Supplementary Material

Tyre wear nanoparticles as test for a nano risk governance framework

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# Supplementary Data

This supplementary material is a summary of the technical reference document that was published with the title “Airborne release of tyre wear particles”, as part of the Case Study Rubber Tyres, carried out within the European Union’s Horizon 2020, Research and Innovation programme NMBP13 (Grant Agreement 814530), within the project NANORIGO. The technical reference document is available on ResearchGate. (Broekhuizen 2022).

For risk governance it is important to carefully distinguish TWP and TRWP, but for real-life sampling this can be complex. The term TWP (tyre wear particles) is used for particles not contaminated with road wear, which is the most likely form to be generated in laboratory experiments (e.g., a road simulator). The term TWP is also used when the release of tyre wear is discussed in general. The term TRWP (tyre and road wear particles) is used for associates (agglomerates or aggregates) of TWP with road wear particles, i.e., the most likely form for environmental sampling and for risk assessment in practice (Halle et al 2020). T(R)WP is used when both forms are likely.

TWP are also classified under the general definition of microplastics. To define ‘nanoplastics’, publications refer to upper size limits of either 100 nm or 1000 nm (Mitrano et al 2021). In analogy with the EC definition for nanomaterials (EC 2022), this study uses an upper limit of 100 nm for the qualification of nanoplastics. Pragmatically, this upper limit is used as well for nano-TWP.

# Introduction

The rubber tyre case study (RTC) is one of the case studies within the European NMBP13 projects, that are active in 2019-2023 with the development and establishment of a solid, well-balanced nano risk governance approach for Europe. The RTC investigates what can be learned from applying this nano risk governance approach to the problem of tyre wear generated by car driving activities. Within the frame of the NMBP13 project there is a special focus on the nanomaterials. As part of the RTC, this technical reference document collects available risk-related scientific information about environmental release of tyre wear particles (TWP) and its hazards. TWP generally associates with road wear to form TRWP (Tyre and Road Wear Particles). The particles’ size distribution concerns coarse, micro-sized and nano-sized particles and its associates.

From this literature research, the main obstacle to clearly understanding the contribution of nano-TWP to the health and environmental problem is the predominant use of the mass metric for measurements, rather than particle numbers. As a result, the 'nano contribution' is often ignored (van Broekhuizen 2022). Still, there are a few recent publications explicitly measuring real-life particle number-based concentrations of nano-TWP (Dahl et al 2006, Mathissen et al 2011, Beji et al 2021). Also, the relevance of a specific focus on nano, within the context of TWP release and the microplastics problem gains growing political attention (OECD 2014, 2020, 2020a), but suffers a huge shortage of relevant real-life measurements (Andersson-Sköld et al (2020), Mennekes et al 2022). TWP associates (agglomeration and/or aggregation) with road wear particles, which are collectively referred to as tyre and road wear particles (TRWP). TWP is estimated to contribute up to 30-50% to the TRWP mass (Simons et al 2016; Järlskog et al 2020).

Also, within the Safe-by-Design policy area a specific focus on ‘nano’ seems the more relevant, since the used nanomaterials in the rubber tread, amongst which carbon black and amorphous silica (up to 30% w/w) and nanoclay (Ullmann 2011, IARC 2012, SCOEL 2016) are generally not released as such, but rather as part of the coarse, micro and nano-sized TWPs, consisting of abrasion wear and conversion products generated by heating, combustion and nucleation (Cadle and Williams 1978, Fukahori et al 2020, Wagner et al 2022). Especially the use of amorphous silica shows an increasing use (Solvay 2015, Evonik 2015,2018,2019). On-going rubber research on nano-additives for rubber tread shows interests in Nanoprene (Nanowerk 2008), carbon nano fibres, graphene and several other MNMs (Felix and SivaKumar 2014; Bridgestone n.d.; INSCX, nd; Nanotech, nd). But according to information from tyre manufacturers (anno 2022) most potential MNM-applications are still mainly in the laboratory phase.

# Generation of nano-sized TWP

Driving behaviour is an important trigger for TWP formation. Decent driving conditions show a low TWP-release, but stressed driving, such as rapid cornering, acceleration, deceleration and full stop braking show larger emissions (Dahl et al 2006; Mathissen et al 2011, Grigoratos 2014, 2018, Beji et al, 2021). Mild braking at 90 km/h can generate more than twice as much T(R)WP as heavy braking at 50 km/h (Beji et al, 2021). The size distribution of released particles from braking is similar to that of acceleration, i.e., higher percentages by number of nano-sized particles than those determined at stabilized speeds (Beji et al, 2021). Under normal, decent driving conditions the average micro-sized mass release (for four tyres) ranges between 90-270 mg/vehicle.km, depending on the type of vehicle, road and tyre and driving behaviour (Unice et al 2019; CEDR, 2018). In particle numbers, for steady straight driving an average release was estimated of 1∙1011 #/vehicle.km (Mathissen et all 2011) with extremes reported of 14∙1011 #/vehicle.km (Emissions Analytics 2022b). Also release particle number concentrations are reported for particle-size < 1µm as high as 1,85∙107#/cm3 (Belkacem et al, 2022).

It is estimated that 10-30% (w/w) of the tread rubber is lost during the tyre life time (Grigoratos, 2014). For Germany (with 47,7∙106 cars in 2019 (Statistica, 2022)) this builds up to 100.000 ton/y of micro-sized TRWP, for the Netherlands (with 8,8. 106 cars and an average distance driven of 12,9∙103 km/car/y (CBS 2022)) this is 17.300 tons and for the EU 1.327.000 ton (Verschoor et al 2016; Wagner et al, 2018). Based on these numbers for NL a particle number release per vehicle of 1015 #/verhicle/y and totally 1022 #/yr can be calculated. In mass these range from 0,23 - 4,7 kg/year, with a global average of 0,8 – 1,2 kg/year (Kole et al, 2017).

Measurements of real-world particle’ numbers at the tyre-road interface show that most transient or Aïtken nuclei mode TRWP (size distribution: 10-100 nm) stem from tyre wear, whereas a large proportion of the accumulation mode (0,1-2μm) and coarse mode (>2μm) TRWP consist of re-suspended road dust and wear particles from the tyre, brake and road surface (Beji et al, 2021). In a number-based approach the inhalable PM10 fraction of TRWP falls mainly in the size modes of <30 nm and 50-300 nm, whereas the mass of TRWP is mainly determined by the size fractions of 100-600 nm and 1,0–15 μm (Beji et al, 2021). In particle numbers, the ultrafine TRWP fraction (<100nm) may contribute up to 92% of the total number of particles in the PM10 fraction (Emissions Analytics, 2022b). Additionally, nucleation products may be formed at the heated tread surface, with generally a bi-modal size distribution: 10–100nm and 0,1–2µm (Mathissen et al, 2011; Grigoratos, 2014). Further environmental fragmentation of microplastics in nanoplastics is suggested through secondary processes influenced by UV-light, oxidation, shear and other aging processes (Enfrin et al, 2020), but so far for this no TWP- or TRWP-specific evidence was found.

# Fate

The fate of TRWP depends on the local geography, such as steepness of the terrain, the proximity of surface water and runoff collection systems. Mass-based estimations are that 67% ends up in the soil, 12% in the air, 6% directly in the surface waters and 15% entering in sewers (which indirectly leads to a 12% end up in surface waters) (van Duijnhove et al 2014). For German highways it is estimated that 11.000 tons/year reach surface waters (Wagner et al, 2018). TRWP concentrations measured in soil range from 0,2 - 160 mg/g dry weight (Baensch-Baltruschat et al, 2020). The relative contribution of TRWP to the total global load of microplastics ending up in oceans is estimated to be <5–10%. In air, 3–7% of the particulate matter (PM2,5) is estimated to consist of TRWP, but here as well the estimations vary widely (Kole et al, 2017). Much lower concentrations are measured as well. Airborne nanoparticles may as well deposit in extreme remote areas, such as the high Alps, and the Polar areas, for which deposits in Greenland firn ice were associated with tyre wear (Materic et al, 2021, 2022).

Occupational exposure measurements of road traffic officers to traffic-generated particles (exhaust and non-exhaust particles) show an exposure to UFPs (nanoparticles) of 7∙104 #/cm3 (8hr-TWA), based on a working day of 3,5h inside- and 3,5h outside the car and 1h at the office, and taking into account that cabin filters and recirculating air may reduce the UFP exposure concentration up to 45-50% (Wander & Verbist, 2017). This 8h-TWA concentration is 5 times higher than the background particles’ number concentration. Short term peak exposures of 2∙106 #/cm3 were measured. Road maintenance workers along the highway show an exposure (exhaust and non-exhaust particles) of 7,5∙104 particles/cm3, with peaks of 3,3∙105 particles/cm3 (Meier et al, 2014).

# Hazard

Mentioned leachable compounds in TWP are Zn-, Cu- and S- compounds, PAHs, benzothiazoles, phthalates, natural resins and paraffins (Sarasa et al 2006, Wik et al 2009, Redondo-Hasselerharm et al 2018; Hasse et al 2021; Emissions Analytics 2022a) and quinones (Tian et al 2020). Wik et al (2009) indicate that T(R)WP may be bioavailable to pelagic filter-feeding organisms, to benthic organisms, and to plants, and that several of the compounds that leach from tire rubber to water are bioavailable to aquatic organisms like fish. Contrary to this Redondo-Hasselerharm et al (2018) could not show effects on aquatic organisms (insects, crustaceans and snails) exposed to TWP at realistic environmental (leachate) concentrations. Effects at a TWP concentration between 500-2000 #/ml on survival, reproduction and growth were observed (Kahn et al, 2019). Tamis et al (2021) proposed a PNEC for TWP-derived microplastics of 0,33 µg/l in water and 100 µg/kg in dry sediment. Observed TWP concentrations in river water range from 0,5-500 mg/l and in river sediments from 500-2000 mg/kg. Acute mortality under (reproducing) adult Coho salmon was associated with the exposure to oxidized 6PPD-quinone (Tian et al, 2020; 2022). This 6PPD-quinone (the oxidised form of the rubber antioxidant 6PPD) is supposed to leach from TWP.

*In vitro* model extracts of TWP show cytotoxicity, genotoxicity and oxidative stress in A549 cells, genotoxicity and inflammatory response in macrophage and lung epithelial cell (Kreider et al, 2019). Animal studies are more equivocal, with some findings of cytotoxicity and inflammation and other findings of small transient effects on inflammation in the absence of cytotoxicity following instillation of TWP (Kreider et al, 2019). For respirable TRWP a NOAEC of 55 μg/m3 was derived, indicating a low potential risk for human cardiopulmonary effects (Kreider et al, 2019).

To date no nano-specific health hazards of T(R)WP have been identified. But, although real-world exposures are likely to be much lower than the *in vitro* studies, there are indications of their health and environmental hazard. These relate to the heterogeneous composition of TWP and TRWP, its hazardous components and leachates, the findings from the (eco)toxicological tests with TWP-extracts and the findings with the Coho-salmon, as well as the generally hazardous nature of persistent nanoparticles. Oxidative stress leading to inflammation is likely to be one of the key mechanisms of exposure to persistent nanomaterials (Shvedova et al 2010; Fadeel et al 2012; Reijnders 2012; Jeevanandam, 2018).

# OELs and Guidance values

No specific OELs have been established for exposure to (fine) rubber dust, neither for occupational exposure to TWP or TRWP, nor for nanoparticles’ exposure (Mihalache et al, 2017). Germany established the most stringent OEL for (undefined) respirable dusts: 1,25 mg/m3.

Provisional Nano Reference Values (NRVs), established in the Netherlands, advise to accept a maximum exposure level of 4∙104 #/cm3 (8h-TWA) for airborne nanoparticles (SER, 2012). These generic NRVs are based on the size, shape and density of the nanoparticles. The German BAUA advises to use 0,5 mg/m3 for nanomaterials without specific toxicity, or 0,1 mg/m3 for nanomaterials with a specific toxicity (Packroff, 2015). So, in principle these approaches are advisable as well for the risk assessment of airborne exposure to nano-sized TWP or nano-sized TRWP. In line with the NRVs, the Dutch Rijkswaterstaat advised to use an OEL of 6∙104 #/cm3 for exposure to airborne particulates along highways (exhaust + non-exhaust particles) (Wander & Verbist 2017).

Air quality guidelines for TWP or TRWP have not been established, nor for UFP (EC, 2007). The WHO-advised mass-based annual average for airborne fine dust PM10 -particles is 20 µg/m3 and 10 µg/m3 for PM2.5 (Orellano et al, 2020; Chen et al, 2020). The EC Euro 5 and 6 standards for exhaust emission of cars set at a limit for PM at 4,5 mg/km, and particle numbers of particles at a maximum 6\*1011 #/km, but the standards do not include non-exhaust emissions such as particulates from tyres and brakes. For this reason, particles generated by brakes and tyres are part of the emissions considered in the impact assessment supporting the preparation of the upcoming Euro 7 pollutant regulation (EP 2022).

The EU tyre labelling system (EC 2020b), as instrument to encourage the 'greening' of tyre innovation and purchasing, combines rankings for the rolling resistance (energy consumption), wet grip (safety in wet conditions) and external noise and includes pictograms for snow and ice grip. Nevertheless, it lacks a ranking for the abrasion resistance as most important indicator for TWP emission reduction.

# Suggested mitigation measures

Policy platforms recognise the potential problems of TWP release and discuss mitigation measures, distinguishing technical, infrastructural and behavioural levels (Verschoor et al, 2016; OECD, 2021; ADAC 2021):

* The technical level of innovation concerns reducing abrasion wear and at the same time optimizing energy-saving, wet-grip and noise performance of the tyres.
* At the infrastructural level, measures concern tasks for government agencies related to the improvement of road surface composition, design and maintenance, while also taking into account the influence of road material and design on abrasion wear, particulate absorption capacity, cleaning and maintenance of road surfaces. roads and efficient drainage of roads.
* The behavioural level focuses on driving behaviour and includes government incentives for optimizing the use and maintenance of cars (and tyres), speed-limiting measures, reducing the total distances travelled and encouraging the purchase of lightweight cars. But it also includes raising consumer awareness about TWP emissions, convincing them of the need to reduce them, and that they themselves play a role in this. In this regard, confounding elements are, for example, the growing market introduction of heavy consumer cars (SUV), as well as the pollution reduction incentive advocated by the purchase of high-weight electric cars.

Notwithstanding the importance of increasing attention to the environmental load of micro and coarse TWP and TRWP, this mass-based predominance hinders a clear focus on the release, hazards and risks of the nanosized TWP emission. So far, the limited number of nano-targeted TWP release studies complicates the qualification and quantification of clearly existing 'nano-risks' and contextualization.

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