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The Policy Relevance of Wear Emissions from Road Transport, Now and in the Future—An International Workshop Report and Consensus Statement

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REVIEW PAPER

The Policy Relevance of Wear Emissions from Road Transport, Now and in the Future—An International Workshop Report and Consensus Statement

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Road transport emissions are a major contributor to ambient particulate matter concentrations and have been associated with adverse health effects. Therefore, these emissions are targeted through increasingly stringent European emission standards. These policies succeed in reducing exhaust emissions, but do not address "nonexhaust" emissions from brake wear, tire wear, road wear, and suspension in air of road dust.

Is this a problem? To what extent do nonexhaust emissions contribute to ambient concentrations of PM_{10} or $PM_{2.5}$? In the near future, wear emissions may dominate the remaining traffic-related PM_{10} emissions in Europe, mostly due to the steep decrease in PM exhaust emissions. This underlines the need to determine the relevance of the wear emissions as a contribution to the existing ambient PM concentrations, and the need to assess the health risks related to wear particles, which has not yet received much attention. During a workshop in 2011, available knowledge was reported and evaluated so as to draw conclusions on the relevance of traffic-related wear emissions for air quality policy development. On the basis of available evidence, which is briefly presented in this paper, it was concluded that nonexhaust emissions and in particular suspension in air of road dust are major contributors to exceedances at street locations of the PM_{10} air quality standards in various European cities. Furthermore, wear-related PM emissions that contain high concentrations of metals may (despite their limited contribution to the mass of nonexhaust emissions) cause significant health risks for the population, especially those living near intensely trafficked locations. To quantify the existing health risks, targeted research is required on wear emissions, their dispersion in urban areas, population exposure, and its effects on health. Such information will be crucial for environmental policymakers as an input for discussions on the need to develop control strategies.

Implications: Road transport particulate matter (PM) emissions are associated with adverse health effects. Stringent policies succeed in reducing the exhaust PM emissions, but do not address "nonexhaust" emissions from brake wear, tire wear, road wear, and suspension in air of road dust. In the near future the nonexhaust emissions will dominate the road transport PM emissions. Based on the limited available evidence, it is argued that dedicated research is required on nonexhaust emissions and dispersion to urban areas from both an air quality and a public health perspective. The implicated message to regulators and policy makers is that road transport emissions continue to be an issue for health and air quality, despite the encouraging rapid decrease of tailpipe exhaust emissions.

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Introduction

Particulate matter (PM) in ambient air consists of tiny particles of solid matter or liquid droplets suspended in the atmosphere. Well-conducted cohort studies in Europe have indicated that air pollution emitted from road traffic, including PM, is of particular concern, and long-term exposure to traffic-related air pollution may shorten life expectancy (e.g., Hoek et al., 2002; Gauderman et al., 2007; Janssen et al., 2003). The principal local source of particulate matter in many European cities is road traffic (e.g., Pey et al., 2009; Lonati et al., 2011). Exhaust and nonexhaust emissions from road traffic result in enhanced levels of PM in urban areas and in particular near trafficked locations (Table 1). In general, trafficked sites are the most likely locations for PM_{10} levels to exceed the European Union (EU) annual limit value of 40 μg m^{-3}.

While engine exhaust emissions have been strongly reduced by EU emission standards in the past decades, wear emissions are unaffected by such measures. Recent studies have identified a significant contribution of nonexhaust emissions to the traffic-related PM_{10} load of the ambient air (Gehrig et al., 2004a; Thorpe and Harrison, 2008), yet very little is known about this fraction in terms of health impacts. This paper describes the results of a 1-day international workshop on nonexhaust emissions held in June 2011 entitled "Policy Relevance of Wear Emissions From Road Transport, Now and in the Future." The workshop, of which the outcomes are reported here, aimed to provide air quality policymakers with a preliminary and indicative assessment of the health relevance of wear emissions from road transport, now and in the future, and the subsequent (possible) need to develop control policies.

Nonexhaust emissions, being brake, tire, and road wear emissions together with traffic-induced resuspension of deposited road dust, may contribute up to 50% of local road traffic emissions of PM_{10} (Bukowiecki et al., 2010). In Scandinavian countries, the contribution may be even up to 90% of PM_{10} along roads in early spring (Johansson et al., 2007a). Since the trend is toward cleaner exhaust through the use of catalytic converters, diesel particulate filters (DPF) and improved fuels and engines, in the near future (if not already) nonexhaust particulates may exceed the exhaust PM in terms of emissions and may become dominant by 2020 (Rexeis and Hausberger, 2009).

The contribution of nonexhaust emissions varies temporally and spatially across Europe. This is illustrated by Figure 1, which presents PM_{10} concentrations at a street canyon in Stockholm, Sweden, during the various seasons of the year.

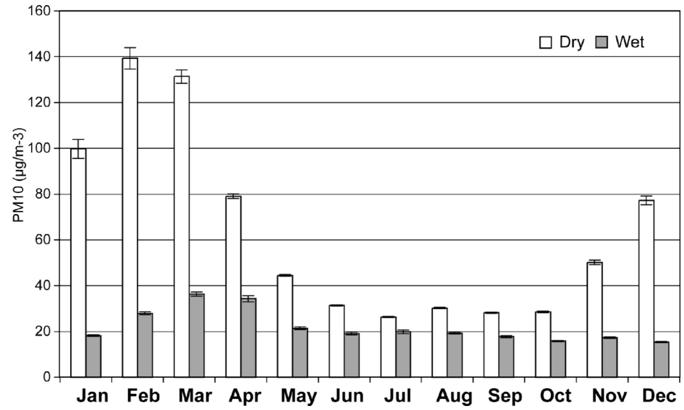

Figure 1. Monthly average PM10 data for 2005–2011 with standard error from Hornsgatan, Stockholm (street canyon), classified according to a wet and dry road surface.

In southern European cities, the contribution of wind-blown soil in nonexhaust emissions is relatively large because of the dry climatological conditions, especially in the summer season. In central Europe, other processes play an important role. In particular, pavement wear contributes to PM_{10}, mostly to the coarse fraction $PM_{2.5\text{-}10}$, even when nonstudded tires are used and especially when driving over damaged pavement in poor condition (Gehrig et al., 2010). These examples show the complexity implicit in temporal and spatial resolution of nonexhaust emissions. The chemical composition of individual nonexhaust particles covers a wide range, potentially making one type of wear particle more toxic than others. Hence, generalization of the health relevance of nonexhaust particles is difficult, if not impossible.

The workshop reported here brought together participants from various disciplines to assess the current state of knowledge regarding wear emissions, from emission source to health impact. The following issues were addressed:

- Concentrations and size distributions of particles from nonexhaust emissions.
- Emissions of nonexhaust particles and their impact on ambient air quality.
- Chemical composition and relative toxicity.
- Exposure and potential health impact.

Table 1. Average annual levels (μgm^{-3}) of PM_{10} and $PM_{2.5}$ recorded at regional background (RB), urban background (UB) and traffic sites (TS) in central, northern and southern Europe

	Central Europe			Northern Europe			Southern Europe		
	RB	UB	TS	RB	UB	TS	RB	UB	TS
PM_{10} (μgm^{-3})	14–24	24–38	30–53	8–16	17–23	26–51	14–21	31–42	45–55
$PM_{2.5}$ (μgm^{-3})	12–20	16–30	22–39	7–13	8–15	13–19	12–16	19–25	28–35

Notes: (compiled from Querol et al., 2004; Putaud et al., 2004; Denier van der Gon et al., 2010).

Emissions, Composition, and Exposure

Emissions

Until the early 1990s, road transport emissions were dominated (80–90%) by exhaust emissions (Figure 2a). Over time this proportion has gradually decreased to ~65% (Figure 2b). This decreasing trend is likely to continue due to further exhaust emission reductions and a continuing growth in vehicle kilometers. The latter is relevant because wear emissions are proportional to vehicle kilometers driven and current legislation does not require a reduction in wear or resuspension emissions per vehicle kilometer traveled.

Figure 2b presents the fraction of road transport emission arising from nonexhaust sources as an average for Europe. This includes a large proportion of extra-urban highway traffic, where wear emissions are expected to be lower than in the urban environment with its frequent stop-and-go driving and cornering. Estimates of the contribution of nonexhaust particles to total vehicle-generated particulate matter are in the range of 35–55% (Harrison et al., 2001; Charron and Harrison, 2005; Harrison et al., 2011). Recently, Rexeis and Hausberger (2009) predicted, using a detailed emission model for the Austrian fleet, that the percentage of PM nonexhaust of the total PM emissions will increase from about 50% between 2005 and 2010 up to some 80–90% by 2020. Hence in the European urban environment, where most Europeans live and spend their time, the contribution of nonexhaust in total exposure to urban road transport PM_{10} emissions easily reaches 50% and is likely to grow.

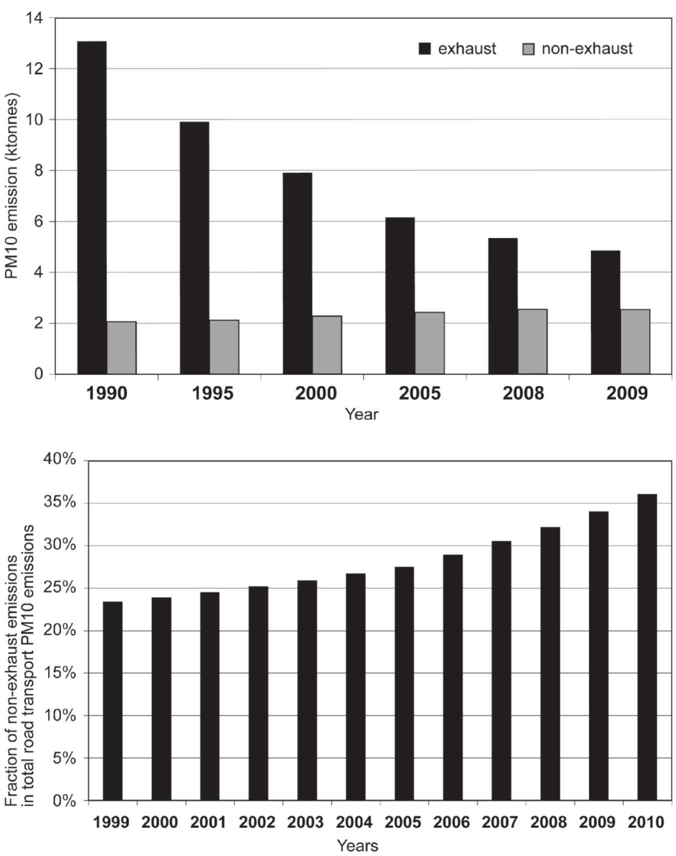

Figure 2. (a) Trend of PM_{10} emission from road transport exhaust and nonexhaust in the Netherlands (source: PRTR, 2011). (b) Average trend in nonexhaust emission for Europe based on extrapolation of base years in the IIASA GAINS model (source: GAINS, 2011).

Formation and composition

The properties of nonexhaust particles differ greatly from those of exhaust particles, but the difference in size, chemistry, morphology, and so on between various types of nonexhaust particles is also substantial. The size of most nonexhaust particles is generally several times larger than exhaust particles. This is due to the processes of formation, which for nonexhaust particles include mechanical abrasion, grinding, crushing, and corrosion, whereas exhaust particles are formed in combustion processes. Important exceptions to the typically rather coarse size of wear particles are nanometer-sized particles from tires, presumably formed due to thermally induced release (evaporation) of tire rubber components that form volatile droplets (Gustafsson et al., 2009b), and submicrometer particles from brake wear (e.g., Sanders et al., 2003). Because nonexhaust particles are large in comparison to exhaust particles, they contribute far more to the overall mass than to the number of particles (see for example Harrison et al., 2011). However, submicrometer particles arise as minor contribution to mass of tire and brake wear, but may represent a dominant contributor to particle number from those sources.

The chemical composition also differs; exhaust particles contain much more carbonaceous material (elemental carbon and organic compounds) as a direct result of fuel combustion and relatively less metal species than nonexhaust. In terms of chemistry the difference between various nonexhaust particles may be substantial depending on their origin. Particles from brake, tire, and road wear each have their unique chemical signature. Other properties that may differ between exhaust and nonexhaust particles are morphology, hygroscopicity, and surface reactivity. Several of the differences mentioned here may contribute to making nonexhaust particles more (or less) toxic.

Tire wear

Tires may generate particles both through the wear of the tire rubber and through the wear of road surfaces. These processes may depend on tire type, size, and age, vehicle speed and weight, road surface properties (stone mineral and size, texture), and meteorological conditions (temperature, road wetness, etc.). Tire wear contributes to PM_{10} even though most of the wear results in larger particles (Kreider et al., 2010). A peak at about 1–2 μm is likely to be related to worn rubber (Fauser, 1999; Gustafsson et al., 2009b), while peaks within the nanometer sizes are thought to be related to high-temperature events (Dahl et al., 2006). In laboratory tests, a nanometer-sized fraction at 20–50 nm was related to the occurrence of tire studs (Gustafsson et al., 2009b). Recently, Mathissen et al. (2011) found no increased nanometer particle concentrations emanating from tires during normal driving, but in maneuvers with significant tire slip, particles within the 30–60 nm range were generated. The chemical components of tire tread

Table 2. Inorganic composition of tyre and/or tyre tread rubber mixes in the 1990s. (Denier van der Gon et al., 2003)

Metal	Concentration range (mg/kg)	Metal	Concentration range (mg/kg)
Ag	0.08	Mg	32–444
As	0.8	Mn	2–14
Al	81–956	Mo	2.8–10
Ba	0.9–4.1	Na	610
Ca	113–1500	Ni	0.9–50
Cd	0.28–4.96	Pb	1–160
Co	0.88–39	Sb	2
Cr	0.4–49	Se	4–20
Cu	1.8–69	Sr	1.16–3.13
Fe	2–2800	Ti	195
K	180	V	1
Li	0.23–2.3	Zn	8000–13500

Sources: Malmquist (1983); Hewitt and Rashed (1990); Brewer (1997); Legret and Pagotto (1999); San Miquel et al. (2002).

particles include silica, sulfur, and zinc, but 46% of the total mass is polymers and 19% carbon black (Kreider et al., 2010).

An average of 5–10% of total tire wear will become airborne (EEA, 2009). Tire formulation and construction is an area of ongoing innovations and the exact composition and formulation are often proprietary. This implies that the chemical tire tread profile determined some 10 years ago (Table 2) may not be relevant anymore. There are no programs that monitor chemical composition of average in-use tires; hence we lack good data to characterize this source and the changes in best tracer elements over time.

Brake wear

Brake wear is due to forced deceleration of road vehicles during which brake linings are subject to large frictional heat generation. Detailed laboratory tests conducted by Sanders et al. (2003) showed that ∼50% of the total wear is emitted as airborne material; the other half directly deposits on the (road) surface and the wheel of the car. Since the phase-out of asbestos, a wide spectrum of formulations is applied in brake pad material, for example, matrices of organic resins, fillers such as barium sulfate or other inorganic material, friction modifiers such as graphite, talc, and antimony, and fibers of steel, glass, and brass (Chan and Stachowiak, 2004; Blau and Meyer, 2003, Garg et al., 2000; Sanders et al., 2003).

In field measurements, the appearances of antimony (Sb), copper (Cu), and barium (Ba) are often used as indicators for brake wear (Gietl et al., 2010; Bukowiecki et al., 2009a). A compilation of data on the copper content of brake pads and linings indicated that the copper contents vary between 1 and 14%; average concentrations reported were 5% for the United States and 10% for Europe (Denier van der Gon et al., 2007). Despite the range in contents, Denier van der Gon et al. (2007) showed that copper is a reliable tracer for brake wear with consistent appearance in reported chemical composition of brake linings, emission factors derived from tunnel studies, and increased elevations of Cu in PM samples sampled close to roads (Figure 3).

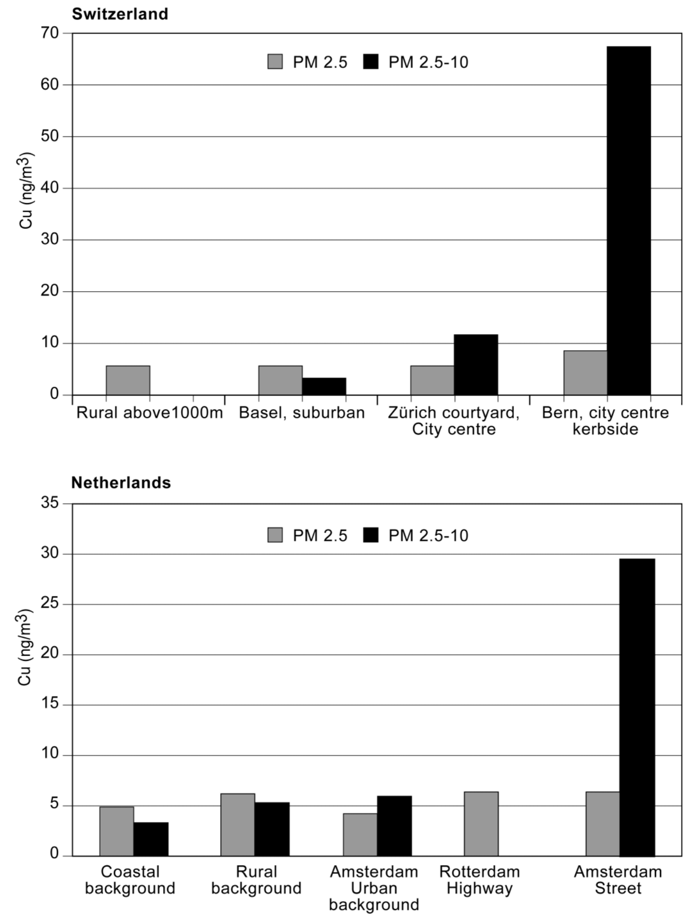

Figure 3. Concentration of copper in ambient PM samples at various locations in the coarse fraction (PM $_{2.5\text{-}10}$) and the fine fraction ($PM_{2.5}$) for Switzerland (top) and the Netherlands (bottom). (Recalculated from data by Hueglin et al., 2005, and Visser et al., 2001, respectively; source: Denier van der Gon et al., 2007).

Brake wear normally forms at high temperature, which may explain why the particle size is relatively small and will change over time due to cooling and agglomeration. Some 50% of the total wear mass of brake pads becomes airborne (EEA, 2009). Concentration peaks have been observed both in the micrometer size range but also at 200–300 nm and at about 100 nm (Wahlström et al., 2010). Note that in Figure 3 the enrichment in Cu (and thus brake wear) near roads is overwhelmingly seen in the coarse fraction, whereas based on various size-resolved emission factor measurements a larger proportion was expected to be in the fine fraction ($PM_{2.5}$). This example stresses the very important point that at present we cannot assess the importance of particle size distribution in combination with chemical composition. The connection between size distribution, chemical composition, exposure, and possible health effects clearly warrants further research.

Road wear

Wear of the road surface may vary drastically depending on the properties of the asphalt as well as tire type, vehicle type, and speed, as well as road surface conditions (covered with, e.g., sand, salt, snow, or ice). Key properties of the pavement are the maximum stone size and type of rock material used, both affecting wear resistance as well as pavement type, for example, densely packed, porous, or rubber-mixed pavement (Gustafsson et al., 2011b; Gehrig et al., 2010). Use of studded winter tires and/or sand/salt makes road wear and subsequent particle emissions increase drastically as seen in Stockholm (Figure 1), but also nonstudded winter tires result in slightly higher wear particle emissions than summer tires. This is likely to be due to the softer, more treaded rubber, increasing the shear forces to the pavement. Road wear, pavement-derived PM_{10}, mainly consists of small mineral fragments and therefore is dominated by crustal elements like Si, Ca, K, Fe, and Al. The composition therefore differs depending on the rock material used. The mass size distribution within PM_{10} normally peaks around 5 μm and very little mass resides below 0.5–1 μm (Gustafsson et al., 2008). Identification of road-wear-related airborne particles is more robust than for tire and brake wear particles, as road surfaces consist of 95–99% rock material and the prime elemental contributors are silicon and aluminum. The disadvantage is that it is nearly impossible to separate primary road wear from other mineral dust deposited on the road. Moreover, road dust can contain material from numerous other sources such as gritting material (sand/salt), corrosion products of vehicle components and street furniture, road maintenance activities, vegetative material, and material from building renovation/construction activities (Luhana et al., 2004; Thorpe and Harrison, 2008). Since sources differ as do the traffic characteristics, climate, and meteorology, the occurrence and properties of road dust are very variable in time and space.

Emission factors and source apportionment

In the past, particle emissions from road traffic were generally assumed to be associated with exhaust emissions only. A Swiss study identified a significant contribution of nonexhaust emissions to the traffic related PM_{10} concentrations in ambient air (Gehrig et al., 2004b). However, quantitative information about the contributions of the individual processes leading to nonexhaust emissions (brake, road, and tire wear, vehicle-induced suspension of deposited road dust) is still scarce. What makes nonexhaust particle emissions in general more complicated to quantify than exhaust is that the former are strongly influenced not only by the vehicle type and traffic conditions, but also by differing material properties and meteorological factors. For more detailed information on processes and controlling factors for nonexhaust particle emissions, see Luhana et al. (2004) and Thorpe and Harrison (2008).

Harrison et al. (2012) estimated the relative contributions of brake wear, tire wear, and resuspension to nonexhaust traffic particles on a busy London highway. The sources were inferred from their size association and chemical signatures in airborne particles sampled into 10 separate size fractions in a street canyon, after subtraction of measurements from a nearby urban background site. An extensive field study providing real-world emission factors for the most important nonexhaust sources for different traffic situations has recently been performed in Switzerland (Bukowiecki et al., 2010). Two sites representing important traffic situations were investigated (urban street canyon with heavy stop-and-go traffic and along a national motorway). To

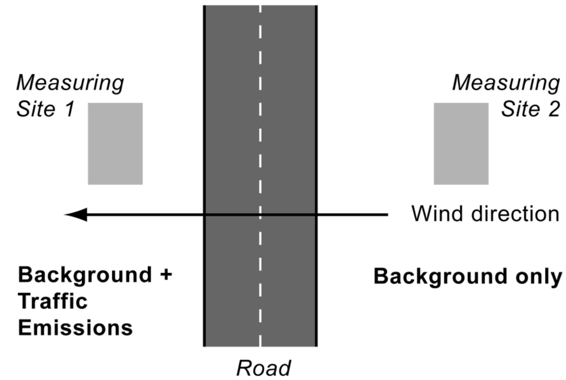

Figure 4. Schematic illustration of the upwind–downwind concept for specific measurements of the contributions of the local traffic.

separate local traffic-related PM_{10} emissions from the total ambient PM_{10} load, simultaneous additional measurements were performed either upwind of the selected measuring location or at a nearby background site (Figure 4). The concentration differences thus represented the contributions of local traffic only. The identification of individual traffic related sources was based on specific elemental fingerprint signatures for the various sources.

These fingerprints were obtained by hourly elemental mass concentration measurements in three size classes (0.1–1 μm, 1–2.5 μm, and 2.5–10 μm) using a rotating drum impactor (RDI) for sampling (Bukowiecki et al., 2009b) and synchrotron radiation x-ray fluorescence spectrometry for elemental analysis (Bukowiecki et al., 2008). For the identification and quantification of the different pollutant sources positive matrix factorization (PMF) was applied to the set of size-segregated and time-resolved elemental mass concentrations.

Emission factors cannot be obtained from concentration measurements alone because these are strongly influenced by changing traffic intensity and meteorological conditions. Using measured NO_x concentration differences (ΔNO_x) and the known exhaust emission factors for NO_x, the dilution factor, d, at any time could be calculated (eq 1) and can, together with traffic counts, be used to establish emission factors for any measured component emitted by the local traffic (eq 2).

$$d = \frac{EF_{NO_x,LDV} \cdot n_{LDV} + EF_{NO_x,HDV} \cdot n_{HDV}}{\Delta NO_x} \qquad (1)$$

$$c_x = EF_{x,LDV} \cdot \left(\frac{n_{LDV}}{d}\right) + EF_{x,HDV} \cdot \left(\frac{n_{HDV}}{d}\right) + C \qquad (2)$$

where $n_{LDV}, EF_{NO_x,LDV}$, and $n_{HDV}, EF_{NO_x,HDV}$ are vehicle frequencies (vehicles h^{-1}) and NO_x emission factors (mg km^{-1}) for light and heavy duty vehicles; c_x= measured mass concentration (μg m^{-3}) assigned to the considered source or emission process x; and C is the fitting constant.

Mass-relevant contributions from wear particles and suspended road dust were found mainly in the size range 1–10 μm. In the street canyon, the traffic-related PM_{10} emissions (light duty vehicles [LDV]: 24 ± 8 mg km^{-1} $vehicle^{-1}$, heavy-duty vehicles [HDV]: 498 ± 86 mg km^{-1} $vehicle^{-1}$) were assigned as 21% brake wear, 38% suspended/abraded road dust, and 41% exhaust emissions. Along the motorway (LDV: 50 ± 13 mg km^{-1} vehicle^{-1}, HDV: 288 ± 72 mg km^{-1} vehicle^{-1}), respective contributions were 3% brake wear, 56% suspended/abraded road dust, and 41% exhaust emissions. No indication for an important PM_{10} contribution of tire wear was observed (Figure 5).

Specific differentiation between PM_{10} emissions due to wear and suspension from the road pavement was not possible based on the field measurements at the traffic sites. This is mainly due to their similar elemental composition and highly correlated variation in time. However, mobile load simulators offered a possibility to tackle the wear versus suspension issue. These devices are designed and used by road engineers to test the properties and durability of road pavements in the field. In a recent study emission rates were derived from measurements on different types of road pavement (asphalt concrete, porous asphalt). The experimental setup allowed for a separate characterization of the emissions caused by fresh in situ wear and by suspension of previously deposited dust (Gehrig et al., 2010).

The measurements showed that compared to PM_{10} suspension "fresh" wear particle emissions from pavements in good condition are quite low in the range of only a few mg km^{-1} vehicle^{-1} if quantifiable at all. Considerable wear emissions, however, can occur from damaged pavements in poor condition. Suspension of deposited dust can cause high particle emissions, depending strongly on the dust load of the road surface (Figure 6). This finding is highly relevant for mitigation of pavement wear but based on limited data. A comparison study to validate the findings of this simulator is recommended.

Porous pavements seem to retain deposited dust better than dense pavements, thus leading to lower emissions from suspension compared to pavements with a compact surface structure (Figure 7). Vehicle-induced suspension is not strictly correlated with traffic counts but is also strongly influenced by available

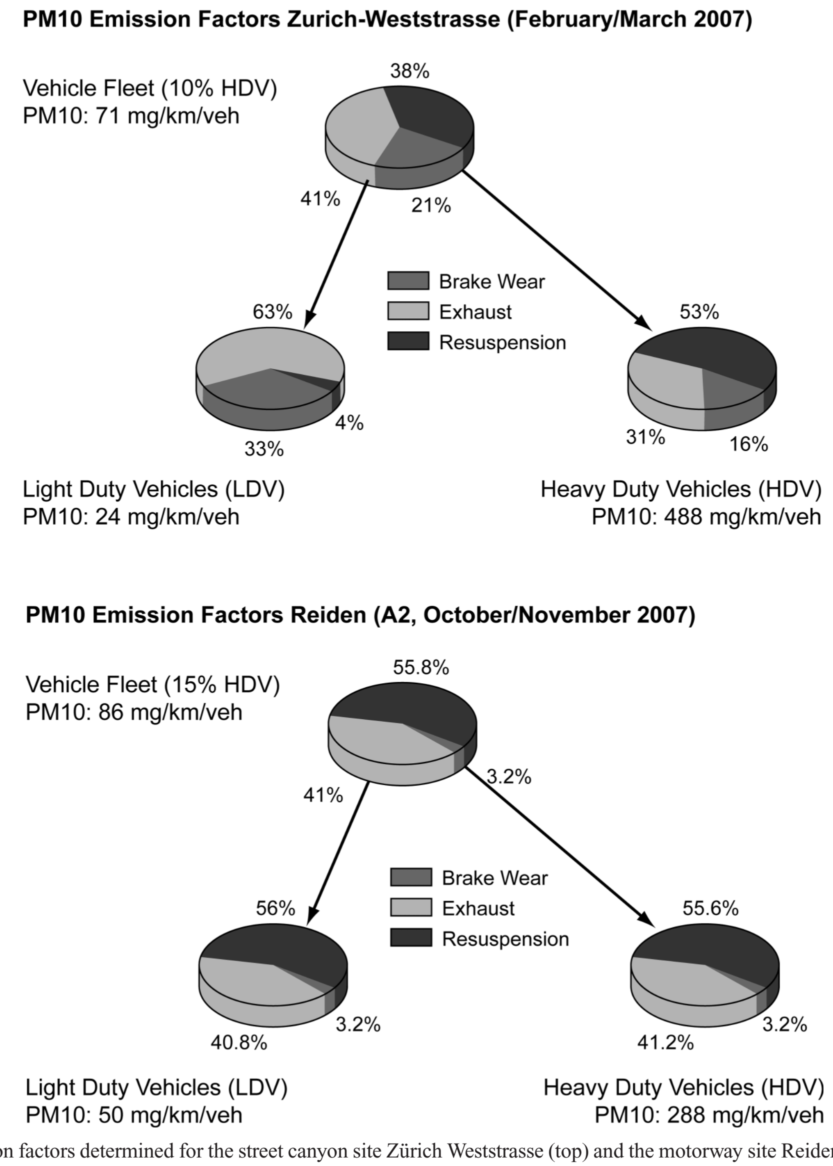

Figure 5. Traffic PM_{10} emission factors determined for the street canyon site Zürich Weststrasse (top) and the motorway site Reiden (bottom) and their composition.

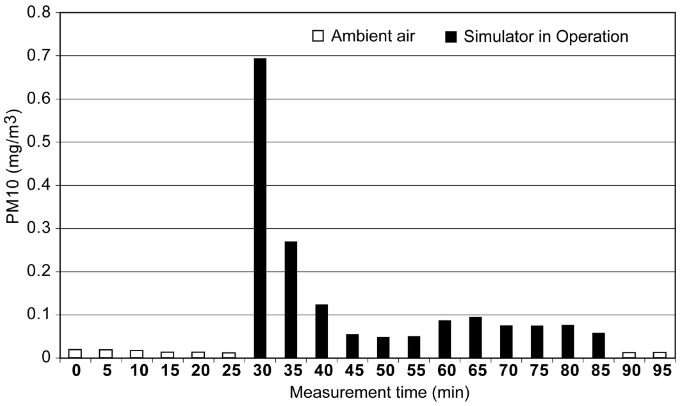

Figure 6. Trend of resuspended and abraded particles during an experiment with a mobile load simulator simulating heavy duty vehicles on a pavement in relatively poor condition with an already slightly damaged surface. Every bar in the plot represents 5 min measurement with a total of 500 double-wheel passages. High concentrations at the start of the simulator operation show the importance of resuspension.

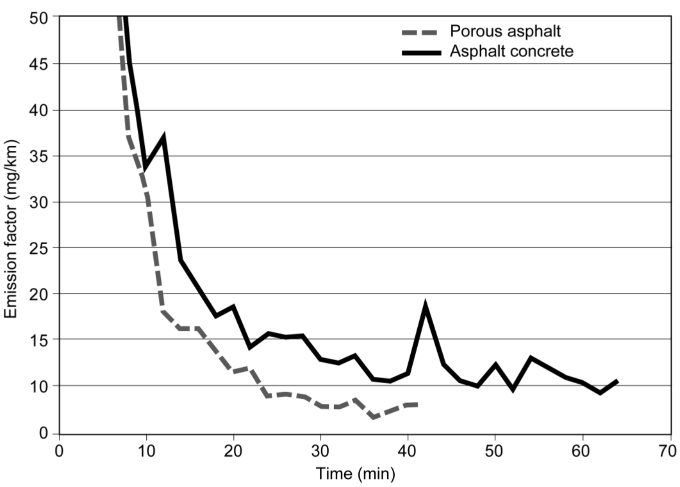

Figure 7. Emission factors as a function of time during a mobile load simulator wear experiment simulating light-duty vehicles comparing the resuspension of fine dust from an asphalt concrete pavement (AC11) and a porous asphalt pavement (PA11).

road dust. It is therefore difficult to describe with conventional emission factor models.

Exposure

Assessment of exposure to air pollution is often based on fixed site measurements provided by the local air quality monitoring network. Because of economic and practical reasons, personal exposure measurements are rare in epidemiological studies. Exposure to air pollution from traffic has been associated both with respiratory and cardiovascular effects (e.g. Hoek et al., 2002; Orru et al., 2009) with fine combustion-derived exhaust particles believed to be the most harmful (Laden et al., 2006; Brook, 2008). However, epidemiological studies differentiating between the coarse particles, PM2.5–10, and the fine particles, PM2.5, have shown that health effects arise from both fractions (Brunekreef and Forsberg, 2005; Meister et al., 2011), clearly highlighting the need to quantify exposure to nonexhaust particles if we are to fully understand the health effects of air pollution from traffic .

After generation and emission, the same meteorological processes that control atmospheric dispersion of exhaust particles also control dispersion of nonexhaust particles. Temporal variability of nonexhaust particle concentrations may, however, differ drastically from exhaust particles due to the influence of weather conditions on the suspension of nonexhaust particles. For example, road dust is suppressed if roads are wet, whereas exhaust particle emissions are unaffected by the road conditions. There may also be geographical differences in exposures to exhaust versus nonexhaust particles, depending on driving conditions, fleet composition, and road pavement variations.

Mitigation measures

Suspension of road dust particles is suppressed drastically if roads are wet. The use of dust suppressants or dust binders, e.g., aqueous solutions of magnesium or calcium chloride, or calcium magnesium acetate (CMA) that keep road surfaces wet has been shown to be efficient in reducing emissions of road dust in many cities in Sweden (Norman and Johansson, 2006), Norway (e.g. Aldrin et al., 2008), Finland (e.g. Kupiainen et al., 2011), Austria (Hafner, 2007), and the United Kingdom (Deakin and Ren, 2011). In Stockholm, use of magnesium chloride or CMA resulted in 20–40% lower PM_{10} levels on dry days (Norman and Johansson, 2006; Amato et al., 2010). On a rural road with lower traffic volumes and few other sources, the dust binder effect was similar, but also shown to have a longer duration than in a busy city street (Gustafsson et al., 2010). In Austria, where the road dust source is weaker, the maximum effect seemed to be a 10–20% decrease on a daily basis.

Vehicle speed increases road and tire wear and influences suspension of road dust. Concentrations alongside roads may be reduced due to increased dilution as vehicles increase atmospheric turbulence, making assessments of vehicle speed impact difficult based on concentration measurements alone. Some controlled field and lab tests have shown that increased vehicle speed increases road surface PM_{10} emissions (Gustafsson et al, 2009b). In Stockholm, when studded tires were used, regulation of vehicle speed was effective for reducing PM_{10} concentrations along highways (Johansson et al., 2009a).

Road sweeping alone, using traditional cleaning machines, did not show any significant reduction of PM_{10} concentrations in most studies (Keuken et al., 2010; Amato et al., 2010). Water flushing and/or dust binding in combination with sweeping have been shown to reduce PM levels, but partly this effect may have been due to the wetting of the surface (Amato et al., 2010). Many studies have shown that sweeping is efficient when it comes to reducing the dust load on the streets, which is likely to affect the particle emissions on a longer time scale. In tests with a known amount of well-characterized stone dust, it was shown that cleaning techniques including high-pressure water washing in combination with vacuuming the road surface might be efficient when it comes to collecting PM from the road surface (Gustafsson et al., 2011a). In normal street conditions the same study found indications of a positive effect on PM_{10} concentrations the day after sweeping, but only when data were filtered to show optimal conditions for sweeping effects. Clearly, more investigations of street cleaning efficiencies for reduction of PM emissions and concentrations are needed.

There are few field studies on the effect of different road pavements upon PM_{10} emissions and concentrations. Mobile measurements on pavements with different maximum stone size but the same mineral composition indicate that smaller stone size gives rise to higher emissions if studded tires are used, but the differences are small and more tests are needed (Johansson et al., 2007b). Laboratory studies using a road simulator showed that there are large differences between different pavement rock materials and maximum stone sizes, especially when using studded tires in the tests. Rocks with higher Nordic ball mill values (a technical wear test value) are more prone to emit PM_{10} than rocks with lower values (more wear resistant) (Gustafsson et al., 2011b). Likewise, using cement pavement may reduce PM_{10} emissions if studded tires are used (Johansson et al., 2009a). Measurements comparing impacts on concentrations or emissions of PM_{10} from porous asphalt as compared to traditional pavements have shown somewhat diverging results. Field studies based on both mobile and stationary measurements in Stockholm have not shown porous asphalt to give lower emissions of PM_{10} compared to traditional asphalts (Johansson et al., 2007b). Some unpublished results from tests in the Netherlands suggested that porous asphalts emit less PM, as a result of road dust moving with water into the pavement pores and being rinsed away. However, more data under varying conditions are needed to be conclusive.

In areas where studded tires are used, an effective measure to reduce nonexhaust contributions to PM_{10} was to reduce the use of studded tires. In Norway, massive reductions in the larger cities have been accomplished through a studded tire fee. Exceedances of PM_{10} limit values have been reduced as a result of this, but this was also the result of simultaneous improved dust binding, sweeping and traffic measures. Local bans of studded tires in certain streets are being evaluated in Sweden and the first two seasons of application of the measure seemed promising. PM_{10} concentrations have been reduced, but again, this has several reasons apart from a reduction in studded tires. Traffic counts have been reduced and extremely cold winters with snow have contributed to the reductions (Johansson et al., 2012).

Health Effects and Risk Assessment

Exposure to traffic-related PM plays an important role in the development of adverse health effects. Traffic-generated particles are associated with cardiovascular and pulmonary effects, as shown by both epidemiological and toxicological studies (Maynard et al., 2007; Schwartz et al., 2005, Riediker, 2007; Riediker et al., 2004a; Riediker et al., 2004b; Gerlofs-Nijland et al., 2007; Peters et al., 2004; Kelly and Fussell, 2011). Some of these studies suggest that brake wear particles might contribute to these adverse effects (Riediker et al., 2004b; Gerlofs-Nijland et al., 2007). In a study performed in North Carolina, highway patrol officers were followed during their work shift (Riediker et al., 2004b). Their health status was assessed with electrocardiograms and blood samples for inflammation and thrombosis factors. A comprehensive exposure assessment inside the cars included the collection on filters of fine particles ($PM_{2.5}$), for which the composition was analyzed and used for source apportionment. The source loadings were then compared to the health outcomes. This showed electrocardiogram and blood parameters to be correlated to a source that had a chemical fingerprint indicative of both brake wear and combustion particles emitted during load-change conditions (it was thus named "stop-and-go source"). A subsequent element-specific analysis (Riediker, 2007) suggested that copper contributed importantly to the observed health effects. In that study, brake pads were the only important source of copper (Riediker et al., 2003); thus, both the source apportionment and the element-specific health analysis were in support of the hypothesis that brake wear significantly contributes to the health effects of traffic particles.

Both tire wear and brake wear contain heavy metals that are known for their toxicity (Wallenborn et al., 2009; Jomova and Valko, 2011). This aspect of the chemical composition of wear particles in relation to health effects clearly warrants more study. A relatively toxic component of tire wear is zinc, while brake wear contains relatively high amounts of the transition metal copper (Gustafsson et al., 2008; Thorpe and Harrison, 2008). The toxicity of tire wear has mainly been tested in vitro using cell cultures rather than an intact organism (Gualtieri et al., 2005; Karlsson et al., 2006; Karlsson et al., 2008; Lindbom et al., 2006; Lindbom et al., 2007). Human alveolar lung cells (A549 cells) were exposed for various time periods (24, 48, and 72 hr) to different concentrations (50, 60, 75 μg/ml) of the organic fraction of pure tire wear particles (Gualtieri et al., 2005). Since it was suggested that toxicity may depend also on the presence of organic compounds, the solvent-extractable organic fraction of tire debris was studied. The organic extracts from tire wear induced a dose-dependent increase in cell mortality and DNA damage. The role of tire wear particles in an inflammatory response was also studied (Lindbom et al., 2006; Lindbom et al., 2007; Mantecca et al., 2010; Karlsson et al., 2006). The results showed that wear particles from studded tires and pavement can induce the release of proinflammatory cytokines from macrophages and human lung epithelial cells (Lindbom et al., 2006; Karlsson et al., 2006). The type of pavement is of importance for this effect. Particles generated from a granite pavement had a significantly higher capacity to induce the release of cytokines compared to quartzite pavement (Lindbom et al., 2006). Karlsson et al. (2011) have examined the toxicoproteomic effects on macrophages after exposure to tire wear particles to elucidate the pathways. This study showed the increase of proteins associated with an inflammatory response, while proteins involved in cellular functions and glycolysis were reduced. Only a few in vivo studies have reported the toxicity of tire wear particles (Gottipolu et al., 2008; Mantecca et al., 2009; Mantecca et al., 2010). The study of Gottipolu et al. (2008) showed that rats exposed to respirable (<5.0 μm) tire wear particles by intratracheal instillation developed acute pulmonary inflammation. This inflammatory response, which was measured by the number of neutrophils in broncheoalveolar lavage fluid, was a reversible effect and not present at 1 week after exposure. The observed acute pulmonary toxicity of tire wear was related to the levels of water-soluble zinc and copper. However, copper is normally not a major component of tire wear but rather of brake wear (Denier van der Gon et al., 2007; Thorpe and Harrison, 2008) as presented in a previous section. Hence, the high amounts of copper in the studied tire wear samples in the study of Gottipolu et al.(2008) could imply a contamination with brake wear particles. The effect of size-fractionated tire particles smaller than 10 μm (tire-PM_{10}) and smaller than 2.5 μm (tire-$PM_{2.5}$) was examined by Mantecca et al. (2009; 2010). Mice were exposed by intratracheal instillation and the proinflammatory and toxic effects were studied 24 hr after exposure. Lung toxicity induced by tire-PM_{10} was primarily due to macrophage-mediated inflammatory events, while toxicity induced by tire-$PM_{2.5}$ appeared to be related more strongly to a direct cytotoxic effect (Mantecca et al., 2009). In a second study with a similar setup Mantecca et al. (2010) compared the acute inflammatory responses of tire particles from two size fractions 3 hr after exposure to elucidate the time course of the lung inflammatory process. While Mantecca et al. (2009) showed that both size fractions induced an inflammatory response, which was still present 24 hr after instillation, the time-course study revealed that coarse particles do not induce an early recruitment of neutrophils (Mantecca et al., 2010).

The health effects of brake wear particles are even less studied than tire wear particles. Perrenoud et al. (2010) developed an experimental setup in which cell cultures could be exposed to freshly generated brake wear particles under controlled conditions. Measurements on particle number and mass concentration from different brakes showed that large numbers of fine and ultrafine particles are released during braking, especially when full stops were simulated. The air-to liquid exposure system developed by Perrenoud et al. (2010) is likely to correspond to reasonable exposure conditions, as also suggested by the absence of any significant cell mortality as assessed with the lactate dehydrogenase (LDH) test (Gasser et al., 2009). Epithelial lung cells were directly exposed for 10 min to particles emitted from two typical braking behaviors (“full stop” and “normal deceleration”). Both the generation of reactive oxygen species (ROS) and the carbon concentration (total and organic carbon) were significantly and positively associated with the inflammation marker interleukin (IL)-8. In addition, a significant decrease of the protein occludin was observed in correlation to exposure to the metals iron, manganese, and copper. Coloring specific for occludin suggested that this tight-junction protein got fractionated. This suggests that the presence of brake wear particles leads to damage to tight junctions well before any significant cell mortality occurs. The authors propose as mechanism the formation of ROS on the particles’ surface, which then led to oxidative damage to occludin and a cellular inflammation response. The ROS-generation potential of metal and metal oxide particles is the subject of ongoing research. Initial results suggest that copper and copper oxide particles are very potent in generating ROS.

Some studies have not only examined the toxicity of wear particles but included the relative toxicity of different sources (Karlsson et al., 2006; Karlsson et al., 2008; Lindbom et al., 2006; Lindbom et al., 2007; Mantecca et al., 2010). Tire/road wear particles were able to cause DNA damage of the same magnitude as particles collected from wood combustion and an urban street (Karlsson et al., 2006). In addition, tire/road wear particles could induce an inflammatory response, although to a lesser extent than urban street particles. However, the study of Lindbom et al. (2006) showed that depending on the type of pavement used and inflammatory marker studied, a similar inflammatory response could be observed. In this study the response of particles from tire/road wear and a traffic-intensive street seemed even of a greater magnitude than that of particles collected directly from the exhaust of a diesel engine. Particles from studded tire/road wear could not only induce an inflammatory response but also are able to induce NO, lipid peroxidation and ROS formation (Lindbom et al., 2007).

With respect to road wear and resuspension, a recent study in Stockholm found significant effects of coarse particles on premature mortality of the population, especially during winter and spring in connection with high emissions of road dust (Meister et al., 2011). The study of Meister et al. (2011) indicates an increased mortality of 1.7% per 10-μg/m^3 increase of coarse particles.

In conclusion, current epidemiological and toxicological (mainly cell culture) research suggests that wear particles lead to negative health effects. The formation of ROS could play an important role, which subsequently could lead to inflammatory and cardiovascular responses. It also suggests that wear particles are an important element in traffic-related particle toxicology and thus an important contributor to adverse health effects associated with exposure to traffic emissions. However, current data do not yet allow quantification of, for example, numbers of deaths or diseases attributable to wear particles. Further research seems not only warranted but urgently needed.

Workshop Outcomes

There is a clear lack of data in the field of road transport wear and suspension emissions to conclusively assess its importance for air quality and the impact on human health. It is uncertain to what extent nonexhaust emissions contribute to ambient concentrations of PM_{10} or $PM_{2.5}$ and how the problem of “nonexhaust” relates to “exhaust” in a relative sense, in terms of both emissions and air quality and in terms of human health. Although generalized mass fractions are available for nonexhaust and exhaust road transport PM emissions, the regional, spatial, and temporal patterns of the former are least known, while the role of suspension versus primary wear emissions is even less characterized. As the trend toward cleaner technologies with reduced exhaust emissions continues through the use of catalytic converters, diesel particulate filters (DPF), and improved fuels and engines, nonexhaust PM will soon surpass exhaust emissions and may well become dominant by 2020 both in terms of emissions and contributions to air quality. The majority of the experts participating in the workshop (55%) ranked the importance of wear and suspension emission compared to exhaust emissions in the future (2020 and beyond) as dominating in terms of PM mass (Figure 8).

It was agreed that nonexhaust emissions are health relevant, also based on material presented during the workshop, but a major knowledge gap is the relative toxicity and health impact of nonexhaust emissions versus exhaust emissions. Tailpipe particle

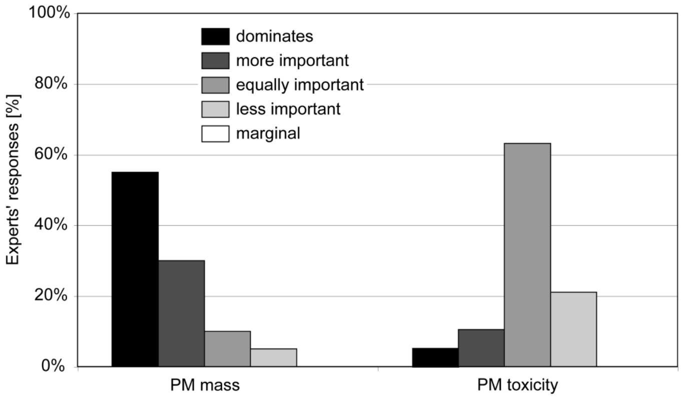

Figure 8. Experts' indications: How will you rank the importance of wear and resuspension emission compared to engine emissions in the future (2020 and beyond) in terms of (a) PM mass and (b) PM toxicity? (Total expert responses: $n = 20$.)

emissions may be thought of as presenting a greater risk for adverse health impacts due to their smaller size and chemical nature, including soot and PAHs, but no firm evidence has been identified to split the contribution to health effects between exhaust and nonexhaust. Furthermore, nonexhaust particles consist of a high fraction of transition metals, which are seen as key players in the production of reactive oxygen species (ROS) causing oxidative stress (e.g., Huang et al., 2004; Charrier and Anastasio, 2012). Based on our current knowledge, no firm evidence on the relative risk of exposure to exhaust versus nonexhaust particles could be established. In the workshop, 63% of experts indicated that nonexhaust PM toxicity may be equally important to exhaust particles based on various studies (e.g., Gustafsson et al., 2008; Lindbom et al., 2006; Lindbom et al., 2007 and Karlsson et al., 2011). A further complication in identifying the unique contribution of exhaust and nonexhaust particles to health effects is the near-perfect correlation between both sources in time and space. There is clearly a need for more studies examining both nonexhaust and exhaust emissions from road transport, the linkage between these emissions, and their separate health impacts.

In theory it might be possible to separate primary wear emissions from resuspension emissions, for example, using tracers or tracer ratios, and to quantify their relative contribution to air quality and/or human health effects. Seventy-six percent of the experts indicated that wear and resuspension emissions can be separated for their relative contribution to air quality, and 55% pointed out that it is possible in terms of human health effects. In a qualitative manner, wear emissions can be separated from resuspension emissions by identifying the distinct chemical marker of particles from brake wear, tire wear, and road dust suspension. However, in the real-world situation the separation is difficult due to the lack of unique chemical tracers of wear emissions and resuspension emissions; it should be borne in mind that wear particles also contribute to resuspension emissions. It was generally agreed that more evidence on the linkage between resuspension and wear emissions and adverse health effects needs to be gathered.

Additional information on the questions addressed during the workshop and the subsequent discussion and conclusions can be found in the supporting material and the workshop report (Denier van der Gon et al., 2012).

Consensus Statements, Conclusions, and Recommendations

Participants agreed that we need to improve our knowledge about emission, exposure, and health effect of wear particles since this fraction of PM cannot be neglected and its relative and absolute importance is still increasing. It is important to stress to regulators and policymakers that road transport emissions continue to be an issue for health and air quality, despite the encouraging rapid decrease of tailpipe exhaust emissions.

The following conclusions were drawn:

- Wear particles, direct and resuspended, are a concern in the context of meeting current PM standards in many road and street environments. In the Scandinavian countries in winter and early spring, the problem is especially pronounced due to the use of studded tires and extensive winter road operations. Also, wear particles may contribute to exceedances of other standards, such as those for heavy metals. Quantitative information from which to estimate the local and regional contributions of wear particles to airborne concentrations is either unavailable or highly uncertain, which seriously impedes modeling. Epidemiological studies have an important impact on policymaking, but it should be realized that it is very difficult to separate health effects from exhaust versus nonexhaust particles in such studies because the correlation between both emission types is extremely high.
- The group agreed that the health risk associated with wear emissions may not be neglected and that at present insufficient evidence is available. It is valuable to put the hazard of wear particles in perspective in relation to, for example, engine exhaust emissions. Improved insight is needed into exposure concentrations and durations to allow epidemiological studies and to link hazard and exposure to estimate the health risks.

This resulted in the following recommendations:

- More toxicological and epidemiological evidence is needed to identify the urgency of tackling wear emissions and guide policymakers. Proper toxicity comparison expressed in comparable units between exhaust and nonexhaust is crucial information but currently lacking.
- A standardized approach to assess hazard of, and exposure to, wear emissions in Europe is needed to compare results from the various studies.
- To be able to define effective and necessary future policies we need to create (research) opportunities to assess the health impact of wear particles so as to put this in the perspective of cost effective abatement measures.
- Analytical methods to properly disentangle the PM of wear sources from other PM fractions are warranted, for example, by use of appropriate markers to allow source apportionment.
- Within the mixture of wear PM, different fractions can be identified for which the hazard is not clear. Simple measures such as chemical reactivity (redox or oxidative potential) may provide insights into both the chemical composition as well as the hazard of various (sources of) wear particles.

To tackle the aforementioned issues, there is an urgent need for a comprehensive research program and work plan. Dedicated funding from the national governments and European Union is necessary that explicitly encourages collaboration between the relevant disciplines, rather than competition for funding. A full consensus about what is most important will never be reached. For example, in terms of priority setting, brake wear appears more important than tire wear but there is no full agreement on this. Some workshop participants argued that the information on tire wear is just too limited. Furthermore, policymakers should be informed about potential no-regret measures, existing possibilities to change brake/tire composition (e.g., ceramic brakes have fewer emissions), and the importance of road maintenance.

In light of the continuous increase of the relative contribution of nonexhaust emissions to ambient PM, where it is becoming the dominant emission process for urban transport, it is more than timely to devote greater effort to properly quantifying nonexhaust emissions and assessing their health relevance.

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