Environmental Pollution 290 (2021) 118102

Contents lists available at ScienceDirect

Environmental Pollution

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

Static modelling of the material flows of micro- and nanoplastic particles caused by the use of vehicle tyres☆

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ARTICLE INFO

Keywords:
Tyre wear particles
Non-exhaust gas emissions
Microplastics
Nanoplastics
Carbon black

ABSTRACT

The emissions of tyre wear particles (TWPs) into the environment are increasing and have negative impacts on the environment and human health. The aim of this study was therefore to establish a mass balance for vehicle tyres und TWP emissions in Austria using static material flow analysis, which enabled a quantification of mass flows of rubber including carbon black as the most mass-relevant tyre filler. Vehicle-specific and mileage-dependent emission factors were used to calculate the TWP emissions. The results for the year 2018 indicate that 80% of the tyre rubber remained in use, while 14% was re-treaded, recycled, incinerated or exported as end-of-life tyres and 6% was emitted as TWPs to air, soil or surface water. Of these 21,200 t/y released and dissipative lost TWPs, 6% were microscale, with a possible size between 0.1 and 10 μm, and 0.3% were nanoscale below 0.1 μm. The mass balance on the substance level shows that the TWPs contained 5,500 t/y of carbon black emitted in the form of airborne TWPs (6%) or entering in the soil or surface waters (47% each). Regarding air pollution from road vehicles, about 3,600 t/y were non-exhaust emissions, including tyre, brake and road-surface wear, which contributed to 9% of total dust emissions across Austria. Scenario analysis for 2050 with regard to e-mobility and the European Green Deal reveals that non-exhaust emissions can only be significantly reduced by a general reduction of the mileage or an environmentally friendly tyre design. This modelling approach provides a solid basis for decision makers in traffic planning as well as for chemical risk assessment. However, dynamic models with higher temporal and spatial resolution are needed to predict future mass flows of TWPs and their environmental fate, including their degradation products and possible accumulation effects.

1. Introduction

Different studies clearly show the significance of tyres as a plastic polluter and in short: microplastics, which are typically smaller than 5 mm, have been found all over the environment including deep-sea sediments of the Arctic (Evangeliou et al., 2020; National Geographic, 2019; Tekman et al., 2020). Tyre and road wear particles are recognized microplastic materials (Unice et al., 2019), although elastomeric materials are not considered to be plastics according to international standards (ISO 472, 2013). Nevertheless, the general definition of microplastics or the classification of abraded tyre rubber is still the subject of scientific discussion (Hartmann et al., 2019). Sommer et al. (2018) highlighted that microplastics from tyres are omnipresent in the environment. Estimations indicate that released tyre wear particles (TWPs) contribute up to 5–10% of plastics subsiding in the oceans (Kole et al., 2017).

Sufficient friction between tyres and road surfaces is required to keep vehicles operating safely on the roads. This leads to high shear stress in the tyres and consequently to the diffuse release of large quantities and differently sized TWPs into the air, soil or surface waters (Kole et al., 2017). In general, the total amount of released tyre wear (TWP_{total}) is the sum of TWP_{air}, TWP_{soil} and TWP_{water}. Various parameters influence the released quantity, including driving style, engine power, vehicle weight, loads, tyre positions, materials properties and age of tyres, road conditions and weather (Boulter, 2006; Hillenbrand et al., 2005; Ntziachristos and Boulter, 2019). Ntziachristos and Boulter (2019) summarized that

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☆ This paper has been recommended for acceptance by Eddy Y. Zeng.
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https://doi.org/10.1016/j.envpol.2021.118102
Received 22 April 2021; Received in revised form 31 August 2021; Accepted 2 September 2021

over the lifetime of a single tyre the TWP release can range from 10 kg for trucks or busses to 1–1.5 kg for passenger cars (PC) and even several hundred grams for two-wheel-vehicles (TWV). For example, a PC tyre of 8 kg is used on average between 50,000–60,000 km and loses within its approximately 3 year lifetime up to one fifth of its mass (Boulter, 2006). Literature data show that the annual emissions of TWPs range from 0.2 to 5.5 kg/capita – the lowest value applies to India and the highest to USA. The emission rate depends mainly on the car density (Baensch-Baltruschat et al., 2020; Kole et al., 2017). A study of the German Federal Highway Research Institute estimated that Germany emits about 1.38 kg/capita-year TWPs (BASt, 2010). Most of these TWPs are deposits and about 7% are airborne dust, each consisting mainly of rubber and polymer components (BASt, 2010). In general, larger particles are deposited in the soil via road runoff or transferred into surface waters via rainwater run-off or effluents from wastewater treatment plants. The environmental distribution and fate of TWPs depend on the road type (highways, urban or rural roads), the sewage systems and technologies for waste water and sewage sludge treatment (Baensch-Baltruschat et al., 2021). During these processes polymeric tyre components and additives may be released (Wagner et al., 2018). Using dynamic probabilistic material flow analysis (MFA), Sieber et al. (2020) showed that in Switzerland 0.96 kg/capita-year of TWPs enter the environment, 74% of which are deposited along roads, 22% enter surface waters, and 4% accumulate in soils. With the assumptions that tyre rubber is persistent over time and thus hardly degrades, about 219,000 t of TWPs have accumulated in Switzerland since 1988 (Sieber et al., 2020).

Since the majority of TWPs ends up in soil or surface waters, rubber particles as well as additives can accumulate in the human food chain (Hüffer et al., 2019; Kole et al., 2017; Sieber et al., 2020). Additionally, TWPs can serve as vectors for associated inorganic or organic co-pollutants (e.g., heavy metals) which can be adsorbed onto the particle surface (Hüffer et al., 2019). Owing to gaps in data, however, the topic of magnification of TWPs and their additives in the food chain as well as sorption properties need further research (Hüffer et al., 2019; Kole et al., 2017). Regarding effects on human health, it is proposed that non-exhaust airborne traffic-related emissions, including airborne TWPs, pose the highest risks (Amato et al., 2014). However, studies on rats indicate only a low likelihood of health risks from inhalation of tyre and road wear particles (Kreider et al., 2012), while in vitro toxicity studies focusing on organic compounds and nanoparticulate debris from tyre and brake wear showed an increase in lung cell toxicity and genotoxicity in peripheral blood lymphocytes (Gualtieri et al., 2008; Kazimirova et al., 2016). A further study on the particle transfer of carbon black (CB) – commonly used as tyre filler – showed that CB can translocate from the mother's lung to the placenta leading to detrimental health effects for the foetus (Bové et al., 2019). Recent reviews concluded that knowledge gaps need to be closed and monitoring or modelling studies are required to estimate the transport of TWPs in rivers and soils (Baensch-Baltruschat et al., 2020; Kole et al., 2017). Moreover, a review by Pant and Harrison (2013) highlighted that quantitative information on exposure is still needed, particularly for research on the contribution of non-exhaust emissions to the total mass of airborne dust.

This study has the objective to provide a sound database on the amount of TWPs released in the year 2018 in Austria including quantitative information on the particle size. The objectives were achieved by the following approach: (i) create a comprehensive static material flow analysis (MFA) at the level of goods (rubber) and substances (CB), (ii) predict the material flows of TWPs in the environment (iii) determine the contribution of TWPs to microplastic pollution as well as of non-exhaust emissions to the quantity of total suspended particles (TSP) in Austria in 2018. Subsequently, scenarios were developed to predict the quantity of TWPs for the year 2050.

2. Material and methods

2.1. Material flow analysis for vehicle tyres

MFA was applied to quantitatively analyse and display the flows and stocks in a system defined in space and time providing an understanding of the functioning of the whole system. Following terms are used based on Brunner and Rechberger (2004):

- *Substances* are any (chemical) element or compound that are composed of uniform units.
- *Goods* are any economic entity of matter with an economic value and are made up of one or several substances.
- *Material* serves as an umbrella term for both substances and goods.
- *Transfer coefficients* describe the partitioning of a substance in a process (for diffuse release into the air the term emission factor (EF) is used)

The main focus for MFA is to establish a mass balance and to provide a complete database at the level of goods as well as at the level of substances. The MFA at the level of goods is instrumental for understanding the overall functioning of the system; MFA at the level of substances is essential to assess aspects regarding the quality of material flows, such as resource flows or emissions into the environment. Combined, these serve to evaluate the transformation, transport, and storage of valuable and hazardous substances, and thus form the base for identifying both resource potentials and risks for human health and the environment (Allesch and Brunner, 2015). Additional fate modelling was not conducted in this study but would be further needed to assess the final fate of non-/degradable rubber particles and their transformation products. It must be mentioned that fate models for microplastics (e.g. Besseling et al., 2017; Nizzetto et al., 2016; Siegfried et al., 2017; Unice et al., 2019) have focused on terrestrial transport in soil, water (rivers), and sediment but further developments on regional air dispersion models are needed. The system boundaries for this MFA are vehicle tyres in Austria in the year 2018 and are summarized in Fig. 1. A static MFA was carried out since dynamic modelling was not possible due to the lack of data on market dynamics (e.g. missing use-phase statistics for life-time-distribution of different vehicle tyres). The software tool "e! Sankey" (IFU Hamburg) was used to perform the MFA on the level of goods (rubber of vehicle tyres) and the software tool "STAN" (TU Wien) was used to display the substance flows of the tyre filler CB. The calculation algorithm of "STAN" uses mathematical methods such as data reconciliation, error propagation and gross error detection to reconcile conflicting data (with data reconciliation) and subsequently to compute unknown variables including their uncertainties (with error propagation) (Cencic, 2016). Since MFA models are usually subject to uncertainties, data uncertainties have been calculated and estimated based on the proposed approach "expert estimations" by Laner et al. (2015) (*see SI* section 1). Especially due to frequent data scarcity, expert estimation is an important source of MFA input data, but still needs to be addressed by uncertainty evaluation.

Traffic statistics were used to estimate the consumption rates of vehicle tyres in Austria during the use-phase (see SI section 2) and waste statistics (BMNT, 2019) were used to calculate the end-of-life phase (EOL) (see SI section 3). Since no tyre production and manufacturing processes take place in Austria, all tyres are assumed as imported. Import statistics on new as well as EOL-tyres were used as MFA input data. All processes and flows from the MFA on vehicle tyres in Austria in the year 2018 and their transfer coefficients on the level of goods and substances are shown in detail in the SI (see section 7). Exported re-granulates and EOL-tyres as well as the calculated amounts of TWPs diffusely entering the air, terrestrial soil or water are represented in the MFA as output data. The quantities of TWPs were calculated based on vehicle-specific emissions factors (EF) and driven mileage (see Table 1 and Table 2). The contribution of non-exhaust emissions (TWP_{air}) to the

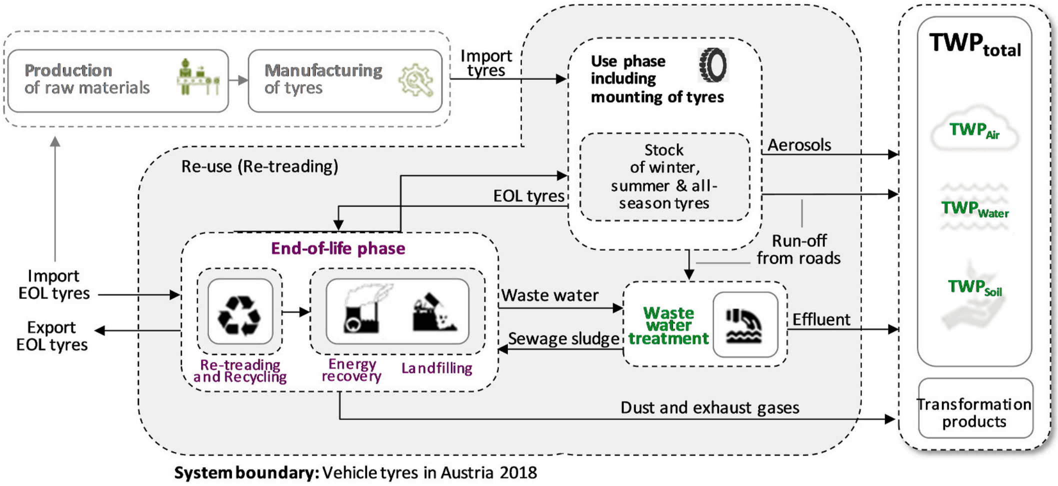

Fig. 1. Illustration of the life cycle of vehicle tyres (grey area = MFA system boundary). White areas are outside of the system boundaries and would require emission-related data during production and manufacturing outside from Austria as well as additional fate models to consider the degradation of tyre wear particles (TWP) and their transformation products.

Table 1
Summary of used vehicle-specific non-exhaust emission factors (EF).

Tyre Wear									
Vehicle-specific EFs[a]	TWP_{total} [g/km][b]	TWP_{air}					TWP_{soil} and TWP_{water}		
		TSP [g/km][c]	PM_{10} [g/km][c]	$PM_{2.5}$ [g/km][c]	PM_1 [g/km][c]	$PM_{0.1}$ [g/km][c]	Micro-particles (<5µm)[d]	Micro-particles (5-1µm)[d]	free Nano-fillers (<100nm)[d]
TWV	0.0625	0.0046	0.0028	0.0019	0.0003	0.0002	1.6% of total TWPs on rainy days and 4.0% on dry days	0.2% of total TWPs on rainy days and 1.7% on dry days	0.045% of TWPs
PC	0.1250	0.0107	0.0064	0.0045	0.0006	0.0005			
LDV [e]	0.7000	0.0169	0.0101	0.0071	0.0010	0.0008			
HDV	1.2000	0.0563	0.0338	0.0236	0.0034	0.0027			
Busses	0.7000	0.0169	0.0101	0.0071	0.0010	0.0008			
PC (BEV 100) [f]	0.1733	0.0111	0.0067	0.0047	0.0007	0.0005			
PC (BEV 300) [f]	0.1295	0.0148	0.0089	0.0062	0.0009	0.0007			
LDV (BEV 100) [f]	1.0355	0.0179	0.0108	0.0075	0.0011	0.0009			
LDV (BEV 300) [f]	0.7421	0.0250	0.0150	0.0105	0.0015	0.0012			

Vehicle brake wear						Road-surface wear		
Vehicle-specific EFs[a]	*TWP_{air}*					*TWP_{air}*		
	TSP [g/km][c]	PM_{10} [g/km][c]	$PM_{2.5}$ [g/km][c]	PM_1 [g/km][c]	$PM_{0.1}$ [g/km][c]	TSP [g/km][c]	PM_{10} [g/km][c]	$PM_{2.5}$ [g/km][c]
TWV	0.0037	0.0036	0.0014	0.0004	0.0003	0.0060	0.0030	0.0016
PC	0.0075	0.0074	0.0029	0.0008	0.0006	0.0150	0.0075	0.0041
LDV [e]	0.0117	0.0115	0.0046	0.0012	0.0009	0.0150	0.0075	0.0041
HDV	0.0328	0.0321	0.0128	0.0033	0.0026	0.0760	0.0380	0.0205
Busses	0.0117	0.0115	0.0046	0.0012	0.0009	0.0150	0.0075	0.0041
PC (BEV 100) [f]	0.0019	0.0019	0.0008	0.0002	0.0002	0.0150	0.0075	0.0041
PC (BEV 300) [f]	0.0027	0.0026	0.0010	0.0003	0.0002	0.0150	0.0075	0.0041
LDV (BEV 100) [f]	0.0031	0.0030	0.0012	0.0003	0.0002	0.0150	0.0075	0.0041
LDV (BEV 300) [f]	0.0045	0.0044	0.0018	0.0004	0.0004	0.0150	0.0075	0.0041

Abbreviations: BEV (battery electric vehicle), HDV (heavy-duty vehicles), LDV (light-duty vehicles), PC (passenger cars), TWV (two-wheel vehicles).

[a] The classification of vehicle types is based on registered vehicles in Austria.

[b] Taken from Hillenbrand et al. (2005) and ADAC (2019).

[c] EFs for TWP_{air} are based on the mass fractions of total suspended particles (TSP) to PM_{10}, $PM_{2.5}$, PM_1 and $PM_{0.1}$ according to Ntziachristos and Boulter (2019) and are derived from studies conducted on dry days.

[d] Factors or transfer coefficients for TWP_{soil} and TWP_{water} are taken from Wohlleben et al. (2016).

[e] Includes trucks “class N3” (<3.5t) and “class N2” (>3.5t <12t).

[f] EFs for BEVs are taken from OECD (2015, 2020).

total TSPs in Austria was determined based on government reports (see SI section 6). For the present study, EFs of mass fraction and particle size classes for airborne TWPs were included in the MFA – i.e., TWP_{air} was calculated based on EFs for TSP and particulate matters (PM_{10}, $PM_{2.5}$, PM_1, and $PM_{0.1}$). Additionally, size-dependent data based on an experimental lab-scale study (Wohlleben et al., 2016) was also included to calculate the flows of TWP_{soil} and TWP_{water} (see SI section 5).

Table 2
Data for Scenario analysis: Total mileage per year. The scenario "with-existing-measures" (WEM) was compared with the "European Green Deal" (GD) scenario projected to 2050.

Vehicle types	Mileage [10^6 km/year]		
	Status quo (2018)	WEM 2050 *(BEV100-scenario) (BEV300-scenario)*	GD 2050 *(BEV100-scenario) (BEV300-scenario)*
PC	68,200	53,600	0
PC (BEV)	300	39,400	93,000
TWV	1700	2500	2500
HDV	5500	7100	1800
Busses	500	600	600
LDV	7800	9500	0
LDV (BEV)	20	0	9500
Total	**84,000**	**112,700**	**107,400**

Abbreviations*: BEV (battery electric vehicle), HDV (heavy-duty vehicles), LDV (light-duty vehicles), PC (passenger cars), TWV (two-wheel vehicles).*

2.2. Data for modelling flows of tyre wear particles

Vehicle tyres are manufactured from elastomers like natural rubbers, polyisobutylene, styrene butadiene rubber, polyurethane etc. (Baensch-Baltruschat et al., 2020; Paul and Robeson, 2008; Wohlleben et al., 2016). Chemical analyses show that the composition of the TWPs can be heterogeneous including fillers (e.g. CB) and agents (mostly mineral oils and Zinc) (BASt, 2010; Kreider et al., 2010). The average plastic content in car tyres is about 85%, including fillers (e.g., CB, silica or chalk), elastomers, plasticisers (oils and resins), vulcanisers, UV stabilisers and other additives. The remaining 15% consists of textile fabrics and steel wires (Continental Reifen Deutschland GmbH, 2013; OECD, 2005). CB in particular is produced in large quantities – about 1, 700,000 t/y in Europe alone – of which about 65% is used for vehicle tyres (European Commission, 2007). The CB content in typical tyres can be up to 45% w/w (BASt, 2010). However, the composition of tyres depends on the vehicle type and it changes by season (summer and winter tyres). Since the mid-nineties, the CB content is decreasing, whereas annealing residues have been strongly increasing due to more frequent use of other fillers like silica (BASt, 2010). This study applied a 22% mass concentration of CB for the whole tyre as the most recent estimated value found by database research. Since only the plastic content of the tyre was considered, the CB content corresponds to about 26% (Continental Reifen Deutschland GmbH, 2013; OECD, 2005). Additionally, other emerging pollutants or dust (e.g., metallic wear from brakes or pigments from road markings) are adsorbed onto the surface of TWPs. It must be noted that this additional mass of adsorbed substances was not considered to calculate the TWP quantity for this study. It is estimated that non-exhaust traffic emissions of medium sized cars consist of approximately 45% road wear, 32% tyre wear and 23% brake wear (Ntziachristos and Boulter, 2019). A literature review (Wagner et al., 2018) shows that the share of TWPs of non-exhaust emissions from traffic is 5–30%. Other studies on aerosols or deposited dust show that the size of TWPs was found to be between 10 nm and several 100 μm (from $PM_{0.1}$ to $PM_{>10}$) (Aatmeeyata et al., 2009; Dahl et al., 2006; Kreider et al., 2010; Panko et al., 2013):

- $PM_{10\text{-}80}$: Analysis of $PM_{10\text{-}80}$ collected samples next to German motorways and highways revealed that the majority of collected aerosol particles (ca. 54% by volume) could be traced back to tyre wear (Sommer et al., 2018). The rubber from tyre wear was roundish or kidney shaped and contained traces of C, Si, Zn which are typical elements in fillers and vulcanization agents for tyres (i.e., CB, silica, or zinc oxide) (Sommer et al., 2018). Contribution of tyre wear to PM_{10} accounts for up to approx. 11 mass-% (Baensch-Baltruschat et al., 2020).
- $PM_{2.5\text{-}10}$: Pant and Harrison (2013) summarized that TWPs are predominantly coarse particles ($PM_{2.5\text{-}10}$), which are generated by shear forces and in turn are influenced by the road-surfaces, tyre types and driving conditions.
- $PM_{1\text{-}2.5}$: Aerosol measurements at the road-tyre interface showed that nanoscale emissions between 5 and 700 nm are generated, which depend on speed and tyre type (Dahl et al., 2006; Foitzik et al., 2018; Gustafsson et al., 2008; Mathissen et al., 2011). Other measurements in an urban street area showed that over 99% of the particle concentrations were found in the size range of 10–300 nm, whereas the particle mass concentration was almost equally distributed between PM_1 and $PM_{2.5}$.
- $PM_{0.1}$: Foitzik et al. (2018) highlighted that ultrafine particulate matter ($PM_{0.1}$) must not be neglected.

Kole et al. (2017) summarize that $PM_{>10}$ and $PM_{2.5}$ mainly contribute to the TSP, which deposit on the ground over time and thus lead to microplastic pollution. Concentrations of TWPs are estimated to range from micrograms per litre in surface water to milligrams per kilogram in sediments, suggesting that sediments act as a sink for TWPs (Wagner et al., 2018). EFs, which are derived from aerosol measurements and already published data, allow TWP_{total} and TWP_{air} to be quantified by multiplying with the annual mileage of each vehicle type (Ntziachristos and Boulter, 2019). Table 1 summarizes the used EFs. The calculated quantities of TWP_{total} and mass-related size distributions for TWP_{air} (given as TSP, PM_{10}, $PM_{2.5}$, PM_1, and $PM_{0.1}$) are shown in the SI (*see* section S6). The vehicle-specific EF for TWP_{total} is based on Hillenbrand et al. (2005). They summarized that the EF for heavy-duty vehicles (HDV) is about 9.6 times higher than for PCs. However, the EF used for PCs is based on a more recent report from the German automobile club ADAC (2019) where more than 120 summer, winter and all-season tyres were tested under real driving conditions. For this test, each tyre was driven for 15,000 km and every 2500 km a measurement of the tread depth and weight loss was carried out using a laser measuring device and digital scales. More than one million data points were evaluated during the tread depth measurements which indicates a high reliability of EF data. Among all tyre types, these tests indicated that the total mass loss (TWP_{total}) was between 73 and 183 mg/km and on average 125 mg/km (ADAC, 2019). TWP_{air} is typically given as TSP which refers to the mass of the total dust (without re-suspended tyre abrasion). The report by Ntziachristos and Boulter (2019) used in this study is updated every year and allows a reliable calculation of the total air emissions. A recent study from German roads (Baensch-Baltruschat et al., 2021) showed that 5% of the total emissions are airborne particles while, 66–76% are "non-airborne" particles transported to road banks and soils including runoff and drift and 12–20% are released to surface waters. In the case of Austria, data on the type of road (highways, urban or rural roads) and resulting road-specific EFs is missing. Therefore, simplifications were needed: based on a life cycle inventory (Simons, 2016) and literature review (Wagner et al., 2018), it was assumed that half of the TWPs (not emitted into the air) is deposited in soils and the other half in water and thus the ratio of TWP_{soil} and TWP_{water} is 1 to 1. Using EFs from Wohlleben et al. (2016) to calculate TWP_{soil} and TWP_{water}, it was also possible to obtain different size classes that mainly depend on weather conditions (rainy or dry days). In general, weathering as a degradation mechanism, which includes mechanical stress, photochemical degradation and hydrolysis (e.g. by rainwater), is seen as an influential factor (Duncan, 2015; Froggett et al., 2014; Koivisto et al., 2017; Noonan et al., 2014). Wohlleben et al. (2016) estimated that on rainy days about 1.6% and on dry days about 4.0% w/w of TWPs below 5 μm (related to the total solids) are emitted directly or time-delayed due to wear ageing. Of these particles, about 0.045% are free nanoparticles (≤100 nm). A discussion about the definition and classification of microplastics can also be found in the SI (*see* section S5) or in Hartmann et al. (2019).

The TWP_{air}, shown in Table 1, indicate that both lighter weight and heavier weight battery electric vehicles (BEVs) emit less PM_{10} than "internal combustion engine vehicles" (ICEV). The reason for this are the lower values for brake and dust wear, although PM_{10} tyre wear is still higher for BEVs. However, for $PM_{2.5}$ this is not the case. BEVs with a longer range, thus a higher weight, emit more $PM_{2.5}$ than lighter ICEVs (OECD, 2020). It must be emphasized that there is currently a lack of measurement data on the EFs for BEVs and therefore more measurement campaigns would be necessary to subsequently reduce the uncertainties in emission models.

2.3. Scenario analysis

Scenario analysis is used to calculate two projections related to TWPs for the year 2050. One projection is based on a scenario called "with-existing-measures" (WEM). WEM includes all policies and measures implemented by 2018. The status quo and current implementation of the measures have been defined at expert level in consultation with the responsible Austrian authorities (Umweltbundesamt, 2019). The second projection is based on the strategies from the European Green Deal to put European transport on track for future sustainable and smart mobility (European Commission, 2020), which was named "green-deal" scenario (GD). Within the GD scenario it is estimated that all PCs and light-duty vehicles (LDV) will use electric drive and that the HDVs will be reduced by 75% due to shifting of road freight to rail. The applied mileages for the status quo and the two scenarios are summarized in Table 2. Within the scenario analysis both, BEVs with a 100-mile range (BEV100-scenario) and a 300-mile range (BEV300-scenario), are considered. It is stressed that BEVs with a longer range are heavier due to the weight of the extra battery capacity.

3. Results and discussion

3.1. MFA on vehicle tyres

3.1.1. Goods - MFA on rubber tyres

The MFA at the level of goods (see Fig. 2) shows that in Austria 28,600 t/y new vehicle tyres were imported. Based on the number of registered vehicles, about 349,000 t/y of vehicle tyres were in stock. Taking summer and winter tyres into account, about 211,000 t/y were actually driven. With regard to the EOL-phase, this MFA shows that about 51,000 t of EOL-tyres were discharged and treated, of which 40,000 t/y EOL-tyres were generated in Austria and an additional 11,000 t/y were imported. In Austria, EOL-tyres are either retreaded for re-use or mechanically pre-treated (shredded) to produce refuse-derived-fuels (RDFs) for energy recovery or re-granulates for recycling. Thus ca. 1,300 t/y of retreaded tyres were placed back in the Austrian market, ca. 13,000 t/y were used for co-combustion as RDFs and ca. 11,000 t/y were recycled and exported as secondary raw material for further use. Based on the calculated quantity of TWP_{total}, the tyres lost 10% of their mass during use (use-phase: 211,000 t/y). This result is consistent with literature data (10–30% of the initial tyre mass) (Kole et al., 2017). The released TWP_{total} is 21,000 t/y of which about 10,000 t/y each entered surface waters (5,000 t/y direct and 5,000 t/y via waste water treatment) and soils, of these 600 t/y are particles <5 μm and 9 t/y are nanoparticles <0.1 μm. The calculations also show a TWP_{air} of 1,200 t/y (cf. Fig. 2). In summary, 80% of all tyre mass remains in the stock, while 6% is dissipated in the environment as TWP_{total} and 14% enters into the circular economy of the Austrian waste management system.

3.1.2. Substances – MFA on carbon black

Fig. 3 shows the MFA at the level of CB, enabling to identify potential emission hotspots during the use and EOL-phase as well as potential sinks in the environment. For parameterization of the exposure model, it was necessary to assume that CB remains in the rubber matrix, thus behaves similar to the polymeric components and deposits. This simplification was needed since no CB-specific transfer coefficients are currently published. With regard to risk assessment it is noted that most CB particles are incorporated into tyres as bulk material (aggregates up to 0.5 μm) and not as engineered nanomaterials (<0.1 μm) since this is currently cost prohibitive (Donnet et al., 1993; European Commission, 2007). Detailed information to the assumptions and data are shown in the SI (*see* section S4). In 2018, about 7,400 t/y CB were imported to Austria and in total 90,000 t/y CB were stocked. Due to the use of vehicle tyres, about 5,500 t/y CB were emitted as TWP_{total}, whereby it can be assumed that 93–99% remain embedded into fragments of their polymeric host material, and only a small amount is released as free (nano-) fillers (BASt, 2010; Wohlleben et al., 2016). Since there are no published studies concerning the behaviour of CB during waste water treatment, it was assumed that C_{60} (fullerene) nanomaterials and CB particles have comparable behaviour in the environment. Accordingly, a study by Wang et al. (2012) on the fate and biological effects of fullerene nanomaterials was used to calculate the input and output flows for this process. The CB contained in TWP_{total} ends up in the environment in the

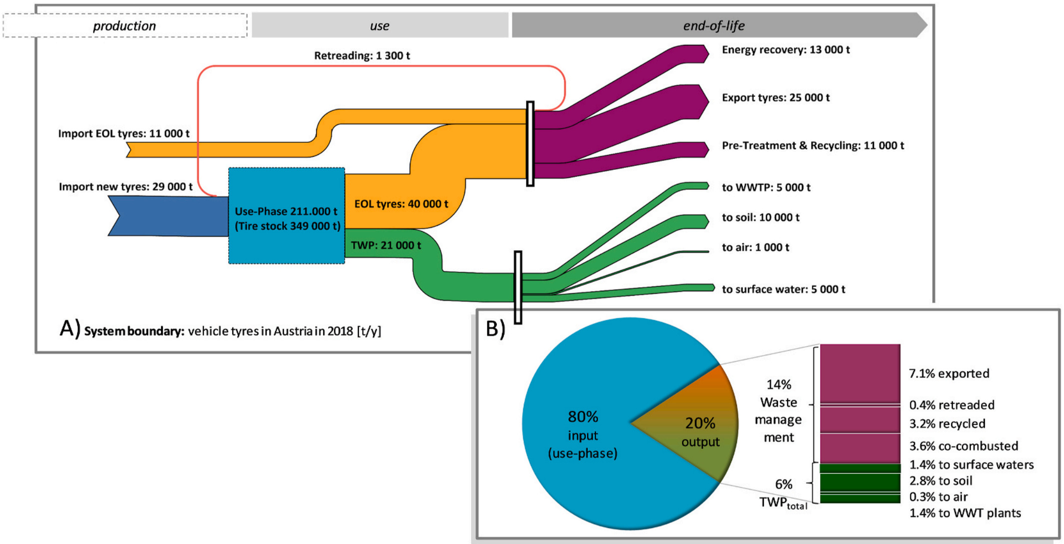

Fig. 2. A) MFA at the level of goods (vehicle tyres related to polymer components only (in tons per year)). B) Total stock of all vehicle tyres and resulting outputs in Austria (2018) (in tons per year).

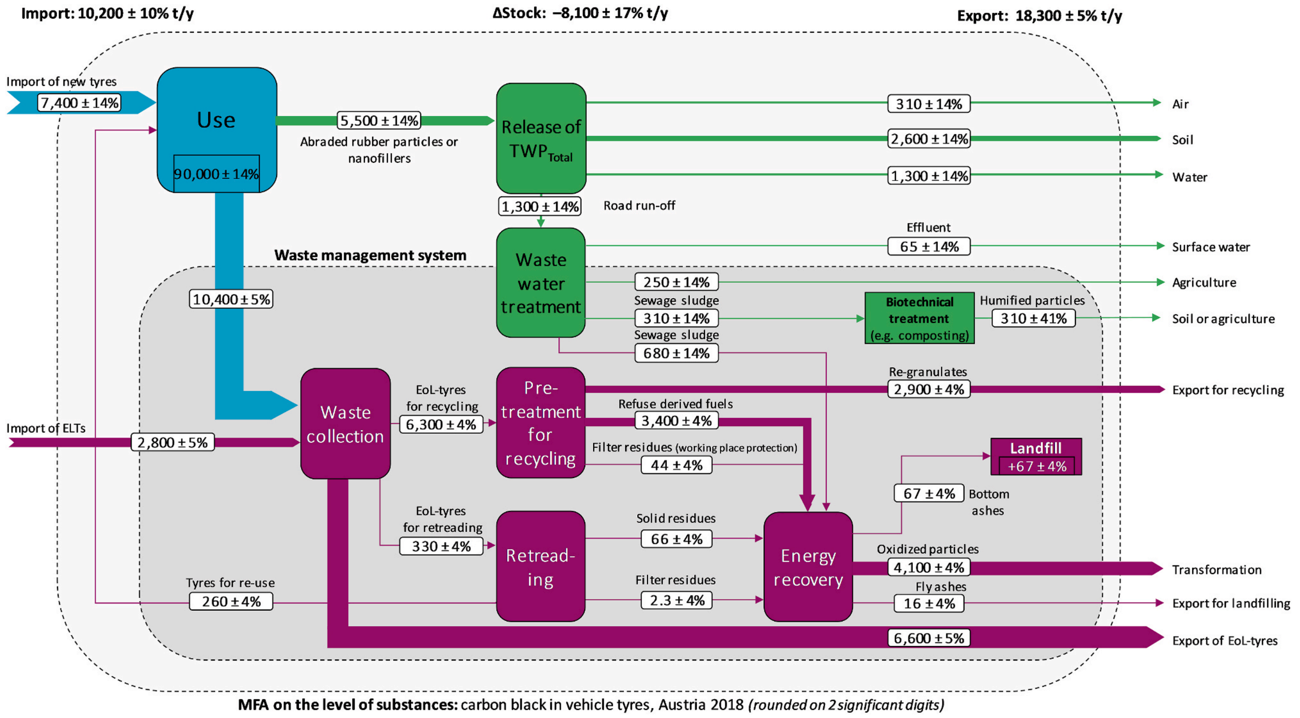

Fig. 3. MFA at the level of substances (CB used in vehicle tyres in Austria (2018) (t/y)).

same way as rubber: 47% in surface water or in waste water treatment plants (half each); 47% in the soil; 6% dispersed into the air.

With regard to the Austrian waste management system, about 10,400 t/y of CB entered directly the waste regime and 2,800 t/y were imported from abroad. About 50% of all collected EOL-tyres were exported, 47% mechanically pre-treated or shredded, and 3% re-treaded. About 2,900 t/y of CB were exported in form of rubber re-granulates (e.g., for new products in the automotive sector or road construction). Generally, the re-granulates from tyres are down-cycled (e.g., asphalt additives) rather than used for new tyres. The produced RDFs contain 3,400 t/y CB used for energy recovery – either in cement production or waste incineration plants. Overall, 4,200 t/y CB were incinerated as RDFs, filter residues or sewage sludge from waste water treatment.

3.2. Non-exhaust emissions

3.2.1. Tyre wear particles (TWP$_{total}$)

Table 3 shows the released TWP$_{total}$ in Austria compared to other studies which use different methods or data for their calculations (Baensch-Baltruschat et al., 2021; Bertling et al., 2018; Kole et al., 2017; Sieber et al., 2020). Methods for bottom-up extrapolation are based on an average measured weight loss during a car's service lifetime and derived mileage-dependent EFs (Baensch-Baltruschat et al., 2020; Sieber et al., 2020). Different vehicle types or fleet composition were included in these studies and therefore hamper comparability of the literature data.

In this study, the benchmark of 2.4 kg/cap was calculated for Austria. This figure is up to 4-timers higher than those for Switzerland or Germany. The difference can be explained by the fact that this study considers additional categories, such as TWVs and particularly the share from transit (freight) traffic in addition to PCs, LDVs and HDVs. This result highlights that transit (freight) traffic, carried by HDVs, plays a significant role in TWP quantification since 16% of the driven total HDV kilometres are caused by transit traffic (BMVIT, 2009).

The model in this study also shows that PCs, HDVs and LDVs are the largest sources for TWP$_{total}$, accounting for 41%, 31%, and 26% respectively, whereas busses (2%) and TWVs (0.5%) play a secondary role. The calculations in this study show that TWP$_{air}$ was 1,200 t/y, and thus on average about 6% of TWP$_{total}$ were released into the air. This percentage is in good agreement with other studies that estimated up to 10% of TWPs can be released as airborne dust (Barlow et al., 2007;

Table 3
Comparison of the total amount of tyre wear particles (TWP$_{total}$) from vehicle tyres in Austria (2018) to other countries.

Method	Area	Vehicle types[b]	TWP$_{total}$ [kg/cap.y]	Sources
Bottom-up extrapolation based on vehicle-specific EFs and mileage	Austria	PC, TWV, LDV, HDV incl. transit share[a]	2.4	This study
Top-down extrapolation based on TWP benchmarks derived from an e-mail survey and meetings	Germany	PC, motorbikes, trucks, skateboards, bikes	1.1	Bertling et al. (2018)
Top-down extrapolation based on average weight loss of 10–30% during a car tyres service life is assumed	Switzerland	PC, LDV, HDV	0.8–1.7	Sieber et al. (2020)
Bottom-up extrapolation based on vehicle-specific EFs and mileage	Germany	PC, motorbikes, busses, LDV, HDV	1.2	Baensch-Baltruschat et al. (2021)
Literature review	Worldwide	Considered vahicle types can vary from country to country.	0.2–5.5	Kole et al. (2017) Baensch-Baltruschat et al. (2020)

[a] Transit traffic is traffic through countries or states which are neither the start nor the destination of the journey.
[b] Abbreviations: *HDV (heavy-duty vehicles), LDV (light-duty vehicles), PC (passenger cars), TWV (two-wheel vehicles).*

Grigoratos and Martini, 2014; Gualtieri et al., 2005). The use of mileage-dependent, vehicle-specific EFs revealed that PCs are responsible for 61%, followed by HDVs for 26%, LDVs for 11%, busses and two-wheel vehicles for 1% of the total TWP_{air}.

3.2.2. Total suspended particles (TSP)

Based on Ntziachristos and Boulter (2019), TWP_{air} mostly consists of PM_{10} (60%) and $PM_{2.5}$ (42%), with smaller shares of PM_1 (6%) and $PM_{0.1}$ (5%) emissions. By including EF data published in Ntziachristos and Boulter (2019) for road vehicle-brake and road-surface wear (*see SI section S6*), it was further possible to calculate the sum of non-exhaust emissions from road traffic into the air which was in total 3,600 t/y. This result on the $TSP_{total,\ non\text{-}exhaust}$ in turn gives a distribution of 33% tyre wear, 22% road vehicle brake wear, and 45% road-surface wear. The calculated share of airborne tyre ($TSP_{tyre} = TWP_{air}$), vehicle-brake (TSP_{brakes}) and road-surface ($TSP_{road\text{-}surface}$) emissions in Austria in 2018 are listed in Table 4.

In summary, all non-exhaust traffic-related emissions ($TSP_{total,\ non\text{-}exhaust}$) contribute 9.3% to the national TSP emissions across Austria, which were about 38,000 t/y in 2018 (Umweltbundesamt, 2021). Literature data show that total traffic-related PM emissions can contribute between 5 and 80% to total airborne emissions, which is mainly influenced by traffic load, populations density and location (Panko et al., 2013; Pant and Harrison, 2013). For the year 2018, the Environment Agency Austria reported that road transportation contributes to 13% (4,980 t/y) of the overall TSP, which is the sum of exhaust and non-exhaust emissions in Austria (Umweltbundesamt, 2021). This figure was used to determine a contribution from non-exhaust traffic-related emissions ($TSP_{total,\ non\text{-}exhaust}$) of approx. 72% to the total TSP from road transportation. These results highlight that it is mainly through the reduction of tyre, brake and road-surface wear that overall non-exhaust traffic emissions can be reduced.

3.3. Scenario analysis 2050

The results of the scenario analysis are summarized in Fig. 4 showing in part (A) the mileage changes within the scenarios status quo (SQ), with-existing-measures (WEM) and Green-Deal (GD), and in part (B) non-exhaust emissions (tyre, brake and road-surface wear) for each scenario differencing BEVs with a 100-mile and 300-mile range. The scenarios indicate an increase of the number of annual kilometres for all vehicle types by 34% (WEM) and 28% (GD) in the period between 2018 and 2050. In both scenarios the numbers of BEV will increase, especially within the GD scenario in which all PCs and LDVs are replaced by BEV and 75% of the road freight (HDV) is shifted to railways.

The scenario analysis shows that in 2050 tyre wear ($TSP_{tyre} = TWP_{air}$) will increase by 35–46% if only existing measures (WEM) will be taken into account. Remarkably, it will also increase in the GD scenario by 17–40%. This development is due to the higher EF for BEV compared to conventional vehicles with combustion engines (ICEVs). Also, road-surface wear ($TSP_{road\text{-}surface}$) will increase by 33% or 7% respectively, due to higher number of kilometres driven. Vehicle brake wear (TSP_{brakes}) will only slightly increase within WEM but will substantially decrease with the GD scenario due to lower brake emissions rate of BEV. In summary, the analysis in this study shows that the Green-Deal targets of reducing the mileage of LDVs and HDVs on the road by 75% will not lead to a reduction in total non-exhaust emissions from road vehicles. This can be explained by the predicted increase of mileage of PCs and the currently higher EFs from BEVs than ICEVs. To reduce emissions in the transportation sector, TSP can only be significantly reduced by a general reduction of the vehicle kilometres travelled or by shifting personal mobility to public transportation, walking or cycling as well as shifting road freight traffic to rail. The latter one is already addressed in the Green-Deal scenario where it is stated that 75% of today's road freight should shift to rail- or waterways. A study by Brand et al. (2021) showed, for example, that only a shift to walking or cycling as mode of transport can lead to a significant CO_2 reduction in the case of personal mobility. The same is most likely true for non-exhaust emissions, since scenario analysis in this study shows that a complete shift to BEV is not enough to reduce tyre wear and non-exhaust emissions.

Table 4
Summary of non-exhaust emissions from road traffic (total suspended particles (TSP), and particulate matter (PM_{10}, $PM_{2.5}$, PM_1, and $PM_{2.5}$)) in relation to the national TSP for Austria 2018.

Air emissions	Total	PM_{10}	$PM_{2.5}$	PM_1	$PM_{0.1}$
$TSP_{tyre} = TWP_{air}$ [t/year]	1,200	720	500	72	57
TSP_{brakes} [t/year]	800	780	310	80	64
$TSP_{road\text{-}surface}$ [t/year]	1,600	790	430	*N/A*	*N/A*
$TSP_{total,\ non\text{-}exhaust}$ [t/year]	**3,600**	**2,300**	**1,200**		
Total national TSP of Austria [t/year]	38,000	26,000	14,000	*N/A*	*N/A*
- Share of $TSP_{total,\ non\text{-}exhaust}$	9.3%	8.7%	8.7%	*N/A*	*N/A*
Total road transportation TSP (both non-exhaust and exhaust emissions) [t/year]	5,000				
- Share of $TSP_{total,\ non\text{-}exhaust}$	72%				

N/A (not applicable because relevant data not available).

4. Conclusions

In Austria (2018), the total amount of emitted tyre wear particles was calculated at 2.4 kg per capita showing that 57% of the resulting total airborne dust emissions are caused by trucks (heavy and light-duty vehicles), and 41% by passenger cars, while busses (2%) and two-wheel vehicles (<1%) currently play a minor role. Since transit traffic plays a significant role in a country like Austria, this certainly should be considered in all future material flow analyses. Our scenario analysis for 2050 shows that both the continuation of existing measures and the shift to battery electric vehicles will lead to an increase of total non-exhaust emissions (i.e. tyre, break and road wear) due to an increase in overall mileage and higher vehicle-specific emission factors. More empirical research is needed on electric vehicles including release mechanisms, the amount of micro- and nanoscale debris, and the hazardous properties of tyre rubber and its components. Standardised analytical methods are needed to determine both particle size distributions and mass concentrations, particularly for nanoscale emissions such as airborne particulate matter ($PM_{0.1}$) or nanoparticles (<100 nm) dispersed in agricultural soil or surface water. In addition, the development of innovative vehicle tyres and eco-friendly rubber formulations is needed to reduce tyre wear and facilitate recyclability. Nanotechnology-based fabrication methods (e.g. shown by Togo et al. (2019)) or the use of engineered nanomaterials (e.g. summarized in OECD (2015)) could allow the reduction of the amount of tyre wear compared to conventional vehicle tyres. In doing so, the principles of "safe and sustainable by design" need to be applied and more exposure studies will be necessary to minimise potential risks. In conclusion, the results of this study provide a solid database useful in traffic planning and for quantitative risk assessment of chemicals. Nevertheless, data uncertainty and comparability can be improved by updating the model with the latest measurement data and by integrating data on temporal dynamics of markets and stocks of vehicle tyres. Dynamic material flow analysis in combination with a fate model adapted to the micro- or nanoparticles of tyre wear would enable the prediction of accumulation effects or the degradation of tyre rubber, which can last for decades. Future exposure and fate model should be extended with regard to potentially hazardous rubber additives or emerging pollutants (e.g. antioxidants such as *p*-phenylenediamines, as shown in Huang et al. (2021)).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

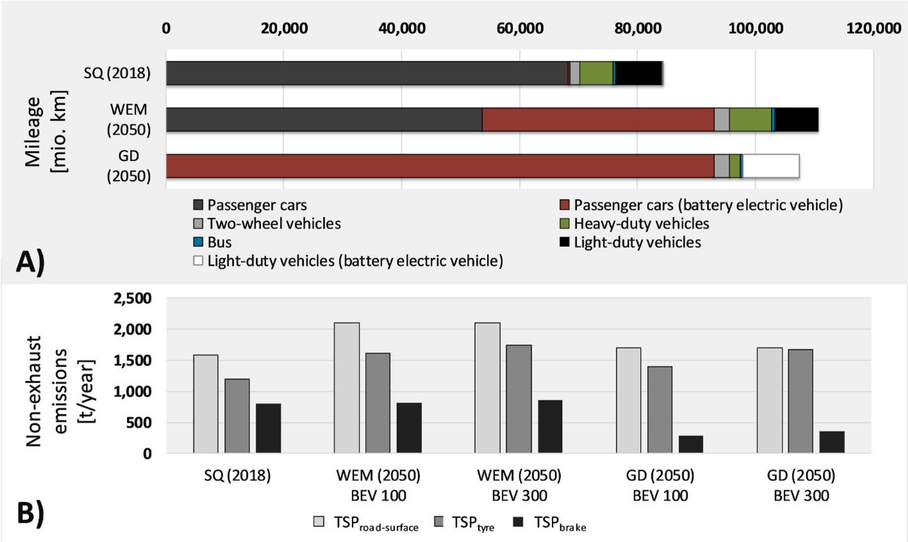

Fig. 4. Scenario analysis: Mileage [km/year] and wear [t/year].

the work reported in this paper.

Acknowledgments

The authors acknowledge the Austrian Federal Ministry of Climate Action, Environment, Energy, Mobility, Innovation and Technology for supporting the project "NanoAdd" (No. 867565), which is part of the national "NANO Environment Health and Safety program". Any findings and conclusions expressed are those of the author(s) and do not necessarily reflect the views of the Federal Ministries.

Thanks to Robert Jones for proofreading the article.

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

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.118102.

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