficked surfaces

Dissertation thesis submitted in partial fulfilment of the requirements for the academic degree of **Dr. nat. techn.** (Doctor rerum naturalium technicarum) at the University of Natural Resources and Life Sciences, Vienna.

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Vienna, June 2018

List of Abbreviations

AADT	Average annual daily traffic
ACY	Acenaphthylene
ACE	Acenaphthene
Al	Aluminium
ANT	Anthracene
Ba	Barium
B[a]A	Benzo(a)anthracene
B[a]P	Benzo(a)pyrene
B[b]F	Benzo(b)fluoranthene
B[k]F	Benzo(k)fluoranthene
B[ghi]P	Benzo(g,h,i)perylene
Cd	Cadmium
CHR	Chrysene
Co	Cobalt
Cr	Chromium
Cu	Copper
D[ah]A	Dibenzo(a,h)anthracene
DOC	Dissolved organic carbon
EC	Electrical conductivity
GAC	Granular activated carbon
Fe	Iron
FL	Fluorene
FLR	Fluoranthene
HNO3	Nitric acid
INP	Indeno(1,2,3-c,d)pyrene
Mn	Manganese
Mo	Molybdenum
MOH	mineral oil hydrocarbons
NAP	Naphthalene
Ni	Nickel
NPAHs	Nitro-polycyclic aromatic hydrocarbons
PAHs	Polycyclic aromatic hydrocarbons
Pb	Lead
PHEN	Phenanthrene
Pt	Platinum
PYR	Pyrene
Rb	Rubidium
Sb	Antimony
Sn	Tin
Ti	Titanium
TNb	Total nitrogen bound
TOC	Total organic carbon
TSS	Total suspended solids
V	Vanadium
Zn	Zinc

Dedication

This dissertation is lovingly dedicated to the memory of my late father, Measho Haile and my mother Lemlem Mesele, who always valued education.

Declaration

I hereby declare that the experimental work, results and conclusions reported in this dissertation are entirely my own effort, except where otherwise acknowledged. All used sources of materials and tools are duly acknowledged as well as the institutions involved for assistance are indicated. I also certify that the work contained in this thesis has not been previously submitted for any other award.

Preface

This thesis is based on two research projects (ÖNORM and SARIT) conducted throughout the years 2011 to 2016 at Institute of Sanitary Engineering and Water Pollution Control (SIG), University of Natural Resource and Life Sciences, Vienna (Austria). The projects were supervised by DI. Dr. Ao. Univ. Prof. Maria Fürhacker and I was the principal project employee responsible for designing and conducting laboratory and field experimental work.

The topics addressed in the ÖNORM (Fördervertrag GZ B100121) project focused on stormwater quality characteristics, performance evaluation of stormwater treatment systems and development of a standardized proof method for testing the applicability and utilization of filter media in stormwater filtration systems. Parts of the project results are published in in peer-reviewed journals and are included in this thesis.

The SARIT project has focused on investigating the efficiency of stormwater filtration systems with adsorptive filter media based receiving parking lot and highway runoff. During the course of the project the filtration systems were evaluated after five to seven years in operation regarding hydraulic performance, pollutant accumulation profile and long-term sorption capacity. Furthermore the performance of filter media (Aquafilt®) was tested in laboratory and at field. Parts of the project results are published in in peer-reviewed journals and are included in this thesis.

The research output of this dissertation will hopefully contribute to increased knowledge regarding the negative environmental and genotoxic impacts of stormwater runoff from vehicle trafficked surfaces, and performances of stormwater filtration systems in mitigating the impacts.

Acknowledgement

At this moment of accomplishment, I praise the Almighty God for the wisdom and perseverance that he has been bestowing upon me throughout my life. Several people have contributed towards the successful accomplishment this dissertation thesis. Therefore, I would like to thank all those people who supported me during the last years.

First and foremost I am very pleased to take this opportunity and express my sincere gratitude to Ao. Univ. Prof. DI. Dr. Maria Fürhacker who supervised my dissertation and gave me the opportunity to work as project employee in numerous projects at the Institute of Sanitary Engineering and Water pollution Control (SIG-BOKU). I am highly indebted for her professional guidance, discussions, advice and constructive comments provided to me throughout the completion of this thesis work. I would like to express my gratitude to Ass. Prof. Dipl.-Ing. Dr.nat.techn. Gerhard Kammerer for his scientific advices and valuable suggestions.

Special thanks go to:

- DI. Dr. Rosa Allabashi and staff members of the chemical laboratory at SIG for the analysis of water and sediment samples as well as for the provision of laboratory facilities.
- Prof. Siegfried Knasmüller, Dr. Miroslav Mišık and MSc. Al-Serori Halh from the Institute of Cancer Research, Department of Internal Medicine I, Medical University of Vienna for conducting the toxicity assays and scientific advices.
- Dr. Tamara Grummt from the Section Drinking Water and Swimming Pool Water Toxicology, Federal Environment Agency (UBA), Germany for her help to the toxicity assays.
- The laboratory at Institute of Hydraulics and Rural Water Management (BOKU) for allowing me to conduct physical and chemical analyses soil and sediment samples.
- Birgit Schärfinger for helping in the field and laboratory experimental works.
- All known and anonymous reviewers for their helpful constructive comments.

Finally I would like to thank my family and friends for motivating and supporting me with helping words. Thanks to my parents, brothers and sister for always being there and the outreaching courage you gave me all the time. I am extremely grateful to my partner Selam Yohannes for her understanding, unconditional love and encouragement during the past 7 years. Many thanks go to my best friends Jonita and Zenebe for their unconditional friendship, brotherhood and support no matter what.

Last, but not least, I would like to thank my friends Firew, Gebremeskel (PhD), Miki and Yohannes for your sincere friendship and moral support.

Abstract

Stormwater runoff from vehicle trafficked areas is polluted with a mixture of contaminants that can cause significant contamination of receiving water bodies. Decentralized treatment techniques such as filtration systems that utilize adsorptive filter media have been widely implemented to reduce the impact of stormwater. Physicochemical characterization alone may not sufficiently explain the toxic effects of roadway runoff. Knowledge with regard to cytotoxic and genotoxic impacts are essential to evaluate the performance of stormwater treatment technologies in removing the toxic pollutants. Therefore, the overall objective of this dissertation was to investigate water quality characteristics of parking lot and highway runoff, performance of three stormwater filtration systems, spatial accumulation of sediments and heavy metals as well hydraulic performance of two operational filtration systems, cytotoxic and genotoxic effects of water and sediment samples, suitability of soil and granular filter media for utilization in roadway runoff filtration systems, and operational performance of sedimentation basin.

Three stormwater filtration systems treating parking lot and highway runoff though a layered filter media and one soil filter system were installed at different sites. Stormwater quality characterization was conducted for parking lots runoff monitored for over 18 months (n=11) and also for grab water samples from non-urban highway. The performance of soil filter versus layered filter media treating parking lot and highway runoff were compared in full-scale filtration systems where equal volumes of runoff events were distributed between the two systems. To allow a direct comparison, the filtration system layered technical filter media (A_f: A_{red} = 1:283) was constructed at one corner of a soil filter (A_f: A_{red} = 1:28.3). Both treatment systems were monitored for two years (n= 12). Treatment performance of the filtration systems was evaluated based on effluent water quality, pollutant removal efficiencies, pollutant accumulation profile, and long-term sorption capacity of core filter media mix from in operation systems. Median effluent concentrations were compared to the Austrian groundwater quality ordinance threshold value while removal efficiencies were compared to the ÖNORM B 2506–3 (2016). The removal of pollutants through filtration and sorption processes depends mainly on the chemical composition and structural stability of the filter media. Accordingly, suitability of five different adsorbents [quartz sand (QS), sandy soil (SS) and three different technical filter media (TF–I, TF–II, TF–III)] for removing metals Cu, Pb and Zn from synthetic highway runoff under high hydraulic loads, long-term performance and effect of raod salt (NaCl) application for winter de-icing were evaluated in column flow-through experiments. A method for filter media lifespan assessment was proposed based on the requirements of the Austrian Standard (ÖNORM B 2506–3, 2016) regarding maximum effluent concentrations (Pb ≤ 9 µg/L and Cu ≤ 1800 µg/L) and minimum removal efficiencies (Cu ≥80% and Zn ≥ 50%) for groundwater protection. In this study, filter media was considered as exhausted when one or a combination of the criteria is not met: effluent Pb exceeded 9 µg/L, Cu removal rate fall below 80% and/or Zn removal rate fall below 50%. The column study results were used to predict lifespan of the filtration systems. According to the ÖNORM B 2506–3 (2016) the term “technical filter media, TF” used in this thesis refers to mixed filter media consisting of different mineral adsorbents. Additionally, the performance of a sedimentation basin receiving highway runoff was investigated regarding particle removal, flow distribution, and flow retention time. Particle retention efficiency was evaluated based on mean influent and effluent concentrations of total suspended solids (TSS) measured during seven runoff event. A series of laboratory experiments were conducted using tracer in a lab-scale sedimentation device to study the effect of baffles on sedimentation device performance regarding hydraulic residence time and particle removal efficiency. Finally, cytotoxic and genotoxic effects of both grab water samples from thee sites (parking lot, rural highway, and urban highway runoff) and sediments deposited with two stormwater filtration systems (parking lot and highway runoff) were tested using a combination

of bioassays to evaluate the impacts of stormwater from traffic areas.

The results of the investigation showed that the concentrations of both organic and inorganic contaminants were higher in untreated runoff compared to concentrations measured at the effluent of operational stormwater filtration systems. Performance of parking lot runoff filtration system in operation demonstrated mean removal efficiencies (n =11) of 85% for TSS, > 75 for Cu, Pb and Zn, 91% for ∑16 PAHs, 95% for MOH, 71% for NH4–N and 52% for TOC. Removal of dissolved (< 0.45 μm) and particulate metal species (e.g. Cu and Zn) was very similar, indicating contaminants are removed within the filtration system by filtration, sorption, precipitation and transformation processes. The removal efficiencies of pollutants were varying from event to event, indicating treatment performance depends on rainfall characteristics and influent concentrations of the pollutant. The average pollutant removal efficiencies were generally low for runoff events with low influent concentrations, thus direct comparisons with the minimum removal efficiency requirements (ÖNORM B 2506–3, 2016) is problematic. Nevertheless, conclusions on water quality can be drawn about effluent concentrations that are evident from the monitoring data. Concentrations of all the following contaminants measured at the effluent of operational filtration systems met the Austrian threshold value for groundwater protection: Pb, Cu, Cd, Cr, Ni and ∑5 PAHs (B[a]P, FA, B[b]F, B[k]F B[ghi]P, INP), for all sampling sites. However, median electrical conductivity of effluent samples from all sites exceeded the threshold value for protection of groundwater. While the influent sampling procedure used in this study primarily focused on the first flush, one plausible explanation is that the road salt (e.g. NaCl) applied in winter seasons are readily soluble in stormwater exhibited higher conductivity and chloride concentration in the first flush. Moreover, the installed sedimentation devices are very small so that temporary storage and dilution of chloride of the first flush runoff volume was not optimal. Furthermore, median effluent concentrations of MOHs were partly exceeding the threshold value, which could be attributed to the very high influent concentrations (960– 1100 mg/L) measured during vehicular accidents. In general, treatment performance of soil filter and technical filter media were compared in dual treatment system and the results demonstrated that all effluent concentrations from both system were comparable. The filtration system with technical filter media was very compact (A_f : A_{red} of 1:283), resulting in significant cost savings with respect to disposal of exhausted material and space for installation.

The particle layer accumulated at the top surface of the filtration system was identified as the largest sink of solids, metals and PAHs, which helped to avoid saturation and clogging of the underlying filter media due to deep filtration of sediments or straining of particle within the pores. Concentrations of heavy metals, particularly Cu, Pb, V, and Zn were highest in the surface deposited particle layer, decreasing with increasing depth. This implies that large fractions of the influent metal concentrations were particulate bound and removal of the fine particles plays an important role for mitigation of stormwater. Column experiments with core filter media from operational filtration system showed high metal sorption capacity and the filter media can remain in operation for up to 34 years based on on-site conditions. In-situ infiltration measurements showed that the hydraulic conductivity of the filtration systems has dynamically decreased over the operational period, yet it is acceptable in accordance to the ÖNORM B 2506–3 (2016) up to seven years of operational time. Decrease in near-saturated hydraulic conductivity was primarily due to accumulation of sediments on the filter surface. Scraping and removal of the accumulated sediment layer was considered as major maintenance procedure to recover the hydraulic performance of the system.

Column study results showed that treatment of synthetic highway runoff by soil and technical filter media both met the requirements of the Austrian regulations regarding maximum effluent concentrations and minimum removal efficiencies for groundwater protection. Breakthrough analysis indicated that load removal at filter media exhaustion were > 95 % for Cu and Pb, and

80 – 97% for Zn, while QS showed very poor performance. The adsorption capacity (mg/kg) of each filter media at column exhaustion point towards individual heavy metal varied significantly which is resulted from the variations in influent concentrations and adsorption affinity. For example, sorption capacity Zn at filter media exhaustion was 45, 265, 2390, 4420, 1558 mg/kg for QS, SS, TF-I, TF-II and TF-III respectively. Consequently, adsorption capacity of filter media was found to be in the order of TF-II > TF-I, TF-III > SS > QS. Based on the adsorption capacities, stormwater filtration systems could be sized to 0.4% (TF-I), 1% (TF-I and TF II) and 3.5% (sandy soil) of their impervious catchment area and predicated lifespan of each filter media was at least 35, 36, 41 and 29 years for SS, TF-I, TF-II and TF-III, respectively. In conclusion, technical filter media are potentially suitable alternatives to soil based filters for installation in compact systems, particularly in urban landscapes where space is very limited. Furthermore, road salt (NaCl) application has a minor effect on the mobilization of heavy metals retained by soil and technical filter media, except for natural quartz sand.

Results of toxicity analysis demonstrated that effluents of filtration systems treating parking lot and rural highway runoffs were neither cytotoxic nor genotoxic, while urban highway runoff exposed to higher vehicle traffic emissions showed genotoxic activities. Sediments deposited within the stormwater filtration system induced mutagenic and genotoxic effects. However, the influents were partly cytotoxic indicating that the installed filtration systems are effective in mitigating the effects. The obtained results emphasise the inclusion of chemical speciation of the runoff water and bioassays in the assessment of stormwater quality and performance of a stormwater treatment system.

The sedimentation basin treating highway runoff prior to its draining into soil infiltration basin showed very poor performance with respect to particle removal and hydraulic conditions. Operation and monitoring of the basin showed that the inflow was not uniformly distributed across the cross-sectional area of the basin and the turbulence conditions resulting from high flow rate gives rise to backflow currents and side eddies. Turbulent flow conditions and mixing of the inflow within the basin that have arisen from poorly inlet and outlet design structures did have a marked influence on particle settling efficiency. The average TSS concentrations measured at the inlet and outlet section of the basin were 89 mg/L and 94 mg/L, respectively. Accordingly, our results confirmed that no removal of settleable solids was observed during the monitoring period indicating all the particle load is transported to the filtration basin. Results of tracer tests in a pilot-scale sedimentation basin show that installation of baffles led to an improved flow distribution across the entire section of the basin, decreased short-circuiting, and increased mean residence time by at least 400%. The inclusion of baffles forces the solids to move faster towards the bottom of the basin and decreases the inlet recirculation zone, thus yielding significantly enhanced settling of particles. The removal efficiency of solids increased from mean particle diameter of 130 µm without baffles to 90 µm when using baffles. Therefore it can be concluded that laboratory simulations are suitable for determining the efficiency of a sedimentation basin unit where conditions and variables can be controlled as much as possible and measurement of the results can be done more accurately.

This study has contributed to an increased knowledge of the cytotoxic and genotoxic impacts of parking lot and roadway runoff, and treatment of stormwater applying filtration systems with adsorptive filter media. Information about pollutant concentration levels and biological effects can then be integrated qualitatively and/or quantitatively for a holistic assessment of filtration systems performance in reducing toxic pollutants. Results of the field investigations together with column breakthrough data can be applied in future design and construction of treatment systems.

Key words: Stormwater, Filtration systems, Filter media, Adsorption capacity, Lifespan, Cytotoxicity, Genotoxicity, Sedimentation.

Kurzfassung

Der Abfluss von Straßen und Parkplätzen ist mit einer Mischung von Schadstoffen verunreinigt, die zu einer erheblichen Verschlechterung der Qualität des aufnehmenden Gewässers führen können. Um die negativen Auswirkungen von Regenwasser auf die Umwelt zu reduzieren werden, dezentrale Behandlungstechniken, wie Filtrationssysteme mit adsorptiven Filtermedien verwendet. Die physikalisch-chemische Charakterisierung alleine kann die toxischen Effekte von Straßenabfluss nicht ausreichend beschreiben. Kenntnisse in Bezug auf zytotoxische und genotoxische Wirkungen sind wesentlich, um die Leistung von Regenwasserbehandlungstechnologien bei der Entfernung der toxischen Schadstoffe zu bewerten. Das Gesamtziel dieser Dissertation war daher die Untersuchung der Wasserqualität in Abflüssen von Straßen und Parkplätzen und der Leistung von drei Regenwasserfiltrationssystemen, die Akkumulation von Sedimenten und Schwermetallen sowie die hydraulische Leistung zweier funktioneller Filtersysteme, zytotoxische und genotoxische Effekte von Wasser- und Sedimentproben, der Eignung von Boden- und körnigen Filtermedien zur Verwendung in Straßenabwasserbehandlungsanlagen und der Betriebsleistung von Sedimentationsbecken.

Es wurden drei Regenwasserfiltersysteme, die das Abwasser von Parkplätzen und Autobahnabflüssen behandeln, mit geschichteten Adsorptionsfiltermedien bzw. einem Bodenfiltersystem an verschiedenen Standorten installiert. Die Charakterisierung der Wasserqualität der Niederschlagsabflüsse wurde für Parkplätze und den damit verbundenen Autobahnabflüssen durchgeführt, die zum Teil mehr als 18 Monate lang überwacht wurden (n = 11). Außerdem wurden Wasserproben von nichtstädtischen Straßen entnommen. Die Leistung eines Bodenfilters wurde mit einem geschichteten technischen Filtermedium in einem Full-Scale-Filtersystem verglichen, bei dem gleiche Volumina von Abflussereignissen zwischen den beiden Systemen aufgeteilt werden. Um einen direkten Vergleich zu ermöglichen, wurde ein Filtersystem mit technischem Filtermaterial (Af: Ared = 1: 283) an einer Ecke eines Bodenfilters (Af: Ared = 1: 28,3) errichtet. Beide Behandlungssysteme wurden über einem Zeitraum von zwei Jahren überwacht (n = 12). Die Behandlungsleistung der Filtrationssysteme wurde auf Grundlage der Qualität des Abwassers, der Schadstoffentfernung, des Schadstoffakkumulationsprofils und der langfristigen Sorptionskapazität des in der Großanlagen verwendeten Filtermediengemisches bewertet. Die medianen Abwasserkonzentrationen wurden mit den Grenzwerten der österreichischen Grundwasserqualitätsverordnung verglichen, die Abscheidegrade mit der ÖNORM B 2506-3 (2016). Die Entfernung von Schadstoffen durch Filtrations- und Sorptionsprozesse hängt hauptsächlich von der chemischen Zusammensetzung und Struktur der adsorptiven Filtermedien ab. Die Anwendbarkeit von technischen Filtermaterialien, deren Langzeitleistung in Bezug auf die Entfernung von Schwermetallen und die Auswirkungen der Anwendung von Auftausalz (NaCl) wurden anhand von fünf verschiedenen Adsorbentien [Quarzsand (QS), Sandboden (SS) und drei verschiedenen technischen Filtermedien (TF I, TFII, TF III)] zur Entfernung von Metallen (Cu, Pb und Zn) aus synthetischem Autobahnabfluss unter hohen hydraulischen Belastungen in Säulen im Labor bewertet. Es wurde eine Methode zur Beurteilung der Standzeit von Filtermedien basierend auf den Anforderungen der österreichischen Vorschriften für den Grundwasserschutz (zulässige Ablaufkonzentrationen: Pb $\leq$ 9 µg/L und Cu $\leq$ 1800 µg/L und minimale Abscheidegrade Cu $\geq$80% und Zn $\geq$50%) vorgeschlagen. Die Ergebnisse der Säulenversuche wurden verwendet, um die Standzeit der Filtrationssysteme vorherzusagen. Gemäß der ÖNORM B 2506-3 (2016) bezieht sich der in dieser Arbeit verwendete Begriff "technische Filtermedien, TF" auf gemischte Filtermedien, die aus verschiedenen mineralischen Adsorbentien bestehen. Des Weiteren wurde die Leistung eines großtechnischen Sedimentationsbeckens, das den Abfluss einer Autobahn aufnimmt, hinsichtlich der Partikelentfernung, der Gleichmäßigkeit der Strömungsverteilung und der

Retentionszeit untersucht. Die Partikelretentionseffizienz wurde direkt basierend auf den mittleren Zulauf- und Ablaufkonzentrationen der abfiltrierbaren Stoffe (AFS) untersucht. Eine Reihe von Experimenten wurde unter Verwendung von Tracern in einem Prototyp einer Sedimentationsvorrichtung im Labormaßstab durchgeführt, um auf die Wirkung von Prallflächen auf die Sedimentationsbeckenleistung bezüglich der hydraulischen Verweilzeit und der Partikelentfernungseffizienz des Sedimentationsbeckens im vollen Maßstab zu schließen. Zusätzlich zu den chemischen Analysen wurden zytotoxische und genotoxische Effekte von Wasserproben von verschiedenen Standorten (Parkplatz, Landstraße und Stadtautobahnabfluss) und den Sedimenten, die in zwei Regenwasserfiltrationssystemen (Parkplatz und Autobahnabfluss) abgelagert wurden, mittels einer Kombination von Bioassays getestet.

Im Allgemeinen waren die Konzentrationen von sowohl der organischen als auch anorganischen Verunreinigungen im unbehandelten Abfluss höher als die Konzentrationen, die am Abfluss von betrieblichen Regenwasserfiltrationssystemen gemessen wurden. Die Auswertung der Regenwasserfiltrationssysteme im Betrieb ergab mittlere Abscheidegrade von 85% für AFS,> 73% für Cu, Pb und Zn, 83% für Σ16 PAK, 95% für MOH, 71% für NH_4-N und 52%. Für die Entfernung von gelösten (<0,45 µm) und teilchenförmigen Metallspezies (z. B. Cu und Zn) war sehr ähnlich, was darauf hinweist, dass Verunreinigungen innerhalb des Filtrationssystems durch Filtration, Sorption, Fällung und Transformationsprozesse entfernt werden. Die Entfernungseffizienz der Schadstoffe variierte von Ereignis zu Ereignis, was darauf hindeutet, dass die Behandlungsleistung von den Niederschlagseigenschaften, den Zulaufkonzentrationen und der Phase des Schadstoffs abhängt. Die mittleren Schadstoffabscheidegrade waren für Abflussereignisse mit geringen Zulaufkonzentrationen im Allgemeinen gering, so dass direkte Vergleiche mit den Mindestanforderungen an die Abscheideeffizienz (ÖNORM B 2506-3, 2016) problematisch sind. Dennoch können Rückschlüsse auf die Qualität der Abflusskonzentrationen aus den Überwachungsdaten gezogen werden. Die Konzentrationen der folgenden Spurenstoffe, gemessen im Abfluss betrieblicher Filtersysteme, erfüllten die österreichischen Vorgaben für den Grundwasserschutz: Pb, Cu, Cd, Cr, Ni und Σ5 PAK (B [a] P, FA, B [b] F , B [k] FB [ghi] P, INP), für alle Probenahmestellen. Der Median drn elektrischen Leitfähigkeit der Abwasserproben aller Standorte überschritt jedoch den Schwellenwert für den Schutz des Grundwassers. Da sich das in dieser Studie verwendete Probenahmeverfahren hauptsächlich auf den ersten Spülstoß konzentrierter, ist eine plausible Erklärung, dass das Streusalz (z. B. NaCl), das im Winter angewendet wird, in Regenwasser leicht löslich ist und eine höhere Leitfähigkeit und Chloridkonzentration im ersten Spülstoß aufweist. Darüber hinaus sind die installierten Sedimentationsvorrichtungen sehr klein, so dass die temporäre Speicherung des ersten Spülabflussvolumens und die Verdünnung von Chlorid nicht optimal war. Darüber hinaus übertrafen die medianen Abflusskonzentrationen von MOHs teilweise den Schwellenwert, was auf sehr hohe Zuflusskonzentrationen (960–1.100 mg/L), die bei Fahrzeugunfällen gemessen wurden, zurückzuführen war.

Die an der Filteroberfläche an einer Vorfiltermatte angesammelte Partikelschicht wurde als die größte Senke von Feststoffen, Metallen und PAKs identifiziert, die dazu beitrug, eine Sättigung und Verstopfung der darunter liegenden Filtermedien oder der Belastung von Partikeln innerhalb der Poren aufgrund der Tiefenfiltration von Sedimenten zu vermeiden. Die Konzentrationen von Schwermetallen, insbesondere Cu, Pb, V und Zn, sind in der oberflächlich abgeschiedenen Partikelschicht am höchsten und nehmen mit zunehmender Tiefe ab. Dies bedeutet, dass große Anteile der zufließenden Metallkonzentrationen partikulär gebunden waren und die Entfernung der feinen Partikel eine wichtige Rolle bei der Behandlung von Regenwasser spielt. Säulenexperimente mit Filtermedien aus Anlage die 7 Jahren im Betrieb waren, zeigten eine hohe Metallsorptionskapazität, und die Filtermedien können basierend auf den Bedingungen vor Ort bis zu 34 Jahre in Betrieb bleiben. In-situ-Infiltrationsmessungen zeigten, dass sich die hydraulische Leitfähigkeit der Filtersysteme über die Betriebsdauer dynamisch verringert hat, jedoch gemäß der Anforderung der ÖNORM B 2506–3 (2016) nach 7 Jahreigen Betrieb noch akzeptabel ist. Die Abnahme der (nahezu gesättigten) hydraulischen Leitfähigkeit war hauptsächlich auf die Oberflächenakkumulation von Sedimenten zurückzuführen. Das Entfernen der an der Oberfläche angesammelten Sedimentschicht wurde als Hauptwartungsverfahren betrachtet, um die hydraulische Leistungsfähigkeit des Systems wiederherzustellen.

Ergebnisse von Säulenuntersuchungen zeigten, dass die Behandlung von synthetischem Autobahnabfluss durch Boden und technische Filtermedien beide den Anforderungen der österreichischen Vorschriften hinsichtlich maximaler Abwasserkonzentrationen und minimalen Schadstoffrückhalt für den Grundwasserschutz entspricht. Die Durchbruchsanalyse zeigte, dass der maximale Frachtrückhalt bei vollständige Ausschöpfung des Filters > 95% für Cu und Pb und 80 bis 97% für Zn waren. Die Adsorptionskapazität (mg/kg) für ein einzelnes Schwermetall variiert zwischen den Filtersystemen signifikant. Zum Beispiel betrug die Sorptionskapazität Zn bei einer Filtermedienerschöpfung 45, 265, 2390, 4420, 1558 mg / kg für QS, SS, TF-I, TF-II bzw. TF-III. Folglich wurde gefunden, dass die Adsorptionskapazität von Filtermedien in der Reihenfolge TF-II> TF-I, TF-III> SS> QS liegt. Basierend auf den Adsorptionskapazitäten konnte die Filterfläche für Regenwasserfiltersysteme auf 0,4% (TF-I), 1% (TF-I und TF II) und 3,5% (Sandboden) der Fläche ihres Einzugsgebiets bemessen werden. Die vorhergesagte Standzeit jedes Filtermediums war mindestens 35, 36, 41 und 29 Jahre für SS, TF-I, TF-II bzw. TF-III. Abschließend ist zu sagen, dass technische Filtermedien potentiell geeignete Alternativen zu bodenbasierten Filtern für die Installation in kompakten Systemen sind, insbesondere in Stadtlandschaften, in denen der Platz sehr begrenzt ist. Darüber hinaus hat die Enteisungsbehandlung (NaCl) nur eine geringe Auswirkung auf die Mobilisierung von Schwermetallen, die von Böden und technischen Filtermedien zurückgehalten werden, mit Ausnahme von natürlichem Quarzsand.

Die Ergebnisse der Toxizitätsanalyse zeigten, dass Abwässer von Filtrationssystemen, die Abflüsse von Parkplätze und Autobahn behandeln, weder zytotoxische noch genotoxische Effekte hatten, während städtische Autobahnabflüsse, die höheren Fahrzeugemissionen ausgesetzt waren, genotoxische Wirkungen zeigten. Sedimente, die im Regenwasserfiltrationssystem abgelagert wurden, verursachten mutagene und genotoxische Effekte. Die Zuflüsse waren jedoch sogar teilweise zytotoxisch, was darauf hinweist, dass die installierten Filtrationssysteme wirksam sind, um die Auswirkungen zu mildern. Die erhaltenen Ergebnisse betonen die Wichtigkeit der Einbeziehung der chemischen Speziation des Abflusswassers und der Bioassays in die Bewertung der Abflussqualität und der Behandlungsleistung eines Regenwasserbehandlungssystems.

Das untersuchte Sedimentationsbecken, das den Autobahnabfluss behandelte, bevor letzterer in das Bodeninfiltrationsbecken abgelassen wurde, zeigte eine sehr schlechte Leistung in Bezug auf Partikelentfernung und hydraulische Bedingungen. Die konstruktive Gestaltung und der Betrieb des Beckens brachten mit sich, dass die Abflussströmung nicht gleichmäßig über die Querschnittsfläche des Sedimentationsbeckens verteilt war und das Turbulenzverhalten aus der hohen Strömungsrate resultierten, führten zu Rückströmungen und Seitenwirbeln, die das Absetzen der Teilchen beeinflussten. Dementsprechend betrugen die durchschnittlichen Konzentrationen an AFS, die am Einlass und am Auslass (des Abschnitts) des Beckens gemessen wurden, 89 mg/L bzw. 94 mg/L, was anzeigt, dass die gesamte Partikelladung in das Filtrationsbecken transportiert wurde. Die Ergebnisse der Tracer-Tests in einem Prototyp-Sedimentationsbecken im Pilotmaßstab zeigen, dass die Installation von Prallblechen zu einer verbesserten Strömungsverteilung über den gesamten Abschnitt des Beckens, zu einem verringerten Kurzschluss und einer erhöhten mittleren Verweilzeit um mindestens 400% führte.

Als Folge konnten mittelgroße bis feine Partikel leicht abgesetzt werden. Der Einbau von Prallflächen zwingt die Feststoffe dazu, sich schneller zum Boden des Beckens zu bewegen und verringert am Einlass Rezirkulationszonen, was zu einem deutlich verbesserten Absetzen von Partikeln führt. Die Abscheidung stieg von mittleren Partikeldurchmessern von 130 µm ohne Prallbleche auf 90 µm bei Verwendung von Prallblechen. Daraus kann gefolgert werden, dass Laborsimulationen geeignet sind, die Wirkungsgrade der Sedimentationsbeckeneinheit zu bestimmen, wo Bedingungen und Variable so weit wie möglich kontrolliert werden können und die Messung der Ergebnisse genauer durchgeführt werden kann.

Diese Arbeit hat zu einem besseren Verständnis der zytotoxischen und genotoxischen Auswirkungen von Straßenabfluss und der Behandlung von Regenwasser mit Filtrationssystemen mit adsorptiven Filtermedien beigetragen. Informationen über Schadstoffkonzentrationen und biologische Wirkungen können dann qualitativ und / oder quantitativ integriert werden, um eine ganzheitliche Beurteilung der Effizienz von Filtrationssystemen bei der Behandlung von Regenwasserabflüssen zu ermöglichen. Die Ergebnisse der Felduntersuchungen zusammen mit den Daten zum Säulendurchbruch können für die zukünftige Planung und Konstruktion von Behandlungssystemen verwendet werden. Um schließlich die Wirksamkeit einer Regenwasserbehandlungsanlage hinsichtlich der Entfernung toxischer Schadstoffe genau zu bewerten, sollten chemische Analysen und Biotests als ergänzende Methoden verwendet werden.

Schlagworte: Regenwasser, Filtrationssysteme, Filtermedien, Adsorptionskapazität, Standzeit, Zytotoxizität, Genotoxizität, Sedimentation.

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List of publications papers related to this thesis

The doctoral thesis is based on four scientific paper published in peer-reviewed journals (Papers I, II, III, V and VII), a Manuscript (Paper VI) and a conference paper (Paper IV). In the thesis, these papers are referred to with the roman numbers in bold font (e.g. paper **I**).

I. Fuerhacker M., **Haile, T.M**., Monai, B., Mentler, A. (2011). Performance of a filtration system equipped with filter media for parking lot runoff treatment. Desalination 275 (1–3), 118–125.

II. **Haile, T.M.**, Hobiger, G., Kammerer, G., Allabashi, R., Schaerfinger, B., Fuerhacker, M. (2016). Hydraulic performance and pollutant concentration profile in a stormwater runoff filtration systems. Water Air Soil. Pollut. 227:34. DOI 10.1007/s11270-015-2736-4**.**

III. **Haile, T.M.** and Fürhacker, M. (2015). Prüfmethode zur Filterwirkung und –eignung. In ÖWAV Veranstaltung Versickerung von Niederschlagswässern: ÖWAV-Regelblatt 45 Rahmenbedingungen, Bemessung und Betrieb von Versickerungsanlagen Gewässerschutzanlagen für Verkehrsflächen. ÖWAV: ISBN: 978-3-902978-62-2.

IV. **Haile, T.M.**, Mišík, M., Grummt, T., Halh, A.S, Pichler, C., Knasmueller, S., Fuerhacker, M. (2016). Cytotoxic and mutagenic activities of waters and sediments from highway and parking lot runoffs. Water Sci. Technol. 73(11), 2772 – 2780.

V. **Haile, T.M.** and Fürhacker, M. (2017). Filtermaterialprüfung: Anwendung der ÖNORM B 2506 Teil 3 für das hochrangige Straßennetz. Österreichische Wasser- und Abfallwirtschaft, 1613-7566, 1-8; ISSN 0945-358X.

VI. **Haile, T.M.** and Fuerhacker, M. (2016). Simultaneous removal of heavy metals from roadway stormwater runoff using different filter media in column studies. (Submitted to Water, mdpi journal).

VII. **Haile, T.M.**, Kammerer, G., Fürhacker, M. (2014). Probleme bei Planung und Betrieb von Absetzbecken für Straßenabwässer. Österreichische Wasser- und Abfallwirtschaft 66, 112-119; ISSN 0945-358X.

Furthermore, the results were presented and discussed in international scientific conferences and national seminar in Austria (ÖWAV and ASI) as follows.

1) Haile, T.M. Mišík, M., Grummt, T., Knasmueller, S., Fuerhacker, M. (2017). Micropollutants and genotoxicity in sediments from stormwater filtration systems. [Micropol & Ecohazard Conference 2017, Vienna, Austria, Sept. 17-20, 2017]. [Micropol & Ecohazard Conference 2017, 10th IWA Specialized Conference on ACHSW, 17-20 Sept. 2017, Vienna, Austria]

2) Haile, T.M., Mišík, M., Grummt, T., Halh, A.S, Pichler, C., Knasmueller, S., Fuerhacker, M. (2015). Cytotoxic and mutagenic activities of waters and sediments from highway and

parking lot runoffs. [Micropol & Ecohazard Conference 2015, 9th IWA Specialist Conference on ACHMSW, 22–25 Nov. 2015, Singapore].

3) Haile, T.M. and Fuerhacker, M. (2013). Heavy metal and PAH concentrations in water, sediment and filter media from street runoff treatment systems. [Poster]. [Proceedings of the 8th IWA Specialized conference on ACHSW: Micropol & Ecohazard 2013, Zurich, Switzerland, 16–20 June, 2013].

4) Haile, T.M. and Fürhacker, M. (2012). Forschungsergebnisse zur Reinigung von Straßenabwässern. In ÖWAV Seminar "Veranstaltung Gewässerschutzanlagen für Verkehrsflächen", 28 – 29 Nov. 2012, Vienna, Austria. ISBN: 978-3-902810-58-8.

5) Fuerhacker, M. and Haile, T.M (2012). Assessment of particle removal efficiency by filter media used in stormwater treatment systems. [Poster]. In: IWA World Water Congress, Busan, South Korea, 16–20 Sept., 2012.

1. Introduction

1.1 Background

Impervious surfaces inhibit the natural infiltration of rainwater into the ground causing stormwater runoff, which in turn might lead to multiple negative hydrologic and ecological impacts for receiving waters (Hatt et al., 2008; Kayhanian et al., 2012a; Kayhanian et al., 2012b; McQueen et al., 2010). Highways, driveways, parking lots and rooftops comprise the bulk of impervious surfaces within a watershed (Barnes et al., 2002). The pollutants accumulated on the paved surface are washed off during rainfall runoff and transported with the stormwater to receiving water.

Stormwater runoff from vehicle trafficked surface often contain elevated concentrations of inorganic and organic pollutants, nutrients, and solids that an impact the health of receiving waters and are susceptible to induce genotoxic effects to the aquatic life and human health. These pollutant constituents are present either in the dissolved phase associated with the runoff water or in the particulate phase bound to particles. Therefore, on site treatment technologies of stormwater aimed at effective removal of both dissolved and particulate pollutants are of particular interest to protect the aquatic environment in the receiving water.

Structural best management practices (BMPs) are widely used to reduce the hydrologic and water quality impacts on receiving water bodies. However, their pollutant removal performance is highly variable depending on system type, design specifications, local hydrologic and climatic conditions, filter media type, and system age (Scholes et al., 2008). BMPs utilising filtration process is one of the most promising technologies for the removal of both dissolved and particulate pollutants providing that effective filtration media is used (Fuerhacker et al., 2011; Genc-Fuhrman et al., 2007; Reddy et al., 2014). Hence, the stormwater from vehicle trafficked areas contains a mixture multiple contaminants a single filter media in not capable of removing all of the runoff pollution constituents (Reddy et al., 2014; Seelsaen et al., 2006). Therefore, the filter media composition plays an important role regarding pollutant removal capacity and efficiency. Studies have shown that filtration systems that utilize layered or combination of different filter media are effective for the simultaneous removal of multiple pollutants from stormwater (Fuerhacker et al., 2011; Pawluk and Fronczyk, 2014; Reddy et al., 2014). As the stormwater percolates through the filter media pollutants are removed by processes such as filtration, sorption, surface complexation, precipitation and transformation (Davis et al., 2001; Grebel et al., 2013; Pawluk and Fronczyk, 2014; Reddy et al., 2014). Previous investigations concerning performance of stormwater treatment facilities focused mainly on removal of pollutant concentrations or loads (Hatt et al., 2009; Hilliges et al., 2013; Li and Davis, 2008). Several studies have also investigated the ecotoxicological effects of highway and parking lot runoffs (Grapentine et al., 2008; Karlsson et al., 2010; McQueen et al., 2010), however only limited research have been conducted on the cytotoxic and genotoxic effects of highway and parking lot runoff. The toxicity results could be essential to evaluate performance of the installed stormwater treatment system.

Performance of stormwater filtration systems to fulfil the requirements regarding minimum removal efficiencies and threshold value for groundwater protection (ÖNORM B 2506, 2016; BMLFUW, 2010) is the subject of research in this dissertation. The key research issues addressed are performance evaluation of full-scale filtration systems and sedimentation basin, cytotoxic and genotoxic effects of water and sediments, suitability soil and mineral based filter media for application in filtration systems treating stormwater from vehicle trafficked areas.

1.2 Research hypotheses and objectives

Research hypotheses

The following hypotheses have been identified for investigation based on literature review:

- ✓ Stormwater runoff from vehicle trafficked areas contains a complex mixture of environmental contaminants which may induce cytotoxic and genotoxic effects.
- ✓ Stormwater pollutants, particularly heavy metals and PAHs are strongly associated with fine particles (<63µm). Therefore, retention of these particles in stormwater filtration systems could significantly improve the effluent water quality.
- ✓ Stormwater filtration systems that incorporate adsorptive filter media can effectively remove the typical parking lot and highway runoff pollutants to meet the requirements for groundwater protection (e.g. ÖNORM B 2506–3, 2016).
- ✓ The hydraulic performance of stormwater filtration systems diminish over service time due to surface deposition of sediments and straining particles within the filter pores. It is hypothesized that the lifespan of a stormwater filtration system is frequently limited by hydraulic performance or clogging.
- ✓ Heavy metals retention capacity of filter media may be significantly reduced over time. The lifespan of filter media in terms of pollutant removal capacity can be simulated in column experiments.
- ✓ The design of sedimentation basin has significant impacts on stormwater flow distribution and particle removal efficiency.

Objectives of the study

The objective of this work was to provide an overall assessment of the water quality characteristics, cytotoxic/genotoxic activities, and treatment performance of stormwater filtration systems. This was carried out both in laboratory and field studies. Based on the aforementioned hypotheses, the main research objectives of this thesis were:

(1) Characterization of parking lot and highway runoff water quality and pollutant removal performance of stormwater filtration systems [Paper **I, II, III and III**].
(2) To assess the hydraulic performance and long-term pollutant removal capacity of two stormwater filtration systems after five to seven years of service time [Paper **II**].
(3) To evaluate the cytotoxic and genotoxic effects of waters and sediments from sites impacted by runoff from highway and parking lots [Paper **IV**]
(4) To assess applicability of the Austrian Standard Method (ÖNORM B 2506–3, 2016) filter materials test procedures for road runoffs with high annual average daily traffic (AADT) [Paper **V**].
(5) To investigate the feasibility, sorption capacity and long-term treatment performance of different filter media types in laboratory column experiments for further use in stormwater filtration systems [Paper **VI**].
(6) To evaluate performance of a full scale sedimentation basin in terms of particle removal, and pilot scale simulation using tracers for design optimization [Paper **VII**].

Each objective is fully addressed in the independent scientific papers as indicated in brackets.

1.3 Outline of the dissertation

This cumulative dissertation consists of 6 chapters. A general introduction, research hypothesis and objectives of this study are presented in chapter 1. Chapter 2 presents a review of literature on the state of the art concerning stormwater quality characteristics, toxicity assessment methods and performance of different treatment technologies. An overview of the materials and methods is presented in Chapter 3. It provides a detail information on site characteristics, sampling strategies and analytical methods within which the published separate papers – are couched. The laboratory and field test methods are explained in this chapter. Chapter 4 presents a summary of results published in peer-reviewed journals and a manuscript, which are the central parts of this cumulative doctoral thesis. Chapter 5 draws the conclusions based upon the main findings of the presented thesis work and discusses recommendations for future research. Chapter 6 comprises seven research articles presenting the results and discussions of the investigation. Three articles were published in scientific peer-reviewed journals listed in the Science Citation Index, two articles were published in the Austrian Water and Waste Association journal (ÖWAV), one conference paper, and one manuscript is submitted for publication in Water, Air, & Soil Pollution journal. This chapter represent the central part of the doctoral thesis while each research article addressing the raised research hypotheses independently.

2. State of the art

2.1 An introduction to stormwater

Stormwater runoff from vehicle trafficked areas (highway, major roads, feeder streets, bridges and car parking areas) is recognized as a critical non-point source of pollution (Davis and Birch, 2010; Göbel et al., 2007). Stormwater generated from traffic areas carries a heterogeneous mixture of pollutants which includes particles, heavy metals, polycyclic aromatic hydrocarbons (PAHs), nitro polycyclic aromatic hydrocarbons (NPAHs), mineral oil hydrocarbons (MOH), nutrients and de-icing salts (Fuerhacker et al., 2011; Göbel et al., 2007; Helmreich et al., 2010; Kayhanian et al., 2012a; Kayhanian et al., 2012b; Shinya et al., 2000). Pollutants accumulated on road surfaces are washed off the surface with runoff water during rainfall events and enter the receiving waters (Brown and Peake, 2006; Gnecco et al., 2005; Helmreich et al., 2010; Opher et al., 2009). Vehicular emission, road wear, atmospheric deposition (wet and dry), road maintenance, the rainfall itself and surrounding land use have been found as most influential sources of pollutants of stormwater runoff from vehicle trafficked areas (Davis et al., 2001; Fürhacker et al., 2013; Göbel et al., 2007; Hemreich et al., 2010; Opher et al., 2009). The typical traffic-derived stormwater runoff pollutants and their major source are summarized in Table 1.

Table 1. Main sources of road runoff pollutants directly related to traffic

Pollutant	Source
N and P	atmosphere, roadside fertilizer application and sediments
Ba	brake pads and linings
Cd	tyre wear, brake pads, lubricating oils, combustion and corrosion
Co	wear of studded tyres, corrosion of bushings, brake wires, and radiators
Cr	moving engine parts, brake linings, asphalt and pavement wear, corrosion of welded metal plating, and paints
Cu	bearing and bushing wear, moving engine parts, brake linings, tyre wear, asphalt and pavement wear, and lubricating oils.
Ni	automobile emission, lubricating oil, corrosion body parts, brake linings, asphalt and pavement wear, and wear of moving parts in engines, catalysts
Pb	vehicle exhaust, tyre wear, lubricating oils, grease, brake linings, bearing wear, asphalt and pavement wear, and wear of moving parts in engines
Pt	Catalysts
Sb	brake pads and linings
Sr	brake pads and linings
Ti	brake pads and linings
V	tyre wear, asphalt and pavement wear
Zn	tyre wear, motor oil, grease, brake pads, asphalt and pavement wear, and lubricating oils
PAHs	tyre wear, vehicle exhaust, asphalt wear/road aging
MOH	motor oil spills , vehicle exhaust, fuels and anti-freeze, volatilization loss

Source: Ball, 1998; Legret and Pagotto, 1999; Davis et al., 2001; McKenzie et al., 2009; Thorpe and Harrison, 2008; Zafra et al., 2011.

Under certain conditions pollutants in highway runoff may exert acute or chronic impact on the chemical and ecological status of the receiving water and soils (Crabtree et al., 2006; Opher et al., 2009). Among the pollutant constituents, heavy metals, PAHs and NPAHs contribute to the toxicity of receiving water bodies to an extent that can be detrimental to aquatic organisms (Kayhanian et al., 2008; Murakami et al., 2008; Shinya et al., 2000; Wium-Andersen et al., 2011). In a recent study in Austria, Clara et al. (2014) reported that that concentrations of heavy metals, PAH`s and other organic pollutants such as di(2-ethylhexyl)phthalate, octylphenol, and bisphenol A present in road runoff were higher as compared to effluents from wastewater treatment plants. Although roads, highways and parking lots may occupy only 20% of urban catchment areas, their drainage water can contribute 50% of the total suspended solids, 30% of total hydrocarbons and 35–75% of heavy metals discharged directly to receiving streams (Scholes et al., 1998).

Numerous efforts have been made to investigate the relationships between stormwater runoff pollution and rainfall characteristics for various catchment areas, such as highways, parking lots and bridges (Gnecco et al., 200; Han et al., 2006, Kayhanian et al., 2007). The pollutant concentrations as well as the correlations among pollutants and storm and site characteristics were different from study to study. Several parameters are found to have significant impacts on roadway and parking lot runoff pollutants constituent and concentrations such as average daily traffic load, climate, atmospheric deposition, seasonality (road salt), type of asphalt, vehicle types, and frequent congestion, braking or acceleration (Kayhanian et al., 2007; Helmreich et al., 2010; Huber et al., 2016a; Sansalone and Buchberger, 1997). Table 2 presents a summary of the total concentration matrix of highway runoff pollutants as reported in the literature and the Austrian chemical water quality ordinance.

Most of the pollutants identified in Table 1, particularly heavy metals and non-polar organic contaminants are partitioned between the dissolved and particulate phases as function of pH, flow rate, average pavement residence time and the nature and quantity of solids present (Sansalone and Buchberger, 1997). Nevertheless it is widely reported that the majority of roadway and parking lot runoff pollutant loads are adsorbed to fine particles and particulate organic matter which act as mobile substrates (Kayhanian et al., 2012b; Helmreich et al., 2010; Murakami et al., 2008; Shinya et al., 2000). For instance, Han et al. (2006) analyzed highway stormwater runoff characteristics for three years in California and suggested that suspended solids were associated with most particulate-bound metals such as chromium, copper, nickel, lead and zinc. Similarly, chemical analysis of highway runoff demonstrated that PAHs and NPAHs are dominantly associated with particulate matter (Murakami et al., 2008; Shinya et al., 2000). Therefore, as many pollutants are associated with sediments, it has been suggested that the removal of suspended sediment is critical for the treatment of a wide range of stormwater pollutants (Dierschke et al., 2010; Hatt et al., 2008).

Table 2: Total concentration matrix of typical pollutants in roadway and parking lot runoff as compared to the Austrian Groundwater and Surface Water Quality Ordinance.

Parameter		**Min.**	**Median**	**Mean**	**Max.**	**QZV-Chemie GW (2010)** [a)]	**QZV-Chemie OG (2010)** [b)]
pH[c)]	-	6.4	7.3	7.6	12.3		
EC[c)]	µS/cm	10.8	[d)]	[d)]	59800	2250 at 20 °C	
TSS[f)]	mg/l	29	-	248	1280	-	-
Cd[e)]	µg/l	0.05	1.3	3.3	40	4.5	0.09 - 0.26
Cr[e)]	µg/l	1.0	10.3	145.9	105	45	9
Cu[e)]	µg/l	5.0	48.9	67.5	430	1800	1.6 - 9.3
Ni[e)]	µg/l	2.0	12.6	20.7	145	18	20.3
Pb[e)]	µg/l	1.4	25	52	380	9	7.4
Zn[e)]	µg/l	21	228	314	2234		8.8 – 53
∑16 PAK[f)]	µg/l	0.016	-	2.1	17.1	0.09 [g)]	0.1[g)], 0.1[h)], 0.1[i)], 2.4[j)]
MOH [f)]	mg/l	0.13	-	2.7	8	0.1	-

[a)] BGBl. 98/2010; Austrian Groundwater Quality Ordinance. [b)] BGBl.II 96/2006 i.d.g.F. BGBl.II 461/2010; Austrian Surface Water Quality Ordinance. [c)] pH and EC values (Fürhacker et al., 2013; Helmreich et al., 2010; Kayhanian et al., 2012a). [d)] Mean and median not applicable due to seasonal changes.[e)] Concentrations of heavy metals in highway runoff (Huber et al., 2016a). [f)] TSS, PAHs (∑16 US-EPA PAK) and MOH concentrations (Fürhacker et al., 2013; Kayhanian et al., 2012a; Wichern et al., 20012). [g)] Total PAH: sum of 5 PAH (reference substances: Benzo(a)pyrene, Benzo(b)-fluoranthene, Benzo(k)-fluoranthene, Benzo(ghi)-perylene, Indeno(1,2,3-cd)pyrene)calculated as carbon. [h)] Anthracene. [i)] Fluoranthene. [j)] Naphthalene.

The grain size distribution and physico-chemical characteristics of roadway runoff particles and sediments deposited on roadway/parking lot surfaces are particularly important factors that govern the mobility of the particles and their associated pollutant loads (Kayhanian et al., 2012b; Zandres, 2005; Zhao et al., 2010). The size distribution of roadway runoff particles is usually below 100 µm (Roger et al., 1998; Furumai et al., 2002 and Li et al., 2006). Several researchers reported that fine particles (less than 50 µm in diameter) accounted for 70 to 80% the TSS load carried by roadway runoff (Andral et al., 1999; Kayhanian et al., 2012b; Haile et al., 2016). Furumai et al. (2002) reported that fine particles with diameter less than 20 µm accounted for more than 50% of the particulate mass for highway runoff samples with TSS concentrations below 100 mg/L. Similarly Li et al. (2006) showed that the particle size distribution a first flush particles from highway runoff were less than 30 µm in diameter and more than 90% of particles were less than 10 µm in diameter.

Concentrations and loads of roadway runoff contaminants in sediments, such as heavy metals and hydrocarbons, have been reported to increase with decreasing particle size (Herngren et al., 2006; Li et al., 2006; Sansalone and Buchberger, 1997; Zandres, 2005; Zhao et al., 2010). It is anticipated that concentrations of particulate bound pollutants strongly vary from site to site in a response to sample type (runoff water or road side deposited sediments), climatic conditions, atmospheric deposition and vehicular density. Kayhanian et al. (2012) noted that due to particle aggregation and the preferential deposition of larger, denser particles, particle size distribution and associated pollutant distributions in sediments are not the same as those of the suspended solids in the runoff. A review on the distribution of particulate bound copper (representative for other heavy metals) in road dust and roadway runoff from different investigation programs from the US and Europe is shown in Figure 1 (Dierschke et al., 2010).

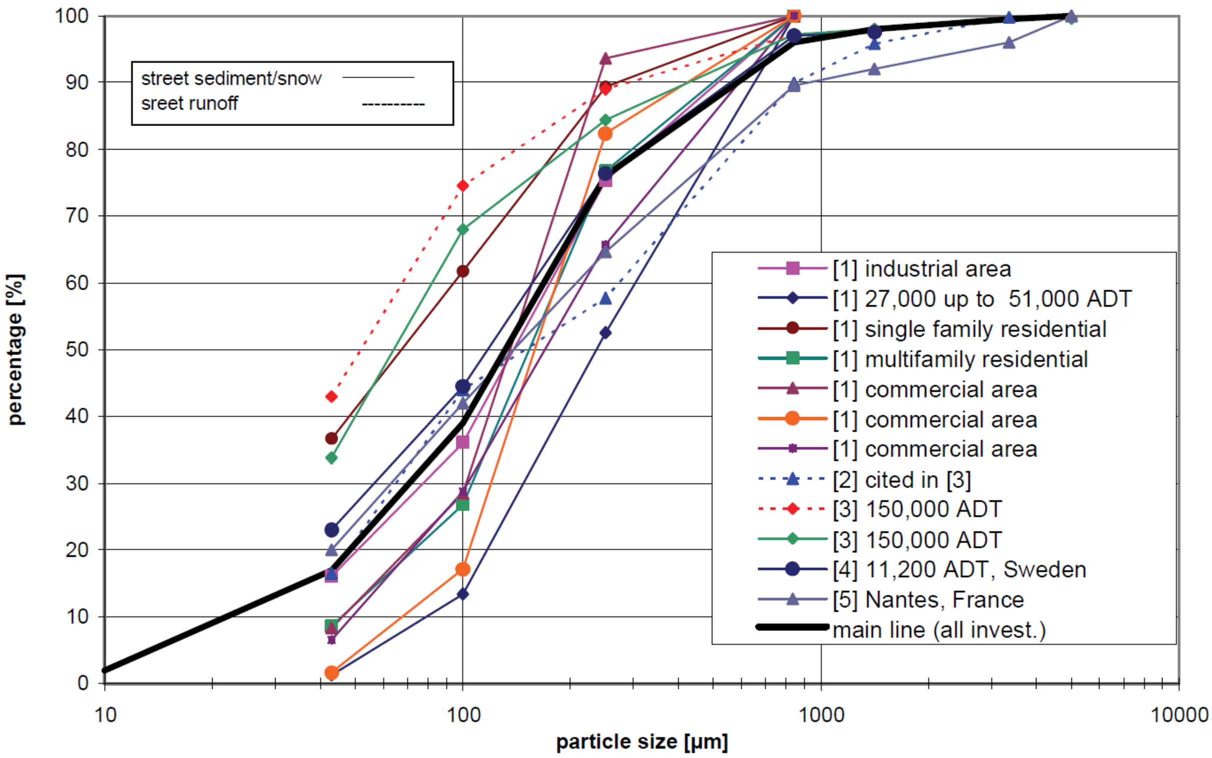

Figure 1. Particulate bound copper distribution as a function of particle size fractions of road dust and road runoff (Schmitt et al., 2010). Copper attached to different particles sizes in road dust and road runoff. [1] Lau und Stenstrom, 2005; [2] Kobriger und Geinopolos, 1984 (cited in [3]); [3] Sansalone und Buchberger, 1996; [4] Germann und Svenson, 2000; [5] Colandini, 1997, ADT: average daily traffic [vehicles/ day] (Source: Dierschke et al., 2010).

Several studies have measured the particle size distribution and associated metal load (Zn, Cu, Pb) in road sediment (Sutherland et al., 2012; Herngren et al., 2006) and stormwater particle (Furumai et al., 2002). Sutherland et al. (2012) studied the fractionation patterns of metal in road sediments and found that the <63 µm grain size class had the highest of Al, Cu, Pb and Zn. Xanthopoulos (1990) has noted that the fine particles with diameters 6 µm to 60 µm accounted for over 80 % of the Cu, Ni and Pb, over 70% of Cd, and 67% of Zn of the pollution load of road runoff. Similarly, Furumai et al. (2002) showed that the loads of certain pollutants such as zinc, lead and copper sorbed to particles smaller than 100 µm in diameter accounted for more than 50% of the total particulate pollutant loads in the runoff. In this regard, fine particles are of more concern than larger particles because they have relatively high surface area, which facilitates the adsorption of pollutants (Herngren et al., 2006; Sansalone and Buchberger, 1997).

Thus, in addition to the obvious water quality impairment caused by particles such as high turbidity, suspended solids act as the transport vector to downstream areas (Herngren et al., 2006). Therefore, the phase partitioning and grain size distribution of stormwater runoff particles have an important implication on the design, construction and performance of a stormwater treatment system. A stormwater treatment system has to ensure both drainage of the runoff and retention of a wide range of particulate and dissolved pollutant loads.

2.2 Toxicity of stormwater

The performance of stormwater treatment systems (BMPs) are usually measured based on removal of pollutant concentrations or mass, as a result less attention has been made to evaluate toxicity (Kayhanian et al., 2008). Therefore, knowledge of roadway runoff toxicity is essential to accurately evaluate BMP effectiveness with regard to removal of the toxic fraction of pollutants. In the literature the majority of studies focus on the ecological effects of stormwater. The ecotoxicological tests were mostly performed using freshwater organisms (Grapentine et al., 2008; Kayhanian et al., 2008; McQueen et al., 2010) and saltwater organisms such as Microtox and Biotox bacteria (Karlsson et al., 2010; Marsalek et al., 1999). Several toxicity tests have been applied to stormwater, for example invertebrate (e.g. Ceriodaphnia dubia, Daphnia magna) (Kayhanian et al., 2008; McQueen et al., 2010), fish (Karlsson et al., 2010; Wu et al., 2013), or algae (Kayhanian et al., 2008). Ecotoxicological evaluations of stormwater from roadways clearly revealed that toxicity may or may not be detected depending upon site, storm conditions and the toxicity test system chosen. For example, McQueen et al. (2010) noted that first-flush samples were more toxic than samples collected later in the storm, thus C. dubia and P. promelas toxicity ranged from no detectable toxicity to complete mortality. However, Karlsson et al. (2010) have shown that none of stormwater derived from a motorway were found to be toxic to the bacteria, V. fischeri. Sediments from a pond and sedimentation tank receiving stormwater runoff from higher traffic intensities exhibited toxic response as pollutants settle and accumulate in the sediments (Grapentine et al., 2008; Karlsson et al., 2010). Sediment toxicity was associated with high concentrations of trace metals and high-molecular weight PAHs, benthic community impoverishment appeared related to high water column salinity (Grapentine et al., 2008). Similarly Karlsson et al. (2010) noted that the determined toxic effects of sediments were in line with their affinity for heavy metals but the role of organic carbon content is highlighted. Kayhanian et al. (2008) investigated the toxicity of stormwater runoff from urban highway sites near Los Angeles, USA. Results indicated that the toxicity to water fleas and flathead minnows of the most toxic samples was mostly, but not entirely, due to copper and zinc. A study of simulated runoff from parking lots were toxic to the purple sea urchin egg fertilization test in all samples tested, with evidence suggesting dissolved zinc as the primary cause of toxicity (Greenstein et al., 2004).

Hence roadway and parking lot runoff is composed of a complex mixture of pollutants a more direct and comprehensive testing methods are required to assess the combined effects on the receiving ecosystem and human health. In the literature different bioassays of genotoxicity such *Salmonella*/microsome assay, micronucleus (MN) assays with human derived cells (HepG2) and plant species such as *Tradescantia* clone 4430 and comet assay with human derived cells (HepG2) have been employed in environmental experiments to test the mutagenic and genotoxic effects of contaminations of soils and waters (Keiter et al., 2006; Mišík et al., 2014; Reifferscheid et al., 2012; Tsukatani et al., 2002; White and Claxton, 2004; Zegur et al., 2009). Genotoxic parameters, such as DNA strand breaks, are currently the most valuable biomarkers for environmental risk assessment and there are many reports linking the DNA damage to subsequent molecular, cellular and tissue level alteration of aquatic organisms (Ohe et al., 2004).

The comet assay with HepG2 also known as single cell gel electrophoresis (SCGE) is one of the most popular tools in genotoxic studies (Knasmüller et al., 1998), and it can be used to assess the genotoxicity of direct and indirect mutagens (Valentin-Severin et al., 2003). This assay has been employed to detect DNA single strand breaks (SSBs), double strand breaks (DSBs), crosslinks and alkali-labile sites (Tice et al., 2000). The SCGE assay is rapid, easy to handle and an excellent tool to detect genotoxic properties of complex mixture of environmental chemicals (Knasmüller et al., 2004; Winter et al., 2008). Such testing method will produce a response to the mixture of chemicals without any prior knowledge of the mixture composition or its chemical properties (Zegur et al., 2009). This method has been widely used to investigate genotoxic effects of pure chemical substances (Uhl et al., 1999), wastewater, surface water and drinking water (Zegura et al., 2009), and sediments (Costa et al., 2014).

The *Salmonella*/microsome assay is the most widely used mutagenicity test procedure for routine screening and was also employed in environmental experiments to study the effects of contaminations of soils (Tsukatani et al., 2002), sediments (Chen and White, 2004; Keiter et al., 2006; Reifferscheid et al., 2012), and waters (Ohe et al., 2004; Keiter et al., 2006). The test is based on the detection of induction of mutations in strains of *Salmonella typhimurium* which lead to histidine auxotrophy (Maron and Ames, 1984). The bacteria are sensitive towards a variety of agents including nitro-aromatic chemicals that are contained in emissions from engines (Rosenkranz, 1982). Therefore, the *Salmonella*/microsome assays are appropriate to assess the mutagenic/genotoxic effect of roadway runoff.

The micronucleus (MN) test with pollen tetrads of Tradescantia is the most widely used plant bioassay for the detection of genotoxins in the environment (for review see Misík et al., 2011). Plants are highly sensitive to metals (Knasmüller et al., 1998), thus MN assay can provide information on environmental effects of stormwater runoff where metals like Pb, Cr, Ni, Cu and Zn are common constituents.

The *in vitro* mutagenic and genotoxic effects of roadway runoff and road side soils have been reported in few studies (Marsalek, 1999; Shinya et al., 2000; Tsukatani et al., 2002). In a study of motorway runoff, Marsalek et al. (1999) found that a sample pre-concentrated 10 times resulted in high cytotoxicity, i.e. 90% inhibition of cell growth, but did not induce genotoxic effects. In this study it was emphasized that sediments from retention ponds may be genotoxic and therefore also particulate matter in the stormwater may show this kind of effects. Shinya et al. (2000) reported that extracts of stormwater particulates elucidated a mutagenic response when tested using the Ames test where the authors ascribed these effects to the presence of PAHs. However, it should be noted that although the water phase was found to give mutagenic effects, it was not possible to relate these effects to any specific compound identified in the water phase (Shinya et al., 2000). Using the comet assay, studies by Lee et al. (2008) have shown a relationship between reproductive abnormalities and increased DNA strand breaks when grass shrimp embryos were exposed to estuarine sediments receiving highway runoff. Reduced embryo production and DNA strand breaks may be linked to high PAH and metal levels. Tsukatani et al. (2002) employed the *Salmonella* assay and found that organic extracts of roadside soils (Kurume City, Japan) were mutagenic both with and without metabolic activation (i.e. +S9 and -S9) in strains TA98 and TA100 derivatives YG1041 and YG1042. The mutagenic effect was more pronounced for soil extracts with higher PAHs and metals concentrations. The mutagenic response in YG1041 and YG1042 strains was associated to nitroarenes and aromatic amines (Tsukatani et al., 2002).

Among the typical roadway runoff pollutants, PAHs and NPAHs are ubiquitous organic contaminants that might have carcinogenic and/or mutagenic effects to the aquatic environment and human health (Huang et al., 2014; Maltby, 1995). It is well documented that NPAHs can have stronger carcinogenic and mutagenic activity than their parent PAHs (Huang et al., 2014; Jung et al., 2001). For example, 1,8-dinitropyrene's mutagenic activity in *Salmonella typhimurium* TA98 is three orders of magnitudes higher than benzo[a]pyrene's, which is often considered one of the most toxic PAHs (Schantz et al. 2000). Moreover, studies indicated that metal–PAH co-toxicity in aquatic systems have more-than-additive mortality, thus such co-exposure to metals and PAHs may produce unexpected effects that exacerbate the combined toxicities (Gauthier et al., 2014; Peng et al., 2015).

In vitro assessment of the cytotoxic, mutagenic and genotoxic effects of stormwater from vehicle trafficked areas and sediments deposited within stormwater treatment systems are screening method that should therefore be regarded as a complementary approach to the chemical analysis commonly used for assessing treatment requirements of stormwater and performance of installed treatment systems.

2.3 Stormwater treatment strategies

The EU Water Framework Directive (WFD) (CEU, 2000) identifies the control of diffuse pollution as a key factor in enabling good ecological status to achieve in aquatic systems. Unless properly managed, runoff water from roadways and car parking lots can carry a heterogeneous mixture of pollutants into receiving waters and contribute significantly to local poor water quality and ultimate failure to achieve WFD targets. As a consequence the reduction of pollution loadings and concentrations are now considered important and are often mandatory in order to meet the requirements of the WFD and from a national regulatory perspective. The impacts of roadways and parking lots runoff on receiving waters can be reduced or avoided through the use of various stormwater treatment technologies commonly known as Best Management Practices - BMPs. Consequently a wide range of best management practices (BMPs) including sedimentation devices (e.g. wet detention ponds), infiltration devices (e.g. infiltration basins, infiltration trenches, infiltration swales and bioretentions), constructed wetlands and filtration systems (Achleitner et al., 2007; Fuerhacker et al., 2011; Gill et al. 2014; Hilliges et al., 2013; Ingvertsen et al., 2012; Wium-Andersen et al., 2011) have been implemented to remove the pollutants in order to meet regulatory water quality requirements. A stormwater BMPs has to ensure both drainage of the runoff and retention of a wide range of both dissolved and particulate pollutant loads.

Easily settable solids and associated particulate pollutants can be effectively removed by treatment technologies such as detention/retention ponds which merely rely on particle settling by gravitation are effective for the removal of solids and particulate bound pollutants (Hossain et al., 2005; Pettersson et al., 1999; Stanley, 1996), but are not suitable for the removal of fines particles or colloids and dissolved pollutants (Scholes et al., 2008; Stanley, 1996). For example the treatment efficency of an urban stormwater detention pond was 71% for suspended solids, 45% for particulate organic carbon and particulate nitrogen, 33% for particulate phosphorus, and 26-55% for metals but no removal of dissolved pollutants was observed (Stanley, 1996). Additionally, some BMPs such as swales and detention ponds suffered a noticeable seasonal performance decline in the winter (Marsalek et al., 2003; Rosee et al., 2009; Semadeni-Davies, 2006). For example Pb, Zn and TSS removal by wet detention ponds dropped from 79%, 81% and 80% to 42%, 48% and 49% respectively (Semadeni-Davies, 2006). Other researchers, such as Grenbel et al. (2013) noted that particle-associated contaminants removed by settling or sedimentation mechanisms may later be released into the dissolved phase due to changes in equilibrium conditions. Thus, a robust and reliable treatment system will ideally possess mechanisms for removal of both particulate and dissolved contaminants. Removal of colloidal/fine particle and dissolved pollutants can only be achieved by infiltration/filtration treatment methods (Scholes et al., 2008).

Infiltration/filtration systems with adsorptive filter media are increasingly considered as an effective alternative for the removal of all typical roadway and parking lot runoff pollutants present in their dissolved and particulate phases (Fuerhacker et al., 2011; Hatt et al., 2008, Li and Davis, 2008; Reddy et al., 2014). BMPs such as soakaways, infiltration trenches, infiltration basins, and filtration systems could utilise adsorptive filter media (i.e. natural sorbents) other than soil to remove pollutants by assimilating and transforming organic, inorganic and toxic constituents through processes such as infiltration, sorption, precipitation, and binding by organic colloidal material or adsorption of metal–ligand complexes (Davis et al., 2001). Despite their effective performance some BMPs such as soil infiltration systems demand large space for their installation which is difficult to fulfil this requirement in urban settings and other land impervious landscapes where space is limited (Hilliges et al., 2013; Reddy et al., 2014). Filtration systems, combined in some cases with dissolved contaminant removal by an adsorptive media are extremely compact and can be retrofitted into existing stormwater collection systems (Brueske, 2000).

The present study focused on BMPs particularly infiltration basins and filtration systems which work by passing the stormwater through filter media (e.g. soil or filter bed layer of zeolite, vermiculite etc.). Especially on understanding the quality of the stormwater runoff and the effect of filtration systems on the effluent water quality. Filtration systems have been recognised as effective techniques to both reduce peak runoff rates and volumes, and maximize the removal of both particulate and dissolved pollutants mitigating pollution of receiving waters (Genc-Fuhrman et al., 2007; Hatt et al., 2008; Reddy et al., 2014). During the passage through the filtration system the stormwater pollutants are removed by several mechanisms, specifically sedimentation, filtration, physico-chemical adsorption, surface complexation and transformation (Grebel et al., 2013; Scholes et al., 2008). This indicates that the long-term performance of infiltration/filtration treatment system mainly depends on the hydraulic performance and adsorption capacity of the filter media.

- Sedimentation of particulate matter and accumulation of pollutants being adsorbed to solid particles occurs due to settling under gravity. Fine particle removal by sedimentation processes can be estimated using particle settling velocities calculated with Stokes' Law (assuming spherical particles). Particulate metal removal efficiency can be calculated using the concentrations for different particle size ranges.
- Physical filtration occurs as particles are deposited in the filter bed due to settling under gravity and straining at pores. Stormwater colloids and chemical contaminants are removed by processes such as attractive forces due to Van der Waals hydrophobic interactions.

- Sorption to filter media which are vitally important processes for the removal of dissolved chemical contaminants are due to processes including chemisorption, surface complexation, ion exchange, surface precipitation, or, in the case of organic contaminants, absorption into organic matrices
- Transformation or degradation of organic pollutants can assure abiotically by hydrolysis, oxidation, or reduction reactions or biotransformation of chemical contaminants by microbial activities
- Plant uptake of dissolved pollutants is envisaged for infiltration basins due to a combination of a natural grass surface with an algal-coated filter media.

The primary biological, chemical and physical processes associated with pollutant removal in filter media based structural BMPs (i.e. filtration system) are summarised in Figure 2.

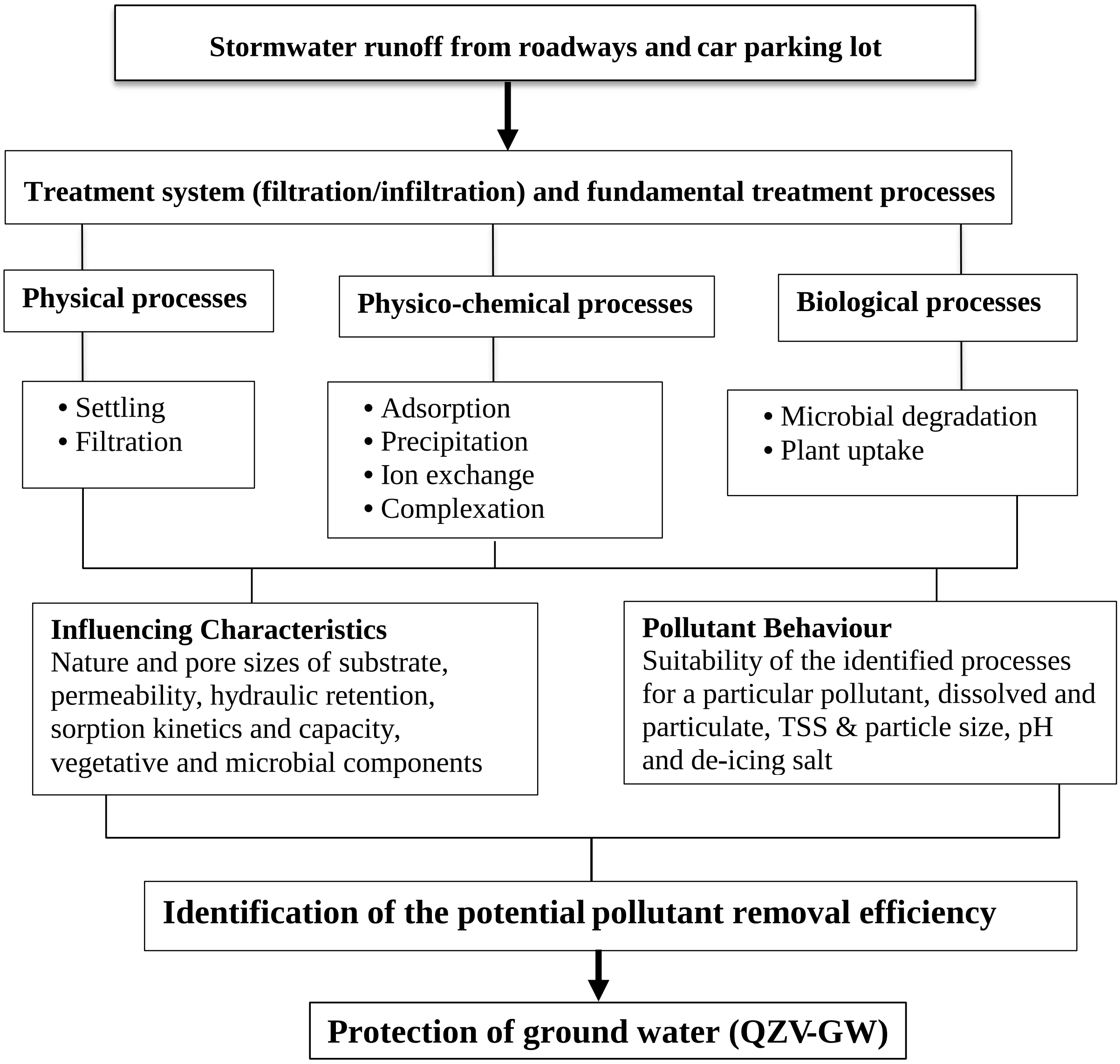

Figure 2. Fundamental unit processes in relation to BMP characteristics and pollutant behaviour (adopted and modified from Scholes et al., 2008).

2.4 Adsorptive filter media

The filter media in stormwater infiltration/filtration systems has received increasing attention as an effective technology for the removal of both particulate and dissolved pollutants through filtration and adsorption processes, using a relatively small space that could be retrofitted in a confined urban environment (Hilliges et al., 2013; Reddy et al., 2014). Adsorption to filter media refers to the physico-chemical adherence of pollutants to a natural substrates or a commercial mixture composed of different media (multi-media filters). There is a significant body of literature demonstrating the potential use of adsorptive filter media in stormwater runoff treatment systems including granular activated carbon, fine sand, zeolite, perlite, vermiculite, limestone, iron filings, steel slags, compost amended soil, multi-media filters; all of which possess good hydraulic properties and are readily available (Hatt et al., 2008; Seelsaen et al. 2006; Reddy et al., 2014; Thomas et al., 2015; Trenouth and Gharabaghi, 2015; Wium-Anderson et al., 2012). Filtration systems with specially selected adsorptive filter media have been found effective in removing suspended solids, nutrients, metals and organic pollutants commonly found in stormwater (Reddy et al., 2014). Adsorbents with large surface area, abundant surface functional groups have high adsorption capacity (Pawluk and Fronczyk, 2014). However, not all of the adsorbents are suitable for use in stormwater runoff filtration system for several reasons including very low hydraulic performance, no single filter media capable of removing all of the contaminants of stormwater (Fürhacker et al., 2013; Fürhacker et al., 2013). Several studies have recognized that multi-media filtration system could effectively remove all of the contaminants of concern in stormwater (Reddy et al., 2014; Trenouth and Gharabaghi, 2015; Wium-Anderson et al., 2012). Therefore, the final selection of a filter media should be verified by testing various combinations or layers of different filter media (Pawluk and Fronczyk, 2014; Reddy et al., 214). Recent studies showed that full-scale filter systems with specially selected sorption materials were presented as a relatively new method for the removal of pollutants from roadways runoff (Fuerhacker et al., 2011; Hilliges et al. 2013).

Seasonal application of road salt have the potential to mobilize heavy metals, soil dispersion and subsequent pollutant mobilization other pollutants bound to soil particles (Bäckström et al., 2004; Norrström, 2005). Heavy metals previously adsorbed/retained within the stormwater treatment system could be mobilized due to ion exchange, chloride complex formation and colloid dispersion as a result of NaCl exposure (Bäckström et al., 2004; DIBt, 2011; Nelson et al., 2009). To ensure the suitability of filter media a test method has been developed to evaluate the remobilization of heavy metals under the influence of road salt (DIBt, 2011; ÖNORM B 2506– 3, 2016).

The mineral components of technical or commercial mixture filter media plays a significant role on the overall performance stormwater infiltration/filtration treatment facilities. Various filter media amendments have been proposed to enhance the adsorption capacity for dissolved contaminant constituents of stormwater, hydraulic performance and reduce mobilization of retained heavy metals due road salt application (. For example, limestone or dolomite used as a filter media amendment improved pH buffering capacity and metal attenuation (Fürhacker eta al., 2015). Therefore, selection of a filter media should be based on the following considerations: (a) consistent in composition; (b) easily/commercially available; (c) environmentally benign, (d) long lasting (non-biodegrable), (e) high permeability, (f) effective in removing multiple contaminants, and (g) no or significantly low (i.e. < 5% of the total load) remobilization of adsorbed heavy metals during road salt application (DIBt, 2011; Fürhacker et al., 2013; Reddy et al., 2014).

3. Materials and methods

In this chapter the case studies and methodologies used are presented in brief. The flowchart in Figure 3 illustrates the investigated thematic analyzed in this dissertation and scientific papers related to these subjects. A detailed description of the methods is available in the papers.

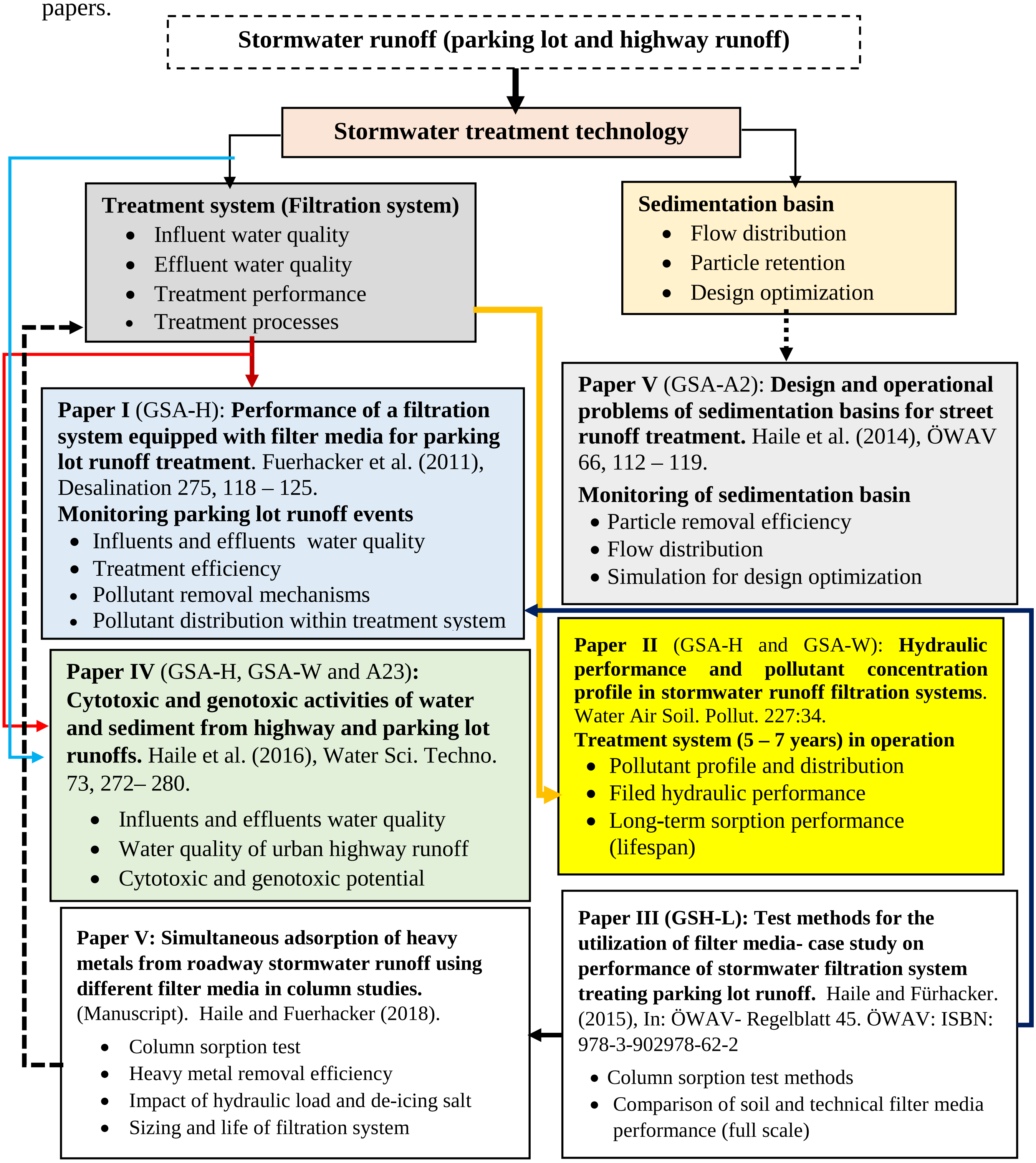

Figure 3: Graphical overview of the workflow from the research background, via system components analyzed in this thesis to the research articles.

3.1 Study sites

Overall four sites, five sites (four treatment systems and one urban highway runoff) were investigated during the course of this thesis. Site characteristics of the five study sites and six installed systems are shown in Table 3. Those sites were used to study water quality characteristics, performance of installed stormwater treatment systems, and toxicity of water and sediment samples. The performance of filtration systems for improving the water quality was evaluated by comparing the influent and effluent concentrations as well as pollutant profiles accumulated in sediments deposited at the filter surface and in the main filter bed of stormwater filtration system. The treatment systems GSA-H and GSA-W are underground concrete structures filled with adsorptive filter media, Aquafilt® (technical filter media) which is produced by SW-Umwelttechnik Austria (SWUT). Despite the sizing variability, the design of the filtration system receiving parking lot runoff (GSA-H) is identical to that of the system treating stormwater from non-urban highway and bridge (GSA-H). The parking lot runoff filtration system at GSA-L is combined plant which is built as a plant – within plant system to allow direct comparison between soil and layered filter media comprised of three different adsorbents (ytong, zeolite and vermiculite). The systems were designed at A_f : A_{red} of 1:283 and 1:28,3 for the soil based and layered filter media respectively. GSA-A2 is a surface filtration basin filled with sandy soil and the infiltration system is planted with grass to enhance treatment efficiency and hydraulic performance.

Table 3. Stormwater runoff source catchment and treatment system characteristics.

Type of catchment	Parking lot and adjacent highway			Non-urban highway & bridge	Urban highway	
Site	GSA-H	GSA-L[a]		GSA-W	GSA-A2	A23
	Lower Austria	Styria		Lower Austria	Vienna	Vienna
AADT (n)	42,147	65,000		25,777	28,549	255,000
A_{red} (m²)	16,800	4,500		17,600	70,000	-
A_f (m²)	40.5	150[b]	15[c]	65	2,360	-
A_{SB} (m²)	17.5	115		28	820	-
Q_{max} (l/s)	209	150	150	180	37	-
A_{red}/A_f (m²/m²)	400	283	28,3	276	29	-
R (mm)	720	870		750	650	650
$K_{sat\text{-}in}$ (m/s)	$5.4\text{x}10^{-4}$	$1.4\text{x}10^{-4}$	$2.5.\text{x}10^{-3}$	$5.4\text{x}10^{-4}$	$1.4\text{x}10^{-4}$	-
$K_{sat\text{-}opr.}$ (m/s)	$1.3\text{x}10^{-6}$ (after 7 yrs)	na	na	$1.3\text{x}10^{-6}$ (after 5 yrs)	$(4.8\text{-}12)\text{x}10^{-4}$ (after 1.5 yrs)	-

AADT: average annual daily traffic; A_{red}: impervious/reduced catchment area; A_f: surface area of the in/filtration system; A_{SB}: surface area of sedimentation basin; Q_{max}: Maximum design flow; and R: mean annual rainfall; [a]: a compact treatment system with adsorptive filter media constructed within a treatment system (Influent volume is equally distributed to the soil Aquafilt® filter bed and RVS-soil)
[b]:soil filter (RVS-soil); [c]:adsorptive filter media (Aquafilt®, SWUT); $K_{sat\text{-}in}$: initial saturated hydraulic conductivity, and $K_{sat\text{-}opr.}$: saturated hydraulic conductivity of treatment system in operation

In this work A_f represents total surface area of filtration system filled with filter media while Ared represents the reduced catchment area (i.e. A_{red} = watershed area x runoff coefficient) connected to the treatment system. The annual runoff volume was determined according to the Simple Method (Schueler, 1987) using the following equation:

$V = P * A_{red} * C$; where V is estimated annual runoff volume treated by stormwater filtration system (m^3); P is annual average precipitation (mm), Ared is total reduced catchment area (m^2), and C is runoff coefficient which is 0.95 for impervious surface.

For further information about the study sites see papers **I**, **II**, **III** and **IV**.

3.1.1 GSA-H

The filtration system receives runoff from a an asphalt car parking lot located in Lower Austria alongside the highway A21, which is one of the main routes of Vienna - northern Austria – lower Austria – Eastern Europe. The parking lot is divided into two areas, one for heavy trucks and for small service automobiles (Figure 4). The parking lot is basically constructed as a resting station for long distance travellers whereby heavy trucks dominate over small service automobiles. A mini-shop and coffee house in this rural area is situated within the yard of the parking lot, which might increase vehicle turnovers using the parking lot. The total reduced catchment area of the site is 1.62 ha and the parking lot has an area of 1.22 ha. The mean annual rainfall of the site is 720 mm and the estimated average runoff for the catchment is 12,000 m^3 per year.

Figure 4. Overview of the parking lot and adjacent highway catchment area (GSA-H), installed filtration system and effluent drainage.

Under the project “Gewässerschutzanlagen für Verkehrsflächenabwässer”, the parking lot runoff filtration system (GSA-H) was installed in 2005. It has three sub-treatment sections: sedimentation tank, oil separator and filter chamber. The filter chamber is filled with Aquafilt®. A schematic of the setup of the filtration system and some pictures after 7 years of operation are indicated in Figure 5. The underground filtration system was designed as filter bed filled with 36 cm (0 – 12 cm ytong, 12 – 24 cm zeolite and 24 – 36 cm vermiculite) layered technical filter media (Aquafilt®, SWUT) and a 20 cm drainage layer of 16/32 mm gravel. The Aquafilt® filter bed and the underlying gravel layer were separated by a polyethylene mesh (mesh size 1,34mm). A geotextile (300 g/m²) was placed at the filter bed surface to capture the particles and prevent clogging of the filtration system. Stormwater draining from the parking lot flows into the connected sewer and manholes and is directed to the sedimentation tank. Then it enters the filter chamber through perforated flow distribution pipes and infiltrates though the 36 cm Aquafilt® filter bed by gravitation. The treated runoff is discharged into a small river. Further details of the study are presented in papers **I, II** and **IV**.

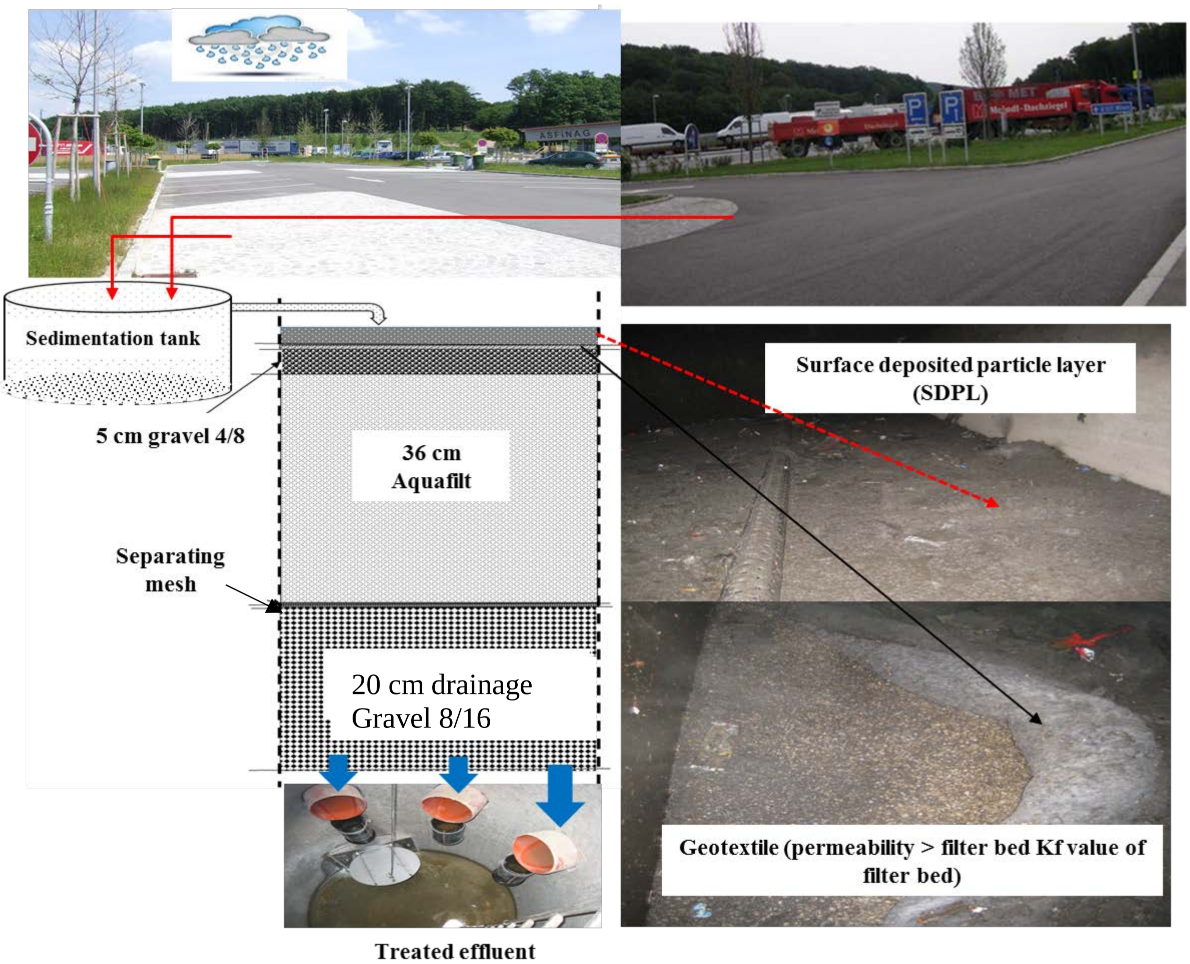

Figure 5. Picture of the parking lot at Hinterbrühl (top), layout of the filtration system, GSA-H (bottom left) and particle layer after 7 years of operation.

3.1.2 GSA-L

This treatment system is located in Styria adjacent to the highway A2 and it receives stormwater from car parking lot and adjacent highway. The treatment system is constructed as a double treatment plant (plant within plant), where 50% of the runoff treated by soil filter (RVS) and 50% through a filter filled with the technical filter media Aquafilt®. The area ratio A_f: A_{red}: is 1: 28.3 for the soil filter and 1: 283 for the technical filter (Figure 6). This type of plant was constructed by the company SWUT to compare the treatment performance of a soil filter system and a technical filter system.

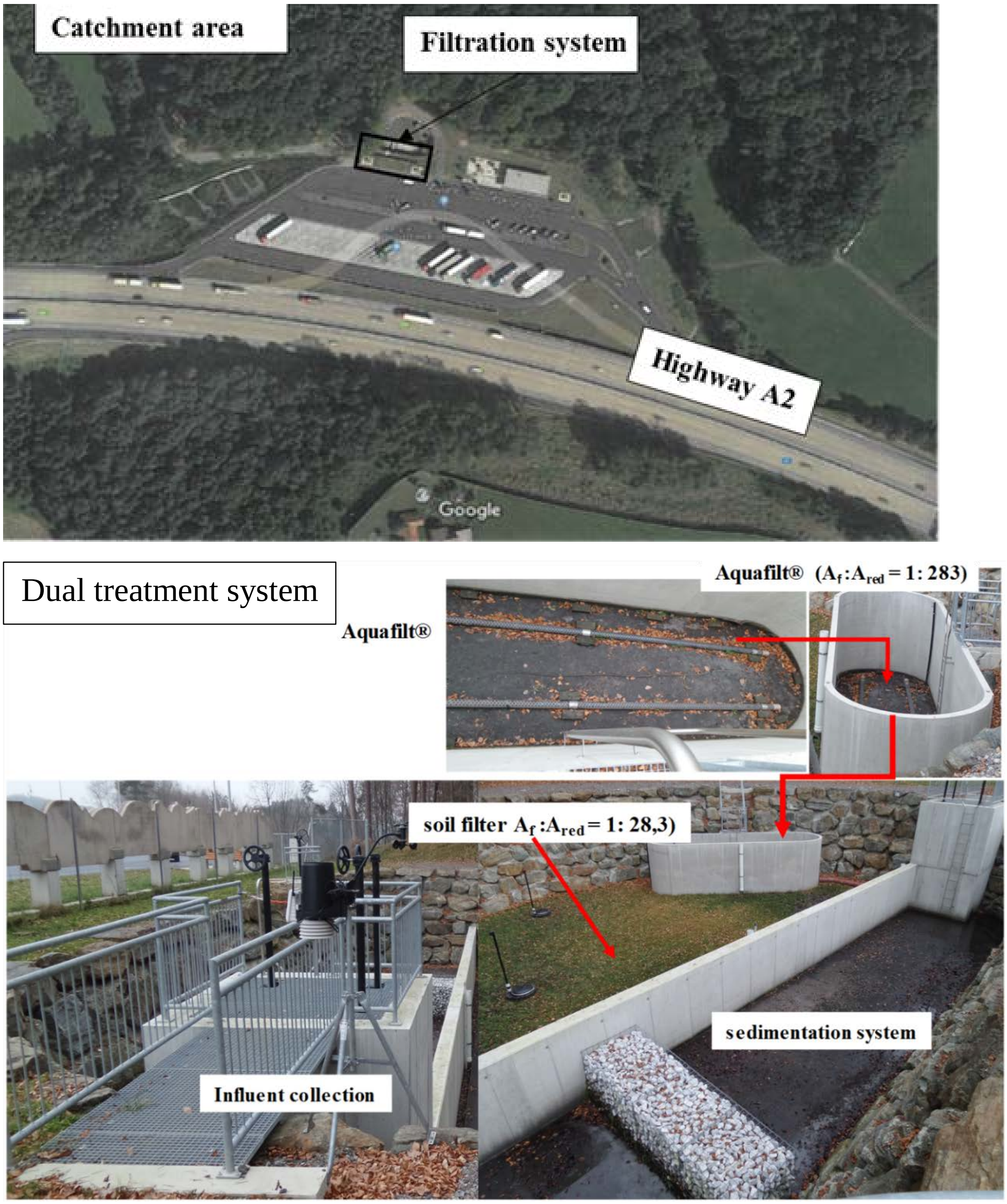

Figure 6. Picture of Lassnitzhöhe catchment area (top) and the dual treatment system, GSA-L (bottom).

During the 2014–2016 sampling period, a total of 11 runoff events were monitored and pollutant removal efficiencies of the filter systems was evaluated. The results are compared with the threshold values define in the Austrian Groundwater Quality Ordinance, i.e. QZV Chemie Grundwasser (QZV-GW, 2010). Note that the QZV threshold values refers to the average values in the groundwater body and a direct comparison of the discharge values with the threshold values of the QZV Chemie GW is not applicable.

Sampling of the influent and the effluent was carried out using a drainage pipe (Figure 7), which allows the sample bottles to be filled when the flow is at a certain discharge height in the pipe. The sampling device in the effluent consists of 3 sampling points at different discharge heights "bottom", "middle" and "top". Depending on the duration and/or intensity of the rain event, the bottles that are connected at different sampling points fill up. Bottles at the bottom sampling points are already filled at very low flow rates, while the sampling points at the center are more likely to sample a medium discharge. A higher outflow would be recorded from the sampling points "top", which was not the case throughout the study period (March 2014 to March 2016). For each sampling point, 3 glass bottles of 250 ml, 500 ml and 1000 ml were connected to the effluent drainage pipe to determine the concentrations of heavy metal contents, mineral oil hydrocarbons and for other relevant stormwater runoff pollutants respectively. Influent samples were collected in 2000 ml bottle using the sampling device indicated in Figure 7. The influent samples, left and right bottles were combined into a mixed sample and concentrations of heavy metals, mineral oil hydrocarbons and other relevant stormwater pollutants were analyzed.

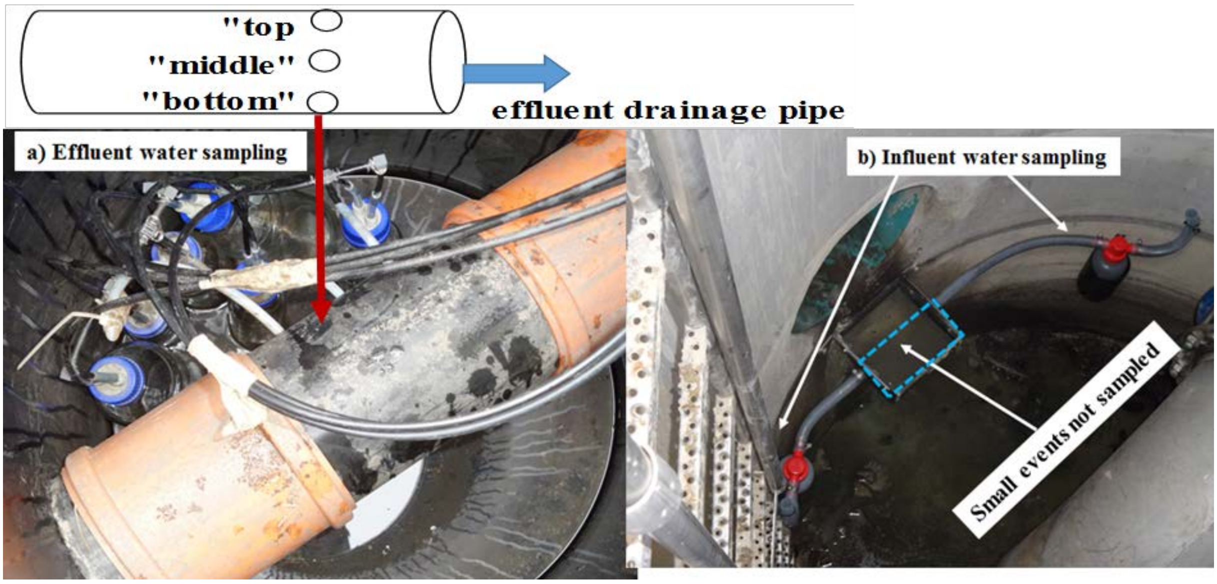

Figure 7. Passive sampling device of effluent (a) and influent (b) installed at GSA-L.

For further details of study objectives, methodologies and results of this site see papers **III**.

3.1.3 GSA-W

This treatment system is located in lower Austria adjacent to highway A1 Lower Austria–Vienna and it receives runoff from a highway and a bridge. It has been in operation since 2007 (Figure 8 and Figure 9). The design layout, filter media composition (Aquafilt®, SWUT) and filter bed thickness is similar to that installed at GSA-H. The runoff collected through connected sewers and manholes, first enters a sedimentation tank and then passes to the filter chamber filled with Aquafilt®. The filter chamber has an area of 67 m^2 and filter bed of 36 cm Aquafilt® and a 20 cm drainage filled with 16/32 mm gravel. The total impervious catchment area is 1.76 ha. GSA-W was sized at A_f to A_{red} ratio of 1:276. The mean annual rainfall of the site is 750 mm and the estimated average runoff for the catchment is 13,000 m^3 per year. The water infiltrates though the filter bed by gravitation and the treated runoff is discharged into surface running waters.

For further details of study objectives, methodologies and results of this site see papers **II** and **IV**.

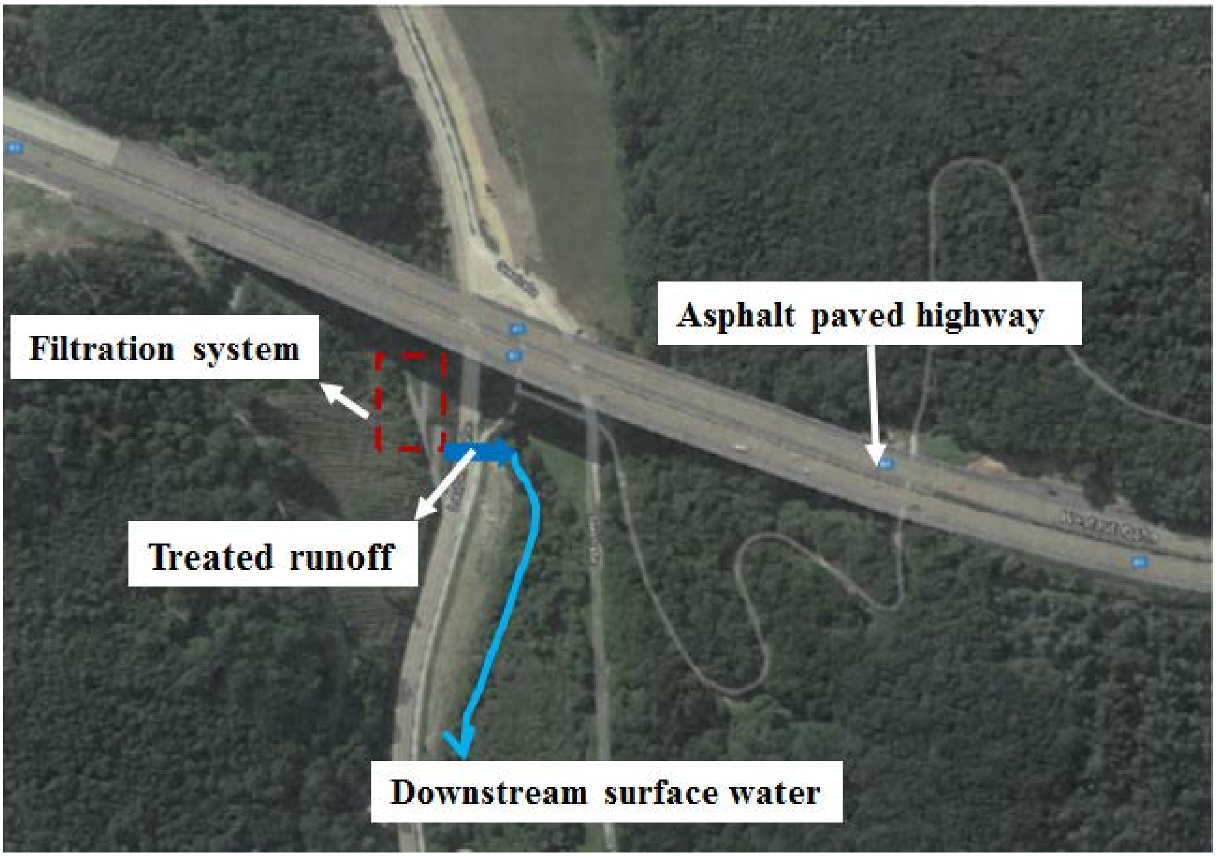

Figure 8. Picture of the non-urban highway catchment area. Dashed red line indicates the underground concrete vault filled with Aquafilt® (GSA-W), the treated highway runoff is discharged (Blue line) to the downstream surface water (light blue line).

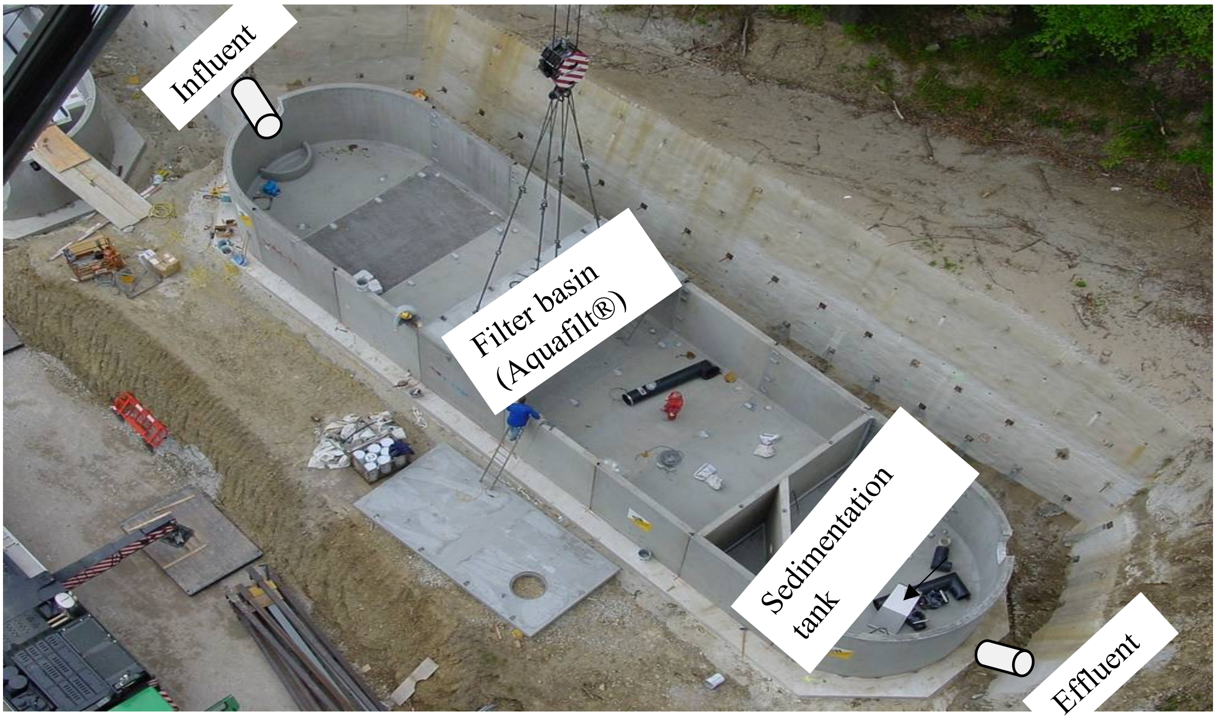

Figure 9. A picture of the filtration system for GSA-W catchment area.

3.1.4 Urban highway (A23)

The highway runoff collection shaft is located adjacent to the highway A23 within the urban city of Vienna (Figure 10). It is a highly trafficked urban road with an average annual daily traffic (AADT) load of 240.000 automobiles and 15.000 lorries. This site was investigated for the characterisation of urban highway runoff quality and also to assess the cytotoxic and genotoxic effects.

Figure 10. An overview of A23 and photo of grab water sample collected in March 2012.

For further details of study objectives, methodologies and results of this site see papers **IV**.

3.1.5 GSA-A2

This stormwater treatment system which receives highway and road runoff from a total impervious catchment of 7 ha, was constructed in 2010 and has been in operation since 2011. It is located along the highway West Autobahn in the vicinity of Vienna. The treatment system includes two major components: sedimentation basin and soil based filter bed (vertical flow filtration) basin. The highway runoff passes through the sedimentation basin where particles would be removed and infiltrates through a soil based filter layer (Figure 11).

The filter basin has an area of 2360 m². It has a filter bed height of 25 cm filled with soil based filter media and a 20 cm 16/32 mm gravel was placed at the bottom as a drainage layer. The soil filter bed and the gravel layer are separated by a geotextile. The soil filter media characteristics (e.g. grain size distribution, particle size uniformity coefficient, pH) and the design of the treatment system was done according to RVS guidelines (RVS 4:04:11; 2011) and the Austrian Standard (ÖNORM B2506-2). The estimated average runoff of this catchment is 50,000 m^3 per year. The treated runoff drains though DN150 pipes and is discharged into a sewer. A spillway drainage system is located at the downstream section of the filter basin to safely convey system overflows.

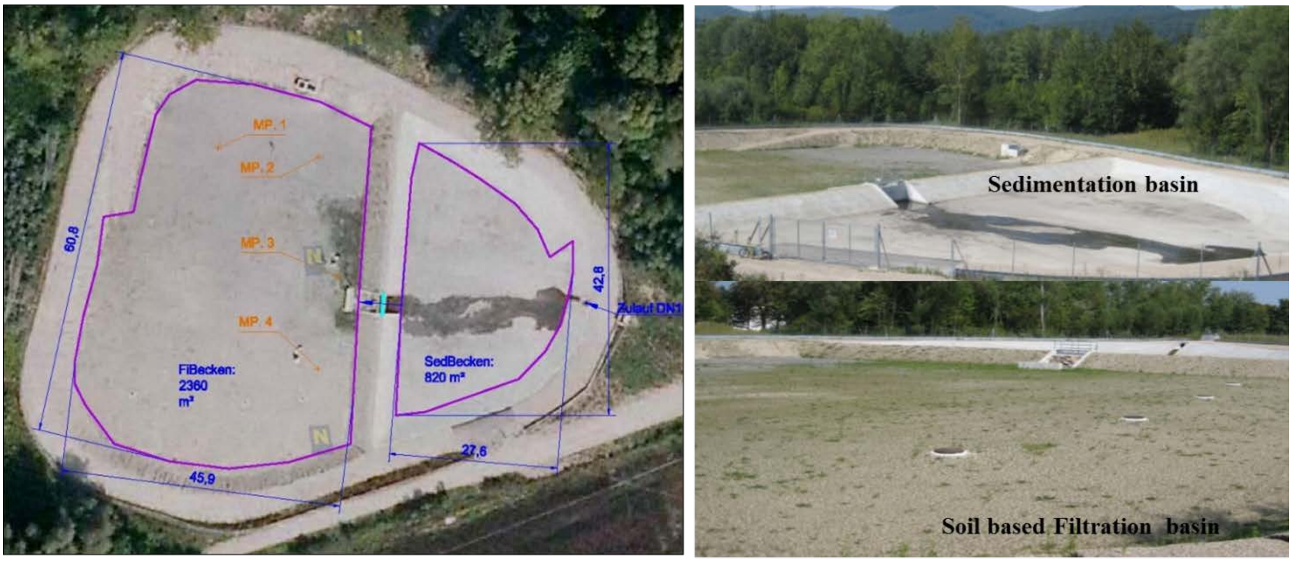

Figure 11. Arial view and photo of the sedimentation basin (GSA-A2).

Sedimentation basins at stormwater treatment facilities provide temporary pools for runoff that allow sediment to settle before the water is discharged into the filtration basin (Al-Nasra, 2013). They are most effective for the removal of coarse to fine particles so that clogging of the filter basin due to particle deposition could be reduced. Unfortunately, sedimentation basins are not efficient when the swift, turbulent water moves along a straight-line flow that takes runoff quickly to the basin's outlet (Figure 12). Basin length to width ratio, ideal basin depth, design of inflow and outflow structures, and installation of baffles in the vicinity of the inlet are the basic design parameters often used for the effective functionality of a sedimentation basin. The installation of baffles can lengthen the flow path and distribute the flow uniformly over the whole basin. This will increase the amount of sediment captured within the basin and also it trap much smaller particles than open basins.

Therefore, one of the objectives of this study were to identify the performance of a sedimentation basin for the removal of particles from highway runoff, causes of non-uniform flow distribution (turbulent flow), settled sediment resuspension and design optimization (use of baffles) to improve the efficiency of the sedimentation basin.

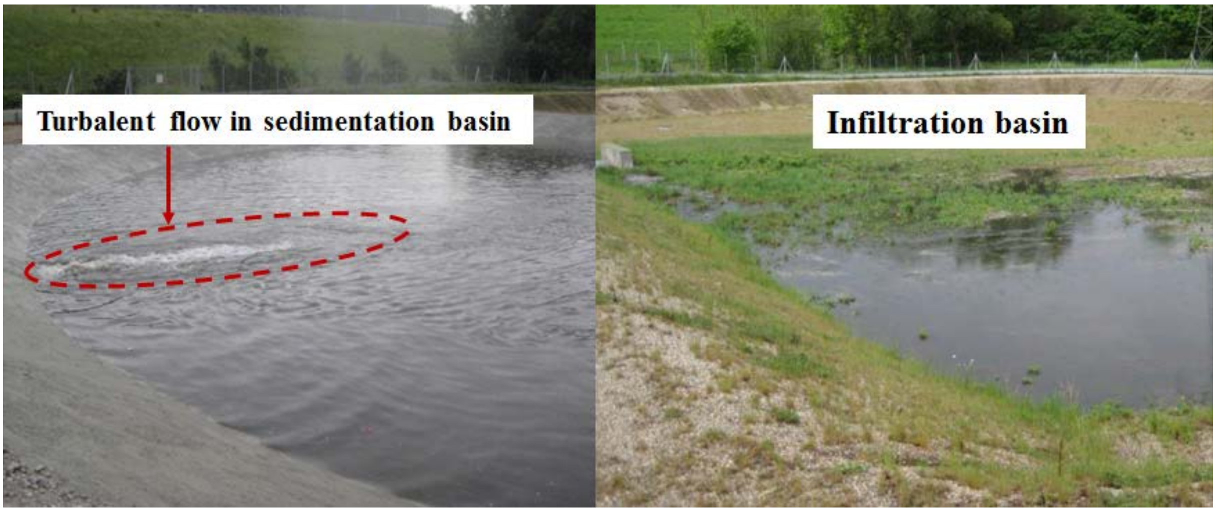

Figure 12. A photo depicting flow distribution for a runoff event on 10.06.2012 at GSA-A2; sedimentation (left) and filter basin (right).

The specific objectives of this site investigation were the following:

(1) Field monitoring

- Assess uniformity of flow distribution for on-site monitored runoff events,
- Determine performance of a sedimentation basin alone for the removal particles by settling processes,
- Evaluate the design optimization/maintenance and operational requirements to achieve laminar flow conditions and improve the basin's efficiency

(2) Laboratory experiment

In a tracer test, the effect of retrofitting parallel baffles at the inlet and outlet of the basin on the performance of the sedimentation basin was evaluated in a lab-scale (prototype) sedimentation basin. The idea of the proposed parallel baffle was to increase the flow residence time by promoting a uniform flow across the entire settling basin. Thus, the objectives of the experiments were following:

- Compare the flow distribution, flow uniformity and flow residence time in experiments with and without retrofitting parallel baffles
- Compare the effect of inlet and outlet structure design with respect to flow uniformity

The details of field monitoring and experimental with lab-scale sedimentation basin are presented in Paper **VII**.

3.2 Sampling

a) Water samples

Stormwater water samples were collected from parking lot runoff (GSA-H), highway and bridge runoff (GSA-W), and urban highway runoff (A23 and GSA-A2). The parking lot runoff treatment system (GSA-H) was monitored over 18 months (December 2005 through May 2007), where influent and effluent samples were collected for eleven runoff events (n=11). The parking lot and highway runoff treatment system (GSA-L) was monitored over 24 months (March 2014 through May 2016), where influent and effluent samples were collected for twelve runoff events (n=12). Details of the sampling procedure are presented in Papers **I** and **III**. Furthermore influent and effluent grab water samples were collected from GSA-H and GSA-W in 2012 (see Paper **II** and **IV)**. The A23, grab water sample collection took place directly from the highway runoff collection shaft during two sampling events; at the end of winter period in 2011 and in the summer period of 2013 (Paper **II)**. To investigate the performance of a sedimentation basin installed at GSA-A2 with regard to stormwater flow distribution and particle retention, five rainfall runoff events were monitored in 2011 (Paper **VII)**.

The pH, electrical conductivity, redox potential and temperature of water samples were measured immediately after collection. Water samples for chemical analyses were collected with glass bottles and samples for the toxicity tests were collected using a 20 L polypropylene jerry cans (Menke Industrieverpackungen GmbH & Co.KG, Beckedorfer Bogen, Germany). For the chemical analyses water samples were preserved and stored in dark room at 4 °C until analyses. Samples for heavy metal parameters were filtered and acidified with HNO_3 suprapure immediately after sample collection. For the toxicity tests water samples were stored at -20°C and before use in the experiments filtered with 0.2 μm filter (Whatman, Sigma St. Louis, USA).

b) Solid samples

Sediment samples from the surface deposited particle layer (SDPL) and filter media core samples (FMC) for the different layers of the filter bed were collected in the summer of 2012 from GSA-H and GSA-W using a precleaned infiltrometer cylinder. The average thickness of the SDPL was 4.5 cm at GSA-H and 2.5 cm at GSA-W respectively. To investigate the vertical pollutants concentration profiles, filter media cores (FMC) were collected at three depths (0–8cm, 8cm– 20cm, and 20cm–36cm). Sediment samples were collected in plastic bags and stored at 4°C before analysis. SDPL and FMC samples were then analyzed for several metal concentrations using acidic digestion to extract the metals. In addition, particle size distribution and concentrations of heavy metals in different particle size fraction were also investigated (Paper **II)**. In addition, a combination of bioassays were employed to evaluate the mutagenic and genotoxicity activity of surface deposited sediments (SDPL) (Paper **II)**.

3.3 Analytical procedures

a) Water quality analysis

Analysis of water samples was conducted at the Chemical Laboratory of the Institute of Sanitary Engineering and Water Pollution Control, University of Natural Resources and Life Science, Vienna (Austria). The analysis has focused on anthropogenic chemical constituents of stormwater runoff from vehicle trafficked surfaces which are the main concern in deteriorating the quality of receiving waters. Measured water quality parameters included heavy metals (barium, cadmium, chromium, copper, lead, nickel, titanium, vanadium and zinc), 16 US-EPA PAHs, MOH and nutrients (i.e., total Kjeldahl nitrogen (TKN), ammonia, nitrate and total phosphorus). Other parameters measured were pH, electrical conductivity, total suspended solids, total organic carbon, biological oxygen demand and chemical oxygen demand. The water quality analytical methods and detection limits are summarised in Table 4. Concentrations of heavy metals were measured using inductive coupled plasma mass spectrometer (ICP–MS) (Elan DRC-e, Perkin-Elmer, Germany) according to DIN EN ISO 17294-2. The concentrations of 16 EPA PAH`s and MOH were determined using gas chromatography mass spectrometry (GC-MS) according to DIN 38407-F39 and DIN EN ISO 9377-2 respectively. The pH, EC, redox-potential, dissolved oxygen content and temperature were measured onsite immediately after the collection of water samples using the WTW Multiline P4.

Table 4. List of parameters with their reporting unit, analytical methods and detection limits.

	Parameter	Unit	Detection limit	Analytical method
Conventional	pH	-	-	DIN 38404 C5
	EC	µS/cm	-	EN 13038
	Chloride	mg/L	0.2	EN ISO 10304-1
	TOC	mg/L	1.0	DIN EN 1484 H3
	TSS	mg/L	1.0	DIN 38409 T2
Nutrients	Phosphorus total	mg/L	0.05	DIN EN ISO 6878
	Ammonium–N	mg/L	0.03	DIN 38406 T5
	Nitrat (NO3-N)	mg/L	0.1	EN ISO 10304-1
Metal	Al total/dissolved	µg/L	5.0	EN ISO 17294-2 E29
	Ba total	µg/L	3.0	
	Cd dissolved	µg/L	0.05	
	Cd total	µg/L	0.1	
	Cr dissolved	µg/L	0.5	
	Cr total	µg/L	1.0	
	Cu dissolved	µg/L	1.0	
	Cu total	µg/L	3.0	
	Fe total/dissolved	µg/L	5.0	
	Mn total	µg/L	1.0	
	Mo total	µg/L	1.0	
	Ni dissolved	µg/L	0.5	
	Ni total	µg/L	1.0	
	Pb dissolved	µg/L	0.5	
	Pb total	µg/L	4.0	
	Sr total	µg/L	1.0	
	Ti total	µg/L	3.0	
	V total	µg/L	2.0	
	Zn dissolved	µg/L	3.0	
	Zn total	µg/L	6.0	
PAH	Acenaphthene	µg/L	0.005	DIN 38407-F39
	Acenaphthylene	µg/L	0.005	
	Anthracene	µg/L	0.01	
	Benzo(a)anthracene	µg/L	0.01	
	Benzo(a)pyrene	µg/L	0.01	
	Benzo(b)fluoranthene	µg/L	0.01	
	Benzo(g,h,i)perylene	µg/L	0.01	
	Benzo(k)fluoranthene	µg/L	0.01	
	Chrysene	µg/L	0.01	
	Dibenzo(a,h)anthracene	µg/L	0.01	
	Fluoranthene	µg/L	0.02	
	Fluorene	µg/L	0.01	
	Indeno(1,2,3)pyrene	µg/L	0.01	
	Naphthaline	µg/L	0.02	
	Phenanthrene	µg/L	0.01	
	Pyrene	µg/L	0.01	
Hydrocarbons	MOH	mg/L	0.1	DIN EN ISO 9377-2

b) Sediment quality analysis

Sediment samples were collected from a particle layer deposited on the surface of stormwater treatment systems (GSA-H and GSA-W). The objective of this sediment sampling was to: (1) characterize the physical composition and particle size distribution, (2) quantify metal element concentrations in bulk surface deposited particle layer (SDPL) and in defined fractions as a function of grain size and (3) test genotoxic potential in different bioassays.

Approximately 98% of the mass of the SDPL was composed of particles smaller than 2000 µm, thus material greater than 2000 µm (2 mm) was discarded. Samples were sorted into grain size fractions of <63, 63–125, 125–200, 200–630, 630–1000 and 1000–2000 µm. The particles with diameter < 2000 µm were wet sieved through stainless-steel sieves into grain size fractions 63–125, 125–200, 200–630, 630–1000 and 1000–2000 µm (ÖNORM L1061-1, 2002). The finer particles with diameter < 63 µm were fractionated into four fractions <2, 2–6.3, 6.3–20, and 20–63 µm by sedimentation using the pipette method after Kubiena (ÖNORM L1061-2, 2002).

Heavy metals were analyzed for the bulk SDPL and FMC at different depth. For the analysis of concentrations of metal elements in the SDPL and FMC, 500 mg of air dried sample was added into 10 mL of ultrapure water and acidified using 3 mL 30% HCl and 2mL 65% HNO_3 in a closed Teflon vessel, and was digested in a high performance microwave digestion unit “mls 1200 mega” (MLS GmbH, Germany). The digested solution was then cooled for 2 hrs. After cooling, the digested samples were filtered through 0.22 µm pore size Phenex RC filter and analyzed for total concentrations using an inductively coupled plasma-atomic mass spectrometry (ICP-MS; Perkin-Elmer, Sciex). Concentrations of PAHs in the sediment samples were analyzed according to methods described in Table 4. The metals used in the sediment quality analysis and their corresponding detection limits are presented in Table 5.

Table 5. The detection limit for trace metals analyses using Perkin Elmer ICP-MS

Metal*	Detection limit (µg/g)	Metal*	Detection limit (µg/g)
Al	0.1	Mn	0.01
As	0.01	Mo	0.01
Ba	0.01	Ni	0.01
Pb	0.01	Pb	0.01
Cd	0.001	Sb	0.01
Cr	0.01	Sn	0.04
Fe	0.1	Sr	0.001
Co	0.002	Ti	0.01
Cu	0.02	Zn	0.02

**The analytical method EN ISO 17294-2 E29 used for all metal parameters*

Moreover, the total concentrations of several heavy metals in the sediment for each particle-size fraction (< 63, 63 – 200, 200 – 630, 630 – 1000, and 1000 – 2000 µm) were analyzed at another institute using a different method. For the analysis of metal elements concentration 100 mg of SDPL dry sample was weighted in Pt-dishes and mixed with 2.5 ml HNO_3 (65%), 2.5 ml $HClO_4$ (60%) and 5 ml HF (40%) and the acid was concentrated near dryness. After that the residue was heated two times with 5 ml HNO_3 (65%) until the fumes were emitted. The last step was to take the residue with 0.5 ml HNO_3 (65%), diluted to 50 ml H_2O and analyzed for total concentrations using ICP-MS 7500 ce (Agilent). The following elements were measured: As, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb, Rb, Sr, Sb, Sn, Sr, V, Zn and Zr. To guarantee the quality of measurement some controlling standards were also measured. All concentrations were reported on a dry-weight basis (mg/kg). Because of their higher relevance regarding stormwater runoff from trafficked areas, particular emphasis was given to Ba, Cd, Cr, Ni, Cu, Pb, Ti, V and Zn (for details see Paper **II**).

3.4 Toxicity tests

In vitro assessment of cytotoxic, mutagenic and genotoxic effects of roadway and parking lot runoff, and sediments deposited within stormwater treatment systems is one of the primary focus of this study. The tests were conducted by the group of Prof. Siegfried Knasmüller, (Institute of Cancer Research, Medical University of Vienna) and Dr. Tamara Grummt (Section Drinking Water and Swimming Pool Water Toxicology, UBA, Germany). All tests were conducted according to existing protocols used for environmental contaminants.

Cytotoxicity of water samples was studied using either reactive oxygen species (ROS) generation, cell proliferation or dye exclusion assay in HepG2. The mutagenicity and genotoxicity of water and sediment samples was evaluated using either *Salmonella*/microsome assays, micronucleus assay or comet assay. These test methods are currently used to evaluate the mutagenicity and genotoxicity of complex mixtures in environmental samples (Mišık et al., 2011; Ohe et al., 2004; White and Claxton, 2014; Zegura et al., 2009). All tests were conducted according to existing protocols used for environmental contaminants.

The *Salmonella*/microsome assay based on the detection of induction of back mutations in different strains of *Salmonella typhimurium* which lead to histidine auxotrophy (Maron DM & Ames, 1994). The tester strains which are currently used, differ in regard to their detection spectrum for different classes of mutagens and have different target genes (for more details see Table 6). The water and DMSO extract was tested with the three strains namely TA98, TA100 and YG1024. The tests were carried out as plate incorporation assays as described by Ames et al. (1973), in presence and absence of metabolic activation mix. The “two fold rule” was applied to evaluate the results (UKEMs) as noted by Kirkland et al. (1990).

Table 6. Test strains used and their characteristics

	Gene affected[1]	**DNA repair**[2]	**LPS**[3]	**Biotin requirement**[4]	**Plasmids** [5]	**Mutational Event**	**Range of his$^+$ colonies**[6]
TA98	*hisD3052*	*uvrB*	*rfa*	*bio-*	pKM101	frameshift	15-60
TA100	*hisG46*	*uvrB*	*rfa*	*bio-*	pKM101	base-pair substitution	75-200
YG1024				*bio-*	pYG219	frameshift	

[1] Target genes in the different strains, leading to histidine requirement.
[2] *uvrB* - Deleted excision repair system, increasing the sensitivity.
[3] The *rfa* mutation changes the properties of the bacterial cell wall and results in partial loss of the lipopolysaccharide (LPS) barrier and increases the permeability of cells to certain types of chemicals. The *rfa* mutation is indicated by sensitivity to crystal violet.
[4] Requirement for biotin.
[5] pKM101, pAQ1– plasmids enhance sensitivity to special types of mutagens.
[6] Number of background colonies according to Kirkland (1990).

Genotoxic effects of water samples with singe cell gel eletrophoresis assay (SCGE Assay) were conducted with a human derived liver cell line (HepG2). These cells were used since these cells have retained the activities of various phase I and phase II enzymes which play a crucial role in the activation/ detoxification of genotoxic procarcinogens Knasmüller et al. (2004). It was shown in numerous earlier investigation that the cells are able to detect of a wide variety of environmental genotoxins.

All results were analyzed by use of GraphPad Prism, version 4.0 (GraphPad Software, Inc., San Diego, CA; USA). Results are reported as means ± standard deviations (SD). The results of the SCGE assays were analyzed by student t-test and one-way ANOVA; p values ≤ 0.05 were considered as statistically significant.

The *Tradescantia* micronucleus assays (Trad MN) were performed according to the protocol of Ma et al. (1994). Clone #4430, a hybrid of *T. subacaulis* and *T. hirsutifolia,* was used in all experiments (Figure 13). The experiments were conducted with young inflorescences. Tap water was used as a negative control, maleic hydrazide (20mg/l) served as a positive control.

The experimental results were analysed with the software Prism 5 (Graphpad Inc., CA, USA) by using one-way ANOVA followed by Dunnett’s multiple comparison test. P-values ≤ 0.05 were considered as significant. The details of the test methods and data analysis are presented in **Paper II**.

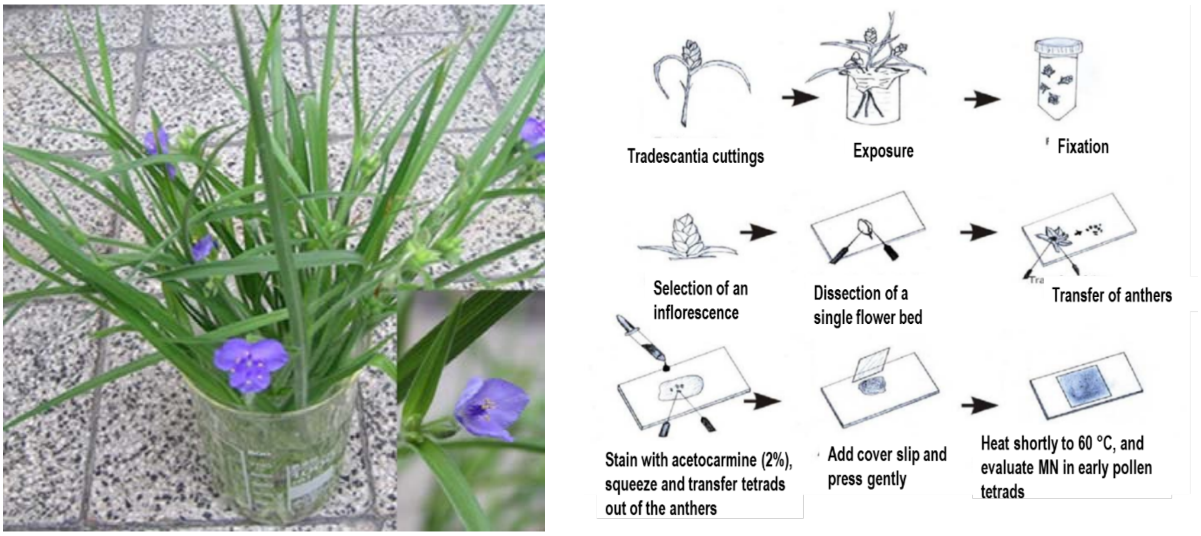

Figure 13. Tradescantia clone #4430 in glass beaker, for assay closed flower buds were used (left) and Scheme of the treatment of the plant material and of the preparation of the slides (right).

3.5 Column sorption experiments

Stormwater infiltration/filtration systems that use adsorptive filter media have proven effective at capturing both dissolved and particulate stormwater pollutants. Column sorption experiments are essential to accurately evaluate the practical applicability of adsorptive filter media. The potential of using natural quartz sand, sandy soil and three mineral based commercial mixes (technical filter media) to remove three heavy metals (Cu, Pb and Zn) from synthetic stormwater solution are another goal of this thesis. The objectives were to evaluate the effect of high hydraulic loads on heavy metal removal efficiency and the effect of de-icing road salt (NaCl) application on the mobilisation of already retained heavy metals in 100 mm inner diameter plexiglass columns (Figure 14). Furthermore, the long-term sorption capacity and expected lifespan of each filter media was evaluated in column breakthrough experiments using 32 mm inner diameter (Figure 15). The experimental breakthrough data determined from column adsorption studies could further be used to predict size of the filtration system relative to its impervious catchment area and the column bed operation lifespan. The influent concentrations of heavy metals in the column feed solution were within the range of levels found in real highway runoff. Moreover, to mimic the minimum pH observed in real highway runoff and avoid precipitation of heavy metals in the solution, the influent pH was adjusted to 5.8±0.2. A filter media was considered as exhausted when effluent concentration exceeded the Austria effluent limit value of 9 µg/L Pb and/or 1800 µg/L Cu for groundwater protection or removal efficiency below the minimum requirement of 80% for Cu and 50% for Zn (ÖNORM B 2506–3, 2016).

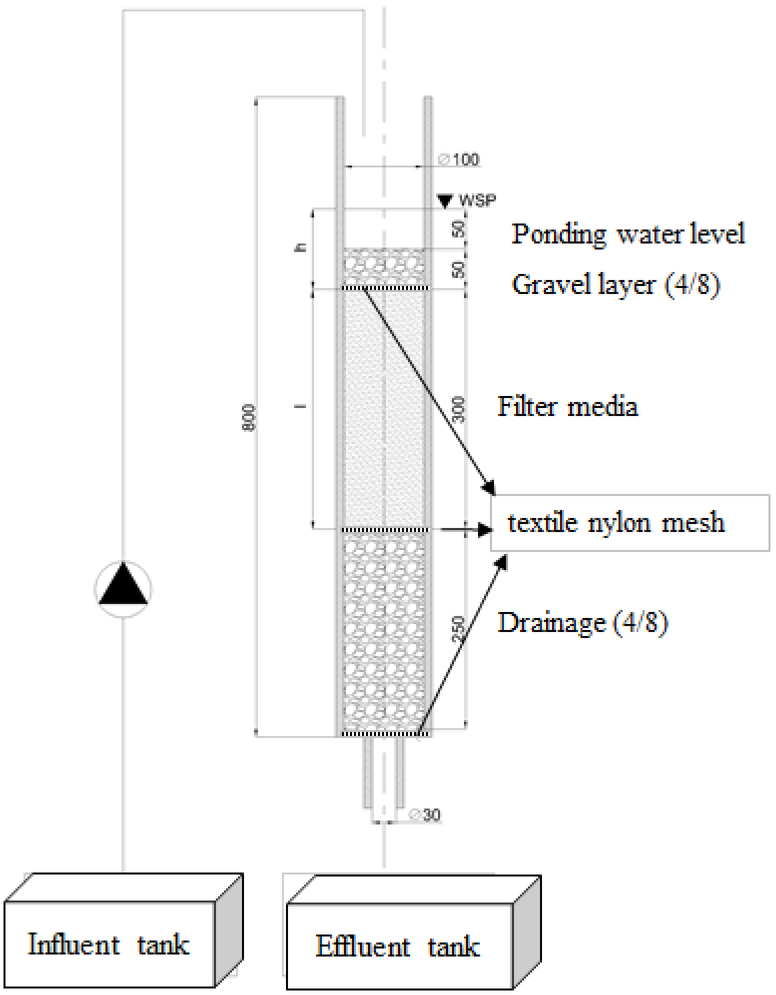

Figure 14. Schematic overview of the column setup to study the effect of hydraulic load on heavy metal removal. Subsequently, the filter media preloaded with heavy metals were flushed with 42 L of NaCl solution (5 g/L NaCl) to evaluate the remobilization of heavy metals.

The design configuration of filtration systems depends on several parameters including depth of the media filter, size of the system relative to its catchment, sorption capacity and hydraulic performance. Therefore, the total mass of each heavy metal retained at filter media exhaustion was used to estimate the size of the system relative to its impervious catchment area. In the laboratory setup, the column acts as a filtration system (A_f) and the pumped water from the tank represents the stormwater runoff volume from impervious surfaces (A_{red}). The ratio between a filtration system to its impervious catchment area (A_f/A_{red}) is often used to size such filters (ÖNORM B 2506–3, 2016).

Further details of the highway runoff characteristics and column sorption experiment are presented in papers **V** and **VI.**

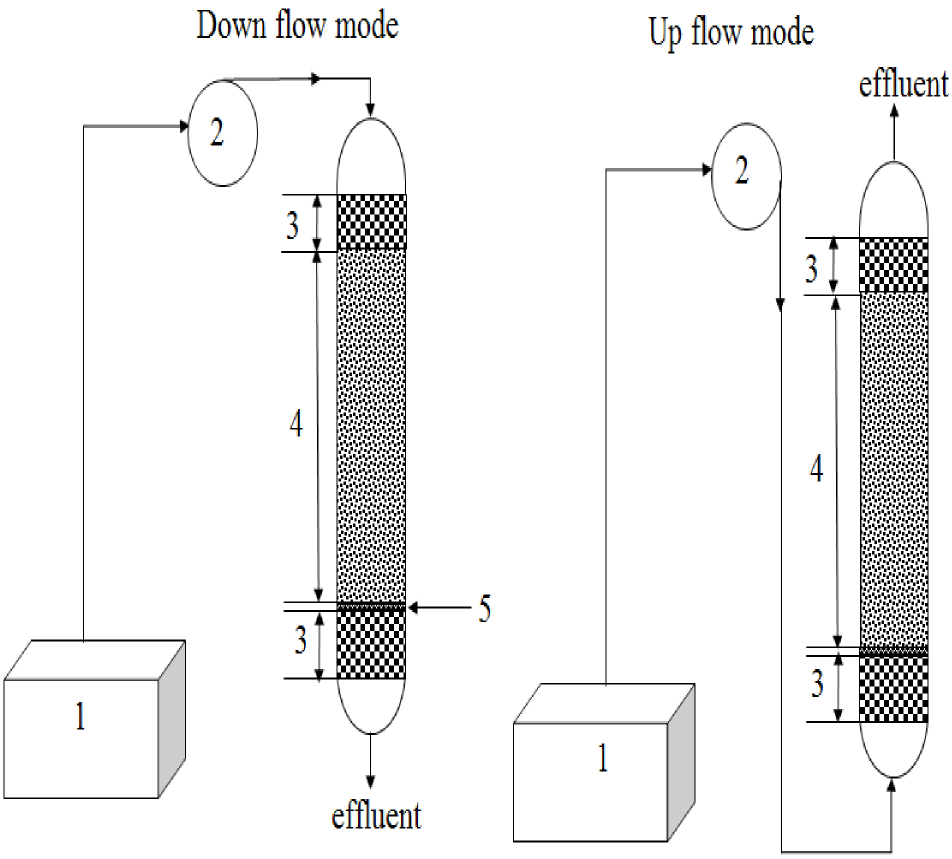

Figure 15. Schematic overview of the column setup to study effect of flow mode and heavy metal sorption capacity at filter media exhaustion. Schematic set-up for column breakthrough experiments: (1) influent feed solution; (2) peristaltic pump; (3) 40 mm glass beads; (4) fritted glass filter ; (5) 200 mm filter media bed; (6) effluent.

4. Summary of research results

4.1.1 Characterization of stormwater runoff

In this study the main pollutants analysed in highway and parking lot runoff were suspended solids (TSS), trace metals (e.g. Ba, Cr, Cu, Ni, Pb, Ti, V and Zn), MOH, nutrients (NH_4–N and H_3PO_4), Cl, DOC, TOC, EC and pH. The selection of this water quality parameters was based on their frequent detection in roadway runoff as reported in previous studies (Göbel et al., 2007, Legret and Pagotto, 1999; Shinya et al., 2000) and their impact in the degradation of receiving water quality (Kayhanian et al., 2008; Marsalek et al., 1999) (see Paper **I, III, IV**). These data were obtained by monitoring runoff from two parking lots and adjacent highways, one rural highway, and one highly trafficked urban highway. The concentrations of pollutants (Pb, PAHs and MOH) measured in runoff grab water samples from highly trafficked urban highway exceeded the Austrian threshold values for groundwater protection. The higher concentrations of heavy metals and Σ5 PAHs observed in urban highway runoff water are in accordance with other studies (Helmreich et al., 2010; Shinya et al. 2000). The measured concentrations of heavy metals from parking lot and rural highway runoff were below the threshold values for groundwater protection. However, median concentrations of MOH and Σ5 PAHs in parking lot and rural highway runoff were exceeding the threshold value (Paper **I, III, IV)**. In addition, the concentration levels of all analysed pollutants in the urban highway runoff were higher than the levels determined in parking lot and rural highway runoff.

Several studies have measured contaminant concentrations on roads and highways runoffs (Helmreich et al., 2010, Hilliges et al., 2013; Pitt et al., 1995) and parking lot runoff (Bannerman et al., 2003; McQueen et al., 2010; Pitt et al., 1995; Tiefenthaler et al., 2003). Others, such as Haile and Fürhacker (2017) and Huber et al. (2016a) compiled runoff concentrations in parking lot and highway runoff across multiple continents. The concentrations of some constituents of parking lot and highway runoff pollutants are summarized in Table 7 and Table 8). Several studies, such as Tiefenthaler et al. (2001) and Bannerman et al. (1993) characterized parking lots as critical source areas to impact receiving water quality. Large fraction of the contaminant loads occur disproportionately in the initial phase of a runoff episode indicating the phenomenon of first flush (Tiefenthaler et al., 2001). The concentrations of pollutants observed in runoff from two parking lots (Paper **I, III** and **IV**) are comparable to parking lot runoff concentrations measured by others globally. For example, the total zinc concentrations measured in this study ranged from < 3 to 264 µg/L for the parking lot at GSA-L and from <LOD to 1000 µg/L for the parking lot at GSA-H, and runoff concentrations from parking lots compiled by Huber et al. (2016a) across multiple continents averaged 201 µg/L (range 39 to

620 µg/L). However, as shown in Table 7 mean concentrations of Zn measured herein in parking lot runoffs were lower the levels observed in other studies (Bannerman et al., 2003; Pitt et al., 1999; Tiefenthaler et al., 2001). The range of concentrations for Cd, Cr, Ni, and Pb measured herein were lower than the global parking lot concentrations compiled by Huber et al. (2016a), in parking lot runoff from simulated rainwater Tiefenthaler et al. (2001) and runoff from parking lot in urban areas (Pitt et al., 1995). While vehicular activity and asphalt pavement ware are the main source of pollution in parking lot runoff, the differences in pollutant concentrations observed in our study compared to literature values may be directly or indirectly attributed to site-specific characteristics such as land use, vehicular traffic density, atmospheric deposition and impervious surface drainage designs (Helmreich et al., 2010; Kayhanian et al., 2007). Moreover, concentrations of pollutants in stormwater runoff also depend on factors such as rainfall runoff event characteristics, the first flush and the dry period preceding a storm event (Kayhanian et al., 2007).

Table 7. Concentrations of constituents in parking lot runoff compared to literature values.

		GSA-H [a] (n=11)	GSA-H [b] (n=1)	GSA-H [b] (n=1)	GSA-L [a] (n=12)	McQueen et al. (2010) [c]	Bannerman et al. (2003) [d]	Tiefenthaler et al. (2003) [e]	Pitt et al. (1995) [f]	Huber et al. (2016a) [g]
		2005 – 2007	2012	2015	2014 - 2016	Concentrations reported in literature				
pH	-	9 (6.6– 12.3)	8.5	7.9	8.9 (7.1– 9.3)	5.7				
EC	µS/cm	105– 59800	12010	1728	680– 100600	89				
Cl	mg/L	267 (53– 789)	402	830	670– 42000	169				
TSS	mg/L	25	21	4						
Al	µg/L				200 (34.6– 639)	910		533.3	3210	
Cd	µg/L	0.4 (0.1– 0.9)	<0.1	<0.05	1.2 (<0.05– 4.3)	3	0.1	2.5	6.3	
Cr	µg/L		5.1	6.3	10.5 (<0.5– 37.3)	23.9	5	3.6	116	0.9
Cu	µg/L	205 (40–430)	14.2	25.4	81 (0.8– 235)	14.3	150	40.3	56	8.6
Fe	µg/L				259 (11.9– 3390)	1543		835		23.4
Ni	µg/L		5.3	2.8	14.6 (3.3– 30.1)	115.1	220	20.7	45	10.5
Pb	µg/L		<4	<4	< 0.5 (<0.5– 0.63)	64		41.8	46	15.5
Zn	µg/L	360 (55– 1000)	81	81.3	76.1 (< 3– 264)	160.1	1780	620	1100	155
Σ16 PAH	µg/L	2.3 (0.41– 7.36)	0.3	<0.2	n.a	12				
MOH	mg/L	1.96 (<0.1– 4.4)	0.3	0.2	3.7 (<0.1– 1100)					

[a]Mean (range) of pollutant concentrations in parking lot runoff; [b] Concentration of pollutants in grab water samples of a parking lot runoff events (GSA-H); [c] Mean concentrations of pollutants in Campus parking lot runoff Clemson University campus, South Caroline (USA); [d] Geometric mean concentrations of pollutants in stormwater runoff from commercial parking lot in Madison, Wisconsin; [e] Mean concentrations pollutants of in parking lot runoff from a simulated rainfall in Longbeach, California (USA); [f] Mean concentrations of pollutants in parking lot runoff from residential, commercial/institutional, and industrial sites in Birmingham, Alabama; [g] Geometric mean concentrations of pollutants in parking lot runoff from global literature review

The concentrations of MOH we observed in two parking lots runoff are comparable to the levels found in runoff from trafficked areas measured by others globally. The concentrations of MOH ranged from < 0.1 to 7-9 mg/L, excluding the high concentrations measured during vehicular accidents (960–1100 mg/L), and runoff concentrations from highly trafficked roads by Göbel et al. (2007) from a literature review ranged 0.51 mg/L to 6.5 mg/L. The concentrations of MOH for parking lot and urban highway runoff were also within the range of mean concentrations in the literature compiled by Haile and Fürhacker (2017) across Austria, Germany and Switzerland averaged 1.9 mg/L(< 0.1 – 7.02 mg/L). The higher concentrations of MOH measured following a vehicular accident confirmed that pollutant wash-off during the initial runoff volume was critical for the occurrence of first flush (Paper **III**).

Table 8. Concentrations of constituents in highway runoff

Paramet.	**unit**	**Grab water samples in this study** [a]				**Göbel et al. (2007)** [b]	**Huber et al. (2016a)** [c]		**Haile & Fürhacker (2017)** [b]	**Helmreich et al. (2010)** [d]	**Hilliges et al. (2013)** [d]
		HNL (GSA-W)		HNL (A23)		Main road	HNL	HUL	HUL/HNL	HUL	
		2012	2015	2012	2013	Global literature review (AADT > 3000)				AADT = 57.000	
pH	-	9	8.3	7.8	7.9					7.5	7.6
EC	µS/cm	930	680	720	1060	108–2436				10.8– 52000	63– 10440
Cl	mg/L	57.6	15			3.9–669					
TSS	mg/L	25	5	825		163				355	67.4
Cd	µg/L	< 0.1	<0.05	1.3	<0.1	3.7	1.2	1.6		<0.5	
Cr	µg/L	7.7	8.7	71.6	11.1	11	12.7	8.6	13.1	191	
Cu	µg/L	39	13	682	131	97	61.8	51.4	86	191	94.9
Ni	µg/L	4.8		68.8	9.8	27	16.1	9.5	13.5	55	
Pb	µg/L	<5	<0.5	112	205	170	18.7	16.8	38.3	56	14
Zn	µg/L	331	81	2560	633	407	248	237	383	847	349
Σ16 PAH	µg/L	<0.2	0.3	18.3		1.9					
MOH	mg/L	0.2	0.3	3	1.7	4.1					

HNL, highway non-urban land; HUL, highway urban land use; [a] Concentration of pollutant in grab water samples of two runoff events from non-urban (GSA-W) and from urban highway (A23); [b] Mean concentrations of pollutants in stormwater runoff from main road and urban/non-urban highway from global literature review; [c] Geometric mean of concentrations of pollutant in stormwater runoff from urban and non-urban highway from global literature review; [d] Mean concentrations of pollutant in stormwater from highly trafficked urban highway.

Traffic related sources such as vehicle exhausts, tyre, brake lining, motor oil and atmospheric deposition contribute to the differences in pollutant concentrations and induced effects among the sites. Metals and other pollutants are deposited on road and parking lot surfaces primarily as a result of atmospheric deposition, vehicular emission (e.g tire wear, brake linings, tyres, motor oil and lubrication oil) and asphalt wear (Table 1). Road salts application during winter months can facilitate the mobility of heavy metals deposited on roadway and parking lot surfaces. One noticeable pattern observed in this research is that, significantly higher pollutant concentrations were observed in runoff grab water samples from an urban highway runoff collected at the end of winter season. This observation was primarily attributed to mobility of heavy metals in response to de-icing salt application which was evidenced by a higher conductivity level of 7200 µS/cm. The range of EC and concentration of Cl we observed in parking lot and urban highway runoffs were comparable to the levels observed in runoff from highly trafficked urban road (Helmreich et al., 2010, Hilliges et al., 2013). This higher conductivity level indicated that the greatest mass of applied de-icing salts was transported by the winter storm first flush events. Therefore, further research is needed to study the accumulation of chloride in impervious surfaces from road de-icing, characterize chloride transport by stormwater runoff, to optimize mitigation efforts.

4.1.2 Treatment performance of stormwater filtration systems

Stormwater filtration systems that utilize adsorptive filter media are effective for the simultaneous removal of dissolved and particulate pollutants typically found in roadway and parking lot runoff (Paper **I, II, III and IV**). As the stormwater passed through a filtration system pollutants can be removed through a combination of processes including filtration, sorption, complexation, precipitation and transformation. Dissolved contaminants are primarily removed by sorption onto surfaces of adsorptive granular filter media. The results of field investigations demonstrated that pollutant removal efficiencies were dependent on their measured influent concentration, nevertheless filtration systems achieved mean removal efficiencies of greater than 85, 75, 73, 95, and 91 percent for TSS, Cu, Zn, MOH and PAHs, respectively. The removal efficiencies of pollutants were varying from event to event, thus treatment performance of the filtration systems closely depends on hydraulic factors (e.g. rainfall characteristics) and influent concentrations of the pollutant. The influent concentration of contaminants affected the performance of stormwater treatment devices with removal efficiency typically lower at lower influent concentrations. Therefore, direct comparisons with the minimum removal efficiency requirements (ÖNORM B 2506–3, 2016) is problematic. It is important to note that the ÖNORM 2506–3 (2016) targets laboratory column experiments for the certification of filter media using relatively high influent concentrations (50–200 µg/L Pb, 100–400 µg/L Cu, 400 – 1600 µg/L Zn and 5 mg/L MOH). For runoff events with high influent concentrations the required removal efficiencies of 80, 80, 50, and 95 percent for TSS, Cu, Zn and MOH respectively, were met. The effectiveness of filtration systems employed for parking lot runoff mitigation depend on the compatibility between their design specifications and actual stormwater quality and quantity characteristics. Therefore, performance the filtration systems primarily focused on the first flush phenomenon.

Treatment efficiency of the parking lot filtration system is in accordance with other studies that found low-impact development (LID) designs such as wetlands, filtration and infiltration designs (Roseen et al., 2006). Roseen et al. (2006) have conducted a comprehensive comparison of eleven stormwater treatment strategies for the treatment of TSS, dissolved inorganic nitrogen, MOH and total Zn deriving from a 3.6 ha asphalt surface car park. Performance evaluations indicate that several LID designs (treatment wetland, filtration and infiltration designs) demonstrated removal efficiencies of 80% to 100%. In contrast, conventional structural BMPs (e.g. swales, retention ponds) perform poorly except for the pond with TSS. Similarly, a bioswale integrating an engineered soil (a mixture of Lava rock and regular soils) and trees installed in a parking lot showed total pollutant load reduction efficiency of 95.4 % for (TSS, heavy metals, nutrients, organic carbon (Xiao and McPherson, 2011). However, performance of parking lot filtration system with determined in this study was higher than the performance of a filtration device filled with a multi-media composed of zeolite, perlite, and granular activated carbon installed at Wisconsin Department of Transportation employee parking lot (Horwatich and Bannerman, 2010). Treatment efficiencies of the stormwater filtration device was 45, 22, 5, and 46 percent for TSS, Cu, Zn and PAHs, respectively, which are up to an order of magnitude lower than the efficiencies achieved by the filtration system evaluated in this study. The variations in treatment efficiencies are caused by variations in filter media composition, inflow concentration, hydraulic loading, retention time and site-specific contaminant loading functions.

Median effluent concentrations measured during the monitoring period (18 to 24 months) were below the threshold values of the Austrian groundwater quality ordinance (BMFLUW, 2010) for the protection of groundwater (Cd < 4.5 µg/L, Cr < 45 µg/L, Ni < 18 µg/L, Pb < 9 µg/L, and ∑5PAHs (B[a]P, FLR, B[b]F, B[k]F, B[ghi]P, INP) < 0.09 µg/L). Despite its higher removal efficiencies (>95%), the effluent concentrations MOH ranged between 0.1 and 2.6 mg/L exceeding the Austrian threshold value of 0.1 mg/L. The mean MOH concentrations, however, are influenced by two high influent concentration events (960–1100 mg/L) measured during vehicular accident that skew average concentration in the effluent results. In addition, the total MOH-index is determined with gas chromatography–flame ionization detector (GC-FID) and that integration of the alkane fractions C10 and C40 occurs. It should be noted that not only petroleum hydrocarbons are detected with this parameter. In addition, measured influent concentrations of copper from all sites were below the Austrian groundwater quality ordinance threshold value of 1800 µg/L, therefore mean removal efficiency noted in the ÖNORM B 2506 –3 (2016) was used as a major criterion for evaluating the treatment performance for Cu. It should be realized that concentration reduction (percent removal) is a function of the relationship between influent and effluent concentrations, that it alone do not provide the full picture regarding treatment performance of stormwater filtration system and should therefore be regarded as a complementary approach to the effluent water quality. In general, the highway and parking lot runoff filtration systems indicated a very robust treatment performance, where all mean heavy metal and PAH effluent concentrations measured during the monitoring period and determined removal efficiencies met the requirements of the ÖNORM B 2506–3 (2016).

Treatment performance of soil filter and technical filter media were compared in dual treatment systems (Paper **III**). The results demonstrated that all effluent concentrations from the filter system with technical filter media were comparable with effluents of the soil-based filter system. The measurements show that the MOH concentrations (values excluded oil spill during vehicular accident) are on average 0.5 mg/L in the effluent of the soil filter and 0.2 mg/L in the effluent of the filter system with a technical filter media. However, MOH concentrations increased up to1.5 mg/L (soil) or o 2.6 mg/L (technical filter media) could be measured in both filter systems due to high influent concentrations from an oil spill during vehicular accident, whereby the removal rates were> 99.8%. The electric conductivity (EC) of parking lot runoff varied greatly between 680 µS/cm and 100600 µS/cm. The effluent EC raged between 240 µS/cm for the soil filter and for the filtration system with technical filter media varied between 503 µS/cm and 8940 µS/cm. Similarly, chloride concentrations of parking lot runoff varied greatly between 370 mg/L and 4200 mg/L µS/cm. The effluent concentrations of chloride raged between 20 mg/L to 2100 mg/L for the soil filter and between 170 mg/L and 3300 mg/L for the for the filtration system with technical filter media. The substantial variations observed for electric conductivity and chloride concentration can be attributed to the application of de-icing salts during the winter season. The application of de-icing salt is reflected in significant increase in electrical conductivity and Cl concentration which is in accordance with other studies that found the application of de-icing salt is reflected in abrupt rises in electric conductivity up to 52 000 µS/cm and Na concentrations of 10 400 mg/L, respectively (Helmreich et al., 2010, Hilliges et al., 2013). The results demonstrated that stormwater filtration systems provide effective removal of typical roadway and parking lot runoff pollutants through mechanisms such as adsorption, filtration, surface complexation, ion exchange and transformation but may not significantly reduce the chloride concentration. The soil filter system has higher retention volume and chloride concentrations of the first flush are likely to be diluted overtime resulting in relatively lower EC value as compared to the technical filter media. Therefore, water retention capacity of the filter media composition is important in improving dilution of the first flush. In addition, sedimentation basin/tank with considerable storage capacity can promote the dilution of chloride concentrations in de-icer laden runoff. The measured values for MOH and

EC were sometimes higher than the threshold values of the Austrian groundwater quality ordinance (BMFLUW, 2010), whose values were used for comparison, but are water quality criterion for whole groundwater body. The mean effluent concentrations of Cd, Cr, Ni and Pb from both systems were below the threshold values of the ÖNORM B 2506–3 (2016). Therefore, stormwater filtration practices that utilize technical filter media can be designed for application at the surface (e.g., infiltration basins) or below ground (e.g., soakaways). An advantage of technical filter media is that they can be installed in compact systems located below parking lots, roadways, or in urban areas where the value of land is very high, this often makes them preferable to soil infiltration systems. Overall the technical filter media based filtration system performed better or comparable to the traditional soil based filtration systems. This is in accordance with results obtained from a laboratory column study using sandy soil and three different technical filter media filters showed that the dissolved Cu Pb, and Zn were effectively removed from synthetic stormwater (Paper **VI**).

4.1.3 Evaluation of filter media for roadway runoff treatment

The simultaneous heavy metal removal potential of five filter media was investigated in column experiments under different operational conditions: high hydraulic loads, effect of de-icing salt and column breakthrough (Paper **VI)**. The tested filter media are natural quartz sand (QS), sandy soil (SS), and three technical filter media (TF-I, TF-II and TF-III) using synthetic stormwater containing three heavy metals (Cu, Pb and Zn). Removal efficiencies under high hydraulic load and breakthrough curves of dissolved metals and sorption capacity were used to compare the performance and lifespan of each tested filter. Filter media exhaustion was set to be equal to target effluent concentration (Pb ≤ 9µg/L) and minimum removal rates (Cu ≥ 80% and Zn≥50%) noted in the Austrian standard (ÖNORM B 2506–3, 2016).

Results show that all filter media were able to maintain high effluent pH level (7.4 – 8.7) over the course of the heavy metal loading except for the column packed with QS. Filter columns packed with sandy soil and technical filter media were very effective for the simultaneous removal of the heavy metals from the synthetic stormwater. The performance of the filter columns decreased over time due to decreasing the total number of adsorption sites. While QS was capable in removing metals in the early stage of dosing, however its performance was very poor relative to other media. Column experiment operated under high hydraulic load showed that soil and all three technical filter media achieved >97 %, 94% and > 80% of Pb, Cu and Zn load removals, respectively, however QS showed very poor performance (34% Cu and 28% Zn). Treatment of synthetic stormwater by the soil and technical filter media met the ÖNORM

B 2506–3 (2016) requirements. Furthermore, the results showed that application of NaCl had only a minor impact on the remobilization of heavy metals from the soil and technical filter media, while the largest release of metals was observed from the QS column. This is in accordance with other studies that found application of NaCl did not show adverse impact on the mobilization of heavy metals previously retained by adsorbent amended filters for treatment of highway runoff in cold climates (Monrabal-Martinez et al., 2017). The adsorption capacity (mg/kg) of each filter media at column exhaustion point towards individual heavy metal varied significantly which resulted from the variations in influent concentrations and adsorption affinity. According to the percentage sorption and sorption capacity values, the selectivity sequence of studied metals by all tested filter media can be given as Zn > Cu > Pb. This shows that adsorption affinity and capacity tend to increase with increasing the influent concentration. For example, sorption capacity Zn at filter media exhaustion was 45, 265, 2390, 4420, 1558 mg/kg for QS, SS, TF-I, TF-II and TF-III respectively. Breakthrough analysis indicated that total load reduction at column exhaustion (SS, TF-I, TF-II and TF-III) were > 95 % for Cu and Pb, and 80– 97% for Zn. Sorption capacity results suggested that the filtration system with the tested filter media can be sized up to $A_{f:}$ A_{red} TF-II (1:200), TF-I and TF-III (1:100) and SS (1:30), respectively. The tested technical filter media can be installed in a very compact system, resulting in significant cost savings with regard to disposal of exhausted material and space for installation. Furthermore, compact treatment systems can be retrofitted in areas urban settings where space is limited or complex topography. Predicted lifespan of each filter media for Cu, Pb and Zn treatment to meet the Austrian regulations is at least 35, 36, 41 and 29 years for SS, TF-I, TF-II and TF-III, respectively. It is suggested that the operational lifespan of stormwater infiltration/filtration systems is mainly limited due to inadequate hydraulic conductivity or clogging (Paper **VI)**.

Several studies showed that most of the pollutants from road runoff are mainly adsorbed on the fine fraction of particulate matter typically smaller than 63 µm (Furumai et al. 2002; Sutherland et al. 2012; Xanthopoulos, 1990), highlighting the need to remove the finer particles transported by stormwater runoff to mitigate receiving environments. Filter media physical parameters such as grain size distribution did have an influence on purification capacity and hydraulic conductivity. Systems with fine filter media, such as soil, are more effective at removing fine particles (TSS) than coarse media but have high head loss characteristics causing higher clogging factors. While systems with coarse filter media provide sufficient infiltration capacity to handle higher flow rates but are less efficient in fine particle removal (Hatt et al., 2008; Kandra et al., 2014). Therefore, filter media through which the stormwater is allowed to infiltrate must be selected to meet two conflicting criteria: have good hydraulic conductivity to treat high runoff events and effective performance to retain fine particles. The selected particle size range is something a trade-off between the hydraulic and treatment performance.

As part of this thesis, laboratory column experiments were conducted using Quarzmehl (Millisil W-4) to evaluate the particle retention efficiencies and hydraulic performance of filter media (natural quartz sand, soil, and technical filter media). 10 to 30 g of Millisil W-4 (1 – 400 µm) were applied at the surface the filter column and tap water was pumped using peristaltic pump in down flow mode. The results showed that all tested technical filter media achieved particle removal efficiencies of greater than 80% but its hydraulic conductivity reduced by only 10%. In contrast, the soil based filter media showed higher particle removal efficiency (90 – 99%) but the infiltration rate reduced to 30% of its initial value (for details see poster abstract, Fuerhacker and Haile 2012). Additionally, a single media may not be efficient to remove multiple pollutants, multiple-media systems can provide effective removal of multiple pollutants, as well as can be amended to improve media effectiveness for the removal of site-specific pollutants (Fuerhacker et al., 2011; Hatt et al., 2008; Reddy et al., 2014). Therefore, filter media should be selected following step by test procedures including laboratory batch and column experiments prior to its application on a full scale. In accordance, the filter media applied in parking lot runoff filtration systems were selected based on the results previous results of column experiments using combinations of individual filter media for testing in mixtures or in a layered configuration (Fürhacker et al., 2013; Fürhacker et al., 2015; Haile, T.M, 2008). Results of the column experiments and full scale filtration systems therefore suggests that mixture or layered filter media have been demonstrated to provide effective capacity for the simultaneous removal of multiple pollutants of parking lot and highway runoff while maintaining high infiltration rates.

4.1.4 Application of the new Standard Method to determine the suitability filter for roads with more than 15000 vehicles per day

Stormwater from roadways with high traffic volume (AADT > 15,000 vehicles/day) are not within the scope of regulations described in ÖNORM B 2506 Parts 1, 2 and 3, and the ÖWAV-RB 45 (2015). For roads with more than 15000 vehicles per day, the rule RVS 04.04.11 (2011) must be considered. This study focused on a literature study (n=55) conducted to collect data on concentrations of the most relevant runoff pollutant constituents (Cr, Cu, Ni, Pb, Zn, MOH and PAHs) in stormwater from highways with an annual average daily traffic load (AADT) of more than 30,000 vehicles across Austria and multiple continents. The objective of this review was to ensure if the

Austrian testing procedure (ÖNORM B 2506-3, 2016) is applicable for certifying filter media usability in stormwater treatment facilities receiving heavily polluted highway runoff.

In Part 3, the requirements and test procedures for all types of filter materials were published in 2016 as Part 3 of the new Standard Method (ÖNORM B 2506-3, 2016), which is based on the results of a research report (Fürhacker et al., 2013). In laboratory-scale column tests, the filter materials can be evaluated to approve their application in stormwater treatment systems receiving runoff from metal roofs (Cu and Zn) or traffic areas after fulfilling several criterion (e.g., initial hydraulic performance and clogging rate; removal of fine particles, heavy metals, MOH; remobilization of adsorbed heavy metals under the effect NaCl and retained fine particles). In Germany, a comparable test procedure has been developed for approving decentralized systems for infiltration/filtration of runoff from traffic areas (DIBt, 2012; DIBt, 2015). Stormwater from roadways with high traffic volume (AADT > 15,000 vehicles/day) are not within the scope of regulations described in ÖNORM B 2506 Parts 1, 2 and 3, and the ÖWAV-RB 45 (2015). For roads with more than 15000 vehicles per day, the rule RVS 04.04.11 (2011) must be considered.

Filter materials performance could be assessed in a lab-scale column tests where several requirements are for the aforementioned parameters. The influent concentrations of fine particles (Millisil-W4), heavy metals (Cu, Pb and Zn) and MOH proposed in the ÖNORM B 2506-3 (2016) were based on a project report conducted by Fürhacker et al. (2013). Several studies have evaluated the performance of filter materials for stormwater treatment (Huber et al., 2016b; Pawluk and Fronczyk, 2014; Trenouth and Gharabaghi, 2015), which are significantly higher the actual concentrations found in runoff from traffic areas (Göbel et al., 2007; Haile and Fürhacker, 2017; Huber et al., 2016a). In contrast, in the new Standard Method (ÖNORM B 2506– 3, 2016) distinct and realistic influent concentrations are used in testing for heavy metals, MOH and de-icing salts for evaluating the suitability of filter materials. The selected influent concentrations for runoff from traffic areas are: 50– 200 µg/L Pb, 100– 400 µg/L Cu and 400 – 1600 µg/L Zn (mixed-metal solution dissolved in deionized water), 5 mg/L MOH and 5 g/L NaCl. According to the ÖNORM B 2506-3 (2016) a filter media could be certified when it fulfilled the following requirements: mean removal efficiency is 80% Cu, 50% Zn, 80% TSS and 95% MOH, and maximum effluent concentrations of 9 µg/L for Pb and 0.25 mg/L for MOH. For the remobilization test, the effluent concentration criteria are $\leq$ 50 µg/L for Cu and $\leq$ 500 µg/L for Zn. Runoff concentration from roadways with more than 30000 vehicles per day compiled by Haile and Fürhacker (2017) across multiple continents (n=55) averaged 86 µg/L Cu, 59 µg/L Pb, 388 µg/L Zn and 4.8 mg/L MOH. These average concentrations are within the range of the influent concentrations proposed in the new Standard Method (ÖNORM B 2506-3, 2016). Results clearly demonstrated that due to the strict test conditions and test criteria chosen in ÖNORM B 2506–3 (2016), the testing of the technical filter media is also suitable to meet the requirements for roads with high annual average daily traffic (AADT).

4.1.5 Accumulation of heavy metals in surface deposited sediments

The particle deposition rates and size distribution of solids transported by stormwater runoff have a significant influence on pollutant adsorption and pollutant accumulation. The geotextile layer placed at the filter bed surface enhanced the capture and retention of fine particles and particulate pollutants (Paper **II**). More than 70% of the mass of sediments accumulated at the surface of stormwater filtration systems were fine particles with a diameter less than 63 µm. The median particle size was 20 µm which is very similar to the particles observed in highway runoff reported by Furumai et al. (2002) and Li et al. (2006). Generally the majority of heavy metals (particularly Cu, Pb and Zn) and PAHs were predominantly accumulated in the surface deposited particle layer (SDPL) with concentrations sharply decreasing with increasing depth (Figure 16) , which is in agreement with findings of other authors (Ingvertsen et al, 2012; Li and Davis, 2008). The results demonstrated that the concentrations of heavy metals, particularly Cu, Pb and Zn, in SDPL were over one magnitude higher than the levels measured in core filter media (FMC) (Figure 16). This implies that the pollutants generated from car parking lot and highway runoffs were mainly particulate-associated and dissolved pollutants were partly adsorbed to the surface deposited sediments. About 60% of the heavy metals (Cu, Pb and Zn) mass accumulated in the surface deposited sediments were associated with the fine particles of less than 63 µm diameter. This is in accordance with other studies that found heavy metal contamination to be confined in the surface layer and upper soil layers of a bioretention system (Li and Davis, 2008). However, concentrations of Cr and Ni slightly increased with depth of filter media suggesting these contaminants predominantly existed in the dissolved form and their removal was mainly by adsorption to the vermiculite layer (24 – 36 cm).

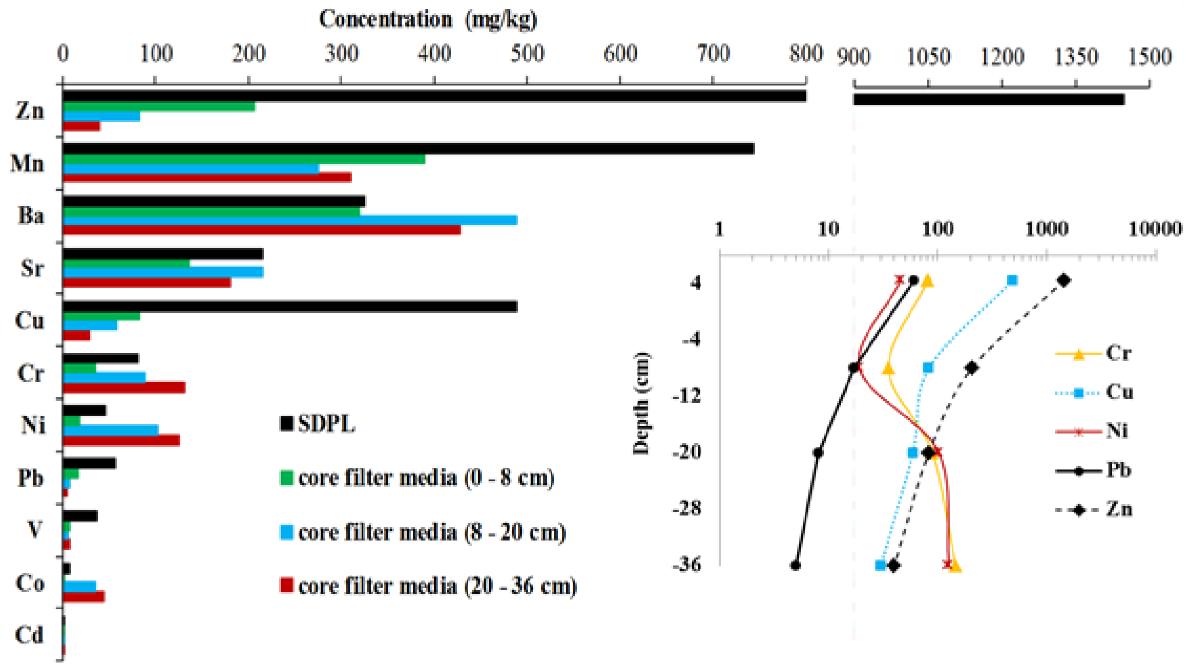

Figure 16. Profiles of heavy metals in surface deposited particle layer (SDPL) and core filter media collected from filtration system treating parking lot runoff (GSA-H).

The concentrations measured in the present study were compared to other similar studies (Table 7). As indicated in Table 9, the mean concentrations of Cr, Cu, Pb and Zn found in the present study were well above the levels reported by several authors (Paus 2999; Ingvertsen et al. 2012; Li and Davis 2008). For example, the mean Cu and Zn concentration in the SDPL observed in this study were 3 to 60 times higher than the values in the top layer (0–5 cm) infiltration systems aged 2 to 16 years reported by Ingvertsen et al. (2012). However, Pb concentrations in the SDPL were within the range of the levels reported by Ingvertsen et al. (2012) and Jensen et al. (2006) but far below the concentration reported by Li and Davis (2008). It is apparent that the pollutant loads entering a stormwater treatment system could significantly vary from site to site due to differences in climatic influences, vehicular traffic density, atmospheric deposition, road maintenance and characteristics of the catchment area. The roadside infiltration swales investigated by Ingvertsen et al. (2012) were sized at 7 to 28 % of their impervious catchment area which is by far larger than the sizing of the filtration systems (i.e. 0.25 to 0.37 %) examined in this study. Therefore, the higher metal concentrations observed in this study could mainly be due to the small size of the filtration system compared to drainage system (i.e. runoff contributing the impervious catchment area), so that input load is high compared to the input loads in bioretentions and road-side infiltration swales. Interestingly, the concentrations of heavy metals in the SDPL were comparable to the levels found in roadside-deposited sediments as reported by Gunawardana et al. (2014). The total concentrations of metals in sediments deposited on urban road surfaces were 11,710 mg/kg Al, 20,310 mg/kg Fe, 30 mg/kg Cr, 400 mg/kg Cu, 420 mg/kg Mn, 30 mg/kg Ni, 170 mg/kg Pb and 910 mg/kg Zn (Gunawardana et al. 2014).

Table 9. Concentrations of heavy metals (mg/kg dry matter) in sediment and filter media layer compared to literature values and sediment quality guidelines.

Reference	Site	Cd	Cr	Cu	Ni	Pb	Zn
Li & Davis, (2008)[a]	Parking lot, (District of Columbia, USA)			75		660	532
Ingvertsen et al. (2012)[b]	Road side swales (Germany)	0.13– 0.71	11 – 67	12 – 57		20– 108	44– 259
Jensen et al., (2006) [c]	Road side swales (Denmark, highways)	0.35– 0.70	13 - 136	114 - 598		49– 44	190– 273
Paus et al. (2014) [d]	Minneapolis, USA metropolitan area	<0.9– 1.87		9.4– 38			44– 87.9
Al Husseini et al. (2013)	Boisbonne basin (A11 highway, France)	0.4– 1.2	43– 54	27–59	34–75	31– 41	132–480
•Boisbonne and Cheviré basin in operation for 17 yrs	Cheviré basin (Nantes ring-road, France)	0.5– 1.2	48– 92	190–366	20–37	70– 368	784–1863
•G08 basin in operation for 37 yrs	G08 basin (A21 highway, France)	9.1– 9.3	111– 129	457– 514	53–56	1064–1235	2508–2595
Fuerhacker et al. (2011) [e]	Parking lot filtration (GSA-H)	0.1	37	230		47	796
GSA-H [f]	This study	0.6	91	490	46	57	1449
GSA-H [g]		0.03– 0.34	35– 131	31– 83	19–126	5.1– 19	40–206
GSA-W [f]		0.49	86	326	52	114	3016
GSA-W [g]		0.05	36.4	21.8	49.8	8.1	241
TEL [h]	Sediment quality	0.596	37.3	35.7	35	18	123
PEL [i]		3.53	90	197	91.3	36	315

[a] Bioretention media in operation for 5 yrs; [b] Infiltrtion swales in operation for 6 to 16 yrs; [c] surface deposits in swales in operation for 16 to 26 yrs; [d] Bioretention media in operation for 2 - 8 yrs; [e] Surface deposited particle layer from GSA-H in operation 2 yrs; [f] surface deposited sediment 3 to 5 cm; [g] core filter media 36 cm; [h] threshold effect level and [i] probable effect level for freshwater ecosystem (MacDonald et al., 2000).

Furthermore, high concentrations of other heavy metals related to vehicular emission such as Ba, Mn, Ti and V were also determined. The average total concentrations of metals in the surface deposited particle layer decreased in the order of, Fe > Al > Zn > Ti > Mn > Cu > Ba > Pb > Cr > Ni > V > Cd. The heavy metals concentrations, particularly Cu, Ni, Pb and Zn were found several times higher in the accumulated sediments compared to their geochemical background concentrations in (Geochemical Atlas of Europe, as cited in Zehetner et al., 2010) indicating vehicular emission and traffic related activities are the major source of these metals.

The sediment quality guideline (SQGs) method for freshwater ecosystems developed by MacDonald et al. (2000) have been used to evaluate potential ecotoxic effects of heavy metals deposited within parking lot and highway runoff filtration systems. SQGs consist of the comparable threshold effects level (TEL) and probable effects level (PEL) (Table 9), which can be used to determine whether a specific metal concentration measured in sediment poses a threat to aquatic ecosystems. Our results revealed that the sediments deposited on both parking lot and highway runoff filtration systems had Cr, Ni, Cu, Pb and Zn concentrations exceeding the threshold levels (TELs) and also the levels of Cu, Pb and Zn were higher than the probable effect levels (PELs) for adverse impacts on biota as shown in Table 9. Therefore, the sediments deposited at the surface of surface stormwater filtration systems will almost definitely exceed guidelines for human and ecological health after several years (> 5) operating life, thus requiring special disposal. Moreover, previous studies focused mainly on heavy metals which are toxic (i.e. Cd, Cu, Cr, Ni, Pb and Zn) (Paus 2014; Ingvertsen et al. 2012; Li and Davis 2008). This study reports additional information regarding the presence of a wide range of heavy such as Ba, Mn, Sr, Ti, and V with high concentrations found in the sediments deposited at the surface of the filtration systems. For example, the metals Ba, Ti, V are included in the Annex 2 of the Austrian groundwater guideline but they are not regulated with target values. Thus, there is a demand to further investigate the behaviour and mode of action of those heavy metals.

4.1.6 Hydraulic performance stormwater filtration systems

The accumulation of particles at the top surface of stormwater filtration systems adversely impacted their hydraulic performance. In-situ (field) infiltration measurements revealed that the hydraulic performance of the filtration systems dynamically decreased after 5 to 7 years of operation (Paper **II**). Initial hydraulic conductivity of the fresh filter media tested at laboratory before its installation ranged 3.8 $x10^{-4}$ to 5.2 $x10^{-4}$ (Haile 2008). The mean hydraulic conductivity of the filter systems reduced to $5.9x10^{-5}$ m/s and $1.4x10^{-4}$ m/s for GSA-H and GSA-W, respectively. The results indicated that hydraulic performance of the filtration system decreased in 1 to 2 orders of magnitudes after 5 to 7 years of operation. The decrease in near-saturated hydraulic conductivity was primarily due to the deposition of sediments at the top surface of the filtration system which acts as a clogging layer or surface clogging (Le Coustumer et al., 2009). The hydraulic conductivity measured in laboratory before installation are used for comparison but could represent the initial field hydraulic performance of the system. It may also be argued that the influence of compaction is important in real systems, where the filter media is often not compacted during construction. In addition, straining of particle within the pores of the filter bed and compaction of the filter media due to hydraulic loading might also contributed to the overall decline in hydraulic performance of the system (Hatt et al., 2008). Le Coustumer et al. (2012) investigated the hydraulic performance of biofiltration systems after long-term use and reported that more than 40 % of the investigated systems had a lower hydraulic conductivity (K) than that recommended in Australian design guidelines, which recommend a K between 50 and 200 mm/h. Scraping and removal of the surface accumulated particle layer, replacement of the geotextile and back flushing of the filter bed approximately every 7 years depending on sizing of the filtration system was recommended as an effective maintenance measures to prevent hydraulic failure. Additionally, removal of the accumulated sediment will also avoid excessive accumulation of pollutants, their leaching or mobilization may otherwise be of concern. The research also showed that placement of a geotextile at the surface of the filter bed appear to be best system configuration for optimal water infiltration and higher efficiencies of treatment, particularly for compact systems with high infiltration rates.

4.1.7 Cytotoxic and genotoxic effects of runoff waters and sediments

In order to evaluate the effectiveness of a stormwater treatment facility with regard to removal of toxic pollutants, chemical analysis and biotests should be used as complementary methods. In this study genotoxic and cytotoxic potential of grab water samples collected from stormwater filtration systems and urban highway runoff, and sediments accumulated at the surface of filtration system were evaluated ((Paper **IV**). The genotoxic activity of water and sediment samples was tested in the *Salmonella*/microsome assays with *Salmonella typhimurium,* micronucleus assay (Trad-MN) with plants and with human-derived liver cells (HepG2), or comet assay with HepG2. Cytotoxicity of water samples was studied using either reactive oxygen species (ROS) generation, cell proliferation or dye exclusion assay in HepG2 (Haile et al., 2016).

The results obtained in the present study demonstrate that stormwater runoff from an urban highway which was exposed to high vehicular traffic volume induced some mutagenic and genotoxic effects. Water samples collected from the influent of the filtration system treating parking lot and rural highway runoff were found to be partly cytotoxic against HepG2 cells in the reactive oxygen species assay and cell proliferation monitored by the xCELLigence system, however there was no clear genotoxicity or mutagenicity effects. In contrast, all effluents did induce neither cytotoxic nor genotoxic effects indicating the investigated stormwater filtration systems were efficient in removing the toxic pollutants.

Several studies have reported the mutagenic and genotoxic effects of roadside soil in different bioassays (Kaur et al., 2014; Schoen et al., 2002; Tsukatani et al., 2002). Furthermore, Jarvis et al. (2018) reported that coarse particulate matter from roadside air elicited a genotoxic response in the normal alveolar cell lines. The aforementioned authors assumed that mutagenic and genotoxic activities of roadside soils correlate significantly with the concentration of PAHs and NPAHs. Similarly, stormwater runoff from traffic areas has been shown to induce mutagenic and genotoxic activities (Shinya et al., 2000). Under heavy traffic conditions, the depositions of toxic compounds such as PHA and NPAHs are usually more important than other stormwater pollutant constituents. Vehicular emissions are the major sources of PAHs and NPAHs which are potentially carcinogenic, mutagenic and genotoxic chemicals. Murakami et al. (2008) reported that particulate fractions of PAHs and NPAHs in street runoff tended to increase with increasing suspended solid concentrations. These compounds can contribute significantly to the carcinogenicity and/or mutagenicity of environmental samples. To evaluate the potential genotoxic and mutagenic impact of stormwater, sediment toxicity bioassay gives a more integrated answer in the test system. Results obtained in the present study demonstrate that sediments (DMSO extracts) accumulated within stormwater treatment systems were either mutagenic in the Trad-MN assay or genotoxic in the *Salmonella*/microsomal assay. The genotoxic and mutagenic activity observed in DMSO extracts suggested that particulate-bound PAH and NPAH compounds existed in the highway and parking lot runoff. It cannot be excluded that also heavy metals contributed to the effects that were detected. It is known that certain metals (such as Cu and Zn) may cause induction of MN in Tradescantia and in some samples the concentration levels of Cu and Zn detected in the sediment sample were sufficiently high to induce MN formation (Steinkellener et al., 1998). Also, in the SCGE experiment with HepG2 cells, metal may have been implicated in the effects, which was found with one of the grab urban highway runoff water sample (A23-2). However, it is unlikely that metals account for positive results in *Salmonella*/microsome assays as they do not induce gene mutation in coliform bacteria, possibly as a consequence of low uptake into the cells (Gatehouse et al., 1990). Therefore, the identified cytotoxic, mutagenic and genotoxic effects were attributed to the presence of NPAHs, PAHs and the combined effect of metals, PAHs and NAPHs. Furthermore, as stormwater runoff contains a complex mixture of organic and inorganic contaminants, the observed mutagenicity and genotoxicity may be attributed to the combined effect of PAHs and metals. Further work is needed to integrate bioassays with analytical methods to better determine and quantify environmental impacts.

4.1.8 Performance of the sedimentation basin

High concentration of pollutants are associated with the finer fraction of solids, thus removal of these particles by sedimentation processes provides significant water quality improvements. Another outcome of this thesis is that the performance of a sedimentation basin constructed as a pre-treatment device on a highway runoff infiltration system was evaluated using six rainfall events monitored in 2012 (April to June) (Paper **VII**). The results show that performance of the sedimentation basin was poor, which indicated that the average influent and effluent TSS concentrations were very similar. This indicates that design optimization of the basin is required to promote settling of sediments through the reduction of flow velocities, increased hydraulic retention time and retention of settled sediments. Moreover, the stormwater entering the basin was not evenly distributed across the whole cross-section of the basin and turbulence conditions resulting from the high flow rate gives rise to backflow currents and side eddies, which reduces particle settling process. Sediment basin efficiency was mainly affected by several design factors, namely basin sizing and geometry, inlet and outlet distribution structure (energy dissipaters) and the length-width ration. Using tracers, results indicated that the installation of a baffle at inlet and outlet structure of the lab-scale sedimentation device improved the uniformity of flow distribution, increase flow retention time, horizontal water flow direction and constant water flow velocity at all points in the settling zone. This combination increases particle settling and retention of the particles captured in the size of sedimentation basin. Sedimentation basins design configuration directly affects the retention of particles and indirectly functionality of the filtration systems (e.g. clogging due to surface accumulation of particles). Therefore, the use of baffles or other retrofits are suggested both to increase particle settling and minimize resuspension of settled particles. The size distribution and density of particles can have a significant impact on the sediment removal performance of sedimentation basin. Therefore, these parameters need to be thoroughly investigated to accurately assess treatment performance of a sedimentation basin.

5. Conclusions and outlook

5.1 Conclusions

This PhD thesis provides an overview of important issues regarding water quality characteristics of stormwater generated from vehicle trafficked areas (parking lot and highway), treatment performance of filtration systems, cytotoxic and genotoxic impacts of runoff from vehicle trafficked areas and sediments accumulated on stormwater filtration systems, and utilization of adsorptive filter media in roadway runoff filtration systems. The research study also established the linkages between treatment efficiency and reduction of toxic pollutants that can induce cytotoxic, mutagenic and/or genotoxic effects. The analysis undertaken was based on field and laboratory studies. Outcomes of this PhD thesis provide essential in-depth understanding to enhance the design, filter media selection and maintenance requirements of stormwater filtration systems.

The results presented in this thesis showed that runoff water from parking lot, rural highway and urban highway were polluted, and several contaminants exceeded the Austrian groundwater threshold values (e.g. Ni, Pb, MOH, PAHs and EC). In addition, the pollutant levels were generally higher in urban highway runoff as compared to parking lot and rural highway runoffs. Stormwater filtration systems that utilize adsorptive filter media as a main treatment component are reliable to remove pollutants for parking lot and highway runoffs ensuring the protection of groundwater. Generally, treatment of parking lot and highway runoff in decentralized stormwater filtration systems met the new Austrian Standard Method (ÖNORM B 2506–3, 2016) requirements regarding minimum removal efficiencies and Austrian threshold values for groundwater protection (BLMFUW, 2010), except for MOH and EC. The pollutant removal efficiency of filtration system treating runoff from parking lot and adjacent highway were 70%–83% for Cu, 66%– 81% for Zn, 82%– 98% for ∑16 EP PAH, and > 95% for MOH. Pollutant removal efficiencies were generally low for runoff events with low influent concentrations, which in turn underestimates the mean and median removal efficiency. For runoff events with relatively high influent concentrations the removal efficiencies fulfil the ÖNORM B 2506–3 (2016) requirements: 80% TSS, 80% Cu, 50% Zn and 95% MOH. In addition, median effluent concentrations of MOH ranged between < 0.1 and 0.4 mg/L which was significantly influenced by the higher influent concentrations (960–1100 mg/L) measured during vehicular accident that skew average concentration results. The road salt (e.g. NaCl) applied in winter seasons are readily soluble in stormwater runoff exhibited higher conductivity and chloride concentration in the first flush. Hence the sampling procedure implemented in this study was devoted to the first flush, average and median electrical conductivity of effluent samples from all sites exceeded the threshold value of 2200µS/cm (at 20 °C) for protection of groundwater.

The technical filter media based filtration systems were very compact (A_f : A_{red} of 1:276 to 1:400), resulting in significant cost savings regarding disposal of exhausted material and space for installation. Such compact treatment systems can be retrofitted in areas where space is limited particularly urban settings or complex topography. The results of laboratory column studies showed that soil and three technical filter media were effective for removal of dissolved Cu, Pb and Zn and loads and application of de-icing salt (NaCl) had a minor effect on the mobilization of retained heavy metals. Adsorption capacity of heavy metals varied to a great extent among the sorbents and concentration of adsorbate. Filtration of synthetic highway and roof runoff in columns packed with soil and technical filter media fulfilled the requirements of the Austrian Standard Method (ÖNORM B 2506– 3, 2016) requirements regarding maximum effluent concentrations (9 µg/L Pb and 1800 µg/L Cu) and minimum removal efficiencies (80% for Cu and 50% for Zn). The column study also show that filtration system that implement technical filter media could be sized at A_f: A_{red} of 1:100 to 1:200. This is in accordance with the size of operation filtration systems. Furthermore, addition of dolomite to the filter media constituent improved adsorption capacity.

Accumulation of sediments and particulate pollutants in the top surface layer of the stormwater filtration system after 5 and 7 years in operation were investigated. More than 70% of the mass of fine sediments with a diameter less than 63 µm was accumulated in surface deposited particle layer. The heavy metals (particularly Cu, Pb and Zn) and PAHs were predominantly accumulated in the surface deposited particle layer with concentrations sharply decreasing with increasing depth. This implies that the pollutants generated from parking lot and highway runoffs were mainly particulate-associated and dissolved pollutants were partly adsorbed to the surface deposited sediments. A geotextile was placed at the surface filtration systems as a barrier to avoid stormwater runoff particles from reaching the pores and clogging the main filter bed.

The present thesis also showed that untreated highway and parking lot runoff as well as sediments deposited within stormwater filtration systems might induce cytotoxic and genotoxic effects. Runoff water samples from parking lot and rural highway were partly cytotoxic against HepG2 cells in the reactive oxygen species assay and cell proliferation monitored by the xCELLigence system. In addition, runoff water from highly trafficked urban highway induced some mutagenic and genotoxic effects. The present study also showed that sediments deposited at the surface of filtration systems exhibited mutagenicity in the Trad-MN assay or genotoxicity in the *Salmonella*/microsomal assay suggesting that particulate-bound carcinogenic and mutagenic PAH and NPAH compounds existed in the highway and parking lot runoffs. The results of the bioassay tests have demonstrated that filtration systems are effective in mitigating environmental impacts from highway and parking lot runoffs.

The performance of field-scale sediment basin receiving highway runoff was evaluated and its performance failed to meet the expectations with regard to hydraulic retention and particle settling or particle removal efficiency. The results of this study has shown that non-uniformity of flow velocity and short-circuiting within the basin induced turbulent flow near the centre of the influent section extending to the effluent. As such, particles settled from previous runoff events were re-suspended and washed out from the basin during high runoff flows. Overall, the influent and effluent mean concentration of TSS were very similar indicting very poor performance of the basin. Therefore, sedimentation basin design optimization is required to enhance hydraulic retention time and permanent retention of settled sediments. Furthermore, a series of experiments were conducted using tracers in a lab-scale (prototype) sedimentation indicated that the installation of a baffle at the inlet and outlet structure of the device enhanced uniformity of flow distribution, hydraulic retention time, laminar flow conditions and constant water flow velocity at all points in the settling zone. This combination increased removal of particles by settling processes and also minimize resuspension of settled particles. Therefore, future testing should indicate how modifications to the sedimentation basin design and installation of baffles in the inlet and outlet structures of the basin enhance the overall performance sediment basin system.

5.2 Outlook

Several recent investigations have evaluated parking lot and roadway runoff water quality, ecotoxicological effects, and performance of stormwater treatment technologies for reducing pollutant input to receiving waters. A complete risk assessment of the fate of stormwater pollutants is still required. This research study has contributed additional new knowledge particularly in relation to long-term pollutant removal and hydraulic performance of filtration systems, and cytotoxicity and genotoxicity effects. The cytotoxic and genotoxic effects of roadway and parking lot runoffs are not somehow understood. In addition to optimization of sedimentation basin design, characterization of stormwater runoff particles is important for evaluating basin performance, pollutant transport and particulate load profiles. As part of this research investigation, a number of areas were identified for further investigation. These include:

- Additional research is needed regarding water quality characteristics of runoff from trafficked areas representing different seasons, first-flush characteristics and throughout the hydrograph of multiple storm events in order to accurately evaluate the effectiveness of filtration systems.
- Prolonged deposition of heavy metals in stormwater filtration system and their possible leaching due to changes in the environmental conditions should further be investigated.
- The cytotoxic and genotoxic effects of parking lot and highway runoff only focused on grab water samples and sediments. Similar research should be conducted throughout the hydrograph of multiple storm events and seasons in order to evaluate filtration system effectiveness with regard to removal of the toxic fraction of pollutants.
- Detailed study of solids transported by stormwater runoff in relation to the composition, size distribution, density, surface charged sites and theoretical settling velocities are all relevant to the effectiveness of treatment facilities.
- The particulate pollutant (sediment) load has significant impact on hydraulic performance of filtration systems. Therefore, detailed filtration system design optimization in relation to ratio of the filter area to its impervious catchment area, and boundary conditions of the site (e.g. load from pollen or solid input loads in response road salt application) is required to minimize clogging of the system due to high solid loading.
- The research study investigated the effect of road salt (NaCl) application on the mobilization of previously retained heavy metals. To simulate the real situation, further investigations are also needed to assess the response when NaCl and multiple heavy metals are applied simultaneously.
- Hence stormwater pollutants are preferentially bound to fine particles, a detailed study is recommended in relation to selecting a target particle size and particle density as an important sedimentation basin design parameters. Additional testing is needed to provide a more comprehensive evaluation of sediment basins performance in relation to size distribution and resuspension of settled particles.

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6. Research papers in the context of this PhD thesis

I. Performance of a filtration system equipped with filter media for parking lot runoff treatment. Desalination 2011, *275*, 118– 125.

II. Hydraulic performance and pollutant concentration profile in a stormwater runoff filtration systems. Water Air. Soil Pollut. 2016, *227:34*, 1–16.

III. Prüfmethode zur Filterwirkung und –eignung: Behandlung Niederschlagsabflüssen von Parkplatz mit Bodenfilter bzw. technischen Filtern. ÖWAV Veranstaltung Versickerung von Niederschlagswässern 2015, ISBN: 978-3-902978-62-2.

IV. Cytotoxic and mutagenic activities of waters and sediments from highway and parking lot runoffs. Water Sci. Technol. 2016, *73(11)*, 2772– 2780.

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6.1 Paper I

Performance of a filtration system equipped with filter media for parking lot runoff treatment.

Fuerhacker M, **Haile, T.M.**, Monai B, Mentler, A. (2011). Desalination 275, 118–125.

Desalination 275 (2011) 118–125

Contents lists available at ScienceDirect

Desalination

journal homepage: www.elsevier.com/locate/desal

Performance of a filtration system equipped with filter media for parking lot runoff treatment

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ARTICLE INFO

Article history:
Received 25 October 2010
Received in revised form 16 February 2011
Accepted 16 February 2011
Available online 16 March 2011

Keywords:
Stormwater runoff
Pollutants
Filtration
Filter media
Removal efficiencies

ABSTRACT

Stormwater runoff from automobile trafficked areas introduces significant loads of pollutants to receiving water bodies leading to an adverse long term effect. To control runoff pollutants from parking lot, a filtration treatment system with a layered filter media activated carbon, composite, vermiculite and zeolite was installed. The removal of the pollutant constituents was evaluated in this study. During a monitoring period of eighteen months eleven runoff samples were collected from the influent and effluent sections and analyzed for the mentioned pollutant parameters. The mean influent annual load of TSS, Cu and Zn were 2600, 2.1 and 3.5 kg respectively. For the monitoring period mean removal efficiencies of 85% for TSS, 75% for Cu, 73% for Zn, 83% for 16 EPA PAHs, 70 to 98% for individual PAHs, 93% for mineral oil, 71% for NH_4–N and 52% for TOC were achieved. Greater than 60% of the Cu load was removed within the filter chambers, but >60% of Zn and TSS loads were removed in the sedimentation tank, oil separator and geotextile filter mainly due to filtration, precipitation and sorption to sediments. Furthermore, except for chrysene heavier molecular weight PAH compounds were effectively removed than lighter molecular weight PAH compounds. The treatment mechanisms include: filtration, precipitation, adsorption and cation exchange.

1. Introduction

Stormwater runoff from paved and automobile trafficked areas such as highways, roads, parking lots and bridges has been identified as critical non-point source pollution to receiving waters. The typical pollutant constituents of primary concern include: TSS, nutrients (nitrogen and phosphorus compounds), heavy metals (e.g. Cd, Cr, Cu, Mn, Ni, Pb, Pt, Ti, Zn, and V); PAHs, mineral oil and grease [1,4,8,9,13,16,19].

If stormwater contaminated with heavy metal is directly discharged into receiving water bodies, the non-biodegradable metals can accumulate in the environment being adsorbed to river bed sediments and soil, causing acute or carcinogenic adverse effects also on human life [10,11,17]. Increasing environmental concerns regarding the spread of pollutants through stormwater runoff from highways, roads and parking areas have led to the construction and installation of different kinds of treatment facilities. Today a large variety of publications exist regarding stormwater runoff treatment methods along with their observed performance. In several studies natural and constructed wetlands have been investigated as practical alternatives for treating runoff from urban areas, highways and roads [5,18,20,22]. These treatment systems allow reducing mainly particulate pollutants. In detention ponds, removal of pollutants in their dissolved form can be improved by clay lining [12]. The construction of wet lands and detention ponds requires large area and were mostly applied for pollution source areas at catchment scale.

Filtration of stormwater through a specially constructed filter system is one possible treatment method (Farm, 2002; [15,21]). Filtration systems comprised of a media layer with an adsorption capacity (e.g. zeolite, peat, granular activated carbon or sand) are relatively recent innovation through which runoff passed and the filtered runoff is collected by an under drain [6]. For isolated and small pollution source areas, filtration treatment devices filled with an adsorbent material have been developed and used at many locations, especially in areas with soils where infiltration (too clayey) or wet detention ponds (too sandy) cannot be used or isolated areas and areas of high imperviousness (e.g. parking lot) [15]. Filtration treatment systems are considered as promising methods for reducing dissolved and particulate phase pollutants provided that the selected filter media has high adsorption capacity towards heavy metals and PAHs. An ion exchange filter media could provide additional stormwater treatment by replacing any toxic heavy metal cations in the runoff with cations such as sodium, calcium or magnesium. This technology could then reduce the heavy metal levels in motorway

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0011-9164/$ – see front matter
doi:10.1016/j.desal.2011.02.041

stormwater through retention within the ion exchange material and surface adsorption [18]. The removal of stormwater pollutants by filtration treatment system equipped with different filter media (eg coated sand, zeolite, granular activated carbon (GAC), and peat) has been evaluated and showed effective performance in treating runoff pollutants [3,7,24]. Gravel and sand filters can generally be used for either large or small catchments whereas small sand filters are best used in areas of high imperviousness. On the other hand reactive filter media such as zeolite and GAC are generally used in filters where high-water quality output is required with removal of organic matter [7].

In a filtration treatment system the removal of dissolved and particulate metals and PAH constituents is primarily accomplished by a variety of physical and chemical processes; sedimentation, precipitation, adsorption, absorption, ion exchange and complexation reactions [12,18,19]. To assess the applicability of filter media for runoff treatment several studies were conducted at laboratory scale and reported excellent removal rates, but full scale application are yet low and most available data are dealt with pilot treatment plants. In the present study we consider literatures mainly dealt with full and pilot scale filtration treatment systems for treating highway runoff from specific roads. One characteristic example is a very interesting publication by Zhou et al. [24] which indicated that the treatment of highway runoff using peat filtration system achieved high removal efficiencies: 95% for PAHs, 70% for dissolved Cu and Zn, 90% for total Cu and Zn, 70% for total Pb as well as promising removal efficiency with respect to TSS. Birch et al. [3] evaluated the efficiency of infiltration basin equipped with 1:6 zeolite/sand mixture. They reported the removal efficiencies of TSS, mineral oil, and total Kjeldahl nitrogen (TKN), Cu and Zn are as moderate to high, ranging from 20% to 80% (mean: 50%), 50 to 75%, 50% to75% (mean: 65%), 49% to 81% (mean: 68%) and −1% to 77% (mean: 52%), respectively.

This paper reports about a study centered on a monitoring program of a filtration treatment system installed for mitigating pollution from parking lot runoff. The main objective was to evaluate the performance of the filtration treatment system in removing TSS, dissolved and particulate heavy metals, PAHs, mineral oil and nutrients from parking lot runoff. The performance of three filter chambers filled with layered filter media of different grain sizes was investigated and the effectiveness of each chamber was determined for the removal of the several pollutant parameters. In the following the description of monitoring data carried out over 18 months in different seasons is reported together with the obtained results.

2. Materials and methodology

2.1. Site and treatment plant description

The pilot treatment plant is located at Gaaden parking lot, near to the highway A21 which is one of the main roads connecting Vienna, Lower Austria-Northern Austria and Eastern Europe. The parking lot basically serves as resting station which is used by long distance travelers whereby heavy lorries dominate over small service cars. It receives stormwater runoff from approximately 4.22 ha in which 1.22 ha (30%) is paved parking area. Owing to its high imperviousness, the largest percentage of the influent was assessed to be runoff from the parking lot.

The filtration treatment system was installed at the end of 2003, covers an area of 45 m² and has three major components: sedimentation and oil separator tanks, three filter chambers, and a control chamber for sampling (Fig. 1). Table 1 presents layering of the filter beds for each filter chamber along with physico-chemical characterization of the filter media. The treatment system was sized to completely capture the initial portion of the runoff volume at a peak flow of 209 L/s resulting from 15 min rainfall with a probability of one event in five years. The sedimentation and oil separator tanks were designed to accomodate a total volume of 22,000 L in which 10,000 L was for settled sediments/sewage sludge. The total volume of the filter chambers was 110,000 L in which 24,000 L was for the filter media. The filter chambers were filled with layers of reactive filter media (composite, zeolite, vermiculite and granular activated carbon (GAC)), and are the components of the treatment device in reducing dissolved and particulate pollutants. Each filter layer is separated by geotextile (300 g/m²). Filter media were selected based on their adsorption capacities towards heavy metals and PAH compounds as well as economic considerations. The runoff is collected by manholes and trench drainage systems and delivered to the treatment plant through a 50 cm internal diameter concrete pipe. The sedimentation tank is located adjacent to the influent pipe and receives the first flow from the influent channel. In the sedimentation tank solids with

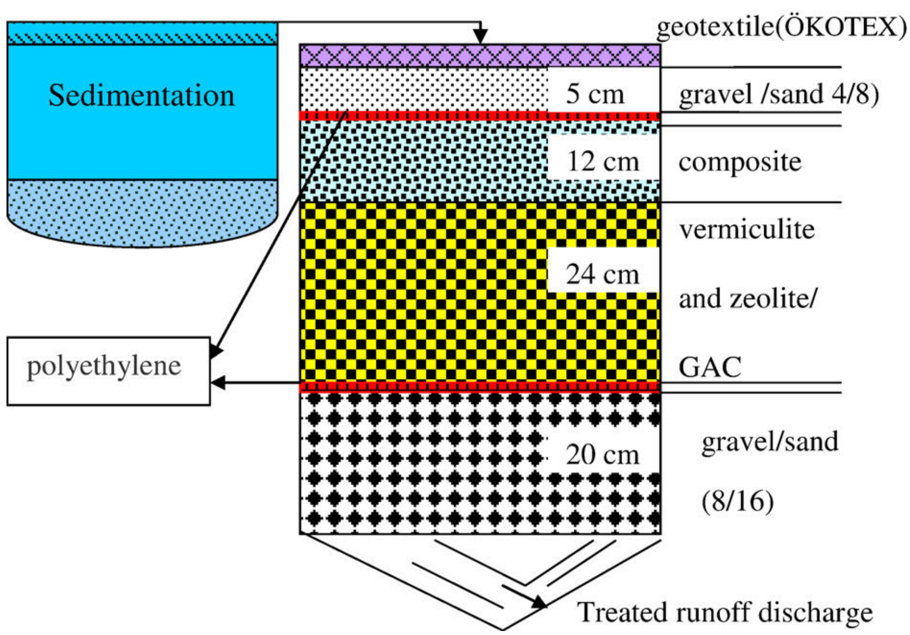

Fig. 1. A general set up of filtration treatment components with layered filter bed.

Table 1
List of the pollutant parameters along with analytical methods and analytical detection limit.

Parameter	Unit	MDL	RDL	Analytical method
Acenaphthene	μg/L	0.17	0.085	DIN38407 T18 (F18)
Acenaphthylene	μg/L	0.002	0.001	DIN38407 T18 (F18)
Anthracene	μg/L	0.005	0.0025	DIN38407 T18 (F18)
Benzo(a)anthracene	μg/L	0.001	0.0005	DIN38407 T18 (F18)
Benzo(a)pyrene	μg/L	0.005	0.0025	DIN38407 T18 (F18)
Benzo(b)fluoranthene	μg/L	0.004	0.002	DIN38407 T18 (F18)
Benzo(g,h,i)perylene	μg/L	0.001	0.0005	DIN38407 T18 (F18)
Benzo(k)fluoranthen	μg/L	0.002	0.001	DIN38407 T18 (F18)
Chrysene	μg/L	0.006	0.003	DIN38407 T18 (F18)
Dibenzo(a,h)anthracene	μg/L	0.001	0.0005	DIN38407 T18 (F18)
Fluoranthene	μg/L	0.013	0.0065	DIN38407 T18 (F18)
Fluorene	μg/L	0.004	0.002	DIN38407 T18 (F18)
Indo(1,2,3-cd)pyrene	μg/L	0.001	0.0025	DIN38407 T18 (F18)
Naphthaline	μg/L	0.011	0.0055	DIN38407 T18 (F18)
Phenanthrene	μg/L	0.017	0.0085	DIN38407 T18 (F18)
Pyrene	μg/L	0.004	0.002	DIN38407 T18 (F18)
pH	–			DIN 38404 T5 (C5)
EC	μS/cm			DIN 27888 T5 (C8)
Mineral oil	mg/L	0.1	0.05	DIN 38409 T18
TSS	mg/L	1	1	(DHIN18 3) 8409 T2 (H2)
TOC	mg/L	0.1	0.1	DIN EN 1484 (H3)
Ammonium–N	mg/L	0.01	0.01	DIN 38406 T5 (E5)
Chloride	mg/L	0.2	0.2	EN ISO 10304
Cd(dissolved)	μg/L	0.02–0.8	0.01–0.4	EN ISO 5961 (E19)
Cd(total)	μg/L	0.2	0.1	EN ISO 5961 (E19)
Cu(dissolved and total)	μg/L	0.5	0.25	DIN ISO38406-7
Zn(dissolved)	mg/L	0.005–0.01	0.0025–0.005	DIN ISO38406-8
Zn(total)	mg/L	0.01–0.11	0.005–0.055	(DEI8N) ISO38406-8

MDL—method Detection Limit , RDL—reporting Detection Limit (if concentration is below detection limit, 50% of MDL were used to calculate the average concentrations).

higher specific density are settled due to gravity effect. After passing the sedimentation tank the runoff flows through the oil separator whereby oil is separated from the runoff water due to variation in density or gravity separation. After flowing through the sedimentation tank runoff filtrates through geotextile filter material of type ÖKOTEX placed on top of the layer filter beds. The geotextile filter material is aided to achieve even distribution of the flow and to reduce sediments or floatable constituents. In the filter chambers, removal of pollutants takes place through processes like adsorption (ion exchange), precipitation, and other complex chemical transformation.

2.2. Sampling and analysis

2.2.1. Water samples

Sampling was conducted at irregular intervals during rainfall events. Eleven runoff event samples were collected from the influent and effluent sections of the treatment plant over a period of 18 months (December 2005–May 2007) with an auto-sampler using a 5 L glass bottle. During a runoff event, influent and effluent samples were collected with the aid of automatic samplers programmed to collect composite samples when flow exceeded a preset level based on signal initiation. Both samplers were started with a difference of 15 min. Water samples were analyzed for content of the following indicator parameters: pH, EC, heavy metals (Cu, Zn, Cd), 16 EPA PAHs, mineral oil, TSS, TOC, NH_4–N, and chloride ion (Cl^-); methods applied and limit of detection (LOD) are given in Table 1.

2.2.2. Sediment samples and assessment of loads

To investigate treatment efficiency of different sections of the treatment plant with respect to removal loads, after 15 months of treatment service on 21.12.2006 and 30.01.2007 three kinds of samples; liquid, solid, and liquid–solid mixture samples were collected from selected sections with in the treatment plant. Sampling points were four from the sedimentation tank, two from the oil separator, and two from the second filter chamber. Sampling from the sedimentation and oil separator tanks was carried out using a vacuum sampling device. To take a representative sample of the whole volume of the tanks, a sampling tube was used to fix (isolate) a defined volume of oil/water/sediment phase from the tank and this sample was sucked out with the help of a vacuum device. Samples were transported to the laboratory and analyzed for metals (Ba, Cd, Cr, Cu, Ni, Pb, Pt, Ti, and Zn) and PAH (fluoranthene , pyrene, benzoanthracene, chrycene, benzo(b) fluoranthene, benzo(a)fluoranthene, benzo(a)pyrene, dibenzo(a,h) anthracene, benzo(g,h,i)perylene, and indo(1,2,3 cd)pyrene) contents. Sampling filter cake from the geotextile filter material was conducted by cutting small square area near to the influent and effluent points. Samples from the geotextile filter material were analyzed only for heavy metals. These sampling techniques enabled the investigation of the amount of pollutants retained in the sedimentation tank, on the geotextile filter material and in the oil separator. The distribution of pollutants within the plant was determined based on area or volume depending on the sample type and sampled section.

2.2.3. Analytical procedures

The pH and electrical conductivity (EC) of runoff event water samples were measured immediately after being transported to the laboratory using pH Meter (WTW pH 196) and conductivity meter (WTW LF196) respectively. Heavy metal concentrations were measured using flame (AAS – Spectr AA 20) and graphite furnace (AAS – Varian Spectr AA 400, Zeeman Graphite Tube Atomizer) Atomic Absorption Spectrophotometer. At its high concentration Zn was measured by inductively coupled plasma optical emission spectrometry (ICP-OES) using a Perkin Elmer Emission Spectrometer Plasma 400. PAHs were extracted and pre-concentrated from runoff samples using liquid–liquid extraction with acetonitrile as an extraction solvent. The concentration of PAHs in the extracts was quantified using reverse phase high pressure liquid chromatography (HPLC), HP1090 Series equipped with fluorescence detection with a capillary column (250 mm × 30 mm id. 5 μm). The concentration of TSS was determined using glass fiber method, TOC with Shimadzu TOC-5000A, NH_4–N using a spectrophotometer (Hitachi U-2000). Detection limits and reference method are given in Table 2.

Results for samples collected from selected sections of the pilot plant were reported both in dry weight (mg kg^{-1}) and wet concentration (mg L^{-1}). Analytical results for wet sludge (filter cake)

Table 2
Composition of filter layers and the physico-chemical properties of the filer materials. K: hydraulic conductivity, ρ_d: bulk density, EC: electrical conductivity.

Filter material		Gaaden				Gravel (sand)	Height
Total volume	5.00 m³	Filter area		13.5 m²		4/8	0.1 m
Total mass	3.46 ton	Filter hight		0.4 m		4/16	0.2 m
Filter material	Grain size	K (m/s)	ρd (g/cm³)	EC (μS/cm)	pH	Mass (kg)	Volume
Chamber 1 and 2							
Composite	1.0–4	3.8×10^{-4}	0.37	859	8.5	616	1665
Vermiculite	1.4–4	3.7×10^{-4}	0.8–1	94	6.5	1415	1665
Zeolite	2–5 (0.6–2)[a]	3.8×10^{-4}	0.86	160	7.3	1432	1665
Total						3463	4995
Chamber 3							
Composite	1.0–4	3.8×10^{-4}	0.37	859	8.5	616	1665
Vermiculite	1.4–4	3.7×10^{-4}	0.8–1	94	6.5	1415	1665
Zeolite	2–5	3.8×10^{-4}	0.86	160	7.3	1432	1665
GAC			0.45			56	125
Total			3519			5120	

[a] grain size of zeolite in chamber 2.

from the geotextile filter material were given in mg kg^{-1} whereas samples from the sedimentation tank and oil separator were given both in mg L^{-1} and mg kg^{-1}.

2.3. Calculation

The efficiency of the system as a whole was evaluated with regard to the concentration difference of indicator parameters at the influent and effluent. The removal efficiency of the filtration treatment device was determined using the following equation:

$$RE = (C_{influent} - C_{effluent}) / C_{influent} \times 100\%$$

where RE is the removal efficiency (in %); $C_{influent}$ is the mean concentration at the influent; and $C_{effluent}$ is the mean concentration at the effluent. For concentrations below detection limit (LOD) a value half of the LOD was assigned for removal quantification. The event mean concentration and total annual load of each parameter measured in the runoff samples was estimated using total rainfall during each event and mean annual rainfall respectively.

For samples from selected sections, the dry matter content of each sampled section was used for conversion dry weight to concentration. The mass of each parameter removed from the filter cake was calculated from dry weight of each parameter, dry matter content (%) and the total area of the pilot plant which is 40.5 m², and masses from the sedimentation tank and oil separator were calculated from the volume of wastewater during the sampling day. The total mass of a parameter removed in the period December 2005 to January 2007 was calculated by summing up the masses removed from each sampled section.

3. Results and discussion

3.1. Temperature, pH and electrical conductivity

Online temperature measurement from the effluent section conducted in the period of October 2006 to February 2007 ranged from 2.9 to 18 °C. In contrast to the online temperature values, meteorological data of the site in winter time ranged from 0 to −5.8 °C. Possible clogging of the filter chambers could occur at the time of low temperatures due to freezing of runoff within the treatment plant. This in turn would result in lowering treatment efficiency and frequent bypass overflow. As can be evidenced from the on-line recorded temperature values, there was no freezing of the runoff even in the winter time. This indicates that the filter chambers of the treatment plant were warmer than the surface temperature or the salt content prevented the freezing.

The pH value of the influent and effluent ranged from 6.9 to 12.3 and 7.9 to 12.2 respectively. The influent and effluent mean and median pH values were 9 and 8.8. The influent and effluent pH values were not statistically different. In most cases the effluent pH was found to be somehow higher than its influent which could be resulted from the composition of the parking lot and leaching of the carbonate rich materials used during parking lot construction.

Measured conductivity values showed high periodic variations over the course of sampling. The influent and effluent conductivities varied from 105 to 59800 µS/cm and from 143 to 14400 µS/cm respectively. Conductivity levels during winter time were one to two magnitudes higher than in summer time. Similarly the concentrations of chloride ion (Cl^-) showed similar variation compared to the conductivity levels and demonstrated a strong relation. Cl^- concentrations ranged from 9.6 to 24700 mg L^{-1} with a mean 5112 and median of 89.4 mg L^{-1}. The higher Cl^- concentrations represent winter samples which are attributed to de-icing salts. For most events the effluent concentrations of chloride were greater than its influent due to the integration of the concentrations in the volume of the tanks. Chloride from road salts caused high EC readings.

3.2. Total suspended solids (TSS)

The influent and effluent concentrations of TSS showed a high degree of variability among event samples. The influent concentration ranged from 53 to 789 mg L^{-1} with a mean and median value of 267 and 188 mg L^{-1} respectively. The effluent concentration ranged from 13 to 133 mg L^{-1} with a mean and median value of 41 mg L^{-1} respectively. The removal efficiency of TSS by the filtration system ranged from 50 to 95% from each filter chamber and during each monitored event. The overall mean removal efficiency of TSS was 85%. Among the filter chambers the second chamber showed an excellent TSS removal (92%) and good (81%) removal of TSS was achieved in the first and third chambers. The removal of TSS during its maximum influent concentration (789 mg L^{-1}) was above 95%. Removal efficiency of TSS was dependent on its influent concentrations, high influent TSS concentrations resulted in higher mean removal efficiency. The mean TSS removal percentage achieved in the present study was generally higher than those obtained in previous studies focused on treatment of urban stormwater and highway runoff [3,5,24]. Birch et al. [3] evaluated the efficiency of an infiltration basin for treating urban stormwater runoff equipped with filter media consisting 1:6 mixture of zeolite and sand reported TSS removal efficiency ranged from 20 to 88% with a mean value of 50% and Zhou et al. [24] concluded promising TSS removal can be achieved with peat filtration for highway runoff treatment. Removal of TSS from highway runoff treated by constructed wetland ranged from 57 to 82% with an average of 69% [5]. The removal mechanisms for TSS are settlement and filtration within the geotextile filter material. Smaller particles that passed the filter fabric are captured in the pore spaces within the filter media in the filter chamber.

3.3. Removal of nutrients, TOC and mineral oil

A summary of mean concentration reduction of mineral oil, TOC and NH_4–N from each filter chamber is presented in Fig. 2. Influent mineral oil concentrations ranged from below LOD (0.1 mg L^{-1}) to 4.4 mg L^{-1} with an equal mean and median value of 2 mg L^{-1}. Except in one event (4.4 mg L^{-1}), the influent concentration of mineral oil was ≤3 mg L^{-1}. The effluent concentrations of mineral oil were below LOD (<0.1 mg L^{-1} or ≤0.5 mg L^{-1}). The mean removal efficiency of mineral oil was >90% in all filter chambers indicating an excellent treatment performance of the filtration treatment device. In a work by Birch et al. [3], the removal efficiency of mineral oil by zeolite-sand filter was moderate (50–75%) which is lower than the efficiency determined in the present study.

TOC is one of the parameters which showed significant concentration variation between event samples. The mean influent and

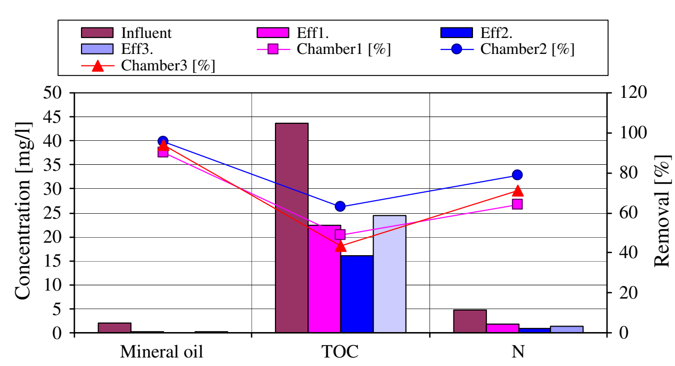

Fig. 2. Comparison of influent and effluent mineral oil, TOC and NH_4–N mean concentrations and their calculated removal efficiency from each filter chamber.

effluent concentrations of TOC were in the range of 2.9 to 113 mg/L and 2.4 to 90.1 mg/L respectively. The mean removal efficiency of TOC was 49%, 63% and 44% from the first, second and third filter chambers respectively. The overall mean removal efficiency TOC is estimated as 52%. TOC, despite of its fair mean removal rate, four event samples (04.06.06, 01.08.06, 04.08.06 and 20.03.07) showed negative removal rates of TOC. This could be due to the integration of TOC in the tank from previous runoff events with high TOC.

The influent concentrations of NH_4–N showed relatively high variation among runoff event samples ranging from 0.73 to 17 mg L^{-1} and the mean effluent concentrations ranged 0.19 to 3.9 mg L^{-1}. For the majority of event samples the effluent concentrations of NH_4–N were below 2 mg/L with a maximum of 2.83 mg L^{-1}. The average removal efficiency of NH_4–N was 64%, 79% and 71% representing the first, second and third chambers respectively. The overall mean removal efficiency of NH_4–N was estimated 71%. The estimated removal efficiency of Kjeldhal nitrogen (TKN) by zeolite-sand filters is moderate (50–75% and mean 65%) [3]. Bulc and Slak [5] reported a mean removal efficiency of 89% for TKN from highway runoff treated by constructed wetland.

3.4. Removal of Cu, Zn and Cd

The performance of the filtration treatment with respect to heavy metals was evaluated for dissolved and total fractions separately. The influent concentration of total Cu ranged from 40 to 430 μg L^{-1} with a mean and median value of 205 and 182 μg L^{-1} respectively. The dissolved Cu ranged from 11 μg L^{-1} to 352 μg L^{-1} with a mean and median value of 129 and 135 μg L^{-1} respectively. The influent concentration of total Zn ranged from <LOD to 1000 μg L^{-1}, a mean and median value of 360 and 294 μg L^{-1} respectively whereas the dissolved Zn ranged from <LOD to 494 μg L^{-1} with a mean value of 182 and 115 μg L^{-1} respectively. The effluent concentration varied between 23 and 99 μg L^{-1} and mean 52 μg L^{-1} for total Cu, 12.3 to 56 μg L^{-1} and mean 35 μg L^{-1} for dissolved Cu, <LOD to 258 and mean 95 μg L^{-1}, and <LOD to 202 and mean 56 μg L^{-1} for dissolved Zn. As indicated in Fig. 3, the removal efficiencies of total Cu and Zn varied between 70 and 83% (mean 75%) and 66 to 81% (mean 73%) respectively. The removal of the dissolved fractions of Cu did not show high variation ranging from 71 to 77 (mean 73%) and 64 to 76 (mean 70%). With respect to Cu one event showed negative removal rate, however the influent concentrations of Cu during this event was the minimum among the monitored events. The removal efficiencies of Cu and Zn were found to be substantially dependent on their influent concentration in which higher removal efficiencies were obtained during higher metal influent concentrations. Overall moderate removal efficiency of Cu and Zn was achieved using filtration treatment device. The influent as well as the effluent concentrations of Cd was below LOD in the majority of samples, therefore Cd removal efficiency was not representative.

The overall performance of the treatment system is the average value of the removal percentages achieved by each filter chamber. In addition to the mean removal a comparison was done between the influent and effluent concentration during each specific event. There is a significant difference between each sampled runoff event for the removal of Cu and Zn, and for Cu low concentrations (<30 μg L^{-1}) could not be removed easily. Removal efficiencies reported in the literature also show variation ranging from negative to >90% removal [3,20].

The mean removal efficiencies of Cu and Zn achieved in the present work were in the same range as reported in literature. For example, Birch et al. [3] reported removal efficiencies varying between 49% and 81% (mean: 93%) and −1 and 77% (mean 52%) for Cu and Zn respectively. Zhou et al. [24] monitored and evaluated the performance of a pilot-scale peat filtration system for treating highway runoff in a karst setting. They reported that at a concentration level of less than 2 mg/L for total Zn, the test indicated a removal efficiency of 90% for total Zn and 70% for dissolved Zn. The filter had similar removal efficiency for copper. Athanasiadis et al. [2] evaluated a pilot infiltration system equipped with clinoptilolite as barrier material for the removal of Cu from roof runoff. The system was able to reduce the concentration of Cu up to 96%. Filtration treatment system using coated quartz achieved removal efficiencies of up to 67% for total Cu and 84% for total Zn, and using sand up to 84% for total Cu and 88% for total Zn [14].

3.5. PAHs

The concentration of total $\sum$ PAH (16 EPA) at the influent ranged from 0.83 to 8.67 mg L^{-1} with a mean value of 3.01 mg L^{-1}. The corresponding effluent ranged from 0.21 to 1.44 mg L^{-1} with a mean value of 0.51 mg L^{-1}. Individual PAH concentrations (fluoranthene, pyrene, and phenanthrene) were comparable to those found in stormwater runoff from a highway (Shynia et al., 2000). PAH concentrations did not show any seasonal variation among summer and winter samples. The mean removal efficiency of $\sum$16 PAHs was as high as 83%. During each monitored events, for individual and $\sum$16 PAHs, there was no occasion whereby the effluent concentration is higher than its influent. Unlike to the heavy metals and TSS, the removal efficiency of PAHs was found to be independent of their influent concentrations. As presented in Fig. 4, the mean removal of individual PAHs ranged from 78 to 99%. The

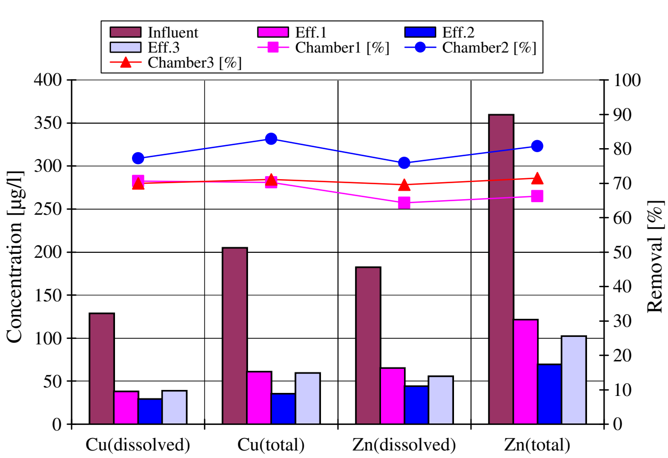

Fig. 3. Comparison of influent and effluent Cu and Zn mean concentrations and their calculated removal efficiency from each filter chamber.

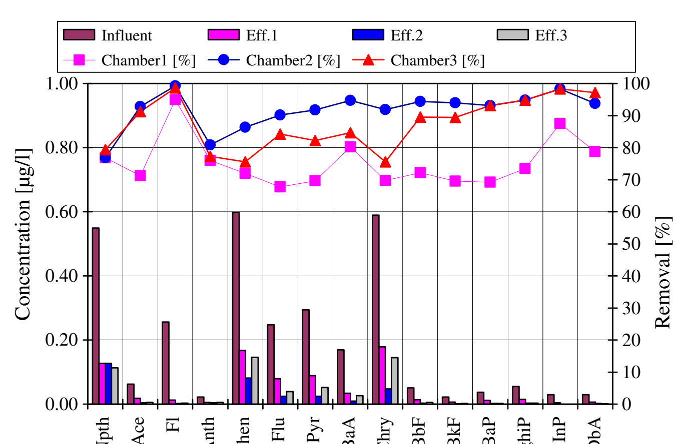

Fig. 4. Comparison of influent and effluent individual PAH mean concentrations and the calculated removal efficiency from each filter chamber.

Table 3
Mass of metal parameters retained in different sections of the treatment plant.

Parameter	Sedimentation tank	Oil separator	Filter cake	Total
Unit	g	g	g	g
Ba	178	17.7	139	334
Ti	175	6.9	168	350
Cd	0.3	0.1	0.4	0.83
Cr	17.4	1.3	19	37.7
Cu	129	11	133	273
Ni	12.8	0.44	12.2	25.5
Pb	27.1	2.84	24.3	54.2
Pt	20.2	2.37	24.4	46
Zn	536	92.8	433	1062
Σ (9 metal mass)	1096	135	953	2183
DM(%)	12	3.4	72	

removal efficiency showed an increasing tendency with increasing molecular mass of PAH compound. Removal efficiencies of PAHs determined in the present study were comparable to efficiencies of infiltration basins [24] and wetlands [22]. In a study by Zhou et al. [24], a pilot-scale peat filtration system for treating highway runoff in a karst setting achieved removal efficiency of as much as 95%. Terzakis et al. [22] investigated the effectiveness of constructed wetland for highway runoff treatment and reported removal efficiencies ranging from 49 to 71% indicating the high performance of filtration treatment method.

3.6. Efficiency of different sections of the treatment plant

3.6.1. Heavy metals

Table 3 and Fig. 5 present the total mass and the percentage distribution of total mass of metals retained in the sampled sections and percent of metal mass distribution removed in the filter cake (Fig. 6). The total mass of nine metal elements (Table 3) retained within the tank and geotextile was 2183 g. The removal mechanisms could be physico-chemical processes, precitation and/or sorption to sediment components of the runoff. The mass of cadmium retained within the sampled sections was insignificant in comparison to other metals whereas the mass of zinc retained is more than one magnitude higher than for all other metals representing 49% of the total mass of metal elements retained in the sedimentation tank and geotextile filter material of the treatment plant. Next to zinc higher amounts of barium, copper and titanium were retained in the sampled sections representing 44% of the total, and lower amounts of chromium, platinum, lead and nickel were retained representing 7%. The sedimentation tank and geotextile filter material showed similar treatment efficiency. The amount removed in the oil separator represents only 1–8% of the total. As calculated from the mean influent concentrations and total runoff volume, the annual load of Cu

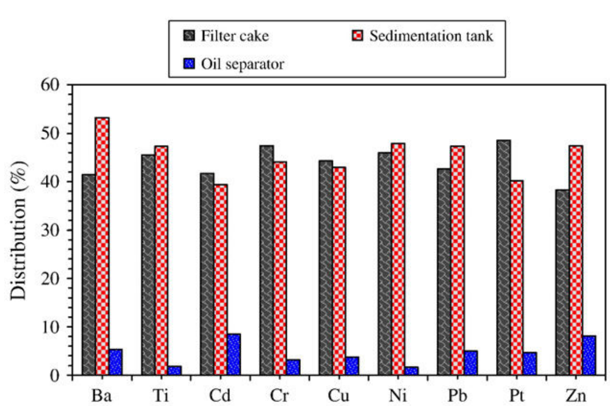

Fig. 5. Distribution of metal parameters removed in different sections of the treatment plant.

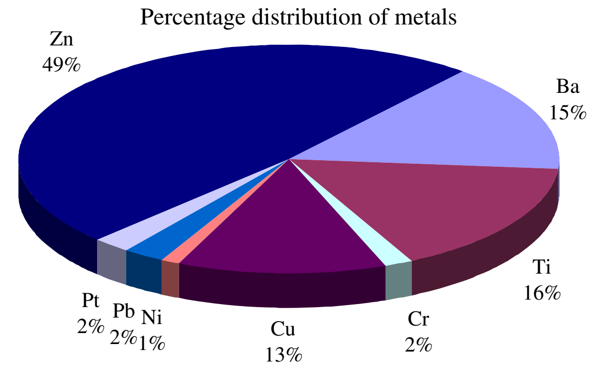

Fig. 6. Mass distribution of metals retained in filter cake (geotextile filter material), sedimentation tank and oil separator.

and Zn were (range) 2.0–2.3 kg/year and (range) 2.9–3.7 kg/year. 60–70% (1.2–1.6 kg) of the annual load of Cu was removed within the filter chambers, but <50% (1–1.7 kg) of Zn within the filter chambers. This indicates that the removal mechnism of Cu was most likely adsorption, ion exchange, or complexetion processes whereas removal of Zn was governed by either physical process such as precipitation or chemical sorbed to the solid component of the runoff.

3.6.2. PAH

The mass of PAH removed within the sedimentation tank and oil separator was calculated from the volume of water, sediments and oil at sampling date, concentrations of PAHs in samples and dry weight of the PAHs (Table 4). The dry weight of the sample was changed to concentration by a converting factor of 0.12 and 0.034 for samples from point Sdt and point Os respectively. Analytical results showed significant variation within replicates which might be resulted due to sampling and handling problem or analytical errors. An average concentration of the replicates was considered for the evaluation. Furthermore samples from the oil separator showed higher concentrations of individual PAH based on dry weight than samples from the sedimentation tank (P). 3.2 g and 856 mg of selected ten PAHs (fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthrene, benzo(a)fluoranthrene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene and indo(1,2,3-cd)pyrene) were removed in the sedimentation tank and oil separator respectively.

The mass distribution percent of PAHs is indicated in Fig. 7. Among the ten assessed PAHs the mass of chrysene removed in the sedimentation tank and oil separator represents 39% of the total mass in the tanks (Σ10PAH = 4057 mg) followed by pyrene with 28%. The amount of benzo(a)pyrene was the lowest and removal of the other PAHs was similar representing 32% of the total. The distribution of the ten PAH in the sedimentation tank and oil separator was similar

Table 4
Mass of PAH's retained in the sedimentation tank and oil separator.

	Sedimentation tank (P)		Oil separator (PA)		Total (P + PA)
Unit	μg/kg	mg (total)	μg/kg	mg (total)	mg
Flu	524	240	4744	139	379
Pyr	1876	860	8728	255	1115
BaA	305	140	1993	58.3	198
Chry	2533	1162	14431	422	1584
BbF	667	306	50	1.46	307
BaF	256	117	1256	36.7	154
BaP	50	22.9	333	9.7	32.7
DbA	367	168	2388	69.8	238
BghiPy	50	23	50	1.46	24.4
InP	50	23	50	1.46	22.4
Total (Σ10 PAH)	6678	3062	34023	995	4057

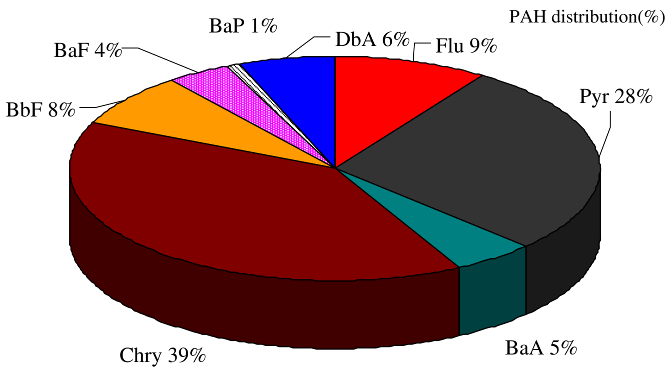

Fig. 7. Measured distribution of PAHs (%) retained in the sedimentation tank.

to the situation of heavy metals. From the total mass of each PAH removed in the sedimentation tank and oil separator, 65 to 96% was removed in the sedimentation tank and the remaining 4 to 35% was removed in the oil separator. No PAH samples were taken from the geotextile filter cake, however considerable mass of total PAH (16 EPA) is believed to be removed in this section either sorbed or attached to the suspended solids.

3.6.3. TSS

The amount of TSS retained (reduced) in the sedimentation tank, oil separator and in the filter cake was calculated based on the volume and dry matter weight of samples. The influent annual load of TSS was 2600 kg. Results showed that 29.3, 458.4 and 506 kg of TSS were removed in the oil separator, sedimentation tank and filter cake respectively. This represents 60% of the influent load; the remaining was removed within the filter chamber. The finer sediments could not be settled easily and the filtrate through the geotextile was being strained within the layered filter media. Mass distribution in the sampled sections (Fig. 8) showed that sedimentation tank and geotextile filter material have similar treatment efficiency.

4. Conclusions

Results of the present study proved that the filtration treatment system installed in Gaaden parking lot, Austria is efficient for the removal several stormwater runoff pollutants. According to the results the system achieved excellent efficiency in removing $\sum 16$ EPA and mineral oil. In addition the system is moderately to highly efficient in removing suspended particulate matter (TSS), heavy metals (Cu and Zn) and nutrient (NH_4–N). Despite its mean fair removal, the removal rate of TOC was significantly varying among events. The observed performance difference among the three filter chambers is due to differences in media mixture composition and grain size. Basically the third chamber with GAC was expected to perform better than the second and first chambers; however our results indicated that the second chamber performed better than both the first and third chambers. The grain size of zeolite in the second chamber was smaller than that of in the first and third. The surface area of a natural sorbent material increases with degreasing grain size which leads to increased adsorption capacity. Hence smaller particles bind slightly more heavy metals than coarser ones. Studies indicated that a decrease in grain size of zeolite causes an increase in adsorption capacity [17,23]. Based on the present results the filter chambers can be ranked as second > third > first. According to results of the present study the grain size of zeolite played a key role with regard to reduction efficiency. Therefore, the variation of these removal efficiencies between the three filter chambers could be due to differences in the filter media grain size and the composition of the layers. Nevertheless, these removal efficiencies are comparable indicating similar mechanisms mainly adsorption could be responsible for the removal of heavy metals and PAHs. It could be concluded that the ability of a filtration treatment system to remove TSS, heavy metals, PAHs, mineral oil and nutrients did not vary significantly. The noted small variability as compared to other similar studies, particularly highway runoff can be a function of pollutant concentration in the runoff, traffic volume, storm duration and its intensity, number of dry period, seasonality, and surrounding land uses. In addition, removal efficiencies of filtration treatment systems depend to a large extent on the capacity of the selected filter media.

Furthermore, the results shown in the present work demonstrate that it is necessary to have an extensive amount of good quality data over a long term to provide useful statistically valid information for the evaluation of a runoff treatment system. The objective of future research will be to offer design criteria for highway runoff and road runoff derived from this present experiences, and linked with the common standards in the literature.

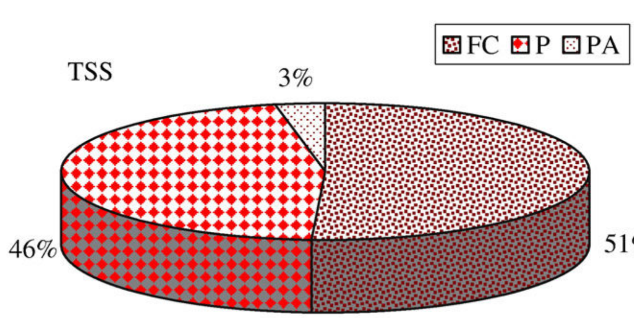

Fig. 8. Distribution of the removed TSS (%) in the filter cake, sedimentation tank and oil separator.

Acknowledgements

This study was conducted under the project "Gewässerschutzanlage für Verkehrflächen." The Project was supported by the FFG project no. 292 and the Company SW Umwelttechnik Stoisser and Wolschner AG, they are gratefully acknowledged for providing funds. The chemical laboratories at the Institutes of Soil Sciences and Sanitary Engineering and Water Pollution Control are acknowledged for conducting the analysis.

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6.2 Paper II

Hydraulic performance and pollutant concentration profile in a stormwater runoff filtration systems.

Haile, T.M., Hobiger, G., Kammerer, G., Allabashi, R., Schaerfinger, B., Fuerhacker, M. (2016). Water Air Soil Pollut (2016) 227:34. DOI 10.1007/s11270-015-2736-4

Water Air Soil Pollut (2016) 227: 34
DOI 10.1007/s11270-015-2736-4

Hydraulic Performance and Pollutant Concentration Profile in a Stormwater Runoff Filtration Systems

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Received: 26 May 2015 / Accepted: 14 December 2015 / Published online: 23 December 2015

Abstract Stormwater filtration system has proven to be effective for the removal of dissolved and particulate pollutants from roadways and car parking areas. However, the long-term treatment performance of filtration systems strongly depends on the hydraulic conductivity and sorption capacity of the filter media. This paper sought to provide information regarding the hydraulic performance, characteristics and metal concentration profiles in sediments accumulated at the surface of filtration systems (SDPL) and core filter media (FMC). Sorption capacity of filter media was used to estimate the lifespan of the filter media. The results showed that saturated hydraulic conductivity of the filtration systems have significantly reduced over the operational time, yet acceptable ($Kf = 5.9 \times 10^{-5}$ to 1.4×10^{-4} m/s). The accumulated sediments (SDPL) were predominantly composed of fine particles with 70 % < 63 μm but the heavy metals were rather uniformly distributed in the different size fractions. The concentrations of heavy metals, particularly Cu, Pb and Zn were significantly higher in the SDPL and decreased with depth of the filter bed. However, Cr and Ni increased with depth of filter media demonstrating their removal was mainly by adsorption. Concentrations of Ba, Mn, Ti and V were comparable to Zn levels indicating comparable concentrations in roadway runoff. Simultaneous adsorption of multiple heavy metals in a column experiment demonstrated that the filter media could remain operational for over 34 years. However, there is a significant concern about their lifespan, particularly due to significant reduction in the hydraulic performance and the possibility of clogging of the systems over time. Therefore, to minimize hydraulic failure, the accumulated sediment be scraped off every 7 years.

Keywords Stormwater · Heavy metals · Filter media · Filtration · Hydraulic conductivity · Sorption

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1 Introduction

Stormwater runoff from vehicle trafficked areas (e.g. highway and parking lot) contains significant loads of metal elements, polycyclic aromatic hydrocarbons (PAHs) and fine particles/suspended solids (Fuerhacker et al. 2011; Göbel et al. 2007; Helmreich et al. 2010). Those pollutants are presented either dissolved in the stormwater or are bound to particulates, particularly in roadway runoff, a large fraction of heavy metals and

PAHs are adsorbed to fine particles (Sansalone and Buchberger 1997; Kayhanian et al. 2012; Gromaire-Mertz et al. 1999; Zgheib et al. 2011). Thus, in addition to the obvious water quality impairment caused by particles such as high turbidity, suspended solids act as the transport vector to downstream areas (Zgheib et al. 2011; Herngren et al. 2006). Numerous previous studies reported that significant loads of heavy metals and PAHs are associated with fine particles typically smaller than 50 μm (Ball et al. 1998; Furumai et al. 2002). For example, (Ball et al. 1998) reported that more than 50 % of the metals be sorbed to particles smaller than 43 μm, although this fine solid fraction only contributed to 5.9 % of the total solids collected. Despite large variations in concentration, most researchers agree that the concentrations of heavy metals and PAHs generally increased with decreasing particle size (Sansalone and Buchberger 1997; Herngren et al. 2006; Zandres 2005; Li et al. 2006; Zhao et al. 2010). In this regard, fine particles are of more concern than larger particles because they have relatively high surface area, which facilitates the adsorption of pollutants (Sansalone and Buchberger 1997; Herngren et al. 2006).

Therefore, the effectiveness of stormwater treatment system depends mainly on the partitioning of pollutants between dissolved and particulate as well as between size fractions of the particles (Kayhanian et al. 2012; Furumai et al. 2002; Zandres 2005). In order to optimize the design and performance of stormwater treatment systems, several researchers have investigated the characteristics of the pollutants in roadside-deposited sediments (Herngren et al. 2006; Zandres 2005; Zhao et al. 2010). Due to particle aggregation and preferential deposition of larger and denser particles, particle size distribution and associated pollutant distribution in sediments are not the same as those of suspended solids in the runoff (Kayhanian et al. 2012; Zandres 2005; Roger et al. 1998). For example, Furumai et al. (2002) showed that particles less than 20 μm accounted for more than 50 % of the particulate mass for runoff samples with TSS concentrations below 100 mg/l. However, for roadside-deposited sediment, Zhao et al. (2010) reported that the median particle size ranged from 100 to 200 μm which is significantly higher than that of highway runoff.

A stormwater treatment system has to ensure both drainage of the runoff and retention of a wide range of particulate and dissolved pollutant loads. A broad review of the international literature has revealed that numerous treatment systems have been constructed, operated and evaluated for the removal of inorganic and organic pollutants (Fuerhacker et al. 2011; Gill et al. 2014; Paus 2999; Ingvertsen et al. 2012). Treatment systems such as sedimentation basin, retention ponds and other structural facilities that mainly rely on sedimentation are not sufficient in the removal of fine particulate and dissolved pollutants. Therefore, different source control strategies such as bioretention (Paus 2999; Li and Davis 2008), infiltration basin (Ingvertsen et al. 2012), constructed wetlands (Gill et al. 2014), media filtration systems (Fuerhacker et al. 2011) and other structural facilities which rely on particle retention and physico-biochemical processes are of interest. Despite their effective performance, some treatment systems demand large areas and are hard to implement in area where available space is limited, for example constructed wetlands.

Infiltration/filtration systems with adsorptive filter media are increasingly considered as an effective alternative for use along highways and parking lots to provide effective removal of dissolved and particulate contaminants (Fuerhacker et al. 2011; Li and Davis 2008; Hatt et al. 2008). These systems are extremely compact and can be retrofitted into existing stormwater collection systems. There are several mechanisms through which contaminants are removed during passage through an infiltration/filtration system: sedimentation as contaminants are attached to solids, filtration, sorption, and transformation (Fuerhacker et al. 2011; Grebel et al. 2013). This indicates that the effectiveness of infiltration/filtration treatment system mainly depends on the hydraulic performance and adsorption capacity of the filter media. Numerous recent investigations relevant to the concentration profiles of heavy metals in in-service stormwater treatment systems showed that metal concentrations decrease with increasing depth of filter bed (Paus 2999; Ingvertsen et al. 2012; Li and Davis 2008). Ingvertsen et al. (2012) investigated the heavy metal profile and hydraulic performance of eight roadside infiltration swales which have been in operation for 6 to 16 years. They reported that the top layer (0–5 cm) of the filtration system was significantly enriched with Cr, Cu, and Zn, but Pb concentrations were similar throughout the soil profile. In another study, Li and Davis (2008) investigated the capture and accumulation of heavy metals in a bioretention media cell receiving parking lot runoff. Results of the study indicated that the heavy metals Cu, Pb, and Zn were predominantly accumulated in the surface deposited street particle layer and significantly decreasing with the media depth.

This research study was undertaken to assess the current state of two media filtration systems receiving parking lot and highway runoff. The objectives of this paper were to investigate (i) hydraulic performance and maintenance requirements,(ii) concentration profiles of metal elements captured in surface deposited particle layer (SDPL) and core filter media (FMC), (iii) size fractionation of surface deposited sediments and concentrations of heavy metals in the different size fractions, (iv) simulate the remaining operational lifespan of the filter media on the basis of the Austrian groundwater target values, QZV-Chemei GW (BMFLU. 98 2010) and or minimum heavy metal removal efficiency requirements as to ÖNORM B 2506-2 (2506-2, ÖNORM B 2012). The study outcomes will contribute to a greater understanding of the treatment performance of filtration systems and in turn enable improved design and maintenance of these systems.

2 Materials and Methodology

In the summer of 2012, a test program was set up to measure the infiltration capacity, composition of surface deposited particle layer (SDPL) and core filter media (FMC) and heavy metal sorption capacity of FMC of stormwater filtration systems.

2.1 Study Sites

Two media filtration systems, namely GSA-H and GSA-W located in upper Austria adjacent to the highway A-21, were investigated after 5 to 7 years of operation. Both systems are underground concrete structures with a sedimentation tank followed by a filter chamber filled with technical filter media, Aquafilt (Fig. 1). GSA-H has been in operation since October 2005 (i.e. 7 years at the time of sampling) and GSA-W has been in operation since 2007 (i.e. 5 years at the time of sampling). GSA-H has a filter drain area of 40.5 m^2 and receives stormwater runoff from a total impervious catchment area of 1.62 ha for which 1.22 ha is paved parking lot. For further details of the detail, see Fuerhacker et al. (2011). GSA-W has a filter drainage area of 65.5 m^2 and receives stormwater runoff from a highway and a bridge covering an impervious catchment of 1.76 ha. The filter media depth in both filter drains was 36 cm. The ratio of the impervious catchment area to the filtration system were 400 for GSA-H and 276 for GSA-W (i.e. GSA-H and GSA-W were sized at 0.25 and 0.37 % of their impervious catchment area, respectively) which is similar to high flow filters sized by Bratieres et al. (2012) to be 0.3 % of their impervious catchment area. To retain suspended solids and prevent early clogging of the system, a geotextile 300 g/m^2 was placed on top of the underlying filter media. Yet, no maintenance activities (e.g. media replacement, geotextile replacement or removal of surface deposited sediment) have occurred at these sites since they were constructed.

2.2 Infiltration Test

The hydraulic performance of the media filtration systems was evaluated by in situ measurement of the infiltration rate using a single ring infiltrometer (14.5 cm inner diameter and 50 cm long) according to Reynolds et al. (2002). Infiltration rate measurements were conducted at the centre of the filter chamber at two points: (a) whole filter bed (i.e. surface deposited particle layer (SDPL) + filter media bed) and (b) filter media bed without surface deposited particle layer. The cylinder was vertically straight pounded (inserted) into the filter bed to a depth of 5 cm and water was poured into the ring keeping a constant head of 10 cm. The volume of water poured into the ring was recorded every 5 min and the test was stopped 15 min after the infiltration rate reached steady state. The saturated hydraulic conductivity of the filter bed was calculated from the infiltration test using the mathematical expression (Reynolds et al. 2002) for a steady flow as follows:

$$K_{\mathrm{fs}} = \frac{q_s}{\frac{H}{(C_1 d + C_2 a)} + \frac{1}{a^*(C_1 d + C_2 a)} + 1}$$

K_{fs} (cm/s): is saturated hydraulic conductivity under field conditions

Q_s (cm/s): measured infiltration rate under submergence conditions

H (cm): height of the stationary water level (constant head)

C_1: dimensionless empirical constant, for $d \geq 3$ cm and $H \geq 5$ cm $C_1 = 0.316\pi$

C_2: dimensionless empirical constant, for $d \geq 3$ cm and $H \geq 5$ cm $C_2 = 0.184\pi$

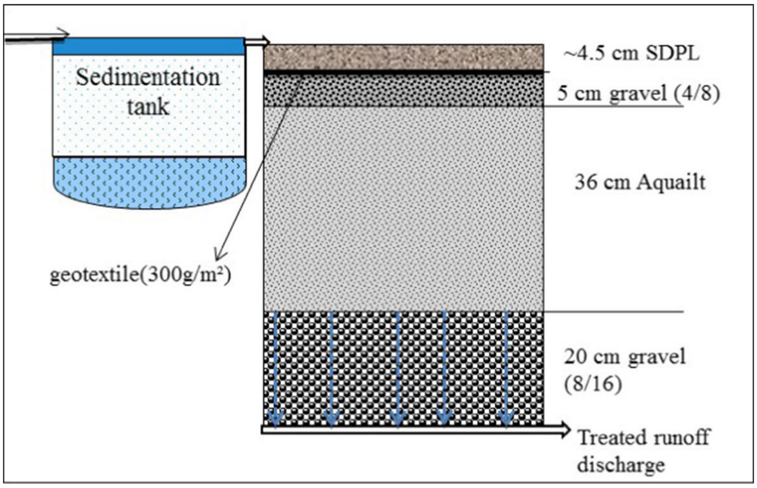


Fig. 1 Overview of a parking lot runoff treatment system GSA-H, Austria

d (cm): inserted depth of the inner infiltrometer

a (cm): inner radius of the infiltrometer

[α*] (cm -1): macroscopic capillary length parameter. α* expresses the relative importance of gravity relative to the capillary force, where large values of α* show the dominance of gravity over the capillary force. α* ranges from 0.01 to 0.36

The determined hydraulic conductivity was then compared to the initial hydraulic conductivity of the filter media measured at laboratory before its installation.

2.3 Sampling

To assess the concentration profiles of pollutants, sediment (SDPL) and filter media core (FMC) samples were collected from GSA-H and GSA-W using precleaned infiltrometer cylinder. The average thickness of the SDPL was 4.5 cm at GSA-H and 2.5 cm at GSA-W, respectively. Since the area of the filtration system was relatively small (40.5 m^2 for GSA-H and 65.5 m^2 for GSA-W) and thickness of the deposited particle layer was uniform, spatial variation of pollutants was not considered. Therefore, two sampling points at the centre of the filter area were considered as representatives for the SDPL. To investigate the vertical variability of pollutant concentrations with depth, filter media cores (FMCs) were collected at three depths (0–8, 8–20, and 20–36 cm). Samples were collected in plastic bags and placed in a cooling room at 4 °C till analysis.

2.4 Particle Size Distribution of the Deposited Sediment

To assess the particle size distribution, the bulk sediment sample collected from the SDPL (GSA-H and GSA-W) and street dirt collected from road-deposited sediment (Lienz, Austria) were air-dried for 2 weeks to a constant weight. The air-dried SDPL and street dirt bulk sample were then gently crushed and dry sieved through a 2000-μm stainless-steel sieve. The particles with diameter less than 2000 μm were wet sieved into two fractions, i.e. particle size of less than 63 μm and greater than 63 μm. The greater than 63 μm sample was further wet sieved into fractions: 63–125, 125–200, 200–630, 630–1000 and 1000–2000 μm. The finer particles with diameter less than 63 μm were fractionated into four fractions <2, 2–6.3, 6.3–20, and 20–63 μm using a pipette method.

The distribution of heavy metals in the SDPL from GSA-H was analysed for fractions; <63, 63–200, 200–630, 630–1000 and 1000–2000 μm. Note that only 2 % of the bulk sediment sample has particle size of greater than 2000 μm in which its effect on the heavy metal fractionation could be negligible.

2.5 Heavy Metal Removal and Lifespan of Filter Media

To quantitatively determine the remaining metal sorption capacity (lifespan) of the filter media in operation, simultaneous adsorption of multiple heavy metals (Cr, Cu, Ni, Pb and Zn) was carried out in a fixed bed column experiment using core filter media (FMC) from GSA-H. The experiment was conducted in a glass column which had an internal diameter of 32 mm and filter bed height of 240 mm (i.e. empty bed volume (BV) of 193 cm^3). To minimize wall effect on sorption test results, Inczédy (1966) suggested that the column diameter on particle diameter ratio should be greater than 10. In this study, the diameter of the column was approximately 20 times greater than the mean particle size of the filter media, so the wall effect could be ignored. The FMC was packed in the column in a systematic way proportional to the layering of the filter bed in real system. A glass bead and fritted glass filter was placed at the bottom of the column to support the adsorbent. Multi-metal column feed solution containing Cr, Cu, Ni, Pb and Zn with desired concentrations was prepared using analytical grade 1000 mg/l stock solutions ($CuCl_2$, $CrCl_3$, $Pb(NO_3)_2$, $NiCl_2$ and $ZnCl_2$) and in distilled water. pH of the influent multi-metal solution was adjusted to 5.8 ± 0.2 using either suprapure HNO_3 or NaOH. This pH value was selected at least to resemble the minimum pH of highway runoff. A summary of the heavy metal concentrations in roadway runoff (Göbel et al. 2007; Helmreich et al. 2010) and column influent, calculated column annual load and filter media exhaustion point is given in Table 1.

The annual load of a target heavy metal in the column experiment was calculated based on the scaling of the filtration system using the following equation:

$$\text{annualload(mg per yer)} = C_{\text{in}}APr$$

Where C_{in} is mean roadway runoff concentration in μg/l obtained from literature (Table 1), A is surface are of the column in m^2, P is mean annual precipitation of the sites (720 mm per year) and r is the ratio of total impervious catchment area to filtration system which is 400 for GSA-W and 276 for GSA-W.

The experiment was conducted in up-flow modus (from bottom to top) which ensures saturated flow conditions and uniform hydraulic distribution of the sorbate to the filters (Athanasiadis 2005). Prior to loading with the feed solution, the column was slowly saturated and flushed with distilled water for 2 h, in order to remove entrapped air bubbles and leachable background concentrations. The feed solution was pumped using a peristaltic pump (ISMATEC IDEX) at a constant flow rate of 50 ml/min. Effluent samples were collected at designated time intervals, filtrated, acidified to pH <2 with 0.01 mM HNO_3 suprapure and analysed for heavy metal concentrations. Influent samples were also collected and handled similar to the effluent samples.

The performance of the FMC in reducing the heavy metal levels from synthetic solutions was assessed using the following equation:

$$\% \text{ metal removal} = [C_i - C_e]\text{x}100$$

where C_i is the measured metal concentration of the column influent (initial) and C_e is the metal concentration of the effluent after a known volume of flow through.

The column was loaded till it reached the exhaustion point. In the present study, a filter media exhaustion was set to be equal or lower than the maximum effluent concentration defined in the Austrian regulation for ground water protection (BMFLU. 98 2010) or removal efficiency is below the minimum requirement according to the Austrian Standard (2506-2, ÖNORM B 2012) as indicated in Table 1.

2.6 Analytical Procedures

For the analysis of concentrations of metal elements in the SDPL and FMC, 500 mg of air-dried sample was added into 10 ml of ultrapure water and acidified using 3 ml 30 % HCl and 2 ml 65 % HNO3 in a closed Teflon vessel, and was digested in a Microwave Digestion System. The digested solution was then cooled for 2 h. After cooling, the digested samples were filtered and analysed for total concentrations using an inductively coupled plasma mass spectroscopy (ICP-MS; Perkin-Elmer, Sciex).

For the analysis of metal element concentration in the different size fractionations, SDPL sample was sieved through stainless-steel test sieves: 2000, 1000, 630, 200, and 63 μm. Material greater than 2000 μm was discarded because it contributes only <2 % of the total mass and also the fine fractions were of particular interest. 100 mg of SDPL dry sample was weighted in Pt-dishes and mixed with 2.5 ml HNO3 (65 %), 2.5 ml HClO4 (60 %) and 5 ml HF (40 %) and the acid was concentrated near dryness. After that, the residue was heated two times with 5 ml HNO3 (65 %) until the fumes were emitted. The last step was to take the residue with 0.5 ml HNO3 (65 %), diluted to 50 ml H2O and analysed for total concentrations using ICP-MS 7500 ce (Agilent). To guarantee the quality of measurement, some controlling standard was also measured. All concentrations were reported on a dry-weight basis (μg/kg). Because of their higher relevance regarding stormwater

Table 1 Concentrations of heavy metals, column annual load and material exhaustion criteria according to discharge limits and removal efficiencies

Metal	Concentration (μg/l)		Annual load (mg/yr)	Filter media exhaustion point	
	Highway runoff (Göbel et al. 2007; Helmreich et al. 2010)	Column	Column	GW limit (μg/l)[a]	RE (%)[b]
Pb	<5–405 (43)	50	9.68	9	–
Cr	6–50 (27)	50	3.93	45	–
Cu	11–604 (100)	250	22.5	1800	80
Ni	4–403 (35)	50	5.1	18	–
Zn	15–3470 (400)	1000	90		50

Values in bracket represent median concentration

[a] Maximum effluent concentration for the protection of groundwater according to the Austrian regulation (BMFLU. 98 2010)

[b] Minimum removal efficiency according to the Austrian Standard (2506-2, ÖNORM B 2012)

runoff from trafficked areas, particular emphasis was given to Ba, Cd, Cr, Ni, Cu, Pb, Ti, V and Zn.

3 Results and Discussion

3.1 Hydraulic Performance

The mean hydraulic conductivity of the whole layer (SDPL plus filter media layer) was 3.5×10^{-6} m/s and 4.8×10^{-5} m/s for GSA-H and GSA-W, respectively. After scraping of the SDPL, mean hydraulic conductivity of the filter media layer was 5.9×10^{-5} m/s and 1.4×10^{-4} m/s for GSA-H and GSA-W, respectively. Hydraulic conductivity of the fresh filter media tested at laboratory before its installation ranged 3.8×10^{-4} to 5.2×10^{-4} m/s (Haile 2008). The results indicated that hydraulic performance of the filtration system decreased in 1 to 2 orders of magnitudes after 5 to 7 years of operation. Infiltration rate at GSA-W was higher than that measured at GSA-H. The operational time and thickness of SDPL at GSA-H (4.5 cm) was relatively higher than at GSA-W (2.5 cm). The observed slight variation in hydraulic performance could be related to the operational time, thickness of SDPL and amount of fine particles strained within the filter media layer. The hydraulic conductivity of the SDPL plus filter bed was one magnitude lower than filter bed. This indicates that SPDL and particles strained in the filter bed have played a vital role in reducing the hydraulic conductivity of the filtration system.

The SDPL has an organic carbon content of 21 %. The very high organic carbon contents were mainly attributed to tire and break pad dusts, oil leaks, asphalt particles and other organic sources such as plant residues. After 7 years in operation, organic carbon content of the original filter media has increased from less than 1 % to 7 %. The increase could be a result of fine particulate matter transported by runoff being deposited in the filtration system. Sieve analysis and mass measurement of core filter media showed that 20 % of the pore volume was occupied by fine particles strained within the filter bed. Since particle straining in the filter bed has resulted in reduction of the pore volume, this in turn decreases the infiltration rate or hydraulic conductivity. The decrease in hydraulic conductivity could be also related to the compaction of the filter media due to hydraulic loading. Despite the significant decrease in hydraulic conductivity, these values still comply with the Austrian design standard value which ranged 1×10^{-3} m/s to 1×10^{-5} m/s (2506-2, ÖNORM B 2012). Thus, stormwater filtration systems like the ones investigated in the present study could remain operational for at least 7 years without maintenance.

3.2 Particle Size Distribution SDPL and Street Dirt

The particle size distributions of SDPL from stormwater filtration systems and street dirt from urban roadway are shown in Fig. 2. The characteristics and particle size distribution of SDPL from GSA-H and GSA-W were similar. The particle size distribution of SDPL ranged from <2 to 2000 μm with a median particle size (d50) of

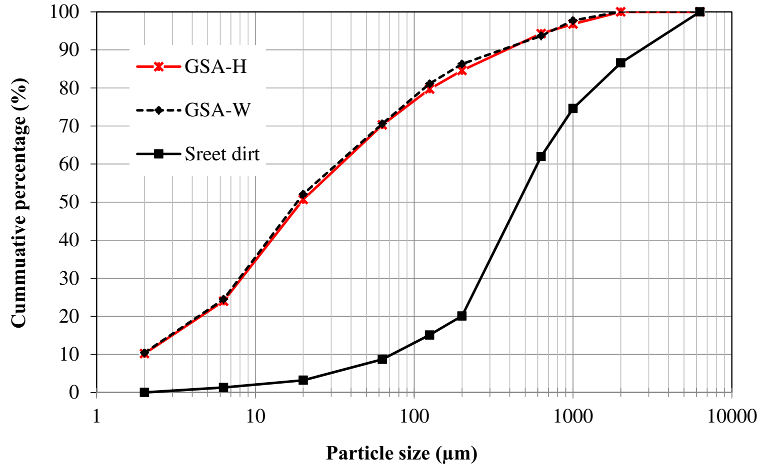


Fig. 2 Mass distributions of ten particle size range SDPL for two stormwater filtration systems, highway runoff and street dirt

20 µm. The particle size of street dirt ranges from <6.3 to 6300 µm and has a d50 of 480 µm.

A histogram of the size fractionations as a function of the particle size is shown in Fig. 3. The particle size distribution results showed that SDPL was predominantly composed of fine particles in which over 70 % the total mass was <63 µm, 23 % was <630 µm and only 7 % was >630 µm. Particle size distribution of the street dirt showed the following characteristics: 9 % of particles were <100 µm, <63 µm, 53 % were <630 µm, 25 % were <2000 µm and 13 % were <6300 µm.

Over all the results revealed that the size distribution for SDPL was narrower than the range for street dirt, indicating less variability in the particle size for SDPL. The observed significant variation between SDPL and street dirt can be explained by two main reasons. Firstly, the street dirt typically have accumulated over extended periods and they are generally enriched with coarser particles; secondly, the finer material having been lost through wind dispersion and/or washed away by rain (Zandres 2005). This means that the fine sediment fraction is underrepresented in street dirt. Secondly, the street dirt typically includes pollutant material derived from a range of sources, not just vehicle activity.

3.3 Metal Content of Surface Deposited Sediments and Core Filter Media

The total concentrations of metals measured in the bulk SDPL and FMC samples area are summarized in Table 2. The vertical distribution of the metals measured in the SDPL and FMC profile are presented in Fig. 4. From the vertical distribution, it can be seen that concentrations of Cu, Pb, V, and Zn were high in the SDPL and decreased with depth of the FMC. The concentrations in the SDPL were 2 to 4 times higher than the levels in the filter

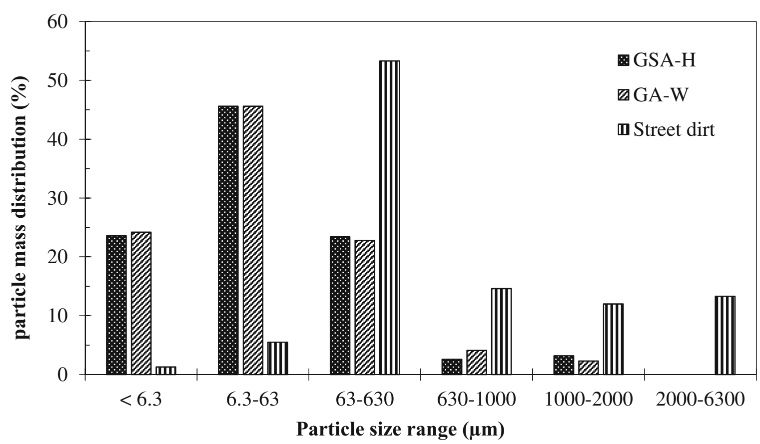


Fig. 3 Histogram of mean mass percentages of sediments from surface deposited particle layer versus grain size fraction

Table 2 Concentrations of metal elements in the SDPL and FMC

Metal	Unit	SPDL		FMC	
		GSA-W	GSA-H	GSA-W	GSA-H
Li	mg/kg	19	18	5.8	13
B	mg/kg	25	27	15	14
Ti	mg/kg	589	565		
V	mg/kg	43.6	37	3.4	7.8
Cr	mg/kg	86	81	37	83
Mn	mg/kg	499	744	160	350
Co	mg/kg	10.3	7.7	16	24
Ni	mg/kg	52	46	50	73
Cu	mg/kg	326	490	22	47
Zn	mg/kg	3016	1149	241	123
Sr	mg/kg	55	216	210	176
Cd	mg/kg	0.49	0.6	0.05	0.19
Ba	mg/kg	135	325	481	404
Pb	mg/kg	114	57	8.1	10.8
Bi	mg/kg	3.4	3	0.21	0.27
Al	g/kg	12.3	10.9	27	26
Fe	g/kg	32.9	37.9	18	28

media layer. Such vertical stratification of the pollutants was expected because as the runoff water infiltrates through the filter media layer, pollutant concentrations should decrease due to removal via filtration, precipitation, sorption and complexation processes. However, the contents of Cr, Co and Ni generally increased with depth from top to bottom (SDPL to FMC). The metals B, Ba, Mn, and Sr were uniformly distributed over the SDPL and filter media.

In the SDPL following Al and Fe, Zn was the next highest whose concentrations were significantly higher (i.e. 3–58 folds) than Cr, Ni, Pb and Cu concentrations (Table 2). This was also comparable to the concentration ratios in highway runoff. With respect to highway runoff, Cu and Zn are the most relevant and frequently measured heavy metals (Göbel et al. 2007; Zandres 2005; Ingvertsen et al. 2012; Li and Davis 2008). Considering the concentration ranges from Göbel et al. (2007), the concentration ratio of Cu/Zn ranged from 0.05 to 0.24. This was similar to the Cu/Zn ratio in the SDPL which ranged from 0.07 to 0.34. However, the Cu/Zn ratio in the FMC samples were higher ranging from 0.34 to 0.9 which indicates that Cu removal by adsorption process in the filter media layer is higher as compared to Zn.

As can be seen in Table 2, the heavy metal concentration dataset did not show any direct relation with the operation time (age of the treatment system), thickness of SDPL, or ratio of impervious catchment area to filter area. The levels of heavy metals, specifically Zn, Pb, and Cu varied among the sites. The thickness of the SDPL as well as age of GSA-H was relatively higher than that of GSA-W; however, the concentrations of Zn and Pb at GSA-W were 2 folds higher than that found at GSA-W, Cu was higher at GSA-H, but Cr, and Ni concentrations were very comparable. Grab runoff water sample analysis indicated that the concentrations of Cu and Zn measured at GSA-W were up to 3 folds higher than the levels at GSA-H (data not indicated).

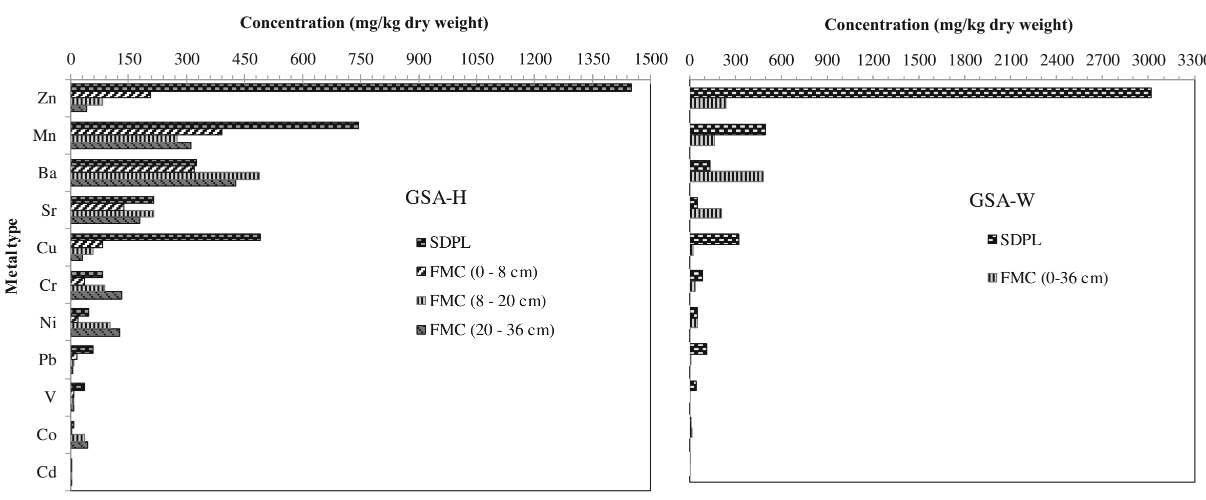


Fig. 4 Profiles of heavy metals in SDPL and filter media layer collected from two media filtration treatment systems

Average daily traffic density, seasonal influences, type of asphalt, motor vehicle types, frequent congestion, braking or acceleration are all factors that cause site-to-site variability in the concentrations of roadway run-off pollutants (Helmreich et al. 2010; Sansalone and Buchberger 1997; Ingvertsen et al. 2012). The variations observed in metal concentrations in SDPL and FMC samples could be attributed to the aforementioned factors. Despite those variations, the distribution of the heavy metals in both stormwater filtration systems was very similar.

3.4 Metal Concentrations as a Function of Particle Size

The average metal concentration in each particle size fraction is shown in Table 3. As can be seen, except for slight higher concentrations in the size fraction 200–630 μm, no clear trend in metal concentrations dependency on size fractionations could be observed. This indicates that the metals were uniformly adsorbed to the whole particle size range (0–2000 μm). The average concentrations of Cr, Cu, Ni, Pb, and Zn in all size ranges were in the range of 131–159, 301–342, 47–8, 105–148 and 2692–3265 mg/kg, respectively. It is important to note that over 90 % of the SDPL was comprised of particles with diameter smaller than 630 μm, thus most of the metal loads were captured in this fraction.

The metal load of the whole sample and the contribution to this load of each particle size fraction has been calculated using the metal concentration data (Table 3) and the particle size distribution (Fig. 2). The metal load contributed by each particle size fraction to this load is shown in Fig. 5. Results showed that the majority of metal masses resided in particles smaller than 63 μm (Fig. 5). In the particle mass distribution analysis, over 71 % of the total mass of SDPL was represented by the smallest fraction (<63 μm) and over 60 % of the trace metals were bound to the fine fraction (<63 μm) of SDPL. Low percentages of metal mass in the larger size fraction (>630 μm) were primarily due to the fact that less than 10 % of total mass of SDPL was associated with the particle size greater than 630 μm. The results indicated that particle treatments, specifically removal of the fine size fraction <200 μm plays a vital role in removing pollutants from roadway runoff. Several researchers reported that highest metal concentrations were consistently found in the fine particle size fractions (<250 μm) (Zandres 2005; Zhao et al. 2010). In road-deposited sediment samples, Zandres (2005) observed that a high percentage of the total metal load was associated with particles smaller than 125 μm (64 % of Zn, 57 % of Cu and 46 % of Pb). Similarly, Zhao et al. (2010) reported that 80 % of the heavy metals in road-deposited sediments were mainly associated with the <250 μm size fraction.

3.5 Remaining Heavy Metal Removal Efficiency

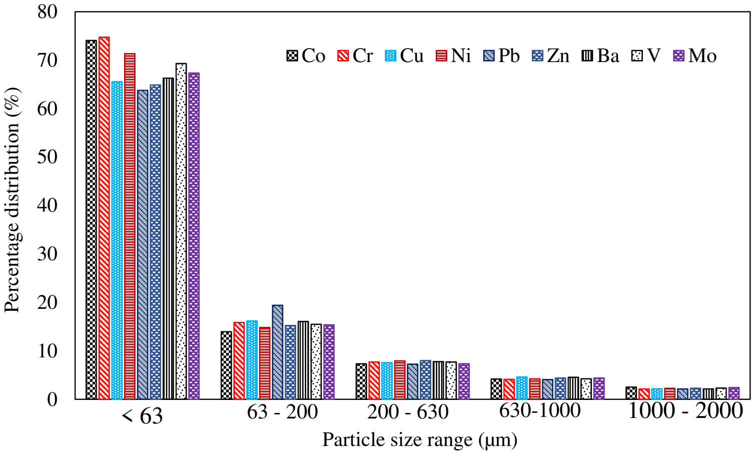
The performance of the core filter media from GSA-H in reducing the heavy metal levels from the synthetic solution is presented in Fig. 6. The column experimental data indicated that the filter media in operation still has high affinity for the removal of Cr, Cu, Pb, Ni and Zn. Over 90 % removal efficiency of all five tested heavy metals was achieved till a total flow through of 2800 BV

Table 3 Total metal concentrations determined for each particle-size fraction of SDPL collected from GSA-H and the metal concentration of the bulk SDPL sample

Particle size (μm)	Total metal concentration (mg/kg)														
	V	Cr	Co	Ni	Cu	Zn	Rb	Sr	Zr	Mo	Cd	Sn	Sb	Ba	Pb
<63	88	157	15	53	301	2692	46	135	153	11	<1	63	31	329	108
63–200	88	150	12	50	335	2847	51	142	122	11	<1	64	31	359	148
200–630	96	159	14	58	342	3265	58	141	114	12	<1	70	35	379	121
630–1000	81	131	13	47	321	2771	54	127	118	11	<1	62	30	342	105
1000–2000	89	136	15	51	304	2900	51	132	114	12	<1	64	31	323	109
Whole sample[a]	88	147	14	52	321	2895	52	135	124	11	<1	65	32	346	118

[a] Due to the uniform heavy metal distribution in different size fractions, the mean values were considered as concentration of the whole sediment

Fig. 5 The contribution of each particle size fraction to the total metal load in SDPL



(i.e. 540 l). A selectivity series can be determined as follows: Pb > Ni = Zn > Cu > Cr. The removal efficiency of Cr, Cu, Ni and Zn slightly decreased after a total flow through of 3500 BV (675 l). However, removal of Cr was significantly reduced to less than 20 % and after a total flow though of 4440 BV (860 l) and full breakthrough (i.e. $Ce/C0 = 1$) was achieved after applying 6520 BV (1260 l). Regarding road runoff treatment, the Austrian standard (2506-2, ÖNORM B 2012) has set a minimum removal requirements of 80 % for Cu and 50 % for Zn removal. After the passage of 5070 BV (980 l), the removal of Cu was consistently below 80 % but Zn removal was still greater than 77 % which is above the requirement.

The effluent concentrations of Cu, Ni, Pb and Zn were reduced to significantly low levels during the early loading but have slightly increased over the course of the experimental run (Fig. 6). Nevertheless, till the end of the experiment, the effluent concentrations of Pb and Ni were still below maximum effluent concentrations set in the Austrian regulation regarding groundwater protection (BMFLU. 98 2010). The column influent and stormwater concentration matrix for Cr is low (<50 μg/l) which is comparable to the maximum effluent concentration set in the Austrian regulation (i.e. 45 μg/l). As a result, Cr values might not be representative for the lifespan estimation. The test was subsequently terminated when 6800 BV (1310 l) feed solution has been applied, as the removal of Cu has remained consistently less than 80 % which is below the minimum requirement as to the (2506-2, ÖNORM B 2012). Therefore, in this study, the sorption capacity of the filter media used in the filtration systems would be limited by the removal of Cu.

3.6 Remaining Lifespan of Filter Media for Heavy Metal Removal

The column sorption data (Fig. 6) and annual load of heavy metals (Table 1) were used to estimate the remaining metal sorption capacities in the field filtration system. In roadway runoff, large fractions of the heavy metals, typically >50 % of Cr, Cu, Ni and Zn and >80 % Pb, exist in particulate form (Helmreich et al. 2010; Gromaire-Mertz et al. 1999; Ball et al. 1998). This was also evidenced in the present study that large fraction of the total load of Cu, Pb and Zn was particle bound being captured in the SDPL (see section 0). Therefore, to calculate the remaining lifespan of the filter media, it was assumed that only 50 % of the metal loads/concentrations are in dissolved fraction. The dissolved fraction of heavy metals could be effectively removed by the filter media mainly via mechanisms such as sorption, filtration, ion exchange and complexation reactions (Fuerhacker et al. 2011; Sansalone and Buchberger 1997). The lifespan of each metal was then obtain by dividing the total amount of a metal adsorbed till FMC exhaustion or experiment termination by the annual load. The remaining lifespan of the filter media simulated in column sorption is shown in Table 4. Over the experimental running time, filter media exhaustion was observed for Cu and Cr, but effluent concentrations of Ni and Pb as well as removal efficiency of Zn met the requirements. The results demonstrated that Cr and Cu sorption capacities are the determinant factors limiting the lifespan of the filter media. Thus, the filter media could remain operation for up to 34 years. It is important to note that removal of the dissolved fractions in the surface deposited sediment is expected by processes

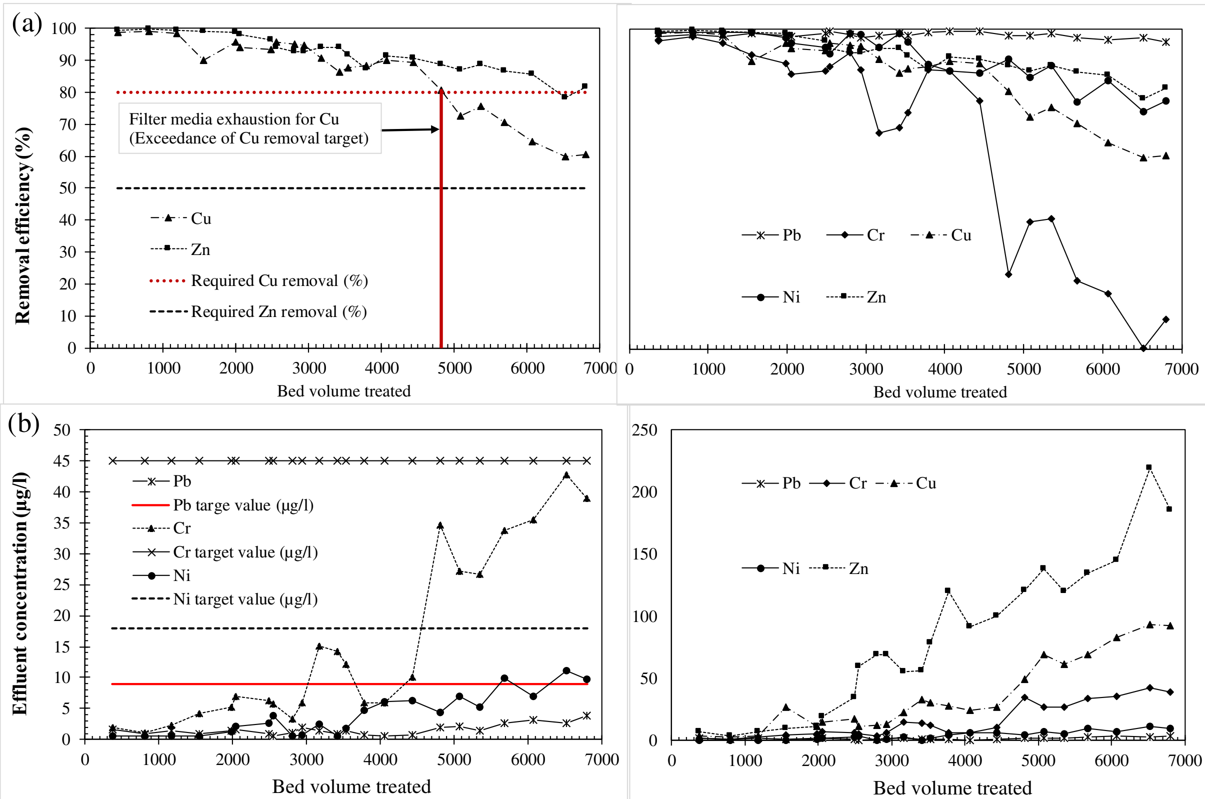


Fig. 6 Removal efficiency (%) of heavy metals (**a**) and effluent concentrations (**b**) versus column flow through number of bed volumes (1 BV = 193 ml). The lines are not fitting functions; they connect points to facilitate visualization

such as surface complexation and precipitation. As a result, the lifespan would be higher than the calculated ones.

Over the experimental running time, filter media exhaustion was observed for Cu and Ni, but effluent concentrations of Ni, Pb and removal of Zn met the requirements. The results demonstrated that Cr and Cu sorption capacities are the determinant factors limiting the lifespan of the filter media. Thus, the filter media could remain operation for up to 34 years. It is important to note that removal of the dissolved fractions in the surface deposited sediment is expected by processes such as surface complexation and precipitation. As a result, the lifespan with respect to heavy metal would be higher than the computed ones.

The long-term sorption capacity and lifespan of a filtration system depends on the composition of the filter media. The Aquafilt used in the investigated filtration

Table 4 Summary of the column sorption results and calculated lifespan of the core filter media from GSA-H

Heavy metal	Cr	Cu	Ni	Pb	Zn
Mean column influent (µg/l)	47	250	50	85	1020
Column annual load (mg)	3.93	22.5	5.1	11.3	90
Total loaded into column (mg)	88	380	91	166	1999
Column maximum effluent (µg/l)	43	93.7	16.1	3.8	219
Targeted maximum effluent (µg/l) (BMFLU. 98 2010)	45	1800	18	9	–
Removal efficiency at FMC exhaustion (%)	<10	<80	68	>97	>77
Minimum removal efficiency (%) (2506-2, ÖNORM B 2012)	–	80 %	–	–	50 %
Years to exhaustion	45	34	36	34	44

systems is composed of several filter media for which zeolite is one of the components. Numerous researchers reported that zeolite has promising performance for the removal of heavy metals from stormwater runoff (Fuerhacker et al. 2011; Wium-Andersen et al. 2012; Genç-Fuhrman et al. 2007). Column sorption experiment with fresh unloaded Aquafilt similar to that installed at GSA-H showed high heavy metal removal efficiencies (data set not indicated). The metal sorption capacity of the filter bed in the filtration system might possibly increase overtime due to the enrichment of organic matter content transported with the incoming runoff particles. In addition, filtration of particles in the pores of the filter bed will cause reduction in infiltration, and this in turn increase the rate so that the retention time of the polluted runoff. The increased retention time may result in greater adsorption of dissolved metals.

4 Discussion

The SDPL acts as a filter itself, which enhances pollutant removal, but it obviously reduces the hydraulic performance of the filtration system. The geotextile was placed on top of the filter media to act as a barrier to remove particles from reaching and clogging the filter bed. The accumulation of the fine particles in the SDPL appears to confirm that the geotextile itself was performing much of the filtration. Removal of the SDPL on a regular basis, (approximately every 7 years) and replacement of the geotextile 300 g/m^2 would be very important to maintain an acceptable infiltration rate. Geotextile layers placed in a stormwater treatment system enhanced treatment performance; however, it adversely impacted the hydraulic performance of the system (Paul and Tota-Maharaj 2015). Back flushing was recommended as an effective technique to remove the clogging particles deposited on the geotextile layer and recover the hydraulic conductivity (Paul and Tota-Maharaj 2015; Koerner and Koerner 1992). Based on laboratory experiments, some researchers (Hatt et al. 2008; Clark and Pitt 2009) suggested that scraping the top layers of sediment from stormwater infiltration systems periodically can reduce clogging and restore hydraulic performance, though hydraulic recovery was reported to be incomplete (Hatt et al. 2008) or unsustainable (Clark and Pitt 2009) in laboratory experiments. Mousavi and Rezai (1999) investigated infiltration basin (used for recharging to groundwater) constructed on sandy-loam soils and observed that scraping off the top 5 cm (deposited sediment layer and top soil) resulted in a restoration to about 40 % of the original infiltration capacity.

The particle size distribution of SDPL was generally in close agreement with the size distribution reported for highway and motorway runoff (Furumai et al. 2002); however, the median particle size of SDPL in this study was lower than those found for sediments accumulated on stormwater detention basin (Sébastian et al. 2014) and road-deposited sediments (Herngren et al. 2006; Zhao et al. 2010). Several previous studies found that in roadway runoff, fine particles less than 50 μm in diameter accounted for more than 70 to 90 % of the weight of the TSS load carried by runoff (Li et al. 2006; Haile et al. 2014; Andral et al. 1999). Furumai et al. (2002) reported that the median particle size of particles from urban highway runoff was 20 μm which is equivalent to the d50 of SDPL. However, the d50 of the SDPL was lower than that of sediments accumulated on stormwater detention basin which ranged from 24 to 50 μm (Sébastian et al. 2014) and d50 of road-deposited sediments ranged from 100 to 200 μm (Herngren et al. 2006; Zhao et al. 2010). This suggests that in road-deposited sediments, the particles with diameter smaller than 200 μm are of particular concern but in roadway runoff, the fine particles (<50 μm) are more important. The results of the present study and literature survey values (Haile et al. 2014) indicate that suspended solids carried by stormwater runoff and sediments accumulated within stormwater treatment systems are usually smaller in size than street dirt and road-deposited sediments. In the literature, the range and median particle size distribution of solids in roadway runoff, road-deposits and sediments accumulated on stormwater detention systems were significantly different from site to site or among different source areas. The reasons for these controversies are differences in sampling method and transport of particle (Sansalone and Buchberger 1997), analytical method (sieving analysis or particle counter method), climatic influences, atmospheric deposition, vehicular traffic density and road maintenance (Helmreich et al. 2010).

Concentrations of metals in sediment deposited within stormwater treatment systems can be highly variable. Several recent studies assessed the concentrations of heavy metals, particularly Cd, Cr, Cu, Ni, Pb, and Zn captured within stormwater treatment systems. Despite the large variations in concentration, most researchers agree that the majority of metals tend to accumulate in the top layer of stormwater treatment systems and concentrations decreased with depth of the filter bed (Paus 2999; Ingvertsen et al. 2012; Li and Davis 2008; Jensen et al. 2006). The concentrations measured in the present study compared to literature values are shown in Table 5. The concentrations of heavy metals, particularly Cu and Zn, in the SDPL were significantly higher than the levels found in FMC sample collected from bioretention and infiltration swales (Table 5).

Concerning the GSA-H, the concentrations of heavy metals in SDPL after 7 years of operation (sampling in 2012) were over two folds higher than the levels determined after 2 years of operation (sampling in 2008) as reported by Fuerhacker et al. (2011) (see Table 5), evidencing surface accumulation of heavy metals in the SDPL over the operation time. The mean concentrations of Cr, Cu, Pb and Zn found in the present study were well above the levels reported by several authors (Paus 2999; Ingvertsen et al. 2012; Li and Davis 2008). For example, the mean Cu and Zn concentration in the SDPL observed in this study were 3 to 60 times higher than those reported for the top layer (0–5 cm) infiltration systems aged 2 to 16 years (Table 5). Pb concentrations in the SDPL were within the range of the levels reported by Ingvertsen et al. (2012) and Jensen et al. (2006) but far below the concentration reported by Li and Davis (2008). It is apparent that the pollutant loads entering a stormwater treatment system could significantly vary from site to site due to differences in climatic influences, vehicular traffic density, atmospheric deposition, road maintenance and characteristics of the catchment area. The higher metal concentrations observed in this study could mainly be due to the small filtration system size compared to the impervious catchment area, so that input load is high compared to the input loads in bioretentions and road-side infiltration swales. For example, the roadside infiltration swales investigated by Ingvertsen et al. (2012) were sized at 7 to 28 % of their impervious catchment area which is by far larger than the sizing of the filtration systems (i.e. 0.25 to 0.37 %) examined in this study. Interestingly, the concentrations of heavy metals in the SDPL were comparable to the levels found in roadside-deposited sediments as reported by Gunawardana et al. (2014). The total concentrations of metals in sediments deposited on urban road surfaces were 11,710 mg/kg Al, 20,310 mg/kg Fe, 30 mg/kg Cr, 400 mg/kg Cu, 420 mg/kg Mn, 30 mg/kg Ni, 170 mg/kg Pb and 910 mg/kg Zn (Gunawardana et al. 2014).

Previous studies by several researchers focused mainly on heavy metals which are toxic (i.e. Cd, Cu, Cr, Ni, Pb and Zn) (Paus 2999; Ingvertsen et al. 2012; Li and Davis 2008). This study reports additional information regarding the presence of a wide range of heavy

Table 5 Concentrations of heavy metals (mg/kg dry matter) in sediment and filter media layer compared to literature values and sediment quality

Cd	Cr	Cu	Ni	Pb	Zn	Source
0.6	81	490	46	57	1449	SDPL (GSA-H)
0.03–0.34	35–131	30–83	19–126	5–19	40–206	FMC (GSA-H)
0.49	86	326	52	114	3016	SDPL (GSA-W)
0.03–0.1	24–49	20–24	31–69	6.1–10.1	152–168	FMC (GSA-W)
0.1	37	230		47	796	Fuerhacker et al. 2011[a]
		75		399–660	114–532	Li and Davis 2008[b]
0.13–0.71	11–67	12–57		20–108	44–259	Ingvertsen et al. 2012[c]
0.35–0.70	13–136	114–598		49–144	190–273	Jensen et al. 2006[d]
<0.88–1.87		9.7–37.8			44–89	Paus et al.[e]

SDPL surface deposited sediment 2.5 to 4.5 cm, *FMC* filter media core 36 cm

[a] Surface deposited particle layer from GSA-H after in operation for 2 years

[b] Bioretention media in operation for 5 years receiving parking lot runoff (USA)

[c] Roadside infiltration swales in operation for 6 to 16 years, (highway, Germany)

[d] Surface deposits in roadside swales in operation for up to 16 years (highways, Denmark)

[e] Bioretention media in operation for 2 to 8 years (metropolitan area, USA)

metals such as Ba, Mn, Sr, Ti, and V with high concentrations found in the sediments deposited at the surface of stormwater filtration systems. For example, the metals Ba, Ti, V are included in the Annex 2 of the Austrian groundwater guideline but not regulated with target values. Thus, there is a demand to further investigate the behaviour and mode of action of those heavy metals.

The concentration of heavy metals as a function of the particle size of sediments (roadway runoff and road-deposited) has been investigated by several researchers and invariably the highest concentrations were consistently found in the fine particle size fractions (Herngren et al. 2006; Zandres 2005; Zhao et al. 2010). Results from this study did not support the assertion that the heavy metals were rather uniformly distributed in the different SDPL size fractions (Table 3). In road-deposited sediment samples, Zandres (2005) observed that a high percentage of the total metal load was associated with particles smaller than 125 μm (64 % of Zn, 57 % of Cu and 46 % of Pb). Similarly, Zhao et al. (2010) reported that 80 % of the heavy metals (Cd, Cr, Cu, Ni, Pb and Zn) in road-deposited sediments were mainly associated with the <250 μm size fraction. The heavy metal distribution of road-deposited sediments indicated that particle retention (removal), specifically removal of the size fraction <250 μm plays a vital role in removing pollutants from roadway runoff. However, results of this study showed that despite the uniform concentration distribution over 60 % of the trace metals loads were bound to the fine fraction (<63 μm) of SDPL.

5 Conclusion

In this study, two stormwater (parking lot and highway runoff) filtration systems were investigated after 5 to 7 years in operation and the following conclusions are drawn:

- In situ infiltration test results showed that saturated hydraulic conductivity of the filtration systems decreased over the course of operational time but still acceptable in accordance with the Austrian standard design guidelines. This is mainly attributed to the deposition of suspended solids, straining of particles in the pores and compaction due to hydraulic loading.
- The hydraulic performance of the system could be recovered through removal of the accumulated particle layer and replacement or back flushing of the geotextile on periodic bases, approximately every 7 years.
- Particle size distribution of the solids deposited at the surface of the filtration system (i.e. SDPL) showed that 70 % of the masses comprised of particles smaller than 63 μm, and the particles smaller than 200 μm accounted for 85 % of the total mass. Metal concentration distributions along the different particle size fractions were uniform; however, the distributions of metal masses across particle size fractions followed patterns of particle mass distribution. For instance, 60 % of the trace metals were bound to the particles smaller than <63 μm.
- Concentration profiles of heavy metals, particularly those relevant in highway runoff (Cu, Pb and Zn), showed a high accumulation in the SDPL significantly decreasing with the depth of filter media. This implies that large fractions of the input metal concentrations were particulate-bound and during stormwater treatment, removal of the fine particles plays an important role. Furthermore, other heavy metals (such as Ba, Ti, V, Sr) with concentrations comparable to the commonly investigated heavy metals (e.g. Pb, cu and Zn) were identified in the SDPL and core filter media which indicates comparable concentrations in runoff. There is still a demand to assess the sources, concentration ranges and potentials for toxic problems.
- Column test with FMC suggested that the filter media in operation is highly efficient for simultaneous adsorption of Cr, Cu, Ni, Pb and Zn from synthetic stormwater runoff. Based on the heavy metal loads applied in column test and computed annual input loads, the filter media can remain in operation for over 34 years. In addition, it is likely that the build of a particle layer on top of the filter media have a tendency for the sorption of dissolved heavy metals and a longer lifespan can be expected. Therefore, lifespans will be dependent mainly on the long-term hydraulic performance of the systems.

Acknowledgments This study was conducted under the projects “ÖNORM and SARIT.” The Project was supported by the

Austrian Federal Ministry of Agriculture, Forestry, Environment and Water Management and the Company SW Umwelttechnik AG. They are gratefully acknowledged for providing funds.

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6.3 Paper III

Test methods for the utilization of filter media– case study on performance of stormwater filtration system treating parking lot and highway runoff. "Prüfmethode zur Filterwirkung und –eignung, in German".

Haile, T.M. & Fürhacker, M. (2015). ÖWAV Seminar "Versickerung von Niederschlagswässern: ÖWAV-Regelblatt 45 Rahmenbedingungen, Bemessung und Betrieb von Versickerungsanlagen", 06 Oct. 2015, Vienna, Austria. ISBN: 978-3-902978-62-2.

Prüfmethode zur Filterwirkung und -eignung

Tadele Measho Haile, Maria Fürhacker

Einleitung

In Hinblick auf ein nachhaltiges Regenwassermanagement muss die Verunreinigung und der Behandlungsbedarf der Niederschlagsabflüsse die Reinhaltung des Grundwassers bzw. Oberflächenwassers ermittelt werden. Der Niederschlagsabfluss von Straßen und Dächern enthält Schwermetalle, abfiltrierbare Stoffe, organische Verbindungen, Spurenstoffe und Nährstoffe (Kasting, 2002; Boller, 2003; Dierkes et al., 2005; Göbel et al., 2007; Helmreich, 2005, Fuerhacker et al., 2013) die entweder gelöst oder partikulär gebunden vorliegen. Diese Niederschlagsbestandteile stammen bzw. entstehen aus Abgasnebenprodukten, Reifen-, Karosserie- und Fahrbahnverschleiß, Abflüssen aus Niederschlägen, nasser und trockener Deposition und Fahrbahninstandhaltungsarbeiten. Die relativen Beiträge der unterschiedlichen, von Fahrbahnen abgeleiteten Verunreinigungen (z.B. das Verhältnis von Kupfer zu Stickstoff zu abfiltrierbaren Stoffen) hängen stark von der Landnutzung rund um die Verkehrsflächen (Choe et al., 2002) sowie vom Regenereignis und der entsprechenden Probenahme ab.

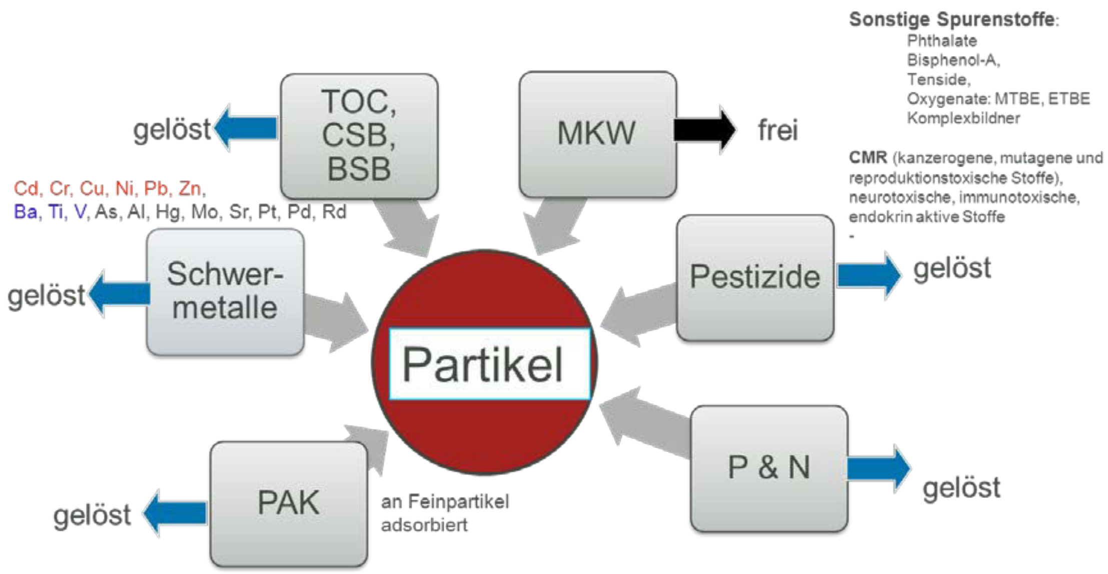

Abb. 1 Mögliche Verunreinigungen von Niederschlagsabflüssen

Sollen belastete Verkehrsflächen- oder Dachabflüsse direkt vor Ort versickert werden, so müssen sie in Abhängigkeit von der Quelle bzw. von der Verschmutzung über ein entsprechendes Behandlungssystem z.B. Rasen oder Filtersystem mit bewachsenem

Oberboden bzw. mit technischem Filter abgeleitet werden (ÖWAV, 2015). Vom bewachsenen Oberboden wird von vornherein eine optimale Reinigung erwartet, allerdings müssen technische Filtermaterialien vor ihrem Einbau geprüft werden und bestimmte Qualitätskriterien erfüllen.

Im Zuge des Projektes „Entwicklung von Methoden zur Prüfung der Eignung von Substraten für die Oberflächenwasserbehandlung von Dach- und Verkehrsflächen“, das vom BMLFUW finanziert wurde, wurden von der Universität für Bodenkultur, Institut für Siedlungswasserbau, Industriewasserwirtschaft und Gewässerschutz gemeinsam mit einer Ad-hoc Gruppe Kriterien und Vorschriften für eine Prüfung von technischen Filtermaterialien erarbeitet. Diese Methode wird als ÖNORM B 2506 Teil 3 Entwurf veröffentlicht werden.

Prüfmethode

Mit der ÖNORM B 2506 Teil 3, die im Entwurf vorliegt, wurde eine Prüfmethode erarbeitet ,um die Leistungsfähigkeit verschiedener Filtersubstrate anhand des Partikelrückhalts, der Schwermetalladsorption und des Mineralölrückhalts für eine Standzeit von mindestens 4 Jahren festzustellen, da nach dieser Zeit eine bewilligte Anlage jedenfalls zu prüfen ist. Diese Leistungsfähigkeit wird anhand von Prüfkriterien, die zum Teil in der ÖNORM B 2506 Teil 2 festgelegt bzw. in Fürhacker et al. (2013) vorgeschlagen wurden, gemessen. Es wurden Prüfungen für verschiedene Herkunftsklassen wie befestigte Flächen, Zink- und Kupferdachabflüsse für verschiedene Flächenverhältnisse (Entwässerungsfläche $A_{red} : A_s$ wirksame Filterfläche zwischen 1 : 15 bis 1 : 250) definiert. Die Prüfung besteht aus 8 Teilprüfungen (Tabelle 1):

Tabelle 1 Überblick über die Teilprüfungen (ÖNORM B 2506 Teil 3 Entwurf)

Nr	Teilprüfung
1	Infiltrationsrate
2	Partikelretention I
3	Schwermetallrückhalt / Kapazitätsprüfung
4	Mineralölrückhalt
5	Partikelretention II
6	Änderung Infiltration
7	Remobilisierung durch NaCl
8	Säureneutralisationskapazität

Die Untersuchungen erfolgen in einer Filtersäule mit einem Innendurchmesser von 100 mm. Für Flächenverhältnisse > 1 : 15 wird die Kapazität für den Schwermetallrückhalt in einer kleineren Säule geprüft. Neben der Prüfung auf Partikelrückhalt, bzw. auf Verstopfung des Materials durch Partikel wird auch der Rückhalt von Schwermetallen und Mineralöl geprüft. Die Prüfkriterien sind in Tabelle 2 angegeben.

Tabelle 2 Anforderungen an Filtermaterialien nach der ÖNORM B 2506 Teil 3 Entwurf

Eigenschaft	Anforderung
Infiltrationsrate	> $1 * 10^{-5}$ m/s
Partikelretention	Rückhalt > 80 %
Schwermetallrückhalt	> 80 % Cu-Entfernung, > 50 % Zn-Entfernung, Pb < 9 µg/l
Mineralölrückhalt	Entfernungsrate > 95 %
Änderung der Infiltrationsrate	> 50 % bzw. 30 % des Ausgangswertes, je nach Flächenverhältnis
Remobilisierungsprüfung für Partikel	< 1,6 g AFS
Schwermetallremobilisierung durch NaCl	Konzentration von Cu < 50 µg/l Konzentration von Zn < 500 µg/l
Säureneutralisationskapazität	pH-Wert > 6,0, während des Durchlaufs des Wasservolumens von 42 l oder während einer Prüfdauer von 30 min

Da diese Filtermaterialien auch Winterwässern mit einem erhöhten Salzgehalt ausgesetzt sind, wird auch die potentielle Remobilisierung der Schwermetalle durch Natriumchlorid erfasst.
Zusätzlich zur Prüfung von frischem Filtermaterial ist auch die Prüfung für die weitere Einsatzfähigkeit eines in Benützung stehenden Filtermaterials in der ÖNORM B 2506 Teil 3 Entwurf definiert.

Prüfung auf Praxisrelevanz

Im Zuge eines Projektes „Entwicklung von Methoden zur Prüfung der Eignung von Substraten für die Oberflächenwasserbehandlung von Dach- und Verkehrsflächen“, wurden fünf Materialien, drei davon aus in Betrieb befindlichen Beckenfilteranlagen (zwei Bodensubstrate BA und BB und ein technisches Filtermaterial TF1) in die Teilprüfungen im Säulenversuch miteinbezogen. Zusätzlich wurde die Praxistauglichkeit der Teilprüfungen auch noch an zwei weiteren Filtermaterialien Quarzsand und dem technischen Filtermaterial TF2 erprobt, das allerdings nicht in einem System in Beckenform eingebaut war. Es zeigte sich, dass jenes Bodenfiltersubstrat (BB), das im Säulenversuch nach kurzer Zeit Kolmationserscheinungen zeigte, auch in der Betriebsanlage Durchlässigkeitsprobleme aufwies. Das technische Filtermaterial ist bereits in mehreren Anlagen eingebaut, wobei die erste bereits 2005 mit einem Flächenverhältnis von $A_{red} : A_s$ =1 : 400 an einem Parkplatz errichtet wurde. Diese Anlage wurde zu Beginn eineinhalb Jahre vom Institut SIG der Universität für Bodenkultur wissenschaftlich begleitet und 2012 und 2015 wieder untersucht. Die Ergebnisse zeigen, dass die Anlage die in der ÖNORM B 2506 Teil 3 Entwurf geforderten Entfernungsraten eingehalten wurden und sich die Ablaufkonzentrationen sowohl hinsichtlich der AFS, Schwermetalle, als auch der polyzyklischen Kohlenwasserstoffe (PAK) über die Zeit verminderten.

Eine zusätzliche Überprüfung der Restkapazität des nach einer Betriebszeit von 7 Jahren entnommenen Filtermaterials zeigte noch eine Reststandzeit für die vor Ort zu erwartende Belastung von 34 Jahren, wobei Cu als limitierendes Element zu sehen ist.

Vergleichbarkeit mit Bodenfiltern

Eine weitere Anlage, die einen direkten Vergleich zwischen Bodenfilter und technischem Filter erlaubt ist eine „Anlage in der Anlage“. D.h. in einem Teil einer Bodenfilteranlage (Flächenverhältnis von $A_{red} : A_s$ =1 : 28,3) wurde eine Filteranlage mit technischem Substrat mit einem Flächenverhältnis von $A_{red} : A_s$ =1 : 283 errichtet. Beide Anlagen werden mit je 50 % der Zulaufwassermenge beschickt. Die Probenahme erfolgt mit automatischen Probenahmesystemen, wobei sich die Probenahmeflaschen je nach Füllstand im Rohr bei geringen, mittleren oder hohen Füllgraden füllen. Da das gleiche System in beiden Ablaufrohren angewendet wird, kann von vergleichbaren Proben ausgegangen werden.

Es wurden bisher 9 Ereignisse beprobt, wobei die Untersuchungen jedenfalls über den Winter weitergeführt werden sollen. Die Ergebnisse der Messungen sind in Tabelle 3 dargestellt.

Tabelle 3 Ergebnisse der vergleichenden Untersuchungen an einem System mit Bodenfilter und mit technischem Filtermaterial

Parameter	Einheit	Zulauf	Bodenfilter RVS	Technisches Filtermaterial	QZV Chemie GW Schwellenwert
pH Wert	-	7,1 – 9,3	6,8 – 8,1	6,8 – 8,1	
Leitfähigkeit	µS/cm	680 - 100600	194 - 6100	356 – 7150	2250 (20°C)
Chlorid	mg/l	670 - 42000	15 - 590	<4,0 - 2000	
AFS	mg/l	110 - 19000	12 - 180	4 - 130	
TOC[1)]	mg/l	113 - 3120	23,4 – 33,2	15,4 – 34,2	
DOC[1)]	mg/l	44 - 2868	7,6 – 22,1	11,1 – 26,4	
TNb	mg/l	9,7 – 22,5	26,4 – 22,2	2,5 – 42,6	
KW-Index	mg/l	7,9 - 1100	0,2 – 2,3	<0,1 – 0,6	0,1
Al gelöst	µg/l	34,6 - 234	<5,0 - 827	<5,0 – 29,2	
Pb gelöst	µg/l	<0,5 – 0,63	<0,5 – 2,7	<0,5 – 3,5	9
Cd gelöst	µg/l	<0,05 – 4,3	<0,05 – 0,07	<0,05 – 0,11	4,5
Cr gelöst	µg/l	<0,5 – 37,3	<0,5 – 20	<0,5 - 22,8	45
Fe gelöst	µg/l	11,9 – 3390	<5,0 - 816	5,3 – 729	
Cu gelöst	µg/l	0,8 – 235	1,6 – 53,8	1,6 – 37,1	1800
Ni gelöst	µg/l	3,3 – 30,1	<0,5 – 10,2	0,8 – 10,3	18
Zn gelöst	µg/l	<3,0 - 264	<3,0 – 24,5	<3,0 - 62,1	

[1)]Es wurden nur jene Werte angegeben, von denen sowohl DOC als auch TOC-Ergebnisse vorlagen
[2)]KW-Index-Werte beim Ölunfall (Bodenfilter 1,5 mg/l und TF Ablauf 2,6) wurden nicht berücksichtigt

Die Messungen zeigen, dass die KW-Index Konzentrationen (Werte während des Ölunfalls ausgenommen) im Ablauf des Bodenfilters im Mittel bei 0,5 mg/l und im Ablauf des Systems mit technischem Filter im Mittel bei 0,2 mg/l liegen. Bei beiden Filtersystemen konnten aber bei einem Ölunfall erhöhte Konzentrationen von 1,5 mg/l (RVS) bzw. bis zu 2,6 mg/l KW-Index (TF) gemessen werden, wobei die Entfernungsraten bei > 99,8 % lagen. Die Messwerte für KW-Index und Leitfähigkeit lagen fallweise höher als die Schwellenwerte der QZV-Chemie GW, deren Werte zwar zum Vergleich herangezogen wurden, aber Immissionswerte für ganze Grundwasserkörper sind. Die teilweise hohen gesamt gebundenen Stickstoffergebnisse (TNb) sind eher auf Ammonium zurückzuführen, wie einzelne Messungen ergaben, die Nitratwerte lagen größtenteils unter der Bestimmungsgrenze von < 1 mg/l Nitrat-N.

Schlussfolgerungen

Zusammenfassend kann gesagt werden, dass die im Entwurf der ÖNORM B 2506 Teil 3 vorgeschlagene Prüfung der Filtersubstrate gute Vorhersagen für die Einsatzfähigkeit der Substrate in der Praxis erlauben. Aus jetziger Sicht, ergibt die Kapazitätsprüfung im Test vermutlich zu kurze Standzeiten an, verglichen mit Anlagen im praktischen Betrieb, weil ein erheblicher Teil der Schwermetalle partikulär, zum Großteil gebunden an Feinstpartikel < 63 µm, vorliegen und sich die Reinigungsleistung mit der Standzeit und zunehmenden Verminderung der Durchlässigkeit erhöht. Ein direkter Vergleich eines Systems mit Boden bzw. mit technischem Filtermaterial unter Praxisbedingungen zeigte, dass die Ablaufkonzentrationen des technischen Filters auch bei einem 10-fach höherem Flächenverhältnis im Bereich des Bodenfilters liegen. In Hinblick auf eine mögliche Kontamination mit Kohlenwasserstoffen und AFS wird empfohlen einen wirksamen Öl – und auch Partikelabscheider den Filterbecken vorzuschalten.

Danksagung
Die dieser Veröffentlichung zugrundeliegenden Arbeiten wurden unter anderem im Rahmen des Projektes „Entwicklung von Methoden zur Prüfung der Eignung von Substraten für die Oberflächenwasserbehandlung von Dach- und Verkehrsflächen“, (finanziert vom BMLFUW) und im Rahmen des FFG Projekts „SARIT“, das von der Fa. SW-Umwelttechnik eingereicht wurde, erarbeitet.

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Anhang der Fallstudie Lassnitzhöhe

In der beprobten Gewässerschutzanlage können die Abläufe eines Bodenfilters (RVS) direkt mit denen eines technischen Filters (TF) verglichen werden, weil der Zulauf nach dem Sedimentationstank geteilt wird und zur Hälfte über den Bodenfilter (Flächenverhältnis 1:28,3) und über den technischen Filter (Flächenverhältnis 1:283) geleitet wird. Die Ergebnisse sind in der Tabelle 1 und Tabelle 2 aufgelistet.

Die Schwellenwerte der QZV Chemie Grundwasser beziehen sich auf durchschnittliche Werte im Grundwasserkörper. Trotzdem werden sie in Tabelle 2 zum Vergleich angeführt.

Schwermetalle:

Am 12.07.2014 wurde händisch und an allen nachfolgenden Probenahmen durch eine automatische Probenahmeeinrichtung Zulaufproben gezogen. Die händisch gezogene Probe wurde bei Trockenwetter aus dem Absetzbecken gezogen und kann daher nur indirekt mit den Ablaufwerten verglichen werden. Die Gesamtprobe bezieht sich auf eine Durchschnittsprobe inklusive Sediment, während die flüssige Phase eine abgesetzte Probe wiederspiegelt. Durch den hohen pH-Wert und die Absetzzeit können sich die Gleichgewichte in Richtung Festphase verschoben haben, wodurch die niedrigen Schwermetallkonzentrationen in der gelösten Phase des Zulaufs zu erklären wären. Auffällig sind die besonders niedrigen Schwermetallkonzentrationen im Zulauf (Probenahme 12.07.2014). Diese können auf den relativ hohen pH-Wert in der Probe zurückgeführt werden, bzw. könnte es sich auch um eine relativ unbelastete Niederschlagsfraktion am Ende eines Regenereignisses handeln.

Die Cadmium-, Chrom-, Kupfer-, Nickel- und Bleikonzentrationen sind in den gelösten Fraktionen des Ablaufs entweder kleiner der Bestimmungsgrenze oder deutlich unter dem Schwellenwert der QZV Chemie Grundwasser. In den Abläufen fallen die Gesamtgehalte der Aluminium- und Eisenkonzentrationen der ersten Probenahme vom 10.03.2014 mit 1200 mg Al/l und 675 mg Fe/l für RVS und mit 313 mg Al/l und 214 mg Fe/l für TF im Vergleich zu den restlichen gemessenen Schwermetallen eher hoch aus und weisen in den gelösten Fraktionen deutlich niedrigere Konzentrationen (827 mg Al/l und <5,0 mg Fe/l für RVS und mit <5,0 mg Al/l und 5,3 mg Fe/l für TF) auf. Die gelösten Eisenkonzentrationen waren bei der Probeentnahmestelle „Mitte“ fast immer höher als bei der Probeentnahmestelle „„unten““ (Ausnahme RVS Probenahme 12.07.2014). Auffällig sind die starken Schwankungen der Eisenkonzentrationen, die zwischen den Probenahmen höher sind als zwischen den Abläufen der verschiedenen Filtermaterialien. Dies könnte auf reduzierte Verhältnisse im Zulauf zurückzuführen sein. Die gelösten Zinkkonzentrationen sind im Ablauf der Filter RVS und TF bei der Probenahmestelle „unten“ höher als bei der Probenahmestelle „Mitte“ vom 16.06.2014, 02.07.2014, 12.07.2014 und vom 18.12.2014. Vor allem die beiden Proben RVS „unten“ und TF „unten“ vom 18.12.2014 weisen Zinkkonzentrationen >10000 mg/l auf und RVS „Mitte“ und Tf „Mitte“ besitzen Zinkkonzentrationen kleiner der Bestimmungsgrenze < 3,0 µg/l. Dies ist durch die Verwendung von verzinkten Verschraubungen und einer damit verbundenen Kontamination bedingt. Diese Werte werden zwar dargestellt, aber nicht in die Betrachtung miteinbezogen. Nach Entnahme der Probenflaschen vom 18.12.2014 wurden die verzinkten

Schraubverbindungen ersetzt werden. Alle später durchgeführten Probeentnahmen, wiesen deutlich geringere Zinkkonzentrationen auf.

Die Ergebnisse zeigen, dass auch bei hohen Salzkonzentrationen im Zulauf bzw. bei abgeminderten Salzkonzentrationen im Ablauf die Schwermetallkonzentrationen in den Abläufen des Bodenfilters und des technischen Filters vergleichbar sind und die Konzentrationen nicht wesentlich über jenen von Proben mit geringer Leitfähigkeit liegen. Daraus lässt sich schließen, dass es zwischen den Sommer- und Winterabflüssen in Bezug auf die gelösten Schwermetalle keinen Unterschied gibt.

Kohlenwasserstoffe:

Der Schwellenwert der QZV Chemie Grundwasser mit 0,1 mg/l KW-Index wird von den meisten Proben überschritten, außer TF Ablauf „unten“ am 17.03.2015 sowie TF Ablauf „Mitte“ am 16.06.2014, am 18.12.2014 und am 17.03.2015. Die Werte im Ablauf der Bodenfilteranlage liegen zwischen 0,1 und 2,3 mg/l KW-Index und die Werte im Ablauf des technischen Filtermaterials zwischen <0,1 und 2,6 mg/l KW-Index. Bei einer sehr hohen Kontamination im Zulauf (ca. 1000 mg/l Probenahme 22.4.2015) konnte zwar mehr als 99 % des KW-Index von beiden Filtern entfernt werden, wie sich zeigt ist es aber möglich, dass bei verschiedenen Zuständen messbare KW-Indexkonzentrationen im Ablauf beider Filter auftreten.

Es sollte auch bedacht werden, dass der KW-Index eine Bestimmung mittels Gaschromatograph und Detektion mittels Flammenionisationsdetektor ist und dass die Integration zwischen C_{10} und C_{40} erfolgt. Es ist bekannt, dass mit diesem Parameter nicht nur Mineralölkohlenwasserstoffe erfasst werden, weil auch die Aufreinigung anders als beim klassischen Parameter Summe-KW erfolgt. Bei der Bestimmung des Parameters Summe-KW, der mittels IR-Photometer bestimmt wird, ist sicherzustellen, dass keine C-O oder C-N Banden vorhanden sind, d.h. dass sich dieser Parameter, im Gegensatz zum KW-Index, nur auf Kohlenwasserstoffe bezieht. Es sollte in weiterer Folge ermittelt werden, ob es sich bei diesen Fraktionen, die bei geringen Abläufen bzw. mit dem ersten Schwall durchgespült werden, um Kohlenwasserstoffe handelt.

Chlorid und elektrische Leitfähigkeit:

Am 22.01.2015 und am 17.03.2015 wurden im Zulauf sehr hohe elektrische Leitfähigkeiten von bis zu 100000 µS/cm und Chloridkonzentrationen von bis zu 42000 mg/l gemessen. Am 22.01.2015 wurden die Zulaufwässer, die zu Beginn der Ereignisse gezogen wurden, durch weitere Regenfälle bzw. durch Wasser in der Filteranlage verdünnt, sodass im Ablauf 6100 bzw. 6400 µS/cm gemessen wurden. Das Regenereignis füllte jedenfalls nur die Probenflaschen an der Probeentnahmestelle „unten“. Am 17.03.2015 konnte in den beiden Zulaufproben (links und rechts) sehr unterschiedliche Konzentrationen gemessen werden. Die Leitfähigkeit war in beiden Flaschen sehr hoch. Am 22.01.2015 wiesen die Proben RVS Ablauf „unten“ 590 mg/l Chlorid (Lf 6100 µS/cm) und TF Ablauf „unten“ 2000 mg/l Chlorid (Lf 6400 µS/cm) auf und liegen höher als der Schwellenwert der QZV Chemie GW mit 150 mg/l Chlorid (Schwellenwert elektrische Leitfähigkeit 2250°µS/cm). Proben vom 17.03.2015 wiesen für RVS Ablauf „unten“ bzw. „Mitte“ 1700 (Lf 5580 µS/cm) bzw. 1600 mg/l Chlorid (Lf

5270 µS/cm) und für TF Ablauf "unten" bzw. „Mitte" 2200 (Lf 7150 µS/cm) bzw. 1300 mg/l Chlorid (Lf 4460 µS/cm) auf. Die beiden folgenden Probeentnahmen vom 22.04.2015 und 28.05.2015 wiesen in den Zuläufen deutlich niedrigere Chloridwerte (bis zu 980 mg/l) und elektrische Leitfähigkeiten (bis zu 3800 µS/cm) auf als jene beiden zuvor gemessenen. Ebenso sieht es bei den Ablaufwerten aus. Am 22.04.2015 wird der Chlorid-Schwellenwert von 150 mg/l von den Proben RVS Ablauf „unten mit 200 mg/l, TF Ablauf „unten mit 690 mg/l und TF Ablauf „Mitte" mit 590 mg/l und der Schwellenwert elektrische Leitfähigkeit mit 2250 µS/cm von den beiden Proben TF Ablauf „unten" mit 3200 µS/cm und TF Ablauf „Mitte" mit 2900 µS/cm überschritten. Am 28.05.2015 halten die Ablaufproben des Bodenfilters (Proben RVS Ablauf „unten" und RVS Ablauf „Mitte") die Schwellenwerte der elektrischen Leitfähigkeit und Chlorid ein. Der technische Filter (Proben TF Ablauf „unten" und TF Ablauf „Mitte") unterschreitet den Schwellenwert der elektrischen Leitfähigkeit, überschreitet mit 170 mg/l und 160 mg/l jedoch den Schwellenwert Chlorid mit 150 mg/l etwas.

Auch im darauf folgenden Probenahmejahr 2016 wiesen die Proben TF Ablauf und RVS Ablauf im Jänner 8920 µS/cm und 7880 µS/cm und im Februar 8940 µS/cm und 6550 µS/cm auf und liegen wiederum deutlich über dem Grenzwert der QZV Chemie GW von 2250 µS/cm. Die Proben TF Ablauf und RVS Ablauf vom 24.03.2016 weisen im Gegensatz zu den beiden vorangegangenen Monaten geringere Leitfähigkeiten auf.

Die hohen Chloridwerte der Probenahme Jänner und März 2015 und Jänner und Februar 2016 lassen auf Winterbetrieb schließen und die noch etwas erhöhten Werte vom 22.04.2015, 28.05.2015 und 24.03.2016 können Restkonzentrationen in den Anlagen sein, die durch den Winterbetrieb in den Anlagen verblieben sind und durch die beiden Regenereignisse vom April und Mai langsam aus dem Filtermaterial ausgewaschen werden.

Ammonium-N:

Da seit der Probenahme vom 17.03.2015 erhöhte Gesamtstickstoffgehalte gemessen wurden, wurden von den Probenahmen vom 22.04.2015, 28.05.2015, 16.02.2016 und 24.03.2016 zusätzlich Ammoniumstickstoff gemessen, um zu eruieren, von wo der Gesamtstickstoffgehalt herrührt. Es zeigt sich, dass die größte Fraktion des Gesamtstickstoffs löslicher Ammoniumstickstoff ist. Der Schwellenwert der QZV Chemie GW mit 0,45 mg/l wird von beiden Anlagen mit bis zu 42 mg/l (TF Ablauf „Mitte") deutlich überschritten.

Phosphor (gesamt) und Phosphat-P:

Zusätzlich zu Ammoniumstickstoff wurde von den letzten 3 Probenahmen auch Gesamtphosphor und Phosphat-P gemessen. Der Schwellenwert der QZV Chemie GW liegt bei 0,30 mg/l Orthophosphat-P und wird von beiden Filteranlagen mit bis zu 2,65 mg/l Phosphat-P überschritten, wobei ein großer Teil des Gesamtphosphors als Phosphatphosphor vorliegt.

DOC:

Auch im Hinblick auf die DOC-Messungen ist festzustellen, dass sich die Ablaufkonzentrationen der Filter zwar unterscheiden und der Bodenfilter tendeziell geringere

Konzentrationen aufweist, dass aber im Fall von höheren DOC-Konzentrationen (um 100 mg/l) beide Filtermaterialien ähnlich reagieren.

<u>AFS Reinigungsleistung (fein Partikel):</u>

Aufgrund der zusätzlich gemessenen Ablaufproben kann durch Vergleich mit den Ablaufwerten für beide Anlagen (TF und RVS) auf eine gute Reinigungsleistung geschlossen werden.

In den Abläufen werden die Schwellenwerte der QZV Chemie GW für sämtliche Parameter von beiden Filtern eingehalten, außer für den Parameter KW-Index, Chlorid und infolge die elektrische Leitfähigkeit, Ammonium-N und Phosphat-P. Der direkte Vergleich der Abläufe zwischen Bodenfilter mit dem Flächenverhältnis 1:28,3 und techischem Filtermaterial mit einem Flächenverhältnis von 1:283 zeigte, dass die Schwellenwerte der QZV Chemie GW von beiden Filtern eingehalten werden, außer für den Parameter KW-Index, Chlorid, Ammonium und Phosphat. Die Werte im Ablauf der Bodenfilteranlage liegen zwischen 0,3 und 2,3 mg/l KW-Index und die Werte im Ablauf des technischen Filtermaterials liegen zwischen < 0,1 und 0,6 mg/l KW-Index bei einem Schwellenwert von 0,1 mg/l KW-Index. Bei der Interpretation der KW-Indexkonzentrationen sollte bedacht werden, dass der KW-Index anders als der klassische Parameter Summe-KW nicht nur Kohlenwasserstoffe erfasst. Es sollte in weiterer Folge ermittelt werden, ob es sich bei diesen Fraktionen, die bei geringen Ablaufvolumina bzw. mit dem ersten Schwall durchgespült werden, tatsächlich um Kohlenwasserstoffe handelt.

Die AFS und DOC Konzentrationen waren bei den Filterabläufen der beiden Filter in vergleichbaren Konzentrationsbereichen.

Tabelle 1: Statistik der Zulaufkonzentrationen im Parkplatzabflüsse (GSA-L) der kombinierten Anlage mit Bodenfilter (RVS) und technischem Filtermaterial (TF)

Parameter	Einheit	Min	Median	Mean	Max	QZV-GW2)
pH		7,1	8,9	8,94	11	
EC	-	680	7840	34006	100600	2250 (20 °C)
Cl	µS/cm	370	6800	15578	42000	
DOC	mg/L	44	112,5	561	2868	
TOC	mg/L	113	493	756	3120	
MOH	mg/L	0,1	58,95	364	1100	0,1
TNb	mg/L	9,7	22,5	39,4	99	
TSS	mg/L	110	818	4411	19000	
Metalle						
Al	mg/L	6,5	71,8	200	639	
Pb	µg/L	0,5	0,5	0,51	0,63	9
Cd	µg/L	0,05	0,395	1,18	4,3	4,5
Cr	µg/L	0,8	8,2	10,5	37,3	45
Cu	µg/L	0,8	77,8	100,7	258	1800
Fe	µg/L	11,9	96,9	792	3390	
Ni	µg/L	3,3	14,2	14,6	30,1	18
Zn	µg/L	13,9	43,8	76,1	264	
Nährstoffe						
NO_3^-	µg/L	4	1	5,0	5	
NH_4^+	mg/L	4,2	13	16,6	40	
H_3PO_4	mg/L	2,19	7,9	7,4	16	
H_2SO_4	mg/L	5,4	44,5	37,9	70	

Tabelle 2: Ergebnisse der Untersuchungen der Ablaufproben der kombinierten Anlage mit Bodenfilter (RVS) und technischem Filtermaterial (TF) bei der Probeentnahmestelle „unten".

Probenahme Datum		**10.03.2014**		**16.06.2014**				**02.07.2014**				**12.07.2014**				**QZV-GW[2]**
Probeentnahmestelle		**„unten"**		**„unten"**		**„Mitte"**		**„unten"**		**„Mitte"**		**„unten"**		**„Mitte"**		
Parameter	**Einheit**	**RVS**	**TF**	**RVS**	**TF**	**RFS**	**TF**	**RVS**	**TF**	**RFS**	**TF**	**RVS**	**TF**	**RFS**	**TF**	
pH		-	-	-	-	-	-	-	-	-	-	7,4	6,8	7,2	7	
EC	-	-	-	-	-	-	-	-	-	-	-	255	692	226	356	2250 (20 °C)
Cl	µS/cm	-	-	-	-			-	-			-	-			
DOC	mg/L	-	-	91	106			43	76			16	80			
TOC	mg/L	-	-	-	-	104	135	43	76	12	62	16	80	13	55	
MOH	mg/L	0,2	0,2	0,3	0,2	0,2	<0,1	0,2				2,3	0,1	0,2	0,2	0,1
TNb	mg/L		-	-				0,4	0,6	0,2	0,1	-	-	-	-	
TSS	mg/L	38	-	61	130	54	71	16	17	29	10	180	18	21	14	
Metalle																
Al	mg/L	1200	313	17,3	5,2	78,9	16,7	17,2	5,3	13,2	15,2	8,4	5,3	11	9,8	
Pb	µg/L	4	4	<0,5	<0,5	0,8	<0,5	<0,5	1,7	2,7	3,5	<0,5	0,5	<0,50	1,8	9,0
Cd	µg/L	2	1,7l	<0,05	<0,05	0,05	0,1	<0,05	<0,05	0,05	0	<0,05	<0,05	0,03	0,1	4,5
Cr	µg/L	2,5	16,8	0,97	1,5	1,5	1,5	0,79	1,5	0,5	1,6	0,5	2	0,64	1,5	45
Cu	µg/L	5,5	3	18,6	6	9,5	4,6	17,1	21,6	6,2	12,7	11,1	14,6	12,9	7,8	1800
Fe	µg/L	675	214	178	401	816	729	95,1	85,9	171	139	16,9	50,2	5,2	96,9	
Ni	µg/L	2,7	2	0,91	3,5	1,6	3,3	2,6	4,9	<0,50	3,3	<0,50	3,5	<0,50	1,6	18
Zn	µg/L	24,8	152	184[1]	678[1]	20,8	8,7	487[1]	386[1]	9	16,8	106[1]	555[1]	9,3	13,9	
Nährstoffe																
NO_3^-	µg/L	-	-	-	-			-	-			-	-			
NH_4^+	mg/L	-	-	-	-			-	-			-	-			
H_3PO_4	mg/L	-	-	-	-			-	-			-	-			
H_2SO_4	mg/L	-	-	-	-			-	-			-	-			

[1] *Kontamination durch Probenahmevorrichtung;* [2]Chemie GW Schwellenwert

Tabelle 2: Ergebnisse der Untersuchungen der Ablaufproben der kombinierten Anlage mit Bodenfilter (RVS) und technischem Filtermaterial (TF) bei der Probeentnahmestelle „unten“ versus „Mitten“.

Probenahme Datum		**18.12.2014**				**22.01.2015**		**17.03.2015**				**22.04.2015**				**QZV-GW²⁾**
Probeentnahmestelle		**„unten“**		**„Mitte“**		**„unten“**		**„unten“**		**„Mitte“**		**„unten“**		**„Mitte“**		
Parameter	**Einheit**		**TF**	**RFS**	**TF**	**RFS**	**TF**	**RVS**	**TF**	**RFS**	**TF**	**RVS**	**TF**	**RFS**	**TF**	
pH		6,8	6,9	6,8	7,2	7,9	7,2	7,9	8,1	7,7	7,9	8,1	8	7,8	7,7	
EC	-	240	503	194	424	6100	6400	5580	7150	5270	4460	1700	3200	1400	2900	2250 (20 °C)
Cl	µS/cm	20	71	15	57	590	2000	1700	2200	1600	1300	200	690	140	590	
DOC	mg/L	22,1	26,4			7,6	11,1	17,5	17,8			15,3	18,4			
TOC	mg/L	33,2	34,2	23,4	23,7	26,4	19,2	17,8	18,5	17,2	17,2	15,8	18,9	15,2	15,4	
MOH	mg/L	1,5	0,4	0,1	<0,1	0,4	0,3	0,3	<0,1	0,7	<0,1	1,5	2,6	1,1	2,3	0,1
TNb	mg/L	7,4	8,8	2,3	2,5	6,2	9,4	16,1	15,8	15	14,8	20,7	40,1	22,2	42,6	
TSS	mg/L	21	4	13	27	12	19	24	11	28	9	31	26	25	24	
Metalle																
Al	mg/L	5,3	2,5	5	<5,0	5	2,5	<0,5	<0,5	14,5	<5,0	<0,5	<0,5	12,1		
Pb	µg/L	<0,5	<0,5	<0,50	<0,50	<0,5	<0,5	9,6	5	<0,50	<0,50	33,9	29,2	<0,50	<0,50	9,0
Cd	µg/L	<0,05	<0,05	0,025	0,05	0,07	0,11	0,07	0,06	0,025	0,06	<0,05	<0,05	0,025	0,05	4,5
Cr	µg/L	20	<0,50	0,25	1,4	2,1	2,8	2,2	2,1	1,7	1,8	5,2	7	2,6	7,6	45
Cu	µg/L	8,1	7	1,6	1,6	27,8	29,4	27,9	37,1	32,4	26,7	53,8	26,4	27	24,3	1800
Fe	µg/L	31,7	10,9	431	316	34,4	39,5	65,3	41,1	61,6	22,3	486	211	482	303	
Ni	µg/L	<0,50	0,8	1,2	1,2	8,8	10,3	10,2	9,2	6,2	7,1	3,6	7,7	4,7	8	18
Zn	µg/L	12600[1)]	10700[1)]	<3,0	<3,0	17,4	40,6	24,5	17,2	18,2	5,3	14,1	5,7	11,7	9	
Nährstoffe																
NO_3^-	µg/L	<1,0	3,7	<1,0	<1,0	<1,0	<10	<1,0	<1,0	<1,0	2	<1,0	<1,0	<1,0	<1,0	
NH_4^+	mg/L	-	-			-	-	-	-			19	39	21	42	
H_3PO_4	mg/L	-	-			-	-	1,11	1,39	1,24	1,17	2,38	2,29	2,11	2,65	
H_2SO_4	mg/L	13	17	5,7	<4,0	20	<40	10	14	10	10	17	39	12	36	

[1)] *Kontamination durch Probenahmevorrichtung;* [2)]Chemie GW Schwellenwert

Tabelle 2: Ergebnisse der Untersuchungen der Ablaufproben der kombinierten Anlage mit Bodenfilter (RVS) und technischem Filtermaterial (TF) bei der Probeentnahmestelle „unten“ versus „Mitten“.

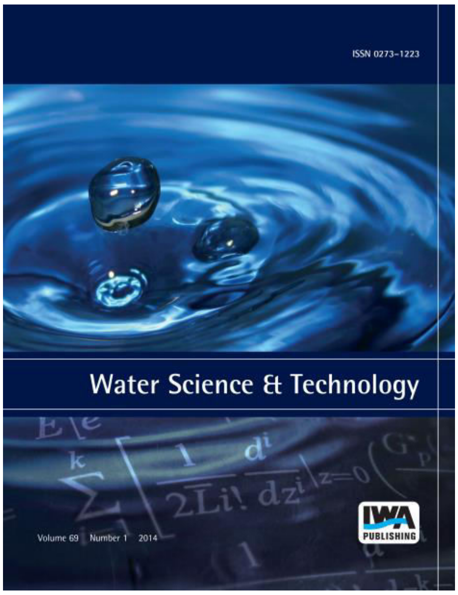
Probenahme Datum		**28.05.2015**				**11.01.2016**		**16.02.2016**		**24.03.2016**		**QZV-GW[2)]**
Probeentnahmestelle		„unten“		„Mitte“		„unten“		„unten		„unten		
Parameter	**Einheit**	**RVS**	**TF**	**RVS**	**TF**	**RVS**	**TF**	**RVS**	**TF**	**RVS**	**TF**	
pH		7,8	7,9	7,6	7,7	7,1	7,1	8,1	8,1	7,2	7,2	
EC	-	1100	1740	980	1650	7880	8920	6550	8940	966	1550	2250 (20 °C)
Cl	µS/cm	56	170	42	160	-	-	2110	3300	140	410	
DOC	mg/L	18	16,9			-	-	-	-	-	-	
TOC	mg/L	18,7	18,9	19,5	18	16,8	17,5	15,1	15,1	6,1	35,5	
MOH	mg/L	0,4	2,6	0,2	0,1	-	0,1	-	-	-	-	0,1
TNb	mg/L	11,6	40,1	8,3	14,7	6,3	12	5,4	5,4	4,1	23	
TSS	mg/L	37	26	25	29	42	31	21	21	10	22	
Metalle												
Al	mg/L	2,5	<0,5	21,8	5	<0,5	<0,5	<0,5	<0,5	0,5	<0,5	
Pb	µg/L	22,8	29,2	<5,0	<5,0	7,5	2,5	5	5	2,5	7,7	9
Cd	µg/L	0,05	0,05	0,05	0,025	0,07	0,05	0,12	0,12	0,05	0,07	4,5
Cr	µg/L	3,3	7	2	3,7	2,5	5,1	2,4	2,4	1,1	5	45
Cu	µg/L	21,1	26,4	9,7	11,9	17,8	30,3	28,7	28,7	33	17,1	1800
Fe	µg/L	340	211	251	257	25,1	78	30,9	30,9	16,1	540	
Ni	µg/L	1	7,7	0,7	1,8	6,2	1,7	8,6	8,6	4,6	13,8	18
Zn	µg/L	19,4	5,7	11,5	11,2	39,6	33,6	42,6	42,6	18,6	15,1	
Nährstoffe												
NO_3^-	µg/L	<1,0	<1,0	<1,0	<1,0	<1,0	<1,0	-	-	3	< 2,5	
NH_4^+	mg/L	9,5	39	7,4	14	11,6	11	-	-	1,4	20	
H_3PO_4	mg/L	1,08	2,29	1,04	1,08	1,05	0,877	-	-	0,5	1,15	
H_2SO_4	mg/L	5,6	39	<4,0	9,3	9,7	20	-	-	7	28	

[1)] *Kontamination durch Probenahmevorrichtung;* [2)]Chemie GW Schellenwert

6.4 Paper IV

Cytotoxic and mutagenic activities of waters and sediments from highway and parking lot runoffs.

Haile, T.M., Mišík, M., Grummt, T., Halh, A.S., Pichler, C., Knasmueller, S., Fuerhacker, M. (2016). Water Science and Technology (in press). doi: 10.2166/wst.2016.137.


ISSN 0273-1223
Water Science & Technology
Volume 69 Number 1 2014
IWA PUBLISHING

Cytotoxic and genotoxic activities of waters and sediments from highway and parking lot runoffs

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ABSTRACT

The genotoxicity of water and sediment samples from stormwater treatment systems and water from urban highway runoff was tested in the *Salmonella*/microsome assays with *Salmonella typhimurium*, micronucleus assay (Trad-MN) with plants and with human-derived liver cells (HepG2), or comet assay with HepG2. Cytotoxicity of water samples was studied using either reactive oxygen species (ROS) generation, cell proliferation or dye exclusion assay in HepG2. Concentrations of several contaminants in the tested samples were also measured. Results suggested that urban highway runoff exposed to severe vehicle traffic emissions caused genotoxic effects in comet assay and in Trad-MN assays. Sediments induced either mutagenic effects in strain YG1024 or genotoxic effects in Trad-MN assay. These effects could be due to the presence of nitro-polycyclic aromatic hydrocarbons (NPAHs) which possess carcinogenic and mutagenic properties. Influent and effluents of stormwater treatment systems did not induce genotoxic activity or effects on HepG2 cell viability; however, the influents were able to induce ROS generation and cell proliferation in HepG2 cells. As the methods require a sterile filtration of the water samples, this could have also removed particulate-associated polycyclic aromatic hydrocarbons (PAHs) and resulted in a less pronounced induction of genotoxicity, as would be expected by PAH contamination.

Key words | contaminants, cytotoxicity, genotoxicity, mutagenicity, stormwater runoff

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INTRODUCTION

Stormwater runoff from vehicle traffic areas (e.g. highways) contains numerous contaminants including metals, polycyclic aromatic hydrocarbons (PAHs) and mineral oil hydrocarbons (MOH) (Shinya *et al.* 2000; Fuerhacker *et al.* 2011) and nitro-polycyclic aromatic hydrocarbons (NPAHs) (Murakami *et al.* 2008). PAHs and NPAHs are environmental contaminants with carcinogenic and mutagenic properties (Shinya *et al.* 2000; Ohe *et al.* 2004; Murakami *et al.* 2008). The genotoxic effects of stormwater runoff from roadways and roadside soils have been investigated by several researchers (Shinya *et al.* 2000; Tsukatani *et al.* 2002). For example, Shinya *et al.* (2000) showed that particulates from highway runoff were mutagenic in bacteria and these effects were attributed to NPAHs which have stronger carcinogenic and mutagenic activities than the parent PAHs (Wantabe *et al.* 1990). Currently, several genotoxicity tests are available, i.e. *Salmonella*/microsome assay, micronucleus (MN) assays with plant species such as *Tradescantia* clone 4430 and comet assay with human-derived cells (HepG2). These models have been used to investigate the genotoxicity of pure chemicals and of environmental samples (Tsukatani *et al.* 2002; Chen & White 2004; Ohe *et al.* 2004; Keiter *et al.* 2006; Žegura *et al.* 2009). The *Salmonella*/microsome assay is currently the most widely used test to detect the mutagenicity of contaminated sediments, soils and waters (Chen & White 2004; Ohe *et al.* 2004; Keiter *et al.* 2006). It is based on the detection of induction of mutations in strains of *Salmonella typhimurium* which lead to histidine auxotrophy (Maron & Ames 1983). The bacteria are sensitive towards a variety of agents including nitro-aromatic chemicals that are contained in emissions from engines (Rosenkranz 1982). Single cell gel electrophoresis (SCGE) is used to measure DNA single strand breaks, double strand breaks, crosslinks and alkali-labile sites (Tice *et al.* 2000). This method has been widely

doi: 10.2166/wst.2016.137

used to investigate genotoxic effects of pure chemical substances (Uhl *et al.* 1999), water (Žegura *et al.* 2009), and sediments (Costa *et al.* 2014).

The MN test with pollen tetrads of *Tradescantia* is the most widely used plant bioassay for the detection of genotoxins in the environment (for review see Misík *et al.* (2011)). Plants are highly sensitive to metals (Knasmueller *et al.* 1998); therefore the MN assay can provide information on environmental effects of stormwater runoff where metals are common constituents.

Only a few studies reported on the cytotoxicity, mutagenicity and genotoxicity of stormwater from roadways (Shinya *et al.* 2000) and roadside soil (Tsukatani *et al.* 2002). Therefore the aim of the present study was to assess the genotoxic effects of runoffs from a highway and a car parking lot, and of sediments accumulated within stormwater runoff treatment systems. To achieve this goal a combination of methods, namely the *Salmonella*/microsome test, comet assays with HepG2 cells and MN assays with plants (*Tradescantia*) as well as HepG2 cells were applied. Furthermore, the concentrations of several contaminants in the tested samples were also measured.

MATERIALS AND METHODS

Sampling sites

Water and sediment samples were collected from three sites. Two sites, GSA-H and GSA-W, are stormwater treatment systems which receive runoff from a car parking lot and a less trafficked highway respectively. Both sites are located in Lower Austria along highway A-21. The treatment systems (GSA-H and GSA-W) consist of a sedimentation tank and a filter bed with Aquafilt® adsorption media and a geotextile layer on top (for details see Fuerhacker *et al.* (2011)). The third site is an urban highway stormwater runoff collection shaft (A23) within the city of Vienna which has a daily average traffic of 255,000 vehicles.

Sampling procedure and preparation

Grab water samples were collected from the influent and effluent of the stormwater treatment systems (GSA-H and GSA-W) and the urban highway runoff collection shaft (A23), at the end of the winter (A23-1, March 2011) and in the summer (A23-2, July 2013). Sediments from the surface of the filter bed of GSA-H and GSA-W were collected in June 2012 in plastic bags and were refrigerated at 4 °C until analyses. Water samples for chemical analyses were collected with glass bottles and samples for the toxicity test were collected using a 20 L polypropylene jerry can (Menke Industrieverpackungen GmbH & Co. KG, Beckedorfer Bogen, Germany). For the chemical analyses water samples were preserved and stored in a dark room at 4 °C until analyses. For the toxicity tests water samples were stored at −20 °C and before use in experiments were filtered with 0.2 μm filter (Whatman, Sigma, St Louis, USA). Dimethyl sulfoxide (DMSO) extracts of sediment samples (48.6–54.4 g) were prepared according to the method of Keiter *et al.* (2006) with 340 mL acetone.

Toxicity tests

Cytotoxicity test

For cytotoxicity testing, the following three procedures were applied.

(a) *xCELLigence* System (Real-Time Cell Analyser, RTCA). The experiments were performed using a real-time xCELLigence cell (impedance system, according to the manufacturer's instructions (ACEA Bioscience ACEA Biosciences, Inc.). HepG2 cells (20,000 cells/well) were cultivated in 96-well multiwell plates (E-plates, OLS Omni Life Science GmbH & Co. KG, Bremen). Details are in Leme *et al.* (2011). Measurements were made hourly. The increasing impedance is plotted against the 24 h exposure period, resulting in a slope. The slope of the test sample is divided by the slope obtained for the negative control or by the slope of the solvent control. A quotient around 1 indicates no effect, quotients <0.7 are indicative of an effect.

(b) Generation of reactive oxygen species (ROS). This experiment was conducted using 2′,7′-dichloro-dihydrofluoresceine diacetate (DCFH-DA). This reagent diffuses into living cells. Dichlorofluorescein (DCF) is oxidised by intracellular ROS to green fluorescent DCF. Fluorescence measurements were carried out with a 'Genios' fluorescence plate reader ($\lambda = 485$ nm and 535 nm) for excitation and emission, respectively. To avoid side scatter effects HepG2 cells (1×10^5/well) were cultivated in black 96-well multiwell plates with flat, transparent bottom. A calibration curve with known DCF concentrations facilitates the quantification of the results. An increase in ROS (oxidative DNA damage) was defined as DCF concentrations higher than 20 nM. 3-Morpholinosydnonimine (0.5 mM) was used as a positive control.

(c) Dye exclusion assay with HepG2 cells. The vitality of human-derived liver cells was determined in each experiment with the trypan blue (0.4%) dye exclusion technique (Lindl & Bauer 1989).

Genotoxicity test

Salmonella/microsome assays. The mutagenicity of water (GSA-H and A23-2) and sediment (GSA-H and GSA-W) samples was assessed in *Salmonella* plate incorporation assays according to the protocol of Maron & Ames (1983). The tests were carried out with tester strains TA98, TA100 and YG1024 as described by Ames *et al.* (1973), in presence and absence of metabolic activation mix (S9 mix). YG1024 is a derivative of strain TA98 which over-expresses an *O*-acetyltransferase gene that confers high sensitivity to the mutagenic action of nitroarenes and aromatic amines (Wantabe *et al.* 1990). The test strains were provided by Dr Tamara Grummt (Federal Environment Agency, Germany). The characteristics of the strains were tested as described by Ames *et al.* (1973). Different amounts of sterile filtered water samples (100–500 μL per plate) or DMSO extract were prepared according the methods of Keiter *et al.* (2006) with 340 mL acetone for 20 g of each sediment. The extracts were diluted with DMSO and a range of different concentrations were tested, which corresponded to 250 to 1,000 mg sediment (per plate). Per experimental point, three plates were made. In all experiments, positive controls were included and dissolved in DMSO. In experiments without metabolic activation mix (−S9) the positive controls were TNF (2,4,7-trinitro-9-fluorenone, 0.1 μg/plate) for TA98 and YG1024, and NaN_3 (sodium azide, 1.5 μg/plate) for TA100. For experiments with metabolic activation (+S9) 2AA (2-aminoanthracene, 1.0 μg/plate for water samples and 0.5 μg/plate for sediment samples) was used as positive control for all three strains.

Single cell gel electrophoresis assays with HepG2 cells

The human-derived liver cell line (HepG2) was used to perform comet assays. The cell line was kindly provided by F. Darroudi (Department of Toxicogenetics, Leiden University Medical Centre, The Netherlands).

The experiments were conducted under standard alkaline conditions which reflect the formation of single and double strand breaks and apurinic sites according to the modified version of the protocol described by Tice *et al.* (2000). The cells were exposed in the control medium, in the undiluted water samples (100%), or in mixtures of the two, containing either 11 or 33% of the test substance for 60 min at 37 °C. From each concentration triplicates were made. Air-dried slides were stained with ethidium bromide (20 μg/mL) and the percentage of DNA in the tail was measured by use of a computer aided image analysis system (Comet IV, Perceptive Instruments Ltd, Haverhill, UK). For each experimental point, three cultures were made in parallel; from each culture one slide was prepared and from each slide 50 randomly distributed cells were evaluated. The experiments were performed in duplicate. DNA damage was only analyzed in cells from cultures in which the viability was ≥80%, as acute toxic effects may cause false positive results.

MN assays with HepG2 cells

The test was performed according to the OECD guideline 487 (OECD 2010). Vitality of the HepG2 cells (1.8×10^4/mL) as determined by trypan blue stain has to be at least 90%. The assay was performed in Quadri-PERM® dishes (Greiner Bio One, Frickenhausen, Germany) with four separate chambers. Cells are exposed for 24 h. Afterwards cells are subjected to a hypotonous solution (1.5% trisodium-citrate solution in phosphate-buffered saline) for 5 minutes, followed by two fixation steps (3:1 mixture of methanol and glacial acetic acid) for 10 minutes each. After air drying a Giemsa stain is the final step of the preparation. Microscopic analyses are performed using a Metafer4 automated scoring device (MetaSystem, GmbH, Altlußheim, Germany). For each experimental point 1,000 cells were evaluated. MN were defined by the following properties: less than 30% of the size of a normal nucleus, nucleus and MN possess the same staining pattern and each MN has to be clearly separated from the main nucleus.

Tradescantia MN assays

Tradescantia MN assays were performed according to the protocol of Ma *et al.* (1994). Clone #4430 was used in this study. Per dose, 15 cuttings were treated for 24 h followed by a 24 h recovery period. The cuttings were placed into glass beakers with 100 mL tap water or with the water samples. After exposure, the inflorescences were fixed for 24 h in a mix of ethanol/acetic acid (3:1) and stored in 70% ethanol. Per experimental point, tetrad preparations of at least five buds were made and stained with 2% acetocarmine; 1,500 early phase tetrads (five inflorescences from individual plants and 300 tetrads per bud) were scored in each experimental group. For water samples from GSA-H and A23-2 the tests were conducted with different concentrations (25, 50 and 100%). GSA-H sediment was used as sampled (field moist); 20 g was suspended in 40 mL of tap water in a 100 mL glass beaker and stirred for 6 hours. The extracts were subsequently diluted with tap water (50, 25 and 12.5%), which was used in all experiments as a control. Maleic hydrazide (MH, 20 mg/L) was used in all experiments as a positive control.

Data analysis of toxicity test

Results of ROS and xCELLigence system are reported as means ± standard deviations (SD). Student's *t*-test was performed between pairs of grouped data and a $P \leq 0.05$ was considered statistically significant.

The results of the SCGE and Trad-MN assays were statistically analysed with the GraphPad Prism 5 software system (GraphPad Software, Inc., CA, USA). All results are reported as means ± SD. Results of SCGE assays were analysed by Student's *t*-test and Trad-MN assays with one-way analysis of variance (ANOVA) followed by Dunnett's multiple comparison. P values ≤ 0.05 were considered as statistically significant.

Results of the MN assay with HepG2 were log transformed. Data were then analyzed by one-way ANOVA. Comparisons against negative controls were conducted by Dunnett's test. P values ≤ 0.05 were considered significant.

Results of the *Salmonella*/microsome tests were evaluated on the basis of the 'twofold rule' (for details see Mahon *et al.* (1989)).

Physico-chemical analyses

The concentrations of metals (Ba, Cd, Cr, Cu, Ni, Pb, Ti, V and Zn) in the water and sediment extracts were measured using an inductively coupled plasma mass spectrometer (Perkin-Elmer, Sciex, Canada). The concentrations of 16 US Environmental Protection Agency PAHs and MOH were determined using gas chromatography mass spectrometry.

RESULTS AND DISCUSSION

Physico-chemical characteristics of the water and sediment samples

The characteristics of the water and sediment samples collected from GSA-H, GSA-W and A23 are summarised in Table 1. Among the metals, Zn has the highest concentrations and this observation is in line with its relatively high content observed in roadway runoff water as reported in several studies (Shinya *et al.* 2000; Fuerhacker *et al.* 2011).

The concentrations of all contaminants in A23-1 (winter season) were higher than the levels in the A23-2 (summer season) and also higher than the concentrations at GSA-H and GSA-W. The concentrations of metals in A23-1 were within the ranges found in heavily trafficked urban roads by Shinya *et al.* (2000). For the water samples collected from A23-1, the concentrations of individual PAHs in the dissolved phase were mostly below detection limit, indicating that particulate-bound PAHs were significant contributors to the PAH loads. A highway runoff study by Murakami *et al.*

Table 1 | Concentrations of metals, PAHs and MOH in water and sediment samples

	Water*						Sediment‡	
	GSA-H		GSA-W		A23			
Parameter	Inf	Eff	Inf	Eff	A23-1	A23-2	GSA-H	GSA-W
EC	1,210	1,300	930	810	7,200	1,060		
Ba	51.3	20.1	25	8.9	471	177	325	135
Cd	<0.1	<0.1	<0.1	<0.1	1.3	<0.1	0.6	0.49
Cr	5	1.7	7.7	1.3	71.6	11.1	81	86
Cu	14.2	14.8	39	10.8	682	131	490	326
Ni	5.3	4.8	4.8	4.2	68.8	9.8	46	52
Pb	<4.0	<4.0	<4.0	<4.0	112	205	57	114
Ti	3	<0.3	34	4.9	630	132	475	589
V	7	3	2.9	2.3	44.6	12.1	37	44
Zn	81.3	7.7	331	20.7	2,560	633	1449	3016
Σ16 PAHs	<0.21	<0.15	0.2	<0.09	18.25	n.a.	10.492	n.a.
MOH	0.2	< 0.1	0.2	< 0.1	3	n.a.	n.a.	n.a.

Data represent concentrations of grab water sample and sediments used for genotoxicity tests. Inf, influent; Eff, effluent; n.a., not analysed.
Reporting units: *water samples, EC (electrical conductivity) in μS/cm, metals and Σ16 PAHs in μg/L and MOH in mg/L; ‡sediment samples, metals in mg/kg and Σ16 PAH in μg/kg.

(2008) found that the particulate fraction of PAHs and NPAHs represented 70–99% of the total concentrations.

The concentration ratios of the metals (e.g. Cu/Zn ratio) in the sediments were comparable to the concentration ratios of roadway runoff, indicating that traffic-related activities are the main anthropogenic pollution sources. The present study reports relevant information regarding the presence of a wide range of metals, with high concentrations found in water and sediments, which are not properly regulated (e.g. Ba, Ti and V). The chemical status of the sediments revealed metal concentrations exceeding the environmental threshold and the probable effect levels (MacDonald et al. 1996). Our findings indicate that there is a demand to investigate the behaviour and mode of action of these metals.

Results of toxicity (genotoxicity and cytotoxicity) test

The cytotoxic and genotoxic effects of water and sediment extracts tested using several assays (see 'Material and methods' section) are summarised in Table 2.

ROS production

Overproduction of ROS can induce oxidative stress and thus affects the viability of cells. In this study, high levels of ROS were generated with the GSA-W influent and A23-1 water samples (see Table 2). After 4 h exposure and at a dilution factor of 1:2 in local tap water, the production of ROS was 3.05- and 6.25-fold higher than the no-effect level. Conversely, the effect of ROS production in HepG2 cells exposed to purified highway runoff (GSA-W effluent) was negative. The tested samples were not filtered and it is likely that the production of ROS was induced by the metals and PAHs which were found in the water samples (GSA-influent and A23-1). Metals such as Cd, Cu, Fe, Ni, Ni and Pb (Birben et al. 2012) and PAHs (Leme et al. 2011) are known to induce ROS formation and thus cause oxidative stress which affects all cellular macromolecules including DNA, lipids, and proteins (Birben et al. 2012).

Effect on cell proliferation

The real-time cell proliferation (RTCA) results show that the influent of GSA-W was able to increase cell proliferation in HepG2 cells (see Table 2). The water samples from A23-1 and GSA-W effluent did not show proliferating effects. The cell proliferation rate is associated with cytotoxicity effects; thus the results of the real-time cell impedance analyses showed that the influent of GSA-W was cytotoxic to the HepG2 cells.

The impact of water samples on the viability of HepG2 cells

Under the different experimental conditions (comet, MN, ROS and cell proliferation assays), the viability of HepG2 cells exposed to water (A23-1, A23-2, GSA-H and GSA-W)

Table 2 | Cytotoxicity and genotoxicity test result

	ROS[1,†]	Cell proliferation[2,†]	Cell viability[†]	MN[†]	Trad-MN	Comet[†]	Ames[3]
Water							
GSA-H (Inf)	nt	nt	–	nt	–	–	–
GSA-H (Eff)	nt	nt	–	nt	–	–	–
GSA-W (Inf)	+ (125 nM)	+ (0.52)	nt	–	nt	nt	–
GSA-W (Eff)	– (13 nM)	– (0.74)	nt	–	nt	nt	–
A23-1	+ (63 nM)	– (1.1)	nt	–	nt	nt	–
A23-2	nt	nt	–	nt	+	+	–
Sediment							
GSA-H	nt	nt	nt	nt	+	nt	–
GSA-W	nt	nt	nt	nt	nt	nt	+

[1]ROS (DCFH-DA) data corrected by the 1:2 dilution with local tap water; – no effect ≤20 nM DCF, +effect >20 nM.
[2]Slope of the electrical impedance measured by the RTCA system, quotient = slope of cells exposed to the test samples divided by the slope of the control cells (dilution of tap water). A quotient around 1 indicates no effect, quotients ≤0.7 are indicative of an effect.
[3]The test was conducted with strains TA98, TA100 and YG1024; only the GSA-W sediment was positive in the strain YG1024.
[†]The experiments were conducted with a human-derived liver cell line (HepG2).
Note: – no effect; + indicative of effect (positive response); nt, not tested; Inf, influent; Eff, effluent.

at different concentrations of the test samples (11–100%) for 24 hours was not significant (data not shown). Only a slight decrease of the vitality was observed after exposure of the cells to the highway runoff (A23-2).

Results of bacterial mutagenicity tests

The mutagenic activity of water samples (A23-1, A23-2, GSA-H and GSA-W) and sediment deposited within the stormwater treatment systems (GSA-H and GSA-W) was evaluated in *Salmonella*/microsomal assays using strains TA98, TA100 and YG1024 (for more details see supplementary material Table A1, available with the online version of this paper). The results showed that none of the samples induce mutagenic effect, except the sediment sample from GSA-W in strain YG1024. In all experimental series, the spontaneous revertant frequencies of the negative controls were in the expected range (Kirkland 1990). Furthermore, clear-cut induction of his^+ revertant colonies was seen with all compounds which were used as positive controls. None of the native water samples induced a significant increase in the number of his^+ revertants. However, with strain YG1024, which is a derivate of TA98 and possesses a high level of *O*-acetyltransferase activity (Wantabe *et al.* 1990), positive results were detected with the GSA-W sediment DMSO extract in the absence of metabolic activation mix at the highest dose (1,000 mg/plate); also with lower doses a higher number of revertants were seen, but this effect did not reach significance (see supplementary material Table A1). The same extract caused also an increase (non-significant) of revertant frequencies in TA98 with and without activation mix. Although the concentrations of NPAHs in the sediments (GSA-W) were not measured, the observed mutagenic effect in YG1024 strain suggests that NPAHs which are activated by *N*-acetyltransferase are present in the extract (Shinya *et al.* 2000; Tsukatani *et al.* 2002).

Some studies found that urban highway runoff and roadside soils induced mutagenic effects in the Ames assay. Shinya *et al.* (2000) reported that extracts of water samples collected from urban highway runoff were mutagenic in strains TA98, YG1021 and YG1024 with or without S9 mix. Tsukatani *et al.* (2002) employed the *Salmonella* assay and found that the extracts of roadside soils (Kurume City, Japan) showed much higher mutagenicity in YG1041 and YG1042 stains than in TA98 and TA100 strains. The mutagenic activity of the roadside soils was correlated with the concentrations of PAHs (Tsukatani *et al.* 2002). No correlation was found between the level of mutagenicity and the concentrations of PAHs in the present study. This could be due to the filtration of the water samples before the mutagenicity tests. Because most roadway runoff pollutants (particularly metals, PAHs and NPAHs) are particulate-bound (Shinya *et al.* 2000; Murakami *et al.*

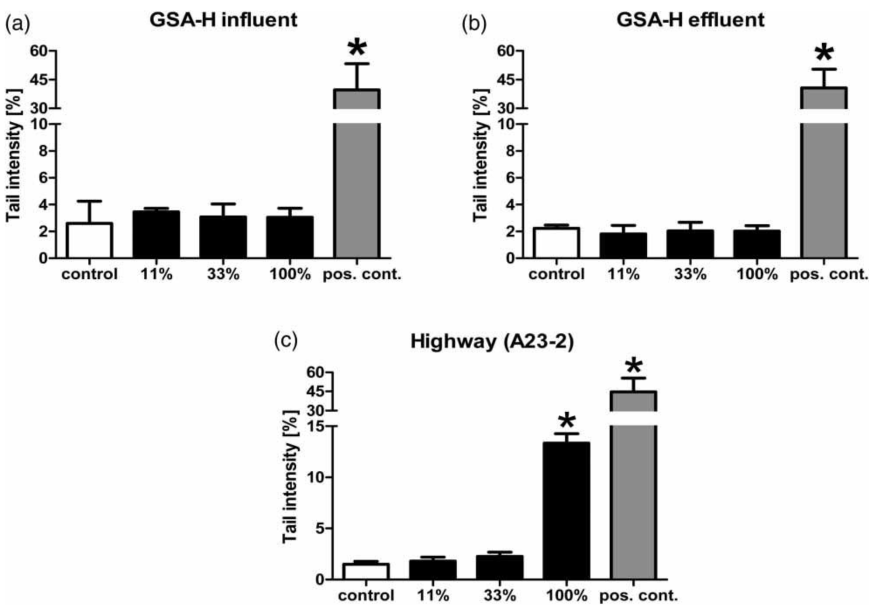

Figure 1 | Results of the SCGE experiments with water sample of GSA-H and highway runoff (A-23). The HepG2 cells were exposed for 60 min (37 °C, 5% CO_2). White bars represent data with untreated control cultures, black bars represent data with cells which have been exposed to different concentrations of the test samples (11–100%) and grey bars represent the positive control (30 μM H_2O_2 treatment). For each experimental point, three cultures were set up in parallel and from each culture, 50 cells were analyzed for DNA damage. Bars represent means ± SD of results obtained with three parallel cultures. Parts (a) and (b) show results of representative experiment. Stars indicate statistical significance (t-test, $*p \leq 0.05$).

2008), filtration through a 0.2 µm pore size filter has important implications on the mutagenicity potential of the water sample.

Induction of DNA damage by the different water samples in HepG2 cell line (SCGE experiments)

The results of the genotoxicity tests with water samples collected from GSA-H and A23-2 in the comet assay in HepG2 cells are indicated in Figure 1.

It can be seen that no significant differences were observed between controls and cells treated with different dilutions of GSA-H (11, 33, and 100%). Overall, no evidence for genotoxic effects was detected after exposure of the cells to the tested water from GSA-H under any test conditions. However, the water sample from A23-2 was genotoxic at the highest test dose (Figure 1).

Induction of micronucleus

The MN frequencies induced by the water and sediment samples in the Trad-MN assay are summarized in Figure 2.

The results indicate that both the influent and effluent water samples from GSA-H did not induce the formation of micronucleated cells at all doses used in the experiments (Figure 2(a)); only a slight (not significant) increase of the

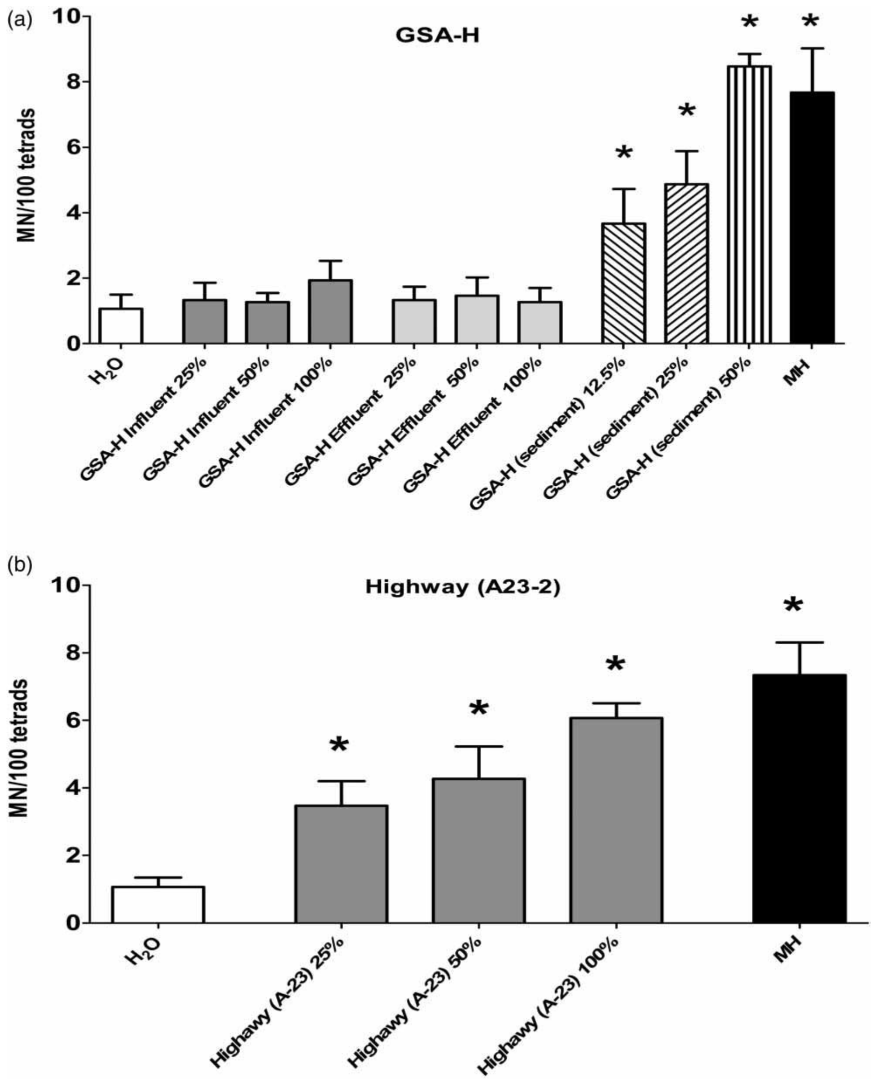

Figure 2 | Induction of MN in early tetrads of *Tradescantia* #4430. Tap water was used as negative control; MH (20.0 mg/L) was used as a positive control. The plants were treated in stem absorption experiments for 24 h followed by a 24 h recovery phase. For each experimental point, five buds (300 tetrads/bud) were analysed for MN formation. Bars indicate means ± SD. Figure shows results of representative experiment. Stars indicate statistical significance (ANOVA, with Dunnett's multiple comparison test, $*P \leq 0.05$).

MN rates was detected at the 100% dose. In contrast, the highway runoff (A23-2) and the sediments (GSA-H) significantly increased the formation of micronucleated cells at all the doses used in the experiments (Figure 2(a) and 2(b)) and a dose–response was evident. For the sediment samples, at dose levels of 12.5, 25 and 50% the respective MN-rates were 4-, 6-, and 9-fold higher as compared to the negative control, respectively. The MN-formation rate at the highest sediment dose (50%) was even more effective than the positive control MH 20 mg/L (Figure 2(b)). In roadway runoff the majority of metals and PAHs are associated with particulate matter; therefore the observed MN frequencies in the GSA-H sediments (Figure 2(a)) could be due to the accumulation of pollutants within the particle layer. The results from this study showed that it is important to carry out both chemical analyses and genotoxicity/mutagenicity tests to be able to correctly evaluate the potential environmental impact of water and sediments contaminated with roadway runoff.

In addition to the Trad-MN assay, the MN assay was performed in HepG2 cells with water samples (influent and effluent of GSA-W and A23-1). In the MN-HepG2 assay after a 24-hour exposure period, none of the tested samples increased significantly the number of MN over the negative control frequency for all test variants.

CONCLUSIONS

The results of the present study lead us to conclude that stormwater runoff from an urban highway which was exposed to high vehicular traffic presented responses indicative of mutagenic and genotoxic effects. The investigated stormwater treatment systems were efficient in removing the toxic pollutants, as effluents did not induce cytotoxicity and genotoxicity effects. The influents were (at least partly) cytotoxic but there was no clear genotoxicity or mutagenicity effects. Sediments (DMSO extracts) deposited within stormwater treatment systems caused either mutagenic effects in the Trad-MN assay or genotoxicity effects in the *Salmonella*/microsomal assay. As the composition of stormwater runoff varies strongly, the sediment gives a more integrated answer in the test system. It is known that NPAHs are found in exhaust gases of cars (Murakami *et al.* 2008). Therefore, the observed genotoxic and mutagenic activity in DMSO extracts suggested that particulate-bound carcinogenic and mutagenic PAH and NPAH compounds existed in stormwater runoff from vehicle traffic areas. It cannot be excluded that also heavy metals contributed to the effects that were detected. It is known that certain metals (such as Cu and Zn) cause induction of MN in *Tradescantia* and in some samples the levels detected were sufficiently high to induce MN formation (see Steinkellener *et al.* 1998). Also, in the SCGE experiment with HepG2 cells, metal may have been implicated in the effects, which was found with one of the sample a (A23-2). However, it is unlikely that metals account for positive results in *Salmonella*/microsome assays as they do not induce gene mutation in coliform bacteria, possibly as a consequence of low uptake into the cells (Gatehouse *et al.* 1990). Furthermore, as stormwater runoff contains a complex mixture of organic and inorganic contaminants, the observed mutagenicity and genotoxicity may be attributed to the combined effect of PAHs and metals. Further work is needed to integrate bioassays with analytical methods to better determine and quantify environmental impacts.

ACKNOWLEDGEMENTS

The Austrian Federal Ministry of Agriculture, Forestry, Environment and Water Management (BMLFUW) and SW-Umwelttechnik are gratefully acknowledged for providing funds. We sincerely thank Alexander Eckhardt and Rita Heinze for their help with the toxicity assays.

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First received 25 September 2015; accepted in revised form 24 February 2016. Available online 14 March 2016

6.5 Paper V

Testing methods for filter materials: Application of ÖNORM B 2506 Part 3 for roads with high annual average daily traffic (AADT). "Filtermaterialprüfung: Anwendung der ÖNORM B 2506 Teil 3 für das hochrangige Straßennetz, in German".

Haile, T.M. and Fuerhacker, M. (2016). Österr Wasser- und Abfallw. 69, :495–502. https://doi.org/10.1007/s00506-017-0427-7

Österr Wasser- und Abfallw 2017 · 69:495–502
https://doi.org/10.1007/s00506-017-0427-7

Filtermaterialprüfung: Anwendung der ÖNORM B 2506 Teil 3 für das hochrangige Straßennetz

Tadele Measho Haile · Maria Fürhacker

Online publiziert: 18. September 2017

Zusammenfassung Verkehrsflächenabflüsse können mit organischen und anorganischen Stoffen belastet sein und als verunreinigt gelten, sodass sie vor Einbringung in den Untergrund gereinigt werden müssen. Die Belastungen stammen bzw. entstehen aus Abgasnebenprodukten, Reifen-, Karosserie- und Fahrbahnverschleiß, Abflüssen aus Niederschlägen, nasser und trockener Deposition und Fahrbahninstandhaltungsarbeiten. Im ÖWAV-Regelblatt 45 und in der ÖNORM B 2506, Teil 1 und 2 wird der Stand der Technik der Reinigung vor Versickerung in den Untergrund mit Bodenfiltern bzw. „technischen Bodenfiltern" (ÖNORM) und „technischen Filtern" (ÖWAV-RB 45) beschrieben. Die Kriterien der Mindestleistungsfähigkeit und deren Prüfung wurden in der ÖNORM B 2506, Teil 3 festgelegt. Da sowohl in der ÖNORM B 2506, Teil 1 und 2 als auch im ÖWAV-RB 45 die hochrangigen Straßen ausgenommen wurden, sollen in diesem Artikel die Grundlagen der ÖNORM B 2506, Teil 3 erläutert und ihre Anwendbarkeit auch auf hochrangige Straßen aufgezeigt werden. Es konnte gezeigt werden, dass aufgrund der in der ÖNORM B 2506-3 gewählten strengen Prüfbedingungen und Prüfkriterien die Prüfung der technischen Filtermaterialien aus wissenschaftlicher Sicht geeignet sind, auch die Anforderungen an die Reinigung von Straßenabwässern von hochbelasteten Straßen mit hohen durchschnittlichen täglichen Verkehrsbelastungen (JDTV), wie jenen des hochrangigen Straßennetzes, zu erfüllen. Es sei noch darauf hingewiesen, dass die Prüfung der technischen Filtermaterialien nach ÖNORM B 2506-3 für die Versickerung in das Grundwasser erstellt wurde.

Schlüsselwörter Straßenabwässer · Materialprüfung · ÖNORM B 2506 Teil 3 · Hochrangige Straßen · JDTV >15.000

Testing methods for filtermaterials: Application of ÖNORM B 2506 Part 3 for roads with high annaual average daily traffic (AADT)

Abstract Street runoff can be contaminated with organic and inorganic substances, and therefore have to be treated before being infiltrated in the underground. The contaminations are from tires, vehicles, roads, precipitation, wet and dry deposition, road maintenance work or are generated by exhaust gas products. The state of the art of cleaning before infiltration into the underground is described in the ÖWAV-Regelblatt 45 and ÖNORM B 2506 Parts 1 & 2, with the help of soil filters or "technical soil filters" (ÖNORM) and "technical filters" (ÖWAV-RB 45). The testing methods and performance criteria for such filters have been defined in ÖNORM B 2506 Part 3. Since both the ÖNORM B 2506 Parts 1 & 2 and the ÖWAV-RB 45 have exempted the high-ranking roads, this article explains the background of ÖNORM B2506 Part 3 and its applicability for run-off treatment of high-ranking roads. It has been shown that due to the strict test conditions and test criteria chosen in ÖNORM B 2506-3, the testing of the technical filter materials is from a scientific point of view also suitable, to meet the requirements for roads with high annual average daily traffic (AADT). It should be pointed out that the testing methods according to ÖNORM B 2506-3 was designed for infiltration into the underground.

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1 Einleitung

Abwässer von Straßen können relativ stark mit Spurenstoffen belastet sein. In der Qualitätszielverordnung Chemie Grundwasser ist ein Verbot der direkten Einbringung von Schadstoffen in das Grundwasser enthalten, wobei unter direkter Einbringung jede dauernde oder zeitweilige Einbringung von Schadstoffen in das Grundwasser ohne Bodenpassage zu verstehen ist. Aus diesem Grund ist die Behandlung von Straßenabwässern besonders bei stark befahrenen Straßen vor der Einleitung in ein Gewässer oder in den Untergrund erforderlich. Im ÖWAV-Regelblatt 45 (ÖWAV-RB 45 2015) und in der ÖNORM B 2506, Teil 1 und 2 wird der Stand der Technik der Reinigung vor Versickerung in den Untergrund beschrieben. Da in den Vorschriften neben Bodenfiltern auch Aussagen zu „technischen Bodenfiltern" (ÖNORM B 2506-2 2012) bzw. zu „technischen Filtern" (ÖWAV-RB 45 2015) gemacht werden, war es notwendig, diese zu charakterisieren bzw. eine Mindestleistungsfähigkeit zu fordern. Die Kriterien der Mindestleistungsfähigkeit und deren Prüfung wurden in der ÖNORM B 2506-Teil 3 (ÖNORM B 2506-3 2016) festgelegt.

Allerdings sind sowohl in der ÖNORM B 2506-Teil 3 als auch im ÖWAV-RB 45 die hochrangigen Straßen nicht berücksichtigt, weil diese durch die Vorschriften der FSG in den RVS konkretisiert wurden. Ziel dieses Artikels ist es, die Grundlagen der ÖNORM B 2506-Teil 3 zu erläutern und ihre Anwendbarkeit auch auf hochrangige Straßen aufzuzeigen.

2 Verunreinigungen von Straßenabwässern

Verkehrsflächenabflüsse können mit organischen und anorganischen Spurenstoffen, Feststoffen (z. B. abfiltrierbaren Stoffen) und Nährstoffen belastet sein (Fürhacker et al. 2013; Geiger-Kaiser und Jäger 2005; Göbel et al. 2007;

Tab. 1 Typische Spurenstoffe in Niederschlagsabflüssen von befestigten Verkehrsflächen und ihre Hauptquellen. (Nach Ball 1998; Davis et al. 2001; Geiger-Kaiser und Jäger 2005; Horstmeyer und Helmreich 2014; Legret und Pagotto 1999; McKenzie et al. 2009; Sansalone und Buchberger 1997; Thorpe und Harrison 2008; Zafra et al. 2011)

Stoffe	Hauptquellen
Ba	Bremsbeläge und Verkleidungen
Cd	Reifenverschleiß, Bremsbeläge, Schmieröle, Verbrennung und Korrosion
Co	Verschleiß von Spikereifen, Korrosion von Buchsen, Bremsdrähten und Heizkörpern
Cr	Bewegliche Motorenteile, Bremsbeläge, Asphalt- und Fahrbahnverschleiß, Korrosion von geschweißten Metallbeschichtungen und Lacken
Cu	Lager- und Buchsenverschleiß, bewegliche Motorteile, Bremsbeläge, Reifenverschleiß, Asphalt- und Fahrbahnverschleiß und Schmieröle
Ni	Automobil-Emission, Schmieröl, Korrosion von Karosserieteilen, Bremsbeläge, Asphalt und Fahrbahnverschleiß und Verschleiß von beweglichen Teilen in Motoren, Katalysatoren
Pb	Fahrzeugabgase, Reifenverschleiß, Schmieröle, Fett, Bremsbeläge, Lagerverschleiß, Asphalt- und Fahrbahnverschleiß sowie Verschleiß beweglicher Teile in Motoren
Pt	Katalysatoren
Sb	Bremsbeläge und Verkleidungen
Sr	Bremsbeläge und Verkleidungen
Ti	Bremsbeläge und Verkleidungen
V	Reifenverschleiß, Asphalt und Fahrbahnverschleiß
Zn	Reifenverschleiß, Motoröl, Fett, Bremsbeläge, Asphalt- und Fahrbahnverschleiß und Schmieröle
PAK	Reifenverschleiß, Fahrzeugabgase, Asphaltabnutzung/Straßenalterung
MKW	Motorölaustritt, Fahrzeugabgase, Kraftstoffe und Frostschutzmittel, Verflüchtigungsverlust

PAK polyzyklische aromatische Kohlenwasserstoffe, *MKW* Mineralölkohlenwasserstoffe

Helmreich et al. 2010; Sansalone und Buchberger 1997). Die Belastung hängt im Wesentlichen von verschiedenen Parametern wie Witterungsbedingungen (Dauer von Trockenperioden und Niederschlagsintensität), Verkehrsfrequenz oder der jahresdurchschnittlichen täglichen Verkehrsbelastung (JDTV) und der Art der Nutzung (z. B. „stop-and go“ im Kreuzungsbereich, Beschleunigungsspur, Parkfläche) ab. Diese unerwünschten Niederschlagsbestandteile stammen bzw. entstehen aus Abgasnebenprodukten, Reifen-, Karosserie- und Fahrbahnverschleiß, Abflüssen aus Niederschlägen, nasser und trockener Deposition und Fahrbahninstandhaltungsarbeiten.

Die Spurenstoffe, die durch den Kraftfahrzeugverkehr in den Gewässerkreislauf eingetragen werden können, sind in Tab. 1 zusammengefasst.

3 Konzentrationen verschiedener Kontaminationen

Die Konzentrationen von Verunreinigungen wie ungelöste Stoffe, Schwermetalle und organische Verbindungen (z. B. MKW und PAK) in Niederschlagsabflüssen von hochrangigen Straßen können so hoch sein, dass diese als verschmutzt eingestuft werden und vor der Einleitung in Gewässer oder Grundwasser zu behandeln sind.

Tab. 2 gibt einen Auszug aus Literaturdaten zu Schwermetallen, MKW und PAK in Verkehrsflächenabflüssen wieder. In Tab. 2 ist erkennbar, dass die Schwermetallkonzentrationen auch innerhalb vergleichbarer JDTV-Belastungen sehr unterschiedlich sind und z. B. für Cu im Bereich 25 bis 682 µg/l schwanken. Tendenziell sieht man eine geringere Konzentration bei kleineren JDTV. Für Pb liegen die älteren Werte im oberen Konzentrationsbereich >50 µg/l. In der Zusammenfassung von Huber et al. (2016), die nur Werte nach dem Jahr 2000 gelistet haben, liegt die mittlere Pb-Konzentration bei 13 µg/l.

Box-Whisker-Plots von der Konzentrationsmatrix aus Tab. 2 ist in Abb. 1 dargestellt. Die Ergebnisse zeigten dass der Verkehrsfrequenz (JDTV) keinen größtmöglichen Einfluss auf die Konzentrationen der Schadstoffe hatte. Die höchsten Gesamtgehalte an Cu, Ni und Zn wurden in Autobahnabflüssen an der A23 in Wien, Österreich gemessen (Fürhacker et al. 2013). Sehr hohe Gesamt-Pb-Konzentrationen (200 bis 250 µg/l) wurden in Autobahnabflüssen an der A23 in Wien, Österreich (Fürhacker et al. 2013), an der A81 (Pleidelsheim) und A6 (Obereisesheim), Deutschland (Stotz 1987), I-94, Minneapolis, USA (Thomson et al. 1997) und 31 Autobahnen in 11 Staaten, USA (Driscoll et al. 1990) gemessen. Die höchsten gesamten 16 EPA-PAK-Konzentrationen in Autobahnabflüssen wurden in absteigender Reihenfolge an der M7 in Monasterevin bypass, Ireland (Desta et al. 2007), an der A23 in Wien, Österreich (Fürhacker et al. 2013), und an der M7 in Kildare, Ireland (Desta et al. 2007) bestimmt (Abb. 1).

Die Spurenstoffkonzentrationen sind nicht nur von den JDTV abhängig. In einigen Studien konnte ein Zusammenhang zwischen Schwermetallbelastung in Autobahnabflüssen und JDTV hergestellt werden (Crabtree et al. 2008; Horstmeyer und Helmreich 2014). Anhand der Schwermetallbelastung von Versickerungsmulden fanden Horstmeyer und Helmreich (2014) einen Zusammenhang zwischen Schwermetallbelastung des Oberbodens und JDTV; über 80.000 JDTV erfolgte keine weitere Zunahme der Kontamination. Die Ergebnisse dieser Literaturstudie zeigen, dass das JDTV keinen signifikanten Einfluss auf die Schadstoffkonzentrationen hat. Ein höheres Verkehrsaufkommen (JDTV >15.000) allein führt somit nicht zu einem Anstieg der Schadstoffkonzentrationen im Straßenabwasser. Auch andere Studien (Herrera 2007; Kayhanian et al. 2003, 2012) ermittelten eine ähnliche Schlussfolgerung.

Die Schadstoffkonzentrationen in Niederschlagsabflüssen von Verkehrsflächen zeigten ortsspezifisch signifikante Unterschiede. Diese Variabilität resultiert nicht nur aus der Verkehrsdichte, sondern auch aus den Unterschieden in der Landnutzung, des gesamten kumulativen Niederschlags, der vorangegangenen Trockenperioden

Tab. 2 Schwermetalle, PAK und MKW-Straßenablaufkonzentrationen aus der Literatur. (Zusammengefasst von Haile und Fürhacker)

Land	Ort	JDTV	Untersuchungszeitraum	Probeanzahl	Cr	Cu	Ni	Pb	Zn	PAK	MKW	Literatur
					µg/l	µg/l	µg/l	µg/l	µg/l	mg/l	mg/l	
AUT	Autobahn A23, Wien	255.000	23.03.2011	1	72	682	69	112	2560	18	3	Fürhacker et al. (2013)
AUT	Autobahn A23, Wien	255.000	11.07.2013	1	11	131	9,8	195	633	–	–	Fürhacker et al. (2013)
AUT	Autobahn A21, Hinterbrühl	42.000	12.2005–05.2008	10	–	205	–	–	360	3,01	2	Fuerhacker et al. (2011)
AUT	Autobahn, Mönchsgraben	60.530	–	–	78	145	29	22	520	–	–	Höfler et al. (2004)
AUT	Autobahn A1, Schwarzenbergkaserne, Salz	>60.000	12.2001–04.2003	29	<1	41	2	8	110	–	0,036	Geiger-Kaiser und Jäger (2005)
AUT	Autobahn A1, Baulos West, Salzburg	>60.000	12.2001–04.2003	29	3	43	3	7	88	–	0,194	Geiger-Kaiser und Jäger (2005)
AUT	Autobahn A1, Baulos Ost, Salzburg	>60.000	12.2001–04.2003	29	6	59	3	11	260	–	0,205	Geiger-Kaiser und Jäger (2005)
ATU	Landesstraße L202 Hard-Bregenz	26.000	01.2005–06.2006	9	14	61	11	9,8	204	–	0,7	Scheffknecht und Prodinger (2007)
ATU	Pilotuntersuchung	26.000–60.500	–	9	10	67	4,6	14	302	–	2,06	Clara et al. (2014)
DEU	Landshuter Alle, München	57.000	11.2003–11.2005	57	–	191	43	56	847	–	–	Helmreich et al. (2010)
DEU	Landshuter Alle, München	57.000	11.2003–11.2005	350	–	194	–	37	933	–	–	Hilliges et al. (2013)
DEU	A3, Köln-Ost	156.000	11.2006–08.2007	20	3	32	2,4	4,9	102	0,15	0,14	Grotehusmann und Kasting (2009)
DEU	A113, Berlin	140.000	11.2006–08.2007	20	5,9	26	2,5	4,9	115	0,28	0,04	Grotehusmann und Kasting (2009)
DEU	Autobahn mit unterschiedlicher DTV	52.000–107.600	20 Woche (1997)	20	–	140	–	17	1250	–	–	Dierkes und Geiger (1999)
DEU	A 555 Widdig	69.368	2005–2006	65	5	27	10	25	106	–	–	Kocher et al. (2010)
DEU	A 61 Meckenheim	73.310	2005–2006	63	5	25	10	25	260	–	–	Kocher et al. (2010)
DEU	A4 Bensberg	71.220	2005–2006	76	5	27	11	25	355	–	–	Kocher et al. (2010)
DEU	Hauptstraße, Literaturstudie	>15.000	–	–	11	97	11	170	407	1,65	4,17	Göbel et al. (2007)
DEU	Autobahn, Literaturstudie	>30.000	–	–	11	36	15	13	217	–	–	Huber et al. (2016)
DEU	Pleidelsheim A81	41.000	–	–	9,6	97	–	200	360	–	7,02	Stotz (1987)
DEU	Obereisesheim A6	47.000	–	–	20	117	–	250	620	–	5,51	Stotz (1987)
DEU	Ulm-West A8	52.100	–	–	5,2	58	–	160	320	–	2,05	Stotz (1987)
DEU	Halenreie, Hamburg	15.000	08.2008–07.2010	8	–	130	–	–	400	–	–	Dobner und Holthuis (2011)
DEU	Berlin	15.000–20.000	–	10	–	127	29	80	500	–	–	Schütte (1997)
CH	N1, Winterthur, Zürich	25.300–73.700	08.1996–11.2000	>10	–	57	–	26	354	–	–	Furumai et al. (2002)
CH	SABA Attinghausen	21.000	03.2007–12.2008	17	13	60	6	9	346	2,7	–	Steiner et al. (2008)
CH	SABA Burgdorf	>17.000	08.2002–11.2004	–	10	57	7	23	299	2,6	1,46	Langbein et al. (2006)
CH	A1 Mattstetten, Bern	60.000	2006–2007	–	12	127	7	19	381	2,3	–	Scheiwiller et al. (2008)
CH	SABA Burgdorf	>17.000	01.2003–02.2004	>20	16	66	7	24	436	–	–	Steiner et al. (2006)
CH	Basel-Landschaft, Birsfelden, N2, Hagnau	>120.000	2008–2012	65	–	57	–	–	188	–	–	Zbinden et al. (2015)
CH	Stark befahrenen Straßen, Literaturstudie	>15.000	–	–	8,9	34	11	15	118	–	0,293	Hürlimann (2011)

Tab. 2 Schwermetalle, PAK und MKW-Straßenablaufkonzentrationen aus der Literatur. (Zusammengefasst von Haile und Fürhacker) (Fortsetzung)

Land	Ort	JDTV	Untersuchungs-zeitraum	Probe-anzahl	Cr	Cu	Ni	Pb	Zn	PAK	MKW	Literatur
					µg/l	µg/l	µg/l	µg/l	µg/l	mg/l	mg/l	
USA	Los Angeles, USA	260.000–328.000	2002–2003	62	10	93	20	33	506	0,4	–	Lau et al. (2009)
USA	Non-urban highways (16), Caifornia	2100–29.000	2002–2003	635	6,5	12	11	17	75,9	–	–	Kayhanian et al. (2007)
USA	Urban highways (8), Caifornia	30.000–100.000	2002–2003	635	6,4	27	7,8	24	134	–	–	Kayhanian et al. (2007)
USA	Urban highways (10), Caifornia	100.000–328.000	–	635	12	50	13	75	261	–	–	Kayhanian et al. (2007)
USA	Ohio, USA	150.000	04.1995–11.1995	5	21	135	43	64	470	–	–	Sansalone und Buchberger (1997)
USA	Austin, Texas area USA	58.150	09.1993–05.1995	–	–	37	25	53	222	–	–	Barrett et al. (1998)
USA	31 Autobahnen in 11 Staaten	>30.000	–	–	–	54	–	234	368	–	–	Driscoll et al. (1990)
USA	I-94, Minneapolis	114.000	–	136	13	47	10	207	174	–	–	Thomson et al. (1997)
UK	M4, Brinkworth Brook	71.930	12.1997–12.1998	10	ND	24	ND	ND	101	–	–	Moy et al. (2003)
UK	A417, River Frome	23.650	06.1998–07.1999	10	12	55	12	51	222	–	–	Moy et al. (2003)
UK	M4, River Ray	36.110	12.1998–03.2000	10	9,1	68	6,7	51	220	–	–	Moy et al. (2003)
UK	M40, Souldern Brook	83.580	08.1999–11.2000	10	7,7	32	4	17	98	–	–	Moy et al. (2003)
UK	A34, Gallos Brook	64.950	09.2000–03.2002	10	4,8	43	4,5	15	149	–	–	Moy et al. (2003)
UK	A34, Newbury	37.190	05.2001–06.2002	10	2,7	24	4,7	4,4	52,5	1,73	–	Moy et al. (2003)
UK	4 Klimaregionen (6 Autobahn/Region)	15.000–>120.000	06.2004–12.2006	240	–	91	–	–	353	7,5	–	Crabtree et al. (2008)
IRL	Motorway, Kildare/Portlaoise	32.000	08.2005–10.2005	6	–	46	–	67	181	–	–	Gill et al. (2017)
IRL	Kildare	25.760	08.2003–12.2005	42	–	120	–	140	660	11	–	Desta et al. (2007)
IRL	Maynooth	29.140	08.2003–12.2005	42	–	–	–	–	70	2,06	–	Desta et al. (2007)
IRL	Monasterevin bypass	18.430	08.2003–12.2005	42	–	40	–	–	150	21,8	–	Desta et al. (2007)
NL	Motorway A1, Laren	–	01.2003–09.2004	80	4,1	117	3,8	29	290	2,36	–	Tromp et al. (2012)
NL	A7, Amsterdam (impervious asphalt)	53.000	07.1994–09.1195	3–6	5	121	5	93	452	5,2	4	Berbee et al. (1999)
NL	A9, Amsterdam (pervious asphalt)	83.00	07.1994–09.1195	3–6	1	40	1	7	47	0,3	<0,1	Berbee et al. (1999)
JP	Autobahn, Osaka	75.000	08.1997–11.1997	4	6,5	66	5,5	34	648	1,28	–	Shinya et al. (2000)
JP	Autobahn, Osaka	62.000	05.1999–08.2000	8	–	68	–	31	713	0,69	–	Shinya et al. (2003)
AUS	Parramatta Road, Sydney	84.500	08.2007–04.2008	8	–	105	–	47	348	–	–	Davis and Birch (2010)
Mittelwert von allen Messungen					*13,1*	*86*	*13,5*	*59*	*383*	*4,8*	*1,9*	–
Mittelwert ohne Extremwerte					*8,8*	*76*	*11,1*	*40,8*	*279*	*2,1*	*1,5*	–

und der maximalen stündlichen Niederschlagsintensität. Atmosphärische Ablagerungen, Probenahmestrategien und Probeanzahl pro Regenereignis sind weitere mögliche potenzielle Einflussfaktoren (Crabtree et al. 2008; Helmreich et al. 2010; Kayhanian et al. 2012). So konnte mit den verfügbaren Literaturdaten kein zusätzlicher statistischer Zusammenhang in Hinblick auf JDTV allein ermittelt werden.

Spurenstoffe in Niederschlagsabflüssen von Verkehrsflächen sind sowohl in gelöster als auch in partikulärer Phase präsent. Die Verteilung der Schwermetalle zwischen der gelösten und der partikulären Phase ist besonders für die Beurteilung der Toxizität des Niederschlagsabflusses sowie für die Entwicklung oder Auswahl von Behandlungssystemen entscheidend (Furumai et al. 2002; Haile et al. 2016; Helmreich et al. 2010; Kayhanian et al. 2012). Trotz einiger Inkonsistenzen in den Literaturdaten zeigen die vorhandenen Monitoringdaten, dass die größten Fraktionen an Schwermetallen (Cr, Cu, Ni, Pb & Zn) und PAK in Verkehrsabflüssen in der Regel überwiegend in der partikulären Phase liegen. Somit liegen die partikulären Anteile für die Schwermetalle (Cr, Cu, Ni und Zn) bei jeweils über 50 % und für Pb bei über

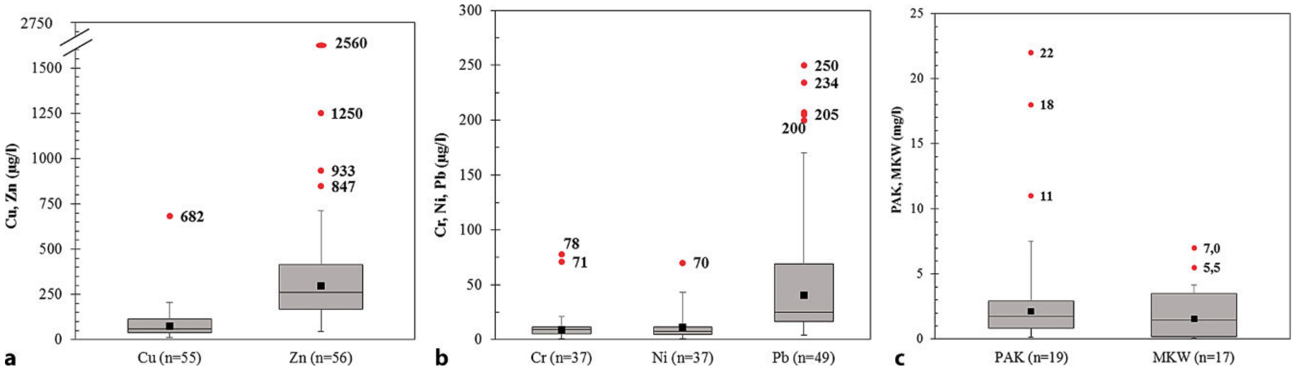

Abb. 1 Schwermetall-, PAK- und MKW-Konzentrationen (gesamt) in Niederschlagsabflüssen von stark befahrenen Straßen mit JDTV >15.000 Fahrzeuge pro Tag (Zusammenstellung der Tab. 1). *Box-Whisker-Plots*: oberes bzw. unteres Ende der Box entspricht dem oberen bzw. unteren Quartil, der Strich in der Box dem Median und das kleine Quadrat (*schwarz*) in der Box der mittleren Konzentration; die kleinen Kreise (*rot*) stellen Extremwerte dar

Tab. 3 Überblick über die Prüfmethoden und Kriterien für Niederschlagsbehandlungssysteme für den Abfluss von Verkehrsflächen

Literatur	Land	Prüfmethode	Prüfsubstanz**	Einleitung in
ÖNORM B 2506-3 (2016)	AUT	Labor	AFS, Cu, Zn, NaCl, MKW	Grundwasser
ÖNORM B 2506-2 (2012)	AUT	Keine Angabe	AFS, Cu, Zn, MKW	Grundwasser
ASTRA (2016a, 2016b)	CH	Anlage	AFS, Cu, Zn	Keine Angabe
ASTRA (2010)	CH	Anlage	AFS, Cu, Zn, PAK, DOC	Einleitung in die Gewässer
Schmidt et al. (2015)	CH	Labor & Anlage	Cu, Zn, Pestizide (z. B. Mecoprop Diuron)	Keine Angabe
DIBt (2011, 2012)	DEU	Labor & Pilotaanlage	AFS, Cu, Zn, NaCl, MKW	Grundwasser
LANUV (2014)	DEU	Labor oder Anlage	AFS, Cu, Zn, NaCl, MKW	Oberflächengewässer
NJDEP (2009)	USA, NJ	Anlage	AFS	Keine Angabe
NJDEP (2013; 2017)	USA, NJ	Labor	AFS	Keine Angabe
Sample et al. (2012)	USA, VA	Labor oder Anlage	AFS, P	Keine Angabe
WDOE (2011)	USA, WA	Labor & Anlage	AFS, Cu, Zn, MKW, P	Grundwasser- und Oberflächengewässer
Boogaard (2015)	NL	Labor & Anlage	AFS	Grundwasser- und Oberflächengewässer
Victorian Stormwater Committee (2006)	AUS	Keine Angabe	AFS, gelösten Stoffe, TN, TP	Keine Angabe
ARC (2003)	NZL	Anlage	AFS	Keine Angabe
Fassman (2012)	NZL	Anlage	AFS, Cu, Zn	Meerwasser
Monrabal-Martinez et al. (2017)	NOR	Labor	Cu, Ni, Pb, Zn	Mäßig verschmutzte Oberflächengewässer

** AFS und MKW werden nach Methode individuell definiert

80 % (Ball et al. 1998; Helmreich et al. 2010; Huber et al. 2016; Kayhanian et al. 2012; Sansalone und Buchberger 1997).

4 Überblick über bestehende Prüfmethoden

Überblick über die internationale Behandlung von Niederschlagsabflüssen von Verkehrsflächen, Prüfmethoden und vorgeschlagene Prüfsubstanzen (Tab. 3).

5 Prüfverfahren nach ÖNORM B 2506-3

In Österreich wurde eine Prüfmethode entwickelt, um die Eignung von technischen Filtermaterialien für die Regenwasser-Sickeranlagen für Abläufe von Dachflächen und befestigten Flächen zu überprüfen (ÖNORM B 2506-3 2016). Die Laborprüfmethode wird verwendet, um die Entfernung von Partikeln (abfiltrierbare Stoffe AFS), gelösten Schwermetallen (Blei, Kupfer und Zink) und Mineralölkohlenwasserstoffen zu beschreiben und die Remobilisierung der Schwermetalle durch Streusalz (NaCl) zu prüfen. Zusätzlich werden auch die Durchlässigkeit des Filtermaterials und seine Veränderung durch Partikelzugabe überprüft.

Die Prüfmethode besteht aus acht Teilprüfungen, die in der angegebenen Reihenfolge durchgeführt werden:

1. Infiltrationsrate und Suffusionsstabilitätstest
2. Partikelretention I
3. Schwermetallrückhalt (Cu, Pb und Zn)
   3.1 Versuche im Überstaubetrieb
   3.2 Kapazitätsprüfung mit 4 Jahresfracht
4. Mineralölrückhalt
5. Partikelretention II
6. Bestimmung der Änderung der Infiltrationsrate und der Remobilisierung der AFS
7. Remobilisierung von Schwermetallen durch Beschickung mit 5 g/L NaCl
8. Säureneutralisationskapazität

Tab. 4 Ausgewählte Konzentrationen der Niederschlagswässer von befestigten Flächen für die Berechnung der Jahresfrachten

Parameter	AFS (mg/L)	Pb (µg/L)	Cu (µg/L)	Zn (µg/L)	Mineralöl (mg/L)	NaCl (mg/L)
Prüfung im Überstaubetrieb	90	50	100	400	5,0	–
Kapazitätsprüfung	–	200	400	1600	–	–
Remobilisierung von Schwermetallen	–	–	–	–	–	5000

Bei der Prüfung wird das Filtermaterial mit vier Jahresfrachten für Schwermetalle bzw. AFS und einer Jahresfracht Mineralöl beschickt. Zur Berechnung der Jahresfrachten wurde eine mittlere Belastung des Niederschlagsabflusses von befestigten Flächen mit Partikeln, Schwermetallen und Mineralöl der Literatur entnommen und die in Tab. 4 angegebenen Konzentrationen ausgewählt.

Die Prüfung gilt als bestanden, wenn ein definierter Mindestrückhalt bzw. eine definierte Konzentration eingehalten wird.

Die Prüfungen werden für unterschiedliche hydraulische Belastungen (Flächenverhältnisse (As: Ared) von 1:15 bis 1:250) definiert. Bei der Prüfung von Anlagen mit Flächenverhältnissen (As: Ared) größer 1:100 ist eine Absetzanlage erforderlich, die zumindest 50 % der Partikel entfernt.

6 Anwendung der ÖNORM B 2506-3 für die Eignungsprüfung von Substraten zur Reinigung von Niederschlagsabflüssen vom hochrangigen Straßennetz

Da die ÖNORM B 2506-3 als Grundlage für die ÖNORM B 2506-1 und -2 und für das ÖWAV-Regelblatt 45 erstellt wurde und der Geltungsbereich dieser ÖNORM Einzugsflächen für die Versickerung von Abflüssen von Dachflächen, befestigten Bodenflächen, wie z. B. Höfen, Zufahrten, Gehwegen, Terrassen, Pkw-Abstellflächen, Lager- und Ladeflächen sowie Verkehrsflächen bis zu einer Belastung von 5000 JDTV (durchschnittliche tägliche Verkehrsstärke), nicht aber den Abfluss von übergeordneten Verkehrsflächen wie z. B. Autobahnen oder Hauptverkehrsstraßen umfasst, und auch das ÖWAV-Regelblatt 45 für JDTV >15.000 Kfz/24 h auf die RVS verweist, stellt sich die Frage, ob die ÖNORM B 2506-3 für die Versickerung von Abwasser von hochrangigen Straßen anwendbar ist.

Dazu ist festzustellen, dass die Schwermetallbelastungen nicht nur von der Verkehrsbelastung abhängen und auch innerhalb vergleichbarer JDTV sehr unterschiedlich sein können (Tab. 2). Die in der Prüfmethode der ÖNORM B 2506-3 verwendeten Schwermetallkonzentrationen und auch die MKW-Konzentrationen sind höher als die mittleren Konzentrationsniveaus von hochrangigen Straßen. Zusätzlich werden die Prüfungen mit gelösten Schwermetallen bei niedrigem pH-Wert (pH 5,5 bzw. 5,8) durchgeführt. Dies bietet eine zusätzliche Sicherheit, da im Straßenabwasser ein erheblicher Teil der Schwermetalle an Partikel gebunden ist und mit diesen entfernt wird. Für die Mineralölprüfung wird Heizöl EL direkt auf die Oberfläche aufgetragen, nach dem Eindringen des Heizöls wird die Säule im Überstaubetrieb beschickt. Für die Prüfung des Partikelrückhalts und der Suffusionsneigung der Materialien werden sehr feine Quarzpartikel (>50 Gew-% <63 µm) verwendet, die weder Mineralöle noch Schwermetalle adsorbieren. All dies stellt eine Verschärfung der Konditionen gegenüber der Praxis dar (Haile und Fürhacker 2015; Haile et al. 2016).

Aus den in der ÖNORM B 2506-3 gewählten Prüfbedingungen und Prüfkriterien für die Prüfung der technischen Filtermaterialien nach ÖNORM B 2506-2 und ÖWAV-RB 45 ergibt sich, dass die erfolgreich geprüften Materialien aus wissenschaftlicher Sicht geeignet sind, auch die Anforderungen an die Reinigung von Straßenabwässern von hochbelasteten Straßen mit hohen JDTV, z. B. des hochrangigen Straßennetzes, zu erfüllen. Es sei noch darauf hingewiesen, dass die Prüfung der technischen Filtermaterialien nach ÖNORM B 2506-3 für die Versickerung in das Grundwasser erstellt wurde.

Erfahrungen aus durchgeführten Projekten am Institut für Siedlungswasserbau der Universität für Bodenkultur Wien zeigen, dass technische Filtermaterialien eine gute Reinigungsleistung für die Behandlung von Spurenstoffen von Straßenabwässern aufweisen.

7 Schlussfolgerungen

Auch wenn die ÖNORM B 2506-3 als Grundlage für die ÖNORM B 2506-1 und -2 und für das ÖWAV-Regelblatt 45 (Anwendungsbereiche JDTV <5000 bzw. <15.000 Kfz/24 h) erstellt wurde, ergibt sich, dass aufgrund der in der ÖNORM B 2506-3 gewählten Prüfbedingungen und Prüfkriterien aus wissenschaftlicher Sicht auch die Anforderungen der Reinigung von Straßenabwässer von hochbelasteten Straßen mit hohen JDTV, wie jener des hochrangigen Straßennetzes, erfüllt werden.

Acknowledgements Open access funding provided by University of Natural Resources and Life Sciences Vienna (BOKU).

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6.6 Paper VI

Simultaneous adsorption of heavy metals from roadway stormwater runoff using different filter media in column studies. Manuscript.

Haile, T.M. and Fuerhacker, M. (2018). Manuscript submitted to *Water* (ISSN 2073-4441).

Simultaneous adsorption of heavy metals from roadway stormwater runoff using different filter media in column studies

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Abstract

Stormwater runoff from roadways often contains a variety of contaminants such as heavy metals that can adversely impact receiving waters. The filter media in stormwater filtration/infiltration systems plays a significant role for the simultaneous removal of multiple pollutants. In this study, the capacity of five filter media –natural quartz sand (QS), sandy soil (SS) and three mineral-based technical filter media (TF-I, TF-II and TF-III)– to adsorb heavy metals (Cu, Pb and Zn) frequently detected in stormwater as well as remobilization due to de-icing salt (NaCl) were evaluated in column experiments. The column breakthrough data were used to predict lifespan of the filter media. Column experiment operated under high hydraulic load showed that all technical filters and sandy soil achieved >97 %, 94% and > 80% of Pb, Cu and Zn load removals, respectively, while natural quartz sand (QS) showed very poor performance. Furthermore, treatment of synthetic stormwater by the soil and technical filter media met the requirements of the Austrian regulation regarding maximum effluent concentrations and minimum removal efficiencies for groundwater protection. The results showed that application of NaCl had only a minor impact on the remobilization of heavy metals from the soil and technical filter media, while the largest release of metals was observed from the QS column. Breakthrough analysis indicated that load removal efficiencies at column exhaustion (SS, TF-I, TF-II and TF-III) were > 95 % for Cu and Pb, and 80– 97% for Zn. Based on the adsorption capacities, filtration systems could be sized to 0.4% to 1% (TF-I, TF-II and TF-III) and 3.5% (SS) of their impervious catchment area and predicated lifespan of each filter media was at least 35, 36, 41 and 29 years for SS, TF-I, TF-II and TF-III, respectively. The findings of this study demonstrate that soil– based and technical filter media are effective in removing heavy metals and can be utilized in full- stormwater filtration systems.

Keywords: Stormwater treatment, Filter media, Heavy metals, Adsorption, De-icing salt, Lifespan.

1. Introduction

Stormwater runoff from vehicle trafficked areas and roofs contains a heterogeneous mixture of pollutants including solids, heavy metals, organic pollutants such as polycyclic aromatic hydrocarbons (PAHs) and mineral oil hydrocarbons (MOH), nutrients, and compounds of de-icing salts that can cause significant hydrological and ecological impacts on receiving waters (Göbel et al., 2007; Hatt et al. 2008; Helmreich et al., 2010). Heavy metals such as cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn) are the most frequently reported pollutants in roadway and parking lot runoffs mainly emitted from vehicles and traffic-related activities (Genc-Fuhrman et al., 2007; Hatt et al., 2011; Huber et al., 2016b; Helmreich et al., 2010). Heavy metals are mobile in natural water ecosystems, non-degradable, and potentially toxic as they can accumulate in the environment causing both short term and long term adverse effects (Marsalek et al., 1999; Wium-Andersen et al., 2011). Dorchin and Shanas (2010) used biological assays and confirmed that road runoff is a major cause of deterioration of aquatic habitat quality in Israel. Furthermore, various studies noted that roadway runoff are likely to induce mutagenic/genotoxic effects due to the combined effects of heavy metals and PAHs (Haile et al., 2016a; Shinya et al., 2000). Consequently, treatment of stormwater has become increasingly important to mitigate its negative ecological effects.

Stormwater infiltration/filtration system that utilize granular adsorptive filter media are receiving increasing interest due to their ability to remove both dissolved and particulate pollutants (Hilliges et al., 2013; Pawluk and Fronczyk, 2015; Reddy et al., 2014). Filter media with high infiltration rates and effective treatment performance are the key design aspect that needs to be considered for the application of filter systems. These treatment technologies could be retrofitted in a confined urban environment or roadways in complex topography where space is limited, where a greater volume of stormwater could be treated in a compact system. The removal of pollutants is achieved via a number of processes including sedimentation, filtration, sorption, ion exchange, surface complexation and transformation (Fuerhacker et al., 2011; Hatt el al., 2008; Pitcher et al., 2004; Scholes et al., 2008).

Studies both under laboratory (Hatt et al., 2008, 2011; Hsieh and Davis, 2005; Huber et al., 2016b; Reddy et al., 2014; Thomas et al., 2014) and field conditions (Fuerhacker et al., 2011; Haile et al., 2016b; Helmreich and Horn, 2010; Hilliges et al. 2013) have proven that adsorptive filter media are effective at capturing both dissolved and particulate stormwater pollutants. For example, Thomas et al. (2014) tested the performance of mixed filter media composed of crushed aggregate and three active ingredients: perlite, dolomite and gypsum in column experiment using synthetic stormwater reported over 90% removal efficiencies of copper and zinc. Bioretention systems with media mixes (sand, soil and mulch) achieved over 96% removal efficiency of oil/grease, suspended solids and Pb (Hsieh and Davis, 2005). Reddy et al. (2014) evaluated the efficiency of mixed media consisted of calcite, zeolite, sand, and iron filings observed that over 90% (Cd, Cr, Cu, Pb and Zn) and 75 – 88 % nutrients were removed from synthetic stormwater. Soil-based filters are efficient for the removal of solids, Cu, Ni, Pb, Zn and PAHs (Hatt et al., 2011; Lim et al., 2015). Overall these studies showed that soil-based and a mixed-media based decentralised stormwater infiltration/filtration system are efficient to treat a wide range of pollutants. Unfortunately, results regarding pollutant removal efficiencies, equilibrium/effluent concentrations and sorption capacities were highly variable among the studies as well as these results may not be comparable to field conditions. These variabilities could be related to many factors including single solute solution versus multi-solute solution, influent concentration, pH, flow rate, flow direction (i.e. upflow vs downflow mode), and filter bed height (Anthandious, 2005; Lim et al., 2015; Nguyen et al., 2015; Wang et al., 2017). Column sorption experiments were mainly conducted with metal concentrations much higher than the levels in real roadway runoff waters (Huber et al., 2016; Monrabal-Martinez et al., 2017; Pawluk and Fronczyk, 2015). Furthermore, as heavy metals often coexist in stormwater there is competition among various metals for adsorption sites, therefore it is important to consider the simultaneous removal of co-existing metals (Nguyen et al., 2015). The candidate filter media should be able to bind and adsorb multiple metals of significantly varying concentrations. In this context the experimental boundary conditions such as influent concentration and pH are very crucial in assessing the suitability and capacity of filter media for application in stormwater filtration systems.

Metals adsorbed to the filter media are not permanently immobilized. Application of de-icing road salts in winter periods may interfere with operation of stormwater treatment facilities, for example release of chemicals (Bäckström et al., 2004; Norrström; 2005). In column study, Norrström (2005) demonstrated that a large part of Pb, Cd and Zn in highway roadside soils are vulnerable to leaching when exposed to a high NaCl (5.84 g/L) concentration. From field studies, Bauske and Goetz (1993) also found a strong effect of NaCl and $CaCl_2$ (1300 mg Na^+, 2700 mg $C1^-$ and 500 mg Ca^{2+} per liter) solution on Cd and Zn release in winter and spring, where cation exchange and the formation of chlorocomplexes are discussed as the responsible mechanisms. Increased mobility of heavy metals coincident with road salt applications have been observed in road side soils by various mechanisms including ion exchange, lowered pH, formation of chlorocomplexes and possible colloid dispersion (Bäckström et al., 2004; Bauske and Goetz 1993; Norrström, 2005). Amrhein (1992) demonstrated that the metals such as Cu, Ni, Cr, Pb, and Fe may be mobilized from roadside soils as a result of road salt (NaCl) application either by direct cation exchange or by mobilization of metal bearing constituents such as organic matter or clays. Bäckström et al. (2004) also demonstrated that the major mobilisation mechanisms of heavy metals (Cd, Cu, Pb and Zn) by deicing salts from roadside soils are ion exchange, lowered pH, chloride complex formation and possible colloid dispersion. Additionally, studies have been conducted to examine the remobilization of heavy metals adsorbed onto filter materials used for stormwater treatment (Huber et al., 2016b; Monrabal-Martinez et al., 2017). In laboratory column experiment, Huber et al. (2016) have shown that pure NaCl (10 g/L) had a minor effect on the remobilization heavy metals. Monrabal-Martinez et al. (2017) also demonstrated that the addition of NaCl (1 g/L) did not show any adverse impact on the release of already adsorbed metals. While Recently, NaCl solution has been used to investigate the remobilization of previously adsorbed heavy metals, which is a crucial test criteria for the certification of filter media in Austria (ÖNORM B 2506-3, 2016) or filtration systems in Germany (DIBt, 2011). Consequently, the simultaneous removal of multiple heavy metals as well as the effect of de-icing salt on the mobilization of adsorbed heavy metals at experimental conditions similar to real roadway runoff should be thoroughly evaluated and well addressed.

The objectives of this study were: firstly to determine the influence of hydraulic loading rate on simultaneously removal of Cu, Pb, and Zn from synthetic stormwater using five different filter media in column sorption experiments; secondly to investigate the impact of de-icing salt (NaCl) on the remobilization of adsorbed heavy metal; and finally long-term performance of each filter media was investigated using column breakthrough curve. To mimic heavy metal adsorption capacity at natural environmental conditions, column sorption studies were conducted with experimental conditions more close to real life of stormwater quality and treatment systems. The column study results were used to predict filter media lifespan based on effluent quality and removal efficiencies.

2. Materials and methods

2.1 Chemicals and analytical instruments

All chemical used were analytical reagent grade (Merck KGaA, Darmstadt, Germany). Synthetic stormwater solutions containing Cu, Pb and Zn were prepared using analytical grade 1000 mg/L stock solutions (Titrisol®) of $CuCl_2$, $Pb(NO_3)_2$ and $ZnCl_2$, respectively and mixed with de-ionised water to obtain the desired concentrations. The initial pH of the test solutions was adjusted to the desired value by using dilute solutions of 0.1 M NaOH and 65% HNO_3. Conservation of samples was performed using 1% volume of 65% HNO_3.

2.2 Filter media

The performance of natural commercial available quartz sand (QS) without pretreatment, sandy soil (SS), and three mineral-based technical filter media (TF-I, TF-II and TF-III) to remove heavy metals (Cu, Pb and Zn) from synthetic stormwater runoff has been investigated through column tests. The sand soil was excavated from a newly constructed highway runoff infiltration basin and the coarse gravel fraction (dimeter over 2 mm) was removed manually. There exist numerous adsorbents of different nature and they can be utilized as mixed-media filter systems. According to the ÖNORM B 2506–3 (2016) mineral-based mixture of adsorptive materials are defined as technical filter media which is here denoted as "TF". Studies showed that a combination of several filter media (for example zeolites, vermiculite, activated carbon, dolomite, sand and soil) are necessary to achieve effective simultaneous removal of multiple contaminants (Hatt et al., 2008, 2011; Pawluk and Fronczyk, 2014; Reddy et al., 2014). The technical filter media (TF-I, TF-II and TF-III) investigated in this study are combinations of various sorbents such as zeolite, vermiculite, dolomite, activated carbon, coconut fiber, expanded clay and soil media. All tested filter media were investigated without any physico-chemical treatment or modification. Physical characteristics and composition of the filter media are summarised in Table 1.

Table 1. Composition and physicochemical properties of filter media used in the study.

Filter media	k_{sat} *(m/s)*	pH (-)	U (-)	PSD (mm)	Porosity (%)	composition
QS	3.7×10^{-3}	6.7	0.95	0.8 (0.71–1.25)	35	Natural quartz sand
SS	4.1×10^{-4}	8.4	14.3	3.1 (0–11.2)	38	Sandy soil + gravel
TF-I	1.8×10^{-3}	8.1	2.3	2.3 (0.5–5.6)	43	Zeolite + vermiculite + crushed concrete
TF-II	1.5×10^{-3}	8.8	2.1	2.3 (0.5–5.6)	39	Zeolite + vermiculite + crushed concrete + dolomite
TF-III	1.4×10^{-3}	8.6	6.7	1.1 (0.02–4.0)	41	Zeolite + coconut fiber + expanded clay+dolomite

PSD= Particle size distribution – d_{50} and range; U) uniformity coefficient (d60/d10)

2.3 Experimental design

The column experiment was carried out using two different sized columns with inner diameters of 32 and 100 mm, respectively. The aim of the 100-mm column experiment was to study the efficiency of metal removal efficiencies under high hydraulic loading rates. Subsequently, the effect of de-icing road salt on the mobilisation of already retained metals was studied by flushing each filter column with sodium chloride (NaCl) solution. In the second set, continuous adsorption experiments were conducted using 32 mm columns to investigate the long-term capacity of the filter media to remove metals and to predict its effective lifespan.

2.3.1 High hydraulic loading conditions

High hydraulic loading may lead to reduced stormwater retention times and could reduce treatment efficiencies. The column test was designed to simulate treatment efficiency of five different filter media at their maximum infiltration rates (saturated hydraulic conductivity, K_{sat}). The experiments were conducted in 800 mm high and 100 mm internal diameter (cross-sectional area of 78.5 cm^2) plexiglass columns and an outlet diameter of 30 mm to allow free flow of water by gravity. The filter media in the column was packed to the desired depth of 300 mm, providing a filter bed volume (BV) of 2.36 L. A drainage layer of 250 mm gravel (4/8 mm) and textile nylon mesh were placed at the bottom of the columns to prevent particle wash out. In order to maintain uniform feed solution distribution and flow rate, 50 mm gravel (4/8 mm) was placed on top of the filter media. The feed solution percolated through the filter columns in downflow mode (from top to bottom) using a precise peristaltic pump (Watson Marlon 520U, UK) dynamically adjusted to a flow rate that resulted in a ponding depth of 50 mm to elucidate peak inflow. The flow rate was 2.1, 0.225, 0.980, 0.820, and 0.770 L/min for QS, SS, TF-I, TF-II and TF-III, respectively. For all technical filter media (TF-I, TF-II and TF-III) and QS the flow rate remained almost constant throughout the experimental period, but for the column packed with sandy soil flow rate slightly reduce over time (0.225 L/min to 0.180 L/min).

The experiments were conducted in five successive runs simulating different stormwater sources and impact of de-icing salt on metal mobility (Table 2). To assess the heavy metal removal efficiency, 84 L of synthetic stormwater was percolated per column per experimental run (Run 1– Run 4), therefore each column received a total stormwater volume of 336 L. After passing this volume of water, the filter columns were allowed to drain for at least 24 hours. Finally, to investigate the impact of de-icing salt on mobilization of retained metals each filter column was flushed with 42 L de-ionised water solution containing 5g/L of NaCl (Run 5). The concentration of NaCl was based on common concentrations found in urban highway runoff in Austria (Fürhacker et al., 2013) and the Austrian Standard Method (ÖNORM B 2506–3, 2016).

Table 2. Influent concentrations of heavy metals (μg/L) and NaCl (g/l) in different experimental runs, and influent pH.

Experiment	Parameters and concentration[1]				pH	Possible source
	Cu	Pb	Zn	NaCl		
Run 1	100	100	100	-	5.8	Roadways (e.g. feeder roads)
Run 2	500	500	500	-	5.8	Urban highway
Run 3	3000	0	500	-	5.5	Cu-roof
Run 4	150	0	5000	-	5.5	Zn-roof
Run 5 [a]				5		

Note[1]: The synthetic stormwater solution used first flush mean concentrations and minimum pH levels reported in the literature (Göbel et al., 2007; Athanasiadis et al., 2005; Huber et al., 2016a); [a]: Influent pH was not adjusted; Run 1- moderately polluted roadway runoff; Run 2- heavily polluted urban highway runoff; Run 3- copper roof runoff; and Run 4- zinc roof runoff.

The influent pH level (Table 2) were selected as the optimum condition, while pH higher than this would cause potential precipitation within the storage tank. Influent water samples were taken at the beginning of every experimental run while effluent samples were collected after every flow through of 28 L from each column (i.e., 3 effluent sample per experimental run per column) and analysed for dissolved concentrations of Cu, Pb and Zn. For the experiments with NaCl solution, one influent sample at the start of the experiment and several effluent samples at designated time intervals were collected in 100 mL glass bottles and preserved with 1% volume of 65% HNO_3. In addition, a mixed sample was collected from total effluent volume of each column. Remobilized metal mass was determined based on the effluent concentrations and effluent volume.

2.3.2 Column breakthrough experiments

Breakthrough curves of Cu, Pb and Zn using five filter media were studied in small-scale plexiglass columns with an inner diameter of 32 mm cm and a length of 300 mm. The filter media was packed to a depth of 200 mm (yielding a bed volume of 160 ml) and used for the continuous flow test. A 20 mm layer of glass beads was placed at the bottom and top of the packed filter column to support the filter media and to ensure uniform flow distribution. The ratio of the inner diameter to mean particle diameter (d_{50}) was at least 10:1 in which wall effect can be negligible (Fanfan et al., 2005). After packing, each column was slowly flushed with approximately 20 bed volumes (BV) of de-ionized water in an upflow mode in order to saturate the filter media and remove the air bubbles entrapped in the sorbent pores to maintain identical experimental conditions.

The column breakthrough experiment was devoted to urban highway runoff, where the target heavy metal concentrations were assumed to be 50 µg/L Pb, 100 µg/L Cu and 400 µg/L Zn at an influent pH of 5.8±0.20 based on a stormwater quality reviews (Göbel et al., 2007; Huber et al., 2016a). The influent solution was prepared in an aquarium tank and pumped in up-flow mode (from bottom to top) using a high precision peristaltic pump (Ismatec IDEX, Laboratoriumstechnik GmbH, Wertheim, Germany). The flow rates were 50% of the flow determined at saturated hydraulic conductivity (K_{sat}) each filter media. Thus, the flow rate was different for each filter media. Firstly, the effect of flow mode on heavy metal removal was examined by conducting column experiment in upflow and downflow mode operated in parallel using TF-II, while maintaining all other experimental conditions constant. Finally, the sorption capacity of all five filter media was evaluated in the upflow mode and their lifespan was predicted using the maximum sorption capacity at filter media exhaustion. The experiments were performed from Monday to Friday, thus during weekends, the filter media were closed and kept saturated without flow in order to maintain similar experimental boundary conditions. The volume of solution kept closed in the filter column over the weekend was insignificant (<< 1%) compared to the total flow though volume. Effluent samples were collected in 100 ml glass bottles from the exit of the column at different intervals, preserved with 1% volume of 65% HNO_3 and analysed for dissolved concentrations of Cu, Pb and Zn.

2.4 Operation criteria

In Austria, purified wastewater should fulfil the criteria of Groundwater Quality Ordinance (QZV) of 9 µg/L Pb and 1800 µg/L Cu (BMFLUW, 2010) and the criterions of the ÖNORM B 2506–3 (2016). Therefore, the operation target of all filter columns was terminated (i.e. filter media exhaustion) when: either Pb concentration in the effluent exceeded 9 µg/L, the Cu removal rate fall below 80%, Zn removal rate fall below 50% or a combination of these criteria.

The expected lifespan (years) of each filter media was determined by dividing the cumulative adsorbed mass of a heavy metal at filter media exhaustion point (section 2.3.2) by the annual load of a heavy metal entering the treatment systems. The annual heavy metal loads entering the treatment systems were calculated under the following assumptions: a filter area of 8.04 cm², a filter media depth of 300 mm, annual precipitation of 700 mm, dissolved runoff concentrations 25 µg/L Pb, 50 µg/L (Cu) and 200 µg/L (Zn) respectively which corresponds to 50% of the total concentration, and size of stormwater treatment system relative to its impervious catchment area. The size of the stormwater infiltration systems relative to its impervious catchment area was estimated from the cumulative heavy metal load retained in the filter column.

2.5 Analytical Procedures

All samples were filtered through 0.45 µm pore size non-sterile Phenex-RC 26 mm syringe filter (Phenomenex LTD, Aschaffenburg, Germany) for analysing dissolved metal concentrations and preserved with 1 % volume of 65% HNO_3 until analysis. Cu, Pb and Zn concentrations were measured using inductively coupled plasma mass spectrometry (ICP-MS) according to DIN EN ISO 17294-2. The detection limits were 1.0, 0.5 and 3.0 µg/L for Cu, Pb and Zn, respectively. For simplicity, effluent concentrations below detection limit were set equal to the detection limit, recognising that this conservative assumption might underestimate the Cu, Pb, and Zn removals by only 1.0%. The pH was measured immediately after sample collection using a glass electrode (WTW pH 197i, Weilheim, Germany) according to DIN EN ISO 10523-C5.

2.6 Data analysis

Metal removal efficiency for a sample taken at time t over the course of the experiment was calculated as follows (1):

$$\boldsymbol{\eta_t}(\%) = \frac{(C_{it} - C_{et})}{C_i} \qquad (1)$$

Where influent and effluent concentrations (µg/L) are denoted as C_{it} and C_{et} respectively, and $\boldsymbol{\eta}$ is metal removal efficiency (%) of a sample taken at time t.

Influent metal load applied to each column till media exhaustion or termination of the experiment, (mg), was calculated as follows (2)

$$\mathbf{q_i} = \frac{\sum(\mathbf{C_i} * \mathbf{V_i})}{\mathbf{1000}} \qquad (2)$$

where C_i is the influent concentration (µg/L) and V_i is influent volume passed through the filter column (L)

Mass of metal adsorbed till filter media exhaustion or termination of the experiment, q_s (mg), was calculated using equation (3):

$$\boldsymbol{q_s} = \frac{\sum(C_i * V_i) - \sum(C_e * V_e)}{\mathbf{1000}} \qquad (3)$$

where C_i and C_e are the influent and effluent concentration (µg/L) and V_i and V_e influent and effluent volumes (L).

Heavy metal sorption capacity (q_e) at column exhaustion per unit dry weight of filter media packed in the column, (mg/g), was calculated using the following equation (4):

$$q_e = \frac{q_s}{M} \qquad (4)$$

where M (g) the total dry weight of filter media packed in the column.

3. Results

3.1 Effluent pH variations during the experiments

Although the pH of the feed multi-metal solution was adjusted to 5.8±0.2 during all column experimental runs, the effluent pH was higher than the influent for all tested filter media (Figure 1). Effluent pH exhibited a general decreasing trend over the course of the experimental period for all columns, decreasing from 8.6 to 7.1 for TF-I, 9.1 to 7.9 for TF-II, 9.1 to 7.8 for TF-III, 9.2 to 7.9 for SS, and 6.7 to 5.6 for QS, respectively. At the end of the experiments the effluent pH of the columns packed with soil based and mineral-based technical filter media remained consistently high ranging from 7.1 to 8.0. However effluent pH of the QS column dropped from 6.7 to 5.6.

The increase in pH suggested that the soil and technical filter media have good pH buffering capacity. The higher effluent pH observed in the technical filter media packed columns was mainly due to calcite (dolomite) additive as an additional amendment in the mixed media. The dissolution of the carbonate phase, impurities present in the filter media, adsorption of hydrogen ions from the solution as well as cationic exchange causes a rapid increment of pH in the solid-water interface (Genc-Fuhrman et al., 2007; Genc-Fuhrman et al., 2008; Pitcher et al., 2004; Reddy et al., 2014). . According to ÖNORM B2506–3 (2016) pH buffering capacity of a filter media tested in column experiments (100 mm inner diameter and 2.36 L bed volume) should achieve a minimum effluent pH of 6.0±0.1 while it is flushed with influent pH of 3.0±0.1 at a flow rate that produces 5 cm ponding level for at least half an hour for filter media with k_{sat} over 2. 5x10^{-3} m/s or a minimum flow through of 42 L when k_{sat} is below 2.5x10^{-3} m/s.. In this regard the investigated technical filter media and sandy soil are suitable for utilization in stormwater filtration/infiltration systems, but QS failed to meet the minimum requirements.

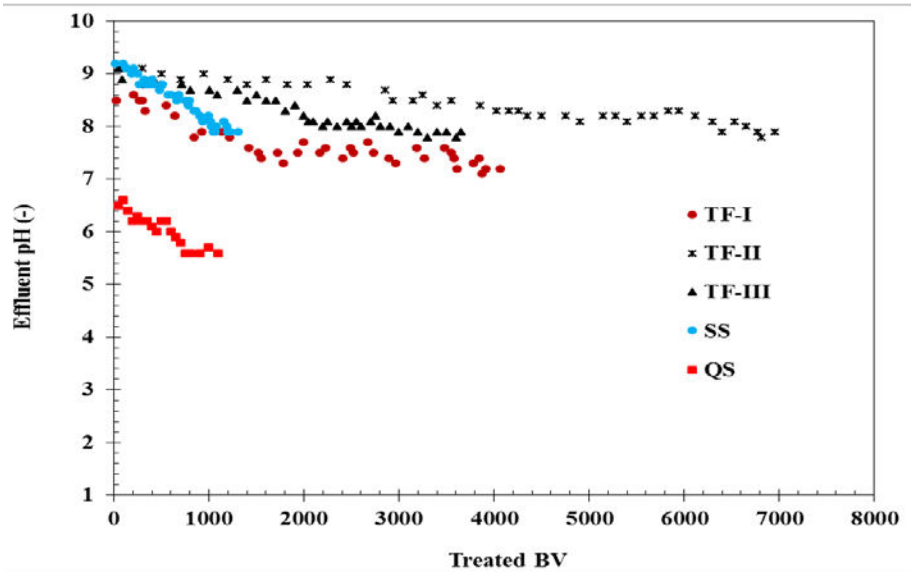

Figure 1. pH drift curves for column experiments conducted at influent pH value of 5.8 ± 0.2.

3.2 Effect of high hydraulic load

The effect of hydraulic load on the adsorption of the selected heavy metals was carried out with synthetic stormwater solutions representing first-flush highway and roof runoff (Table 2). As shown in Figure 2, effluent heavy metal concentration trends suggest that the soil and technical filter media were able to maintain high removal performance for all experimental runs (Run 1 – Run 4). For the soil and technical filter media effluent Pb and Cu concentrations were below the maximum allowed (9 µg/L for Pb and 1800 µg/L for Cu) and the required minimum removal efficiencies (80% for Cu and 50% for Zn) were reached during the whole experimental period. As can be seen in Figure 2, the influent heavy metal concentration had a minor influence on treatment performance of sandy soil and technical filter media. The QS filter column managed to effectively remove all three heavy metals from the synthetic highway runoff (Run 1 and Run 2), however for experiments with synthetic roof runoffs (Run 3 and Run 4) effluent concentration of Cu as well as removal efficiencies of Cu and Zn were not met. It should be mentioned that the natural quartz sand (QS) turned out to contain some metal iron impurities, which could potentially serve as adsorption sites for heavy metal ions through surface complexes on iron oxyhydroxides. Metal ions that form outer-sphere complexes are readily exchangeable and are expected to be more easily displaced from the adsorbent surface (Bradl, 2004). For the experiment with zinc roof runoff (Run 4), effluent concentrations of Cu was significantly higher than its inlet concentration (150 µg/L) and exceeded the required levels of 1800 µg/L (BMFLUW, 2010). This phenomenon was due to the displacement of weakly adsorbed Cu from previous dosings (Run 1 – Run 3) in favor of the increased Zn influent concentration. The displacement of Cu can also be related to the relatively low influent pH, strength of complexation and adsorption order. The extent of simultaneous adsorption of heavy metals is influenced by adsorbate concentration and presence of competing metal ions (Nguyen, 2015). This competitive adsorption also showed that adsorption of Cu was decreased significantly when high concentration of Zn was added to the influent. The effect competitive heavy metal ions on adsorption efficiency was more pronounced in the QS filter column. For example, in the experiment with copper roof runoff (Run 3), effluent Cu concentration reached 70% of the inlet concentration while the effluent Zn concentration reached 100% of its inlet concentration (500 µg/L). Accordingly, Cu outcompetes Zn in occupying available sorption sites of QS. This is in agreement with the findings of Atanassova (1995) that in a multi-component system, an increase in the Cu concentration reduced the uptake of other heavy metals such as Ni, Cd, and Zn.

In general, subsequent dosings of the columns with synthetic runoff showed that the soil and technical filter media were able to remove the heavy metals, thus significantly reducing the concentrations of Cu, Pb and Zn (Figure 2). The extent of heavy metal removal depends on the initial heavy metal concentration and filter media type or composition (Genc-Fuhrman et al., 2007; Wang et al., 2017). The performance of each filter media in reducing the heavy metal levels was assessed based on the influent and effluent concentrations. All filters removed more than 98% of Pb. The mean removal efficiency of Cu was 89.6%, 97.4%, 98.5% and 90.5% through the filter columns packed with SS, TF-I, TFII and TF-III, respectively. The mean removal efficiency of Zn was 93.4%, 96.6%, 98.7% and 89.2% through the filter columns packed with SS, TF-I, TFII and TF-III, respectively. The results indicated that the mean removal efficiencies of Cu and Zn by the sandy soil and technical filter media are not statistically different. Nevertheless, tt seemed that composition of the studied technical filter media have played an important role in treatment efficiency. The mineral composition of TF-I and TF-II were similar, except for the 3% dolomite in the case of TF-II. However, the technical filter media with dolomite (TF-II) provided the best treatment performance indicating that carbonate content enhanced the removal of the studied metals.

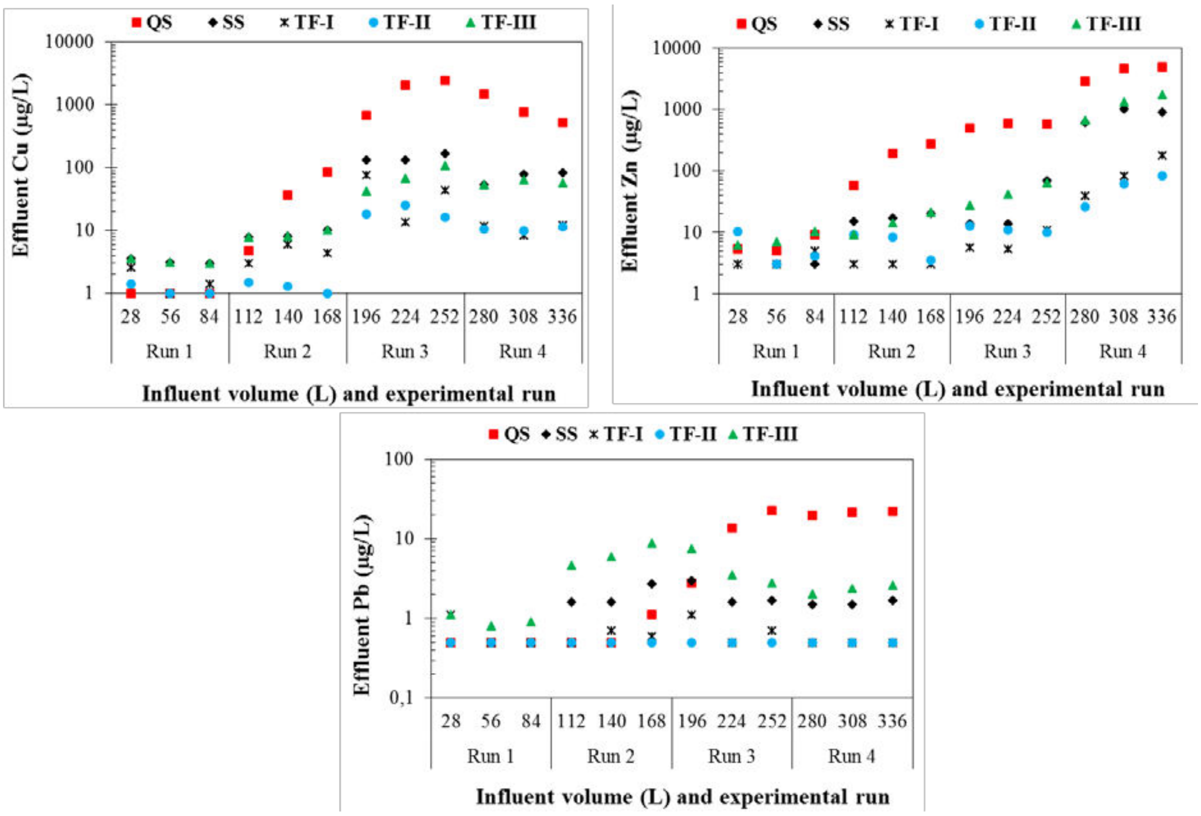

Figure 2. Effluent concentrations of heavy metals with different synthetic stormwater dosings (Run 1 – Run 4, see Table 2) under high hydraulic load experimental conditions as a function of treated volume.

Overall cumulative metal removal efficiency of each filter media was determined using the total influent and effluent loads (Run 1 to Run 4). Results of the cumulative removal efficiencies and the corresponding adsorption capacity are presented in Figure 3, and the influent load added to each column was 51.8 mg, 334.6mg and 542.9 mg for Pb, Cu and Zn, respectively. Load removal efficiencies through the soil based and technical filter media (SS, TF-I, TF-II and TF-III) were > 95 % for Cu and Pb, and more that 87% for Zn. These results demonstrate that all filter media were effective for the simultaneous removal of heavy metals, except for QS which had significantly lower removal efficiency of Cu and Zn. Similar to the findings in our study, Hatt et al. (2008, 2011) showed that a wide range of media compositions (i.e., combinations of sand, sandy loam, vermiculite, perlite, compost, mulch, charcoal) achieved more than 90% removal of Cu, Pb and Zn from synthetic stormwater. Results indicated that the natural quartz sand (QS) has lowest sorption capacity compared to soil based and technical filter media which is attributed to its low surface area and few sorption sites (Huber et al., 2016; Genc-Fuhrman et al., 2007; Wang et al., 2017). Sand used in stormwater in/filtration has relatively limited ability to remove dissolved contaminants compared with other adoptive media mainly due its low specific surface area and cation exchange capacity (Grebel et al., 2013). To enhance its treatment performance quartz sand media can be amended by mixing with a suitable adsorbent media (Wang et al., 2017).

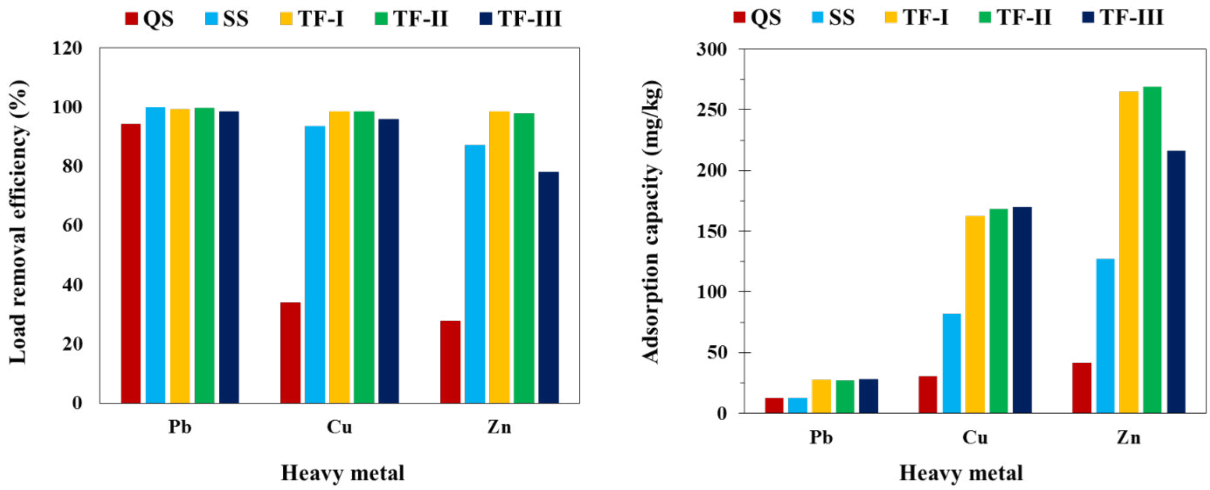

Figure 3. Overall removal efficiency (left) and adsorption capacity (right) of heavy metals by each filter media type.

3.3 Remobilization of heavy metals

Table 3 shows the mean and range of heavy metal concentrations measured at the effluent after each column was flushed with 5 g/L NaCl solution. Heavy metals were remobilized with different intensities depending on metal and filter media type. Regardless of the filter media type, remobilization of metals decreased in the following order Zn > Cu > Pb, which followed the levels of influent concentration and preloading of the filter columns. Effluent concentrations of metals measured after the passage of one bed volume were higher, but decreased successively over time with continued flushing. This suggested that precipitated or slightly adsorbed fractions of the retained metals were remobilized easily during the initial passage of NaCl solution. As displayed in Table 3, the de-icing salt (NaCl) solution had similar effects on the soil based and technical filter media and the three metals. Except for effluents from QS, remobilizations of metals were low and comply with the requirements noted in the ÖNORM B 2506–3 (2016). This relative low release indicates that adsorption was stable and salts would not have a major influence on the remobilization of previously retained metals. However, the heavy metals that had retained in the QS filter column were released in highest amounts indicating a major effect of NaCl so that this filter media is not feasible to utilize as adsorbent in stormwater filtration systems. The mechanism for the removal of metals by QS is outer-sphere complexation or non-specific electrostatic adsorption to negatively charged functional group sites on the sand particle surfaces.

Table 3. Remobilization of previously adsorbed metals during road de-icing salt application (42 L solution of 5 g/L NaCl). Mean effluent metal concentrations are indicated in bold numbers and the Italic values in bracket are ranges.

	TF I	TF-II	TF-III	SS	QS	ÖNORM (2016)
Pb	**4.2** *(3.1–7.3)*	**3.3** *(2.7–5.4)*	**6.4** *(2.7–13.9)*	**3.6** *(2.7–5.9)*	**83** *(63.2–203)*	**9**
Cu	**8.6** *(5.1–24)*	**6.8** (*4–17.9*)	**20.3** *(13.8–39)*	**7.0** *(4–15.8)*	**127** *(43–305)*	**50**
Zn	**23.7** *(12–47)*	**17.3** (*12–37*)	**160** *(121–186)*	**16.5** *(12–30)*	**825** *(676–1377)*	**500**

The mass and load fractions of each heavy metal remobilized from the filter columns as compared to the total mass previously retained by each filter media are presented in Table 4. As shown in Table 4, the effect of NaCl application was more pronounced for QS. The results showed that an extensive mobilization of heavy metals from the QS column (5.4% Cu, 6.8% Pb and 22% of Zn the total retained) occurred in response to NaCl application. Conversely, only a small fraction (< 2.0 %) of the retained heavy metals were mobilized from the soil and technical filter media. This implies that chemisorption was the principal metal removal mechanism and salts would not have a major effect on metal mobilization. Similar results were reported from column studies using alternative filter media other than soil for treatment of highway runoff (Huber et al., 2016b; Monrabal-Martinez et al., 2017). Monrabal-Martinez et al. (2017) observed a small release of Cd, Cu, Pb, and Zn (<3%) by NaCl from filter columns (pine bark, olivine, and charcoal) preloaded with about 50 mg of each metal. Conversely, other studies with soils containing 17 – 50% clay reported an extensive remobilisation of heavy metals as a result of exposure to high concentration of NaCl (Nelson et al., 2009; Norrström, 2005). This could be attributed to the fact NaCl promotes the dissolution of organic matter and/or clay which favours mobilization of heavy metals. Norrström (2005) evaluated the impact of de-icing salt on remobilization of Cd, Cu, Pb, and Zn from two soils from infiltration trenches for highway runoff (1.5– 2.7 mg/kg Cd, 155– 194 mg/kg Cu, 171– 324 mg/kg Pb, and 607– 781 mg/kg Zn). They reported that 37– 45 % of Cd, 0.1– 0.2% of Pb, and 4.7– 5.0% of Zn were leached by NaCl.

Table 4. Heavy metals adsorption and their remobilization/desorption using 42 L solution of 5 g/L NaCl.

Filter media	Adsorbed mass (mg)			Remobilized mass (mg)			Remobilized fraction (%)		
	Pb	Cu	Zn	Pb	Cu	Zn	Pb	Cu	Zn
QS	48.6	98.5	157	2.8	2.3	25.2	6.8	5.4	22
SS	52.1	303	526	0.15	0.29	0.86	0.29	0.1	0.16
TF-I	51.9	342	507	0.18	0.36	1	0.34	0.11	0.20
TF-II	49.8	342	542	0.13	0.29	0.95	0.26	0.08	0.18
TF-III	54.6	309	393	0.28	0.85	12.6	0.51	0.28	1.7

Remobilization of heavy metals is a function of several mechanism including cation exchange, colloid dispersion, chloride complex formation, metal characteristics, and total concentration of metals in the media (Bäckström et al., 2004; Nelson et al., 2009; Norrström, 2005). Overall, results of the present study indicated that the heavy metals (Cu, Pb and Zn) are strongly attached to the soil and technical filter media.

3.4 Effect of flow mode on heavy metal removal

The breakthrough curves for Cu, Pb and Zn removal at two different flow modes are shown in Figure 4. It has been observed that heavy metal removal efficiencies in the upflow mode were generally high as compared to the downflow mode. As shown in Figure 4 both the shape and gradient of the breakthrough curves were different with variations in flow direction. The breakthrough point for Cu and Zn, set at Ce/C_i =10 %, was almost 2300 BV for the downflow mode and 7600 BV for the upflow mode. At 20% breakthrough of Cu 9700 BV of synthetic stormwater was treated by TF-II operated in the upflow mode, and the requirements of the Austrian regulation regarding Pb maximum effluent concentration of 9 µg/L and Zn minimum removal efficiency of 50% were met. Accordingly exhaustion (lifespan) of the filter media (TF-II) was limited by Cu removal. The corresponding adsorption capacities of TF-II at the 20% breakthrough point of Cu were 573.8 mg/kg, 1182 mg/kg and 4669 mg/kg for Pb, Cu and Zn respectively. On the contrary, in the downflow mode 20% breakthrough of Cu was achieved at 7100 BV and exhaustion (lifespan) of the filter media was limited by Cu removal at nearly 7100 BV. The sorption capacity at exhaustion point was 447 mg/kg, 771mg/kg and 2771 mg/kg for Pb, Cu and Zn respectively. Similar to our findings a short breakthrough time and low adsorption capacity of metal ions was reported for a downflow mode as compared to the upflow mode (Athanasiadis, 2005; Inglezakis et al., 2001). For example, Athanasiadis (2005) reported that the adsorption capacity of clinoptilolite for Cu obtained at breakthrough point (C_e/C_i=10%), was 1144 mg/kg for the downflow mode and 1906 mg/kg for the upflow mode, respectively.

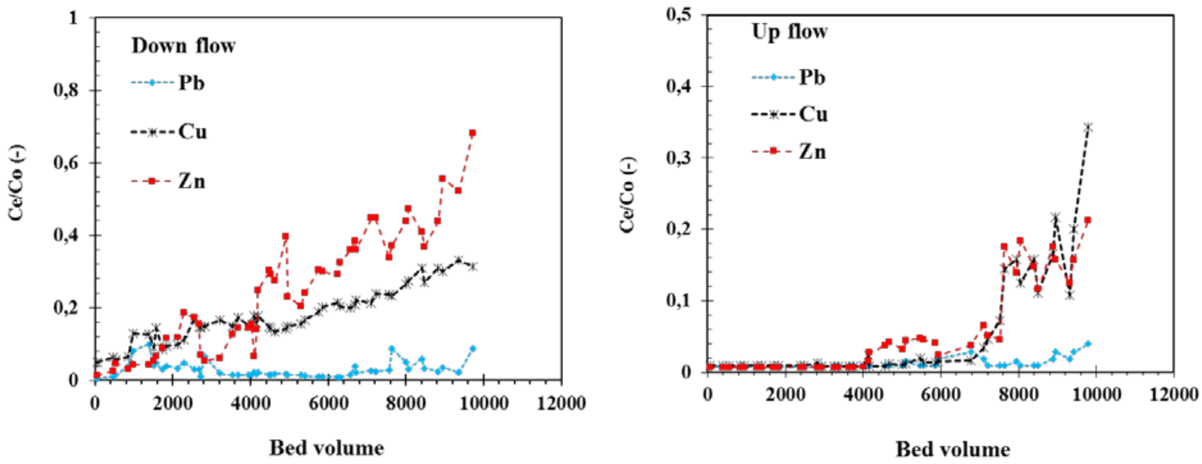

Figure 4. Comparison of breakthrough curves of Cu, Pb and Zn in downflow and upflow mode as a function of bed volume at a volumetric flow rate of 50 mL/min (50% of its maximum saturated flow rate) and filter bed volume of 160 mL. Note that to facilitate visibility of the graph, the Y-axis is different in each case.

The observed performance difference between the downflow and the upflow mode is explained by variabilities in the liquid holdups and liquid maldistribution (Inglezakis et al., 2001). The upflow mode allows saturation of all the vacant metal binding sites which leads to the achievement of higher equilibrium sorption process. These differences are attributed to the liquid holdup in the upflow mode is 100%, while for the downflow mode liquid holdup time is only a function of volumetric flow rate. Furthermore, feeding the multi-metal solution in the upflow mode ensures saturated flow conditions and uniform hydraulic distribution of the sorbate. Accordingly, under the same experimental conditions it becomes apparent that the upflow mode resulted in a more effective use of the filter media. Results of the present study demonstrated that the upflow mode was more efficient in maintaining a saturated flow through condition leading to higher sorption capacity. Therefore, to predict the life span of filter media based on sorption capacity, column experiment operating in upflow mode would be more appropriate. Composition of the filter media TF-II was similar to the filter applied in parking lot runoff filtration system. Results of the upflow mode were comparable to the results obtained for filter media collected from in operation filtration system (Haile et al., 2016b). Thus, column experiment in the upflow mode can be used to determine lifespan and the changes in the long-term treatment efficiency of filter media by considering the three parameters (Cu, Pb and Zn) noted in the new Austrian Standard Method (ÖNORM B 2506 – 3, 2016).

3.5 Breakthrough curves

The breakthrough curves of Cu, Pb and Zn in the column experiments are presented in Figure 5. Subsequent dosing of the columns with synthetic roadway runoff showed that treatment by technical filter media (TF-I, TF-II, and TF-III) and sandy soil (SS) filters are effective in removing Cu, Pb, and Zn simultaneously to effluent levels below analytical detection limit (i.e. Ce/Ci < 0.01). After the breakthrough (i.e. Ce/Ci = 0.1) metal effluent concentrations from all filter media started to increase overtime as a function of treated bed volumes. The patterns of metals breakthrough curves (Figure 5) were similar for all filter columns and the steepness of the breakthrough curves decreased in the order of Pb > Cu > Zn for all filter media.

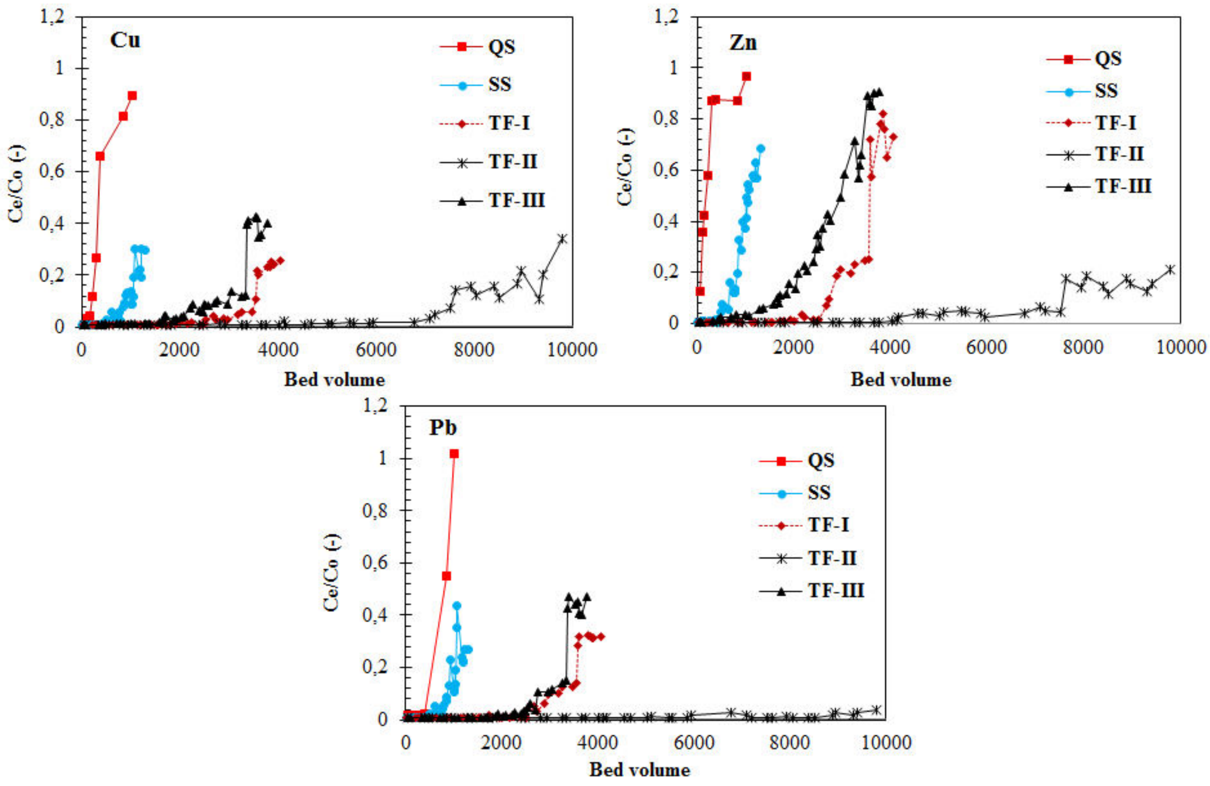

Figure 5. Breakthrough curves of column experiments for metal mixtures. The lines are not fitting functions; they simply connect points to facilitate visualization.

The volume of stormwater treated by all five filter media was different depending on the sorption capacity and total flow through volume at exhaustion. It has to be noted that TF-II treated more stormwater before breakthrough as compared to other filter media types. Zn breakthrough (Ce/Ci < 0.1) occurred in the QS filter media first, followed by SS, TF-III, TF-II and TF-II, respectively (Figure 5). The number of bed volumes to breakthrough was 55, 680, 1700, 2700 and 7600 for QS, SS, TF-I, TF-III and TF-II, respectively. Zn breakthrough generally occurred faster as compared to Cu and Pb which demonstrates that the influent metal concentration has a significant effect on breakthrough curve. This was in good agreement with earlier studies which showed Zn is relatively mobility than Pb and Cu (Hatt et al., 2011). As indicated in Figure 5 full breakthrough (i.e. Ce/Ci = 1) of Pb, Cu, and Zn was not observed in all filters within the experimental running time of three months, except for QS filter column.

Experimental results with the QS filter column indicated a breakthrough of Cu, Pb, and Zn beginning at 50 BV and was nearly complete ($C_e/C_i \approx 1.0$) after a total flow though of 1000 BV. The filter column QS was found to be the worst with fair metal removal in the early dosing stages, but shortly effluent concentration of Pb exceeded the groundwater quality criteria value of 9 µg/L (BMFLUW, 2010). Similar to our findings, Genc-Fuhrman et al. (2007) reported that among 11 sorbents, sand has a minor efficiency for the removal of heavy metals which is attributed to its low specific surface area and cation exchange capacity. Removal of heavy metals increased with increasing pH by processes such as surface complexation between dissolved species and oxide– and hydroxide groups (Genc-Fuhrman et al., 2007). The decrease in metal removal was in line with the pH drift curve (Figure 1). Measured effluent pH levels showed that QS has very limited pH buffering capacity, consequently its performance on heavy metal removal was very poor as compared to other filter media (i.e. SS, TF-I, TF-II, and TF-III).

The filter columns packed with soil and technical filter media showed a maximum breakthrough values (C_e/C_i) of around 0.3 to 0.5 for Pb and Cu, and 0.21 to 0.91 for Zn. The column data suggest that TF-II has high affinity for all tested heavy metals and the magnitude of the sorption (up to 7100 BV) remained constant with removal rates of over 93%. Initial metal ion concentration has a significant effect on breakthrough time and filter media exhaustion. The breakthrough curves determined in this study were slower and least steep, thus the number of BV passed at breakthrough time reported here are much higher than those reported in other studies. It is difficult to compare these results directly to those of other investigators because the influent concentrations are often higher than the levels used in this study (Huber et al., 2016b; Nguyen et al., 2015; Pawluk and Fronczyk, 2014).

It is possible that the adsorption capacity of the filter media may be exhausted before clogging occurs, resulting in high effluent concentrations exceeding discharge water quality and low removal efficiencies of heavy metals (Hatt et al., 2011). The effluent concentration of Pb was exceeding the maximum allowable of 9 µg/L for groundwater protection (BMFLUW, 2010) after the passage of 300, 1060, 3360 and 3600 bed volumes for QS, SS, TF-I, and TF-III, respectively. Comparable bed volumes were treated to reach the 20% and 50% breakthrough of Cu and Zn, respectively. On the contrary, for the filter column packed with TF-II 20% breakthrough (80% removal) of Cu was achieved after the passage of 97000 BV while effluent concentration of Pb, and Zn removal efficiency fulfil the requirements throughout the entire experimental duration. Therefore, lifespan of TF-II was limited by Cu removal and after the treatment of 9800 BV this filter media was considered as exhausted.

The cumulative heavy metal loads applied to each column, mass retained in the filter column, cumulative load removal efficiencies and adsorption capacity at filter media exhaustion point are summarized in Table 5. Due to the differences in volumetric flow rate and the exhaustion point, the total flow though volume and total influent heavy metal loads were different each filter. It can be seen that the influent load of individual contaminants applied into the QS and SS was significantly lower in comparison to the loads applied to the technical filter media. Nevertheless, the load removal efficiencies of each filter column were comparable. Over 90% Cu and Pb dosed into the columns were retained in the filter media, while Zn removal ranged from 62.6 % (QS) to 93% (TF-II).

Table 5. Removal efficiencies and sorption capacity of each filter column at filter media exhaustion for Cu, Pb and Zn

Column	q_i (mg)			q_s (mg)			Load removal (%)			q_e (mg/kg)		
	Cu	Pb	Zn	Cu	Pb	Zn	Cu	Pb	Zn	Cu	Pb	Zn
QS	3.92	1.92	17.9	3.39	1.87	11.2	83.8	97.5	62.3	13.7	7.6	45
SS	20.3	9.6	87.1	19.5	9.2	77.4	96.3	95.8	88.8	66.8	31.5	265
TF-I	68.4	32.3	306	66.9	31.3	306	97.9	96.8	94.5	523	245	2390
TF-II	172	83.3	678	163	82.2	641	94.6	98.6	94.5	1124	567	4420
TF-III	64.0	30.2	286.3	60.9	29.1	228	95.2	96.1	79.5	430	205	1558

The adsorption capacity (mg/kg) of each filter media at column exhaustion point towards individual heavy metal varied significantly. Values of breakthrough show that the adsorption capacity decreases in the following order: Zn > Cu > Pb. This variability is possibly related to the differences in influent concentrations, adsorption affinity (selectivity sequence) as well as weight of filter media. Consequently, adsorption capacity of filter media was found to be in the order of TF-II > TF-I, TF-III > SS > QS. The adsorbent mixture components of TF-I and TF-II (Table 2) were similar, despite for the 3% dolomite addition in TF-II. Comparison of the adsorption capacities of breakthrough curves evidenced that adsorption of Cu, Pb and Zn onto technical filter media was enhanced in the presence of dolomite. The results of this study eventually supported the theory that presence of dolomite increased the pH of the solution above solubility point which caused metals to precipitate as metals oxide and probably metals carbonate Reddy et al., 2014). The lowest adsorption capacities observed in the filter column packed with natural quartz sand (QS) could be due to its low affinity and non-reactive characteristics which is in agreement with a previous study using sand for metal removal (Genc-Fuhrman et al., 2007, Seelsaen et al., 2006).

3.6 Filter media lifespan

The lifespans will be dependent on needed removal efficiencies and effluent water quality requirements. Size of the stormwater treatment system relative to its impervious catchment area allows designers to predict lifespan of a filter media regarding adsorptive removal of heavy metals. Based on the cumulative heavy metal loading (Table 5) the investigated filter media could be sized at 4% (SS), 1% (TF-I and TF-III), and 0.4% (TF-II) of their impervious catchment size. In order to meet the required removal efficiencies of 80% for Cu and 50% for Zn, predicated lifespan of the filter media were at least 35, 36, 41 and 29 years for SS, TF-I, TF-II and TF-III, respectively. The lifespans determined in the present study are relatively high compared to other studies (Monrabal-Martinez et al., 2017; Thomas et al., 2014). For example, mixed media composed of perlite, dolomite and gypsum showed an estimated lifespans from 14 to 22 years for Cu and Zn (Thomas et al., 2014). The variability of estimated lifespan might be attributed to the filter media composition, influent concentration, filter bed depth, size of the treatment system relative to its impervious catchment area.

In practice, lifespan of stormwater infiltration/filtration is usually highly dependent on mitigating sediment input to the system. Solids in stormwater might settled out at the surface of filtration system forming a cake layer or are removed in the pores of the filter bed via filtration are considered to play a vital role in reducing the hydraulic performance of the filtration system due to physical clogging. Clogging of filter media is recognised as the main limiting factor regarding the operational lifespan of stormwater infiltration/filtration systems (Hatt et al., 2008; Haile et al., 2016b). A previous study of stormwater filtration systems constructed with filter media similar to TF-I showed a significant decrease in the infiltration capacity of the systems after five to seven years of operation due to the formation of a clogging layer at the surface of the filters, while the lifespan regarding heavy metal removal was 30 years (Haile et al., 2016b). The authors suggested that the hydraulic performance of the system could be recovered by scraping off the surface accumulated particle layer, and replacement or back flushing of the geotextile on periodic bases, approximately every 7 years. Further research should seek to understand the clogging phenomena of filter media receiving particles and contaminants that mimic the real conditions

4. Conclusion

In the present study, the simultaneous adsorption of heavy metals (Cu, Pb and Zn) from synthetic stormwater runoff aqueous solutions using quartz sand (QS), sandy soil (SS) and three mineral-based technical filter media (TF-I, TF-II and TF-II)) was investigated. The column study result were also used to evaluate effect of de-icing salt (NaCl) on the mobility of retained metals, size of treatment system relative to its imperious catchment area and predict infiltration/filtration system lifespan. The results demonstrate that soil based and mineral-based technical filter media are potentially efficient for the removal of heavy metals under high hydraulic loading conditions. All effluent concentrations measured during the infiltration of synthetic highway and roof runoff fulfilled the requirements of the Austrian regulations (9 µg/L Pb and 1800 µg/L Cu). Additionally, required removal efficiencies for Cu (80%) and Zn (50%) were met during the whole experimental run. However, effluents of Cu from QS column was exceeding the exceeded the required levels of 1800 µg/L as well as required removal efficiencies Cu and Zn were not met. Application of the de-icing salt (NaCl) minor effect on the remobilization for most heavy metals adsorbed heavy metals from the sandy soil and all technical filter media columns and all effluent concentrations fulfil the Austrian regulations. However, results from the natural quartz sand (QS) column showed approximately 6.8%, 5.2% and 22 % of the retained Pb, Cu and Zn respectively, were leached in response to NaCl application as well as effluent concentrations of Pb and Cu exceeded the maximum allowable concentration.

Results of long-term treatment performance (Breakthrough curves) demonstrated that mineral-based technical filter media are able to treat a higher volumes of stormwater in small filtration systems relative to their impervious catchment area (0.4% to 1.0%) so that they are potentially suitable for utilization in compact stormwater treatment, particularly in urban landscapes where space is very limited. Breakthrough of Cu, Pb and Zn is not expected occur during the operating life of a filter. Physical clogging will occur first limiting the lifespan of infiltration/filtration treatment system. The authors suggested that a geotextiles could be installed at the top of the filtration system to act as a barrier to avoid stormwater runoff particles from reaching and clogging the main filter bed. The geotextile filter and accumulated particle layer could be scraped off and replaced at a desired time interval depending on site constraints and clogging rates.

5. Acknowledgment

The Austrian Federal Ministry of Agriculture, Forestry, Environment and Water Management (BMLFUW) and SW-Umwelttechnik are gratefully acknowledged for providing funds. We sincerely thank the chemical laboratory at the Institutes of Soil Sciences and Sanitary Engineering and Water Pollution Control for conducting the analysis.

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6.7 Paper VII

Design and operational problems of sedimentation basins for street runoff treatment. "Probleme bei Planung und Betrieb von Absetzbecken für Straßenabwässer, in German)".

Haile, T.M., Kammerer, G. Fürhacker, M. (2014). Österreichische Wasser- und Abfallwirtschaft, 66, 112-119; ISSN 0945-358X.

Österr Wasser- und Abfallw (2014) 66:112–119
DOI 10.1007/s00506-014-0138-2

Probleme bei Planung und Betrieb von Absetzbecken für Straßenabwässer

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Online publiziert: 21. Februar 2014

Zusammenfassung Das Absetzbecken einer Gewässerschutzanlage dient dem Rückhalt von Partikeln und erfüllt die Aufgabe eines Retentionsraumes. Die Erfüllung dieser Aufgaben hängt stark von der baulichen Ausführung des Absetzbeckens ab. Anhand des konkreten Beispiels einer Gewässerschutzanlage wurden Planung und Ausführung der Anlage verglichen. Dabei wurde ein signifikanter Unterschied der Höhenlage des Verbindungsrohres zwischen Absetz- und Filterbecken festgestellt. Die ausgeführte Anordnung bewirkt ein Trockenfallen des Beckens, geänderte Fließgeschwindigkeiten und somit ein nicht plangemäßes Sedimentationsverhalten im Becken. Um die Fließwege und die mittlere Aufenthaltszeit im Becken herauszufinden, wurden Modellversuche mit einem Farbtracer durchgeführt. Dabei wurden die ausgeführte Variante sowie die Wirkung von Einbauten betrachtet. Die wesentliche Erkenntnis bestand darin, dass der Einbau einer Prallwand zu längeren Aufenthaltszeiten im Becken, einer gleichmäßigeren Durchströmung und zu einer kleineren mittleren Geschwindigkeit führt. Dadurch könnten mehr Sedimente abgesetzt werden. Die ermittelten Spurenstoffkonzentrationen im Zulauf der Anlage liegen im Bereich von Literaturwerten. In der ausgeführten Form ist durch das Sedimentationsbecken kein dauerhafter Rückhalt der partikulären Fracht gegeben. Die bauliche Ausgestaltung und die Betriebsweise verursachen starke Strömungen und Turbulenzen im Becken. Dadurch kommt es im anschließenden Filterbecken zu einer zusätzlichen Belastung durch hohe Feststoffeinträge. Die negativen Auswirkungen dieser Belastung zeigen sich in der Abnahme der hydraulischen Durchlässigkeit und der Filterleistung des Filtersubstrats.

Im Rahmen einer weiteren Studie wurde die Reinigungsleistung des Absetzbeckens in Hinblick auf die Konzentration von abfiltrierbaren Stoffen (AFS) im Zu- und Ablauf ausgewertet. Die Gewässerschutzanlage wurde ein Jahr überwacht; die Ergebnisse zeigten keine Partikelansammlung im Absetzbecken bzw. keine dauerhafte Beibehaltung der Partikelfracht. Die mittlere AFS-Konzentration im Zulauf des Absetzbeckens betrug 89 mg/L und im Ablauf 94 mg/L. Weiters war festzustellen, dass Aufbau und Betrieb des Systems zu ausgeprägt turbulenten Strömungsverhältnissen im Absetzbecken und zu einer geringen Sedimentation von feinen und mittelgroßen Partikeln führen. Das Resultat ist eine Beladung des Substrats im Filterbecken mit diesen Teilchen. Konkret werden die Partikel entweder auf der Oberfläche des Filters oder im Porenraum abgelagert. In beiden Fällen steigt der Bedarf an hydraulischem Potenzial bzw. an Überstauhöhe für die Durchströmung des Mediums, die Anlage ist bei zunehmend kleineren Niederschlagsereignissen hydraulisch überlastet. Daher muss der Filter regelmäßig überwacht und gewartet werden, und zwar im ersten Fall durch Entfernen der Kolmationsschicht an der Oberfläche und im zweiten Fall durch einen Austausch der Filterschicht.

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Design and operational problems sedimentation basin for street of runoff treatment

Abstract Sedimentation basins are incorporated into runoff infiltration systems and serve as a retention system, in which the sedimentation of particles occurs as a result of gravitational force. The effectiveness of sedimentation basins in terms of removing particles is highly dependent on design parameters such as their dimensions and drainage times. To evaluate the flow distribution within sedimentation basins, a conceptual model was developed using a tracer and compared to the actual condition at the treatment system. Here, a significant difference in the height of the connecting tube between settling basin and filter basin was found. This leads to a drying out of the basin, which significantly affects the flow rates and hence the sedimentation there. Based on a model experiment with colour tracer, the flow paths were visually represented in the basin, and both the concrete variant and the effects of internal structures were considered. It was found that the incorporation of a baffle led to an improved utilization of the basin, and to water being retained for longer times. Due to the lower flow rate and these longer times, fine and medium-sized sediments could be easily settled.

In a field study, the performance of a sedimentation basin for the removal of particles was evaluated based on the influent and effluent concentrations of total suspended solids (TSS). The street runoff treatment plant was monitored for one year and results showed that there was no particle accumulation in the sedimentation basin (there was no permanent retention of the particle loads). The mean TSS concentrations were 89 mg/L at the inlet of the sedimentation basin and 94 mg/L at the end of the settling tank, respectively.

The structural design and operation of the system showed higher flow and turbulent flow conditions in the sedimentation basin, in which settlement of fine and medium sized particles was very low; as a result, the soil filter basin was loaded with runoff particles. Thus the particulate matter had to be removed either on the surface of the filter basin or within the porous space of the filter. The build-up of such particles can result in a significant increase of head loss due to clogging; therefore, filters must be maintained by removing the accumulated particles on a regular basis.

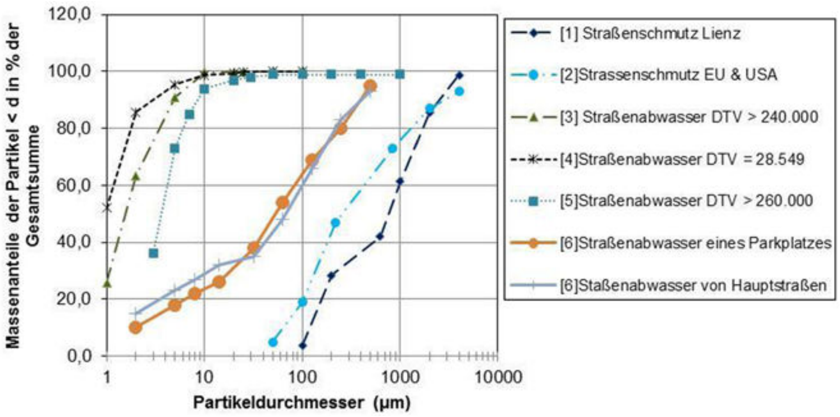

Abb. 1 Partikelgrößenverteilung im Straßenabwasser bzw. Straßenschmutz: *[1]* eigene Untersuchungen 2013; *[2]* Dierschke et al. 2010; *[3]* Fürhacker et al. 2013; *[4]* Lins 2013; *[5]* Li et al. 2005; *[6]* Selbig and Bannerman 2011

1. Einleitung

Die Versickerung von Niederschlagswässern ist in vielen Fällen eine ökologisch sinnvolle Maßnahme. Die europäische Wasserrahmenrichtline (WRRL) definiert Güteziele, die sicherstellen sollen, dass sich alle Oberflächengewässer und Grundwässer in einem guten Zustand befinden und dieser auch erhalten wird. Für Grundwasser fordert die WRRL eine Verhinderung bzw. eine Begrenzung der Einleitung von Spurenstoffen zur Herbeiführung eines guten chemischen Zustandes bzw. der Verhinderung einer Verschlechterung des chemischen Zustandes. Einbringungen von verunreinigtem Niederschlagswasser in den Untergrund sind Maßnahmen, die die Beschaffenheit von Grundwasser großflächig und nachhaltig beeinträchtigen können, weshalb für z. B. Straßenabwässer in verschiedenen Regelwerken (z. B. RVS 0404.11 bzw. ÖNORM 2506, Teil 1 und 2) eine Reinigung vorgesehen ist.

Partikel auf der Straßenoberfläche spielen eine wichtige Rolle beim Transport von chemischen Bestandteilen im Niederschlagsabfluss. Neben der offensichtlichen Beeinträchtigung der Wasserqualität durch Sedimente und hohe Trübung, wirken remobilisierte Sedimente als Substrat für andere chemische Spurenstoffe (Herngren et al. 2006). Untersuchungen von Partikeln im Autobahnabfluss zeigten, dass ein erheblicher Anteil der Schadstoffe wie Schwermetalle, polyzyklische aromatische Kohlenwasserstoffe (PAK) und mineralische Öle und Fette an feine Partikel gebunden sind (Boller 2004; Dierschke et al. 2010; Wichern et al. 2012; Xanthopoulos 1990). Die Größenverteilung von Partikeln sowie ihre Zusammensetzung können einen wesentlichen Einfluss auf die Wasserqualität haben. Studien zur Partitionierung von Schwermetallen in verschiedene Kornfraktionen ergaben, dass die meisten der anorganischen und organischen Verunreinigungen mit den feinen Partikeln assoziiert sind. Schorer (1997) untersuchte die Konzentration von anorganischen und organischen Schadstoffen in verschiedenen Größenfraktionen im Bereich von <2 µm bis 200 µm und fand eine lineare Korrelation zwischen Schwermetallgehalt und Partikelgröße. Die Analyse der Konzentrationen von Zn, Fe, Pb, Cd, Cu, Cr, Al und Mn in abgelagerten Straßensedimenten in fünf Partikelgrößenfraktionen aufgeteilt zeigten, dass die maximale Konzentration der Schwermetalle im Partikelgrößenbereich von 0,45 µm bis 75 µm auftrat. Herngren et al. (2006) und Xanthopoulos (1990) berichteten, dass mehr als 80 % Cu, Ni und Pb und mehr als 70 % Zn und Cd im Bereich zwischen 6 µm und 60 µm sorbiert sind. In diesem Zusammenhang sind feine Partikel von größerer Bedeutung als gröbere, weil sie eine viel größere spezifische Oberfläche besitzen, die die Adsorption der Schadstoffe erleichtert (Herngren et al. 2006; Sansalone et al. 1998).

Herngren et al. (2006) zeigten auch, dass der Anteil an organischen Stoffen eine wichtige Rolle bei der Partitionierung der Metalle in unterschiedliche Korngrößen spielt. Mehrere andere Forscher berichteten, dass der organische Kohlenstoffgehalt in Feinsedimenten größer ist als in groben Sedimenten (Andral et al. 1999; Roger et al. 1998). Die bedeutende Rolle der feineren Partikel im städtischen Regenwasser wird also durch den hohen Gehalt an organischem Kohlenstoff noch verstärkt.

An Partikel gebundene Spurenstoffe können entweder durch Absetz- oder Filtrationsprozesse entfernt werden. Um die Wasserqualität des Straßenabflusses durch eine effektive Anwendung von Behandlungssystemen zu verbessern, ist es notwendig, die Verteilung der Schadstoffkonzentrationen und massen auf die Partikelgrößen und frachten zu verstehen, um die Partikelentfernung durch die gewählten Behandlungsverfahren (z. B. Abscheidung im Absetzbecken und Filtration durch ein spezifisches Filterbett) zu optimieren. Beispiele für Verteilungen von Partikeln im Straßenabwasser (Studie von Fürhacker et al. 2013; Li et al. 2005) und im Straßenschmutz (Literaturzusammenfassung von Dierschke et al. 2010; Selbig and Bannerman 2011 sowie eigene Untersuchungen) sind in der Abb. 1 zusammengestellt.

Die Literaturzusammenfassung von Dierschke et al. (2010) ist repräsentativ für Straßenschmutz in den USA und in der EU.

Es wäre unwirtschaftlich, die Filtereinrichtungen auf die bei Niederschlagsereignissen auftretenden kurzzeitigen Spitzenabflüsse zu bemessen. Die Differenz zwischen dem Volumenstrom des Straßenabwassers beim Zulauf zur Gewässerschutzanlage und dem durch die Filtereinrichtung hindurchtretenden Volumenstrom muss daher zwischengespeichert werden. Dies kann entweder in Retentionsbecken oder Absetzbecken erfolgen. Absetzbecken in Gewässerschutzanlagen dienen dem Rückhalt von Partikeln und erfüllen die Aufgabe eines Retentionsraumes. Der Erfüllungsgrad dieser Aufgaben hängt stark von der baulichen Ausführung der Absetzbecken ab.

Der Gesamtgehalt an Spurenstoffen wird durch den Absetzmechanismus der verschiedenen Partikelgrößen beeinflusst (Deletic et al. 2000; Sansalone et al. 1998). Das Absetzverhalten nicht-kolloidaler

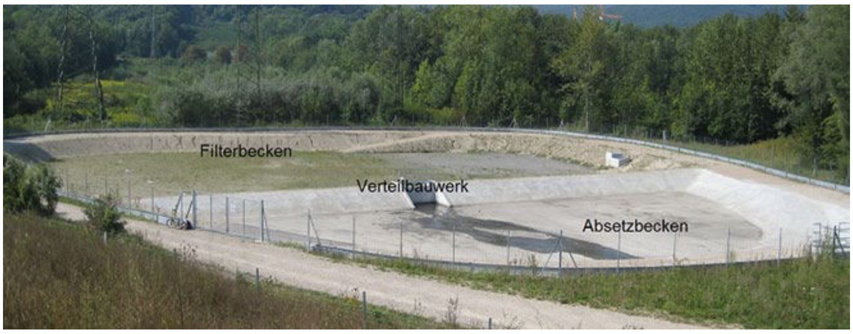

Abb. 2 Übersicht die Anlage im Betrieb (März 2012)

Partikel im Abflusswasser ist nicht nur für die städtische Regenwasserbehandlung von großer Bedeutung. Die Vertikalbewegung der Partikel im Absetzbecken kann entweder numerisch abgeschätzt – und zwar mit Modellansätzen, die auf der Stokesschen Gleichung beruhen und bei denen die Partikelgrößenverteilung (PSD) die wesentliche Eingangsgröße ist – und/oder durch Absetzversuche physikalisch bestimmt werden.

Berechnungen des Absetzverhaltens können entweder direkt für die Auslegung von Absetzbecken oder für die Abscheidung bestimmter Partikelgrößen herangezogen werden, oder indirekt zur Bestimmung kritischer Geschwindigkeiten eingesetzt werden.

Die Ziele dieser Ausführungen bestehen darin, die Partikelverteilung im Straßenabwasser und Berechnungsgrundlagen des Absetzverhaltens darzulegen, die sich daraus ergebenden Probleme beim Bau und im praktischen Betrieb zu diskutieren und Verbesserungen vorzuschlagen. Anhand eines konkreten Beispiels werden Planung und Ausführung verglichen und es wird der Wirkungsgrad durch Probenahme bestimmt. Anhand eines Modellversuchs werden mögliche Verbesserungsmaßnahmen untersucht.

2. Material und Methoden

Für die Versuche und Betrachtungen wurde eine Gewässerschutzanlage (GSA) in der Nähe von Wien ausgewählt. Es wurden einerseits Errichtungspläne mit der Ausführung in der Praxis verglichen und andererseits Modellversuche an einem nachgebauten Gipsmodel untersucht und Messungen an der GSA in Betrieb durchgeführt.

2.1. Planung und Ausführung der GSA

Die betrachtete Gewässerschutzanlage wurde zur Behandlung von Straßenabwässern einer Autobahn errichtet. Die Form der Anlage und Details der beiden Becken sind in der Abb. 2 ersichtlich. Das Einzugsgebiet der Anlage umfasst eine Fläche von 49 801 m².

Die Anlage ist eine Kombination aus einem Absetzbecken und einem Filterbecken. Beide Elemente haben eine Retentionsfunktion. Im Absetzbecken soll das Volumen des Spülstoßes abgefangen werden. Dieses Wasser wird über eine Drosselstrecke ins Filterbecken geleitet und die Abflussspitze dadurch gedämpft. Das den ersten Spülstoß bzw. das Bemessungsvolumen des Absetzbeckens überschreitende Wasservolumen wird über eine Überlaufschwelle in das Filterbecken eingeleitet. Als Bemessungsgrundlage wurde ein 5-jährliches Regenereignis herangezogen. Durch dieses ergibt sich ein Bemessungsspeichervolumen von 4 800 m³.

Im Regelfall wird das gefilterte Wasser nach der GSA von einem Schmutzwasserkanal aufgenommen. Bei Extremereignissen über dem 5-jährlichen Regenereignis erfolgt eine direkte Einleitung in den Vorfluter.

Um bessere Aussagen über die bestehende Anlage treffen zu können, wurde sie begangen und vermessen. Anschließend wurden die Einreichpläne mit der ausgeführten Variante verglichen.

2.2. Modellversuche

Zur Beurteilung der Strömung des Straßenabwassers und ihrer räumlichen Verteilung im Absetzbecken wurde im Rahmen einer Projektarbeit ein physikalisches Modell gebaut (Kessler und Steurer 2012). In den Modellexperimenten wurden die Fließwege im Becken mit einem Farbtracer visuell dargestellt. Es wurden die ausgeführte Variante sowie die Wirkung von Einbauten untersucht. Dabei wurde festgestellt, dass der Einbau einer Prallwand zu einer gleichmäßigeren Durchströmung des Beckenquerschnittes und somit längeren Aufenthaltszeiten führt. Durch die geringere Geschwindigkeit und längere Aufenthaltszeit könnten mehr Sedimente abgesetzt werden.

Das Absetzbecken wurde im Längenmaßstab zwischen Modell und Natur von 1:50 nachgebaut. Da die zu simulierenden Strömungen mit freier Oberfläche stattfinden, wurde das Ähnlichkeitsgesetz von Froude bei der Durchführung und Analyse der Versuche angewandt. Die Froude-Zahl *Fr* ist folgendermaßen definiert:

$$Fr = \frac{v}{\sqrt{g \cdot L}} \qquad (1)$$

Das Ähnlichkeitsgesetz von Froude besagt, dass die Froude-Zahl in der Natur und im Modell gleich groß sein muss.

Die verwendeten Materialien waren Holz und Gips. Der mit Wasser beaufschlagte Bereich wurde lackiert. Für die Tracerversuche wurde Lebensmittelfarbe verwendet.

Der Durchfluss der Versuche wurde mittels der Durchflussmaßstabszahl auf das Modell umgerechnet. Als maßgeblicher Durchfluss in der Natur wurde die Bemessungsregenspende $r_{n,\,t}$ verwendet. Die Zuflussdaten entstammen dem technischen Bericht der GSA.

Über die Durchflussmaßstabszahl P_Q (siehe Tab. 1 und den Spitzenzufluss wurde der für das Modell adäquate Durchfluss von 0,183 L/s bestimmt.

Im Modell wurden folgende strömungsberuhigende Maßnahmen ausprobiert:

- Beruhigungsrechen,
- Prallwand.

Durch die teilweise Zugabe der Lebensmittelfarbe konnte die Strömung erheb-

Tab. 1 Maßstabszahlen für die Modellversuche

Beruhigungsmaßnahme	Bezeichnung	Wert
Längenmaßstabszahl	P_L	50
Flächenmaßstabszahl	P_A	2500
Geschwindigkeitsmaßstabszahl	P_v	7,1
Zeitmaßstabszahl	P_T	7,1
Durchflussmaßstabszahl	P_Q	17678
Längenmaßstabszahl	P_L	50

lich besser veranschaulicht werden. Als Referenz wurde vor dem Einbau jeder Maßnahme jeweils der unbeeinflusste Zustand betrachtet.

Ein wesentliches Kriterium für die Beurteilung diverser Modellvarianten ist ihr Absetzverhalten. Um die Sinkgeschwindigkeit von suspendierten Partikeln in einer viskosen Flüssigkeit zu beschreiben, geht man am besten vom Gesetz von Stokes aus, wonach die auf einen kugeligen Körper wirkende Reibungskraft F_R (dim $F_R = MLT^2$) von der dynamischen Viskosität η des Fluids (dim $\eta = ML^{-1}T^{-1}$), in dem sich der Körper befindet, und der Relativgeschwindigkeit des Körpers zum Fluid v (dim $v = LT^{-1}$) abhängt:

$$F_R = 6\pi r \eta v \quad (2)$$

Die Reibungskraft wirkt dabei entgegengesetzt der Geschwindigkeit. Weiters unterliegt der Körper noch der Erdanziehung $F_G = \rho_s \cdot g \cdot V$ (mit der Feststoffdichte ρ_s, dim $\rho_s = ML^{-3}$, der Fallbeschleunigung g, dim $g = LT^{-2}$ und dem Partikelvolumen V, dim $V = L^3$) und der Auftriebskraft $F_A = \rho_l \cdot g \cdot V$, wobei ρ_l die Dichte der Flüssigkeit ist. Für ein sinkendes Teilchen im Kräftegleichgewicht gilt $F_G = F_A + F_R$, woraus sich mit dem Kugelvolumen $V = 4\ r^3\ \pi/3$ folgende Formel für die Sinkgeschwindigkeit ergibt:

$$v = \frac{2r^2 \cdot g \cdot (\rho_S - \rho_l)}{9\eta} \quad (3)$$

Das ist die Stokessche Gleichung, wonach die Sinkgeschwindigkeit eines kugeligen Teilchens in einer (unendlich ausgedehnten) ruhenden Flüssigkeit quadratisch von seinem Radius bzw. Durchmesser abhängt und zur Differenz zwischen der Feststoff- und der Flüssigkeitsdichte direkt und zur dynamischen Viskosität des Fluids verkehrt proportional ist. Die Fluideigenschaften ρ_l und η sind ihrerseits von der Temperatur abhängig. Für nichtkugelige Partikel ist r als jener halbe Äquivalentdurchmesser aufzufassen, mit dem ein kugeliges Teilchen dieselbe Sinkgeschwindigkeit ergibt. Bewegt sich die Flüssigkeit, hängt die Sinkgeschwindigkeit noch von der herrschenden Turbulenz ab (Burton und Pitt 2002; Raudkivi 1990).

Ersetzt man die Sinkgeschwindigkeit in der Stokesschen Gleichung durch den Quotienten aus dem zurückgelegten Vertikalweg bzw. der Höhendifferenz Δh und der Fall- bzw. Absetzzeit t_{abs}, ergibt die Auflösung der Gleichung nach der Absetzzeit

$$t_{abs} = \frac{9\eta \cdot \Delta h}{2r^2 \cdot g \cdot (\rho_S - \rho_l)} \quad (4)$$

Somit kann bei einem bestimmten Δh die Größe der kleinsten absetzbaren Partikel innerhalb der Aufenthaltszeit berechnet werden. Je größer die Aufenthaltszeit der Suspension aus Abwasser und Schwebstoffen im Absetzbecken ist, umso kleinere Partikel können abgesetzt werden. Für eine Optimierung des Absetzbeckens gilt es daher, die Aufenthaltszeit zu maximieren.

Das Stokessche Gesetz ist nur für eine laminare Strömung gültig, bei der sich Fluidpartikel in Schichten oder Lamellen bewegen. Ob das Strömungsverhalten turbulent oder laminar ist, kann mit der dimensionslosen Reynolds-Zahl *Re* bestimmt werden. Diese ist das Verhältnis zwischen den Trägheits-und Reibungskräften in der Flüssigkeit. Die Reynoldszahl errechnet sich aus

$$Re = \frac{2r \cdot v\rho_l}{\eta}, \quad (5)$$

wobei v die Relativgeschwindigkeit zwischen Partikel und Fluid bzw. die Fallgeschwindigkeit im ruhenden Fluid ist. Als charakteristische Länge ist der Partikelradius angesetzt.

Für den Fluss eines viskosen und inkompressiblen Fluids um eine Kugel gilt das Stokessche Gesetz für Reynolds-Zahlen kleiner 1,0. Dieser Grenzwert für *Re* ergibt sich in Wasser bei 20 °C für ein fallendes Teilchen mit einem Äquivalentdurchmesser von 100 µm unter der Annahme einer Feststoffdichte von 2680 kg/m³, die der von Quarz entspricht. Für größere Partikel kann die Fallgeschwindigkeit nach Rubey (1933) oder mit der allgemeinen Formel von Newtons zweitem Bewegungsgesetz bestimmt werden. Alternativ gibt Weber (1972) die folgenden beiden Formeln für die Sinkgeschwindigkeit in Abhängigkeit von der Reynolds-Zahl an

$$v = 2{,}32 \cdot (\rho_S - \rho_l) \cdot r^{1,6} \cdot \rho_l^{-0,4} \cdot \eta^{-0,6} \quad (6)$$

$$\text{für } 1 < Re < 1000 \text{ (laminare Strömung)}$$

$$v = 1{,}82 \cdot \sqrt{\left(\frac{\rho_S - \rho_l}{\rho_l}\right) \cdot r \cdot g} \quad (7)$$

$$\text{für } Re > 1000 \text{ (turbulente Strömung)}$$

Mithilfe der obigen Gleichungen kann unter bestimmten Annahmen von der Aufenthaltszeit auf den Grenzdurchmesser absetzbarer Partikel geschlossen werden.

Tab. 2 Beprobungsumfang der Wasseruntersuchung

Probenahmestelle	Anzahl
Zulauf Absetzbecken	12
Ablauf Absetzbecken	6

2.3. Feldstudie

In dieser Studie wurde die real existierende Gewässerschutzanlage auf ihre Funktion und Wirksamkeit im Betrieb überprüft. Es wurden die Zuläufe zum Absetzbecken und zum Filterbecken während eines Regenereignisses beprobt. Das Hauptziel war es, die Entfernung der AFS im Absetzbecken zu bestimmen.

Probenahme

Das primäre Interesse galt der Reinigungsleistung der Absetzstufe. Zu diesem Zweck wurde die Anlage vor und nach dem Sedimentationsbecken beprobt. Um den Verlauf der Stoffkonzentration während einzelner Niederschlagsereignisse zu beobachten, wurden Stichproben zu mehreren Zeitpunkten des Abflussgeschehens gezogen. Insgesamt wurden 18 Proben untersucht.

Die Wasserproben wurden mit einem Probenschöpfer aus Polyethylen entnommen, und zwar beim Anlagenzulauf (Zulauf zum Absetzbecken), aus dem Zulaufrohr im Ablauf des Absetzbeckens (am Verteilbauwerk VBW). Einen Überblick über die Anzahl der gezogenen Proben an den verschiedenen Entnahmepunkten gibt Tab. 2.

Berechnung der Sedimentationsleistung

Durch die Probenahmen des Zulaufs zum Absetzbecken (zu) und des Ablaufs vom Absetzbecken können Rückschlüsse auf die Wirksamkeit der Sedimentationsstufe gezogen werden. Der Wirkungsgrad wird nach Formel 9 anhand der AFS-Konzentration im Ablauf des Absetzbeckens (Ab) und der gewichteten Zulaufkonzentration ($\hat{C}_{Zu}$) errechnet.

$$\hat{C}_{zu} = \frac{\left(V_{zu,i} \cdot C_{zu,i}\right) + \left(V_{zu,i-1} \cdot C_{zu,i-1}\right) + \cdots + \left(V_{zu,1} \cdot V_{zu,1}\right)}{\sum V_{zu}} \quad (8)$$

$$\eta = \left(1 - \frac{C_{AbB,i}}{\hat{C}_{zu}}\right) \quad (9)$$

$\hat{C}_{Zu}$ ist hierbei die gewichtete AFS-Zulaufkonzentration in mg/L, C_{VAb} die AFS-Konzentration am Verteilbauwerk in mg/L, η

der Wirkungsgrad in % und i die Anzahl der Zeitschritte vor der Messung am Ablauf des Absetzbeckens (Ab).

Für die Berechnung der gewichteten Konzentration wird die AFS-Konzentration C_{Zu} der Zulaufproben nach deren Zulaufvolumen V_{Zu} gewichtet (Formel 10). Liegt nur eine Zulaufprobe vor, wird mit dieser der Wirkungsgrad berechnet. Somit wird die volumengemittelte Zulaufkonzentration mit der Ablaufkonzentration direkt verglichen.

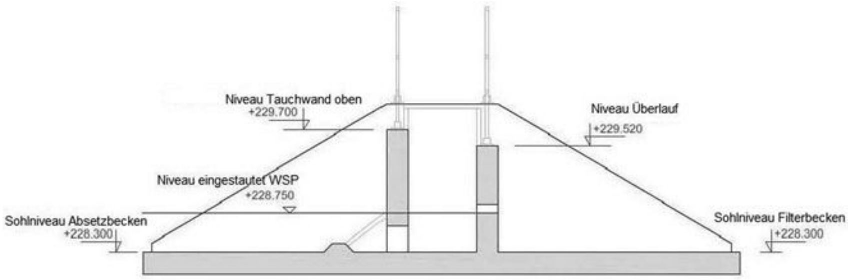

Abb. 3 Schematische Darstellung des Mönchs laut Planungsunterlagen

3. Ergebnisse

3.1. Planung und Ausführung der GSA

Beim Vergleich von Planung und Ausführung konnte anhand des konkreten Beispiels einer Gewässerschutzanlage insbesondere ein signifikanter Unterschied in der Höhenlage des Verbindungsrohres zwischen dem Absetzbecken und dem Filterbecken festgestellt werden. Die ausgeführte Variante bewirkt ein Trockenfallen des Beckens, was sich bedeutend auf die Fließgeschwindigkeit und somit die Sedimentation im Becken auswirkt. Die wesentlichen Unterschiede zwischen den Planungsunterlagen und der letztlichen Ausführung werden im Folgenden beschrieben.

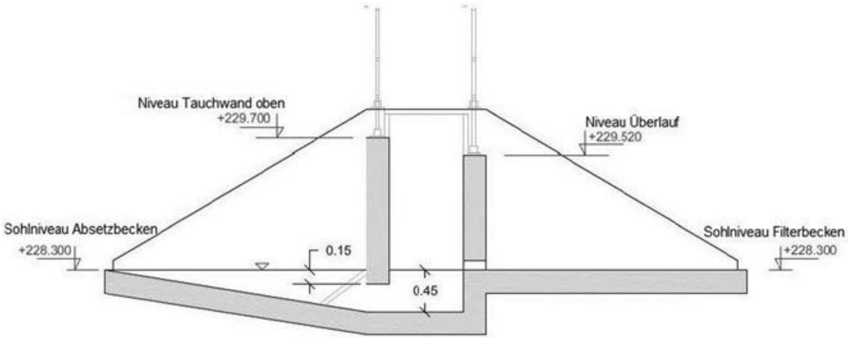

Abb. 4 Schematische Darstellung des Mönchs in ausgeführter Form (Kessler und Steurer 2012)

Geplante Variante
Das Absetzbecken der GSA wurde dauernass geplant, was bedeutet, dass das Becken dauernd eingestaut sein sollte. Der dauernasse Absetzbereich sollte der Abscheidung von absinkbaren Stoffen und Leichtstoffen dienen. Die Fläche des dauerhaft eingestauten Wasserspiegels beträgt in etwa 980 m^2. Die Tauchwandunterkante liegt 15 cm unter dem eingestauten Wasserspiegel. Vom Beckenboden bis zur Unterkante des Drosselrohres im Mönch war ein Höhenunterschied von 45 cm geplant (Abb. 3). Der vertikale Abstand zwischen dem Drosselrohr und dem Beckenboden verhindert eine Resuspension bereits abgesetzter Stoffe.

Abb. 5 Absetzbecken bei Trockenwetter (**a**) und Durchfluss am Tag der Begehung (**b**) (Aufnahmedatum: 19. März 2012)

Ausgeführte Variante
Bei der Begehung konnten zwei entscheidende Planabweichungen festgestellt werden. Im Bereich des Mönchs und dessen Zulauf wurde der Beckenboden vertieft und ein Sumpf geformt. Die Unterkante der Tauchwand des Mönchs liegt 15 cm unter dem oberen Rand des Sumpfs bzw. unter dem Sohlniveau des Absetzbeckens (Abb. 4). Das Drosselrohr, welches das Absetzbecken mit dem Filterbecken verbindet, befindet sich auf dem gleichen Niveau wie die Sohle des Absetzbeckens. Wenn kein Zufluss stattfindet, hat dies das vollständige Leerlaufen des Absetzbeckens bis auf den Sumpf zur Folge.

Durch die Abweichungen zwischen der geplanten Ausführung und dem Bestand werden das Absetzverhalten, die Speicherkapazität von Leichtstoffen und die hydraulischen Eigenschaften des Beckens maßgebend beeinflusst.

Bevorzugte Ablagerungsbereiche
Wie in Abb. 5 (a) zu erkennen ist, lagert sich praktisch kein Material im Absetzbecken ab. Die organischen Ablagerungen zwischen Einlauf und Mönch entstanden in Perioden mit sehr geringem Zufluss und werden vermutlich beim nächsten größeren Abflussereignis zum Mönch transportiert. Bei der Begehung war deutlich zu sehen, dass im Sumpf beim Mönch Sedimente und organisches Material liegen bleiben. Durch den geringen Durchflussquerschnitt im Mönch treten hier vergleichsweise hohen Fließgeschwindigkeiten in Kombination mit Turbulenzen auf. Weiters konnte bei der Begehung eine oberflächliche Ablagerung von Feinmaterial im Filterbecken festgestellt werden. Trotz des sehr geringen Zuflusses (Abb. 5 (b) während der Begehung war ein teilweiser Aufstau im Filterbecken vorzufinden.

Die Ergebnisse zeigen, dass sich im Sumpf des Mönchsbauwerks Feinsedimente ablagern. Mögliche Turbulenzen

Tab. 3 Gegenüberstellung der Maßnahmen

Beruhigungsmaßnahme	t_M in s	t_N in s	d_P in m
Keine	5	35	$1{,}9 \times 10^{-4}$
Beruhigungsrechen	11	77	$1{,}3 \times 10^{-4}$
Prallwand	21	148	$9{,}5 \times 10^{-5}$

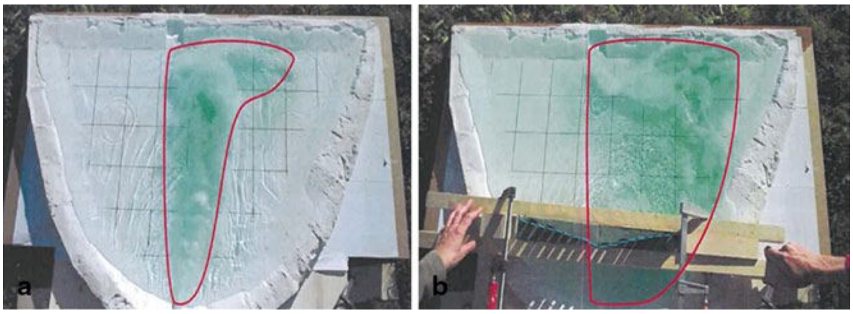

Abb. 6 **a** Modellversuch vor Einbau des Rechens; Verweildauer Farbtracer im Modell 5 s (in Natur 35 s) und **b** Modellversuch mit Rechen; Verweildauer Farbtracer im Modell 11 s (in Natur 77 s) (Kessler und Steuer 2012)

in diesem Bereich beim nächsten hohen Durchfluss machen einen Weitertransport der Sedimente in das Filterbecken sehr wahrscheinlich. Das Absetzverhalten nicht nur im Bereich des Mönchs, sondern im gesamten Becken ist bei dieser Variante als mangelhaft anzusehen. Ein Weitertransport von Material aus dem Absetzbecken ins Filterbecken ist grundsätzlich problematisch, da es zu einer verfrühten Kolmation des Filtermaterials kommt.

Maximale Speicherkapazität für Leichtstoffe

Die Speicherkapazität für Leichtstoffe im Absetzbecken ist einerseits für Notfälle (Unfall mit Leichtstoffaustritt) und andererseits für den kumulierten Eintrag von Leichtstoffen maßgeblich. Die Unterkante des Verbindungsrohres zwischen dem Absetz- und dem Filterbecken liegt auf derselben Höhe wie die Sohle des Absetzbeckens in der Nähe des Mönchs. Wenn kein Zufluss stattfindet, fällt das Absetzbecken bis auf den Sumpf im Bereich des Mönchbauwerks (siehe Abb. 3) trocken. Dies trifft also für die überwiegende Zeit zu. Wie bereits erwähnt ragt die Unterkante der Tauchwand 15 cmin den Sumpf des Mönchs. Im Folgenden wird die Eignung der Tauchwand für den Rückhalt von Leichtstoffen überprüft. Als realistisches Beispiel wird der Eintrag von Mineralöl mit einer Dichte von 820 kg/m³ betrachtet. Es wird angenommen, dass im Absetzbecken vor der Tauchwand (in Abb. 3 links) Mineralöl ansteht und hinter der Tauchwand bzw. im Mönch das verdrängte Wasser. Nachdem der hydrostatische Druck von beiden Flüssigkeitssäulen auf der Unterkante der Tauchwand derselbe sein muss, ergibt sich durch den Dichteunterschied zwischen Wasser und Mineralöl die Stauhöhe des Mineralöls bis zum Durchtritt unter der Tauchwand aus der folgenden Gleichung:

$$h_{\text{Öl}} = \frac{\rho_{\text{w}} \cdot h_{\text{w}}}{\rho_{\text{Öl}}} = \frac{\left(1000\,\text{kg}\cdot\text{m}^{-3}\right)\left(15\text{ cm}\right)}{\left(820\text{ kg}\cdot\text{m}^{-3}\right)} \quad (10)$$
$$= 18{,}3\text{ cm}$$

Daraus ergibt sich ein Höhenunterschied Δh von 3,3 cm. Der Sumpf im Bereich des Mönchs und die umgebende Sohle des Absetzbeckens werden also nur geringfügig überstaut. Bei der entsprechenden Stauhöhe beträgt die Flüssigkeitsoberfläche laut Aufnahmedaten ca. 50 m² und weist eine mittlere Einstauhöhe von 2 cm auf. Zusammen mit dem Speichervolumen im Mönch ergibt das ein maximales Speichervolumen für Leichtstoffe von ca. 3,0 m³.

3.2. Ergebnisse der Modellversuche mit Verbesserungsmaßnahmen

Bei den Modellversuchen wurde die Strömung im Absetzbecken mit und ohne Verbesserungsmaßnahmen im Bereich des Beckeneinlaufs simuliert (Kessler und Steuer 2012). Es sollte festgestellt werden, ob mit den Maßnahmen ein qualitativer Effekt erzielt werden kann. Eine detaillierte Optimierung dieser Varianten wurde nicht durchgeführt. Die Abb. 5 zeigt die Strömung im Modell ohne Einbau (Abb. 5 (a)) und mit Prallwand (Abb. 5 (b)). Im Vergleich ist deutlich zu erkennen, dass der Einbau der Prallwand eine signifikante Beruhigung der Strömung mit sich brachte. Eine ähnliche Verbesserung erreicht man mit dem Einbau eines strömungsberuhigenden Rechens (siehe Abb. 5 (a) und (b)).

In der Tab. 3 sind die Ergebnisse der Modellversuchsvarianten zusammengefasst. Die gemessene Zeit zwischen der Zugabe des Farbtracers und dem Erreichen des Beckenauslaufs ist durch t_M angegeben. Die Verweildauer in der Natur t_N wurde aus t_M berechnet. Der kleinste Durchmesser der abgesetzten Sandpartikel (Dichte 2650 kg/m³) bei einer Einstauhöhe von 1,2 m ist durch d_P gegeben. Für die Berechnung von d_P wurde Formel 4 herangezogen.

Beruhigungsrechen

Der Beruhigungsrechen besteht in der Natur aus mehreren 1 m breiten Rechenstäben mit einem Stababstand von 1,25 m, deren Anordnung im Modell in Abb. 6 (b) zu sehen ist. Die Verweildauer der Teilchen im Becken wird durch den Rechen mehr als verdoppelt und es könnten somit Teilchen abgesetzt werden, die rund 30 % kleiner sind als beim Ist-Zustand (siehe Tab. 3). Diese Teilchen müssten somit nicht vom Filterbecken aufgenommen werden. Insgesamt brachte der Einbau des strömungsberuhigenden Rechens deutliche Verbesserungen.

Prallwand

Die Prallwand des Modells wurde mit einer Breite von 7 m in der Natur ausgeführt (Kessler und Steuer 2012). Sie wurde lotrecht auf die Einströmungsrichtung des Wassers in einem Abstand von 8 m zum Einlauf angeordnet (siehe Abb. 6 (b)). Die Verweildauer der Partikel im Absetzbecken wurde mit dieser Maßnahme im Vergleich zum Ist-Zustand mehr als vervierfacht. Dies hat zur Folge, dass das kleinste absetzbare Teilchen halb so groß ist, wie das der momentan betriebenen Variante (siehe dazu Tab. 3. Aus dem Vergleich der Abb. 6 vor Einbau der Prallwand (a) und Modelversuch mit Prallwand (b) ist ersichtlich, dass der Einbau einer Prallwand eine signifikante Beruhigung der Strömung mit sich bringt. Ähnliche Verbesserungen bringt der Einbau eines strömungsberuhigenden Rechens (siehe Abb. 7 (a) und (b)).

3.3. Feldversuche

Partikelanzahl und Partikelgrößenverteilung

Beim Regenereignis vom 22.05.2012 wurden Stichproben am Zulauf und Übergabebauwerk gezogen. Von den Proben

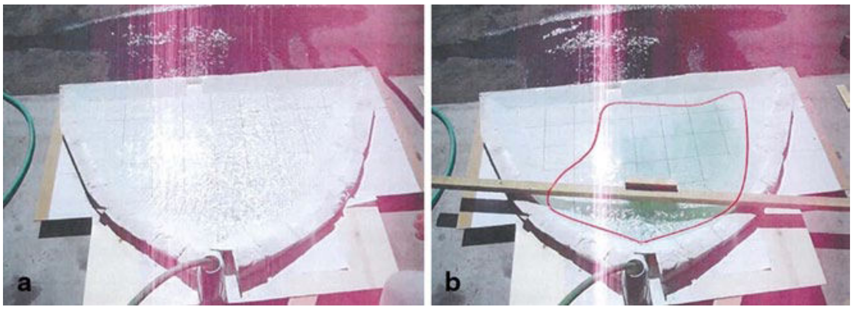

Abb. 7 Modellversuch vor Einbau der Prallwand (**a**) und Modellversuch mit Prallwand (**b**) (Kessler und Steurer 2012)

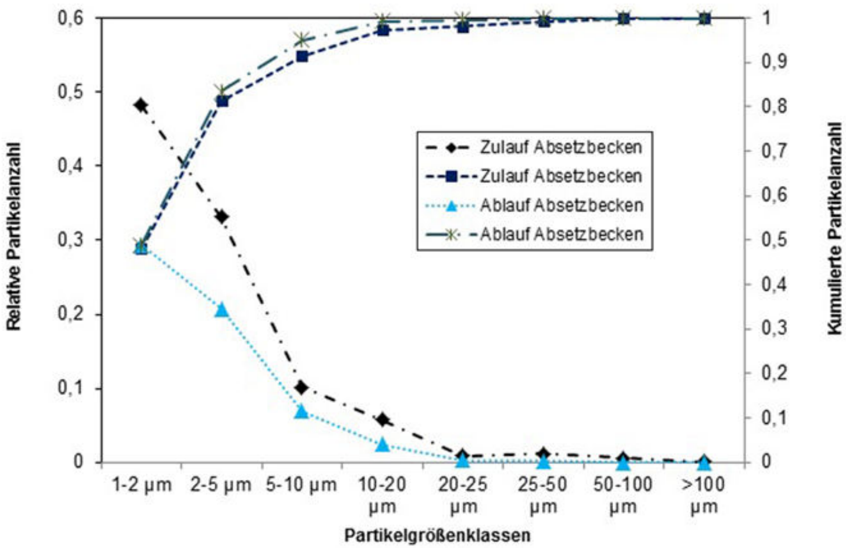

Abb. 8 Relative Partikelzahl und kumulierte relative Partikelanzahl der einzelnen Partikelgrößenklassen für die Stichprobe während des Ereignisses am 22.05.2012

wurden die AFS und die Partikelverteilung bestimmt. Die Partikelanzahl der Partikel im Größenbereich von 1 µm bis 100 µm lag zwischen 6 660/mL und 90 100/mL im Zulauf zum Absetzbecken und zwischen 4 381/mL und 23 245/mL im Ablauf. Um die Partikelgrößenverteilung quantitativ zu beschreiben, wurden nicht die Massenanteile bestimmt, sondern es wurde die Gesamtanzahl n der Partikel einer Größenklasse bzw. in einem bestimmten Größenbereich [1 µm, p] auf die Gesamtanzahl der Partikel n [1 µm, 100 µm] bezogen und als relative Partikelanzahl n_{rel} bezeichnet:

$$n_{\mathrm{rel}}[1\ \mu\mathrm{m},\ p] = \frac{n\ [1\ \mu\mathrm{m},\ p]}{n\ [1\ \mu\mathrm{m},\ 100\ \mu\mathrm{m}]} \quad (11)$$

Die relativen Anzahlen der einzelnen Partikelgrößenklassen und die kumulierten relativen Anzahlen für das Ereignis vom 22.05.2012 sind in der Abb. 8 aufgetragen.

Aus dem Vergleich der Partikelanzahlen der Stichproben (Abb. 8) wird deutlich, dass die Anzahl der Partikel im Zulauf zum Absetzbecken höher war als die Anzahl der Partikel im Ablauf vom Absetzbecken, was auf eine zumindest vorübergehende Entfernung von Feinpartikeln schließen lässt.

AFS

In der Tab. 4 sind die Ergebnisse der AFS im Zulauf zum Absetzbecken im Vergleich zu den Ablaufproben des Absetzbeckens gegenübergestellt. Die statistische Auswertung in Form von Mittelwert, Median, Maximal- und Minimalwert der Konzentrationen sind in Tab. 5 aufgeführt.

Die beobachteten AFS-Konzentrationen im Zulauf des Absetzbeckens betrugen zwischen 20 mg/L und 291 mg/L. Die mittlere AFS-Konzentration liegt bei 89 mg/L, der Median bei 53 mg/L. Die AFS-Konzentrationen im Ablauf des Absetzbeckens lieferten Werte zwischen 26 mg/L und 366 mg/L. Der Mittelwert lag bei 94 mg/L, der Median bei 45 mg/L.

Tab. 4 Vergleich die AFS-Konzentrationen im Zulauf und Ablauf des Absetzbeckens

Probenahmestellen	Datum	Zeit (hh:mm)	AFS (mg/L)
Zulauf Absetzbecken	04.05.2012	10:20	48
	22.05.2012	11:00	26
		11:50	53
		12:10	60
		12:40	54
	04.06.2012	14:20	41
	10.06.2012	18:20	40
		18:30	105
		18:40	237
		19:00	291
		19:20	98
	21.06.2012	06:00	20
Ablauf Absetzbecken	04.05.2012	10:30	43
	22.05.2012	12:00	52
		12:20	46
	04.06.2012	14:30	29
	10.06.2012	18:50	366
	21.06.2012	06:10	26

Tab. 5 Statistische Auswertung der AFS-Konzentrationen

AFS (mg/L)	Min	Max	Mittelwerte	Median
Zulauf des Absetzbeckens	20	291	89	53
Ablauf des Absetzbeckens	26	366	94	45

Durch die Neigung des Beckens wird das Wasser nicht über die gesamte Filterfläche verteilt und versickert in einem kleinen Bereich, der nur wenig mehr als 10 % der gesamten Filterfläche ausmacht. Nach einer Betriebszeit von knapp zwei Jahren sind in diesem Bereich des Beckens die Sedimentablagerungen an der Oberfläche deutlich sichtbar, ein erhöhter Schlämmkornanteil in der obersten Schicht messbar und dessen Wirkung auf die hydraulische Durchlässigkeit deutlich feststellbar. Durch den ständigen Zufluss von Fremdwasser (Dränwasser) konnte sich hier auch keine Vegetation entwickeln.

4. Schlussfolgerungen und Empfehlung

Im Laufe der Projektarbeit konnte festgestellt werden, dass die vorhandene Anlage von den vorliegenden Planungsunterlagen abweicht. Insbesondere wurden Unterschiede in der Ausführung des Übergangsbauwerkes zwischen Absetzbecken und Filterbecken (Tauchwand mit Mönch) entdeckt. Auch wenn diese Unterschiede im Vergleich zur Gesamtgröße der Anlage auf den ersten Blick marginal erscheinen mögen, beeinflussen sie die ursprünglich geplante Funktion des Absetzbeckens jedoch wesentlich. Faktisch ist die verwirklichte Variante ein trockenfallendes Absetzbecken, das nach den Kriterien eines dauernassen Beckens ausgelegt wurde. Das maximal mögliche Speichervolumen für Sedimente und Leichtstoffe wurde durch die Abweichungen drastisch reduziert.

Durch den Modellversuch konnte gezeigt werden, dass die Strömungsverteilung im Absetzbecken nicht optimal ist, da sich ein geradlinig durchströmter Teilquerschnitt vom Zulauf zum Ablauf ausbildet. Zur Abhilfe wird hier der Einbau einer Prallwand empfohlen, mit der bei geringem Aufwand ein positiver Effekt erreicht wird. Die Versuche zeigen nur Möglichkeiten auf, die allerdings noch optimiert werden sollten.

Der Sedimentationsstufe kann keine dauerhafte Wirksamkeit attestiert werden. Die sedimentierten Stoffe werden nur kurzzeitig im Becken festgesetzt. Beim Spülstoß zu Beginn eines Regenereignisses wird das zuvor abgelagerte Sediment wieder aufgewirbelt und über das Drosselrohr aus dem Becken ausgetragen. Bei starker hydraulischer Belastung kommt es zudem zu einer Kreisströmung im Sedimentationsbecken, das Sediment bleibt dadurch in Schwebe. Die fehlende Strömungsberuhigung am Zulauf, die schlecht gewählte Beckengeometrie und der Betrieb des Sedimentationsbeckens im Dauerstau können als mögliche Ursachen für die schlechte Sedimentationswirkung genannt werden. Somit erreicht die gesamte Sedimentfracht die zweite Reinigungsstufe, das Filterbecken. Das Filterbecken wird bei Zulaufvolumina unter dem 1-jährlichen 15-Minuten-Bemessungsereignis ausschließlich über die Rohrdrossel mit einem Zulauf von <37 L/s beschickt.

Die mangelnde Wirksamkeit des Absetzbeckens konnte auch durch die Messungen an der Gewässerschutzanlage im Betrieb festgestellt werden.

Jedenfalls wird aber die Retentionsfunktion von der Anlage erfüllt und die Reinigungsleistung erfolgt in diesem Fall im nachgeschalteten Filterbecken.

5. Danksagung

Einige der Untersuchungen für diesen Beitrag wurden im Rahmen des Projektes ÖNORM durchgeführt, für dessen Förderung wir uns sehr herzlich beim Lebensministerium bedanken. Weiterer Dank geht an Christian Kessler und Tobias Steurer für die Modellversuche und an DI Alexander Lins für die Feldversuche und Beprobung der Gewässerschutzanlage. ■

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6.1 Further, relevant publications related to this thesis

I. Fuerhacker, M. and Haile, T.M. (2012). Assessment of particle removal efficiency by filter media used in stormwater filtration treatment systems. [Poster]. IWA World Water Congress, Busan, South Korea, 16– 21 September 2012.

II. Fürhacker, M., Haile, T.M., Schärfinger, B., Kammerer, G., Allabashi, R., Magnat, S., et al. (2013). Entwicklung von Methoden zur Prüfung der Eignung von Substraten für die Oberflächenwasserbehandlung von Dach- und Verkehrsflächen, Fördervertrag GZ B100121, Wien, Österreich. pp. 1–240 (in German).

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Assessment of particle removal efficiency by filter media used in stormwater filtration treatment systems

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INTRODUCTION

Stormwater runoff has been identified as one of the major non-point source pollution for the deterioration of the quality of receiving waters (Fig. 1) (Clara et al., 2009). There is a new awareness about this issue in Austria and authorities and practitioners. A great portion of pollutant mass (e.g. heavy metals, PAH`s etc.) is associated with fine particles, (Fig. 2) (Xanthopoulos, 1990). Removal of particles could provide significant water-quality improvements. But a high removal rate compares with an early clogging of the filter. The objective of this study was to develop a proofing method for the selection of filter media based on pollutant removal efficiency (particle, heavy metal, mineral oil and nutrient), hydraulic performance and clogging process removal for stormwater treatment systems.

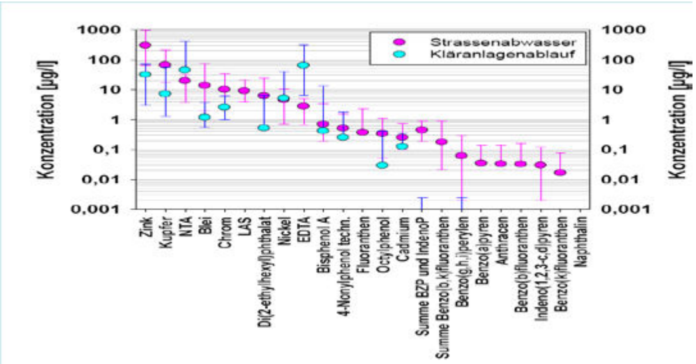

Fig. 1: Comparison of concentrations of pollutants in WWTP effluents and road runoff

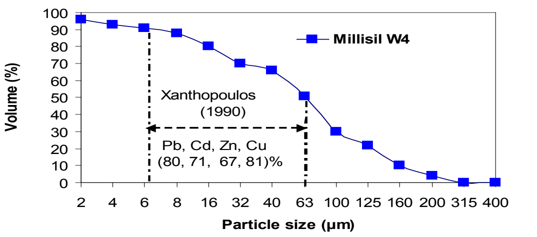

Fig. 2: Particle size distribution of Millisil W4 and particulate fraction of heavy metals as a function of particle size

MATHERIALS AND METHODS

Five filter media (see Table 1) were tested in a column experiment (Ø=100mm). Particle removal rate was evaluated by direct spreading 10 to 30 g of Millisil W4 uniformly on the surface of the column and tap water was pumped through the column. As great portion of the pollutant loads are attached to the fine particles (6 – 60 µm), Millisil W4 (d50= 65µm) was selected as a particle reference substance. For a comparison 40 L of highly polluted highway runoff water ((ATD>250,000) with TSS concentration of 825 mg/l was infiltrated through each column filled with fresh media. Remobilization of the retained particles was investigated under alternate wetting and drying conditions as follows: 15 minutes break and 5 minutes flow through. The performance of the media was monitored by turbidity, total suspended solid (TSS), particle size measurement and visual assessment. The change in infiltration rate due to the retention of applied particles was measured at the beginning and end of the experiment. Removal efficiency of PAH`s by the filter media was evaluated by analysing the concentrations in the highly polluted highway runoff and the infiltrate from each column.

Table 1. Characteristics of the filter media

Filter media	Ks (m/s)	U (d60/d10)	PSD (mm)
QS	$4.6x10^{-3}$		0.71 – 1.25
TF Ia	$2.3x10^{-3}$	2.1	1.25 – 5.6
TF Ib	$1.6x10^{-3}$	2.3	0.5 – 2.5
TF IIa	$7.4x10^{-4}$	4.8	0.063 – 5.6
TF IIb	$5.2*10^{-4}$	4.1	0.063 – 5.6
BA	$8.5*10^{-5}$	14.3	0 – 11.2
BB	$2.3*10^{-5}$	28.6	0 – 11.2

RESULTS AND DISCUSSION

The effluent particle concentration and the effect of wetting and drying on the remobilization of retained particles is presented in Figure 3. For column with unrestricted flow at a ponding depth of 2 to 5 cm and one time 10 g Millisil W4 application, the effluent TSS concentrations in the first outflow was significantly higher for all filters (163 to 407mg/l) and it decreased consistently over time. The effluent TSS concentrations were higher during every rewetting of the column following the dry period indicting the remobilization of fine particles (Fig. 3). All columns showed particle retention efficiency ≥ 80% for single dry Millisil W4 (10g) application, with higher particle retentions at continuous application (Figure 4). The removal efficiency improved consistently with increasing Millisil W4 application. The soil based substrates showed better performance with regard to particle removal rate (> 95%), however the total volume treated was much lower than that treated by the technical media. The infiltration rate decreased from 7.6 mm/min to 2.5 mm/min in BA on a passage of 100 L, whereas the BB column was completely clogged after applying 20g of the Millisil W4 and passage of 60 L. We observed that a clogging layer was formed at the interface between the applied Millisil W4 and underlying soil media irrespective of the flow rate.

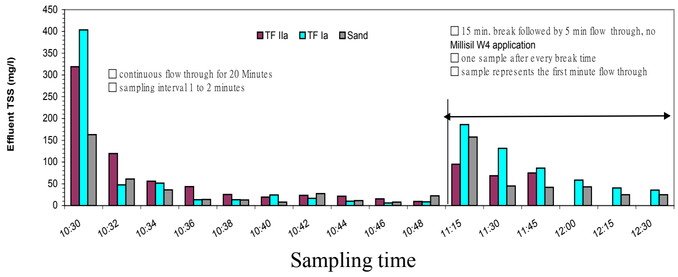

Fig. 3: TSS effluent concentration over time and effect of drying and rewetting of filter media

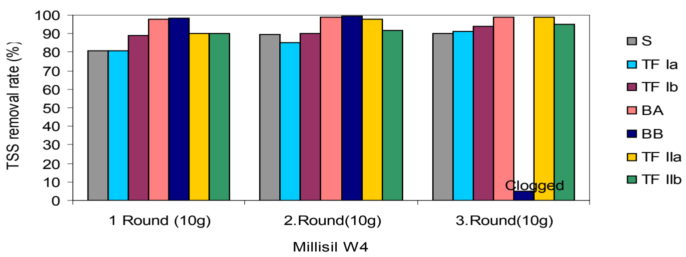

Fig. 4: Particle removal rate by the five filter media during subsequent Millisil W4 application

A comparison of particle removal rate by the filter media for direct spreading of Millisil W4 and infiltration of real highway runoff is presented in Fig. 5. Only 50% to 63% of the particles in suspension were retained by the filters as compared to >80% of the Millisil W4. Therefore, further studies are needed to understand more precisely on the application mode of particles and experiments with large volumes of highway runoff. Column effluents of the infiltrated highway runoff was also evaluated for the removal of pollutants (heavy metals, PAH`s, mineral oil). The filter media TF Ib, TF IIb and BA retained the PAHs very well, with treatment efficiencies of 80 – 90% (Fig. 6). Despite this higher removal rate, the effluent PAH concentrations were much higher than the Austrian ground water guideline (0.09 mg/l). Removal of the fine particles (<25 µm) was not possible, therefore the effluent PAH`s are attached to this particles. The concentration of mineral oil from highway runoff was 3 mg/l. Treatment performance of mineral oil from was similar the situations observed for PAH`s. The removal rate ranged 30 – 70% and the effluent concentrations ranged 0.9 – 2.1 mg/l. Column experiment with diesel showed the filter media were very effective in removing mineral oil (≥ 96%, influent 5 mg/l and effluent ≤0.2 mg/l). Diesel has short chained (volatile) compounds resulting to high removal rate and the low removal from highway runoff indicates large fraction of the mineral oil is attached to the finer particles.

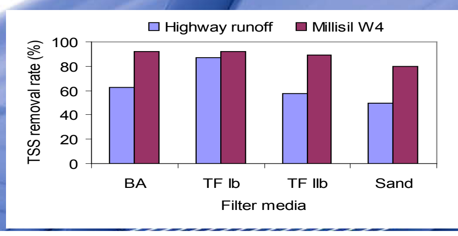

Fig. 5: Comparison of particle removal rate from real highway runoff and Millisil W4

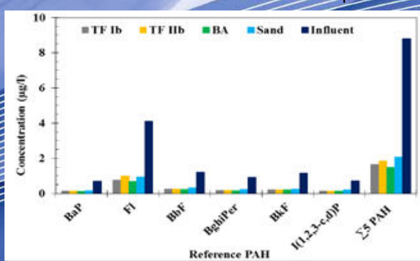

Fig. 6: PAH removal efficiency using real highway runoff

CONCLUSION

The soil based filters showed better treatment performance but the hydraulic performance reduced significantly (or clogged). Filter with high hydraulic conductivity (Ks) allows treatment of more runoff volume, but for a better pollutant (TSS) removal Ks need to be low enough. Therefore there is a trade-off between service time and treatment efficiency of the filter. The filter media with hydraulic conductivity (Ks) of ≤1,5*10-3 m/s can successfully remove the fine particles with a removal rate of >80%. Infiltration systems equipped with fine filter media and effective Ks can more effectively remove contaminants from the infiltrating stormwater runoff.

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IWA-11734: Heavy metal and PAH concentrations and toxicity in water and sediment from street runoff treatment systems

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Abstract. Water, sediment and filter media samples collected from highway and parking lot runoff treatment system were analysed for a range of physico-chemical parameters, heavy metals, mineral oil hydrocarbons (MOH), and polyaromatic hydrocarbons (PAH). Results of the water samples showed that the systems were operating well for the removal of heavy metals and PAH`s with effluent concentrations below the Austrian groundwater guideline. The sediment and filter media samples had elevated concentrations of Ba, Cr, Ni, Cu, Mn, Sr, Ti, V, and Zn indicating those heavy metals generated from road runoff are sorbed to the sediment and filter media. The concentration levels of some heavy metals were compared with sediment quality guidelines. For sediment samples concentrations of Cr, Cu, Pb and Zn were higher than the Canadian Environmental Quality Guideline (CCME) for sediment quality, classifying them as potential sources of pollution. Currently in Austria Cd, Cu, Ni, Cr, Pb, and Zn are regulated for surface and ground water quality. The accumulation of Ba, Mn, Ti and V was comparable to Zn levels indicating comparable concentrations in highway and parking lot runoff. Ba, Mn, Ti and V are not regulated in surface or ground water quality guidelines. Therefore, there is a demand to investigate the concentration levels and their associated risk for applying a guideline.

Keywords: Highway runoff, Heavy metals, Treatment system, Filter media, Sediment quality.

Introduction. Stormwater runoff from highways, roads, parking lots and bridges has been long recognised as the main non-point source pollutant input carrying significant loads of pollutants, including heavy metals, PAH´s, MOH and nutrients[1–3]. The European Water Framework Directive (WFD)[4] aiming at achieving good ecological and chemical quality of receiving water bodies by 2015 and the prevention of deterioration requires to reduce pollution from diffuse sources. Runoff treatment facilities designed as a vertical flow filter from layers of different filter media showed an effective performance for the removal of particulate and dissolved pollutants[3]. The objective of this study is to investigate the performance of treatment systems filled with reactive filter media after 7 years of service time and determine the concentrations of heavy metal, PAH`s and MOH in water and solid phase.

Material and methodology. The treatment systems (GSA-W and GSA-H) consist of underground chambers filled with technical filter media layers to 36 cm. A geotextile 300 g/m² is placed on top of the filter media to held back sediments and prevent from early clogging of the filter bed. Water, sediment and filter media were sampled in July 2012. Water samples were collected in acid washed glass bottles; representative sediment samples were obtained from the deposited sediment layer on top of the geotextile; filter media samples representing the whole filter bed were collected in plastic bags and placed at 4°C until analysis. Chemical analyses were conducted at according to standard procedures. Limit of quantification (LOQ): PAH 0.01 µg/l for individual compound and MOH 0.01 mg/l.

Results. The concentrations of pollutant in present study were lower than previous findings determined during an 18 month monitoring for GSA-H[3] and the literature values from[2], particularly for copper (Table 1).

Table 1. Concentrations of pollutants in stormwater runoff treatmen systems compared to literature and Austrian surface and ground water guideline values

Parameter	Unit	GSA-H		GSA-W		[5]		[2]	Austrian Guideline	
		Influent	Effluent	Influent	Effluent	Influent	Effluent		SWG[a]	GWG[b]
pH	-	7.7	6.2– 7.6	9	9.4	6.9 – 12.3	7.6 – 12	7.4		
TOC	mg/l	57	42 – 46	11.7	24.9	2.9 – 113	3.3 – 72	153		
TSS	mg/l	21	45	25	7	53 - 789	9 – 203			
Ba	µg/l	51.3	20.1	24.9	8.9			1.6		
Cd	µg/l	<0.1	<0.1	<0.1	<0.1	<0.1		11	0.09 – 0.26	4.5
Cr	µg/l	5	<2.5	7.7	<2.5			97	9	45
Cu	µg/l	14.2	14.8	39.1	10.8	40 – 430	19 – 129	11	1.6 – 9.3	1800
Ni	µg/l	5.3	4.8	4.8	4.2			137	20.3	18
Pb	µg/l	<4	<4		<4				7.4	9
Ti	µg/l	3	<0.3	33.9	4.8					
V	µg/l	7	3	2.9	2.4			407		
Zn	µg/l	81.3	7.7	331	20.7	55 – 1000	19 – 280	1.65	8.8 – 53	
Σ16 PAH`s	µg/l	<0.21	<0.15	0.18	<0.09	1.1 – 8.67	0.04 – 1.4	4.17		
MOH	mg/l	0.2	<0.1 – 0.3	0.2	<0.1	<0.1 – 4.4	<0.1 – 0.4	7.4		0.1

SWG[a]: Austrian surface water quality guideline; GWG[b]: Austrian groundwater quality guideline

The results showed that the systems were operating well for the removal of heavy metals. The only exception is Cu in the GSA-H treatment plant where no removal was observed indicating this metal appears to be problematic at low concentration (<30 µg/l). The effluent PAH`s were below detection limit for both sites indicating the treatment facility is operating well in removing these contaminants. The concentrations of the heavy metals in the filter media and sediment measured in this study are shown in Figure 1 and compared to literature[5] and CCME[6] (Table 2). The study reports new knowledge regarding the presence of a wide range of heavy metals with high concentrations found in the solids which are not properly regulated (for e.g. Ba, Ti, V are include in the Annex 2 of the Austrian groundwater Guideline). Thus, there is a demand to further investigate the behaviour and mode of action of those heavy metals. The concentration levels of Cr, Cu, Pb and Zn in the sediment exceeded the CCME probable effect levels (PEL)[6] in both sites. All the sediments have at least four metal concentrations higher than the guideline values, classifying them as potential sources of pollution. Although the sediments contained metal contaminants above probable effect levels, toxicity test did not reveal any effect in bioassays. Thus there was no or weak relation between contaminant concentration and toxicity.

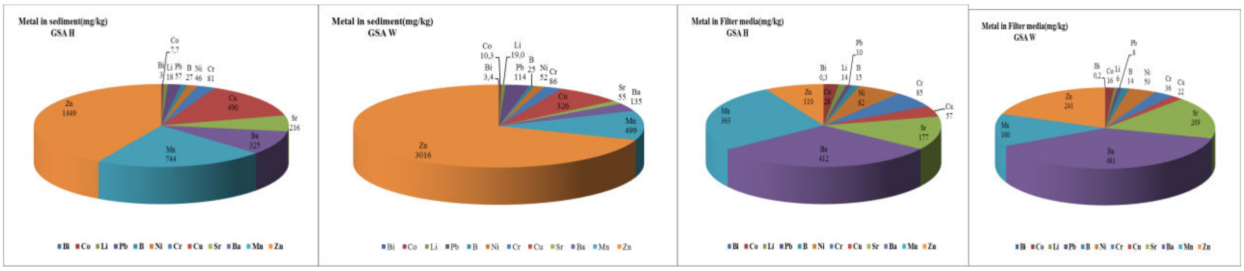

Figure 1. Heavy metal concentrations in sediment and filter media (mg/kg dw).

Table 2 Heavy metal concentrations in sediments and filter media compared to literature and guideline values.

Heavy metal	unit	GSA-H		GSA-W		5	6
		Sediment	Filter media	Sediment	Filter media		
dm	%	50	56	50	60		
B	mg/kg	27	15	25	14		
Ba	mg/kg	325	412	135	481		
Cd	mg/kg	0.6	0.14	0.49	0.05	0.1 – 4	0.6
Co	mg/kg	7.7	28	10	16		
Cr	mg/kg	***81***	85	***86***	36	10 – 97	37.3
Cu	mg/kg	***490***	57	***326***	22	9 – 190	37.7
Li	mg/kg	6.2	14	19	6		
Mn	mg/kg	744	363	499	160		
Ni	mg/kg	46	82	52	50		
Pb	mg/kg	***57***	10	***114***	8	15 – 120	35
Sr	mg/kg	216	177	55	209		
Ti	mg/kg	475		589			
V	mg/kg	37		44			
Zn	mg/kg	***1149***	110	***3016***	241		123

Conclusion. The concentrations of heavy metals in effluents were generally lower than the Austrian groundwater quality guideline, thus the treatment systems were operating well. The metal concentrations of the solid phase samples indicate additional potentials for toxic problems. Nevertheless, this question needs further investigation and efforts for risk assessment.

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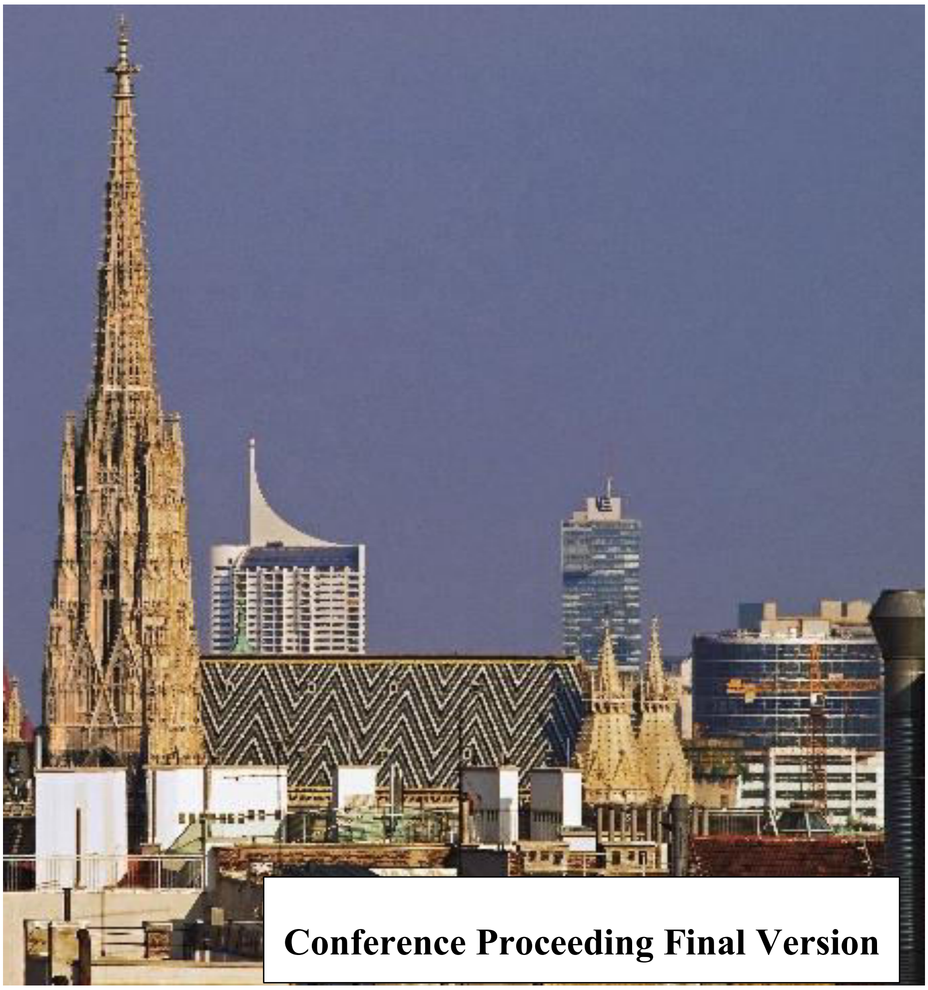

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Oral presentation

Micropollutants and genotoxicity in sediments from stormwater filtration systems

(Abstract ID: 3754489)

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Abstract: Highway and parking lot runoff can induce various negative effects in biocoenoses by a variety of heavy metals and organic micropollutants emitted from vehicles and traffic-related activities. Nevertheless, the sampling of runoff filtration systems is difficult as the duration and intensity of the local rainfall cannot be predicted. The aim of the study was to determine adverse effects of roadway runoff by investigating the genotoxic effects (Salmonella/microsome (TA98, TA100s and YG1024) and Tradescantia micronucleus assay (Trad-MN)) of the sediments deposited at the surface of stormwater runoff filtration systems. Moreover, the thresholds in Sediment Quality Guidelines (SQGs) have been used to evaluate the potential adverse effects of heavy metals and polycyclic aromatic hydrocarbons (PAHs) on sediment-dwelling organisms. Results showed that the micropollutants generated from the highway and parking lot surface are accumulated in the particle layer. From an ecotoxicological point of view, the concentrations of heavy metals (Cr, Cu, Ni, Pb and Zn) and single PAHs exceeded both the threshold/probable effect level (TEL/PEL) for adverse biological effects. Sediment samples induced some mutagenic/genotoxic effect. The bioassays proved to be a suitable tool to detect mutagenic and genotoxic activity of sediments contaminated with anthropogenic pollutants from vehicular emission and therefore are useful to predict the activity of roadway runoff.

Keywords: stormwater; sediments; genotoxicity

In: Fürhacker et al. (eds.), 2017. Micropol & Ecohazard Conference 2017, 10th IWA Specialized Group on Assessment and Control of Hazardous Substances in Water (ACHSW), Vienna, Austria, Sept 17-21, 2017.

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Publications

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