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Engineered nanomaterials in plastic products

A material flow analysis using the example of vehicle tires containing carbon black

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Declaration on oath

I declare that I have independently produced this work and have not used any other aids than those indicated. I have referenced all phrasings and ideas taken from unprinted sources, printed literature or from the internet in accordance with the guidelines of good scientific practice and marked them with footnotes or exact reference in the text.

This work has not yet been submitted in the same or a similar form to any other examiner.

I am aware that any violation constitutes scientific misconduct and will be prosecuted (“use of unauthorized aids”) and may result in further legal action.

Stefanie Prenner

Vienna, 20th of March 2020

Abstract

Due to the demand for “advanced” plastics, the benefits of engineered nanomaterials (ENMs) are increasingly recognized by research and industry. Hence, the number of such polymer nanocomposites (PNCs) available at household level is continually rising. Since ENMs may also pose hazards to humans and the environment, it is crucial to know about their behaviour and fate in the environment. This work aims at generating an overview of the current state of knowledge in the field of PNCs. Furthermore, a material flow analysis (MFA) was carried out on a product example, which had been selected based on the availability of product information. The literature review shows, that Carbon Nanotubes (CNTs), nanoclay, TiO_2, Ag, SiO_2 and CB are the most frequently used ENMs in plastics. However, some of these PNC products refer to a technology readiness level (TRL) of only 1-4 and have not been released to the market yet. In addition, possible release mechanisms of ENMs from plastics are summarized, of which matrix degradation, diffusion, desorption or dissolution are the most relevant. However, only a few experimental studies have been able to analytically proof a release of single nanoscale particles. The majority of the released fragments consist of plastic debris with incorporated or associated ENMs. For this study, a nano-specific MFA on the level of goods as well as on the substance level was conducted using the example of vehicle tires containing carbon black (CB), because this ENM-type is already present on the market. Since data availability is limited, a data uncertainty analysis has been included to characterize input data in a transparent manner. The results indicate that in Austria in 2018, micro- and nanoscale particles were released during several life cycle stages: Especially during the use phase, a large quantity of micro- and nanoplastics was emitted (15 550 t), particularly into the air (8 270 t), because of vehicle tire abrasion. Based on literature data it can be stated that the majority of these released particles is in the size range between 10 nm to several 100 µm and mainly consists of rubber associated with ENMs and other additives or impurities. A predicted small fraction of 0.2 t will be released as free nanofillers. Overall, more release studies are necessary to improve and validate the emission and material flow models. Furthermore, there is an urgent need for appropriate measures to reduce the ultrafine dust load.

Kurzfassung

Aufgrund der steigenden Nachfrage nach verbesserten Kunststoffprodukten wird der vorteilhafte Einsatz von technisch hergestellten Nanomaterialien (ENMs) zunehmend von Forschung und Industrie erkannt. Daher steigt auch die Anzahl der am Markt erhältlichen Polymer-Nanokomposite (PNCs) kontinuierlich an. Da ENMs aber auch eine Gefahr für Mensch und Umwelt darstellen können, ist es wichtig über ihr Verhalten und ihren Verbleib in der Umwelt Bescheid zu wissen. Ziel dieser Arbeit ist es, einen Überblick über den aktuellen Wissensstand im Bereich von PNCs zu geben. Darüber hinaus wurde eine Materialflussanalyse (MFA) an einem Produktbeispiel durchgeführt, das aufgrund der verfügbaren Produktinformationen ausgewählt wurde. Die Literaturanalyse zeigte, dass Kohlenstoff-Nanoröhrchen (CNTs), Nano-Ton, TiO_2, Ag, SiO_2 und Industrieruß (CB) die derzeit am häufigsten verwendeten ENMs in Kunststoffen sind. Einige dieser PNC-Produkte weisen jedoch erst einen Technology Readiness Level (TRL) von 1-4 auf und sind deshalb noch nicht am Markt erhältlich. Weiters wurden mögliche Freisetzungsmechanismen von ENMs aus Kunststoffen zusammengefasst, wobei Matrixabbau, Diffusion, Desorption sowie Auflösung die relevantesten sind. Allerdings konnten nur wenige experimentelle Studien eine Freisetzung einzelner nanoskaliger Partikel analytisch nachweisen. Die Mehrheit der freigesetzten Fragmente besteht aus Kunststoffresten mit eingebundenen oder assoziierten ENMs. Für diese Arbeit wurde eine nanospezifische MFA sowohl auf Güterebene als auch auf Substanzebene für CB-haltige Fahrzeugreifen durchgeführt, da dieser ENM-Typ bereits am Markt erhältlich ist. Aufgrund begrenzter Datenverfügbarkeit ist eine Datenunsicherheitsanalyse enthalten. Die Ergebnisse zeigen, dass in Österreich im Jahr 2018 in verschiedenen Lebenszyklusphasen mikro- und nanoskaliger Reifenabrieb freigesetzt wurde. Vor allem während der Nutzungsphase wurde eine große Menge an Mikro- und Nanokunststoffen (15 550 t) in Form von Reifenabrieb, insbesondere in die Luft (8 270 t), emittiert. Literaturwerte geben an, dass die Mehrzahl dieser freigesetzten Partikel im Größenbereich von 10 nm bis mehrere 100 µm liegen und hauptsächlich aus Kunststoff in Verbindung mit ENMs und anderen Additiven oder Verunreinigungen bestehen. Es wurde geschätzt, dass ein Anteil von 0,2 t in Form von freien Nanofüllstoffen freigesetzt wird. Generell sind weitere Freisetzungsstudien notwendig, um Emissions- oder Materialflussmodelle zu verbessern und validieren. Darüber hinaus besteht ein dringender Bedarf an geeigneten Maßnahmen zur Reduktion der Feinstaubbelastung.

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Abbreviations

AAS	Atomic absorption spectroscopy
AF4	Asymmetric flow field-flow fractionation
AFM	Atomic force microscopy
Ag	Silver
Ag_2S	Silver sulphide
AgCl	Silver chloride
Al	Aluminium
$Al(OH)_3$	Aluminium hydroxide
Al_2O_3	Aluminium oxide
Au	Gold
BMLFUW	Bundesministerium für Land- und Forstwirtschaft, Umwelt und Wasserwirtschaft
BMNT	Bundesministerium für Nachhaltigkeit und Tourismus
CB	Carbon black
CdTe	Cadmium telluride
CeO_2	Cerium dioxide
CLP	Classification, labelling and packaging of substances and mixtures
CNF	Carbon nanofibers
CNR	Cellulose nano-reinforcements
CNT	Carbon nanotubes
Cu	Copper
CuO	Copper oxide
CV	Coefficients of variation
CVD	Chemical vapour deposition
DM	Dry matter
DPMFA	Dynamic probabilistic material flow analysis
EC	European Commission
EFM	Environmental fate model
EFSA	European Food and Safety Authority
EHS	Environmental health and safety
ENM	Engineered nanomaterial
EOL	End-of-life
EU	European Union
EVA	Ethylene-vinylacetate copolymer
Fe_2O_3	Iron(III) oxide
FG	Flake graphite
FINE	Forecasting the Impacts of Nanomaterials in the Environment
FMPS	Fast mobility particle sizer
GaN	Gallium nitride
GO	Graphite oxide
HDPE	High-density polyethylene
IARC	International Agency for Research on Cancer
ICP-AES	Inductively coupled plasma atomic emission spectroscopy
IIR	Butyl rubber
ITO	Indium tin oxide

LDPE	Low-density polyethylene
LED	Light emitting diode
MBWTP	Mechanical-biological waste treatment plant
MgF_2	Magnesium fluoride
MgO	Magnesium oxide
MMT	Montmorillonite
WIP	Waste incineration plant
MWCNT	Multi-wall carbon nanotubes
n.i.	no information
NH_3	Ammonia
NIOSH	National Institute for Occupational Safety and Health
NM	Nanomaterial
nm	Nanometre
NP	Nanoparticles
PA	Polyamide
PAH	Polycyclic aromatic hydrocarbons
PE	Polyethylene
PEC	Predicted environmental concentration
PET	Polyethylene terephthalate
PIB	Polyisobutylene
PM	Particulate matter
PMFA	Probabilistic material flow analysis
PNC	Polymer nano-composites
PP	Polypropylene
PS	Polystyrene
PUR	Polyurethane
PVC	Polyvinyl chloride
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals
Sb	Antimony
SBR	Styrene-butadiene rubber
Sb_xO_x	Oxides of antimony
SCF	Scientific Committee on Food
SEM	Scanning electron microscopy
Si	Silicon
SIMS	Secondary ion mass spectrometry
SiO_2	Silicon dioxide
SP-ICP-MS	Single particle inductively coupled plasma mass spectrometry
SWCNT	Single-wall carbon nanotubes
Ta_2O_5	Tantalum pentoxide
TEM	Transmission electron microscopy
TC	Transfer coefficient
TiCl	Titanium chloride
TiN	Titanium nitride
TiO_2	Titanium dioxide
TOC	Total organic carbon
TRL	Technology readiness level
TSP	Total suspended particles

UBA	Umweltbundesamt
UV	Ultraviolet
WHO	World Health Organization
WPC	Wood plastic composites
wt%	Weight percent
WTP	Waste treatment plant
WWTP	Waste water treatment plant
XPS	X-ray photoelectron spectroscopy
Zn_2SiO_4	Zinc silicate
ZnO	Zinc oxide

1. Introduction

Polymer nanotechnology is used to design and produce advanced plastic products with enhanced properties. Up to now, various products have been developed that contain different engineered nanomaterial (ENM) types and thus provide different improved characteristics. However, these incorporated or associated ENMs may be released from their original polymeric matrix due to several release mechanisms (Duncan, 2015; Duncan and Pillai, 2015). Since released ENMs may pose risks to humans and the environment, it is important to know about their release behaviour along different product life cycles. For this purpose, material flow analyses (MFAs) can be used to predict and identify release hotspots and to generate a basic knowledge of different processes and flows of certain systems under consideration. Additionally, sinks can be located with MFAs and addressed with specific measures in the event of an increased risk of exposure (Baccini and Brunner, 1991).

As vehicle tires with incorporated carbon black (CB) are in use for many years and due to the recent findings of CB particles in the foetal side of human placenta (Bové et al., 2019), CB-containing tires are chosen as a representative PNC for the performance of an MFA. This MFA shall give a more detailed insight into the behaviour of CB along the whole product life cycle of vehicle tires. In addition, the analyzed ENM flows and release hot spots may be useful for environmental policy decision as well as forming the basis for further calculations and modelling.

In the following subchapters important terms for this work will be defined, the regulatory framework for ENMs will be discussed and the research objectives and scope will be presented.

1.1 Definition of terms

In order to assure a uniform use of terms and to clearly define these in advance, all important terms used in this work will be defined in the following subchapters.

1.1.1 Terms of nanotechnology

Nanomaterials (NMs) can occur naturally in the environment like for example volcanic ash, DNA or aerosols. However, due to the various product enhancing characteristics of NMs they are also designed artificially for specific purposes like for instance CB for the use in vehicle tires. In the latter case one speaks of ENMs (ISO/TS 80004-1:2015). In this work, the focus will be on ENMs.

NMs have at least one dimension in the nanoscale (1-100 nm). There are two subcategories of NMs: nano objects (external dimension in the nanoscale) and nanostructured materials (internal or surface structure in the nanoscale). The former is, again, divided into three subcategories which are all primary particles and depend on the number of the external dimensions in the nanoscale. Nanoplates have one, nanofibers have two and spherical nanoparticles (NPs) have three external dimensions in the nanoscale. These primary particles may also form aggregates or agglomerates (BPF, 2009; ISO/TS 80004-1:2015; Steinbach, s.a.a).

According to Greßler and Gazsó (2014) ***nanoproducts*** are products (for consumers, industry, trade and medical sector) which are NMs or contain ENMs according to the manufacturers specifications. Moreover, products whose description or promotion establishes a reference to nanotechnology are also *nanoproducts*.

Nanowaste can be generated along the whole life cycle of a product (Boldrin et al., 2014). However, only when ENMs come into contact with solid waste and can be collected separately one speaks of *nanowaste* (Greßler et al., 2014). Thus *nanowaste* can be categorized into by-product waste (intentional), nano-contaminated waste (unintentional) and end-of-life (EOL) nanoproducts (intentional) (Boldrin et al., 2014). In this context, it is important to make a conceptual distinction to ***nano-pollution***. *Nano-pollution* consists of unintentionally released ENMs during different stages of a product life cycle which cannot be collected (Boldrin et al., 2014).

1.1.2 Terms of polymer sciences

Plastics are made of polymers which consist of macromolecules and usually are derived from petrochemicals. However, there are also natural plastics which are acquired from nature, like natural rubbers or chitosan. Because of their physical properties, plastics are divided into three different groups: *thermoplastics, thermosets and elastomers*. *Thermoplastics* consist of linear or branched macromolecules which can be deformed when heated. This process is reversible. *Thermosets* consist of monomers or oligomers which are also deformed when heated, but this process is non-reversible. *Elastomers* consist of macromolecules which are widely bonded to each other and have a rubber-like behaviour (Ehrenstein, 2011).

ENMs which are fully incorporated into a polymer matrix form so called ***polymer nano-composites*** (PNCs) (Bott et al., 2014). Nanocomposites in general are multiphase materials which consist of two or more components. These components include a matrix (continuous phase) and a nano-dimensional phase (discontinuous phase) (Azeredo et al., 2011). PNCs are produced by the dispersion of an inert nanoscale filler through a polymeric matrix (Adame and Beall, 2009; Duncan, 2011). These filler materials include a wide variety of ENMs and provide enhanced material characteristics for different products (for more information see also chapter 3.1 Manufacturing of ENMs and PNCs). The use of ENMs in polymeric matrices is subsumed under the term polymer nanotechnology (Silvestre et al., 2011).

1.1.3 Terms related to MFA

MFAs are used to investigate and illustrate different processes, flows and stocks of resources within a predefined system (Baccini and Brunner, 1991; Brunner and Rechberger, 2004; Laner et al., 2016).

A ***substance*** according to chemical sciences is defined as any (chemical) element or (chemical) compound which is consisting of homogenous units. Every substance is unique and has an identical constitution by which it can be characterized (Sax and Hawley, 1987; ÖNORM. S 2096-1, 2005). The (chemical) elements are atoms and (chemical) compounds are molecules (Brunner and Rechberger, 2004).

Goods are a composition of several substances and are specified as important economic entities to which a positive or a negative economic value can be ascribed. Only a few goods don't have such an economic value. With respect to MFA the term "goods" is only used for material goods (Brunner and Rechberger, 2004; ÖNORM. S 2096-1, 2005).

In the context of MFA the term ***material*** is used as an umbrella term for *substances* and *goods* (Brunner and Rechberger, 2004; ÖNORM. S 2096-1, 2005).

The term ***process*** is divided into three different meanings: *transformation, transport* and *storage of materials* (Baccini and Brunner, 1991). Different materials (substances or goods) can be *transformed* qualitative, quantitative or/and in its chemical composition on different levels throughout the whole life cycle of the material in question (Baccini and Brunner, 1991; Brunner and Rechberger, 2004). *Transportation* refers to the movement of materials over a certain distance without transformation (Brunner and Rechberger, 2004). *Storage of materials*, as the third type of process, is used for quantitative description of materials. All materials stored in a process are defined as the stock of materials. The mass of the stock with the physical unit kilogram (kg) and the rate of change (accumulation or depletion) of the stock per unit time are two important parameters. Stocks, where materials have a long residence time are called sinks (Baccini and Brunner, 1991; Brunner and Rechberger, 2004).

A distinction has to be made between the terms ***flows*** and ***fluxes***. *Flows* are defined as a "mass flow rate" like for example kg/sec or t/year. In contrast, *fluxes* are defined as a "flow per cross section" like for instance kg/(sec*m^2) or ENMs/(m^2*year). Hence, a flux refers to a specific flow and thus is better suited for comparison amongst different processes and systems (Brunner and Rechberger, 2004; ÖNORM. S 2096-1, 2005).

The partitioning of a substance in a process is described by ***transfer coefficients*** *(TCs)*. These coefficients are substance specific and specified for each process output good. The sum of all these output TCs has to be 1. Due to the possible dependency of these TCs on variables like input compositions and process conditions (e.g. temperature, humidity) they can be constant or non-constant (Brunner and Rechberger, 2004; ÖNORM. S 2096-1, 2005).

The whole system under consideration must be delimited by ***system boundaries***. An MFA system is defined by a group of interacting elements that consist of one or more processes which are connected through flows. Moreover, such a system is defined in time and space. The temporal boundaries (i.e. time span) are defined according to the given problem. Spatial system boundaries are normally set in accordance with the geographical area in which the process of interest is located. There are "open systems" which interact with their surrounding area and "closed systems" which are isolated from external influences (Baccini and Brunner, 1991; Brunner and Rechberger, 2004).

1.2 Regulatory framework

There are many different legal definitions for ENMs. These definitions are only legally binding within their specific regulatory context (Rauscher et al., 2017). Within this work the definition from ISO/TS 80004-1:2015 “NMs have at least one dimension in the nanoscale (1-100 nm)” is used (for more information see section 1.1).

On the level of the European Union (EU) there are some regulations with relevance to ENMs. The regulations of Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) and the classification, labelling and packaging of substances and mixtures (CLP) can be seen as general regulations for ENMs (REACH, 1907/2006; CLP, 1272/2008). Within these regulations the use of ENMs is covered implicitly, because it applies to all chemicals/substances in the EU regardless of their size, shape or physical state (Rauscher et al., 2017). The focus of REACH lies on the tonnage of a substance that is manufactured or produced in one year. Substances with a tonnage over 1 t per year have to be registered and substances with a tonnage over 10 t per year have to undergo a safety assessment (REACH, 1907/2006). REACH thus includes ENMs of high volume like for example titanium dioxide (TiO_2) or silicone dioxide (SiO_2) (Laux et al., 2018). The CLP regulation addresses the classification of chemicals and their labelling on product packaging (CLP, 1272/2008).

According specific product regulations for ENMs there are three different product categories under regulation. One of these categories applies to *cosmetic products* with the underlying cosmetic products regulation No 1223/2009 of the European Parliament and of the Council. This regulation requires a pre-market notification of ENM-containing products including the size of the used particles, an estimation of the used quantity of the ENM, chemical and physical properties, a toxicological profile, safety data of the ENM and potential exposure conditions (1223/2009). Moreover, there is a statutory declaration obligation of the components contained in the product on the packaging in accordance with paragraph 46 of the cosmetic regulation. This should lead to more transparency for the consumer (1223/2009).

Another specific product regulation is that for *biocidal products*, No 528/2012 of the European Parliament and of the Council. This regulation requires a risk assessment for the authorization of ENM-containing products (528/2012). Furthermore, there is also a statutory declaration obligation in accordance with article 58 paragraph 3d of the biocidal products regulation for the contained ENMs on the label of the product (528/2012).

The third category applies to the category of *food*. This is the category with the most regulations concerning the use of ENMs. There are the novel food regulation No 2015/2283, plastic food contact materials regulation No 10/2011, active and intelligent food contact materials regulation No 450/2009, food additives regulation No 1333/2008 and the provision of food information to consumers regulation No 1169/2011.

In the context of regulations for the food sector, the European Food and Safety Authority (EFSA) plays an important role. There is a requirement in the novel food regulation that addresses the verification of the use of the most up-to-date methods for the assessment of the safety of ENMs through EFSA (2015/2283). Moreover, all plastic food contact materials as well as active and intelligent food contact materials in the EU are subjected to a safety assessment of the EFSA (450/2009). Additionally, ENMs that are incorporated or associated in food contact materials and used in the

EU are explicitly authorized in Annex I of the plastic food contact materials regulation including some limitations concerning the mass and size of these ENMs (10/2011). Within the food additives regulation one main goal is to achieve a high level of protection of human life (1333/2008). The provision of food information to consumers regulation has consequences for ENMs with regard to their indication on the label of all ENMs used in a product (1169/2011).

For medical devices, a proposal of regulation exists which deals with ENM-containing products that can release ENMs into the user's body. Furthermore, this proposal contains a mandatory indication on the label that there are incorporated or associated ENMs in the product. This proposal regulation also requires a separate safety assessment of the ENMs used in a specific medical product (Rauscher et al., 2017).

Not all the above-mentioned regulations are relevant for this thesis. With regard to all plastic products available at household level, the regulations of REACH, 1907/2006, CLP, 1272/2008, biocidal products No. 528/2012, active and intelligent food contact materials No 450/2009 and plastic food contact materials No 10/2011 have to be considered. For the specific case of vehicle tires analyzed in the applied MFA, only the regulations of REACH, 1907/2006 and CLP, 1272/2008 are relevant.

1.3 Research objectives and scope

In the sector of nanotechnology the production of PNCs has become an emerging field of interest in the current research and development activities (Paul and Robeson, 2008). ENMs have been used in various plastic products during the last few years because they provide several advantageous effects. Nevertheless, ENMs may as well pose hazards to humans and the environment when they are released from their original polymer matrix. Especially on the level of household products humans are most likely to be exposed to ENMs. These facts and considerations have led to the first two research questions of this thesis:

1. Which ENM-types are used in plastic products for household use?
2. What mechanisms lead to the release of ENMs from the polymer matrix?

An MFA will be used to identify sources of ENM release (“emission hotspots”) along the product life cycle. Moreover, ENM flows are calculated and sinks will be located within the performance of an MFA (Baccini and Brunner, 1991; Brunner and Rechberger, 2004). For this reason, this master's thesis also seeks to answer two more research questions:

3. Which different methodological approaches exist for MFAs on ENMs?
4. Which additive is most frequently used in PNCs and therefore probably causes the highest emissions?

The answer to the last research question is intended to provide more detailed insights into the life cycle of one specific PNC and its incorporated or associated ENMs. For this purpose, a case study of an ENM-containing plastic product will be carried out using an MFA.

2. Materials and methods

This thesis encompasses a theoretical and an empirical part. The theoretical part is based on already existing literature and serves as a basis for the empirical part. The empirical part consists of an MFA on a plastic product with incorporated ENMs.

2.1 Data collection of ENM-containing plastic products

In the field of ENM-containing plastic products, an extensive literature research for scientific articles about ENM-containing plastic products was performed. In this context the term “plastic product” refers to household products (e.g. paints, electrical devices, packaging) but also to products from the medical and the automotive industry, which can be purchased and used by households (e.g. vehicle tires, bandages, gloves).

In order to find relevant scientific articles, a systematic literature research was carried out using different online databases (BOKU Lit-Search, Scopus and Google Scholar) and directly on homepages of “NanoImpact” (Elsevier B.V.) and “Environmental Sciences: Nano” (Royal Society of Chemistry). Relevance in this context means at least a specification of the plastic product (category or product specification) *and* the indication of a specific embedded ENM within the respective scientific article. For this literature research the keywords “nanomaterial”, “plastic”, “polymer”, “embedded”, “polymer nano-composite”, “Kunststoffprodukte”, “Kunststoff” and “Nanomaterial” were used. These keywords were combined in various ways. After this first systematic research in online databases there was another research according to the snowball principle based on the previously found literature.

The resulting relevant articles were then analyzed in detail quantitatively and qualitatively and the findings were added to a table (c.f. Appendix 1). The primary focus is on the analysis of the frequency of nomination of different plastic products and their embedded ENMs. Furthermore, the products were divided into different categories. These product categories are similar to the categories which are used in the screened articles. Moreover, if available, examples for producing companies and product trade names were added to the table. For the further establishment of the table, some of the plastic products were also categorized according to the previously mentioned different plastic groups (thermoplastics, thermosets and elastomers) and moreover specified with regard to their source-based name (e.g. polyethylene (PE), polystyrene (PS)). The source-based name has also to be seen as an example. Furthermore, the mass (weight percent (wt%) or concentration), size of the primary particles, aggregate size and agglomerate size were listed if this information was available in the respective article. Since ENMS are embedded into plastic products due to their enhancing characteristics, these improved features were listed, as well. Another important fact refers to the Commission Regulation No 10/2011 for plastic products intended to come into contact with food. This information affects only products from the category *food and beverage packaging.* It includes information on whether the ENM used is allowed by the European Commission and also whether there is a limitation concerning its mass or size. All these findings are listed in Appendix 1.

After this literature research and the following establishment of Appendix 1, there was an evaluation of all the data found. This evaluation included an analysis of all the different types of the ENMs embedded in plastic products as well as of the distinct ENM-containing products attributed to different categories. Additionally, there was a more detailed analysis of specific product categories and their corresponding plastic products. However, this analysis was only performed for the four product categories with the most ENM-containing products.

It is important to mention that, in the evaluation, multiple responses were taken into account only once. For example, if within one scientific article different sporting racquets (e.g. tennis, badminton), which all include carbon nanotubes (CNTs), are mentioned separately, these findings were only counted as one sporting equipment that includes CNTs. Moreover, plastic products currently under development (marked green in Appendix 1), were not included in the evaluation. Products were only highlighted as future applications if this is explicitly mentioned in the respective article and if there is no evidence for an already existing product.

2.2 Overview of ENM release mechanisms from PNCs

The establishment of an overview of ENM release mechanisms from PNCs was based on recently published literature. In order to find relevant information, a literature research in different online data bases (BOKU Lit-Search, Scopus and Google Scholar) was carried out. For this purpose, the keywords “nanomaterial”, “release”, “mechanism”, “polymer”, “polymer nano-composite” and “plastic” were used. These keywords were combined in various ways. After this first systematic research in online databases, there was another research according to the snowball principle based on the previously found literature mainly focussing on reviews. Additional relevant data with regard to the empirical part were collected. To be considered relevant, the data had to refer to the release of specific ENMs, release rates along distinct product life cycle stages of ENM-containing plastic products, composition of released particles as well as TCs that are needed for the MFA.

2.3 Methodology for the MFA

One specific ENM-containing plastic product had been chosen, for which the MFA was conducted. Three different selection criteria have been applied: (1) The results of the data evaluation of ENM-containing plastic products were taken into account. From this data, one of the most frequently nominated ENMs with its corresponding specific host product was used for the MFA. (2) The density/availability of information of published data about ENMs and their hosting plastic products that are of relevance for a further MFA (e.g. release mechanisms, information on the TC, used mass) was taken into account. (3) It was considered whether MFAs have already been carried out for a specific ENM-containing plastic product or not. This criterion relies on a research in different online databases (BOKU Lit-Search, Scopus and Google Scholar). For this purpose, the terms “material flow analysis”, the name of the respective ENM (e.g. “CNT”) and the product specification (e.g. “tennis racquet”) were used. The goal was, to identify an ENM-containing plastic product for which no MFA has been performed so far.

Within this thesis the general terms and aims defined by Baccini and Brunner (1991), Brunner and Rechberger (2004) and the Austrian Standard S 2096 - 1+2 (ÖNORM. S

2096-2, 2005; ÖNORM. S 2096-1, 2005) were used. These terms have already been briefly explained in section 1 and 1.1. For more detailed information, please refer to these sections and to the references listed above. Besides these general definitions and aims, various different model types and uncertainty considerations for the application of an MFA exist. These different methodological approaches were addressed and summarized within this thesis. With regard to ENM specific MFA, an overview on different model types was provided. The establishment of this overview was mainly based on an evaluation of environmental exposure models performed by Nowack (2017). Nevertheless, some other references found by the snowball principle were also taken into account.

In this thesis, the model type of static probabilistic material flow analysis (PMFA) based on Gottschalk et al. (2010) was applied. The underlying uncertainty consideration was based on the modelling approach of Laner et al. (2016). All data used for the MFA was assessed with the proposed approach for expert estimations. In sections 4.2.4 and 4.1.2.6, PMFA as well as the uncertainty modelling approach will be explained in more detail.

The MFA was performed on two distinct levels: on the "*substance level*" as well as on the *"level of goods".* On the s*ubstance level*, the software tool "STAN" was used. STAN builds a graphical model for the system under consideration with predefined components (e.g. processes, flows). Moreover, with calculation algorithms that use mathematical statistical tools, data uncertainties can be included (TU Wien, 2019). Substances in this context refer to "CB". On the *level of goods,* the software tool "e!Sankey" was used to perform an MFA on vehicle tires in Austria in 2018. This software tool allows to build graphical models with predefined components for a certain system under consideration (IFU, s.a.). Uncertainties are not included by "e!Sankey" but can be seen in Appendix 4. "Vehicle tires" in this context refers to the share of the used elastomer inclusive plasticizers, chemicals for vulcanization and anti-ageing agents, other chemicals as well as fillers (Continental Reifen, 2013).

The input data for the MFA was obtained from already cited articles within previous chapters and subchapters (e.g. chapter 3 release of ENMs), through literature research and expert discussions. Various statistical databases (e.g. Statistik Austria) as well as different online databases (BOKU Lit-Search, Scopus and Google Scholar) were used for literature research.

3. Engineered nanomaterials in plastic products

ENMs are incorporated into polymer matrices because they improve their physical (electrical and thermal), chemical and mechanical properties. These properties can be further improved through addition of other functionalities, like for example sensorics (Adame and Beall, 2009; Hübner, s.a.). Therefore, the number of ENM-containing plastic products is rising (Sánchez et al., 2014). However, there are also some concerns about the environmental, health and safety aspects of ENM-containing plastic products due to their potential nanotoxicity (e.g.Foss Hansen et al., 2008; Laux et al., 2018; EUON, 2019).

3.1 Manufacturing of ENMs and PNCs

There are various methods of manufacturing ENMs. These methods can be subdivided into two main categories, into *top-down* (larger units are disintegrated) or *bottom-up* (molecular entities) processes (Mihranyan et al., 2012; Laux et al., 2018). Due to the need of energy to overcome internal bonding forces, *top-down* approaches are energy intensive processes. Some examples for top-down processes are high-pressure extrusion or microfluidization*.* *Bottom-up* approaches are based on spontaneous self-assembly of molecules and thus require just a moderate energy input. Examples for this method are layer-by-layer assembly or sol-gel procedures. These two main approaches are sometimes combined in manufacturing processes (Mihranyan et al., 2012).

ENMs can exist as primary particles, aggregates or agglomerates (c.f. Appendix 1 for more information). Primary particles can form aggregates. Hence, aggregates are a rigid and discrete colloidal entity that consist of various primary particles. In the following, aggregates can form agglomerates. It is important to note that different ENMs tend to form aggregates more easily than others. Aggregates and agglomerates are held together by van der Waals forces (Wang et al., 2002). Aggregates and agglomerates can occur during manufacturing processes as well as during different EOL processes (e.g. during incineration) (Environment Canada and Health Canada, 2013).

Manufactured ENMs are then incorporated into different polymer matrices. For the manufacturing of PNCs there are also some common methods like graft polymerization, sol-gel methods, surface modification of the NPs, solution blending techniques etc. (Díez-Pascual et al., 2015). ENMs can also be embedded into products as composites of more than one ENM, e.g. CB and CNT in car tires (Wohlleben et al., 2016).

There are two important types of embedding ENMs into a polymer matrix as it is shown in Figure 1. These types are to a certain extent responsible for the product enhancing properties. In the *intercalated type*, the polymer chains are between the parallel arranged ENM layers with a separation of a few nanometres (nm) between these layers. In the *exfoliated type*, the ENM-particles are separately dispersed in a polymer matrix with larger distances between the nanoadditives and the polymer chains. Due to the high aspect ratio of the platelets there is a large interface between matrix and ENMs that results in enhanced material characteristics (Weiss et al., 2006; Díez-Pascual et al., 2015). In most cases, exfoliated PNC morphologies result in better mechanical properties (Díez-Pascual et al., 2015).

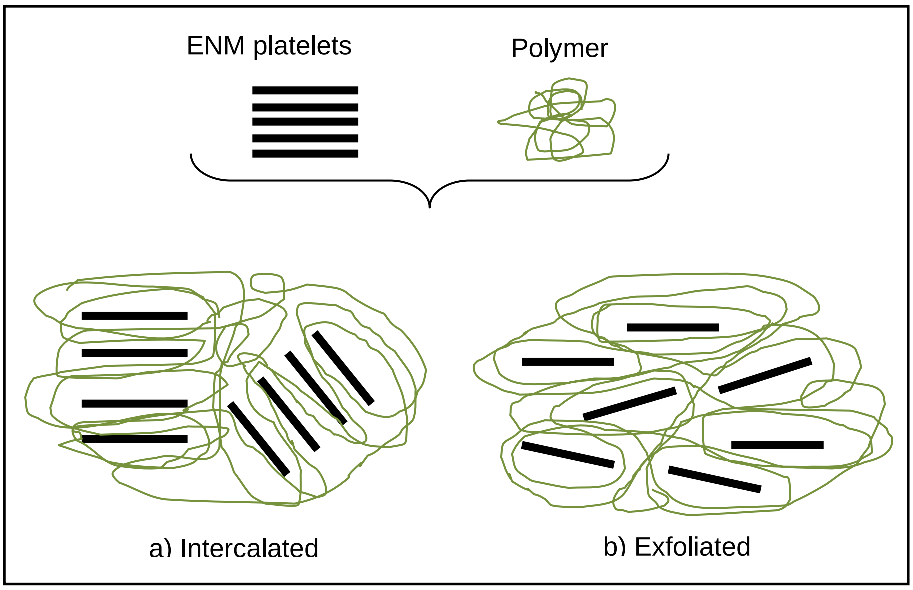

Figure 1: ENM platelets and polymer structure can be combined to (a) intercalated and (b) exfoliated PNC morphologies, modified from Weiss et al., 2006; Duncan, 2011.

For the production of exfoliated types, four main techniques exist, namely in-situ intercalative polymerization, solution intercalation, melt intercalation and in-situ template synthesis (Díez-Pascual et al., 2015). Exfoliated PNC morphologies are mentioned in the literature in connection with nanoclay (Weiss et al., 2006; Duncan, 2011; Potts et al., 2011; Díez-Pascual et al., 2015), graphene (Potts et al., 2011; Hu et al., 2014), graphite and graphite oxide (GO) (Díez-Pascual et al., 2015).

3.2 Data evaluation of ENM-containing plastic products

The collected data listed in Appendix 1 was evaluated according to the most frequently mentioned ENM types in plastic products. Furthermore, these listed ENM-containing products were assigned to different product categories, which have been established in accordance with the analyzed scientific literature. The findings of these two evaluations were combined in further consequence to give a more detailed insight into the specific ENMs from each of the four product categories with the most nominations of ENM-containing products. Overall, 40 relevant international articles (excluding four articles with future applications) were used for the data evaluation.

3.2.1 ENM types and their frequency of use in plastic products

Figure 2 shows the different ENM types mentioned in the 40 analyzed scientific articles as well as their corresponding number of nominations. A total of 214 nominations of 36 different ENMs has been received.

CNTs are the most frequently mentioned ENMs in plastic products with 38 nominations in total. They are used to improve the tensile strength, Young's modulus, electrical conductivity, electromagnetic shielding, stiffness and flexibility. Moreover, it is possible to produce lightweight products with the incorporation of these ENMs (Weiss et al., 2006; Allsopp et al., 2007; Paul and Robeson, 2008; Boysen et al., 2011a; Nowack et al., 2013). In comparison to steel, CNTs provide a 100 times stronger and at the same time 6 times lighter performance (Verma, 2013). CNTs can also be applied as antimicrobial components, because due to their long and thin structure they can puncture microbial cells and thus cause irreversible damages to them (Silvestre et al., 2011).

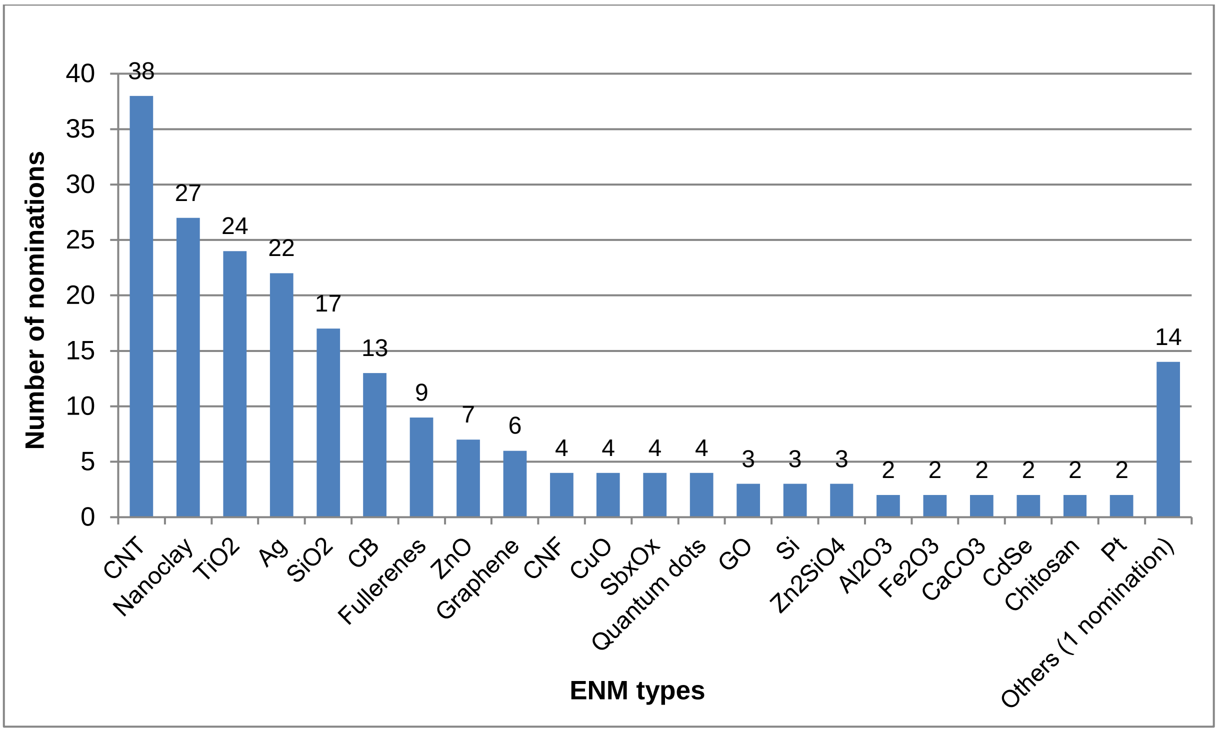

Figure 2: Types of ENMs contained in plastic products and number of their nominations in international scientific articles. ENMs with only one nomination have been summarized as “others (1 nomination)”. Basis: 40 international scientific articles, Σ different ENM types: 36, Σ nominated ENMs: 214.

With 27 nominations, nanoclay has the second most mentions, followed by nano-TiO_2 with 24 and nano-silver (Ag) with 22 nominations. ***Nanoclay*** has various material improving effects. For example enhancing effects in the mechanical properties of plastic products, like an improvement of the Young's modulus, stiffness and tensile strength (Paiva et al., 2008; Paul and Robeson, 2008). But it also exhibits product enhancing effects with respect to thermal properties, namely in thermal stability and thus an increased resistance to fire (e.g. Weiss et al., 2006; Silvestre et al., 2011; Sánchez et al., 2014). With regard to physical material properties, there is an improvement of gas barrier properties which leads to slower loss of air in sporting goods and an increased aroma stability of food and beverages. Thus, both of these products have a longer life span (e.g. Boysen et al., 2011a; Díez-Pascual et al., 2015; Han et al., 2018). This latter effect is the most frequently mentioned enhancing effect in the analyzed papers.

Nano-TiO_2 has photocatalytic effects, like the decomposition of organic compounds and their sterilization. Thus nano-TiO_2 provides disinfective and antimicrobial characteristics (e.g. Fujishima et al., 2000; Allsopp et al., 2007; Silvestre et al., 2011). Furthermore nano-TiO_2 offers ultraviolet (UV) protection for plastic products (e.g. Greßler et al., 2014; Greßler et al., 2017; Laux et al., 2018). Nano-TiO_2 also enables a self-cleaning effect, but only in combination with UV light and water (rain) (Quilitz et al., 2008; Greßler et al., 2010; UBA, 2014). Moreover, this ENM improves the power and flexibility of plastic products (Paul and Robeson, 2008; Verma, 2013).

The principal reason of embedding ***nano-Ag*** into plastic products is its antimicrobial effect (e.g. Allsopp et al., 2007; Duncan and Pillai, 2015; Hincapié et al., 2015). There are also some other effects like the suppression of bad odours (Allsopp et al., 2007; Duncan, 2011), improved strength (Duncan, 2011; Duncan and Pillai, 2015) or absorption and decomposition of ethylene and thus an extended shelf life of diverse food products (Azeredo et al., 2011).

More than half of all the nominations (i.e. 52 %) is attributable to the ENM types CNTs, nanoclay, Ag and TiO_2. As shown in Figure 2 these ENMs are then followed by the results of SiO_2 with 17 and CB with 13 mentions.

The incorporation of the ENM ***SiO_2*** improves the stability, flexibility, tensile strength, abrasion resistance, gas barrier properties and stiffness of a plastic product (e.g. Paul and Robeson, 2008; Duncan and Pillai, 2015; Greßler et al., 2017). There is also a resulting self-cleaning effect (Greßler et al., 2014; UBA, 2014; Duncan and Pillai, 2015) through the incorporation of nano-SiO_2 and a reduction of stray light and reflection (Wendling et al., 2009; Schulz, 2015).

Nano-CB improves the tensile strength, Young's modulus, UV protection as well as grip, abrasion and wear resistance (e.g. Verma, 2013; Wohlleben et al., 2016; Zhang et al., 2017). Furthermore, there is an improvement in the electrical properties of a plastic product, namely an increased electrical conductivity (Wang et al., 2002; Environment Canada and Health Canada, 2013; Zhang et al., 2017). Nano-CB is also used as black pigment in diverse plastic products (BMLFUW, 2013; UBA, 2014).

About two thirds (66 %) of the whole nominations are attributable to the ENMs CNTs, nanoclay, TiO_2, Ag, SiO_2 and CB. These six ENM types are followed by the results of fullerenes with nine, zinc oxide (ZnO) with seven and graphene with six mentions. The rest of the nominated ENMs incorporated in plastic products were mentioned between one and four times. ENMs that have been mentioned only once, namely aluminium hydroxide ($Al(OH)_{3)}$), gold (Au), cadmium telluride (CdTe), cerium dioxide (CeO_2), cellulose nano-reinforcements (CNRs), copolymers (butadiene, ethyl acrylate, methyl methacrylate, styrene), flake graphite (FG), gallium nitride (GaN), indium tin oxide (ITO), magnesium oxide (MgO), nanokaolin, antimony (Sb), silicates and titanium nitride (TiN), were summarized in the category “others (1 nomination)”.

3.2.2 Classification of ENM-containing plastic products

All these 36 above-mentioned different ENMs are incorporated into 214 products. These products have been categorized into seven different product categories: *automotive industry, coatings, electronics, food and beverage packaging, medical equipment, sporting equipment and textiles*. Products from each category can further be subdivided according to their product specification. However, this latter information was not available for all of the products (see Appendix 1 for more information).

As shown in Figure 3, the product category with the most ENM-containing plastic products is ***coatings***. There are 54 different plastic products with embedded ENMs in this category. The most frequently stated product specifications in this product sector are paints (e.g. exterior, interior) (e.g. Vermeiren et al., 2002; Duncan and Pillai, 2015; Singh et al., 2019). According to the analyzed articles, ENMs in paints are solely embedded in the plastic type thermosets and, more specifically, in resins (Greßler et al., 2010; UBA, 2014). For example, the company “Sto” offers products such as Lotusan® and StoColor Photosan® (Sto, s.a; Greßler et al., 2010). There are also some other plastic products in this category like adhesives, sealants, inks etc. (Chin et al., 2010; BMLFUW, 2013; Bott et al., 2014).

Food and beverage packaging is the sector with the second highest number of ENM-containing plastic products. Packaging in this category refers to wrapping for products like baked goods, meat, carbonated drinks, beer, fruit, vegetables, cheese etc. (Vermeiren et al., 2002; Silvestre et al., 2011; Greßler et al., 2017). For this packaging, mostly the polymers PE (low-density polyethylene (LDPE) and high-density polyethylene (HDPE)), polyethylene terephthalate (PET) and polypropylene (PP) are used (e.g. Silvestre et al., 2011; Sánchez et al., 2014; Watson-Wright et al., 2017). Producers of ENM-containing food and beverage packaging are for example “Süd-Chemie” with Nanofil® for PET or “Laviosa Chemica Mineraria” with Dellite® (Paiva et al., 2008).

Nearly half (i.e. 49 %) of the ENM-containing plastic products that have been analyzed were attributable to the two product categories *coatings* and *food and beverage packaging*.

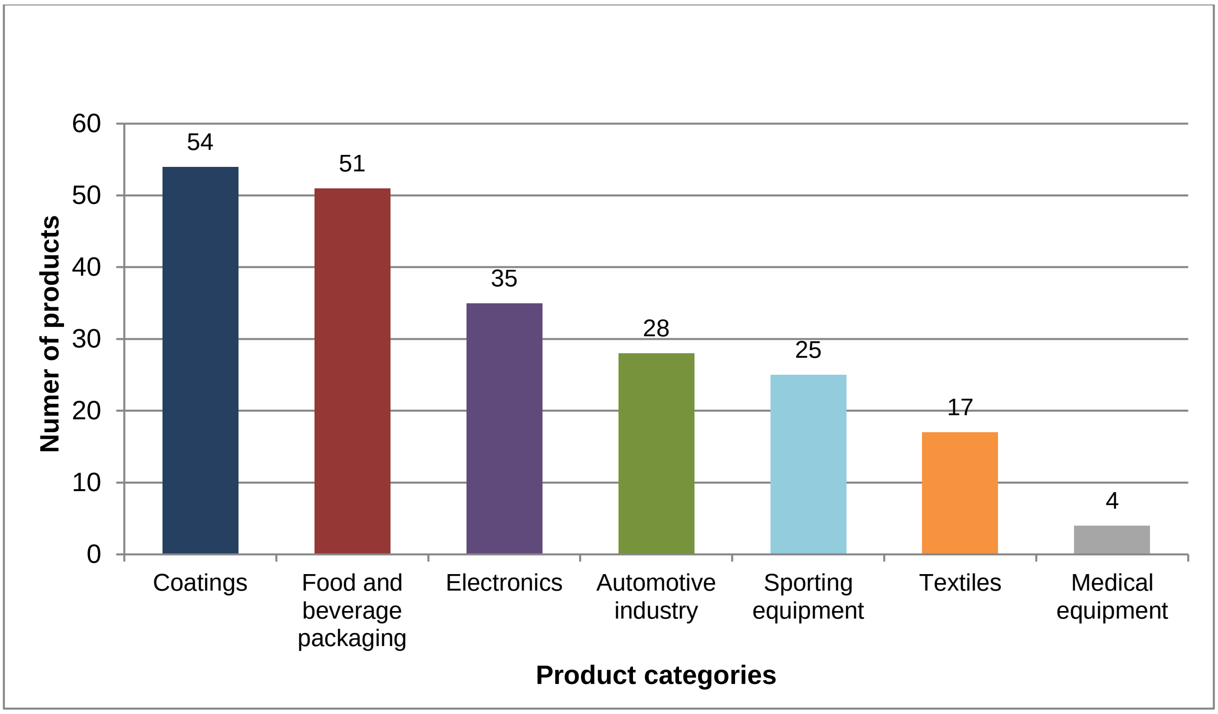

Figure 3: Classification of ENM-containing products into product categories. Basis: 40 international scientific articles, Σ product categories: 7, Σ products: 214.

As shown in Figure 3, the *electronics* category features the third most frequent mentions. This category counts 35 products and is followed by the categories *automotive industry* with 28, *sporting equipment* with 25, *textiles* with 17 and *medical equipment* with 4 products. Electrical and electronic equipment such as refrigerators, vacuum cleaners, washing machines, computer mice, mobile phones or tablets were mentioned in the category electronics (e.g. Allsopp et al., 2007; Duncan, 2011; Duncan and Pillai, 2015). Furthermore, this category also includes light emitting diodes (LEDs) and photovoltaics (e.g. Peet et al., 2009; Lee et al., 2010; Potts et al., 2011). Companies that produce ENM-containing electronics are for example “Daewoo”, “LG Electronics”, “IOGEAR”, etc. (Allsopp et al., 2007).

In the ***automotive industry,*** the most quoted specific products from the analyzed articles are vehicle tires. Tires are solely manufactured from elastomers like natural rubbers, polyisobutylene (PIB), styrene butadiene rubber (SBR), polyurethane (PU) etc. (Paul and Robeson, 2008; Wohlleben et al., 2016). “InMat LLC.” is a company selling ENM-containing plastic products that are to a certain extent used in vehicle tires (InMat Inc., s.a; Paul and Robeson, 2008). Other products in this category are for example fuel cells, assist steps, bumpers, structural components etc. (e.g. Paul and Robeson, 2008; Duncan and Pillai, 2015; Laux et al., 2018).

There is a lot of PNC-containing ***sporting equipment*** in the area of tennis, like tennis balls and racquets. Some producers in this sector are “InMat LLC.” for “Wilson” with their product Double Core^{TM} and “Yonex” with Nanolok^{TM} (e.g. Paul and Robeson, 2008; Boysen et al., 2011a; Greßler et al., 2014). Furthermore there are ENM-containing plastics such as fishing rods, arrows for archery, hockey sticks, skis, footballs, canoes, golf balls etc. (Paul and Robeson, 2008; Boysen et al., 2011a; Verma, 2013). In this category mainly thermosets in the form of epoxy are used (Paul and Robeson, 2008; Boysen et al., 2011a; Verma, 2013).

PNC products, such as carpets with plastic fibres, have been found for the category ***textiles*** (Vermeiren et al., 2002), also tents, uniforms (Fujishima et al., 2000), or different kinds of clothing (e.g. Fujishima et al., 2000; Greßler et al., 2014; Duncan and Pillai, 2015) etc. The company “DuPont” with their product MicroFree^{TM} is an example as a producer of ENM-containing plastic textiles. These textiles can be made of thermoplastics or thermosets in the form of polyester (Greßler et al., 2010).

As shown in Figure 3, these six product categories with double-digit nominations are followed by the category *medical equipment* with four nominations. ***Medical equipment*** for households includes plastic products such as wound care, bandage or latex gloves (Paul and Robeson, 2008; Duncan, 2011). Paul and Robeson (2008) mentioned Curad® as a product trade name for ENM-containing bandages.

3.2.3 Detailed analysis of ENMs in different product categories

The four product categories with the most ENM-containing plastic products have been analyzed in more detail according to their incorporated ENMs as shown in Figure 4. In the category of ***coatings,*** the most frequently embedded ENM type is TiO_2 with nine nominations, followed by SiO_2 with seven, Ag with six, CB and CNTs with five respectively. The rest of the embedded ENMs was only mentioned between one ($Al(OH)_3$, CeO_2, Iron(III) oxide (Fe_2O_3), FG, fullerenes, MgO, Si and zinc silicate (Zn_2SiO_4)) and three times.

Nanoclay has the most mentions in the category of ***food and beverage packaging*** with about a quarter (i.e. 26 %) of the nominations of the embedded ENMs. Nano-Ag was mentioned seven, CNTs six, nano-TiO_2 five, and nano-SiO_2 four times. The remaining ENMs were nominated once (Au, CNRs, copolymers, copper oxide (CuO), Fe_2O_3, nanokaolin, TiN and Zn_2SiO_4) or twice.

With seven nominations CNTs have the highest number of mentions in the product category ***electronics***, followed by nano-graphene with four and nano-Ag, nano-fullerenes and quantum dots with three nominations each. The rest of the ENMs were mentioned once (CdTe, GaN, ITO, Sb, oxides of antimony (Sb_xO_x), Si and ZnO) or twice.

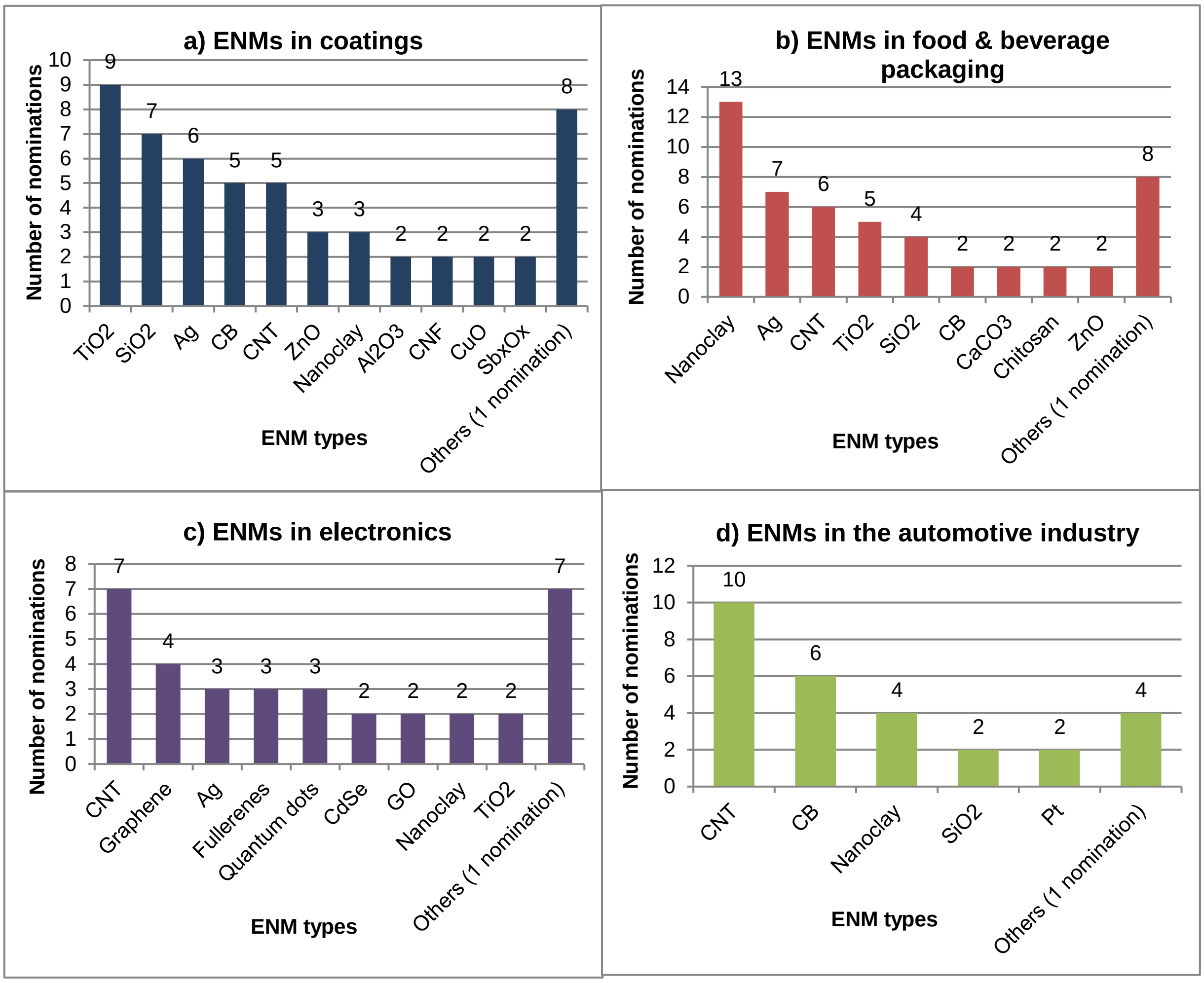

Figure 4: Nomination of different ENMs in the categories (a) coatings, (b) food and beverage packaging, (c) electronics and (d) automotive industry. ENMs with only one nomination have been summarized as “others (1 nomination)”. Basis: 40 international scientific articles.

The fourth category which has been analyzed in more detail refers to ***automotive industry***. In this category, CNTs are responsible for about a third (i.e. 36 %) of the nominations, followed by nano-CB with six nominations. These two ENMs account for more than half (57 %) of the nominations in this product category and are followed by nanoclay with four nominations. The remaining ENMs were mentioned once (carbon nanofibers (CNFs), fullerenes, graphene and quantum dots) or twice.

3.2.4 Detailed description of main ENMs in plastic products

In this section the six most frequently mentioned ENMs from the 40 analyzed international scientific papers are described in more detail according to their material specification, production processes, surrounding polymer matrices, mass content (wt% or concentration) in the polymer matrix, primary-, aggregate- and agglomerate-size. In the context of polymer matrices, it is important to know that a lot of embedded ENMs could not be allocated to a specific polymer type (marked in Appendix 1 as “n.i.”).

3.2.4.1 CNTs

CNTs are long cylinders which consist of carbon atoms. They are subdivided into two types: single-wall carbon nanotubes (SWCNTs) and multi-wall carbon nanotubes (MWCNTs). SWCNTs represent single graphene sheets and MWCNTs in contrast consist of nested graphene cylinders. Both of them are used as ENMs in plastics to achieve enhanced product characteristics like an improvement of electrical conductivity, stiffness, tensile strength etc. (Kim, 2003; Moniruzzaman and Winey, 2006).

CNTs can be produced through different bottom-up processes (Laux et al., 2018). Depending on the process, the results are either low-purity CNTs or high-purity CNTs. Low-purity CNTs are formed during a conventional CB production process through thermal synthesis (Laux et al., 2018). High-purity CNTs can be produced through a chemical vapour deposition (CVD) of carbon containing gases (bottom-up process) like e.g. methane. The thereby used catalysts have nanostructured surfaces on which the CNTs grow. The size of the catalyst is responsible for the diameter size of the CNTs (Kim, 2003; BMLFUW, 2013; Díez-Pascual et al., 2015).The preparation of polymer-CNT composites includes different processes, such as solution mixing, melt-compounding or in-situ polymerization etc. (Díez-Pascual et al., 2015).

CNTs are used in thermoplastics, thermosets and elastomers. They are mainly incorporated in thermoplastics (e.g. polyamide (PA), PE, PP, PU) (e.g. Chen et al., 2005; Watson-Wright et al., 2017; Zhang et al., 2017). For thermosets, CNTs were only used in epoxy (Paul and Robeson, 2008; Boysen et al., 2011a; Verma, 2013).

CNTs of high-purity (< 99 %) were mainly used in the framework of lab-scale studies in order to produce reinforced materials (Kim, 2003). With regard to the wide range of CNTs used in PCNs (0.1-50 wt%) (c.f. Appendix 1), such high quantities would be too costly, to be able to manufacture such products on an industrial scale (e.g. Wohlleben et al., 2016; Watson-Wright et al., 2017; Zhang et al., 2017). Regarding particle size, studies about the production of CNTs show that fibre diameter and length can range from 2 to 20 nm in diameter and 10 to 30 µm in length (Paul and Robeson, 2008; Zhang et al., 2017). Only one scientific paper specified numbers for the aggregate size with 10-20 nm in diameter and for the agglomerate size with 200 µm in diameter (BMLFUW, 2013).

The literature review showed that CNTs have been used most frequently in the *automotive sector*, specifically in vehicle tires or battery cells.

3.2.4.2 Nanoclay

Nanoclay has a nanolayer structure. These layers are separated by interlayer galleries (Sinha Ray and Okamoto, 2003; Silvestre et al., 2011). Nanoclay is mostly applied as montmorillonite (MMT) in plastic products (e.g. Weiss et al., 2006; Paiva et al., 2008; Silvestre et al., 2011). MMT is a composite of aluminium (Al), silicate and clay with a 2:1 layered structure and a high surface area. In this layered structure each clay platelet consists of two tetrahedral silica layers between which there is a single octahedral Al (Weiss et al., 2006; Duncan and Pillai, 2015). MMT is naturally available and can be derived from volcanic rocks and ash (Silvestre et al., 2011). It has the chemical formula $(Na,Ca)_{0.33}(Al,Mg)_2(Si_4O_{10})(OH)_2 \cdot nH_2O$ (Weiss et al., 2006). Polymer-nanoclay composites are formed through numerous processes, like for example in-situ polymerization, solution or melt processing (Paul and Robeson, 2008).

Nanoclays are high-aspect-ratio platelets, which means they may be in the nm-range in their thickness but a few thousand times larger in their other two lateral dimensions (Duncan and Pillai, 2015). Nanoclay is applied as intercalated and exfoliated platelets. As shown in Figure 5 there is an existence of a so called “tortuous pathway” in the case of exfoliated nanoplatelets, which leads to a slower diffusion of molecules. This circumstance results in an improved gas barrier and thus in a longer product shelf-life for food and beverage articles (e.g. Weiss et al., 2006; Azeredo et al., 2011; Duncan, 2011). Moreover, plastic products containing nanoclay show more “topographical hills”. These hills are visible with atomic force microscopy (AFM) and cause a lower free volume in the matrices. Thus, there are lower diffusion rates, which is another reason for an enhanced gas barrier property (Adame and Beall, 2009). Furthermore, this altered diffusion is to a certain extent responsible for the increased flame resistance, because the volatile decomposition products diffuse more slowly (Silvestre et al., 2011). Another reason for its property as flame retardant is the presence of a nanoclay-rich layer at the surface of host polymers after heat exposure. This nano-layer can reduce the additional heat exposure by slowing down the heat transfer between the PNC and the environment (Duncan, 2015).

Within the analysis of the scientific papers, nanoclay has been used most frequently in *food and beverage packaging*.

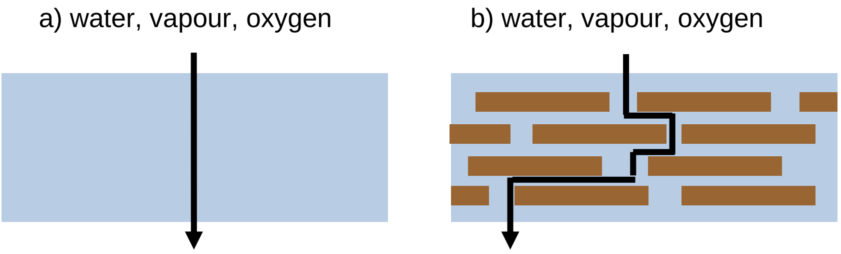

Figure 5: Formation of a “tortuous pathway” by the incorporation of exfoliated nanoplatelets into a polymer matrix film. In a film of (a) only polymer, diffusing gas molecules on average migrate via a pathway perpendicular to the polymer film orientation or (b) nanocomposites the diffusing gas molecules have to navigate around the clay platelets, modified from Duncan, 2011.

According to the literature, nanoclay is used in all three different types of plastic. However, its major appliance is in thermosets (e.g. PA, PE, PET, PP) (e.g. Paiva et al., 2008; Paul and Robeson, 2008; Han et al., 2018) and in elastomers (e.g. ethylene-vinylacetate copolymer (EVA), butyl rubber (IIR), PIB) (e.g. Paul and Robeson, 2008; Silvestre et al., 2011; Verma, 2013).

Literature data shows that the content of nanoclay in PNCs range between 0.1-10 wt% (e.g. Adame and Beall, 2009; Greßler et al., 2017; Han et al., 2018). In the lateral dimension, the primary particle size is stated as few hundred nm to µm (Díez-Pascual et al., 2015; Greßler et al., 2017) as well as 1.7 nm (for PA, PP, PS) to about 3 nm (for PP) in diameter (Paiva et al., 2008; Han et al., 2018). According to the aggregate and agglomerate size, there is no available information in the evaluated literature.

Within the analysis of the scientific papers, nanoclay has been used most frequently in *food and beverage packaging*.

3.2.4.3 Ag

Nano-Ag is generally mined as a metal (= “solid Ag”) and more infrequently in the form of Ag-salt (Steinbach, s.a.b). This ENM is usually nanoscale in all three dimensions (Duncan and Pillai, 2015).

The ENM Ag provides natural antibacterial effects (Allsopp et al., 2007). It has a broad toxicity spectrum to numerous strains of bacteria, algae, fungi etc. (Azeredo et al., 2011; Duncan, 2011; Silvestre et al., 2011). The antimicrobial properties of nano-Ag depend on a release mechanism of Ag^+. These ions bind to electron donor groups in different biological molecules that contain oxygen, nitrogen or sulphur (Azeredo et al., 2011).

Nano-Ag can be mainly found in thermoplastics (e.g. PE, PET, PP) (e.g. Vermeiren et al., 2002; Silvestre et al., 2011; Duncan and Pillai, 2015) and in thermosets (e.g. resin, chitosan) (Allsopp et al., 2007; Duncan, 2011; Hincapié et al., 2015). Just one paper mentioned nano-Ag in combination with elastomers (Paul and Robeson, 2008).

Regarding the mass content of Ag in plastic products, there is different data in the literature available: Duncan (2011) speaks of 2-4 mg/m^3 for different plastic products, Duncan and Pillai (2015) state 1.5 mg/m^3 for *coatings,* whereas Hincapié et al. (2015) refers to > 1 mg/kg for *coatings*. The primary particle size is stated between 10-500 nm (Duncan, 2011; Duncan and Pillai, 2015). With regard to the aggregate and agglomerate size, there is no information available within the analyzed articles.

Within the evaluation, nano-Ag has been found mostly in the categories of *coatings* and *electronics* and more specifically in paints and electrical and electronic equipment.

3.2.4.4 TiO_2

70 % of all TiO_2 in Europe is extracted from natural minerals using the sulphate method and 30 % is extracted through the chloride method. After the extraction of TiO_2, the nanoscale form has to be produced in separate processes. Some possible processes are hydrolysis in combination with thermal processes or through a reaction of titanium chloride (TiCl) compounds with ammonia (NH_3) under the influence of heat. The crystal structures of the ENM TiO_2 can vary due to the different arrangements of TiO_2-atoms. These arrangements are a result of the chosen production technique. There are rutile and anatase modifications (Steinbach, s.a.c).

When nano-TiO_2 is exposed to light, it can be used to break down organic compounds through photocatalysis (Fujishima et al., 2000; Boysen et al., 2011b; Silvestre et al., 2011). This characteristic of nano-TiO_2 therefore is used for antimicrobial appliances (Othman et al., 2014). The antimicrobial effect is based on a peroxidation of phospholipids of microbial cells (Hübner, s.a; Azeredo et al., 2011). It is advantageous that the ENM remains stable during the process of photocatalysis (Greßler et al., 2010).

The ENM TiO_2 is also used in all three groups of plastics, but mainly in thermoplastics (e.g. PET, PE, polyvinyl chloride (PVC)) (e.g. Othman et al., 2014; Sánchez et al., 2014; Watson-Wright et al., 2017).

With respect to the mass content of nano-TiO_2 there are similar numbers in the analyzed articles. Hincapié et al. (2015) speak of 0.1-5 wt% for *coatings*, Watson-Wright et al. (2017) refer to a mass content between 2-5 wt% for *medical equipment* and

Othman et al. (2014) speak of 4-9 wt% for *food and beverage packaging*. Only one article stated a primary particle size, namely 25 nm, for the category *food and beverage packaging* (Othman et al., 2014). There is no information available on the sizes of aggregates and agglomerates within the articles under investigation.

Within the analysis of the scientific papers, nano-TiO_2 has been used most frequently in the category *coatings* and more specifically for paintings.

3.2.4.5 SiO_2

Nano-SiO_2 can be manufactured through bottom-up processes by pyrogenic flame synthesis or precipitation (Laux et al., 2018). PNCs with SiO_2 can be prepared through blending, in-situ polymerization and sol-gel processes (Díez-Pascual et al., 2015).

The ENM SiO_2 is mainly found in thermosets (e.g. epoxy, resin) (e.g. Boysen et al., 2011a; Verma, 2013; Hincapié et al., 2015) and thermoplastics (e.g. PA, PET, PP) (e.g. Wendling et al., 2009; Silvestre et al., 2011; Singh et al., 2019).

Depending on the application, the mass content of nano-SiO_2 in plastic products ranges between 0.1-20 wt% (Hincapié et al., 2015; Singh et al., 2019). With regard to the primary particle-, aggregate- and agglomerate-size of nano-SiO_2, there are restrictions from the European Commission by the Regulation No 10/2011. Regarding nanomaterial classification, the primary particle size is set between 1-100 nm, the aggregate size is regulated with 0.1-1 µm and the agglomerate size is specified as ranging from 0.3 µm to mm size.

According to the literature, nano-SiO_2 has mostly been used for *coatings*, specifically in paints.

3.2.4.6 CB

The ENM CB is chemically very stable and thus often applied in plastic products. It has an amorphous, quasigraphitic structure and is principally composed of elemental carbon (Wang et al., 2002; Díez-Pascual et al., 2015). CB is produced by pyrolysis at high temperatures. Through quenching with water, the combustion reaction remains incomplete so that CB is formed. About 65 % of the world's CB production are used in tire production (IPPC, 2007). During the combustion reaction, carbon nodules (5-100 nm) are formed. However, these nodules do not retain their original shape for very long. They immediately cluster into aggregates (70-500 nm) (Donnet et al., 1993). Within these aggregates, the particles are bound together by covalent bonding mechanisms. Moreover, these aggregates also form agglomerates (10-100 µm) with electrostatic bonds (Rwei et al., 1991; IPPC, 2007). For the use in plastic products, these primary particles, aggregates and/or agglomerates are incorporated into a polymeric matrix. According to the screened articles, nano-CB is applied in all three different plastic types, but mainly in elastomers (e.g. natural rubbers, PU, SBR) for the use in vehicle tires (e.g. Paul and Robeson, 2008; Bott et al., 2014; Wohlleben et al., 2016). For this, CB (nano- and microscale) is formed into pellets. During this process, larger agglomerates may break down into smaller aggregates (70-500 nm) (Rwei et al., 1991; IPPC, 2007).

Based on an evaluation of the Scientific Committee on Food at the European Commission (SCF), a maximum allowed use level of 2.5 wt% for CB in *food and beverage* *packaging* has been set (Bott et al., 2014). In other products, the stated mass content ranges from 3-12 wt% in the case of *coatings* (Zhang et al., 2017), or between 22-40% in vehicle tires (Continental Reifen, 2013; Environment Canada and Health Canada, 2013; Singh et al., 2019).

In general, the primary particle size of CB depends on the application (Bott et al., 2014): For *food and beverage packaging,* the primary particle size was regulated by the European Commission within the Regulation No 10/2011 between 10-300 nm. In the field of *automotive industry* (i.e. vehicle tires) and *coatings* containing CB, the primary particle size is between 10-500 nm (e.g. Wang et al., 2002; Paul and Robeson, 2008; Voll and Kleinschmit, 2010). These primary particles are important for the morphology parameters of CB because they determine the specific surface area, which is important regarding the desired end-use application. The smaller the primary particles of CB, the higher the specific surface area. This leads to different effects of embedded CB on the properties of a product. For example, CB with higher specific surface area leads to better abrasion resistance in vehicle tires. If there is a lower specific surface area, the dispersibility and consequently the performance, e.g. in inks, is better than with larger surface area. In this context, it is crucial to know that these primary CB particles, although important for the morphology, only exist to a very small quantity as discrete entities in CB-containing products as they have a strong tendency to form aggregates and agglomerates (Wang et al., 2002). In the case of *food and beverage packaging*, the size of the chemical substance “CB” was regulated by the Regulation No 10/2011 with 100-1200 nm. For other product categories, the aggregate size was stated between 10-hundreds of nm (Wang et al., 2002). The size of the agglomerates for food contact material was again regulated within the Regulation No 10/2011 between 300 nm-mm range.

The literature review showed that CB (nano- and microsized) has most frequently been used in the category *automotive sector* and here especially in vehicle tires. CB is mainly incorporated in tires because it provides UV protection as this material adsorbs UV radiation (Wohlleben et al., 2016). Furthermore, CB has also been used widely in the category of *coatings*.

3.2.5 Future applications of PNCs

There are many different PNC applications under development on laboratory scale (technology readiness level (TRL) 1-4) or on the level of prototypes (TRL 4-8). Even if there was to be an important discovery in this field of research, it often takes decades until this invention reaches large commercial impact (TRL 9) (Paul and Robeson, 2008; EC, 2014). Nevertheless, some examples of future applications will be presented here (c.f. appendix 1, green marked products).

A possible future application of PNCs is in antireflective polymer optics. For this purpose, various nanoscale metal oxides (TiO_2, aluminium oxide (Al_2O_3), tantalum pentoxide (Ta_2O_5), magnesium fluoride (MgF_2) and SiO_2) are currently under investigation. These ENMs can be incorporated in optical lenses for cameras or headlights to reduce the stray light and thus provide anti-reflective properties (Wendling et al., 2009; Schulz, 2015).

At the moment, there is a lot of research in the field of food and beverage packaging, particularly on “intelligent packaging” (Weiss et al., 2006; Azeredo et al., 2011; Duncan, 2011). This smart packaging can detect gasses, chemical contaminants, aromas and pathogens through electrochemical or optical (colorimetric or fluorimetric) methods. Thus, an improvement of quality as well as food and beverage safety is possible (Duncan, 2011). Moreover, there is currently some research concerning time and temperature indicators for packaging, which could also lead to improved quality and quality control (Azeredo et al., 2011). There are already a few of these nano-sensors available like for example ripeSense®[1] (Duncan, 2011). With respect to the product of ripeSense®, there was no information on the used ENMs available. Furthermore, there is currently some research concerning antimicrobial applications for the food and beverage sector with the ENMs MgO, ZnO (Silvestre et al., 2011) and TiO_2 (Othman et al., 2014) as well as for the sector of coatings with the ENMs copper (Cu) and CuO (Delgado et al., 2011).

3.3 Release of ENMs from polymeric matrices

Regarding environmental health and safety aspects, it is essential to understand the potential release mechanisms of the incorporated ENMs because due to the unique physical and chemical properties of ENMs, they may be toxic for humans and the environment. However, the toxicological profiles of different ENMs may vary in accordance to their macroscale matter (Silvestre et al., 2011; Nowack et al., 2012; Duncan, 2015). Furthermore, released ENMs can change their form and may not be nanoparticulate anymore after release, which in turn can lead to distinct toxicological properties (Keller et al., 2013). For more information on nano-toxicological effects see section 3.4.

Especially in the field of plastic household products, it is crucial to understand the ENM release mechanisms, because humans can directly be exposed. It is also important to consider the location of the ENMs in a polymer matrix (Foss Hansen et al., 2007; Foss Hansen et al., 2008; Duncan and Pillai, 2015). A release from the polymer matrix can take place along the entire life cycle of an ENM-containing plastic product (Greßler et al., 2014).

3.3.1 General release mechanisms

According to Noonan et al. (2014), there are four different release pathways for ENMs from PNCs: *diffusion* release, *dissolution* into external media, *desorption* and a release through *matrix degradation.* These pathways are shown in Figure 6. Within this framework, Duncan (2015) speaks of two roughly distinct ENM release paradigms, because the first three release pathways (desorption, diffusion and dissolution) can be seen as passive ones whereas the fourth (degradation of matrix) is caused through active external influences.

Regarding all four different release mechanisms, a positive correlation between an ENM release and product life time was found. This implies an increased amount of ENMs as a function of product life time (Šimon et al., 2008; Duncan and Pillai, 2015; Han et al., 2018).

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[1] http://ripesense.com/

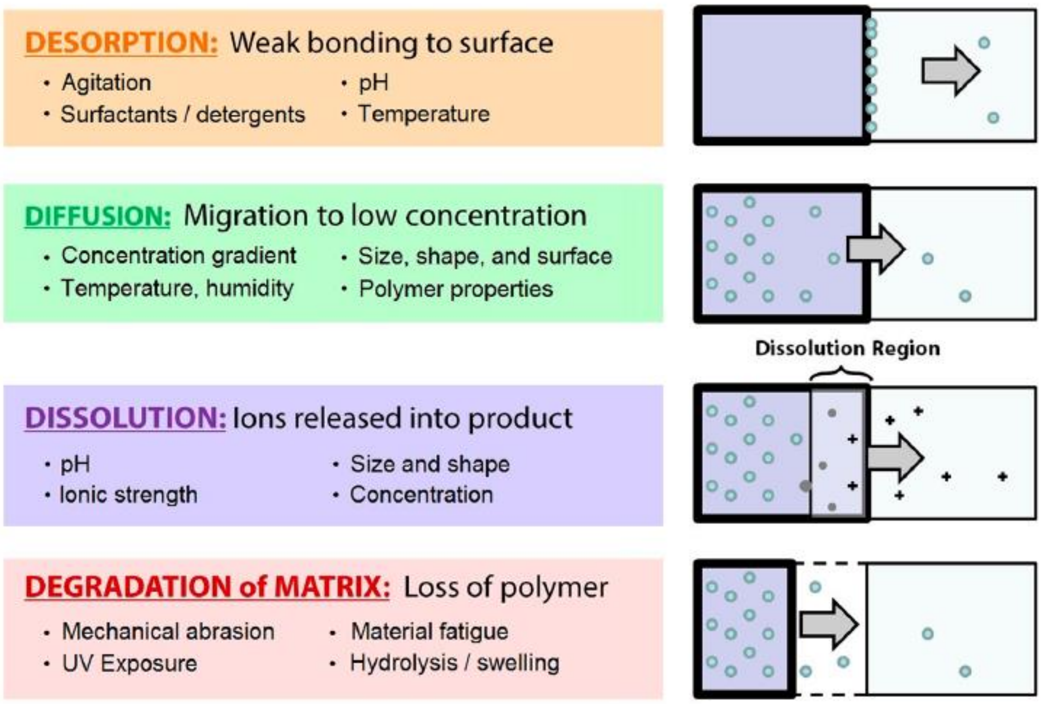

Figure 6: Four possible ENM release pathways (desorption, diffusion, dissolution and degradation of matrix) and respective crucial parameters for taking effect, according to Noonan et al., 2014.

3.3.1.1 Diffusion

As depicted in Figure 6, in the case of a diffusion release there is a relocation of ENMs from the interior of a PNC to the interface between the polymer and the exterior medium (e.g. environment, food) followed by its release into the respective medium. Random chain movement creates free volume holes in the polymer matrix which enables a relocation of ENMs. The process of diffusion release is also known as *migration* and depends on different parameters, such as the molecular weight, size and shape of the migrant or the temperature, polarity or humidity of the exterior. Furthermore, complex interactions with the host matrix can also have effects on the ENM release. Overall the diffusion release pathway plays a dominant role in the field of PNCs (Šimon et al., 2008; Noonan et al., 2014; Duncan and Pillai, 2015).

Šimon et al. (2008) and Duncan and Pillai (2015) found a positive correlation between the migratability of ENMs and the ambient temperature at which plastic products are stored. These findings are based, inter alia, on a theoretical diffusion model applying the Stokes−Einstein equation at 4 °C and 25 °C for LDPE, HDPE, PP, PET and PS. At a temperature of 25 °C, there was a higher diffusion constant than at 4 °C (Duncan and Pillai, 2015).

Šimon et al. (2008) concluded in their analysis that the diffusion of ENMs in polymers is very slow and can be expected mainly in the case of very small nanoadditives (radius < 1 nm) from polymeric host materials with low dynamic viscosity. This corresponds for example to nano-Ag from polyolefins. The migration of bigger nanoadditives with high dynamic viscosity is not very likely. This applies for instance to nanoclay in PET or PS.

3.3.1.2 Desorption

Adsorbed ENMs can also be released spontaneously into an external liquid medium as it is shown in Figure 6 (Noonan et al., 2014; Duncan and Pillai, 2015).

Released ENMs within the two processes of *diffusion* and *desorption* remain in their nanoparticulate form. Although there may be an alteration in their surface characteristics, morphology or composition during or after their release (Duncan and Pillai, 2015).

3.3.1.3 Dissolution

In the case of dissolution mechanisms, the release of internally embedded or surface-bound ENMs is caused by dissolution of these ENMs into constituent atoms/ions. These atoms/ions then may diffuse into an external medium (Noonan et al., 2014; Duncan and Pillai, 2015). ENMs that have been released though dissolution are no longer in a nanoparticulate form after their release. However, the dissolved constituents may once again reform NPs (Duncan and Pillai, 2015).

3.3.1.4 Matrix degradation

An ENM release due to matrix degradation can be caused by mechanical exposure (e.g. machining, wear) and/or chemical aging (e.g. photodegradation, hydrolysis) as shown in Figure 6. *Weathering* as a degradation mechanism includes mechanical stress (e.g. wind), photochemical degradation and hydrolysis (e.g. rain) (Froggett et al., 2014; Noonan et al., 2014; Duncan, 2015; Koivisto et al., 2017).

Reviews about an ENM release from PNCs through matrix degradation performed by Froggett et al. (2014) and Duncan (2015) showed that in most of the analyzed studies the released particles were agglomerates of the ENM filler and the respective polymeric host product. These findings are similar to those of Nowack et al. (2013) who stated, that the likelihood of release of solely nanostructured material is small. This is because for an individual ENM release, a high energy input would be needed. Hence, a release of fragments of PNC material is more likely.

Mechanical exposure is one of the most typical release processes in the category of matrix degradation (Koivisto et al., 2017). There are several influencing factors on the release of ENMs through mechanical exposure. One possible factor are the *nanoadditives* themselves (Duncan, 2015; Singh et al., 2019). Some of the studies Duncan (2015) analyzed stated an increase of ENM release due to a higher ENM content, others recognized a decrease. It is assumed that the influence of ENMs on the host material properties are more important than their intrinsic properties. In this context, the dispersion characteristics of the ENMs in the polymer matrix are important (e.g. exfoliated nanoclay). Well distributed ENMs have more attractive forces than poorly distributed ENMs and thus they are less likely to become released as free ENM particles (Duncan, 2011, 2015). However, with the current state of knowledge, no general statement on the influence of ENMs on particle release can be made. More research is needed to gain significant and comparable results (Duncan, 2015).

Furthermore, the *host material* can have effects on the ENM release. This is related to the mechanical properties such as strength, fracture toughness, hardness etc. of different polymers (Duncan, 2015). For instance, the abrasion of hard lacquer coating with ENM fillers releases more PNC particles compared to softer ENM-containing acrylic paints. This is due to the fact that softer PNCs can transfer some of the sheer energy of the abrasion into deformations (Koponen et al., 2011). However, there are various interrelated factors (e.g. humidity, molecular weight, presence of additives like ENMs) which the properties of a host material depend on. Hence, the prediction of the influence of a host material on ENM release is very complex and difficult to make. Up to now, there have been just a few studies which deal with this topic (Duncan, 2015).

Moreover, the *type of machining process* (e.g. sanding, cutting, sawing, shredding) has an effect on the size and concentration of the ENM release from PNCs. More aggressive processes can result in higher release rates than softer ones. Therefore, an important factor in this context is the amount of sheer energy applied (Duncan, 2015). Another positive correlation was found with the speed of mechanical processing (e.g. drilling speed) (Bello et al., 2010). In the case of machining processes, a release of matrix components with and without ENMs as well as a release of individual ENMs have been found (Froggett et al., 2014). However, also in this area only a few studies have investigated the relationships between the different types of machining processes and PNCs to draw broader conclusions (Duncan, 2015). Nevertheless, this release mechanism is important to keep in mind especially in relation with recycling processes.

Overall, in the articles reviewed by Duncan (2015), a release of free, aerosolized fibrous ENMs (e.g. CNTs) with respect to mechanical exposure was observed, whereas no release of free ENMs from PNCs with small, spherical nanoadditives (e.g. SiO_2) was detected. However, it is possible that these spherical ENMs adhere to larger surfaces once they are released and thus could not be detected. Furthermore, the particle release rates from mechanical exposure are case specific and thus dependent on the experimental design and concentrations of a particular ENM (Koivisto et al., 2017).

Regarding ***chemical degradation*** of the polymeric host material there are different mechanisms such as photochemical and thermochemical degradation, hydrolysis or enzymatic degradation (Froggett et al., 2014; Duncan, 2015).

In the case of *photochemical degradation* of the host matrix, complex and polymer specific chemical reactions can lead to the release of ENMs (Duncan, 2015; Koivisto et al., 2017). If polymers are exposed to UV light, the matrix around the ENMs may begin to degrade and thus enable ENMs to accumulate on the surface. These ENMs may be released under mechanical influence. In case of fibrous ENMs (e.g. CNTs) there is a formation of entangled networks on the surface of the host material and thus these ENMs are less likely to release whereas spherical ENMs (e.g. SiO_2) cannot form entangled networks and thus easily fall off the surface after their exposure (Nguyen et al., 2011). However, ENMs which collect on the surface after photodegradation may filter UV radiation and thus protect the remaining polymer matrix from further UV degradation (e.g. CNTs) (Duncan, 2015). Incorporated ENMS (e.g. nanoclay) can also accelerate photodecomposition (Grigoriadou et al., 2011; Duncan, 2015). Clearly the degradation processes increase with increasing duration of UV exposure (Froggett et al., 2014).

Overall, a release of fragments (plastic debris) with and without ENMs induced by chemical degradation could be observed. Especially through UV degradation a release of individual ENMs is possible (Froggett et al., 2014; Koivisto et al., 2017). In combination with mechanical processing, the possible ENM release may be increased even further. Up to now, there are only a few studies that have directly investigated the release of ENMs through UV decomposition of polymer matrices. New analytical techniques on microscopic levels are needed to generate a better understanding (Froggett et al., 2014; Duncan, 2015).

With respect to *thermochemical degradation* there are no studies on a related ENM release so far. However, it can be assumed that under thermal influence, polymers begin to decompose and thus nanoadditives are released from the matrix. Bearing this in mind, also mechanical processing may be relevant because mechanical forces generate frictional heating (Duncan, 2015).

In the case of *hydrolysis,* the focus so far has been on washing processes of nano-Ag-containing plastics (e.g. textiles) (Froggett et al., 2014). Some ENMs accelerate the hydrolysis of PNCs and some slow down this process (Bikiaris, 2013; Duncan, 2015). During hydrolysis PNCs can undergo different changes when exposed to a liquid. There are slight changes like absorption of the liquid, swelling of the PNC and more but also serious alterations such as oxidation or complete degradation. In the latter case free ENMs may be released from the polymeric host matrix (Duncan, 2015). However, until now, most studies could not observe free ENMs released from plastics through hydrolysis (Froggett et al., 2014).

Enzymatic degradation is especially interesting for PNCs in landfills. Due to different enzymatic processes of biodegradation, matrix nanoscale additives may be released into the surrounding area. Particularly products that were designed to degrade quickly can play an important role in this context. Here too, ENMs can promote or inhibit particle release (Bikiaris, 2013; Duncan, 2015).

3.3.2 Methods to assess the release of ENMs

Regarding different assessment methods for released ENMs, some experimental and theoretical approaches exist (Duncan and Pillai, 2015).

According to Bott et al. (2014), in the case of experimental methods, a sensitive technique is required to quantify the potential migration of ENMs from plastic matrices. This technique has to fulfil the requirements of separation and detection of ENMs in migration solutions (Bott et al., 2014). Hence, they suggest using an asymmetric flow field-flow fractionation (AF4) for the investigation. The main underlying principle of the AF4 technique was established by Giddings (1993) and has since been further developed by Messaud et al. (2009). The AF4 process takes place in a parabolic flow profile in which there is a size-dependent diffusion of particles. This diffusion happens against a contrary separation force field (cross-flow).

An experimental approach that is already in use is the assessment of the migration of ENMs from thin polymer films. For this method, sections of test film containing the potential contaminant are submerged into a fluid. This fluid is then analyzed for the presence of the migrant (Duncan and Pillai, 2015). Another example for an experimental method is the assessment of released ENMs from plastic films into liquid media via liquid diffusion cells. For this method, a neat, polymeric film is inserted between two chambers. Both chambers are filled with test liquid and one of them is additionally charged with the potential migrant. The kinetics of migration can then be assessed by removal of aliquots of the liquid medium from the chamber with originally no migrants. This is done at regular time intervals (Duncan and Pillai, 2015).

Methods to quantify the ENM content and measure the particle size distribution in these experimental approaches are, for example, single particle inductively coupled plasma mass spectrometry (SP-ICP-MS), inductively coupled plasma atomic emission spectroscopy (ICP-AES), fast mobility particle sizer (FMPS) or atomic absorption spectroscopy (AAS). Methods to provide information on the particle composition and morphology are, for instance, transmission electron microscopy (TEM), scanning electron microscopy (SEM). (Duncan, 2015; Duncan and Pillai, 2015; Brame et al., 2018). Usually a combination of instruments is used to capture the particles, measure their number concentration and size and to assess their morphology and composition (Duncan, 2015).

In the field of theoretical approaches, there is some modelling based on fundamental diffusion physics and physicochemical properties of the polymeric material (Duncan and Pillai, 2015).

3.3.3 General ENM release rates along the life cycle of a plastic product

The above-mentioned ENM-containing plastic products or some parts of them may at some point be released as nano-pollution or end up in the waste regime as nanowaste. As shown in Figure 7, a generation of nanowaste and nano-pollution can occur in every stage of the life-cycle of a PNC (Greßler et al., 2014). As already mentioned, so far, there are only a few studies on the release of ENMs from plastic products. Furthermore, there are even less studies on the release rates of individual ENMs from PNCs which leads to a wide variation within the available results (Keller et al., 2013). In the following sections, the main life cycle stages of ENM-containing plastic products with the corresponding ENM release rates will be shown. These values are no specific values for certain types of ENMs. The current state of knowledge on type specific ENM release rates will be presented in section 3.3.4. In addition to the release rates the generation of nanowaste and nano-pollution in each step was analyzed. Special focus was placed on the different EOL stages.

In the context of ENM-containing plastic materials, there are five different relevant EOL stages in Austria in which intentional shipments (mainly nanowaste) and unintentional emissions (mainly nano-pollution) of ENMs are possible: (1) Some of the ENM-containing plastic products (e.g. PET-bottles, car tires) enter a *recycling station* after their use-phase. If ENM-containing plastic products (e.g. sporting racquets, medical bandages) get into the residual household waste, these products either enter (2) a *mechanical-biological waste treatment plant* (MBWTP) or (3) a *waste incineration plant* (WIP) to fulfil the requirements for (4) a subsequent disposal on a *landfill*. Moreover, ENM-containing plastic products or parts of them (e.g. facade painting, textiles) can enter (5) *waste water treatment plants* (WWTPs) over diffuse emissions.

3.3.3.1 Primary manufacturing

As shown in Figure 7, the first main life cycle step of ENM-containing plastic products is the primary manufacturing of ENMs. In this phase, ENMs are not yet incorporated into a product but are still the pure ENMs. The rate of free ENM release in this life cycle stage is between 0.1-2.0 % (Keller et al., 2013). Nanowaste generated during primary manufacturing can be collected separately and in further consequence also be internally recycled or transferred to a waste treatment plant (WTP) (Nowack et al., 2013; Song et al., 2017). In the case of plastic products, WTPs refer to recycling plants, MBWTPs, WIPs as well as WWTPs (diffuse emissions).

According to Keller et al. (2013) 20-80 % of the nanowaste from this life cycle stage finally ends up in a landfill. Unintended and not collectable nano-pollution from the primary manufacturing of ENMs may be emitted to the air (10-40 %) or the water prior to a WWTP (10-40 %) (Keller et al., 2013; Part et al., 2018a). These figures are not only valid for PNCs but have to be seen as general release rates for ENMs from different host matrices.

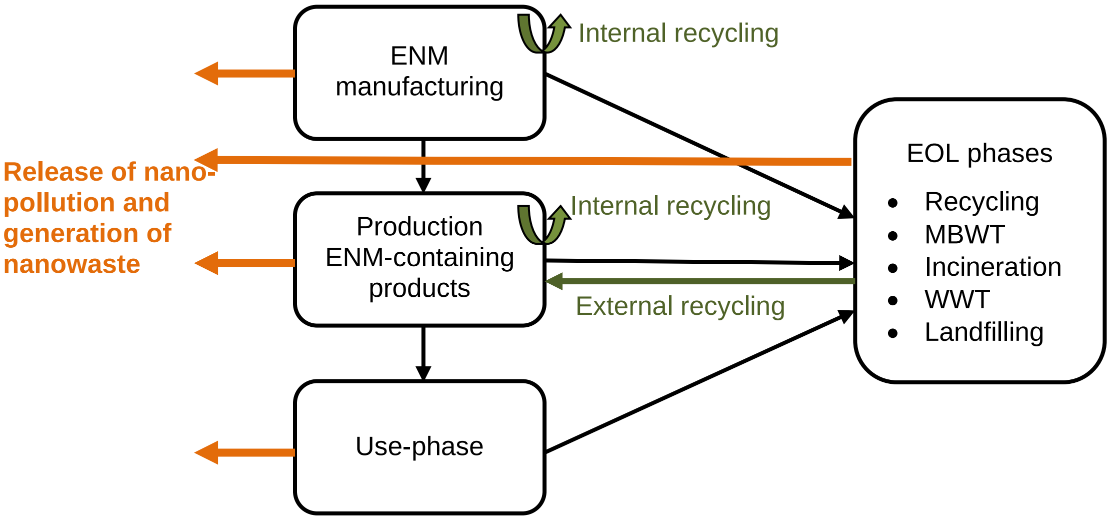

Figure 7: Main life cycle stages of ENM-containing plastic products with corresponding release pathways of nano-pollution and nanowaste, modified from Nowack et al., 2013.

3.3.3.2 Production of ENM-containing products

As depicted in Figure 7, the next life cycle step is the production of ENM-containing products. In this phase, the ENMs are incorporated into the polymer matrix of a specific product. The release in this phase is mainly due to mechanical stress and inefficient use of ENMs. During the production of ENM-containing products there is an estimated overall free ENM release rate of 0.1-2.0 % (Keller et al., 2013).

In this life cycle phase by-product nanowaste can occur and subsequently be internally recycled or directly transferred to a WTP. Additionally, unintended nano-contaminated waste can arise. If this waste is collectable, it can also be transferred to a WTP (Nowack et al., 2013; Part et al., 2018a). Overall, 20-80 % of the collectable nanowaste from this life cycle step will end up in a landfill. Emissions in form of nano-pollution from the production of ENM-containing products may enter the air (10-40 %) or water prior to WWTP (10-40 %) (Keller et al., 2013). In this life cycle stage as well, the figures have to be seen as general ENM release rates from different host matrices.

3.3.3.3 Use-phase

These first two life cycle steps are followed by the use-phase of a PNC. According to Keller et al. (2013) the rate of ENM release in this phase is between 0-95 %. Typical release mechanisms in the use-phase of an ENM-containing plastic product are mechanical stress, photo-oxidation and/or weather related abrasion (Keller et al., 2013; Song et al., 2017). Koivisto et al. (2017) examined the release of ENMs through UV exposure. At a dose of about 150 MJ/m^2 they found a release rate of fragments and ENMs between ca. 10^{-4} to 10^3 mg/m^2.

Released fragments from the use-phase can either be collectable (i.e. nanowaste) and consequently enter a waste collection system intentionally; or they can be uncollectable in the form of diffuse emissions (i.e. nano-pollution) (Part et al., 2018a). The latter can unintentionally find its way into the waste water collection system through different washing processes (5-95 %). Furthermore, there is a possibility that the uncollectable nano-pollution is released into the air (0-5 %) and into the soil (0-95 %). In the use-phase it is important to keep the different periods of use in mind. On the one hand this can be just a few seconds regarding plastic packaging whereas, on the other hand, decades like it is the case for different coatings (Keller et al., 2013; Nowack et al., 2013; Boldrin et al., 2014).

3.3.3.4 Recycling

PNCs that enter recycling facilities enter this life cycle stage intentionally (Duncan, 2011; Silvestre et al., 2011). Generally, ENM-containing plastics can be recycled chemically, thermally or physically. These different recycling types in further consequence influence the fate of ENMs (Part et al., 2018a).

During mechanical pre-treatment processes (e.g. sieving, shredding) PNCs can release ENMs into the air. Within this context, the release mechanisms of matrix degradation from section 3.3.1.4 are of relevance. During recycling processes mainly the workers in recycling plants are exposed to released ENMs (Nowack et al., 2013; Part et al., 2018a). Post-consumer uses after recycling processes could lead to novel release pathways through cross-contamination effects, as, for example, is the case with recycled plastics used in textiles (Nowack et al., 2013).

Overall, the type of stress applied and the location of the incorporated ENMs (see also section 3.3.6 for more information) are important in the context of ENM release during recycling processes (Part et al., 2018a).

3.3.3.5 Waste water treatment

ENMs can enter WWTPs through different pathways, but mainly as unintended nano-emissions. These emissions can either be free ENMs or composites of ENMs and polymers (Part et al., 2018a). One possible way of unintended release is during the washing of different plastic products like textiles which contain ENMs (Greßler et al., 2014). Another unintended way into WWTPs is through weathering processes of ENM-containing plastic products such as exterior paints. The thereby released ENM fraction can then get into the municipal canalization system via the rainwater (Part et al., 2015).

In WWTPs free ENMs as well as still embedded ENMs can be released into different environmental compartments during distinct treatment processes (e.g. different preparation and water cleaning steps, biological degradation processes, chemical reactions) (Keller et al., 2013; Song et al., 2017). However, the major part of the ENMs enters the sewage sludge after sedimentation processes (75-97 %) and a smaller part stays in the effluent (3-25 %). ENMs contained in the effluent can be released into the environment after leaving the WWTP (Keller et al., 2013). Because of legal requirements in Austria, most of the sewage sludge from WWTP is burned. Only a small part is used for agricultural purposes (BMNT, 2017a). The figures in this section are not explicitly valid for PNCs but have to be seen as general rates of release for ENMs from different matrices.

3.3.3.6 Mechanical-biological treatment

Nanowaste is intentionally sent to MBWTPs. These treatment plants can be operated aerobically or anaerobically. ENM-containing plastic waste enters this EOL stage mainly as post-consumer waste from the residual waste collection system. The aims of such a treatment plant can be different. In the context of this thesis there are two main important targets. The first one applies to the creation of biologically stable landfill material. This stability is regulated in the Austrian Landfill Regulation with a content of total organic carbon (TOC) < 5 % DM and with an energy content < 6 600 kJ/kg DM (DVO, 2008). Secondly, a high caloric fraction for a further incineration with energy and heat recovery shall be generated (DVO, 2008).

During different *mechanical process* steps (e.g. shredding, sieving) in a MBWTP the release mechanisms stated in section 3.3.1.4 are of relevance (Nguyen et al., 2011; Froggett et al., 2014; Duncan, 2015; Koivisto et al., 2017; Singh et al., 2019). Considering the *biological treatment steps*, in existing studies reviewed by Part et al. (2018a) transformations of all ENM surfaces were observed but no explicit ENM release was stated.

Overall, only very few studies on the release of ENMs from aerobic and anaerobic treatment plants have been performed so far (Part et al., 2018a).

3.3.3.7 Incineration

The EOL phase incineration also includes intentionally introduced nanowaste. Waste with a high organic fraction (e.g. household waste) which cannot be composted or treated in biogas plants has to be pre-treated before landfilling, according to the Austrian Landfill Regulation (DVO, 2008). Incineration serves as an opportunity to fulfil the requirements of this regulation. In a WIP, waste is oxidized at high temperatures (850-1100 °C). Moreover, an energy and/or heat recovery is possible in combination with an incineration process (Part et al., 2018a). Especially high calorific plastic waste is well suited for energy recovery. Due to this fact, also some cement kilns, paper industry etc. use plastic waste to generate energy and/or heat. For the purpose of simplicity, these co-incineration plants as well as municipal solid WIPs are subsumed under WIPs in this work.

ENMs which enter a WIP can be released through the mechanisms of mechanical pre-processing and the incineration process itself (Greßler et al., 2014). The release mechanisms depend on the chemical specification of the ENM, its mass-percentage in the PNC and its size (Singh et al., 2019). Polymeric host matrices of ENMs mostly undergo a complete thermal decomposition in a WIP (Johnston et al., 1988).

Due to the different chemical and physical properties of ENMs, Mueller et al. (2013) state two distinct factors that determine the behaviour of ENMs during combustion. The first factor considers the type of binding. If there is a loose binding, ENMs can easily be released from their host matrix and in further consequence these ENMs are likely to be discharged together with the flue gas whereas, if there is a tight bonding, ENMs are not likely to be released and thus these ENMs will probably accumulate in the bottom ash. As a second factor, the melting and boiling behaviour of specific ENMs plays an important role. In case of thermally unstable ENMs (mainly organic nanoadditives), the boiling is reached during the incineration process. Thus, it is likely that these materials first evaporate and then recondense in the flue gas stream after cooling. It is assumed that, following this process, a reformation into the pristine nanoscale form is unlikely. Additionally, a reformation of molten ENMs into their original nanoscale form is not likely. Especially ENMs with isolated carbon should completely oxidize during the incineration process (Mueller et al., 2013; Part et al., 2018a). Thermally stable nanoadditives (mainly inorganic ENMs) whose melting and/or boiling point is not reached are likely to end up in the bottom ash. These ENMs mostly remain morphologically unchanged and the size of the primary ENM particles stays the same. All aerosolized ENMs may be released into the air if they are not retained in the filtering systems (Mueller et al., 2013; Greßler et al., 2014; Watson-Wright et al., 2017; Part et al., 2018a; Singh et al., 2019). However, according to Förster et al. (2016) in the flue gas duct, up to 80 % of the ENMs can be scavenged.

ENMs released during the incineration process can build aggregates amongst each other as well as with ash components due to encapsulation. It was also observed that, because of melting processes, the initial size of ENM particles can increase. In both cases, the overall size is increased and thus the filtration efficiency heightened (Vejerano et al., 2014; Förster et al., 2016; Part et al., 2018a). Generally, the ENM size in the fly ash plays an important role. ENMs < 100 nm are only partially retained by the filter. ENMs > 100 nm can be retained more efficiently (Greßler et al., 2014). Furthermore, ENMs may undergo chemical transformation processes and reactions with other waste fractions (Vejerano et al., 2014; Förster et al., 2016; Part et al., 2018a) or new additional nano-emissions may occur through secondary re-formation processes (Part et al., 2018a).

Singh et al. (2017) concluded that the release of ENMs may also depend on the specific combustion conditions. When there are insufficient oxygen conditions, a reduced ENM release could be observed. Regarding final temperature, an increased heating led to an increase in the NP number whereas increased temperatures during the heat treatment of released aerosols decreased the NP number. A crucial factor for the latter one may be the additional combustion of carbonaceous compounds to CO and CO_2.

Nowack et al. (2013) and Keller et al. (2013) stated that the ENM release rate to the air is about 0.05-1 %, into the bottom ash between 1-50 % and/or into the fly ash among 50-98 %. These figures are general release rates and do not specifically refer to PNCs.

Overall, the majority of ENMs are found in the solid combustion residues after incineration processes with the largest share in the bottom ash and only small amounts in the fly ash (Vejerano et al., 2014; Part et al., 2018a). In Austria, these two resulting waste fractions are afterwards deposited on appropriate landfills.

3.3.3.8 Landfilling

In the case of landfilling, there is an intentional shipment of nanowaste into this life cycle phase. ENMs can enter this EOL stage directly (manufacturing waste or incorporated in products) or as secondary waste from MBWTPs, WIPs or WWTPs (Part et al., 2018a).

Landfill waste in Austria has to fulfil several storage criteria according to the Austrian Landfill Regulation (DVO, 2008). Especially household waste has to be pre-treated to reach specific threshold-values before landfilling. Hence, the majority of the ENMs from plastic products enters landfills as secondary waste from MBWTPs and WIPs. Another important legal requirement for landfills in Austria in the context of ENM release is a surface cover and a bottom-liner system with a leachate collection for wastes from MBWT and incineration (DVO, 2008).

Several mechanical and chemical mechanisms (e.g. washout, natural weathering, degradation) may lead to an ENM release from deposited nanowaste and/or transformations of ENMs. Especially degradation mechanisms like hydrolysis and enzymatic degradation play an important role with respect to landfills (Bikiaris, 2013; Duncan, 2015).

Landfill internally released ENMs may be in further consequence emitted with liquids into the leachate or into the air in a gaseous state. In this context, installing specially engineered barriers makes a possible ENM release more difficult. In a controlled landfill the resulting leachate is collected and thus an unintended release into the soil and subsequently into the groundwater is unlikely. Furthermore, if there is a surface cover an ENM release into the air (e.g. due to wind erosion) is also unlikely. Contrary to that, if a landfill is rudimentary and not well controlled (there are a lot of open-dumps worldwide) there could also be a release from weathered not pre-treated PNCs into the air via dust or into the soil and consequently into the groundwater. Also dissolved ENMs in leachate can be introduced into the soil and groundwater in poorly controlled landfills (Nowack et al., 2013; Reinhart et al., 2016; Part et al., 2018a).

In the case of leachate, the location of the ENM in the polymer matrix is important. Most ENMs are tightly embedded into their polymeric host matrix and thus a release into the leachate is unlikely. However, under harsh environmental conditions (e.g. high or low pH), ENMs may be released into the leachate due to chemical processes (Reinhart et al., 2016; Part et al., 2018a). So far, some studies have already observed nanoparticulate fractions in the analyzed leachate. However, it is important to consider that these fractions may also be generated from natural sources and not only from ENMs (Part et al., 2018a). In recent studies, heteroaggregation of natural colloids with ENMs in liquid media has been given an important role considering the fate of ENMs (Praetorius et al., 2012).

Various factors like environmental conditions, leachate characteristics, pH value, water content, type of landfill etc. determine the ultimate fate and remaining of ENMs in a landfill (Part et al., 2015; Part et al., 2018a). Chemical and biological processes like reduction, aggregation, agglomeration and degradation processes also play a role within that context (Keller et al., 2013; Song et al., 2017; Part et al., 2018a).

3.3.4 Release of specific ENMs

In this section, the release and migration of the six most frequently nominated ENMs from the data evaluation (c.f. section 3.2) are described in detail. This selection is justified by the fact that these six ENMs count for about two thirds (i.e. 66 %) of the total nominations of ENMs in plastic products mentioned in the analyzed scientific papers. Moreover, these ENMs are generally the most frequently utilized ones in those products that most of the currently available release studies focus on.

3.3.4.1 CNTs

In general, CNTs can be released as free ENMs, aggregates, agglomerates and/or CNT particles still embedded into plastic debris (Nowack et al., 2013). Overall, there are two main scenarios for a potential release of CNTs out of a polymer matrix. The first one is in accordance with *high energy processes* (e.g. recycling, incineration) (Nowack et al., 2013). The second release scenario involves release due to *low-energy processes* (e.g. consumer use, environmental degradation in landfills) over a long time scale (Nowack et al., 2012; Nowack et al., 2013).

In a study performed by Wohlleben et al. (2016), the release of CNTs from elastomers during the use-phase of tires was analyzed. In their experiment, they mechanically and chemically imitated the wear of vehicle tires. Overall, CNT reinforced tires suppressed the release of elastomer components in contrast to tires without nano-reinforcement. In a TEM analysis conducted by Wohlleben et al. (2016), a release of free CNTs could rarely be observed. Within this experimental framework, the authors state that the release of free CNT particles ranges from 0-1 %. Overall, it should be noted that the experimental setup did not fully reflect the circumstances of the real world.

Brame et al. (2018) performed a release study on MWCNT-enabled coatings, where samples were exposed to UV weathering and artificial abrasion. In a following SEM analysis protruding MWCNTs from the matrix were found but no individually released MWCNTs could be observed. Within the framework of the performed analysis, the authors concluded that CNTs can be released in five different ways: (1) completely or (2) partially embedded within the polymer matrix, as (3) individual or (4) fragmented particle or as (5) agglomerated CNT structures. Although no release of free CNTs was found in this experiment, some other analyses already detected the release of free individual CNTs during mechanical influence (Schlagenhauf et al., 2012; Duncan, 2015).

If a PNC gets into a WIP as nanowaste, the incorporated CNTs should theoretically be burned and mineralized. Nevertheless, some of the CNTs might survive for a short period of time at lower temperatures (e.g. poorly controlled incineration < 600 °C) and thus may lead to an airborne CNT release depending on the type of WIP and the respective filter systems (Roes et al., 2012; Watson-Wright et al., 2017). In an experiment performed by Bouillard et al. (2013) not combustible CNTs were observed. Regarding CNT release during recycling processes, CNT-containing matrix fragments were found, but no free CNTs could be detected (Boonruksa et al., 2015; Part et al., 2018a). Considering CNTs in landfills, there can be a release through degradation and/or transformation processes of the composite. This release depends on the structure of a landfill and whether there are some barriers, such as base sealings etc. (Nowack et al., 2012).

3.3.4.2 Nanoclay

There are some concerns about a potential release of nanoclay platelets, nanoscale platelet fragments or ionic residuals of nanoclays (Duncan and Pillai, 2015). In the context of ionic residual release, it is important to note again the chemical formula of MMT $(Na,Ca)_{0.33}(Al,Mg)_2(Si_4O_{10})(OH)_2 \cdot nH_2O$ which is mostly applied as nanoclay in plastic products (Weiss et al., 2006).

In a review performed by Duncan and Pillai (2015) concerning the release of different ENMs from PNCs, some studies about nanoclay were included. There was a general implication that processing conditions during and after the manufacturing of PNCs can have a significant impact on the release of ENMs. Overall, the studies showed a higher release of Si compared to plastic products with no embedded nanoclay. Furthermore, some of these studies reported a release of Al but only under extreme conditions (e.g. high pressure, high temperature) (Bott et al., 2014; Duncan and Pillai, 2015). The higher release of Si can be explained through the direct exposure of the MMT silica layers to the external environment. A release of whole nanoplatelets seems mostly possible for surface-bound nanoclay platelets (Duncan and Pillai, 2015).

An analysis with TEM performed by the EFSA (2015) on the migration of nanoclay from PNCs into food materials showed some released and partially exfoliated layers of nanoclay. However, it is not expected from EFSA that nanoplatelets which are nano-sized in only one dimension migrate from a polymer matrix. This is because of the large lateral size of nanoclays in the other two dimensions (Duncan and Pillai, 2015). Han et al. (2018) also performed a TEM analysis on the release of nanoclay platelets from two aged LDPE food packaging samples under influence of accelerated weathering (UV irradiation and ozone exposure). They observed a release of clay NPs with a measured size between 2 to 8 nm. In a further investigation with ICP-AES it was found that the released mass of elements was much higher for ozone aged samples than for UV aged samples (almost 10 times higher). This is due to the fact that UV photo-oxidation is a surface-limited reaction. It was also observed that PNCs with a higher amount of nanoclay undergo larger property changes and have enhanced ENM releases compared to PNCs with less incorporated nanoclay platelets. Thus, the authors concluded that there is an accelerated photo-oxidation degradation with the presence of more clay mineral oxide ions within nanoclays. When interpreting the results of this experiment, it is important to note that real life ageing processes are usually more complex than those under experimental conditions. Hence, there is just a limited use of these models for practical applications (Han et al., 2018).

In an incineration experiment on nanoclay-containing plastics performed by Ounoughene et al. (2015), a thermal ablation of the polymer (i.e. PA6) has been found. As a result of this, an increased concentration of nanoclay on the surface was observed which delayed the complete oxidation of the PNC.

3.3.4.3 Ag

During different release mechanisms (e.g. washing, weathering) nano-Ag particles can undergo alteration processes. Thus, it is important to consider not only an elemental particle release but also a possible release of Ag-ions (Nowack et al., 2012; Bott et al., 2014; Duncan and Pillai, 2015). Furthermore, in a sulphurous environment nano-Ag can be reduced to silver sulphide (Ag_2S) and in washing processes with detergents a conversion of the incorporated nano-Ag to silver chloride (AgCl) has been found (Impellitteri et al., 2009; Nowack et al., 2012; Mitrano et al., 2016; Part et al., 2018a).

So far, a few studies on the release of nano-Ag from PNCs have been carried out. Most of these studies were dealing with the release of ENMs from polyolefins (i.e. PP, PE (HDPE, LDPE), PET, PS, PVC) (Duncan, 2011; Bott et al., 2014; Duncan and Pillai, 2015). Abrasion, dissolution, adsorption and desorption, microbial transformation and oxidation are the primary processes which lead to nano-Ag release (Nowack et al., 2012).

In a review of Duncan and Pillai (2015) there are some examples for the release of nano-Ag. There may be an ENM release from textiles into washing water, facade paintings into rainwater, plastic food containers into liquid food etc.

Regarding textiles with incorporated nano-Ag, Keller et al. (2013) stated a possible ENM release between 0-100 % during washing processes. For this release, no specification of the incorporated Ag was given. Koivisto et al. (2017) stated a nano-Ag release during washing processes of 0.5-35 %, of which the main part is in ionic form whereas Geranio et al. (2009) demonstrated an ionic Ag release from textiles in the nano-size range of < 450 nm as well as a particle release in the nano-size range of > 450 nm. This release study was performed at a high pH value. The latter size range may be due to mechanical abrasion. Several other studies performed on the release of nano-Ag particles through hydrolysis could not observe free released ENMs. This may be due to the fact that nano-Ag forms salt in aqueous environments and thus it is difficult to distinguish between the release of a salt and Ag-ions (Froggett et al., 2014).

In the case of facade paintings, a substantial release of nanoscale Ag particles during weathering processes has been found (Kaegi et al., 2010; Hincapié et al., 2015).

Šimon et al. (2008) studied the migration of ENMs from polymer packaging. In their calculations of diffusion rates, they found that there is a migration of nano-Ag from food packaging to the food after one year of contact at different temperatures.

If nano-Ag gets into a WIP, it is likely that the majority of this ENM remains in the bottom ash and only a small fraction will evaporate (Mueller et al., 2013; Part et al., 2018a). In Austria, the bottom ash is brought to an adequate landfill where the nano-Ag particles may be released into the leachate depending on the existence and quality of barriers within the landfill (e.g. sealing). A study by Mitrano et al. (2016) observed a Ag concentration in the low µg/L scale with associated nano-particulate matter (PM) in the leachate.

Considering biological treatment, existing nano-Ag particles adsorbed or complexed with solid natural organic matter may be transformed to stable Ag_2S or AgCl resulting in a reduced mobility and thus reduced ENM release (Gitipour et al., 2013; Part et al., 2018a).

Overall, the released amount of nano-Ag varied in the performed experiments. Sometimes no released quantities were measured at all. This is due to the different applied experimental conditions and test materials used. Moreover, the units of measurement differ between these studies (e.g. µg/g, mg/l, ppm) (Bott et al., 2014; Duncan and Pillai, 2015; Part et al., 2018a).

3.3.4.4 TiO_2

In the long term it is likely that released nano-TiO_2 particles do not stay in the nanoscale, unless these particles have been stabilized significantly during their manufacturing process (Keller et al., 2013).

Regarding release of nanoscale TiO_2 during washing processes, Koivisto et al. (2017) stated an agglomerate release of 0.01-3.4 %. Nowack et al. (2012) concluded in a scenario analysis that nano-TiO_2-containing paintings have to undergo mechanical or chemical degradation mechanisms before an ENM release is possible. This is because the ENMs are incorporated into a solid polymeric matrix in the case of paintings. However, if there are external influences as those mentioned above, a release of free ENMs and other particles is possible. So far, most of the studies on nano-TiO_2 release of coatings concluded that a release of ENMs is possible but the rates seem to be low (Kaegi et al., 2010; UBA, 2014; Hincapié et al., 2015).

Due to the high boiling point of TiO_2 (2900 °C) this ENM remains morphologically unchanged during incineration processes (Mueller et al., 2013). Hence, it is very likely that nano-TiO_2 stays in the bottom-ash after the incineration process (up to 91-96 %) (Börner et al., 2016). However, a part of the ENM still can be aerosolized and in further consequence released to the air if it is not retained by a filter (Singh et al., 2019). Watson-Wright et al. (2017) speak of concentrations about < 0.03 wt%. In some recent studies, it was found that aerosolized TiO_2 went through a chemical transformation and generated toxic and carcinogenic polycyclic aromatic hydrocarbons (PAHs) during incineration (Singh et al., 2017; Singh et al., 2019).

In Austria, nano-TiO_2 particles which are present as bottom ash residues after an incineration process are deposited on landfills. These ENMs may then be released to the leachate according to the presence of barrier systems in the respective landfill. A study has observed ENMs in an analyzed leachate consisting to a certain extent of TiO_2 (Mitrano et al., 2017).

3.3.4.5 SiO_2

According to a safety assessment of the EFSA (2014) no migration of nanoscale SiO_2 into food and beverages could be observed. Moreover, in a review performed by Duncan (2015) no individually released nano-SiO_2 particles were detected. However, it is assumed that there could be a release of free nano-SiO_2 with a subsequent adherence of these particles to larger abraded particles which means that free ENMs cannot be detected. Considering the particle itself, it is expected that nano-SiO_2 remains in its particulate form due to its tendency of insolubility (Keller et al., 2013). Kaegi et al. (2010) speak of little release of nano-SiO_2 from coatings (especially exterior facade coatings).

3.3.4.6 CB

One of the first migration studies on nano-CB from PNCs was performed by Bott et al. (2014). In their investigation under ambient conditions (no mechanical, chemical or physical influence), they could show with an AF4 experiment that there is no nano-CB migration from either an LDPE nor a PS matrix.

In an evaluation of EFSA (2005), it was concluded that nanoscale CB does not migrate out of PNCs into the environment.

Wohlleben et al. (2016) performed an analysis on the release of nano-CB mimicking the wear of car tires made of PU and natural rubbers. Compared to vehicle tires made of elastomers without nano-reinforcement, tires with embedded nanoscale CB have reduced release rates of elastomer-components. This is because of the photo-protective filler properties of nano-CB. However, the possibility of a release of free NPs from tires has been detected, although this process could rarely be observed with TEM in the performed analysis. With respect to a possible ENM release, secondary release events have to be considered as well (e.g. tire abrasion on the road which is exposed to sunlight and thus releases ENMs). Overall, Wohlleben et al. (2016) stated a possible release of free nanoscaled particles of 0-0.045 wt%. Nevertheless, it has to be considered that the experimental setup does not fully reflect the circumstances of the real world.

3.3.5 General composition and morphology of released particles

In reviews performed by Froggett et al. (2014) and Duncan (2015) it was found that mainly composites of matrix components were released during different exposure scenarios (e.g. machining, weathering, incineration). In nearly all analyzed articles, at least a part of the released components contained some ENMs. These incorporated or associated ENMs were partially or fully embedded into the released fragments of the host material and mostly irregular in shape (Duncan, 2015). Furthermore, ENMs that are still incorporated in polymeric fragments may protrude from them. These protrusions are material-dependent in relation to the roughness of the polymeric host matrix (Hirth et al., 2013). Highly elastic materials (e.g. Nylon, PE) tend to inhibit the formation of protrusions. This is because the polymeric host material will encapsulate the ENMs rather than break off and expose them (Duncan, 2015). Indeed, despite multiple findings of released matrix composites, a release of free ENMs has also been observed in several studies after exposure to various external forces (Froggett et al., 2014; Duncan, 2015).

Overall, it is important to mention, that not all of the observed released nanoscale fractions are pristine ENMs that have been embedded in the polymer matrix before. Nanoscale particles can also arise out of degraded fractions of the polymeric host matrix itself (Froggett et al., 2014; Duncan, 2015).

3.3.6 Categorization framework for the location of ENMs in a matrix

As potentially hazardous effects of ENM-containing products depend to a significant extent on the location of the nanostructure in a matrix, it is essential to know about this internal position of the ENMs to identify likely exposure pathways and make realistic evaluations (Foss Hansen et al., 2007; Foss Hansen et al., 2008). Such a localization of the nanoscale structure in a matrix is enabled by the categorization framework of Foss Hansen et al. (2007). Within this framework there are three main categories with corresponding subcategories:

1. Nanostructure in the *bulk* (nanostructured in three dimensions)
    a. Only one type of material
    b. Two or more different materials
2. Nanostructure on the *surface* (different nanoscales)
    a. Surface is structured on nanoscale (surface and bulk same material)
    b. Un-patterned film on a substrate (film in nanoscale, different material)
    c. Patterned film on a substrate (either film or pattern in nanoscale)
3. Nanostructured *particles in a matrix* (min. two dimensions in nanoscale)
    a. NPs bound to a surface of a solid structure
    b. NPs are suspended in a liquid
    c. NPs are suspended in solids
    d. NPs are suspended in air/gas (=airborne NPs)

ENMs situated at the interface between PNC and the exterior medium can be released directly through the mechanisms of dissolution and/or desorption. Contrary to that, ENMs incorporated in the interior of a PNC would first have to diffuse to the interfacial boundary before a release to an exterior medium is possible (Duncan and Pillai, 2015). Within the framework of Foss Hansen et al. (2007) this means that surface-bound ENMs are released more easily than ENMs bound in a matrix or in the bulk.

3.4 Additional notes on nanotoxicity

All released ENMs are capable of inducing toxic effects (EC, 2012). Due to different exposure pathways, these toxic effects may pose a risk to humans and nature (Tsuji et al., 2009; EC, 2012).

With respect to human beings, there are four different exposure pathways of released ENMs from a (polymeric) host matrix, in particular oral, dermal, by inhalation or by injection. This exposure depends on different factors such as the characteristics of the released ENMs, the matrix of the product itself, the quantity of ENMs used in a specific product, the particle size distribution, the frequency and type of use of the respective ENM-containing product etc. (Tsuji et al., 2009; EC, 2012).

With regard to nanotoxicity, the material-specific and intrinsic toxicity of ENMs are of great importance (Donaldson and Stone, 2003; Hoet et al., 2004). In the category of material specific properties, the shape of the constituents (particles, fibres or platelets) of the used ENMs play an important role (Tsuji et al., 2009). With respect to the intrinsic toxicity, different ENMs have different toxic properties. The most common effects studied in different experiments are those of causing oxidative stress, inflammatory responses and genotoxic effects (EC, 2012). Depending on the specific ENMs, there are, for example, effects on the lung (e.g. impairment, inflammation) that can be attributed to the ENMs Ag and TiO_2 (Heinrich et al., 1995; Bermudez et al., 2004; Sung et al., 2008). Moreover, carcinogenic effects have been observed in connection with nanoscale TiO_2 and CB (IARC and WHO, 2010; EC, 2012).

In a risk assessment performed by the NIOSH (2005) it has been established that nanotoxicity is a function of dose in relation to the surface. Despite this finding, the dose-response ratio of ENMs is still relatively difficult to predict and has so far mainly been studied in rats, hares and mice. However, the concentrations used within these experiments are usually higher and have a more targeted effect on the subject than it is the case in reality. Thus, these results may not be directly transferable to humans (Tsuji et al., 2009; EC, 2012). Nevertheless, these results should not be underestimated and must be taken as a first starting point for further experiments and activities in accordance with the precautionary principle.

4. Material flow analysis

Figure 8 shows the system components of an MFA as well as the relation to environmental fate models (EFMs). With an *MFA,* all life cycle stages of an ENM-containing product can be taken into account. These stages are: the manufacturing of ENMs, the production of ENM-containing products, the use-phase (different service lives for different products) and finally several EOL stages.

For *EFMs*, transport processes are also taken into account as shown in Figure 8. Furthermore, within the performance of EFMs a specification of ENMs is done and behavioural changes of ENMs are considered (e.g. chemical, physical). Hence, these models describe the ultimate distribution and fate of ENMs in different environmental compartments. EFMs can be based on the values obtained from a preliminary MFA (Nowack, 2017). In this thesis EFMs are not considered in detail, but only mentioned for the sake of completeness.

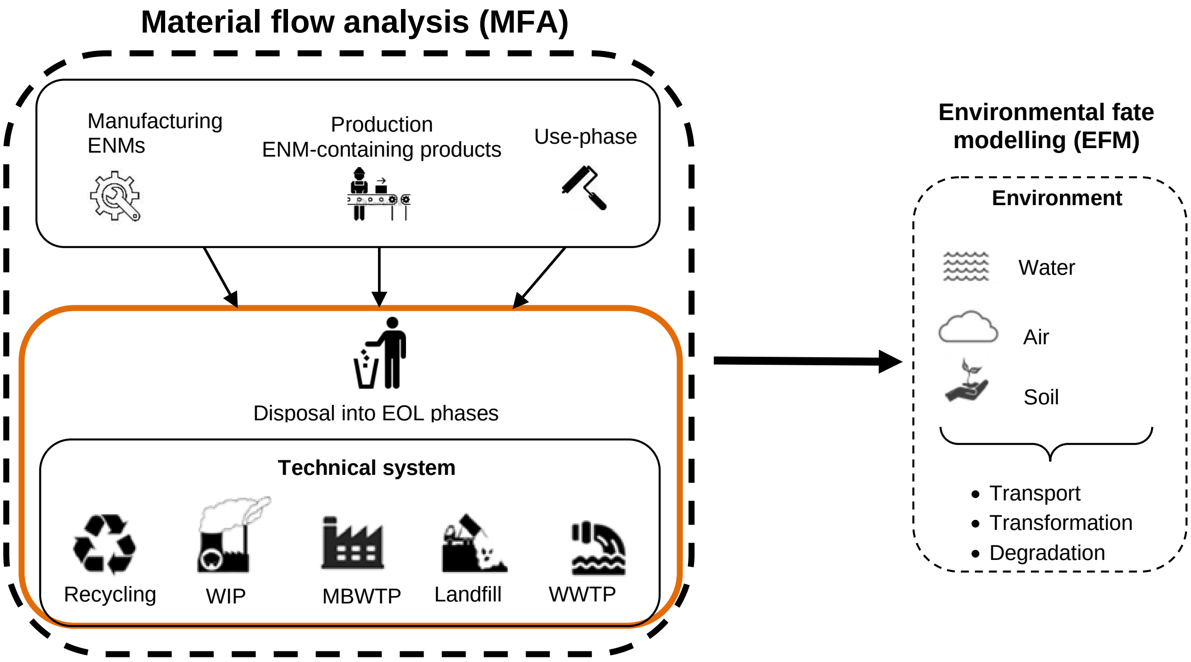

Figure 8: System components of an MFA with special focus on the technical EOL system and the relation to EFMs with its components, modified from Nowack, 2017.

4.1 MFA in general

MFA as an exposure assessment tool is capable of tracking and assessing flows and stocks of materials within a spatially and temporally predefined system (Brunner and Rechberger, 2004). This predefined system for instance can be an entire life cycle of a product from the production stage to the use-phase and up to the different EOL stages within one year (Nowack, 2017). Due to the fact, that MFAs are based on the law of conservation of matter (Brunner and Rechberger, 2004), matter cannot be destroyed and thus cannot get lost within the defined system (Hill, 1981).

For the performance of an MFA, different input data and information is needed. There are three different categories of information availability: (1) Some MFAs are based on available information only. This type of MFA is the most efficient, simplest and least expensive. The information available is used to perform an MFA of a certain system (Baccini and Brunner, 1991). (2) MFAs based on field measurements are particularly important, if there is no or just little information available for a certain field and its processes of interest. Especially in the field of waste management there are often unidentified inputs and outputs (e.g. into incineration plants). Unfortunately such field measurements are not always possible (e.g. for monetary or technical equipment reasons) (Baccini and Brunner, 1991). (3) Depending on the availability of information also a combination of existing information with field measurements is possible (Baccini and Brunner, 1991).

After defining a system and integrating all available information, the inputs and outputs of a specific system can be analyzed. During this analyzing process sources, pathways and sinks are identified and become visible. Furthermore, the potential accumulation or depletion of stocks and possible environmental release hot spots can be detected (Brunner and Rechberger, 2004).

According to Brunner and Rechberger (2004) there are five different main objectives of MFAs: (1) the depiction of a system by clearly defined terms, (2) a major reduction of the complexity, (3) a quantitative assessment of all relevant flows and stocks, (4) the delivery of transparent and reproducible results and (5) the use of these outcomes as a sound basis for future decisions.

4.1.1 MFA types

Different model types are characterized by their basic model structure and their underlying mathematical modelling approach (Nowack, 2017). There are static and dynamic model types. Static MFAs consider only a certain period of time without temporal fluctuations in different input parameters. Dynamic MFAs on the contrary, visualize the time-dynamic performance of a complex system under consideration. It is possible to add up flows over several periods of time, consider life span dependent behaviour of products and locate emission hot spots as well as environmental sinks (e.g. soil, air, water) (Bornhöft et al., 2016; Nowack, 2017; Song et al., 2017).

Overall, MFA models are quite robust. Limitations might be given by a lack of data or by the uncertainty of different input parameters (Nowack, 2017).

4.1.2 Data reconciliation and uncertainty analysis

Subjectivity is an inherent part of uncertainty consideration in MFAs. Thus, a robust approach for the translation of the data quality obtained by calculations from MFAs into associated uncertainties is necessary (Laner et al., 2016).

In the following section different methods of uncertainty considerations will be presented. It is important to mention that the listed methods are only generic. The selection of the presented methods is based on frequently mentioned uncertainty considerations in various scientific articles dealing with MFAs.

4.1.2.1 *Statistical methods*

Statistical methods serve as a tool to generate knowledge about data obtained from several inaccurate samples. If data is available for one specific entity, different statistical calculations can be applied. Thus, it is possible to calculate the mean value, standard deviation and create an overview of the distribution of the used samples. However, if this data is strongly deviating or has a skewed distribution, instead of the mean value and the standard deviation the median value and interpercentiles may be used (Brunner and Rechberger, 2004).

Nonlinear data reconciliation problems can be addressed using iterative techniques based on successive linearization and analytical solution finding of a linear reconciliation problem. In each of the iterations, a Gauss-Jordan elimination process of the original linear/linearized constraint matrix is applied. With the method of error propagation, corresponding uncertainties can be determined (Cencic and Rechberger, 2008).

Sometimes there are only single values available. These values may be acquired from an experiment, interview, measurement etc. In this case, an uncertainty estimation of this particular value can be made by an evaluation of its original source or context (Hedbrant and Sörme, 2001; Brunner and Rechberger, 2004).

4.1.2.2 *Sensitivity analysis*

Sensitivity is defined as the relative change in output induced by a unit relative change in input parameters (Morgan and Henrion, 1990; Finley and Paustenbach, 1994; Gottschalk et al., 2010). In order to determine each parameter's contribution (percental and relative) to the total sensitivity the data obtained from a sensitivity analysis can be summed up. Thereby, the resulting ranking of the modelling parameters serves as an identification tool of the most significant influential parameters. These most significant parameters may in further consequence be studied in more detail in order to take measures to reduce ENM releases and resulting environmental impacts (Brunner and Rechberger, 2004; Gottschalk et al., 2010).

If the available data for the performance of a sensitivity analysis is uncertain, then the reliance in the exposure assessment has to be placed with care as well. Contrary to that, if the available data is robust, the credibility of the results may be higher (Gottschalk et al., 2010).

4.1.2.3 *Scenario modelling*

Different scenarios can be modelled in order to cover a wide range of possible occurrences and behavioural patterns. Hence, the uncertainty in input parameters can be addressed. The most notable scenarios are: a minimum and maximum scenario or a realistic and worst case scenario respectively (Nowack, 2017).

4.1.2.4 *Monte Carlo simulation*

Monte Carlo simulation is a tool to incorporate the propagation of uncertainties into already obtained results of an MFA. This tool is used if there are large deviations or if the data is not normally distributed (Brunner and Rechberger, 2004).

Within the Monte Carlo simulation, it is assumed that the statistical distributions of the input variables are known. A computer algorithm creates a random number for each of these input variables. These numbers are in accordance with the distribution and are used in a mathematical function. If this function is calculated *x*-times, then the result will also be *x* possible outcomes. In further consequence these outcomes are evaluated with different statistical methods (Hebbeker, s.a; quoted from Brunner and Rechberger, 2004).

4.1.2.5 Bayesian modelling

Bayesian network is an uncertainty consideration approach developed for probabilistic modelling and ENM risk forecasting (Money et al., 2012). For the performance of this uncertainty modelling approach the authors developed a model named FINE (Forecasting the Impacts of Nanomaterials in the Environment). Within this model there are four different components. The first component addresses the particle behaviour of ENMs. It consists of a network of particle-related characteristics (e.g. ENM coating, diameter/fractal dimension), environmental factors (e.g. pH, temperature), fate and transport variables (e.g. biodegradation, dissolution). As a second component the exposure potential of ENMs is addressed. This part of the model is directly connected to the first component via dissolution, biodegradation and deposition. As a third part of the model the hazards and potential toxicological effects of the ENMs in question are included. They are based on expert elicitation. Fourthly, the exposure potential and hazard variables are related to a quantitative risk forecast (Money et al., 2012).

After applying the Bayesian modelling network with certain input data, the results may be valid for a considerable period of time. However, in case new data and information on ENM behaviour becomes available, it is possible to adjust and update the model (Money et al., 2012).

4.1.2.6 Modelling approach by Laner et al., 2016

Laner et al., 2016 proposed a modelling approach for the characterization of data uncertainty in MFAs. Their approach includes a data quality assessment and/or an estimation of uncertainty ranges and it is based on an assessment established by Weidema and Wesnæs (1996) as well as on an evaluation proposed by Hedbrant and Sörme (2001). The modelling approach is suitable for data obtained by expert estimations as well as from published sources like reports, studies etc. (Laner et al., 2016).

With regard to expert estimations only one data quality dimension is applied. These expert estimations need to be categorized into one of four categories which all refer to different kinds of uncertainties according to the quality of the estimation. With regard to the quality of the estimate, parameters such as the type of data used (e.g. empirical data, estimated data), transparency of the estimate, knowledge of the experts and practitioners in the respective field etc. are taken into account. Category 1 corresponds to an uncertainty of 5 %, category 2 to 14 %, category 3 to 41 % and category 4 to an uncertainty value of 125 % (Laner et al., 2016).

For published sources, the data quality assessment is based on five quality indicators: reliability, completeness, temporal correlation, geographical correlation and other technological correlation. Each of these indicators is evaluated and corresponding scores are created (Weidema and Wesnæs, 1996; Laner et al., 2016).

Based on the scores, uncertainty estimations are calculated. There are different possibilities to translate individual scores into uncertainty estimations. They can be carried out by calculation coefficients of variation (CV). These CVs assume normal distributions and contain either exponential or linear functions. Furthermore, uncertainty estimations can be expressed by applying log-normal distributions where exponential or linear functions are applied as well (Laner et al., 2016).

4.2 MFA for ENMs – model types

Due to the fact that nanotechnology and nano-based products have evolved quite rapidly in the past few years, concerns have arisen about their ultimate fate and remaining. MFAs have been used to track the flows of ENMs and visualize sources, sinks and pathways. Even if there is a lack of analytical data, as it is often the case with ENMs, MFAs serve as relevant tools to get an initial assessment (Nowack, 2017).

4.2.1 Single equations MFA

Due to the fact that about a decade ago very little data was available on the use of ENMs and their consequential potential environmental impacts, Boxall et al. (2007) created a series of algorithms to estimate the potential environmental exposure of ENMs. These algorithms, which take the form of single equations, are capable of including distinct environmental compartments. In the analysis performed by Boxall et al. (2007), several single equations were established for the environmental compartments of air, soil and water. The equations may be used to generate individual results or can be combined to achieve a comprehensive overview.

4.2.2 Excel based MFA

There are several methods for Excel based MFAs as Nowack (2017) stated in his evaluation of exposure models. All these models created in Excel are based on equations and calculations. Furthermore, some of these Excel based models use add-on packages like @Risk or Matrix (O’Brien and Cummins, 2010; Nowack, 2017).

To run these models, relevant data must be collected and added in the Excel program. Then, the model can be started and it runs for a multitude of iterations (10 000 in the analysis of O’Brien and Cummins (2010)). This iterative process leads to a fully representative convergence of the results that represent concentrations of ENMs referring to different environmental compartments (O’Brien and Cummins, 2010).

4.2.3 Web based MFA

Another model type that is available for the analysis of ENMs is web based MFA. Nowack (2017) mentioned one applied web based method in his evaluation which was performed by Liu et al. (2015). They used a simulation tool that enables rapid “what if?”-scenario analysis. It consists of different elements, for example a fate and transport model for an estimation of environmental concentration values. Within this fate and transport model various environmental compartments are linked. By means of equations, the mass of ENMs in the relevant compartments is obtained. Moreover, the web-based MFA of Liu et al. (2015) includes an assessment of the average release of ENMs into different environmental compartments. Additionally, there is a predefined parameter database available within their simulation tool. In the end, all the outcomes of the different calculation models are combined to generate a comprehensive overview. This model type is also applicable for the environmental compartments of air, soil and water (Liu et al., 2015).

4.2.4 Probabilistic MFA

Regarding PMFA, the method of MFA is extended by the production of probability distributions (Gottschalk et al., 2010). Thus with these models it is more likely to capture reality (Nowack, 2017).

PMFAs consist of a linear mathematical model with different elements in order to describe distinct processes of a defined system. Within the mathematical model an estimation of the emissions to environmental compartments (e.g. to soil, water, air) is possible (Gottschalk et al., 2010). The team of Gottschalk et al. (2010) programmed the underlying mathematical equations and carried out the PMFA with the computational tool R. Up to now, some other studies have used this proposed model structure in order to analyse material flows of different ENMs e.g. Sun et al., 2014; Caballero-Guzman et al., 2015; Sun et al., 2015 etc.

Due to the fact, that PMFAs include probability calculations, different methods of uncertainty analysis like Monte Carlo simulations and sensitivity analyses are applied (Gottschalk et al., 2010). Methods for uncertainty analysis are explained in more detail in section 4.1.2.

4.2.5 Dynamic probabilistic MFA

Dynamic probabilistic material flow analysis (DPMFA) is an analysis method based on PMFA (Gottschalk et al., 2010; Nowack, 2017) extended by the level of dynamic modelling (Bornhöft et al., 2016).

Bornhöft et al. (2016) created the DPMFA model consisting of different static building blocks which must be linked together in order to allow a process simulation and evaluation. In a further step, the authors propose a three-layer process. The first one is concerned with uncertainty analyses of the flow dependencies between the distinct system components and the absolute annual inflow. Therefore, the Bayesian probability distribution is applied. The resulting uncertainties are then run through the model using Monte Carlo techniques. Secondly, there is a layer which focuses on the time-dynamic behaviour of the model. Within this second layer, successive periods (e.g. years) represent time. For each of these periods the external inflows into the model, ENM accumulation in different stocks and ENM releases are determined. The last layer calculates the absolute material flows for a certain period of interest. For the performance of the dynamic model Bornhöft et al. (2016) implemented a software called Python.

At the moment the model type DPMFA is the most comprehensive one to track ENM flows and visualize sinks of ENMs in the environment (Nowack, 2017).

5. Case study: MFA on an ENM-containing plastic product

Before carrying out an MFA, an appropriate ENM-containing plastic product as well as relevant information and data based on the theoretical parts of this masters' thesis had to be collected and summarized for the performance of a specific case study. Therefore, in this chapter, selection criteria for the choice of one ENM-containing product will be defined. Furthermore, parameters for the planned MFA, such as system boundaries, model type and its corresponding uncertainty analysis, input data as well as TCs will be presented.

5.1 Selection of an ENM-containing plastic product

The selection of one specific ENM-containing plastic product was based on three different selection criteria which all had to be met by the selected PNC.

5.1.1 Selection criteria

Firstly, **already existing MFAs** for the specific product of interest had to be ascertained. Ideally, an ENM-containing plastic product should be chosen for which no MFA has been performed yet. As shown in Table 1, for vehicle tires containing CB as well as for nano-Ag- and CNT-containing food and beverage packaging, no MFA has been performed yet. In contrast, for the remaining four ENM-containing plastic products, in particular for all paints and for nanoclay-containing food and beverage packaging, some MFAs have already been performed.

Secondly, for the selection of an appropriate ENM-containing product the **information density** was taken into account. This refers to already existing information and useful input data for the planned MFA which could be obtained from scientific articles or other statistics. Here, information on the incorporated or associated ENM itself (e.g. wt%, size), already performed ENM release studies, TCs etc. were of importance. Considering the products listed in Table 1, for nearly all of them plenty of information (i.e. more than five relevant articles) is available. This particularly applies to the PNCs CB-containing tires, Ag-, TiO_2- and SiO_2-containing paints as well as to Ag-containing packaging. Less information is available only for CNT-containing food and beverage packing.

Thirdly, the **nominations of specific ENM-containing products** served as another selection parameter. The evaluation of these nominations was based on the information of Appendix 1. For this purpose, all product specifications were screened according to their absolute nominations as it is depicted in Table 1. Within this analysis only products with a total number of nominations equal to or greater than five were taken into account. The amount of five nominations has been chosen in order to reduce to quantity of eligible products to less than ten. As it is shown in Table 1, nanoclay-containing food and beverage packaging has 13 nominations and has been mentioned the most. All other listed PNCs, namely CB-containing tires, TiO_2-, SiO_2-, and Ag-containing paints, Ag- and CNT-containing food and beverage packaging, range between five and seven nominations.

Table 1: Evaluation of specific ENM-containing plastic products according to the corresponding available information as well as references of already existing MFAs. Only products with nominations greater than or equal to five are chosen for this evaluation. Basis: 40 international scientific articles listed in Appendix 1.

Product category	**Product Specification**	**ENM**	**≥ five nominations**	**Available information[1)]**	**Reference to already existing MFA** (exemplary)
Automotive industry	Tires	CB	6	🟩	No existing for MFA on CB in tires.
Coatings	Paints	TiO_2	5	🟩	Arvidsson et al., 2012
Coatings	Paints	SiO_2	7	🟩	Wang et al., 2016; Wang and Nowack, 2018
Coatings	Paints	Ag	5	🟩	Part et al., 2017
F&B packaging	F&B packaging	Nanoclay	13	🟩	Project study SafeNanoKap Part et al., 2018b
F&B packaging	F&B packaging	Ag	7	🟩	No existing MFA on nAg in F&B packaging.
F&B packaging	F&B packaging	CNT	6	🟧	No existing for MFA on CNTs in F&B packaging.

[1)] Explanation of the colours: red - no information = 0 relevant articles; orange - little information = between 1 and 5 relevant articles; green - plenty of information = more than five relevant articles.
"*Relevant*" refers to information needed for the planned MFA (e.g. release mechanisms, wt% of ENMs, TCs.).

5.1.2 Result

Because all the selection criteria listed in Table 1 had to be fulfilled by the PNC selected for the MFA, only CB-containing tires, Ag- and CNT-containing food and beverage packaging were eligible for selection. There is plenty of information (i.e. more than five relevant articles) available on CB-containing tires as well as on nAg-containing food and beverage packaging, whereas for food and beverage packaging containing CNTs only little information (i.e. between one and five relevant articles) is available. Consequently, the choice had to be made between CB-containing vehicle tires and nano-Ag-containing food and beverage packaging.

Carbon black **ENM-containing tires**

Figure 9: CB and CB-containing vehicle tires, modified from Surip et al. (2013).

Although the latter one has more nominations in the evaluation, CB-containing tires have been selected (see Figure 9) because the analysis of nano-Ag-containing packaging would have been less relevant for Austria as it is not explicitly authorized in Annex I of the plastic food contact materials regulation (10/2011). In Austria, materials containing nano-Ag should therefore not be produced and/or used as packaging for food and beverages.

CB particles (nano- and microsized) are already commonly used in vehicle tires in Austria and many other countries. Therefore, a lot of information and data on these materials has been published. Moreover, according to the EC (2012) CB used as tire reinforcement is the most frequently incorporated ENM in plastic products. The annual global market value for CB used in tires is about 7.3 billion EUR (EC, 2012; Wohlleben et al., 2016). Due to this high market share, there is also a high waste management as well as environmental, health and safety relevance for this additive. Although, in the empirical part of this thesis, the focus was on products available on household level, for the performance of the MFA tires of “trucks”, “articulated lorries” and “busses” have also been taken into account in order to create the most realistic results possible.

5.2 System definition

In order to perform an MFA on CB-containing vehicle tires, system boundaries had to be defined. These system boundaries are illustrated in Figure 10.

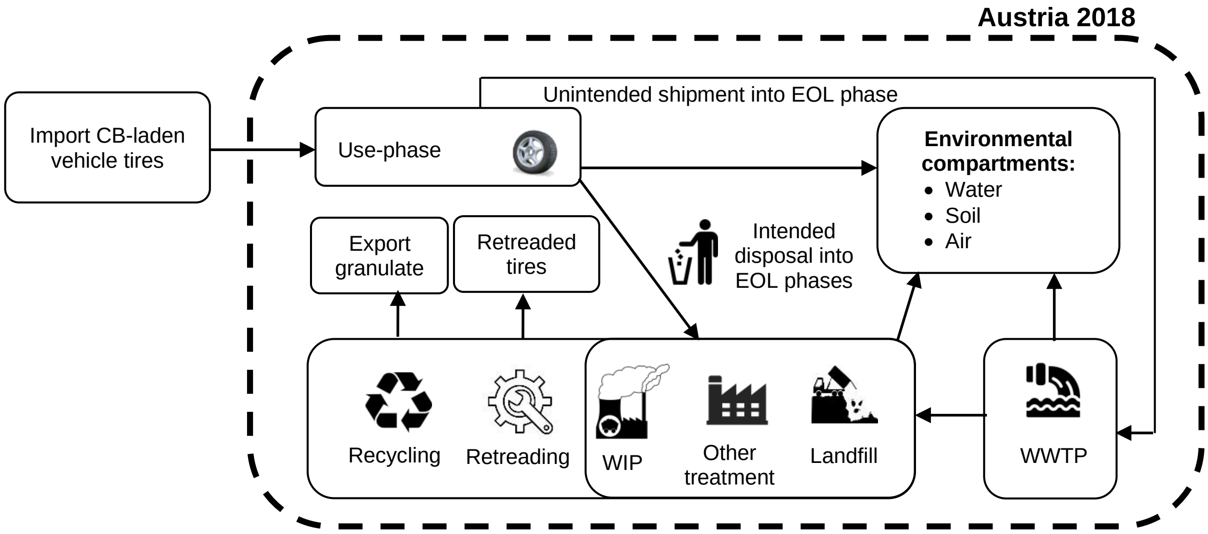

Figure 10: System components MFA on vehicle tires containing CB. The system boudaries are spatially set to the area of Austria and temporally to the year 2018, modified from Nowack, 2017.

With respect to the ***temporal boundaries,*** the period under consideration is the year 2018. A lot of already performed MFAs used an observation period of one year. This is mainly because a lot of statistics and data are collected on an annual basis (Brunner and Rechberger, 2004). In order to set ***spatial boundaries***, the area of Austria was chosen.

Due to the fact that there are no manufacturers of CB nor any vehicle tire producing facilities in Austria, the MFA starts with the import of CB-containing tires (see Figure 10). During the use-phase, parts of the CB-containing vehicle tires (i.e. plastic debris with embedded CB or individual CB-particles) are emitted as wear and tear release to the environment. Moreover, these abrasion particles can enter WWTP through the sewage system as unintended diffuse emissions (Part et al., 2018a). Subsequently, CB and parts of the original product can either stay in the effluent after clarification processes or enter the sewage sludge. The former leads to an ENM release into the hydrological cycle whereas the remaining sewage sludge is transferred either to a WIP, to other treatment facilities (e.g. bio-gas plants) or is used for agricultural purposes (Keller et al., 2013; BMNT, 2017a; Song et al., 2017).

Besides this unintentional entry into the EOL phase of WWTPs, vehicle tires containing CB can also enter different EOL stages intentionally after their use-phase. EOL tires can either be disposed of at a specialized retailer or at municipal waste collection points (BMNT, 2017a). According to the “Federal Waste Management Plan of Austria”, EOL tires enter either a material recycling plant or a WIP. Additionally, CB-containing tires that are still intact and not broken can be retreaded (BMNT, 2017b). However, in Austria only tires of trucks are retreaded because there is no economic benefit in retreading other tires (Eppel, 2019). Retreaded tires can be put back on the domestic market. EOL tires, that enter WIPs can either be burned in co-incineration plants (e.g. in cement kilns) or in municipal WIPs (BMNT, 2017b). There is no distinction between these two different incineration plants in the MFA at hand because it is assumed that the behaviour of the ENMs will not be significantly different from each other (Adam and Nowack, 2017). The residues of WIPs are sent either to landfills or underground landfills, depending on the hazardousness of the waste. In several of these different EOL stages with the corresponding process steps, composites of CB and the polymeric host material as well as free ENMs may be released to different environmental compartments (Nowack et al., 2013; Greßler et al., 2014; Part et al., 2018a).

5.3 Input data and associated limitations

For the MFA on vehicle tires containing CB for the year 2018 in Austria several different sources of input data were taken into account.

To begin with, it is important to note that the applied MFA was performed on two distinct levels. On the one hand on the *substance level* which applies to “CB” that is incorporated in tires, on the other hand on the *level of goods* which refers to the “product of vehicle tires”. However, in the context of “vehicle tires” only the elastomer part of tires, including plasticizers, chemicals for vulcanization and anti-ageing agents, other chemicals as well as fillers (e.g. CB, Silica) were taken into account. These components constitute 85 % of the weight of an average vehicle tire. Other reinforcing materials (e.g. steel, rayon) of vehicle tires were not relevant for a further consideration of CB-particles as these particles are solely incorporated in the plastic part of a tire (Continental Reifen, 2013). This assumption facilitated the MFA, because otherwise there would have been additional flows for “reinforcing material” from each EOL process. In the upcoming sections the considered part of vehicle tires is referred to as either “plastic part” or “plastic share”. It was assumed that all vehicle tires under consideration (e.g. passenger car tires, truck tires) include CB and have the same tire components (Eppel, 2019).

Overall it was assumed that CB content makes up for 22 wt% of the average vehicle tire, including all parts (Continental Reifen, 2013; Environment Canada and Health Canada, 2013). Hence, for the average plastic share of vehicle tires a CB content of 26 % was calculated.

In the next subchapters the input data for the specific processes of the applied MFA will be listed. As already mentioned before, all of these processes were analyzed on two different levels. However, in order to keep it overseeable, only the level of goods will be described in the following. As the values for CB either refer to the figures of goods, except that they are multiplied with the wt% of the CB content, or to a nano-specific TC instead of a stochastic TC, there is no need for a separate description of this level.

A comprehensive list of all input data with a corresponding detailed description as well as assumptions is available in Appendix 2.

5.3.1 Import of tires

The number of imported tires in 2018 in Austria refers to the tires of all new registered vehicles in this year, including a secondary set (i.e. winter/summer tires) for “passenger cars” and “busses”. For each type of vehicle, the specific number of tires was taken into account. For example, passenger cars have four winter as well as four summer tires and motorbikes have only two summer tires.

In order to get the whole mass of imported tires, the resulting figures of each vehicle category were then multiplied with an appropriate tire-weight. Overall, only two distinct types of tire were differentiated and taken into account, namely an average value for light tires as well as an average value for heavy tires. The weight of the average light tire was assumed to be the same for “passenger cars”, “motorbikes”, “campers” and “others”. Heavy vehicle tires refer to “trucks”, “articulated lorries” and “busses”. For both of these two types, the same plastic share (i.e. 85 %) was assumed.

For more information see Appendix 2 section a).

5.3.2 Stock and use-phase

For the calculation of all tires in stock, all registered vehicles in Austria in the year 2018, assuming a secondary set (i.e. winter/summer tires) for “passenger cars” and “busses”, were taken into account. Regarding the stock of tires, the same calculations as for the imported tires were made. Again, for each type of vehicle the specific number of tires was taken into account. The resulting number was multiplied with the respective weight of the tires (i.e. light or heavy tires) to calculate the whole mass of the plastic share of tires in stock.

Also, for the use-phase, the same calculations were made, expect that for this process only one set of tires was taken into account. Passenger cars, for example, were ascribed four tires because only one set (either summer or winter) can be in use at once.

For more information see Appendix 2 section a).

5.3.3 Wear and tear release

During the use-phase there is a non-exhaust wear and tear release from vehicle tires induced by driving. This wear and tear release consists of differently sized particles (10 nm – several 100 µm) which are emitted into environmental compartments (Kole et al., 2017). However, according to Düring et al. (2004) the majority of the released particles is larger than 2.5 µm.

Table 2 shows the various amounts of wear and tear release from the different types of vehicle tires that are used for the MFA. The values for “passenger cars” as well as for “campers” have been taken from an experimental study performed by the ADAC (2019), where the test vehicle was a VW Golf. During this test, the vehicle was driving over 15 000 km under realistic conditions. Every 2 500 km, the profile depth and weight loss were measured using a laser measuring device and digital scales.

All other figures for wear and tear have been found in Hillenbrand et al. (2005). The values from this study are based on a review of already existing literature.

Table 2: Calculated annual wear and tear release of vehicles in Austria 2018 based on the number of vehicles in use and their annual mileage.

	Number of registered vehicles in 2018	**Mileage in 2018 [x10^6 km]**	**Wear and tear [mg/km]**	**Total wear and tear 2018 [t/year]**	**Reference**
Passenger car	5 011 330	73 000	125	9 130	BMVIT, 2009b; Statistik Austria, 2018a; ADAC, 2019
Truck and articulated lorry incl. transit traffic	495 231	5 970	700	4 180	Hillenbrand et al., 2005; BMVIT, 2009a; Statistik Austria, 2018a
Motorbike (2 and 3 tires)	814 877	1 930	45	90	Hillenbrand et al., 2005; KBA, 2018; Statistik Austria, 2018a
Bus	10 037	470	700	330	Hillenbrand et al., 2005; Statistik Austria, 2018a; Salmhofer, 2019
Camper	28 022	400	125	50	BMVIT, 2009b; Statistik Austria, 2018a; ADAC, 2019
Other$^{2)}$	1 345 133	9 800	180	1 770	Hillenbrand et al., 2005; BMVIT, 2009b; Statistik Austria, 2018a
Total	**7 704 630**	**91 570**		**15 550**	

For the calculation of the annual wear and tear release in Austria in 2018 the annual mileage of each vehicle type category was needed. Table 2 illustrates these figures. Due to the lack of data, transit traffic was included only for the category “trucks” and “articulated lorries”. Moreover, for the categories “campers” and “others” there was no data on the annual mileage available at all. For this reason, these numbers were calculated based on the figures for passenger cars. Based on the different scientific as well as statistical sources of input data, the total annual wear and tear release calculated for Austria in 2018 was 15 550 t (see Table 2).

In the case of wear and tear release, the value of total suspended particles (TSP) is important to mention. TSP refers to the mass of the total dust. The frequently used PM_{10} and $PM_{2.5}$ values are a subset of TSP. As it is shown in Table 3 the total calculated TSP for the year 2018 in Austria induced by wear and tear release was 8 260 t (Luekewille et al., 2002). This amount was calculated in the same way as the 15 550 t/year from Table 2. Thus, a simple comparison of the numbers was made which shows that 53 % of the released abrasion particles has been emitted as airborne emissions. It was assumed that in the long run these airborne particles are deposited on the soil or in surface waters.

Table 3: Calculated annual TSP and wear and tear release from vehicle tires in Austria 2018 based on different scientific sources.

Type of calculation and emission	**Area under investigation**	**Vehicle type**	**Original figure**	**Unit**	**Annual wear and tear Austria [t/year]**	**Reference**
Based on a mail-survey (experts, laymen) as well as discussion. No information on type of emission.	Germany		1 095	g/(cap*year)	9 700	Bertling et al., 2018
Calculation of wear and tear based on weight loss tires and mileage. Total emissions.	Germany	Passenger cars	90	mg/km	13 000	Hillenbrand et al., 2005
		Trucks	700	mg/km		
		Motorbikes	45	mg/km		
		Busses	700	mg/km		
		Others	180	mg/km		
Calculation of wear and tear based on measured airborne emissions. TSP.	Europe	Light vehicles	0.0661	g/km	8 300	Luekewille et al., 2002
		Motorbikes	0.0282	g/km		
		Heavy vehicles	0.4208	g/km		

Furthermore, Table 3 shows two other calculated values for the non-exhaust wear and tear release in Austria 2018. The first one applies to a study of the Fraunhofer Institute for Environmental, Safety and Energy Technology (UMSICHT) performed by Bertling et al. (2018). In this study wear and tear release values for “passenger cars”, “motorbikes”, “trucks”, “skateboards” and “bikes” were included. The individual release values for the vehicles were based on an e-mail survey in which about 250 scientists of the Fraunhofer Institute UMSICHT and 142 experts and laymen were interviewed. This survey was supplemented by discussions at meetings. Overall, they concluded that in Germany per capita about 1 230 g tire abrasion are released annually. For the calculation of the values in Table 3 it was assumed that in Austria the annual wear and tear release per capita is the same. Multiplied with the inhabitants in Austria 2018 this resulted in a total wear and tear release of 10 800 t/year. To make it more comparable to the figure from Table 2 the values of “skateboards” and “bikes” were subtracted. This results in an annual wear and tear release of 9 700 t. This figure is lower than the value from Table 2 (i.e. 15 550 t/year). This may be due to the fact that Bertling et al. (2018) took their values from expert estimations whereas the figures of ADAC (2019) and also the values obtained from the review of Hillenbrand et al. (2005) were based on tire tread loss measurements. Moreover, in the calculations of Table 2 the vehicle types “busses”, “campers” and “others” were included which were not taken into account in the study of the Fraunhofer Institute. Additionally, in Table 2 for “trucks” and “articulated lorries” the transit traffic in Austria was taken into account.

Considering the second calculation from Table 3 based on the study of Hillenbrand et al. (2005) there are strong similarities to Table 2. The only difference is the value for “passenger cars” which is 125 mg/km in one and 90 mg/km in the other calculation. This difference may be due to different experimental conditions. For the MFA of this paper, the result of Table 2 with the higher amount of wear and tear release was used because the value of the ADAC study is more recent.

For more information see Appendix 2 section c).

5.3.4 EOL treatment

The figures for the EOL treatment steps of recycling, retreading, incineration, export of EOL tires, other treatment as well as landfilling were taken from the current “Federal Waste Management Plan of Austria” (BMNT, 2019).

5.4 Transfer coefficients and uncertainties

Overall, there were two different types of TCs used as is shown in Table 4. There are *nano-specific TCs* which explicitly refer to ENMs and *stochastic TCs* which were calculated with respect to product specific data.

On the level of “vehicle tires”, the TCs used were mainly calculated based on tire specific data. Only the TCs used for the wear and tear emissions into “surface water”, “soil” and “WWTP” are nano-specific TCs of Gottschalk et al. (2015). These selected TCs specifically refer to CB from tires. For the purpose of simplicity, it was assumed that these nano-specific TCs are also valid for the plastic part of vehicle tires. Due to a lack of data, for the diffuse emissions into air (TSP), other sources, which do not explicitly refer to the level of ENMs, were taken into consideration (see Table 4).

For the process “EOL treatment” and its corresponding sub-processes “tires for recycling”, “tires for retreading” and “export EOL tires”, no nano-specific TCs have been found. Consequently, stochastic TCs based on the present “Status Report of the Austrian Federal Waste Management Plan” were used (BMNT, 2019). For the process of WWT, nano-specific TCs were available for the treatment process itself. These TCs were taken from a study of Wang et al. (2012) where the fate of C_{60} fullerenes was simulated. For the further distribution of CB, figures from the current “Status Report of the Austrian Waste Management Plan” (BMNT, 2019) were taken into account.

Regarding “retreading”, there was very little data available. Hence, it was assumed that during the process of retreading 20 % of the top-layer of tires is mechanically peeled off. During this process some emissions occur, which are all retained by a filter. The TC for the filter residue was taken from a study of Wohlleben et al. (2016). However, this TC does not specifically refer to retreading processes, but to mechanical exposure of tires in general. In addition, this TC from Wohlleben et al. (2016) was also applied for the filter residue during the recycling of tires. All other TCs for the process of “recycling” were based, again, on figures of the current “Status Report of the Federal Waste Management Plan of Austria” (BMNT, 2019).

Table 4: Stochastic and nano-specific transfer coefficients as well as uncertainty values for the different flows and processes.

Name of flow or process	**"stochastic" transfer coefficients* [%]**	**Nano-specific transfer coefficients [%]**	**Uncertainty based on Laner et al., 2016 [%]**	**Reference**
Import tires			14.0%	
All vehicle tires (summer/winter)			14.0%	
Tires in use			14.0%	
EOL tires AT			5.0%	
Use phase			14.0%	
Wear and tear release			14.0%	
Re-use vehicle tires for the next season			14.0%	
Wear and tear release				
Diffuse emissions surface water		25.0%	14.0%	Gottschalk et al., 2015
Diffuse emissions soil		12.5%	14.0%	Gottschalk et al., 2015
Diffuse emissions air (TSP)	53.2%		14.0%	ADAC, 2019; Hillenbrand et al., 2005; Luekewille et al., 2002
Diffuse emissions WWTP		9.4%	14.0%	Gottschalk et al., 2015
Import EOL tires			5.0%	
EOL treatment				
EOL tires for recycling	47.7%		5.0%	BMNT, 2019
EOL tires for retreading	2.5%		5.0%	BMNT, 2019
Export EOL tires	49.8%		5.0%	BMNT, 2019
WWTP				
Effluent surface water		5.0%	5.0%	Wang et al., 2012
Sewage sludge for agriculture (soil)		19.0%	5.0%	BMNT, 2019
Further treatment	23.7%		5.0%	BMNT, 2019
Sewage sludge for incineration	52.3%		5.0%	BMNT, 2019
Retreading			-	
Retreaded tires	79.3%		41.0%	Wohlleben et al., 2016; Own assumption
Filter residue	0.7%		14.0%	Wohlleben et al., 2016
Shredding residue	20.0%		41.0%	Wohlleben et al., 2016; Own assumption
Recycling				
Shredding residue	53.6%		5.0%	BMNT, 2019
Filter residue	0.7%		14.0%	Wohlleben et al., 2016
Export granulate for new products	45.7%		5.0%	BMNT, 2019
WIP				
Slags and ashes		1.6%	5.0%	Mueller et al., 2013
Export fly ash to underground landfill		0.4%	5.0%	Mueller et al., 2013
Elimination		98.0%	5.0%	Mueller et al., 2013

For the process of "incineration" TCs from a study of Mueller et al. (2013) were applied. During this investigation the authors modelled the flows of different ENMs during waste handling and specifically during incineration. However, values were available only for CNTs. Due to the fact that CB as well as CNTs consist of carbonaceous compounds, they have a similar behaviour during incineration processes (Mueller et al., 2013; Part et al., 2018a). Consequently, the TCs for CNTs were used within this work for CB.

Data uncertainties are also shown in Table 4. All flows and processes were assessed with the proposed approach for expert estimations. The attributed uncertainties range between 5 and 41 %.

5.5 Results of the MFA – level of "vehicle tires"

As shown in Figure 12, 29 750 t of vehicle tires were imported by Austria in the year 2018. These tires were either stored in a stock or used depending on the season (i.e. winter and summer tires). The stock also contains tires from the years prior to 2018. Overall, 264 610 t of tires from the stock were used within the time period under consideration. The figures for "import", "total tires stock" as well as "in use" have a 14 % uncertainty.

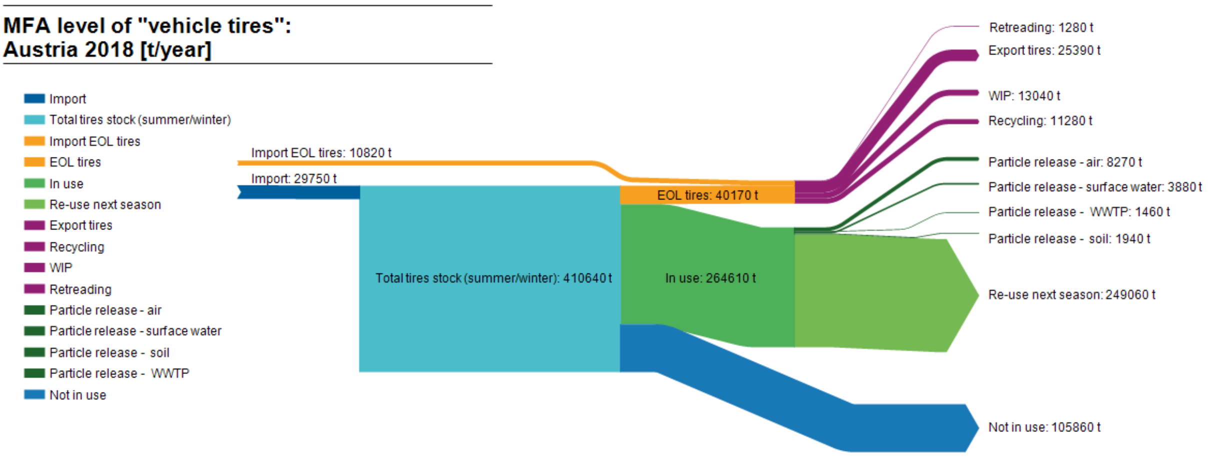

Figure 11: Results of the MFA on the level of "vehicle tires" where the system boundaries are spatially set to the area of Austria and temporally to the year 2018 [t/year]; created with e!Sankey.

During the use-phase, there is a wear and tear release of differently sized particles (10 nm – several 100 µm) into environmental compartments (Kole et al., 2017). This abrasion is always a direct result of friction (i.e. on the ground). It is physically not possible to transfer power without friction. Consequently, there is no tire in existence without any abrasion particle release at all (Eppel, 2019). The mass of the released particles during driving depends on different parameters. An important factor within this framework are the properties of the tire material themselves. Hard materials have a generally higher abrasion resistance (Hillenbrand et al., 2005).

Furthermore, the released amount of wear and tear depends on the appropriate use of tires according to the season. On warm days, winter tires release more abrasion particles than summer tires and vice versa. Another important parameter is the retrieved power of the vehicle (i.e. to the way of driving). If there is a constant acceleration with subsequent abrupt braking (e.g. on winding roads), there is more wear and tear release than whilst driving with constant and moderate speed on straight streets (e.g. on motorways) (Eppel, 2019). In a study by Foitzik et al. (2018) on this subject, the authors stated a reduced release of ultra-fine PM (i.e. diameter < 100 nm) from vehicle tires during moderate driving compared to higher speeds, higher longitudinal forces and higher slip angels. Moreover, the abrasion can depend on the type of vehicle and more specifically on its weight. To exemplify: electric vehicles are heavier than fuel-powered ones. With more weight there is more need for power to accelerate and decelerate a vehicle which results in more wear and tear release. Since electric vehicles are more torque intensive, this can also lead to more abrasion if the power is fully exploited by the driver (Kole et al., 2017; Eppel, 2019). However, there are currently no specific wear and tear release studies on electric vehicles available.

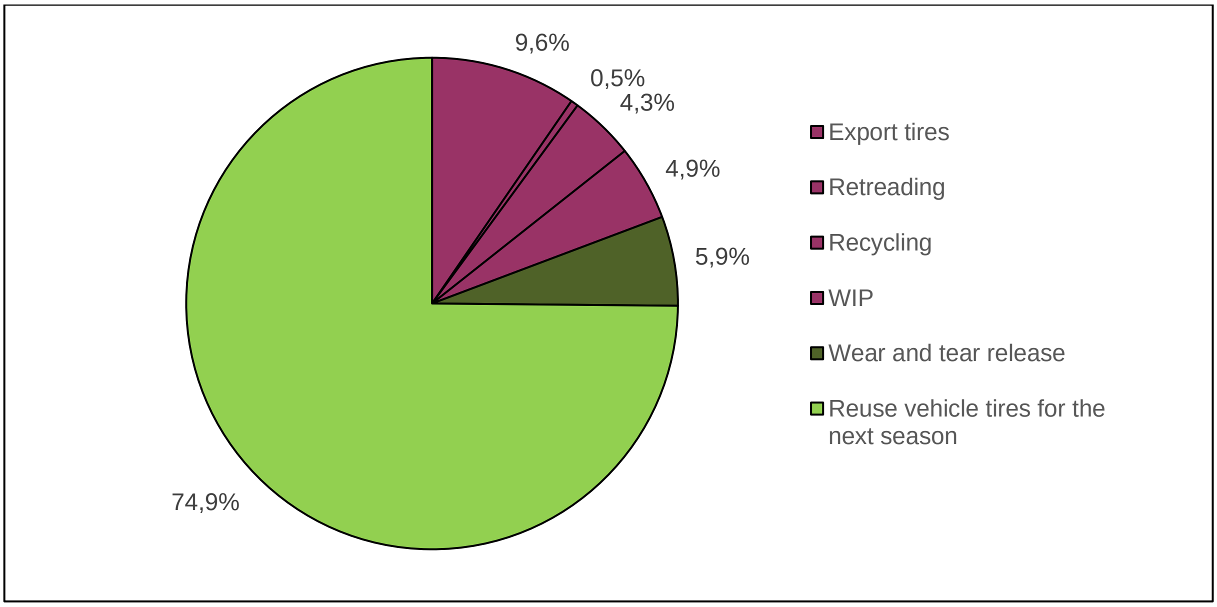

Figure 12: EOL tires, Wear and tear release from vehicle tires and re-use vehicle tires for the next season in relation to all tires in use.

Figure 11 presents a total wear and tear release of 15 550 t in Austria for 2018. This amount corresponds to about 0.5 Mio. average truck tires (only plastic share) or to 2.1 Mio. average passenger car tires (only plastic share). Because CB tends to form aggregates and agglomerates very fast (Donnet et al., 1993; IPPC, 2007), it was assumed that the nano-specific TC taken from Gottschalk et al. (2015) can also be applied to wear and tear release on the level of tires. Hence, on the level of goods these TC were applied to the released plastic particles with incorporated CB. Taking these TCs into account, there was a particle release of 3 880 t into surface waters, 1 940 t into the soil and 8 270 t into the air (i.e. TSP) in the year 2018. The remaining quantity of 1 460 t/year, which was not emitted into environmental compartments, was transported via the sewage system into WWTPs. All of these wear and tear release values show an uncertainty of 14 %. Since this PMFA only considers one year, “surface waters”, “soils” as well as the “air” constitute sinks for the emitted particles.

With regard to EOL treatment, there was an import of 29 750 t of EOL vehicle tires into Austria in 2018. Of those, together with the amount of EOL tires generated within the country itself, about 50 % were then exported, 22 % were recycled, 26 % were incinerated and 2 % were retreaded. Retreaded tires may be put back on the domestic market and may return to the stock/use phase in the following years. All of these EOL-processes and their corresponding output flows have an assigned uncertainty of 14 %.

The flow “not in use” from Figure 11 applies to all vehicle tires from the stock that are not used due to seasonal conditions but still remain in the stock and can be used in the following year(s).

Putting all these calculated figures into relation with “tires in use”, as it is depicted in Figure 12, about 6 % were emitted as diffuse emissions into environmental compartments, about 18 % entered an EOL-stage and 76 % are re-used again in the following year(s).

5.6 Results of the MFA – level of “carbon black”

First of all, it has to be noted that on the substance level not exclusively free CB NPs are addressed with the MFA. The major part of CB is still incorporated into the polymeric matrix of vehicle tires. Furthermore, due to aggregation and agglomeration processes as well as quality requirements the incorporated CB-particles are nanosized only to a small extent (Donnet et al., 1993; IPPC, 2007).

As it is shown in Figure 13, on the substance level, Austria imported 7 700 t of CB incorporated in vehicle tires in 2018. Again, these numbers have a 14 % uncertainty. This imported amount of CB was either stored in the stock or used for driving, depending on the season (i.e. winter or summer tires). CB from the years before 2018 was also included in the stock. The stock serves as a sink for CB within this PMFA.

In 2018 there were 68 490 t of CB in use as components of different vehicle tires. 4 020 t of this amount was emitted as wear and tear release into different environmental compartments. 2 140 t (53 %) of the abrasion particles were emitted into the air, 1 000 t (25 %) were released into surface waters, 500 t (13 %) into soils and 380 t (9 %) into WWTPs through the sewage system. Since in this PFMA only one year is considered, the environmental compartments air, soil and surface water serve as sinks for CB particles. All these wear and tear release values show a 14 % uncertainty.

In WWTPs CB particles undergo different treatment processes. After these processes, 95 % of the CB particles remain in the sewage sludge and 5 % in the effluent. Consequently, 20 t of CB were released into surface waters via the return of the treated effluent water. The sewage sludge to a large extent was burned in WIPs. However, 70 t of CB (20 %) of the sewage sludge were used as fertilizer in agriculture. Soils serve as sinks for CB particles. 25 % of the sludge was treated in other EOL treatment facilities (e.g. MBWTPs, biogas plants). Within the framework of this PMFA the process “other treatment” serves as a sink for CB particles. All output flows referring to WWTPs have an assigned uncertainty of 5 %.

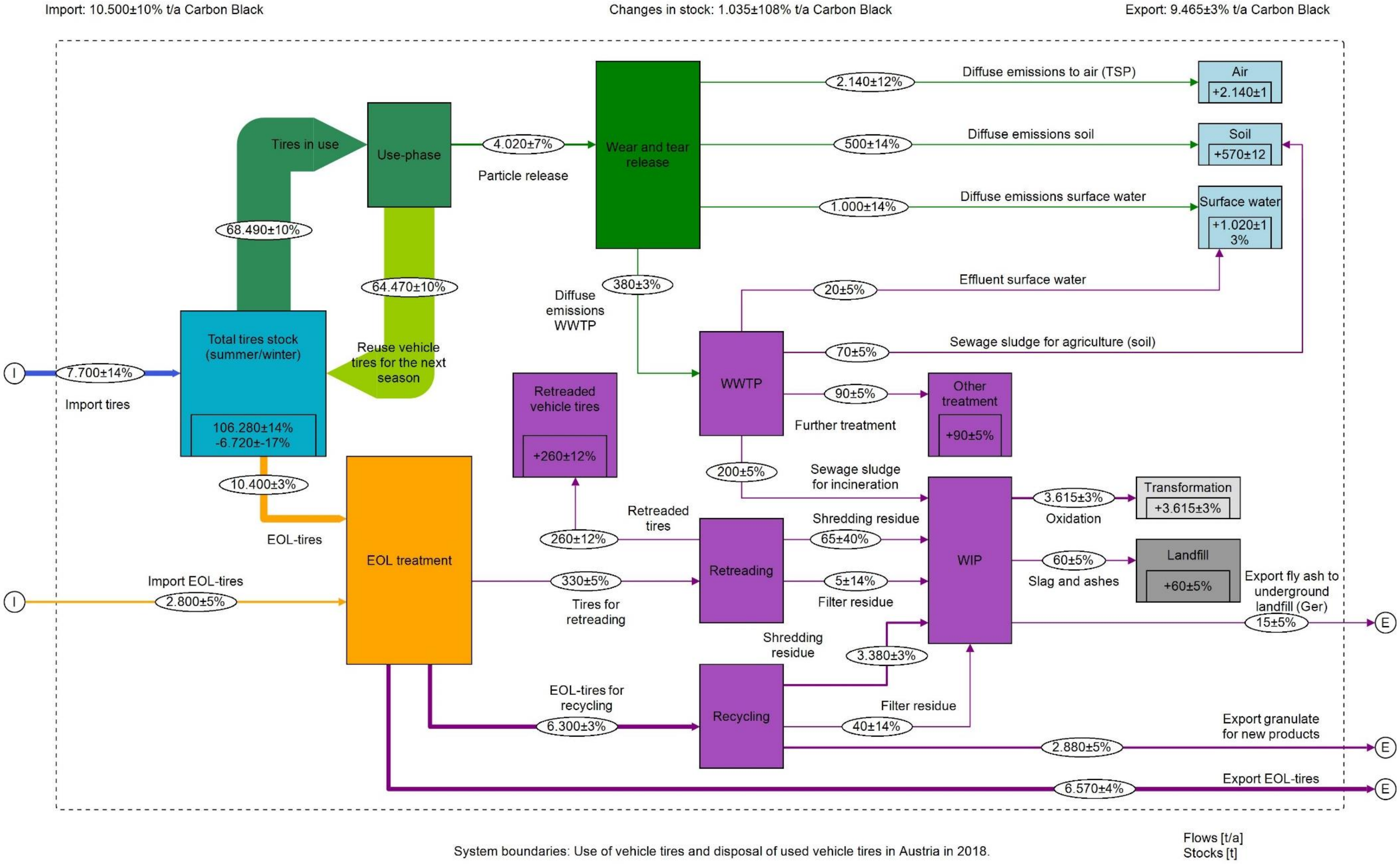

Figure 13: Results of the MFA on the level of “carbon black” where the system boundaries are spatially set to the area of Austria and temporally to the year 2018 [t/year]; created with STAN.

Considering all other EOL stages, there were about 10 400 t of CB directly entering the Austrian waste regime and 2 800 t of CB that were imported from other countries. This results in an overall amount of 13 200 t of CB incorporated in EOL tires. As shown in Figure 13, 6 570 t of this total CB were exported, 6 300 t were recycled and 330 t were retreaded. Due to a lack of data, especially on retreading processes, the output flows are inherently uncertain. The same applies to output flows of recycling. During recycling, granulate including 2 880 t of CB was generated which was exported to different countries to use in new products for the automotive industry, the leisure and sports sector, road construction etc. Shredding residues as well as the filter residues from recycling and retreading processes are burned in WIPs because these residues do not fulfil the requirement of TOC < 5 % DM according to the Austrian Landfill Regulation (DVO, 2008).

Overall, in the year 2018 about 3 690 t of CB were burned. During incineration processes nearly all CB is eliminated. As CB is a carbon-based material it is supposed to completely oxidize under the prevailing conditions in a WIP. However, a few CB particles may still survive in enclosed compartments (Mueller et al., 2013). Thus, a small share of CB remains in the bottom ash and in the fly ash. The fly ash is exported to an underground landfill in Germany whereas the incineration slag and ashes are landfilled in Austria. Landfills serve as sinks for CB particles. Because of existing studies on the behaviour of carbonaceous compounds in WIPs the uncertainties for this process with its related output flows are 5 %.

5.7 Free nanofiller release from tire tread

As already mentioned before, CB particles tend to form aggregates and/or agglomerates (Donnet et al., 1993; IPPC, 2007). Hence, there is only a small share of nano-sized particles within the polymeric host matrix of tires. Furthermore, released abrasion particles to a large extent consist of fractions of polymeric materials (nm-mm) with incorporated fillers. Thus, there is only an infinitesimal share of free nano-CB particles released during the use-phase of tires.

In the case of free nanofiller release, there were only very few studies available and specifically for tires containing CB only one relevant article of Wohlleben et al. (2016) was found. In this study the authors explored the pathway of secondary environmental fragmentation by applying different intensities of UV and hydrolysis after mechanical fragmentation (i.e. sanding) of car tires. For this purpose, two different scenarios were established: Scenario 1 refers to “rainy days” and Scenario 2 refers to “days without rain”. The sampled tires were made from natural rubber with 40 wt% of CB (N550 type Carbon black).

On rainy days (i.e. Scenario 1) the released abrasion particle mass deposited on streets was directly washed off, either to surface waters, soils or into the sewage system. However, due to processes of hydrolysis, already released fragments have been further reduced in size. For this process Wohlleben et al. (2016) determined 1.6 % of CB particles < 5µm of the total released wear and tear mass. On days without rain (i.e. Scenario 2) there was an enhanced UV irradiation on the already released particles deposited on the streets coupled with a delayed wash-off into surface waters, soils or sewage system. Because of this enhanced UV irradiation, already released wear and tear particles were additionally reduced in their size. For this scenario the authors observed 4.0 % of CB particles < 5 µm of the total released wear and tear mass. These emitted particles smaller than 5 µm correspond to PM_{10} emissions. For both scenarios a TEM analysis was performed. Based on the findings of the TEM analyses Wohlleben et al. (2016) stated an overall maximum free nano-CB release of 0.045 % of the aged wear and tear mass.

In a further step these findings of Wohlleben et al. (2016) have been combined with the figures calculated within this thesis. Although the results of Wohlleben et al. (2016) only apply to car tires, due to lack of data, it was assumed that for all other vehicle tires the same rates of secondary fragmentation will occur.

In order to correctly allocate the secondary environmental fragmentation caused by hydrolysis or UV irradiation, the average days with rain and without rain respectively were calculated for Austria. Overall, there were 152 rainy days and 213 days without rain in 2018 (Tulun, s.a.). This resulted in a release of about 100 t of CB on rainy days as well as 360 t of CB on days without rain < 5 µm of the total emitted abrasion particle mass. Hence, in the year 2018 in Austria 460 t of CB < 5 µm were released into environmental compartments due to secondary fragmentation processes. Regarding free nanofillers, a release of 0.2 t was calculated for the year 2018 within the area of Austria.

Emitted free nano-CB particles pose a risk to human health and the environment. In particular, inflammatory as well as carcinogenic effects can occur. Especially CB particles of smaller sizes are more potent at inducing these negative effects (IARC and WHO, 2010; EC, 2012; ETRMA, 2012). However, CB particles incorporated in a rubber matrix are not expected to be released and are therefore unlikely to pose a risk to humans (ETRMA, 2012).

6. Discussion

With regard to ENMs, it is crucial to take into account that not all particles termed “nanomaterials” actually meet the definition of NMs used within this thesis which states that “at least one dimension must be between 1-100 nm” (ISO/TS 80004-1:2015). However, this problem is not restricted to this particular thesis, but rather should be regarded as a general problem. A widely accepted and commonly used definition of NMs is desirable if not necessary.

6.1 ENMs in plastic products

Considering the evaluation of distinct ENM-types used in plastic products, it is difficult to differentiate between products that are already available to consumers and products that are currently only tested in laboratories. However, based on some expert discussions, it was assumed that the majority of the ENM-containing plastic products mentioned in the screened scientific articles have so far only been tested on a laboratory scale and thus correspond to a TRL of 1-4. Nevertheless, some of the products also match higher TRLs. CB or nano-TiO_2 for example are already used in tires or surface coatings respectively, which attests them with a TRL of 9.

Most analyzed scientific articles only addressed the enhancing properties of PNCs, but not environmental issues. Especially in the field of plastics, a material saving effect can be achieved through the incorporation or association of ENMs because lighter materials with less polymer content but with the same functionalities can be produced (Paul and Robeson, 2008; Azeredo et al., 2011). Moreover, plastic products containing ENMs can be manufactured from monolayer polymers instead of the usual multilayer polymers. This could have positive effects on the recycling sector because the use of components and additives can be reduced (Duncan, 2011). However, it has to be kept in mind that ENMs are also additives themselves and their fate and behaviour during different recycling processes has not yet been sufficiently investigated. Another environmental issue that may be addressed is the substitution of hazardous materials. A prominent example of this is nanoclay, which can be used as a flame retardant and, thus, as a substitution for halogens (e.g. Duncan, 2011; Duncan and Pillai, 2015; Hincapié et al., 2015). Nevertheless, there are also concerns about the safety aspects of ENMs because of their potential nanotoxicity (e.g. Foss Hansen et al., 2008; Laux et al., 2018; EUON, 2019). This nanotoxicity is particularly relevant in connection with the release of ENMs into the environment. So far, only a few studies have tried to answer the question, whether free ENMs are released or not.

There are several challenges that need to be considered and addressed. To begin with, it is not possible to detect free ENMs solely through analyzing the size distribution of aerosolized particles. These aerosolized particles do not only contain ENMs but also other nanoscaled particles (e.g. polymer particles released by abrasion). Moreover, some instruments are not sensitive enough to detect free ENMs (Schlagenhauf et al., 2012; Duncan, 2015). Additionally, the released individual ENM particles could accumulate on the surface of larger particles and in further consequence may not be detected by conventional particle sizing methods. Furthermore, it has to be kept in mind that ENMs may undergo transformation processes by photochemistry, adsorption of natural organic matter, aggregation, oxidation, biotransformation etc. during and/or after their release. Some of them will then no longer be nano-sized (Nowack et al., 2013). In order to address these challenges sophisticated surface analytical techniques, such as x-ray photoelectron spectroscopy (XPS) and secondary ion mass spectrometry (SIMS), are needed (Duncan, 2015).

Up to now, the majority of ENM release experiments carried out were case specific. Regarding the “experimental setup” and “aim of the analysis”, these studies mainly focussed on aerosol and airborne particle analysis to generate potential inhalation exposure. Regarding the “material under investigation”, so far, the focus of the performed particle release studies was primarily placed on carbon nanoforms, nano-Ag and nano-TiO_2 under different use scenarios (Duncan, 2015; Part et al., 2018a). With regard to release studies for waste treatment facilities, up to now, only a few studies have been performed at large scale and under real-world conditions (Part et al., 2018a). Considering the “analytical methods” also different techniques have been applied in the studies carried out. This means that the experimental set up, environmental conditions, used test materials as well as the applied analytical methods, differ between the studies. Hence, the comparison of the findings from these analyses is complicated if not impossible. These facts might explain the wide range between the release rates of some of the results. To overcome this problem the introduction of consistent standards is necessary. In order to do so, the most appropriate analytical techniques need to be assessed and, after their approval, set as standards (Silvestre et al., 2011; Clark et al., 2012; Nowack et al., 2013; Froggett et al., 2014; Duncan, 2015; Koivisto et al., 2017). Furthermore, it is crucial to understand how the experiments performed in laboratories are related to real-world conditions (Froggett et al., 2014).

In the case of used test materials, some upstream issues may occur. Used test materials are often poorly characterized or sometimes even uncharacterized. In many cases this can be attributed to the fact that manufacturers of PNCs are not willing or unable to provide information on their materials used. Without profound information on the used ENMs and their structure-function relation to the polymeric host materials, it is difficult to provide reliable results on the release of ENMs from PNCs (Duncan, 2015).

With regard to different EOL stages some studies are available for the release during incineration as well as for landfills. Regarding incineration, the results obtained so far cannot directly be compared to each other, because the waste fractions are relatively inhomogeneous and the chemical conditions in a WIP are highly complex. Moreover, it is difficult to distinguish between pristine ENMs, ENMs generated due to combustion processes and naturally occurring ENMs. Regarding landfilling, no in-situ studies have been performed so far. Additionally, with respect to leachate analysis, it is also difficult to differentiate between naturally occurring ENMs and ENMs introduced through plastic products (Part et al., 2018a). However, with EFMs the ultimate fate of ENMs in landfills, which is dependent on various internal as well as external factors, can be investigated.

Overall, significantly more research has to be done on the release mechanisms of ENMs from polymeric host materials in order to generate reliable data. This data should be comparable, reflect real-world conditions and take complex interacting parameters into account in order to create generally valid results (Clark et al., 2012; Duncan, 2015; Duncan and Pillai, 2015).

6.2 MFA on vehicle tires in Austria in 2018

Both MFAs, on the substance level as well as on the level of goods, contain uncertainties due to a lack of data concerning general information on vehicle tires and annual mileage of different vehicle types, figures on transit traffic, the amount of wear and tear release for different vehicle types, TCs as well as detailed information on certain processes. In consequence, assumptions had to be made and the uncertainties were taken into account by the modelling approach of Laner et al. (2016). On the substance level, these uncertainties were modelled with STAN and can be seen in Figure 13. However, on the level of goods, where the modelling tool e!Sankey was used, uncertainties were not considered within the software tool but within a separate calculation. The values from this calculation are listed in Table 4 and Appendix 4. Overall, the prevailing uncertainties of the input data mainly range between 5 % and 14 %, and only a few flows in the EOL-stages have higher ones. Nevertheless, the results of both MFAs serve as a first assessment of emission hot spots and therefore as a starting point for further investigations.

Considering the substance level, the main part of the incorporated CB in vehicle tires is not nanoscaled due to agglomeration and aggregation processes (Donnet et al., 1993; IPPC, 2007). However, some of these larger particles are still nanostructured (ETRMA, 2012). Furthermore, it has to be considered that on the substance level the CB particles are not always “free CB particles” but rather still incorporated in the polymer matrix of tires for the majority of the processes (e.g. recycling). The same is true for wear and tear particles, where the filler material CB is not isolated from polymeric compounds to a large extend too. Especially free NPs are rarely present in the different life cycle stages of vehicle tires (Wohlleben et al., 2016).

Since the two performed MFAs are static PFMAs and only one year was taken into account, “landfills”, “soils”, “surface waters” and “air” were considered to be sinks. Furthermore, the processes “other treatment” and “stock of vehicle tires” were described as sinks because the flows into this process were not observed further within the MFA. In reality, there are additional processes going on in landfills as well as in the different environmental compartments. For a further investigation of the behaviour and ultimate fate of released tire particles as well as nanofillers, EFMs should be performed. Particularly with regard to aggregation and agglomeration processes of CB, these models could give important insights into the behaviour of CB during different processes and provide information on final sinks.

7. Conclusion and outlook

In the past decades, a lot of research has been conducted in the fields of polymer nanotechnology. Hence, a wide variety of ENM-types used in plastic products for household use is to be found in the literature. According to scientific articles, mainly CNTs are embedded into plastics. These ENMs promise a lot of product enhancing characteristics, such as the improvement of tensile strength, electrical conductivity, or stiffness. Furthermore, nanoclay, nano-TiO_2, nano-Ag, nano-CB and nano-SiO_2 are frequently used ENMs in plastic products available to consumers. These ENMs are associated or incorporated into a variety of different products. Coatings and more precisely paints are the most prominent examples of such PNCs. Also, in the sectors of food and beverage packaging, electric and electronics or the automotive industry, many ENM-types are used in different plastic products for household use. Besides these six most frequently used ENM-types in plastic products, 30 more types were found within the screened scientific literature, such as other carbon-based ENMs (e.g. graphene, fullerenes), metallic ENMs (e.g. Cu, Au), nano-metal oxides (e.g. $Al(OH)_3$, Al_2O_3, ZnO) and organic ENMs (e.g. chitosan). However, many of the PNCs stated in the literature only apply to a TRL of 1-4. Besides that, there are still several ENM-containing products that already have a TRL of 9, such as vehicle tires or paints containing nano-CB or nano-TiO_2, respectively. In the future, it is expected that the electronic sector will play an important role especially with regard to nano-sensors.

ENMs do not only provide product-improving properties, they can also have a negative impact on humans and the environment. In particular, after a release of ENMs from the polymeric host material, nanoscale particles or fragments are likely to enter biological systems and cause toxic effects. Thus, it is important to know what mechanisms lead to the release of ENMs from the polymer matrix.

Various mechanisms that can result in a liberation of ENMs have been described in the literature. However, these released particles are agglomerates/aggregates or plastic debris/fragments, onto which ENM are associated rather than single ENM-particles. Up to now, several studies exist that show, for example, inflammatory or carcinogenic effects after exposure to different ENMs. Although these studies were mainly conducted on mice or rats, the results should still be taken into account for different regulations in order to guarantee the consideration of safety aspects. In the case of regulations for ENMs, there is an overall deficiency, only in the case of cosmetic, biocidal and food and beverage products, some legislations are available that restrict the use of certain ENMs or request their declaration and safety assessments.

Considering the different mechanisms that lead to a release of ENMs from a polymer matrix, there is one active and three passive release pathways: Matrix degradation as an active release mechanism is the most relevant for PNCs. Diffusion, desorption and dissolution as passive pathways play minor roles when it comes to PNCs. Especially during different end-of-life processes, mechanical exposure may lead to a particle and ENM release from plastic products. The release of ENMs during recycling and waste incineration processes needs to be further investigated in future research projects. However, in all other life cycle stages of plastic products, also a liberation of ENMs induced by different mechanisms may occur. Despite the distinct release mechanisms, the liberation of ENMs is also dependent on several other factors such as the dispersion of the ENMs within the matrix. If ENMs are homogeneously and well distributed in a polymeric matrix, these substances are less likely to be released.

Nevertheless, the released amount of ENMs increases with a longer product life time. Additionally, the host polymer itself as well as the type of ENM have an influence on the release behaviour. Also, high temperatures lead to an increased fragment and ENM release. All these above-mentioned factors can interact with each other. Until now, there are still knowledge gaps about the mechanisms of release and the behaviour of ENMs in the environment. For a more detailed insight, experimental studies would be needed, especially regarding safety assessments.

In order to predict the amount of released substances or ENMs, MFAs can be conducted. Therefore, one needs to know what different methodological approaches exist for MFAs on ENMs in order to choose an appropriate one. Overall, there are several different methodologies available: starting from single equation MFAs, where a series of algorithms is used, Excel-based MFAs, which are also based on equations, web-based MFAs to MFAs which take probability distributions (probabilstic MFAs) and additionally time-dynamic behaviour (dynamic probabilistic MFAs) into account. Inherently uncertain data because of data limitations, particularly on release rates and nano-specific transfer coefficients, can be addressed through different methodological uncertainty consideration approaches. There are statistical methods, sensitivity analyses, scenario modelling approaches, Monte Carlo simulations, the Bayesian modelling network as well as the modelling approach by Laner et al. (2016).

To provide a more detailed insight into the life cycle of one specific PNC and its incorporated ENMs, a case study of the most frequently used PNC at household level was carried out using an MFA. This is particularly important with regard to the emissions that can be generated throughout the entire life cycle. These emissions will probably be the highest for the most commonly used PNC as compared to less frequently used ones. The selection of the most frequently used PNC was based on different selection criteria and on a study by the EC (2012) which identified CB as the most commonly used additive in plastic products. In addition, a recent study found CB particles in the foetal side of human placenta, which shows a high relevance in terms of environmental, health and safety aspects. At the moment CB is especially used in vehicle tires, not only nano- but also microsized. Based on these facts, CB-containing vehicle tires have been chosen as a representative example for the case study.

Based on the methodological approach of probabilistic MFA and the uncertainty consideration proposed by Laner et al. (2016), the MFA within this thesis has been conducted on two different levels: On the “level of goods” the results indicate an overall wear and tear release caused by abrasion during the usage phase, consisting of agglomerates of polymers and CB-based fillers (nano- and microscale), of 15 550 t. These emissions in form of tire abrasion particles were emitted into different environmental compartments, in particular, 53 % were released into the air, 12 % were emitted as diffuse emissions into soils and 35 % into surface waters and waste water treatment plants. Regarding the “substance level” a total of 1 000 t of CB emits into surface waters, 500 t into soils, while 2 140 t of CB are airborne emissions. These released ENMs are not only free nanofillers but mainly still incorporated into fractions of polymeric components. For Austria, about 0.2 t of released CB nanofillers have been estimated.

All obtained results from this MFA can be used for further in-depth analysis using environmental fate modelling that generally aims to model the species and forms (e.g. dissolved or aggregated nanoparticles) of the released particles. This information is in turn very important for toxicological studies. Of particular interest are the identified sinks such as soils, surface waters, landfills and the air. Here, an investigation of the behaviour and ultimate fate are valuable, in order to provide, for example, a risk assessment and answers to the following questions: *Are there further processes or exposure pathways that may lead to the release of ENMs from polymeric components? What toxicological impacts might there be on humans and the environment?* In addition, a more comprehensive investigation of sinks would be important. *Are ENMs released from such sinks or will they stay there for decades?*

Since the results of the MFAs on both levels have some uncertainties resulting from the uncertainty of model input data and the lack of release data, the performed MFAs have to be updated and improved in the future. Especially if new and more accurate data is available, the MFAs can be adjusted and re-calculated with new input data. In conclusion, uncertainty and sensitivity analysis should be included in each MFA study and standardized in the future. In addition, results from measurements from environmental monitoring studies should be used as a benchmark to validate and consequently calibrate the emission or material flow model.

Overall, the results obtained from the applied MFAs can be used as an incentive for tire production companies and the corresponding research facilities in order to develop new vehicle tires with less wear and tear release (production of “green tires”). The use of ENMs as reinforcing additives and fillers in vehicle tires, which reduces the wear and tear, is a good example that nanotechnology can make vehicle tires “greener”. Nevertheless, more studies and reliable release data are necessary to minimize potential risks and ensure the safe use of ENMs in plastics (e.g. reinforcing fillers in the rubber of vehicle tires). Moreover, the results can be used for political decision makers in the field of traffic planning or for the development of appropriate end-of-life management strategies to minimize human and/or environmental exposure. In particular, there is a need for appropriate measures to reduce the ultrafine dust load. Finally, the large amount of 15 550 t of emitted tire wear and tear which was calculated for the year 2018 could be an incentive for everybody to use other means of transportation whenever possible.

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Appendices

Appendix 1: List of ENM-containing products with detailed information on each product category and product specification, specific plastic type and its source based name, EU Regulation No 10/2011, mass content, primary particle size, aggregate size, agglomerate size and enhanced characteristics.

Explanation colours: orange=not regulated by the Commission Regulation No 10/2011, green=future application.

Explanation of footnotes: 1) 10/2011; 2) Voll and Kleinschmit, 2010; 3) Hyperion Catalysis, s.a., 4) Green Dot Bioplastics, 2018, 5) Xiao and Mead, 2000, 6) Sto, s.a., 7) own calculation based on figures from the article.

Reference	Product category	Product specification (exemplary)	Company or product trade name (exemplary)	Plastic type	Source-based name (exemplary)	Embedded ENM	(EU) No 10/2011	Mass (wt% or concentration)	Primary size (diameter)	Aggregate size (diameter)	Agglomerate size (diameter)	Enhanced characteristics
Bott et al., 2014	Food and beverage packaging	-	n.i.	Thermoplastic	PE (LDPE), PS	CB	yes	max. 2.5 wt%[1] 2.5 and 5 wt% (experiment)	10-300 nm[1] 10-500 nm[2]	100-1200 nm[1]	300 nm - mm[1]	n.i.
Bott et al., 2014	Automotive industry	Tires	n.i.	Elastomer	n.i.	CB	n/a	n.i.	10-500 nm[2]	n.i.	n.i.	n.i.
Bott et al., 2014	Coatings	Coatings	n.i.	n.i.	n.i.	CB	n/a	n.i.	10-500 nm[2]	n.i.	n.i.	n.i.
Bott et al., 2014	Coatings	Inks	n.i.	n.i.	n.i.	CB	n/a	n.i.	10-500 nm[2]	n.i.	n.i.	n.i.
Silvestre et al., 2011	Food and beverage packaging	e.g. baked goods, meat, carbonated drinks, beer, etc.	n.i.	Thermoplastic	PA, PE, PET, PP, PS	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Improvement of gas barrier and mechanical properties, thermal stability and thus resistance to fire.
Silvestre et al., 2011	Food and beverage packaging	e.g. baked goods, meat, carbonated drinks, beer, etc.	n.i.	Thermoset, Elastomer	Epoxy resins, PU	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Improvement of gas barrier and mechanical properties, thermal stability and thus resistance to fire.

Reference	**Product category**	**Product specification** (exemplary)	**Company or product trade name** (exemplary)	**Plastic type**	**Source-based name** (exemplary)	**Embedded ENM**	**(EU) No 10/2011**	**Mass** (wt% or concentration)	**Primary size** (diameter)	**Aggregate size** (diameter)	**Agglomerate size** (diameter)	**Enhanced characteristics**
Silvestre et al., 2011	Food and beverage packaging	e.g. baked goods, meat, carbonated drinks, beer, etc.	n.i.	Elastomer	EVA	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Improvement of gas barrier and mechanical properties, thermal stability and thus resistance to fire.
Silvestre et al., 2011	Food and beverage packaging	e.g. baked goods, meat, carbonated drinks, beer, etc.	n.i.	n.i.	n.i.	TiO_2	yes	n.i.	n.i.	n.i.	n.i.	Photocatalytic disinfection-effect.
Silvestre et al., 2011	Food and beverage packaging	e.g. baked goods, meat, carbonated drinks, beer, etc.	n.i.	Thermoplastic	PA	Ag	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Silvestre et al., 2011	Food and beverage packaging	-	n.i.	Thermoplastic	PP	ZnO	yes	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Silvestre et al., 2011	Food and beverage packaging	-	n.i.	n.i.	n.i.	MgO	yes	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Silvestre et al., 2011	Food and beverage packaging	e.g. baked goods, meat, carbonated drinks, beer, etc.	n.i.	Thermoplastic	PA, PLA, PP	CNT	no	n.i.	n.i.	n.i.	n.i.	Improvement of Young's modulus, tensile strength, water vapour transmission rate, antibacterial properties.
Silvestre et al., 2011	Food and beverage packaging	e.g. baked goods, meat, carbonated drinks, beer, etc.	Fresher Longer™	Thermoplastic	PET, PP	SiO_2	yes	n.i.	1-100 nm[1]	0.1-1 µm[1]	0.3 µm - mm size[1]	Improvement of mechanical and/or gas barrier properties.

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Paul und Robeson 2008	Food and beverage packaging	-	Nanocor (Imperm^{TM})	Thermoplastic	PET, PP	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Increased Young's modulus, stiffness, improvement of thermal properties and thus flammability resistance, gas-barrier properties, weight advantage.
Paul and Robeson, 2008	Electronics	LED	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Electrical conductivity.
Paul and Robeson, 2008	Electronics	Electrical devices	n.i.	n.i.	n.i.	CNT (SWCNT, MWCNT)	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of electrical conductivity, charge transport.
Paul and Robeson, 2008	Electronics	Electrical devices	n.i.	n.i.	n.i.	Graphene	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of electrical conductivity, charge transport, barrier.
Paul and Robeson, 2008	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	CdSe	n/a	n.i.	n.i.	n.i.	n.i.	Improved charge transport.
Paul and Robeson, 2008	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	CdTe	n/a	n.i.	n.i.	n.i.	n.i.	Improved charge transport.
Paul and Robeson, 2008	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	Si	n/a	n.i.	n.i.	n.i.	n.i.	High efficiency, improved stability.
Paul and Robeson, 2008	Coatings	Paints, adhesives	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of viscosity.
Paul and Robeson, 2008	Automotive industry	Fuel cells: e.g. inclusion in proton exchange membranes, binder for electrodes	DuPont (Nafion®)	n.i.	Nafion	CNT (SWCNT) + Pt (in combination)	n/a	n.i.	2-5 nm	n.i.	n.i.	Improvement of mechanical properties, proton conductivity.

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Paul and Robeson, 2008	Automotive industry	Fuel cells: e.g. inclusion in proton exchange membrane	n.i.	n.i.	Polymers	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of methanol cross-over.
Paul and Robeson, 2008	Automotive industry	Fuel cells: e.g. inclusion in proton exchange membrane	DuPont (Nafion®), Ube	Thermoplastic	Nafion, PA	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of barrier properties.
Paul and Robeson, 2008	Automotive industry	Timing belt cover	Toyota, Ube	Thermoplastic	PA	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stiffness.
Paul and Robeson, 2008	Automotive industry	Assist steps	General Motors	Thermoplastic	Polyolefins	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stiffness and tensile strength.
Paul and Robeson, 2008	Automotive industry	Tires	InMat LLC.	Elastomer	PIB	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of barrier properties.
Paul and Robeson, 2008	Automotive industry	Tires	n.i.	Elastomer	Natural rubber, PIB, SBR	CB	n/a	n.i.	20-100 nm	n.i.	n.i.	Improved strength, wear and abrasion resistance.
Paul and Robeson, 2008	Sporting equipment	Hockey sticks	Montreal (Nitro Hybtonite®)	Thermoset	Epoxy	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stiffness and tensile strength.
Paul and Robeson, 2008	Sporting equipment	Tennis balls, footballs	Wilson - InMat LLC. (Double Core^{TM})	n.i.	n.i.	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Gas barrier which slows down the loss of air and upholds pressure, the ball bounces twice as long as other ones.
Paul and Robeson, 2008	Sporting equipment	Tennis racquets	Wilson	Thermoset	Epoxy	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stability.
Paul and Robeson, 2008	Sporting equipment	Tennis and badminton racquets	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.

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Paul and Robeson, 2008	Sporting equipment	Tennis and badminton racquets	n.i.	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.
Paul and Robeson, 2008	Sporting equipment	Tennis and badminton racquets	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.
Paul and Robeson, 2008	Medical equipment	Wound care, bandage	Curad®	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial properties.
Paul and Robeson, 2008	Medical equipment	Latex gloves	n.i.	Elastomer	Natural rubber	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial properties.
Paul and Robeson, 2008	Electronics	Computer disk drives[3]	Hyperion	n.i.	n.i.	CNT (MWCNT)	n/a	n.i.	n.i.	n.i.	n.i.	Improvement electrical conductivity.
Paiva et al., 2008	Food and beverage packaging	-	Süd-Chemie (Nanofil®) Laviosa Chemica Mineraria (Dellite®)	Thermoplastic	PA, PE, PP, PS	Nanoclay	yes	n.i.	1.67 nm (PA, PP, PS) 2.98 nm (PP)	n.i.	n.i.	Improvement of chemical, gas barrier and thermal properties, tensile strength and Young's modulus.
Adame and Beall, 2009	Food and beverage packaging	-	n.i.	n.i.	Polymers	Nanoclay	yes	max. 5 wt%	n.i.	n.i.	n.i.	Gas barrier properties.
Weiss et al., 2006	Food and beverage packaging	-	n.i.	n.i.	n.i.	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Improvement of gas barrier properties thermal stability, resistance to fire, stability of packaging.
Weiss et al., 2006	Textiles	Protective clothing	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improved mechanical and thermal properties, large surface-to-volume ratio.

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Weiss et al., 2006	Electronics	Electrical devices	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improved mechanical, electrical and thermal properties, large surface-to-volume ratio.
Sánchez et al., 2014	Food and beverage packaging	-	n.i.	Thermoplastic	PE, PET, PP	$CaCO_3$	no	4 wt%[7]	n.i.	n.i.	n.i.	n.i.
Sánchez et al., 2014	Food and beverage packaging	-	n.i.	Thermoplastic	PE, PET, PP	Nanoclay	yes	4 wt%[7]	n.i.	n.i.	n.i.	Improvement of thermal stability, resistance to fire.
Sánchez et al., 2014	Food and beverage packaging	-	n.i.	Thermoplastic	PE, PET, PP	Ag	no	4 wt%[7]	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Sánchez et al., 2014	Food and beverage packaging	-	n.i.	Thermoplastic	PE, PET, PP	ZnO	yes	4 wt%[7]	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Sánchez et al., 2014	Food and beverage packaging	-	n.i.	Thermoplastic	PET, PLA	TiO_2	yes	n.i.	n.i.	n.i.	n.i.	Antimicrobial properties, UV protection, improved strength.
Zhang et al., 2017	Coatings	WPC: Siding, roofing, windows, door frames, outdoor furniture, etc.[4]	n.i.	Thermoplastic	PE, PP	CB	n/a	3-12 wt%	35 nm	n.i.	n.i.	Improvement electrical property, tensile strength, Young's modulus, decreases of volume and surface resistivity values.
Zhang et al., 2017	Coatings	WPC: Siding, roofing, windows, door frames, outdoor furniture, etc. [4]	n.i.	Thermoplastic	PE	CNT (MWCNT)	n/a	3-12 wt%	10-20 nm (diameter) 10-30 µm (length)	n.i.	n.i.	Improvement electrical property, tensile strength, Young's modulus, decreases of volume and surface resistivity values.

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Zhang et al., 2017	Coatings	WPC: Siding, roofing, windows, door frames, outdoor furniture, etc. [4]	n.i.	Thermoplastic	PE	FG	n/a	3-12 wt%	32-100 nm	n.i.	n.i.	Improvement electrical property, tensile strength, Young's modulus, volume and surface resistivity value decreases
Othman et al., 2014	Food and beverage packaging	-	n.i.	Thermoplastic	PE (LDPE)	TiO_2	yes	4.03-8.86 wt%[7]	25 nm	n.i.	n.i.	Improved antimicrobial properties in combination with UV light.
Schulz, 2015	Antireflective polymer optics	e.g. optical lenses for cameras, headlights	n.i.	Thermoplastic	COP, PC	MgF_2	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of stray light and reflection.
Schulz, 2015	Antireflective polymer optics	e.g. optical lenses for cameras, headlights	n.i.	Thermoplastic	COP, PC	Ta_2O_5	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of stray light and reflection.
Schulz, 2015	Antireflective polymer optics	e.g. optical lenses for cameras, headlights	n.i.	Thermoplastic	COP, PC	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of stray light and reflection.
Wendling et al., 2009	Antireflective polymer optics	e.g. optical lenses for cameras, headlights	n.i.	Thermoplastic	COP, PC	Al_2O_3	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of stray light and reflection.
Wendling et al., 2009	Antireflective polymer optics	e.g. optical lenses for cameras, headlights	n.i.	Thermoplastic	COP, PC	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of stray light and reflection.
Wendling et al., 2009	Antireflective polymer optics	e.g. optical lenses for cameras, headlights	n.i.	Thermoplastic	COP, PC	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of stray light and reflection.
Wendling et al., 2009	Antireflective polymer optics	e.g. optical lenses for cameras, headlights	n.i.	Thermoplastic	COP, PC	Ta_2O_5	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of stray light and reflection.

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Wang et al., 2002	Automotive industry	Tires	n.i.	n.i.	n.i.	CB	n/a	n.i.	n.i.	n.i.	n.i.	Reinforcement of structure, abrasion and wear resistance, improvement of strength.
Wang et al., 2002	Automotive industry	Structures for catalyst support	n.i.	n.i.	n.i.	CB	n/a	n.i.	n.i.	n.i.	n.i.	Chemically inert support, mixture of CB and polymers create hydrophilic and hydrophobic pore structures.
Wang et al., 2002	Coatings	Coatings	n.i.	n.i.	n.i.	CB	n/a	n.i.	n.i.	n.i.	n.i.	Reinforcement of structure, ultraviolet absorption and electrical conductivity.
Wang et al., 2002	Coatings	Inks (conductive)	n.i.	Thermoset[5]	Epoxy resins[5]	CB	n/a	n.i.	n.i.	n.i.	n.i.	Good and easy dispersion, electrical conductivity.
Chen et al., 2005	Food and beverage packaging	-	n.i.	Thermoplastic	PVA	CNT	no	0.5-9.1 wt%	n.i.	n.i.	n.i.	Improvement of tensile strength, Young's modulus, toughness, water vapour transmission rate.
Vermeiren et al. 2002	Food and beverage packaging	e.g. vacuum packaged-products such as meat, cheese, fruit, etc.	n.i.	Thermoplastic	LDPE	Ag	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Vermeiren et al., 2002	Food and beverage packaging	e.g. vacuum packaged-products such as meat, cheese, fruit, etc.	n.i.	Thermoplastic	LDPE	CuO	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.

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Vermeiren et al., 2002	Food and beverage packaging	e.g. vacuum packaged-products such as meat, cheese, fruit, etc.	n.i.	Thermoplastic	LDPE	Zn_2SiO_4	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Vermeiren et al., 2002	Textiles	Textile and carpet fibres	DuPont (Micro-Free^{TM})	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Vermeiren et al., 2002	Textiles	Textile and carpet fibres	DuPont (Micro-Free^{TM})	n.i.	n.i.	CuO	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Vermeiren et al., 2002	Textiles	Textile and carpet fibres	DuPont (Micro-Free^{TM})	n.i.	n.i.	Zn_2SiO_4	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Vermeiren et al., 2002	Coatings	Paints	DuPont (Micro-Free^{TM})	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Vermeiren et al., 2002	Coatings	Paints	DuPont (Micro-Free^{TM})	n.i.	n.i.	CuO	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Vermeiren et al., 2002	Coatings	Paints	DuPont (Micro-Free^{TM})	n.i.	n.i.	Zn_2SiO_4	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Fujishima et al., 2000	Textiles	e.g. tent material, uniforms, etc.	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Photocatalytic decomposition of organic compounds and sterilization.
Fujishima et al., 2000	Coatings	e.g. plastic surfaces, bathroom and kitchen components, etc.	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Photocatalytic decomposition of organic compounds and sterilization.
BMLFUW, 2013	Automotive industry	e.g. car interior trim, bumpers, cables, tubes, etc.	n.i.	n.i.	n.i.	CNT	n/a	n.i.	3-4 nm	10-20 nm	200 µm	Improvement of tensile strength at lower density, antistatic effects due to electrical conductivity.
BMLFUW, 2013	Coatings	Paints	n.i.	n.i.	n.i.	CB	n/a	n.i.	n.i.	n.i.	n.i.	Pigment.

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BMLFUW, 2013	Coatings	Paints	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
BMLFUW, 2013	Coatings	Paints	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
BMLFUW, 2013	Coatings	Adhesives, sealants	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
BMLFUW, 2013	Coatings	Inks	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
BMLFUW, 2013	Coatings	Inks	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
BMLFUW, 2013	Coatings	Inks	n.i.	n.i.	n.i.	CB	n/a	n.i.	n.i.	n.i.	n.i.	Pigment.
Boysen et al., 2011a	Sporting equipment	Tennis balls	Wilson - InMat LLC. (Double Core^{TM})	Elastomer	IIR, PIB	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Gas barrier which slows down the loss of air, the ball bounces twice as long as other ones.
Boysen et al., 2011a	Sporting equipment	Tennis racquets	Wilson (nCode^{TM})	Thermoset	Epoxy	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stability.
Boysen et al., 2011a	Sporting equipment	Tennis and badminton racquets	Yonex (Nanolok^{TM})	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.
Boysen et al., 2011a	Sporting equipment	Tennis and badminton racquets	Yonex (Nanolok^{TM})	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.
Boysen et al., 2011a	Sporting equipment	Cycle handlebars and cranks	Easton Cycling	Thermoset	Epoxy	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stiffness, lightweight product.
Boysen et al., 2011a	Sporting equipment	Fishing rods	St. Croix Rod Company (NSi^{TM})	Thermoset	Epoxy	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Stronger and flexible rod, lightweight product.
Boysen et al., 2011a	Sporting equipment	Gel coat for kayaks	ReTurn AS	Thermoset	Epoxy	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of abrasion and crack resistance.

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Boysen et al., 2011a	Sporting equipment	Arrows for archery	Easton Archery (N-FUSED CARBON AXIS^{TM})	Thermoset	Epoxy	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Stronger arrow with better vibration control, lightweight product.
Nowack et al., 2013	Sporting equipment	n.i.	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Nowack et al., 2013.	Electronics	Small WEEE: Laptops, mobile phones, tablets (housing and cables)	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Nowack et al., 2013	Automotive industry	Tires	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Nowack et al., 2013	Automotive industry	Small components of cars	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Verma 2013	Sporting equipment	Tennis and badminton racquets	Yonex (Nanolok^{TM})	n.i.	n.i.	Silicates	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of flexibility, lightweight racquets.
Verma, 2013	Sporting equipment	Tennis balls	Wilson - InMat LLC. (Double Core^{TM})	Elastomer	IIR, PIB	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Gas barrier which slows down the loss of air, the ball bounces twice as long as other ones.
Verma, 2013	Sporting equipment	Tennis racquets	Wilson	Thermoset	Epoxy	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stability.
Verma, 2013	Sporting equipment	Tennis and badminton racquets	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.
Verma, 2013	Sporting equipment	Tennis and badminton racquets	n.i.	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.
Verma, 2013	Sporting equipment	Tennis and badminton racquets	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of power and flexibility, lightweight racquets.

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Verma, 2013	Sporting equipment	Cycle handlebars and cranks	Easton Cycling	Thermoset	Epoxy	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of stiffness, lightweight product.
Verma, 2013	Sporting equipment	Fishing rods	St. Croix Rod Company (NSi^{TM})	Thermoset	Epoxy	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Stronger, more flexible, lightweight product.
Verma, 2013	Sporting equipment	Arrows for archery	Easton Archery (N-FUSED CARBON AXIS^{TM})	Thermoset	Epoxy	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Stronger arrow with better vibration control, lightweight product.
Verma, 2013	Sporting equipment	Skis	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Flexibility control, slicker rides and turns.
Verma, 2013	Sporting equipment	Footballs	InMat LLC.	Elastomer	IIR, PIB	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Gas barrier which slows down the loss of air and upholds pressure.
Verma, 2013	Sporting equipment	Canoes, race-boat hulls/masts	n.i.	n.i.	n.i.	GO	n/a	n.i.	n.i.	n.i.	n.i.	Stronger and lighter, increase of glide.
Verma, 2013	Sporting equipment	Canoes, race-boat hulls/masts	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Stronger and lighter, increase of glide.
Verma, 2013	Sporting equipment	Breaks	n.i.	n.i.	n.i.	CNF	n/a	n.i.	n.i.	n.i.	n.i.	Improved thermal resistance, aerodynamic drag reduction.
Verma, 2013	Coatings	Race-car paints	n.i.	n.i.	n.i.	CNF	n/a	n.i.	n.i.	n.i.	n.i.	Improved thermal resistance, aerodynamic drag reduction.
Verma, 2013	Sporting equipment	Bowling balls	n.i.	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of chipping and cracking.
Verma, 2013	Sporting equipment	Golf balls	n.i.	Elastomer	IIR	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of resilience and bounce.
Verma, 2013	Automotive industry	Tires for racing	n.i.	Elastomer	n.i.	CB	n/a	n.i.	n.i.	n.i.	n.i.	Decreased rolling resistance, grip improvement.

Reference	Product category	Product specification (exemplary)	Company or product trade name (exemplary)	Plastic type	Source-based name (exemplary)	Embedded ENM	(EU) No 10/2011	Mass (wt% or concentration)	Primary size (diameter)	Aggregate size (diameter)	Agglomerate size (diameter)	Enhanced characteristics
Dimitrakakis et al., 2009	Electronics	Small WEEE	n.i.	Thermoplastic	ABS, PC, PP, PS, PVC	Sb	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of heat stability and thus properties of flame retardant.
Chin et al., 2010	Coatings	Paints	n.i.	n.i.	n.i.	Sb_xO_x	n/a	n.i.	2-500 nm	n.i.	> 100 nm	Improvement of heat stability and thus properties of flame retardant, white pigment - “antimony white”.
Chin et al., 2010	Coatings	Adhesives	n.i.	n.i.	n.i.	Sb_xO_x	n/a	n.i.	2-500 nm	n.i.	> 100 nm	Improvement of heat stability and thus properties of flame retardant.
Chin et al., 2010	Coatings	Sealants	n.i.	n.i.	n.i.	Sb_xO_x	n/a	n.i.	2-500 nm	n.i.	> 100 nm	Improvement of heat stability and thus properties of flame retardant.
Chin et al., 2010	Textiles	Back coatings	n.i.	n.i.	n.i.	Sb_xO_x	n/a	n.i.	2-500 nm	n.i.	> 100 nm	Improvement of heat stability and thus properties of flame retardant.
Chin et al., 2010	Electronics	LED	n.i.	n.i.	n.i.	Sb_xO_x	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Vuković et al., 2014	Coatings	Paints	n.i.	n.i.	n.i.	Sb_xO_x	n/a	n.i.	n.i.	n.i.	200 nm (diameter) 400 nm (length)	Improved thermal insulation.
Allsopp et al., 2007	Automotive industry	Fuel cells	Motorola	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Reduction of static electricity, hydrogen storage.
Allsopp et al., 2007	Automotive industry	Fuel cells	Sony	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Allsopp et al., 2007	Automotive industry	Fuel cells	n.i.	n.i.	n.i.	Pt	n/a	n.i.	n.i.	n.i.	n.i.	n.i.

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Allsopp et al., 2007	Automotive industry	Fuel cells	n.i.	n.i.	n.i.	Quantum dots	n/a	n.i.	2-100 nm	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	Small WEEE: Mobile phones	LG Electronics	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antibacterial characteristics.
Allsopp et al., 2007	Electronics	Small WEEE: Mobile phones	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Electromagnetic shielding.
Allsopp et al., 2007	Electronics	LED	Eikos Inc. (Invisicon^{TM})	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Electrical conductivity.
Allsopp et al., 2007	Electronics	LED	EuroAsia Semi-conductor	n.i.	n.i.	Quantum dots	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of colours, consumption of less energy.
Allsopp et al., 2007	Electronics	LED	n.i.	n.i.	n.i.	CdSe	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	LED	n.i.	n.i.	n.i.	ZnO	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	LED	Norel	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	LED	n.i.	n.i.	n.i.	GaN	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of conductive properties.
Allsopp et al., 2007	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	CdSe	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	Quantum dots	n/a	n.i.	2-100 nm	n.i.	n.i.	n.i.
Allsopp et al., 2007	Electronics	Large WEEE: Refrigerator	Daewoo	Thermoset	Resin	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics, restrained growth of bacteria, suppression of bad odours.

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Allsopp et al., 2007	Electronics	Large WEEE: Vacuum cleaner	Daewoo	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics, removement of bacteria, suppression of bad odours.
Allsopp et al., 2007	Electronics	Large WEEE: Washing machine	Daewoo	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antibacterial characteristics.
Allsopp et al., 2007	Electronics	Small WEEE: Computer mouse	IOGEAR	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Antibacterial characteristics.
Allsopp et al., 2007	Electronics	Small WEEE: Computer mouse	IOGEAR	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antibacterial characteristics.
Allsopp et al., 2007	Electronics	Small WEEE: Mobile phone	LG Electronics	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antibacterial characteristics.
Greßler et al., 2010	Coatings	Paints (facade painting)	Sto (Lotusan®)[6]	Thermoset	Silicone resin	Si	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect when in contact with water (rain).
Greßler et al., 2010	Coatings	Paints (facade painting)	Sto (StoColor Photosan®)[6]	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Photocatalytic self-cleaning effect in combination with UV light and water (rain), antimicrobial effect, decomposition of NO_X.
Greßler et al., 2010	Textiles	e.g. awnings, parasols, sails, tents, etc.	n.i.	Thermoplastic or thermoset	Polyester	Si	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect when in contact with water (rain).
Greßler et al., 2010	Coatings	Coatings	n.i.	Thermoplastic	PVC	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Photocatalytic self-cleaning effect in combination with UV light and water (rain), antimicrobial effect.

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Quilitz et al., 2008	Coatings	Coatings	n.i.	Thermoplastic	PVC	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Photocatalytic self-cleaning effect in combination with UV light and water (rain), antimicrobial effect.
Peet et al., 2009	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	P3HT	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	Higher performance levels.
Lee et al., 2010	Automotive industry	Fuel cells	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of electron shuttle properties, higher performance.
Lee et al., 2010	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of electron shuttle properties, higher performance.
Lee et al., 2010	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	Improvement of electron shuttle properties, higher performance.
Lee et al., 2010	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Higher performance.
Lee et al., 2010	Electronics	LED	n.i.	n.i.	n.i.	Quantum dots	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Lee et al., 2010	Coatings	Paints	n.i.	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics.
Potts et al., 2011	Electronics	Electrical devices	n.i.	n.i.	P3HT	GO	n/a	0.15-14.8 wt%	30-80 nm	n.i.	n.i.	Non-volatile memory storage system, barrier for inter-lamellar electron-hole recombination.
Potts et al., 2010	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	P3HT	GO	n/a					Improved energy storage, high electrical conductivity.
Delgado et al., 2011	Coatings	Antibacterial application	n.i.	Thermoplastic	PP	Cu	maybe	5 wt%	10 nm	n.i.	100 nm	Antimicrobial effects.

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Delgado et al., 2011	Coatings	Antibacterial application	n.i.	Thermoplastic	PP	CuO	maybe	5 wt%	40 cm	n.i.	> 100 nm	Antimicrobial effects.
Duncan 2011	Medical equipment	e.g. medical catheters, bandages, etc.	n.i.	n.i.	n.i.	Ag	n/a	2-4 µg/mL	40-50 nm	n.i.	n.i.	Antimicrobial effects.
Duncan, 2011	Coatings	Coatings food industry: Cutting boards, food storage containers, etc.	n.i.	n.i.	n.i.	Ag	n/a	2-4 µg/mL	40-50 nm	n.i.	n.i.	Antimicrobial effects.
Duncan, 2011	Textiles	n.i.	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
Duncan, 2011	Food and beverage packaging	-	n.i.	Thermoplastic	PVA	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Improved thermal properties, flame resistance, strength.
Duncan, 2011	Food and beverage packaging	-	n.i.	Thermoset	Chitosan	Ag	no	2-4 µg/mL	40-50 nm	n.i.	n.i.	Cumulative antimicrobial effects with chitosan, improved gas-barrier and tensile strength.
Duncan, 2011	Food and beverage packaging	Intelligent packaging	n.i.	n.i.	n.i.	Au	no	n.i.	n.i.	n.i.	n.i.	Detection of gasses, chemical contaminants, aromas, pathogens and thus improvement of quality and food safety.
Duncan, 2011	Food and beverage packaging	Intelligent packaging	n.i.	n.i.	n.i.	CNT	no	n.i.	n.i.	n.i.	n.i.	Detection of gasses, chemical contaminants, aromas, pathogens and thus improvement of quality and food safety.

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Duncan, 2011	Food and beverage packaging	Intelligent packaging	n.i.	n.i.	n.i.	Chitosan	no	n.i.	n.i.	n.i.	n.i.	Detection of gasses, chemical contaminants, aromas, pathogens and thus improvement of quality and food safety.
Duncan, 2011	Electronics	Large WEEE: Refrigerator	n.i.	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial characteristics, restrained growth of bacteria, suppression of bad odours.
Azeredo et al., 2011	Food and beverage packaging	-	n.i.	n.i.	Polymers, biopolymers	Nanoclay	yes	n.i.	1 nm (diameter)	n.i.	n.i.	Improved barrier properties, mechanical strength.
Azeredo et al., 2011	Food and beverage packaging	-	FMC BioPolymer (Avicel® PH)	Thermoset	Chitosan	CNR	yes	0-20 wt%	n.i.	n.i.	n.i.	Lightweight product, improved strength, tensile properties, gas barrier.
Azeredo et al., 2011	Food and beverage packaging	-	n.i.	Thermoplastic	PA, PE, PP, PVA	CNT (SWCNT, MWCNT)	no	n.i.	n.i.	n.i.	n.i.	Improvements of Young's modulus, tensile strength, decrease of water vapour transmission rate, antimicrobial effect.
Azeredo et al., 2011	Food and beverage packaging	-	n.i.	Thermoplastic	PP, PVA	SiO_2	yes	n.i.	n.i.	n.i.	n.i.	Improvement of tensile strength, oxygen barrier, thermal and mechanical properties.

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Azeredo et al., 2011	Food and beverage packaging	-	n.i.	n.i.	n.i.	Ag	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect, absorption and decomposition of ethylene and thus extended shelf life.
Azeredo et al., 2011	Food and beverage packaging	-	n.i.	n.i.	n.i.	TiO_2	yes	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect through photocatalysis, oxygen scavenger.
Azeredo et al., 2011	Food and beverage packaging	-	n.i.	n.i.	n.i.	Chitosan	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect.
Díez-Pascual et al., 2015	Electronics	Electrical devices	n.i.	Thermoset, Thermoplastic, Elastomer	Chitosan, Epoxy, PE, PET, PMMA, PU, PVA	Graphene	n/a	0.05-6.0 wt%	1 nm (diameter) 100 nm-µm (lateral)	n.i.	n.i.	Improvement of thermal conductivity, electrical conductivity, strength, Young's modulus.
Díez-Pascual, 2015	Food and beverage packaging	-	n.i.	Thermoset, Thermoplastic, Elastomer	Chitosan, Epoxy, PA, PE (HDPE, LDPE), PLA, PP, PS, PU	Nanoclay	yes	0.5-10 wt%	1 nm (diameter) 100 nm-µm (lateral)	n.i.	n.i.	Improvement of strength, toughness, increased tear and fire resistance, barrier properties.
Díez-Pascual, 2015	Automotive industry	Structural components	n.i.	Thermoset, Thermoplastic, Elastomer	Chitosan, Epoxy, PA, PE (HDPE, LDPE), PLA, PP, PS, PU	Nanoclay	n/a	0.5-10 wt%	1 nm (diameter) 100 nm-µm (lateral)	n.i.	n.i.	Improvement of strength, toughness, increased tear and fire resistance, barrier properties.
Díez-Pascual, 2015	Electronics	Electrical devices	n.i.	Thermoset, Thermoplastic, Elastomer	Chitosan, Epoxy, PA, PE (HDPE, LDPE), PLA, PP, PS, PU	Nanoclay	n/a	0.5-10 wt%	1 nm (diameter) 100 nm-µm (lateral)	n.i.	n.i.	Improvement of strength, toughness, increased tear and fire resistance, barrier properties.

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Duncan and Pillai, 2015	Coatings	Paints (exterior)	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Coatings	Paints (exterior)	n.i.	n.i.	n.i.	Ag	n/a	1.5 mg/m²	< 15 nm	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Coatings	Paints (exterior)	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Coatings	Paints (exterior)	n.i.	n.i.	n.i.	CNF	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Coatings	Adhesives	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Coatings	Adhesives	n.i.	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Coatings	Adhesives	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Coatings	Adhesives	n.i.	n.i.	n.i.	CNF	n/a	n.i.	n.i.	n.i.	n.i.	Self-cleaning effect, improved strength, antimi-crobial effect.
Duncan and Pillai, 2015	Automotive industry	Structural components	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improved heat shielding, strength, wear and abrasion resistance, light weight.

Reference	**Product category**	**Product specification** (exemplary)	**Company or product trade name** (exemplary)	**Plastic type**	**Source-based name** (exemplary)	**Embedded ENM**	**(EU) No 10/2011**	**Mass** (wt% or concentration)	**Primary size** (diameter)	**Aggregate size** (diameter)	**Agglomerate size** (diameter)	**Enhanced characteristics**
Duncan and Pillai, 2015	Automotive industry	Structural components	n.i.	n.i.	n.i.	CNF	n/a	n.i.	n.i.	n.i.	n.i.	Improved heat shielding, strength, wear and abrasion resistance, light weight.
Duncan and Pillai, 2015	Automotive industry	Structural components	n.i.	n.i.	n.i.	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improved heat shielding, strength, wear and abrasion resistance, light weight.
Duncan and Pillai, 2015	Food and beverage packaging	-	n.i.	n.i.	n.i.	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Improved barrier properties.
Duncan and Pillai, 2015	Food and beverage packaging	-	n.i.	n.i.	n.i.	SiO_2	yes	n.i.	n.i.	n.i.	n.i.	Improved barrier properties.
Duncan and Pillai, 2015	Food and beverage packaging	-	n.i.	Thermoplastic	PE (HDPE, LDPE), PP	Ag	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect.
Duncan and Pillai, 2015	Food and beverage packaging	-	n.i.	n.i.	n.i.	CNT	no	n.i.	n.i.	n.i.	n.i.	Improved electromagnetic shielding.
Duncan and Pillai, 2015	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improved electrical and thermal conductivity.
Duncan and Pillai, 2015	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	n.i.	Graphene	n/a	n.i.	n.i.	n.i.	n.i.	Improved electrical and thermal conductivity.
Duncan and Pillai, 2015	Electronics	Electrical devices	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improved electrical and thermal conductivity.
Duncan and Pillai, 2015	Electronics	Electrical devices	n.i.	n.i.	n.i.	Graphene	n/a	n.i.	n.i.	n.i.	n.i.	Improved electrical and thermal conductivity.
Duncan and Pillai, 2015	Electronics	Large WEEE: Washing machine	n.i.	n.i.	n.i.	Ag	n/a	n.i.	10 nm	n.i.	n.i.	Antimicrobial effect.
Duncan and Pillai, 2015	Sporting equipment	n.i.	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improved strength, toughness, stiffness.

Reference	**Product category**	**Product specification** (exemplary)	**Company or product trade name** (exemplary)	**Plastic type**	**Source-based name** (exemplary)	**Embedded ENM**	**(EU) No 10/2011**	**Mass** (wt% or concentration)	**Primary size** (diameter)	**Aggregate size** (diameter)	**Agglomerate size** (diameter)	**Enhanced characteristics**
Duncan and Pillai, 2015	Sporting equipment	n.i.	n.i.	n.i.	n.i.	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improved strength, flame retardancy.
Duncan and Pillai, 2015	Sporting equipment	n.i.	n.i.	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect.
Duncan and Pillai, 2015	Textiles	Clothing	n.i.	n.i.	n.i.	Ag	n/a	57.7 µg/g	10-500 nm	n.i.	n.i.	Antimicrobial effect.
Duncan and Pillai, 2016	Textiles	Clothing	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect.
Greßler et al., 2014	Textiles	Outdoor clothing, lingerie, socks, etc.	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
Greßler et al., 2014	Textiles	Outdoor clothing, lingerie, socks, etc.	n.i.	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect.
Greßler et al., 2014	Textiles	Outdoor clothing, lingerie, socks, etc.	n.i.	n.i.	n.i.	ZnO	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
Greßler et al., 2014	Textiles	Outdoor clothing, lingerie, socks, etc.	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Dirt and water-repellent effect.
Greßler et al., 2014	Coatings	Paints and lacquers	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
Greßler et al., 2014	Coatings	Paints and lacquers	n.i.	n.i.	n.i.	Ag	n/a	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect.
Greßler et al., 2014	Coatings	Paints and lacquers	n.i.	n.i.	n.i.	ZnO	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
Greßler et al., 2014	Coatings	Paints and lacquers	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improved abrasion resistance.
Greßler et al., 2014	Coatings	Paints and lacquers	n.i.	n.i.	n.i.	Al_2O_3	n/a	n.i.	n.i.	n.i.	n.i.	Improved abrasion resistance.
Greßler et al., 2014	Sporting equipment	Tennis racquets	n.i.	n.i.	n.i.	CNT	n/a	n.i.	n.i.	n.i.	n.i.	Improved tensile and breaking strength.
Greßler et al., 2014	Sporting equipment	Tennis racquets	n.i.	n.i.	n.i.	Fullerenes	n/a	n.i.	n.i.	n.i.	n.i.	Improved tensile and breaking strength.
Greßler et al., 2014	Sporting equipment	Tennis racquets	n.i.	n.i.	n.i.	Graphene	n/a	n.i.	n.i.	n.i.	n.i.	Improved tensile and breaking strength.
Greßler et al., 2014	Electronics	Electrical devices	n.i.	n.i.	n.i.	Quantum dots	n/a	n.i.	n.i.	n.i.	n.i.	n.i.

Reference	Product category	Product specification (exemplary)	Company or product trade name (exemplary)	Plastic type	Source-based name (exemplary)	Embedded ENM	(EU) No 10/2011	Mass (wt% or concentration)	Primary size (diameter)	Aggregate size (diameter)	Agglomerate size (diameter)	Enhanced characteristics
Hu et al., 2014	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	PEDOT, P3HT	Graphene	n/a	n.i.	n.i.	n.i.	n.i.	Improved electrical conductivity, power conversion efficiency.
Hu et al., 2014	Electronics	Photovoltaics' (solar cells)	n.i.	n.i.	PEDOT, P3HT	ITO	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Hu et al., 2014	Electronics	Electrical devices	n.i.	Thermoset	Epoxy	Graphene	n/a	5-40 wt%	n.i.	n.i.	n.i.	Improved thermal conductivity.
Hu et al., 2014	Electronics	Electrical devices	n.i.	Thermoplastic	PP	GO	n/a	n.i.	n.i.	n.i.	n.i.	Improved electrical properties, mechanical strength.
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Silicone resin	Ag	n/a	> 0.1 wt%	n.i.	n.i.	n.i.	Antimicrobial effect.
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Resin, silicon resin	SiO_2	n/a	0.1-5 wt%	n.i.	n.i.	n.i.	Water and dirt repellence, antimicrobial effect, scratch resistance, improved flame resistance.
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Silicone resin	TiO_2	n/a	0.1-5 wt%	n.i.	n.i.	n.i.	UV protection, photocatalytic degradation of dirt, water repellent, antimicrobial effect.
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Silicone resin	ZnO	n/a	0.1-5 wt%	n.i.	n.i.	n.i.	UV protection, antimicrobial effect, water repellent.
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Silicone resin	Al_2O_3	n/a	0.1-5 wt%	n.i.	n.i.	n.i.	Improved scratch resistance, flame resistance.
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Silicone resin	CeO_2	n/a	0.1-5 wt%	n.i.	n.i.	n.i.	UV protection.
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Silicone resin	MgO	n/a	0.1-5 wt%	n.i.	n.i.	n.i.	Antimicrobial effect.

Reference	Product category	Product specification (exemplary)	Company or product trade name (exemplary)	Plastic type	Source-based name (exemplary)	Embedded ENM	(EU) No 10/2011	Mass (wt% or concentration)	Primary size (diameter)	Aggregate size (diameter)	Agglomerate size (diameter)	Enhanced characteristics
Hincapié et al., 2015	Coatings	Facade coating (interior and exterior)	n.i.	Thermoset	Silicone resin	Nanoclay	n/a	0.1-5 wt%	n.i.	n.i.	n.i.	Water repellent, scratch resistant, flame resistant.
Kotal and Bhowmick, 2015	Automotive industry	Structural components	n.i.	n.i.	n.i.	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improved heat resistance.
Kotal and Bhowmick, 2015	Food and beverage packaging	-	n.i.	n.i.	n.i.	Nanoclay	yes	n.i.	n.i.	n.i.	n.i.	Improved barrier properties.
Kotal and Bhowmick, 2015	Electronics	Electrical devices	n.i.	n.i.	n.i.	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improved physical and thermal properties.
Kotal and Bhowmick, 2015	Medical equipment	Medical devices	n.i.	n.i.	n.i.	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	
Kotal and Bhowmick, 2015	Coatings	Adhesives	n.i.	Thermoplastic	PVA	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improved tensile modulus, gas barrier properties.
Wohlleben et al., 2016	Automotive industry	Tires	n.i.	Elastomer	PU	CB	n/a	> 40 wt% of rubber content	n.i.	n.i.	n.i.	Improved photoprotection, increased mileage, improved road grip.
Wohlleben et al., 2016	Automotive industry	Tires	n.i.	Elastomer	PU	CB+CNT	n/a	40 wt% (CB) 4 wt% (CNT) -> both refer to rubber content	n.i.	n.i.	n.i.	Improved photoprotection, increased mileage, improved road grip.
Wohlleben et al., 2016	Automotive industry	Tires	n.i.	Elastomer	PU	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Increased mileage, improved road grip.
Greßler et al., 2017	Food and beverage packaging	e.g. crisps, cheese, etc.	n.i.	n.i.	Polymers	SiO_2	yes	n.i.	1-100 nm[1]	0.1-1 µm[1]	0.3 µm- mm[1]	Improved stiffness, tensile strength, barrier properties.
Greßler et al., 2017	Food and beverage packaging	e.g. beverages, sauces, dressings, sausages, pet food, etc.	n.i.	Thermoplastic	EVOH	Nanokaolin	yes	< 12 wt%	< 100 nm (thickness)	n.i.	n.i.	Improved barrier properties.

Reference	**Product category**	**Product specification** (exemplary)	**Company or product trade name** (exemplary)	**Plastic type**	**Source-based name** (exemplary)	**Embedded ENM**	**(EU) No 10/2011**	**Mass** (wt% or concentration)	**Primary size** (diameter)	**Aggregate size** (diameter)	**Agglomerate size** (diameter)	**Enhanced characteristics**
Greßler et al., 2017	Food and beverage packaging	-	n.i.	n.i.	n.i.	CB	yes	max. 2.5 wt%[1]	10-300 nm[1]	100-1200 nm[1]	300 nm- m[1]	UV protection.
Greßler et al., 2017	Food and beverage packaging	-	n.i.	Thermoplastic	PET	TiN	yes	max. 20 mg/kg[1]	20 nm[1]	n.i.	100-500 nm[1]	Improvement of thermal stability.
Greßler et al., 2017	Food and beverage packaging	-	n.i.	Thermoplastic	PVC	Copolymers: Butadiene, ethyl acrylate, methyl methacrylate, styrene	yes	max. 10 wt%[1]	n.i.	n.i.	n.i.	Impact modifier.
Greßler et al., 2017	Food and beverage packaging	-	n.i.	n.i.	n.i.	Nanoclay	yes	1-5 wt%	few nm (diameter) few 100 - few 1000 nm (length)	n.i.	n.i.	Improved shelf life, aroma stability.
Greßler et al., 2017	Food and beverage packaging	-	n.i.	n.i.	n.i.	ZnO	yes	n.i.	n.i.	n.i.	n.i.	UV protection.
Greßler et al., 2017	Food and beverage packaging	-	n.i.	n.i.	n.i.	Ag	no	n.i.	n.i.	n.i.	n.i.	Antimicrobial effect.
Greßler et al., 2017	Food and beverage packaging	-	n.i.	n.i.	n.i.	$CaCO_3$	no	n.i.	n.i.	n.i.	n.i.	Improved tensile strength, thermal resistance.
Greßler et al., 2017	Food and beverage packaging	-	n.i.	n.i.	n.i.	TiO_2	yes	n.i.	n.i.	n.i.	n.i.	UV protection, antimicrobial effect.
Greßler et al., 2017	Automotive industry	Tires	n.i.	Elastomer	n.i.	CB	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Laux et al., 2018	Automotive industry	Structural components	n.i.	Thermoset	Epoxy	Graphene	n/a	n.i.	n.i.	n.i.	n.i.	Improved tensile strength.
Laux et al., 2018	Coatings	Paint	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Laux et al., 2018	Coatings	Coatings	n.i.	n.i.	n.i.	SiO_2	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Laux et al., 2018	Coatings	Hybrid coatings	n.i.	n.i.	Polymers	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Photocatalytic effect.

Reference	**Product category**	**Product specification** (exemplary)	**Company or product trade name** (exemplary)	**Plastic type**	**Source-based name** (exemplary)	**Embedded ENM**	**(EU) No 10/2011**	**Mass** (wt% or concentration)	**Primary size** (diameter)	**Aggregate size** (diameter)	**Agglomerate size** (diameter)	**Enhanced characteristics**
Laux et al., 2018	Textiles	n.i.	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	UV protection.
Watson-Wright et al., 2017	Automotive industry	n.i.	BASF	Thermoplastic	PC, PU	CNT	n/a	0.1 and 3 wt%	n.i.	n.i.	n.i.	Improved scratch resistance, tensile strength, Young's modulus, conductivity.
Watson-Wright et al., 2017	Textiles	n.i.	BASF	Thermoplastic	PU	CNT	n/a	0.1 wt%	n.i.	n.i.	n.i.	Improved scratch resistance, tensile strength, Young's modulus, conductivity.
Watson-Wright et al., 2017	Food and beverage packaging	-	MARINA	Thermoplastic	PE	Fe_2O_3	yes	n.i.	n.i.	n.i.	n.i.	Improved magnetic, optical, electrical properties.
Watson-Wright et al., 2017	Food and beverage packaging	-	n.i.	Thermoplastic	PP	CNT	no	n.i.	n.i.	n.i.	n.i.	Improved scratch resistance, tensile strength, Young's modulus, conductivity.
Watson-Wright et al., 2017	Food and beverage packaging	-	n.i.	Thermoplastic	EVA	TiO_2	yes	n.i.	n.i.	n.i.	n.i.	Improved magnetic, optical, electrical properties.
Watson-Wright et al., 2017	Sporting equipment	n.i.	n.i.	Thermoplastic	PC, PP, PU	CNT	n/a	0.1 and 3 wt%	n.i.	n.i.	n.i.	Improved scratch resistance, tensile strength, Young's modulus, conductivity.
Watson-Wright et al., 2017	Electronics	n.i.	n.i.	Thermoplastic	PC, PP	CNT	n/a	3 wt%	n.i.	n.i.	n.i.	Improved scratch resistance, tensile strength, Young's modulus, conductivity.

Reference	Product category	Product specification (exemplary)	Company or product trade name (exemplary)	Plastic type	Source-based name (exemplary)	Embedded ENM	(EU) No 10/2011	Mass (wt% or concentration)	Primary size (diameter)	Aggregate size (diameter)	Agglomerate size (diameter)	Enhanced characteristics
Watson-Wright et al., 2017	Medical equipment	Biomedical devices	n.i.	Thermoplastic	EVA	TiO_2	n/a	2 and 5 wt%	n.i.	n.i.	n.i.	Improved magnetic, optical, electrical properties.
Brame et al., 2018	Coatings	Anti-corrosive coating	n.i.	n.i.	Polymers	CNT (MWCNT)	n/a	n.i.	n.i.	n.i.	n.i.	Improved strength, conductivity.
Brame et al., 2018	Coatings	n.i.	n.i.	n.i.	Polymers	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Brame et al., 2018	Textiles	n.i.	n.i.	n.i.	n.i.	CNT (MWCNT)	n/a	n.i.	n.i.	n.i.	n.i.	Improved strength, conductivity.
Brame et al., 2018	Automotive industry	n.i.	n.i.	n.i.	n.i.	CNT (MWCNT)	n/a	n.i.	n.i.	n.i.	n.i.	Improved strength, conductivity.
Han et al., 2018	Food and beverage packaging	e.g. bread, Meal-Ready-to-Eat, etc.	Perkin Elmer Inc.	Thermoplastic	PE (LDPE)	Nanoclay	yes	3.3 and 5.4wt%	3.16 nm	n.i.	n.i.	Improved barrier properties, thermal properties, antimicrobial effect.
Singh et al., 2019	Coatings	Coatings for cars, facade painting, etc.	n.i.	n.i.	n.i.	TiO_2	n/a	n.i.	n.i.	n.i.	n.i.	Improved surface cleanability.
Singh et al., 2019	Coatings	Coatings for cars, facade painting, etc.	n.i.	Thermoplastic	PA	SiO_2	n/a	0.5, 1.5, 10, 20 wt%	n.i.	n.i.	n.i.	Improved corrosion protection, thermal stability.
Singh et al., 2019	Coatings	Coatings for cars, facade painting, etc.	n.i.	n.i.	n.i.	ZnO	n/a	n.i.	n.i.	n.i.	n.i.	Improved corrosion protection.
Singh et al., 2019	Coatings	Coatings for cars, facade painting, etc.	n.i.	Thermoplastic	PA	Fe_2O_3	n/a	0.3, 1.5 wt%	n.i.	n.i.	n.i.	Improved coloristic effect.
Singh et al., 2019	Coatings	Coatings for cars, facade painting, etc.	n.i.	n.i.	n.i.	$Al(OH)_3$	n/a	n.i.	n.i.	n.i.	n.i.	n.i.
Singh et al., 2019	Coatings	Coatings for cars, facade painting, etc.	n.i.	Thermoplastic	PA	CuO	n/a	0.3, 1.5 wt%	n.i.	n.i.	n.i.	Improved coloristic effect.
Singh et al., 2019	Coatings	Coatings for cars, facade painting, etc.	n.i.	n.i.	n.i.	Nanoclay	n/a	n.i.	n.i.	n.i.	n.i.	Improved thermal stability.

Appendix 2: Supplementary information and assumptions for the MFA.

a) Vehicle tires	**Description**	**Source**
Passenger cars	Passenger cars include the categories “Personenkraftwagen Kl. M1”, “Vierrädrige Kraftfahrzeuge Kl. L7e” and “Vierrädrige Leichtkraftfahrzeuge Kl. L6e”. They all have four tires.	Statistik Austria, 2018a; BMDW, 2019
	The average mileage of 2018 is an interpolated value which is calculated based on the average mileage of 2015 and of the forecast value of 2025 (scenario 1 - business as usual) from the study “Verkehrsprognose Österreich 2025”.	BMVIT, 2009b
	All passenger cars have summer and winter tires which are relevant for the calculation of the stock.	Own assumption
	Transit traffic of passenger cars is not included in the MFA due to lack of data.	-
	The average weight of one passenger car tire is based on the weight of 205/55 R 16 91V ContiPremiumContact 5.	Continental Reifen, 2013
Motorbikes (2 and 3 tires)	It is assumed, that motorbikes with 2 and 3 tires drive the same average distance per year. This annual mileage is taken from a German study performed for the year 2015. It is assumed that Austrian motorbikes drive the same annual distance as German ones. Since this figure is from 2015, the value is interpolated with the percental annual increase calculated for the category of passenger cars.	BMVIT, 2009b; KBA, 2018; BMDW, 2019
	Motorbikes are not used during the winter.	Own assumption
	Transit traffic of motorbikes is not included in the MFA due to lack of data.	-
	Motorbike tires have the same weight as passenger car tires.	Own assumption
Trucks incl. transit traffic	Trucks include the categories “Kl. N1 (bis 3,5t Gesamtgewicht)”, “Kl. N2 (über 3,5t bis 12t Gesamtgewicht)” and “Kl. N3 (über 12t Gesamtgewicht)”. They all have four tires.	Statistik Austria, 2018a; BMDW, 2019
	The average mileage of 2018 is an interpolated value which is calculated based on the average mileage of 2015 and of the forecast value of 2025 (scenario 1 - business as usual) from the study “Verkehrsprognose Österreich 2025”. The annual mileage already includes transit traffic and refers to Austrian as well as foreign trucks. It is assumed, that the share of foreign trucks in the year 2018 is the same is in the year 2002.	BMVIT, 2009a
	All trucks drive with all-round tires.	Eppel, 2019
	An average value is used for the weight of truck tires.	IAC Publishing, s.a; Michelin, 2019

Articulated lorries	All articulated lorries have 18 tires.	Wikipedia, 2018
	The average mileage of 2018 is an interpolated value which is calculated based on the average mileage of 2015 and of the forecast value of 2025 (scenario 1 - business as usual) from the study “Verkehrsprognose Österreich 2025”. The annual mileage already includes transit traffic and refers to Austrian as well as foreign articulated lorries. It is assumed that the share of foreign articulated lorries in the year 2018 is the same is in the year 2002.	BMVIT, 2009a
	All articulated lorries drive with all-round tires.	Eppel, 2019
	Tires of articulated lorries have the same weight as trucks.	Own assumption
Campers	Campers have four tires.	BMDW, 2019
	Campers are not used during the winter.	Own assumption
	The average mileage is calculated with the figure of passenger cars.	Own assumption
	Transit traffic of campers is not included in the MFA because of lacking data.	-
	Tires of campers have the same weight as passenger car tires.	Own assumption
Busses	Busses have four tires.	BMDW, 2019
	Busses have summer and winter tires which is relevant for the calculation of the stock.	Own assumption
	The average mileage is based on a study from the BMVIT.	Salmhofer, 2019
	Transit traffic of busses is not included in the MFA because of lacking data.	-
	Bus tires have the same weight as truck tires.	Own assumption
Others	This category includes “Anhänger Kl. O und R”, “Wohnanhänger”, “Zugmaschinen”, “Motor- und Transportkarren”, “Selbstfahrende Arbeitsmaschinen”, “Erntemaschinen”, “Sonstige Kraftfahrzeuge”, “Landwirtschaftliche Arbeitsanhänger”, “Anhänger Arbeitsmaschinen” and “Sonderanhänger”.	Statistik Austria, 2018a
	Vehicles of the category other vehicles have all four tires.	Own assumption
	There are no separate tires for winter and summer.	Own assumption
	The average mileage is calculated with the figure of passenger cars. It is assumed, that other vehicles have half the mileage of passenger cars.	Own assumption
	Transit traffic of this category is not included in the MFA because of lacking data.	-
	Tires from the category other vehicles have the same weight as passenger car tires.	Own assumption
Spare tires	Due to the fact that there is no legislation on spare tires in Austria they are not considered in the calculation for the stock of tires.	Eppel, 2019

Composition of an average vehicle tire	The composition of a 205/55 R 16 91V ContiPremiumContact 5 passenger car tire is assumed to be the same for all vehicle tires in Austria.		Own assumption
	Rubber (natural and synthetic rubber)	41%	Continental Reifen, 2013
	Fillers (Silica, Chalk, etc.)	8%	Continental Reifen, 2013; Environment Canada and Health Canada, 2013
	CB	22%	Continental Reifen, 2013; Environment Canada and Health Canada, 2013
	Reinforcing materials (steel, polyester, rayon, nylon)	15%	Continental Reifen, 2013
	Plasticizers (oils and resins)	6%	Continental Reifen, 2013
	Chemicals for vulcanisation (sulphur, zinc oxide, various other chemicals)	6%	Continental Reifen, 2013
	Anti-ageing agents and other chemicals	2%	Continental Reifen, 2013
Calculation weight of vehicle tires	Only the plastic share including fillers (i.e. 85 wt%) is considered. The plastic share of a vehicle tire includes the rubber, plasticizers, chemicals for vulcanization as well as anti-ageing agents and other chemicals. Fillers include mainly CB (nano- and microsized) as well as to a smaller share Silica, Chalk or the like. Other reinforcing materials are not relevant for a further consideration of CB-particles as these particles are solely incorporated in the plastic part of a tire. This assumption also facilitates the whole system under consideration. Otherwise, there would be additional flows from each EOL process that would incorporate “reinforcing materials” like for instance steel mesh.		Continental Reifen, 2013; own assumption
	The average weight of a passenger car tire is 8.5 kg (i.e. light tire). This figure corresponds to the 205/55 R 16 91V ContiPremiumContact 5 passenger car tire.		Continental Reifen, 2013
	The average weight of a truck tire is 35.15 kg (i.e. heavy tire).		IAC Publishing, s.a; Michelin, 2019
	The assumed plastic type for all vehicle tires is styrene-butadiene rubber (SBR).		IISRP, 2012

b) CB	Description	Reference
wt% whole vehicle tire	A CB-share of 22 wt% for all vehicle tires is assumed. This CB share applies to a whole tire including all its parts.	Continental Reifen, 2013; Environment Canada and Health Canada, 2013
wt% plastic part vehicle tire	Based on the CB-share of 22 wt%, a CB-share of about 26 wt% is calculated for the plastic part (i.e. 85 wt%).	Continental Reifen, 2013; Environment Canada and Health Canada, 2013
Calculation weight	The total weight of CB incorporated 2018 in Austrian tires (stock and use-phase) is calculated based on the total weight of the plastic share incl. fillers of all tires. For the stock all registered vehicles of 2018 incl. winter and summer tires are taken into consideration and for the use-phase only one set of vehicle tires.	IAC Publishing, s.a; Wohlleben et al., 2016; Statistik Austria, 2018a; Michelin, 2019
CB-content	All tires include CB.	Eppel, 2019, expert discussion

c) Wear and tear release	Description	Reference
Calculation annual wear and tear release different vehicle tires	The figures for wear and tear release of motorbikes, trucks, articulated lorries and busses are taken from a German study. The values from this study are based on a review of already existing literature. It is assumed, that in Austria the wear and tear emissions for these vehicle types are the same. Each vehicle specific amount of wear and tear [mg/km] is multiplied with the respective annual mileage from the vehicle type under consideration.	Hillenbrand et al., 2005
Calculation annual wear and tear release passenger cars	The figures for the wear and tear release of passenger cars, campers and others are taken from a German ADAC study. In this study the test vehicle was a VW Golf. While conducting the experiment different tires were used (e.g. summer, winter, sport). One of the tires used was also a 205/55 R 16 tire. During the test the vehicle under consideration was driving over 15 000 km under real conditions. Every 2 500 km, the profile depth and weight loss were measured using a laser measuring device and digital scales. In the end an average value of the wear and tear release was calculated. It is assumed, that in Austria the wear and tear emissions are the same.	ADAC, 2019; Eppel, 2019
Share CB	26 wt% of the total wear and tear release is CB, as a homogenous composition of the vehicle tires (fillers and polymer) is assumed. This figure results from the assumption, that 85 % of an average vehicle tire consists of plastic parts and fillers.	Continental Reifen, 2013Environment Canada and Health Canada, 2013
Size	The major part of the wear and tear release is larger than 2.5 µm.	Düring et al., 2004

c) Calculation free nanofiller release	**Description**	**Reference**
Scenario 1	Scenario 1 - “rainy days”: This scenario applies to a direct run-off of the released wear and tear into the sewage channel system. This scenario applies to all rainy days.	Wohlleben et al., 2016
	Wear and tear release < 5 µm with direct run-off is 1.6 % of the total solids released per year.	Wohlleben et al., 2016
	The percentage of rainy days is calculated as a mean value of the annual average rainy days of each of the 9 capital cites of the federal states of Austria.	Tulun, s.a.
Scenario 2	Scenario 2 - “days without rain”. This scenario applies to a delayed run-off of the released wear and tear into the sewage channel system. This scenario applies to all days without rain.	Wohlleben et al., 2016
	Wear and tear release < 5 µm is 4.0 % with delayed run-off of the total solids released per year.	Wohlleben et al., 2016
	The percentage of days without rain is calculated in the same way as for days with rain.	Tulun, s.a.
Free nanofiller content	Free nanofiller release is 0.045% of the released fragments. These released fragments refer to the figures calculated in S1 and S2 with the percental shares of 1.6 and 4.0%.	Wohlleben et al., 2016
Particle behaviour free nanofillers	It is important to keep in mind, that CB tends to form aggregates and agglomerates.	Kole et al., 2017

d) Processes and flows for MFA	**Description**	**Reference**
CB content	For the calculation of the mass of all different processes and flows on the substance level a CB content of 22 % of the whole vehicle tire is assumed. With respect to the plastic part of a vehicle tire, the CB content is 26 wt%.	Continental Reifen, 2013; Environment Canada and Health Canada, 2013
I1 Import tires	The figure for imported vehicle tires [t] is based on the new registered vehicles 2018 incl. winter and summer tires for passenger cars and busses. Only the plastic part of the vehicle tires is taken into account.	Statistik Austria, 2018b; Eppel, 2019
P1 Total tires stock (summer/winter)	The stock of all vehicle tires [t] is calculated based on all registered vehicles in the year 2018 incl. winter and summer tires for passenger cars and busses. Only the plastic part of the vehicle tires is taken into account.	Statistik Austria, 2018a; Eppel, 2019
F1.1 Tires in use	The figure for vehicle tires in use [t] is based on all registered vehicles in the year 2018. Only the plastic part of the vehicle tires is taken into account.	Statistik Austria, 2018a
F1.2 EOL tires	The amount of EOL tires (import, export, EOL tires Austria) is taken from the “Status Report of the current Austrian Federal Waste Management Plan”.	BMNT, 2019

P 2 Use phase	Vehicle tires in the use phase [t] are calculated based on all registered vehicles in the year 2018. Only the plastic part of the vehicle tires is taken into account.	Statistik Austria, 2018a
F2.1 Wear and tear release	This figure is based on wear and tear release studies.	Hillenbrand et al., 2005; ADAC, 2019
F3.1, F3.2, F3.3, F3.4 Diffuse emissions to surface water, soil, air (TSP) and WWTP	These figures are based on different TCs (see appendix 3 and 4 for more information on TCs.)	Luekewille et al., 2002, Hillenbrand et al., 2005; Gottschalk et al., 2015; ADAC, 2019
F4.1 EOL tires for recycling	It is assumed that EOL tires (except tires for retreading) are sent into a recycling plant in a first step. There, the still useable parts are processed into granulate (F7.3 Export granulate for new products). No longer useable parts are sent to a WIP (F7.1 Shredding residue and F7.2 Filter residue).	BMNT, 2019; own assumption
F4.2 Tires for retreading	The figure of vehicle tires for retreading is based on the “Status Report of the current Austrian Federal Waste Management Plan”. It is important to notice that only truck tires are retreaded in Austria.	BMNT, 2019; Eppel, 2019
F5.1 Effluent surface water	This figure is based on a TC.	Wang et al., 2012
F5.2 Sewage sludge for agriculture (soil), F5.4 Sewage sludge for incineration	These figures are based on TCs and on the current “Status Report of the Austrian Federal Waste Management Plan”.	Wang et al., 2012; BMNT, 2019
F5.3 Further treatment	This figure is based on a TC and on the “Status Report of the current Austrian Federal Waste Management Plan”. Further treatment refers to a treatment in MBWTP, biogas plants etc.	Wang et al., 2012; BMNT, 2019
F6.1 Retreaded tires	The share of retreaded vehicle tires is based on the assumption that 20 % of the top-layer of tires is mechanically peeled off (i.e. F6.3 Shredding residue) and 0.7 % are retained by a filter (F.6.2 Filter residue). The remaining 79.3 % are put on the market again and will be sold in the upcoming years.	Wohlleben et al., 2016; own assumption
F6.2 Filter residue, F7.2 Filter residue	During the mechanical processes of retreading and recycling a release of particles is assumed. Moreover, it is assumed, that all released particles are retained by a filter. The amount for filter residues for retreading and recycling is calculated based on a figure from Wohlleben et al., 2016. However, this used figure does not specifically refer to retreading or recycling processes. Nevertheless, it is still used due to lack of other data. Furthermore, it is assumed that these filter residues do not fulfil the requirement of TOC < 5 % DM according to the Austrian Landfill Regulation. Consequently, they are burned in a WIP.	Wohlleben et al., 2016; DVO, 2008; own assumption
F6.3 Shredding residue, F7.1 Shredding residue	It is assumed that the shredding residues of the processes retreading and recycling do not fulfil the requirement of TOC < 5 % DM according to the Austrian Landfill Regulation. Consequently, these shredding residues are burned in a WIP.	DVO, 2008; own assumption

F7.3 Export granulate for new products	It is assumed that the whole granulate produced is exported in order to produce new products. The granulate is used for new vehicle tire production, asphalt and bitumen production, in the leisure and sport sector etc.	MRH, s.a.; BMNT, 2019; own assumption
F8.1 Slag and ashes	This figure is based on a TC. In Austria slag and ashes from WIP are deposited on an adequate landfill.	Mueller et al., 2013; DVO, 2008
F8.2 Export fly ash to underground landfill (Ger)	This figure is based on a TC. The hazardous fly ash has to be brought to an underground landfill. As a lot of hazardous waste from Austria is brought to Germany, in this case it is also assumed that the underground landfill is situated in Germany.	Mueller et al., 2013; DVO, 2008
F8.3 Elimination	This figure is based on a TC.	Mueller et al., 2013; DVO, 2008

Appendix 3: Mass balance MFA level of “carbon black”; performed with STAN.

Name of flow	"stochastic" transfer coefficients* [%]	Nano-specific transfer coefficients [%]	Uncertainty based on Laner et al., 2015 [%]	Reference	Calculated quantity [t/a]	Internal/external system flows
I 0 Import tires			14.0%	Statistik Austria, 2018b	7 700	P 1
P 1 All vehicle tires (summer/winter)			14.0%	Statistik Austria, 2018a	106 280	
F 1.1 Tires in use			14.0%	Statistik Austria, 2018a	68 490	P 2
F 1.2 EOL tires AT			5.0%	BMNT, 2019	10 400	P 3
P 2 Use phase			14.0%	Statistik Austria, 2018a	68 490	
F 2.1 Wear and tear release			14.0%	ADAC, 2019; Hillenbrand et al. 2005	4 020	F4.1, F4.2., F4.3, F4.4 (diffuse emissions)
F 2.2 Re-use vehicle tires next season			14.0%		64 470	
P 3 Wear and tear release				ADAC, 2019; Hillenbrand et al., 2005	4 020	
F 3.1 Diffuse emissions surface water		25.0%	14.0%	Gottschalk et al., 2015	1 000	Output (surface water)
F 3.2 Diffuse emissions soil		12.5%	14.0%	Gottschalk et al., 2015	500	Output (soil)
F 3.3 Diffuse emissions air (TSP)	53.2%		14.0%	ADAC, 2019; Hillenbrand et al., 2005; Luekewille et al., 2002	2 140	Output (air)
F 3.4 Diffuse emissions WWTP		9.4%	14.0%	Gottschalk et al., 2015	380	P 5
I 4 Import EOL tires			5.0%	BMNT, 2019	2 800	P 3
P 4 EOL treatment					13 200	
F4.1 EOL tires for recycling	47.7%		5.0%	BMNT, 2019	6 300	P 7
F4.2 EOL tires for retreading	2.5%		5.0%	BMNT, 2019	330	P 6
F 4.3 Export EOL tires	49.8%		5.0%	BMNT, 2019	6 570	Output (export)

P 5 WWTP					380	
F 5.1 Effluent surface water		5.0%	5.0%	Wang et al., 2012	20	Output (surface water)
F 5.2 Sewage sludge for agriculture (soil)	19.0%		5.0%	Wang et al., 2012; BMNT, 2019	70	Output (soil)
F 5.3 Further treatment	23.7%		5.0%	Wang et al., 2012; BMNT, 2019	90	-
F 5.4 Sewage sludge for incineration	52.3%		5.0%	Wang et al., 2012; BMNT, 2019	200	P 8
P 6 Retreading			-		330	
F 6.1 Retreaded tires	79.3%		41.0%	Own assumption	260	P 1
F 6.2 Filter residue	0.7%		14.0%	Wohlleben et al., 2016	5	P 8
F 6.3 Shredding residue	20.0%		41.0%	Own assumption	65	P 8
P 7 Recycling			-		6 300	
F 7.1 Shredding residue	53.6%		5.0%	BMNT, 2019	3 380	P 8
F 7.2 Filter residue	0.7%		14.0%	Wohlleben et al., 2016	40	P 8
F 7.3 Export granulate for new products	45.7%		5.0%	BMNT, 2019	2 880	Output (export)
P 8 WIP					3 690	
F 8.1 Slags and ashes		1.6%	5.0%	Mueller et al., 2013	60	P 10
F 8.2 Export fly ash to underground landfill (Ger)		0.4%	5.0%	Mueller et al., 2013	15	Output (export)
F 8.3 Elimination		98.0%	5.0%	Mueller et al., 2013	3 615	Output (air)
P 9 Further treatment					90	
P 10 Landfill					60	
P 15 Retreaded vehicle tires					260	

Legend

Assumption based on official statistics

Assumptions based on data from the waste management sector without specification for nanomaterials (stochastic assumptions*)

Assumptions based on scientific articles and studies (experimental investigations, authors' assumptions)

Own assumptions

Appendix 4: Mass balance MFA level of “vehicle tires”; performed with e!Sankey.

Name of flow	**"stochastic" transfer coefficients* [%]**	**Uncertainty based on Laner et al., 2015 [%]**	**Reference**	**Calculated quantity [t/a]**	**Internal/external system flows**
Import		14.0%	Statistik Austria, 2018b	29 750	
<u>Total tires stock (summer/winter)</u>		14.0%	Statistik Austria, 2018a	410 640	
In use		14.0%	Statistik Austria, 2018a	264 610	
<u>EOL treatment</u>					
Import EOL tires		5.0%	BMNT, 2019	10 820	
Retreading		5.0%	BMNT, 2019	1 280	Output (retreading)
Recycling		5.0%	BMNT, 2019	11 280	Output (recycling)
WIP		5.0%	BMNT, 2019	13 040	Output (incineration)
Export tires		5.0%	BMNT, 2019	25 390	Output (export)
<u>Use phase</u>				264 610	
Particle release surface water	25.0%	14.0%	Gottschalk et al., 2015	3 880	Output (surface water)
Particle release emissions soil	12.5%	14.0%	Gottschalk et al., 2015	1 940	Output (soil)
Particle release emissions WWTP	9.4%	14.0%	Gottschalk et al., 2015	1 460	Output (WWTP)
Particle release emissions air	53.2%	14.0%	ADAC, 2019; Hillenbrand et al., 2005; Luekewille et al., 2002	8 270	Output (air)
Re-use tires next season		14.0%		249 060	Used again next season
Not in use		14.0%		105 860	Used again next season

<u>Legend</u>

Assumption based on official statistics

Assumptions based on data from the waste management sector without specification for nanomaterials (stochastic assumptions*)

Assumptions based on scientific articles and studies (experimental investigations, authors' assumptions)

Appendix 5: Interview protocol DI Friedrich Eppel, ÖAMTC.

Interview

Herr DI Friedrich Eppel, ÖAMTC - stv. Leiter Technik, Test, Sicherheit; Konsumentenschutz & Mitgliederinteressen

Datum und Uhrzeit: 09.07.2019, 16:00 Uhr

Ort: Baumgasse 129, 1030 Wien

FE = DI Friedrich Eppel; AP = Anna Pavlicek, MSc, Msc; SP = Stefanie Prenner, BSc

FE: Sprechen wir gleich über die Reifenabriebmengen. Bei dem Abriebtest vom ADAC sind unterschiedliche Ergebnisse zu verzeichnen, im Durchschnitt sind es 125 g pro 1 000 km. Aber die Spannweite reicht von 183 bis 73 g.

SP: Sind hier auch unterschiedliche Reifenarten mitberücksichtigt worden, sprich LKW-Reifen, PKW-Reifen?

FE: Nein, das sind alles nur PKW-Reifen, Sommer-, Winterreifen und Ganzjahresreifen. Aber unterschiedliche Dimensionen. Eher sportliche, bei denen man sieht, dass der Abrieb doch deutlich höher ist. Die Normalreifen, wo der Abrieb etwas weniger ist. Aber es ist schon einmal eine interessante Größenordnung, die für Sommer- und Winterreifen gilt. Wobei die Winterreifen im Winter getestet werden und die Sommerreifen im Sommer. Die Reifen werden nicht nur mit Laser vermessen, sie werden auch bei jeder Vermessung, das ist alle 2 500 km, werden sie gewogen.

SP: Ist der Reifenabrieb pro Reifen oder pro Fahrzeug?

FE: Das ist pro Fahrzeug, eines Fahrzeuges, Vorder- und Hinterachse. Zu den Kilometerleistungen haben wir leider keine Daten zur Verfügung, das einzige was man weiß, ist die verbrauchte Kraftstoffmenge. Damit könnte man dann auf die Kilometerleistung schließen, weil man ja weiß, was so der durchschnittliche Kraftstoffverbrauch ist. Da haben wir aber das Problem, dass wir nur Diesel und Benzin haben. Diesel ist ein großer Anteil LKWs dabei, wahrscheinlich zu 3/4. Oder vielleicht 70 % LKW, 30 % PKW. Das heißt, da wirds dann auch schwer, auf die Kilometerleistung zu kommen. Aber bei PKWs sind das so ca. 12 000, 13 000 km ist der Schnitt PKW-Jahreskilometer. Aber die Zahlen sind bekannt. Beim Transit wirds dann schwierig.

SP: Zum Transitverkehr der LKWs gibts vom BMVIT Daten.

FE: Für PKWs wird das BMVIT auch Daten haben und zwar über die Maut. Da werden sie das wissen. Und über die getankte Kraftstoffmenge. Die Zusammensetzung dessen, was hier an Abrieb entsteht, würde uns selbst auch interessieren. Das ist ein bisschen ein Problem, man hört viel, vor allem weil wir ja auch Reifentests mitmachen und im Konsortium da ziemlich wichtig dabei sind, hören wir viel aus der Branche, aber ich habe noch keine belastbare Quelle dafür. Ich habe dazu kurz recherchiert, die ETBMA hat sich damit auch schon beschäftigt, die haben da einiges schon geforscht. Da gibts dann verschiedene Meinungen, ist das überhaupt Mirkoplastik oder wie schnell wird es absorbiert, wie schnell wird es auch wieder umgewandelt. Weil es gibt ja auch sehr viele organische Stoffe im Reifen. Alles was Gummi ist, ist organisch. In jedem Reifen ist zusätzlich noch Black Carbon drinnen, Silica ist wieder eher ein Mineralstoff. Wir beobachten das sehr interessiert und sind dankbar über alles Wissen, das wir bekommen. Aber selber haben wir einen sehr begrenzen Wissensstand dazu, weil wir uns gar nicht damit beschäftigen können, aufgrund unserer Position und unseren Kapazitäten her.

SP: Es gibt dazu momentan auch noch recht wenig Information, wo sich der Abrieb dann ablagert bzw. wo er dann schlussendlich hinkommt.

FE: Ich habe gehört, dass viel eigentlich gleich neben der Straße bleibt. Dass es eben nicht airborne ist, sondern eher schnell in den Boden hineingeht. Dort dann auch bald nicht mehr nachweisbar ist. Aber das sind alles nur Gerüchte, ich habe dazu nichts Belastbares.

SP: Das war für mich auch spannend, ich habe dann noch versucht über den TSP mir die Mengen auszurechnen. Da habe ich dann vom gesamt TSP in Österreich den Verkehrsteil heraus gerechnet und davon dann noch den Anteil der Reifen.

FE: Der Abrieb durch die Bremsen macht da auch extrem viel aus. Das sind auch nicht so angenehme Partikel von der Bremsscheibe, sie sind zwar nicht mehr asbesthaltig, aber es sind halt nicht sehr viel organische Stoffe dabei. Bei dem Reifenabrieb weiß ich nicht, wie lange diese Partikel dann airborne Partikel sind, ob sie bald wieder in den Boden gehen oder ob sie in der Luft bleiben. Es beschäftigen sich alle Hersteller damit, es beschäftigen sich natürlich Umweltschutzorganisationen damit, so sie Kapazitäten dafür haben. Was studieren Sie genau?

SP: Umwelt- und Bioressourcenmanagement, mit Schwerpunkt auf Abfallwirtschaft. Deshalb wird auch ein Fokus der Materialflussanalyse auf den verschiedenen EOL Prozessen liegen.

FE: Michelin hat sich mit diesem Thema immer schon sehr stark beschäftigt, da könnte man vielleicht auch was zum Recycling auf den Seiten von Michelin finden, eher auf den internationalen. Wieder zum Reifenabrieb zurück, ich könnte mir vorstellen, dass bei nasser Fahrbahn, dass da sehr viel gleich in den Boden geht. Der Reifenabrieb ist bei nasser Fahrbahn mehr als bei trockener Fahrbahn. Auch etwas, was man eher nicht denkt. Weil der Schlupf einfach bei der nassen Fahrbahn höher ist. Wir sehen das bei unseren Tests, das sieht man dann sogar wirklich, welche der Zyklen bei Nässe gefahren sind.

SP: Das hätte ich jetzt auch nicht gedacht.

FE: Bei Regen werden sehr viele Partikel sofort in den Boden gewaschen. Die ganze Reifenabriebthematik war lange Zeit, interessanterweise, gar kein Thema. Die ist erst seit kurzer Zeit aufgekommen.

SP: Ja und es ist doch recht viel, mengenmäßig betrachtet.

FE: Wahrscheinlich sind die Mengen, die Schuhsohlen abreiben, mehr. Das darf man nicht unterschätzen, aber zu dem Reifenabrieb gibt es wahrscheinlich a bisserl mehr Informationen. Bei unserem Reifenabriebtest, was ich da sehe, da war sicherlich ein Großteil Golf. Da hat man dann doch eine relativ eingeschränkte Reifendimension. Aber man hat dann einmal eine Größendimension.

SP: Irgendwo muss man Einschränkungen machen, das kann ja dann erwähnt werden. Macht es auch einen Unterschied, ob Sommer oder Winter ist, beim Reifenabrieb?

FE: An wärmeren Tagen nützen sich Winterreifen natürlich mehr ab und umgekehrt, in der Kälte nutzen sich Sommerreifen mehr ab. Wenn man Reifen nicht der Saison adäquat verwendet, ist der Reifenabrieb höher. Es hat immer damit zu tun, der Reifenabrieb ist direkte Folge von Schlupf, also vom Durchrutschen am Boden. Man kann keine Kraft übertragen, ohne dass man durchrutscht. Das geht physikalisch nicht, deswegen gibt es keinen Reifen ohne Abrieb. Es gibt kein Vorwärtskommen ohne Schlupf. Selbst ein Stahlreifen auf der Schiene bei der Eisenbahn schlupft ein bisschen. Es sind zwar ganz andere Größenordnungen, aber ein bisschen doch. Und wenn der Reifen mehr schlupft, dann reibt er mehr ab. Also bei Nässe, das hab ich schon gesagt, aber der Sommerreifen schlupft bei kalten Temperaturen mehr, weil er hart wird und der Winterreifen schlupft bei warmen Temperaturen mehr, weil er zu weich ist. Die Größenordnung liegt jetzt da wahrscheinlich bei 10-20 %, nicht relevant fürs Szenario dann.

SP: Es macht ja auch ein Unterschied, ob es ein LKW-Reifen oder ein PKW-Reifen ist. Kommt es da dann hauptsächlich auf das Gewicht vom Reifen an, dass bei LKW-Reifen dann beispielsweise mehr Reifenabrieb stattfindet wie bei PKW-Reifen?

FE: Gefühlsmäßig würde ich sagen, dass ein LKW-Reifen weniger Abrieb hat bei weniger spezifischem Antrieb. Das hat viele Faktoren, die Aufstandsfläche, die Antriebsleistung, das sind alles Faktoren, die den Abrieb beeinflussen. Ich weiß aber nicht wie groß das beim LKW ist, das kann ich nicht sagen. Gefühlsmäßig ist es ein bisschen weniger. Aber ich hatte mit LKW-Reifen bisher relativ wenig zu tun.

SP: Beim LKW sind es dann natürlich insgesamt mehr, weil ja mehr Reifen als beim Auto vorhanden sind.

FE: Ja, das stimmt. Die Antriebsreifen sind sicherlich ganz anders beim LKW, also die Reifen, die nur so hinten nachlaufen, die nur die Last tragen. Der Antriebsreifen hat dann wahrscheinlich zehnmal so viel Abrieb wie alles andere. Beim PKW, in dem Fall ist es einfach, das sind Frontantriebsfahrzeuge gewesen, die haben vorne den Antrieb, die haben vorne die Lenkung. Das heißt, da ist sicher der Abrieb vorne am höchsten. Beim LKW ist das kompliziert, da kommt es dann darauf an, wie viel Führungsaufgabe müssen die nicht betriebenen Reifen übernehmen. Da kommt dann der Verschleiß nur über die Führungsaufgaben und über das Bremsen wird natürlich auch verschleißt. Das ist dann wieder bei allen Rädern beim LKW.

SP: Es gibt auch schon unterschiedliche Studien, die unterschiedlichen Reifenabrieb angeben, je nachdem ob man jetzt auf einer Landstraße fährt oder auf einer Autobahn.

FE: Naja, wenn ich jetzt gerade auf der Autobahn fahre, egal bei welchem Tempo, habe ich einen sehr niedrigen Verschleiß. Es können auch 130 sein, die Kraft, um ein Fahrzeug auf der Geschwindigkeit zu halten, ist sehr gering. Die notwendige Leistung und damit Kraft. Beschleunigen, bremsen ist natürlich ganz was anderes, daher kann man sagen, Landstraßen, wo es kurvig ist, wo man immer wieder bremsen und beschleunigen muss, da ist der Verschleiß natürlich wesentlich höher wie auf der Autobahn, weil ich da ja mehr oder weniger nur dahin rolle. Und da ist 130 immer noch ein Rollen für den Reifen. Das ist unsere Erfahrung, die wir aus den Verschleißversuchen haben, weil wir eben auch Verschleiß auf einem Prüfstand machen, wo auf dem Prüfstand genau das Fahrprofil nachgefahren wird. Eben auf der gleichen Prüfstrecke, wo dann auf der Straße gefahren wird und wo halt dann, um das Ganze zu komprimieren, Teile mit kaum Verschleiß oder mit wenig Verschleiß einfach rausgenommen werden. Und das sind die geradeaus Fahrteile, auch auf der Autobahn. So wurde mir das erzählt. Wenn ich mit 200 auf der Autobahn fahre, ist das was anderes. Je höher die Leistung, die notwendig ist, um das Fahrzeug auf der Geschwindigkeit zu halten, desto höher ist der Verschleiß. Und bei 200 ist natürlich der Fahrtwiderstand, der Luftwiderstand oder der Rollwiderstand so hoch, dass ich natürlich mehr Leistung brauche. Aber bei 100 ist eine relativ geringe Leistung notwendig, um es zu halten. Nicht aber, um dorthin zu beschleunigen, das ist natürlich etwas anderes.

SP: In der Literatur, da war eben einmal angeführt, dass auf der Autobahn mehr Verschleiß stattfindet oder mehr Reifenabrieb, aufgrund dessen, wenn man wieder schnell runterbremsen muss.

FE: Klar, das war jetzt rein Konvoi-Versuch gedacht, wo man diese Dinge ja vermeidet. Da muss man halt auf die Quelle schauen, ob sie angeben, wie sie zu den Daten gekommen sind.

SP: Das war dann eben noch im Vergleich gestellt zum Abrieb Innerorts, wo ganz gemütlich geradeaus gefahren wird, ohne Kurven.

FE: Je mehr Leistung abgerufen werden muss, desto mehr ist der Verschleiß. Ganz einfach.

SP: Ich habe bei meiner Arbeit nur den Kunststoffanteil der Reifen berücksichtigt. Wissen sie wie viel Anteil Kunststoff ein Reifen durchschnittlich hat?

FE: Da kommt es darauf an, was alles als Kunststoff kategorisiert wird.

Herr DI Eppel holt ein Modell eines Reifens.

FE: Der Kunststoff ist das, das ist entweder Reyon oder Polyester. Diese drei Lagen sind Stahl und da ist auch noch ein Kunststoff drüber auf der Seite. Das ist meiner Meinung nach der hauptsächliche Anteil an Kunststoff im Reifen. Wie weit jetzt wirklich da in der Mischung etwas drinnen ist, was man als Kunststoff bezeichnen kann, das weiß ich nicht.

SP: Aber die Mischung ist dann schon aus Kautschuk, oder?

FE: Ja, das ist im Prinzip Kautschuk plus Carbon Black plus Silica plus Schwefel und andere Additive. Möglicherweise gibt es dazu auf der Homepage von Continental, unter “Know How” oder so Informationen dazu.

SP: Ich hab aus einer wissenschaftlichen Quelle einen Wert herausgenommen, der angibt, dass 65 Gewichtsprozent Kunststoff sind. Hier ist Kautschuk als Kunststoff miteinberechnet.

FE: Ja, das kann schon hinkommen. Im PKW-Reifen ist nur mehr sehr wenig Naturkautschuk drinnen, da ist sehr viel künstlicher Kautschuk drinnen. Im LKW-Reifen ist der Anteil von Naturkautschuk noch sehr viel höher.

SP: Warum?

FE: Da geht es um Rollwiderstand und solche Sachen. Wenn man den Synthesekautschuk als Kunststoff sieht, dann ist natürlich schon einiges drinnen, ganz klar.

SP: Im Rahmen meiner Masterarbeit habe ich das ebenso klassiert, dass Synthesekautschuk als Kunststoff gilt. Zu den Carbon Blacks noch: in der Literatur sind dazu eben Werte angeben, wie viel Gewichtsprozent im Kunststoffanteil enthalten ist. Da schwanken die Werte zwischen 20 und 40 Gewichtsprozent. Haben Sie da auch Informationen dazu?

FE: Nein, aber das scheint mir vernünftig.

SP: Eine generelle Frage zu den Reifen noch, eine Ersatzreifen-Pflicht gibt es nicht mehr für PKWs oder gibt es die noch?

FE: Nein, die hat es nie gegeben.

SP: Achso, das wusste ich nicht. Das wäre bei unserer Betrachtung ansonsten spannend gewesen, weil wir von einem Bestand ausgehen, in den wir dann die Reservereifen miteingerechnet hätten, aber auch Winter- und Sommerreifen.

FE: Es werden auch immer weniger. Man sieht es bei den Neuwägen, die haben kaum mehr Reservereifen dabei. Entweder eine Dichtung oder so etwas, maximal das. Es ist jetzt nicht Null, auch beim Neuwagen nicht Null. Oft Spare Tires, also kleinere. So der vollwertige Ersatzreifen gibt es kaum mehr. Es wäre der einzige Bereich des Autos, wo man einen Ersatz dabei hat, vielleicht noch das Lämpchen. Ich hab keine Ersatzkeilriemen mit und so weiter. Aber das ist Tradition, da gibt es keine gesetzliche Verpflichtung dazu.

SP: Ich kann Ihnen dazu meine bisher erstellte Materialflussanalyse zeigen.

Betrachtung der bereits erstellten Materialflussanalyse.

SP: Für den Bestand haben wir Winter- sowie Sommerreifen bei PKWs und LKWs mitberücksichtigt.

FE: Bei LKWs wird das nicht unbedingt notwendig sein. LKWs fahren im Prinzip durch mit den gleichen Reifen.

SP: Sind das Allround-Reifen bei LKWs?

FE: Es wird immer mehr, dass es auch Winterreifen für LKWs gibt in den letzten Jahren. Meistens geben sie aber an der vorderen Achse im Herbst neue Reifen drauf und bis zum nächsten Herbst sind die wieder herunten und man kriegt wieder neue Reifen drauf. Da muss man aber schauen, also jetzt den normalen Wechsel wie bei PKW würde ich beim LKW jetzt nicht ansetzen.

SP: Okay. Und von diesem Bestand geht dann ein Teil in die Nutzungsphase. Von dort aus gehen dann verschiedene Pfade in die Umwelt. Der Abrieb findet sich im Boden, im Oberflächengewässer oder in der Kanalisation wieder. Vom Bestand aus geht dann wiederum jedes Jahr ein Teil in die verschiedenen End-of-life Prozesse. Ein Teil der Reifen wird runderneuert.

FE: Runderneuerung betrifft nur LKWs. Vielleicht in sehr unbedeutenden Mengen PKWs, das ist nicht mehr wirtschaftlich. Bei LKWs aber schon.

SP: Gut zu wissen. Ein Teil geht dann weiter in die Verbrennung und ein Teil ins Recycling. Am Schluss dann von der Verbrennung auf die Deponie.

AP: Mich würde noch interessieren, wir sind eigentlich über Frau Kral zu diesem Interview gekommen, sie hat uns gesagt, dass bei Elektroautos ein höherer Reifenabrieb zu erwarten ist. Gibt es dazu schon Erkenntnisse?

FE: Nein. Elektroautos sind drehmomentstärker und schwerer, das ist das Problem. Das heißt, wenn ich ein Elektroauto immer mit Freude am Fahren bewege und die schöne Schubkraft des Elektroautos immer ausnutze, werde ich mehr Reifenabrieb haben. Wenn ich ein Elektroauto gleich fahre wie ein anderes Auto gleichen Gewichts, sollte physikalisch nichts anderes herauskommen. Natürlich wird das Thema Elektroauto in der Reifenbranche auch so behandelt, die haben mehr Abrieb, weil sie drehmomentstärker sind, ja. Wenn der Fahrer oder die Fahrerin das Drehmoment abruft, wird der Reifen mehr Abrieb haben, das ist einfach so. Wir werden mit einem Benzinmotor schon gar nicht, mit einem Dieselmotor schwer, dieses Drehmoment auf die Straße bringen, was so ein Elektroauto haben kann. Wenn ich ganz zart mit so einem Elektroauto fahre, dann habe ich nicht mehr Abrieb, das ist so. Aber meistens will man ja die Leistung dann ausnutzen. Ja, aber so generell ist das nicht der Fall, dass Elektroautos mehr Abrieb haben, man kann sie so nutzen, dass sie mehr Abrieb haben oder man nutzt sie vielleicht anfänglich so, dass sie mehr Abrieb haben. Das hat aber dann auch mehr Stromverbrauch.

SP: Aber das wäre ja dann bei den normalen PKWs auch so, wenn ich auf der Autobahn immer extrem beschleunige, dann werde ich auch mehr Abrieb haben.

FE: Genau, wenn ich immer einen Kavalierstart mache, habe ich natürlich einen höheren Abrieb. Und Elektroautos haben ein höheres Gewicht, das muss man schon sagen, sie haben für die jeweilige Klasse deutlich mehr Gewicht. Das kann gleich bis zu 200 kg mehr sein, das bewirkt natürlich auch wieder mehr Abrieb. Dann brauche ich wieder mehr Kraft um das Auto zu beschleunigen und wieder abzubremsen. Es gibt ja auch noch nicht so viele Elektroautos. Es wird wahrscheinlich prozentuell nicht so viel Unterschied machen, wenn man das mitberücksichtigt.

SP: Wie wird sich zukünftig die Reifenzusammensetzung ändern, wird sich da was ändern?

FE: Wir warten alle auf das Wunder, so ein Wunder, was wir bei Silica hatten. Da war wirklich ein Quantensprung im Verhältnis Rollwiderstand zu Nassbremsverhalten. Das war die Silica-Mischung, die Michelin erfunden hat. Das ist jetzt aber auch schon 20-30 Jahre her. Und das nächste Wunder gibt es derzeit noch nicht. Es wäre das nächste Wunder notwendig, um den Rollwiderstand runterzubringen und die Nasshaftung trotzdem auf gutem Niveau zu halten, ich sehe es bis jetzt nicht. Es kommen so Dinge wie Löwenzahn statt Kautschuk, das ist sicherlich auch eine Weiterentwicklung. Das nächste wäre der luftlose Reifen. Das wird glaub ich noch ein bisschen dauern.

SP: Vielen Dank, dass Sie sich Zeit genommen haben.