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Non-Exhaust Vehicle Emissions of Particulate Matter and VOC from Road Traffic: A Review

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ABSTRACT

As exhaust emissions of particles and volatile organic compounds (VOC) from road vehicles have progressively come under greater control, non-exhaust emissions have become an increasing proportion of the total emissions, and in many countries now exceed exhaust emissions. Non-exhaust particle emissions arise from abrasion of the brakes and tyres and wear of the road surface, as well as from resuspension of road dusts. The national emissions, particle size distributions and chemical composition of each of these sources is reviewed. Most estimates of airborne concentrations derive from the use of chemical tracers of specific emissions; the tracers and airborne concentrations estimated from their use are considered. Particle size distributions have been measured both in the laboratory and in field studies, and generally show particles to be in both the coarse ($PM_{2.5-10}$) and fine ($PM_{2.5}$) fractions, with a larger proportion in the former. The introduction of battery electric vehicles is concluded to have only a small effect on overall road traffic particle emissions. Approaches to numerical modelling of non-exhaust particles in the atmosphere are reviewed. Abatement measures include engineering controls, especially for brake wear, improved materials (e.g. for tyre wear) and road surface cleaning and dust suppressants for resuspension. Emissions from solvents in screen wash and de-icers now dominate VOC emissions from traffic in the UK, and exhibit a very different composition to exhaust VOC emissions. Likely future trends in non-exhaust particle emissions are described.

KEYWORDS: non-exhaust emissions; road traffic; particulate matter, wear, resuspension, emissions, volatile organic compounds

1. INTRODUCTION

Road traffic has long been viewed as a major contributor to urban air pollution. The advent of three- way catalytic converters on gasoline vehicles has led to a huge reduction in exhaust emissions of carbon monoxide, hydrocarbons and NO_x (Winkler et al., 2018). Reductions in emissions from diesel exhausts came much later, and these remain a major source of particulate matter and NO_x pollution in cities globally (Harrison et al., 2021). Diesel Particle Filters are a highly efficient means of controlling particulate matter emissions from diesels, and the latest particle number emissions standards in Europe require the use of Gasoline Particle Filters. Consequently, particulate matter emissions from traffic exhaust in developed countries, where new vehicles have to adopt these technologies, have declined rapidly, and as a consequence, non-exhaust emissions from road vehicles now widely exceed exhaust emissions (AQEG, 2019). According to estimates reported by OECD (2020), the total amount of non-exhaust particulate matter ($PM_{2.5}$) emitted by passenger vehicles worldwide is likely to rise by 53% by 2030 from a 2017 baseline, mainly due to increased vehicle mileage.

Non-exhaust emissions of particles arise mainly from four separate sources. Three of these arise from abrasion: brake rear, tyre wear and road surface wear. The fourth, particle resuspension, arises from road dust particles being suspended into the atmosphere by passing traffic. There are also emissions of VOC from use of screenwash, which are rarely considered. All of these emission types are generated by battery-electric vehicles as well as internal combustion engine vehicles, and although their magnitude may change, road traffic will continue to be a source of particulate matter and VOC emissions, even for a fully electric vehicle fleet.

Measurements of non-exhaust emissions come from both controlled laboratory studies and field experiments. Brake wear, tyre wear and road surface wear can all be evaluated in the laboratory. Brake wear can be determined under highly controlled conditions using a brake dynamometer, and standardised methods allowing comparative studies of materials are under development. Methods for evaluating tyre and road surface wear are more difficult to standardise and there are a range of test methods, with relatively few data available. Such studies are, however, important as they are the basis for deriving the emissions factors which form the building blocks for emissions inventories, and are essential for numerical modelling of airborne concentrations. Resuspension is more difficult to estimate. It is often not included in emissions inventories, and most estimates are based upon algorithms using vehicle mass and road surface dust loading as input data. Non-exhaust particles can also be measured in the atmosphere by use of chemical tracers, and by using receptor modelling methods such as Positive Matrix Factorization. This is challenging and relatively few data exist, although these have increased recently.

Past reviews of this topic, or specific aspects of it, have previously been published by Thorpe and Harrison (2008), Grigoratos and Martini (2015), Amato et al. (2014) and AQEG (2019). It is however, a topical and rapidly moving field, and hence we feel it valuable to provide a brief review of the latest state of knowledge.

2. INVENTORIES OF NON-EXHAUST EMISSIONS

Emission inventories for many countries include estimates of emissions of PM_{10} and $PM_{2.5}$ from tyre wear, brake wear and road abrasion and, in some cases, for road dust resuspension. The methods used are mainly suited for estimating emissions on a national or regional scale as they simplify the effect of vehicle size, type and technology, driving style and road condition. This is normally done out of necessity when national statistics on vehicle population and mileage and travel on different types of roads at different average speeds are used to estimate emissions on a large scale. These approaches are less well suited to estimating emissions on a particular road section at a particular time where emissions are expected to be highly variable.

In Europe, countries report emission inventories annually to conform with guidelines set out by the European Environment Agency. This is to ensure consistency and comparability in inventories reported by EU Member States. The EMEP/EEA Emissions Inventory Guidebook (EMEP/EEA, 2019) provides methods and emission factors for estimating emissions from tyre wear, brake wear and road abrasion for different vehicle types. Emission factors are in mg/km and correction factors are provided to account for different average speeds for mileages done on urban, rural and highway roads and different vehicle loads in the case of heavy duty vehicles. Emissions from resuspension of previously deposited material are not required to be reported and no method is provided in the Guidebook. This is partly because this would be considered a double-count of an emission for accounting purposes, though it is recognised that this source does have to be considered when considering the full impact of traffic on ambient concentrations of PM in urban environments.

The emission factors provided in the EEA Guidebook are mostly based on analysis of data from literature sources of nearly 20 years ago on wear rates for tyre and brake material combined with estimates of the quantities that become airborne in different particle size ranges, with some data from direct measurement using a simulated wheel or brake operation in the laboratory. The factors may not well represent emissions from modern vehicle technologies and tyre and brake systems and materials. Countries are permitted to use country-specific factors if these can be justified and take account of national circumstances. For example, the Netherlands and Sweden consider a smaller proportion of tyre wear and brake wear PM emitted in the $PM_{2.5}$ range relative to PM_{10} compared with the values given in the Guidebook. Sweden and Finland apply a larger emission factor for PM_{10} from road abrasion to account for the effect of studded tyres relative to non-studded tyres and use a much smaller $PM_{2.5}/PM_{10}$ ratio for this source to reflect the high proportion of these emissions in the coarse particle range (see for example, Finland's 2020 Informative Inventory Report for Transport at https://www.environment.fi/en-

US/Maps_and_statistics/Air_pollutant_emissions/Finnish_air_pollutant_inventory_to_the_CLRTAP). Further details on emission factors used in Europe are given in AQEG (2019).

Without any regulation on non-exhaust emissions or understanding of how modern vehicle technologies have changed emissions, the emission factors used in Europe remain constant over time. As a consequence, all inventories show emissions from these sources increasing over time as the numbers of vehicles and kilometres travelled have increased. In contrast, exhaust emissions in most countries have been decreasing as newer vehicles meeting tighter emission standards enter the fleet, particularly in recent years as increasing numbers of diesel vehicles are fitted with particulate filters.

Figure 1 shows the trend in exhaust and non-exhaust emissions of $PM_{2.5}$ for all EU28 countries combined from 2000-2018 taken from inventories officially submitted by the Parties to the UNECE Convention on Long-range Transboundary Air Pollution (EEA, 2021). Inventory data submitted by countries combine emissions from tyre and brake wear in a single category, but road abrasion emissions are reported separately. Whilst there are some differences between individual countries, most are showing the same general trends as Figure 1. For most countries, each of the three non-exhaust sources contribute roughly similar amounts and there is no dominant source, though countries such as Sweden and Finland have a much greater contribution of road abrasion to the PM_{10} inventory.

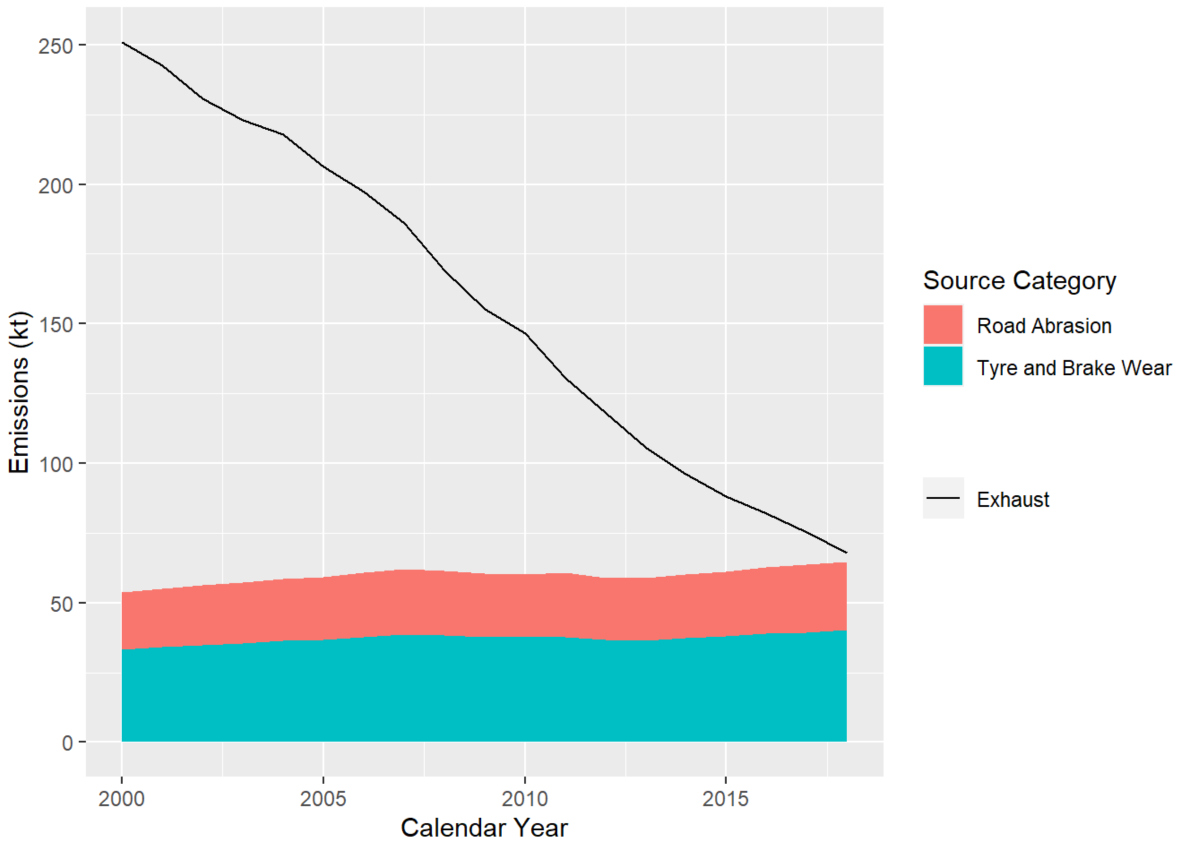

Figure 1: Total exhaust and non-exhaust road transport emissions of $PM_{2.5}$ from EU28 countries from emission inventories reported by Parties under the Convention on Long-range Transboundary Air Pollution in 2020, covering years up to 2018 (EEA, 2021).

Figure 2 shows non-exhaust emissions of $PM_{2.5}$ and PM_{10} for EU28 countries combined as a percentage share of total road transport and of all national primary emissions. These trends imply that the share of road transport emissions from non-exhaust sources has been increasing with time, rising from 18% in 2000 to 49% in 2018 in the case of $PM_{2.5}$ and 31% to 66% in the case of PM_{10}. As a share of all primary emissions in the EU28, non-exhaust emissions increase from 2.9% in 2000 to 5.2% in 2018 in the case of $PM_{2.5}$ and 4.0% to 6.7% in the case of PM_{10} .

Figure 2: Percentage share of total non-exhaust emissions of $PM_{2.5}$ and PM_{10} for EU28 countries combined, as a percentage share of total road transport and of all national primary emissions (shown in the figure marked 'Total'). Based on data from EEA (2021).

However, the range in the contribution made by non-exhaust emissions to total road transport emissions between countries is very wide, according to the official inventories of EU28 Member States (EEA (2021), these include the UK for the purpose of these comparisons). For $PM_{2.5}$, the range in 2018 varies from 11% to 87% and may reflect the different transport activities in each country as well as country-specific circumstances, such as the use of studded tyres in Sweden or it may reflect the use of different emission factors. The range in the $PM_{2.5}$ share in emissions is smaller for the largest European countries (UK, France, Germany, Spain, Italy), from 42% to 67% of all road transport emissions in 2018.

The National Emissions Inventory for the U.S. includes emissions of PM_{10} and $PM_{2.5}$ from tyre and brake wear and resuspension. Emissions from tyre and brake wear are included with exhaust emissions in the 'On-road – Mobile' category and cannot be viewed separately; emissions from resuspension are reported separately under 'Paved road dust'.

Tyre and brake wear emissions are estimated by the USEPA MOVES tool (USEPA, 2020a). Tyre wear emissions in MOVES are derived from wear rates correlated with average vehicle speed, similar to the approach used in Europe. It assumes that 8% of tyre wear is emitted as PM_{10}, 1.5% as $PM_{2.5}$. This leads to an average PM_{10} factor for passenger cars of 5 mg/km which is comparable to the values used in Europe, but the average value for trucks is lower than European values at less than 20 mg/km. This may be due to different assumptions about vehicle sizes and average speed and how tyre wear emissions are correlated. The US approach implies a much smaller tyre wear $PM_{2.5}/PM_{10}$ ratio of 0.15 on average (USEPA, 2020a), compared with 0.7 in Europe (EMEP/EEA, 2019).

MOVES uses g/hour emission rates for brake wear based on brake dynamometer and wind tunnel measurements at different deceleration rates for US-specific brake materials (USEPA, 2020a). These are combined with estimates of the amount of braking done at different deceleration levels for different real-world vehicle operating modes defined in terms of vehicle specific power, a more detailed approach than the Guidebook approach used in Europe. The average brake wear emission factor for PM_{10} calculated in MOVES for passenger cars in the US inventory is quoted as 14 mg/km which is higher than the UK average of 7mg/km derived from the UK's national inventory (NAEI, 2020) using factors from EMEP/EEA (2019), but MOVES implies a much smaller brake wear $PM_{2.5}/PM_{10}$ ratio of 0.12 on average (USEPA, 2020a), compared with 0.4 in Europe (EMEP/EEA, 2019).

Emissions from resuspension in the U.S. inventory, categorised under the term 'paved road dust' are calculated in a method described in the USEPA's AP-42 compilation manual (USEPA, 2011).

A near-linear empirical equation is used that relates an emission factor in g/km to vehicle mass and silt loading of the road surface. A correction is made for the number of precipitation days in the model period. Road surface and environmental conditions are highly variable and the factors from this approach are highly uncertain and not necessarily universal as it applies only to the range of variables from which the equation was derived. The applicability to other conditions of vehicle speed, mass and road conditions may be limited. AP-42 provides an alternative equation for unpaved roads.

Comparing non-exhaust emissions from on-line national inventory data sources for the U.S. and Europe is difficult on a like-for-like basis because the scope of coverage and accessibility of data are different. Europe shows tyre and brake wear emissions (combined) and road abrasion emissions separately, but excludes resuspension; the U.S. includes resuspension (paved road dust), but combines tyre and brake wear with exhaust emissions. A report by Panko et al. (2013) states that according to the USEPA, tyre wear contributed 15% of total vehicle emissions of PM_{10} in 2008 which compares with a value of 17% in the UK for the same year (NAEI, 2020). On-line inventory data are available for the State of California where emissions for all sources can be seen with tyre and brake wear and resuspension (paved road dust) viewed separately. Combining data extracted from the CARB mobile emissions data source EMFAC (CARB, 2017) and the emissions tool CEPAM (CARB, 2016) for all other sources in 2018, the percentage contributions of tyre and brake wear and resuspension relative to all road transport emissions and as a percentage of all California emissions can be derived.

Table 1 compares these contributions for the UK, Europe, California and the U.S. as far as possible on a like-for-like basis. A comparison between the UK, Europe and California excludes resuspension as this is not covered in European inventories. Only emissions for resuspension can be shown for the U.S. The table shows comparisons in contributions as a percentage of all road transport emissions in the country/region and as a percentage of all primary PM emissions for the year 2018, apart from the U.S. where the most recent data is for 2017.

Table 1: Contribution of non-exhaust sources to road transport and all primary sources of PM emissions in Europe and the U.S. These are according to current estimates from emission inventories for the respective regions, with comparisons done on a like-for-like basis, as far as is possible. Data are derived for the UK from NAEI (2020), for EU28 from EEA (2021), for California from CARB (2016) and CARB (2017) and for the USA from USEPA (2020a).

				$PM_{2.5}$	**PM_{10}**
UK	2018	Tyre, brake, road abrasion	as % of all road transport emissions (exc resuspension)	67%	79%
		Tyre, brake, road abrasion	as % of all primary source emissions (exc resuspension)	8%	9%
EU28	2018	Tyre, brake, road abrasion	as % of all road transport emissions (exc resuspension)	49%	66%
		Tyre, brake, road abrasion	as % of all primary source emissions (exc resuspension)	5%	7%
California	2018	Tyre & brake	as % of all road transport emissions (exc resuspension)	71%	85%
		Tyre & brake	as % of all primary source emissions (exc resuspension)	7%	5%

				$PM_{2.5}$	**PM_{10}**
USA	2017	Resuspension	as % of all road transport emissions	65%	79%
		Resuspension	as % of all primary source emissions	4%	5%
California	2018	Resuspension	as % of all road transport emissions	46%	72%
		Resuspension	as % of all primary source emissions	7%	11%

Emissions as a fraction of all primary emissions in the country/region will reflect the quantity of emissions from non-transport sources which can be quite variable. Even so, it is clear to see that resuspension alone makes a large contribution to area-wide emissions from road transport of $PM_{2.5}$ and PM_{10} in the U.S., according to the inventories. In Europe, where emissions from resuspension are excluded from inventories, it is clear how even emissions from the other non-exhaust sources make large contributions to total road transport emissions of PM, often more than 50% in 2018, and this is consistent with the situation in California where the contributions from tyre and brake wear may be even greater. The inventory for California suggests that when all non-exhaust sources are accounted for, including resuspension, they contribute to 96% of all emissions of PM_{10} from road traffic and 15% of primary PM_{10} emissions from all sources in 2018 (CARB, 2016, 2017).

There are a few reported estimates of non-exhaust emissions from road transport in China. Zhang et al. (2020) recently conducted a comprehensive study combining tunnel measurements, laboratory dynamometer and resuspension experiments and chemical mass balance modelling to estimate road dust resuspension, tyre wear and brake wear emissions in four Chinese megacities. The experiments yielded emission factors for $PM_{2.5}$ not dissimilar to the factors used in Europe for tyre and brake wear, but the authors acknowledged their factors could be underestimates, not typical of most traffic in China because of the smooth traffic flow and low amount of braking in the tunnels. The authors also concluded a total inventory for non-exhaust emissions of $PM_{2.5}$ in China of 11.4 ktonnes based on measurements done in 2017/18, of which 8.1 ktonnes were from resuspension (Zhang et al, 2020). Although it is not clear whether this value only applied to light duty traffic, the value seems low when one considers the tyre and brake wear and road abrasion emissions alone in the UK were 8.7 ktonnes in 2019 (NAEI, 2020), emissions from resuspension on paved roads were 209 ktonnes in the U.S. in 2017 (USEPA, 2017) as well as the fact this would be small relative to estimates of total road transport emissions of PM_{10} in China ranging from 40 – 400 ktonnes in 2008, depending on inventory source (Saikawa et al., 2017).

An air pollutant emissions inventory is available for road transport in 2013 in Australia (Smit, 2014), developed using the same method as used in Europe, from the EMEP/EEA Emissions Inventory Guidebook. The inventory suggests non-exhaust emissions were responsible for 26% of all road transport emissions of $PM_{2.5}$, similar to the contributions in Europe for 2013 (EEA, 2021).

The OECD has published a highly detailed review of non-exhaust emissions of PM which considers the causes, consequences and policy responses to tackle these emissions globally (OECD, 2020).

The report considered the limitations of emission inventories, including the inclusion or otherwise of resuspension, and reviews inventories for different regions of the world, mainly Europe and the U.S., but also in parts of Latin America. Inventories for Mexico, Bogota in Colombia, and Santiago in Chile, all highlight the importance of road dust resuspension with contributions to total primary emissions from all sources ranging from 16% to 54% (PM_{10}) and 9% to 56% ($PM_{2.5}$) in these regions. The contribution of tyre and brake wear emissions to total emissions in Bogota was much less than that, at 1.4% for PM_{10} and 0.8% for $PM_{2.5}$.

In a review of tyre wear and its impact on the environment, Kole et al. (2017) estimated the global emission of tyre wear to be 5.9 million tonnes per year. On the assumption that 3-7% of this becomes airborne $PM_{2.5}$ emissions (Kole et al., 2017; Grigoratos and Martini; 2014), a global emission rate of 175-410 ktonnes $PM_{2.5}$ from tyre wear is implied.

The OECD conclude that globally in 2014 at least 50% of total traffic emissions of PM_{10} and 5% of primary PM_{10} from all sources were non-exhaust emissions, even when resuspension is excluded. Corresponding figures for $PM_{2.5}$ are 34% and 4%, respectively. When resuspension is taken into account, non-exhaust emissions represent 95% of all road traffic emissions and 15% of all source emissions, as stated earlier for California.

Non-exhaust sources do not only contribute to airborne PM_{10} and $PM_{2.5}$, but also various metals and polyaromatic hydrocarbons (AQEG, 2019). Emissions are generally related to the composition of materials used in tyres and brake linings. Inventories include emissions of several different metals and PAH species based on factors from fairly old literature sources, though some countries use rates based on more recent composition studies. Some countries take account of the recent prohibition of "PAH-rich" extender oils used in tyre production leading to a reduction in PAH emissions from tyre wear (AQEG, 2019).

There are large uncertainties in emission inventories for non-exhaust sources. This is in part due to the difficulty in measuring emissions, but also because of the high variability in emissions and the factors which influence them. These are referred to in the studies mentioned above and in Guidebooks and reports describing inventory methodologies. Emissions depend on driving style, speed and the amount of acceleration and deceleration and cornering, weight and load of the vehicle, the design of the brake system, type and composition of tyre and brake materials, the condition and roughness of the road surface and ambient temperature, precipitation and humidity. Inventories necessarily approximate and average out their effects or parameterise them in fairly simple ways. Emissions from brake wear are not continuous but are inevitably greatest where braking events are most significant. Methods have been developed to map brake wear emissions at high spatial resolution where there are areas of high braking intensity according to changes in vehicle specific power based on real-world vehicle measurements data associated with different road situations (FAT, 2019). However, these methods have not yet been applied to inventories.

3. PHYSICAL PROPERTIES OF NON-EXHAUST EMISSIONS

3.1 Brake Wear

Brake wear particles arise from wear of both the brake lining (pad) and the rotor (disc) or drum. There are three major types of brake lining, referred to as non-asbestos organic (NAO), low metallic (LM) and semi-metallic (SM) (Kukutschova and Filip, 2018), each with a different composition and favoured in different markets. Ceramic linings are also used on some of the more expensive vehicles (Ostermeyer and Muller, 2008). The lining is made up from four main components: binders, fibres, fillers and friction modifiers, and according to Roubicek et al. (2008), more than 3000 materials are used across brands. This is reflected in a very wide range of chemical composition exemplified by the study of Hulskotte et al. (2014). The compositional differences affect the rate of particle emissions (Kim et al., 2020), and most probably the physical properties of emissions, but this has not been systematically studied. At normal brake operating temperatures

(<150ºC), brake wear particles are formed by abrasion due to friction between the pad and rotor or drum. They are irregular in shape with strong angular features, and contain multiple elements in the same particle (Wahlstrom et al., 2010a). Exceptionally, at higher temperatures, ultrafine particles are formed, and appear broadly similar in shape to the larger particles (Wahlstrom et al., 2010a).Many studies have been conducted under controlled conditions, giving a range of results for median diameters, which may arise from different designs of measurement system, or different brake materials. In tests of various brake linings including both NAO and SM types using a brake dynamometer, Garg et al. (2000) reported highly varied particle mass size spectra, which were attributed by Sanders et al. (2003) in part to particle losses in the sampling system. Garg et al. (2000) reported that between 7 – 48% of the particles became airborne, and at a brake temperature of 100ºC, of the particles sampled by a MOUDI impactor, 80%, 68% and 26% of mass was below 10 µm, 2.5 µm and 0.1 µm respectively, with a mass median diameter of 2.49 +/- 3.47 µm. Sanders et al. (2003) measured from NAO, LM and SM types on a brake dynamometer and found particle mass size spectra with medians at around 5 -6 µm and modes at 3-4 µm, despite the use of different braking scenarios. Iijima et al. (2008) studied NAO linings and reported a mass concentration mode at 3 - 6 µm. In tests of NAO linings, Hagino et al. (2016) found 2 – 21% of brake wear particles became airborne, and particle mass was predominantly in the 0.8 – 2.2 µm size range. Various brake pad and disc materials were tested by Wahlstrom et al. (2009; 2010b) in a laboratory pin-on-disc rig, who found a mode in the mass distribution around 3 µm.

Brake wear size distributions have also been estimated from field measurements. In a receptor modelling study, Wahlin et al. (2006) apportioned a range of elements to the brakes source: chromium (Cr), iron (Fe), copper (Cu), zinc (Zn), zirconium (Zr), molybdenum (Mo), tin (Sn), antimony (Sb), barium (Ba) and lead (Pb), and reported that particles created by brake abrasion had mass median aerodynamic diameters around 2.8 µm. In a Positive Matrix Factorization (PMF) study of wide range particle size spectra at a roadside site, Harrison et al. (2011) identified a source of airborne particles which had a mode at 3 µm diameter and was associated with traffic-influenced wind directions and heavy duty vehicle numbers which they concluded was caused by brake wear. Similarly, analysing metals data from the same site (Marylebone Road, London), Harrison et al. (2012) report a mass modal diameter of 3 µm for Ba, used as a tracer of brake wear. Similar modes were seen for other elements such as Fe, Cu and Sb, associated with brake wear.

Sanders et al. (2003) in dynamometer and test track studies reported considerable difference in total brake wear rates with low metallic (LM) pads emitting 3-4 times the material of semi-metallic (SM) and non-asbestos organic (NAO) linings, but a consistent presence of Fe, Cu and Ba and a mass median diameter of ca. 6 µm. Working in road tunnels in the United States, Lough et al. (2005) measured mass modes at 3-6 µm diameter for brake-associated metals (Cu, Ba, Fe, Sb). According to Gietl et al. (2010), the dominant mass mode of iron, copper, barium and antimony found at Marylebone Road, London was between 1.2 and 7.2 µm aerodynamic diameter, and peaked at around 3 µm. In analysis of antimony mass size spectra, Iijima et al. (2009) identified a bimodal structure and attributed a mode at 3.6-5.2 µm as being due to brake dust. In the vast majority of these studies, the modal diameter of brake wear particles is close to 2.5 µm, the diameter conventionally taken in regulations as the boundary between fine and coarse particles. The consequence is that when apportioning particulate matter mass into fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) fractions, significant quantities of brake wear particulate matter is recorded in both fractions. Minor differences in experimental conditions can therefore result in substantial differences in the $PM_{2.5}$:$PM_{2.5-10}$ ratio as seen in studies above (Garg et al., 2000; Iijima et al., 2008). Nosko and Olofsson (2017) report brake wear particles to have an effective density (0.75 +/- 0.2 g cm^{-3}), far lower than the bulk density of the constituent material, and concluded that the particles are porous.

There has been much recent interest in emissions of nanoparticles from brake wear. Since these are very small (defined as <100nm diameter), when present they typically account for a very large number of the particles, but very little mass. In an early study of the wear of brake components, Roubicek et al. (2008) identified the emission of nanoparticles, using emission of CO_2 from the brake pad as a measure of wear. Using a laboratory pin-on-disc apparatus, Wahlstrom et al. (2009; 2010b) found modes in the number distribution of particles emitted from various brake pad and disc materials of around 350 nm and 550 nm, with a tail extending to much smaller sizes. As the nanoparticle fraction is likely to arise from vaporisation-condensation of metals, rather than abrasion, it is unsurprising that subsequent research has shown that emissions increase sharply above a critical temperature (160-190ºC) (Mathissen et al., 2018; Gonet and Maher, 2019). It is reported that this temperature is not normally achieved during on-road vehicle usage (Farwick zum Hagen et al., 2019a,b). In a pin-on-disc study, such particles could be as small as 1.3-10 nm diameter (Nosko et al., 2017). Consistent with this, Mathissen et al. (2011) identified a unimodal particle number size distribution near the disc brake, with a maximum at 11 nm.

In measurements within a road tunnel, Lough et al. (2005) observed a sub-micron mode in atmospheric measurements of Cu, Fe, Pb and Ca, while Gietl et al. (2010) reported a secondary mass mode of Cu and Sb in roadside tests at a diameter of 0.2 – 0.4 µm. These appear likely to be associated with brake wear, although other sources, such as combusted lubricating oil may also contribute for some elements such as Ca and Zn which are present in oil additives. Iijima et al. (2008) reported a number concentration mode at 0.8 µm in brake dust spectra, but in a subsequent paper, Iijima et al. (2009) suggest that a mode in the Sb size spectrum at 0.5 – 0.7 µm may have been due to fly ash from waste incineration.

3.2 Tyre Wear

There are limited data on tyre wear particle emissions when compared with brake wear. Abrasion occurs due to shear and friction forces arising from the contact between the vehicle wheel and the road surface, and there is observational evidence of particle emissions containing fragments of road surface material embedded in tyre wear particles, referred to as Tyre Road Wear Particles (TRWP). The rate of tyre wear is affected by road surface roughness by factors of up to two to three (Kennedy et al., 2002), and increased emissions may occur when the road surface is unpaved, or especially when studded tyres are in use (Gustafsson et al., 2009). The latter are used to give grip in icy conditions, and work has been carried out in Scandinavia (Johansson et al. 2012; Denby et al., 2013) to predict the emissions from roads subject to damage from studded tyres and the addition of grit to aid traction during winter. This will include both tyre and road surface wear.

Electron microscopy can reveal the composition and morphology of tyre dust particles. Studies by Kreider et al. (2010) and Panko et al. (2013) have shown from on-road studies the association of tyre rubber internally mixed with other constituents such as road surface wear, road dust or other traffic-related sources within individual particles, known as Tyre Road Wear Particles (TRWP). Aerosol Time-of-Flight Mass Spectrometry (ATOFMS) is an alternative form of single particle analysis and has also shown an association of crustal elements with particles derived from tyre dust (Dall'Osto et al., 2014). The particles are typically elongated with sausage-like shapes (Baensch-Baltruschat et al., 2020; Klockner et al., 2021).

The majority of the mass of tyre wear particles is relatively large (Nielsen et al., 2003; Adachi and Tainosho, 2004; Kreider et al., 2010) including elongated particles in excess of 100 µm in size, with metal and mineral particles embedded within them. Kennedy et al. (2002) estimated that 80% of tyre wear debris is deposited on the carriageway surface, with only 20% becoming airborne (as TSP). According to ten Broeke et al. (2008), 5% of tyre wear occurs in the PM_{10} fraction. Pischinger and Rauterberg-Wulff (1999) used elemental carbon and low volatility organic carbon to distinguish between diesel exhaust and tyre debris in PM_{10} and $PM_{2.5}$ samples collected in a highway tunnel, and report tyre wear particles only in the coarse fraction ($PM_{2.5-10}$).

Laboratory studies have examined the airborne particles arising from tyre wear. Dominant mass modal diameters of tyre emission particles have been found at 2 µm (Sjodin et al., 2010) and at 2 – 5 µm (with an average size of 3.8 µm) by Raisenen et al. (2003). Kupiainen (2007) observed a mass size spectrum with mode at 5 – 8 µm when operating a friction tyre on the rig used by Gustafsson et al. (2009). Raisenen et al. (2003) found that the particle size distribution was not dependent on asphalt or aggregate mineralogy. In tests on a laboratory rig and in on-road tests, Kreider et al. (2010) found particle volume size spectra with modes at 50 µm to 100 µm, and bimodal particle number concentrations with modes at 25 µm and 5 µm in the on-road data. They do not directly report the fraction of particles below 10 µm or 2.5 µm, but it can be inferred from the volume size spectra which show it to be very small. Park et al. (2018) working on a tyre wear simulator observed a mass modal diameter of around 2 µm, which broadened under harsh friction conditions. Kim and Lee (2018) found tyre wear modal diameters of 3 and 4 µm in unimodal distributions, which is close to the mode in Zn concentrations attributed primarily to tyre wear measured on Marylebone Road, London by Harrison et al. (2012).

Tyre wear is also able to generate nanoparticle emissions (<100 nm diameter). Mathissen et al. (2011) measured particle number concentrations in the 6 – 562 nm size range near the contact point on the road surface of a car travelling on a test track. This revealed a mode in the number size distribution at 30 – 60 nm (mobility diameter) during acceleration, braking and extreme cornering. During full lock braking, smoke was visible behind the tyre and the modal diameter increased to 60 – 70 nm. There was a general relationship observed between particle number concentration and speed. An ultrafine (nanoparticle) mode in the tyre wear size spectrum has also been reported by Kwak et al. (2014), Kim and Lee (2018), Foitzik et al. (2018) and Park et al. (2017). When measuring ultrafine particle number emissions of studded and winter tyres on a laboratory rig, Dahl et al. (2006), found that particle number emissions in the 15 – 700 nm size range increased with vehicle speed, and that the smallest particles are volatile. This implies that the nanoparticle fraction arises from a vaporisation/ condensation process involving oils used in the tyre, in contrast to the larger particles which arise from abrasion (Kwak et al., 2014). This is consistent with a threshold tyre tread temperature of 160ºC for generation of this mode reported by Park et al. (2017), with increased nanoparticle generation as the temperature increased.

3.3 Road Surface Wear

As explained above, road surface wear particles are normally emitted internally mixed in Tyre Road Wear Particles (TRWP) and hence particle size spectra measured for tyre wear under field or realistic laboratory conditions are likely to be a good representation for road surface wear. However, there have been studies with a mobile load simulator and a road simulator. Gehrig et al. (2010) operated a mobile load simulator and found road wear particles to have a main mode at 6 – 7 µm, and a smaller shoulder at ca. 12 µm. Operating a road simulator and studded tyres, Gustafsson and Johansson (in Gustafsson, 2018) reported a mode at 5-6 µm, with particle mass orders of magnitude greater than the mobile load simulator with conventional tyres. The main constituent of road surfaces is mineral material (Gustafsson, 2018) which is unlikely to be differentiated from resuspension on the basis of its chemical composition.

3.4 Resuspension Particles

A number of studies have sought sample resuspension by placing a sample inlet behind a tyre on a moving vehicle. These have found a mode at 3 – 5 µm, regardless of whether a standard (summer) or studded tyre was used (Hussein et al., 2008). Lee et al. (2013) report a number mode at 800 nm with a concentration that did not vary with vehicle speed, while the mass mode at 2 – 3 µm saw a slight increase in concentration with vehicle speed. A larger mass mode in the 6 – 10 µm diameter range was found from sampling in a road tunnel by Lough et al. (2005) for the geological elements magnesium (Mg) and calcium (Ca), most likely attributable to resuspension, or possibly road surface wear. Harrison et al. (2011) applied Positive Matrix Factorization to wide range particle size spectra measured on Marylebone Road, London, and identified a factor with a mode at diameter >10 µm which occurred at traffic-influenced wind directions, and during the morning rush hour when Heavy Duty Vehicle flow was greatest. Using Si as a tracer, Harrison et al. (2012) identified the contribution of resuspension emissions on Marylebone Road, London, finding a mass modal diameter of around 5 µm, coarser than the brake or tyre wear, extending to sizes beyond 10 µm.

4. CHEMICAL COMPOSITION OF NEE

4.1 Brake Wear Particles

Brake linings of frictional braking systems can be described as metallic, semi-metallic and non-asbestos organic (NAO) according to the proportions of metallic and mineral and/or organic material present (Chan and Stachowiak, 2004; Sanders et al., 2003). Across these classifications, the chemical components in the brake linings typically derive from the following five categories of function, listed here in decreasing order of contribution by mass (Grigoratos and Martini, 2015; Thorpe and Harrison, 2008).

- Fillers (15-70%): cheaper materials to provide bulk and which include minerals such as barite ($BaSO_4$), calcite ($CaCO_3$) and mica (silicates), and particles of rubber.
- Binders (20-40%): to maintain structural integrity of the brake linings under mechanical and thermal stress and which typically comprise phenolic resins.
- Fibres (6-35%): to provide mechanical strength and which may be composed of various metals, carbon, glass, Kevlar and, to a lesser extent, ceramic fibres.
- Friction modifiers or lubricants (5-29%): to stabilise the frictional properties and which are typically composed of graphite, metal sulphides such as antimony trisulphide (Sb_2S_3), and metal oxides.
- Abrasives (up to ~10%): to increase friction and which may comprise a variety of substances including aluminium oxide, iron oxides, silicon oxides (quartz), zirconium oxide and zirconium silicate.

The broad range and quantities of chemical components listed above reflects the wide variety of formulations used in brake linings in practice (Blau, 2001; Chan and Stachowiak, 2004), which is also driven by commercial confidentiality. The plethora of materials used in brake liners means that particles emitted from the braking process likewise contain a multitude of chemical components (Grigoratos and Martini, 2015; Kukutschová et al., 2011). In addition, because of the wide variation in resistance to mechanical disintegration and volatilisation across the chemical constituents, the composition of brake-wear particles significantly differs from that of the original lining material (Kukutschová et al., 2011; Österle et al., 2001). A substantial increase in ultrafine particle number emissions when localised brake temperatures exceed ~170°C, which is attributable to evaporation and condensation of brake binder material (Perricone et al., 2018), may not contribute significantly to mass of brake-wear emissions.

As the elemental content of brake pads is extremely variable (Hulskotte et al., 2014), almost the complete suite of metallic elements (plus Sb and As) have been detected in brake linings and in brake dust (Hulskotte et al., 2014; Grigoratos and Martini, 2015; Thorpe and Harrison, 2008). However, certain elements stand out in brake-wear particles, either because they are in greater abundance or because brake wear is a dominant source of these particles in ambient air and therefore they act as useful tracers for brake-wear emissions. The four elements most readily identified in brake-wear particles are Fe, Cu, Ba and Sb, which also tend to correlate well with each other (e.g. Gietl et al., 2010; Lawrence et al., 2013) indicating a common source. Formerly, Pb was also a significant component of brake wear but this is no longer the case due to the replacement of Pb in modern linings (Grigoratos and Martini, 2015).

For Cu, Ba and Sb, brake wear emission is now their major source into ambient air, away from specific industrial point source emissions (Hjortenkrans et al., 2007). For example, the UK National Atmospheric Emissions Inventory (NAEI) estimates that, in 2016, 90% of UK national emissions of

Cu derived from non-exhaust emissions, a percentage that will be higher still in areas with large road traffic activity. This is consistent with the 80-90% estimates made for the UK by Denier van der Gon et al. (2007). Brake-wear emissions are also an important source of atmospheric emissions of Sb (Bukowiecki et al., 2009; Iijima et al., 2008; Varrica et al., 2013): many brake linings contain up to a few % of stibnite (Sb_2S_3), which can be oxidised to Sb_2O_3 during the braking process. The three elements Cu, Ba and Sb, have all been used as chemical tracers for brake wear in source apportionment studies (Alves et al., 2020; Amato et al., 2010a; Charron et al., 2019; Dong et al., 2017; Dongarrà et al., 2008; Gietl et al., 2010; Hicks et al., 2021; Lawrence et al., 2013; Lough et al., 2005; Pant et al., 2015). Where a mass ratio of the element to brake-wear particles can be estimated or assumed, then quantification of that element in ambient air provides a quantitative measure of the contribution of brake-wear emissions at the measurement location. For example, Gietl et al. (2010) used Ba as a tracer for the contribution of brake-wear particles to different size fractions of ambient PM at a roadside and an urban background site in central London. These authors estimated that the mean proportion of Ba in brake wear emissions in PM_{10} (for the UK, at that time) was 1.1%. Hicks et al. (2021) also used Ba as a tracer, applying a scaling factor of 91 to their roadside increment in Ba concentration to derive the contribution to PM_{10} from brake wear. Charron et al. (2019) report that ratios involving copper (Cu/Fe and Cu/Sn) could be used as brake-wear emissions tracers as long as brakes with Cu remain in use.

Other studies that applied multivariate statistical techniques to a suite of PM elemental composition data have likewise identified Cu, Ba and Sb (together with Fe) as associated with a brake-wear source (e.g. Beddows et al., 2015; Crilley et al., 2017; Visser et al., 2015). (The multiple sources of Fe in airborne PM mean it is not so useful as a source tracer.) In a harmonised study of five major southern European cities, Amato et al. (2016a) identified by PMF a factor showing broadly similar composition within all of the cities, separate to a vehicle exhaust factor, which they attributed to vehicular non-exhaust emissions. The factor showed chemical features typical of brake wear (Cu, Ba and Sb), tyre wear (Zn) and road dust (Si, Al and Ca). Since these elements do not derive exclusively from one source, there are ambiguities and uncertainties in quantification of individual non-exhaust emissions sources by these statistical approaches. It is also not possible unambiguously to distinguish from the composition of ambient PM between direct emissions of brake (and tyre and road) wear particles and the vehicle-induced resuspension or entrainment of brake dust deposited on the road surface or road verges.

Compared with elemental abundances, there is very limited information concerning the presence of organic compounds in brake wear particles (Grigoratos and Martini, 2015). This reflects a number of factors: organic compounds are in lower abundance than metallic and mineral compounds in brake linings and brake wear; quantification of individual organic species is analytically more challenging than for individual elements; and the existence of elemental markers for brake wear reduces the motivation to undertake speciation of organic compounds. Emissions of organic compounds during braking derive from the high-temperature and high-pressure reactions of graphite and phenolic resin components. Plachá et al. (2017) report BTEX, PAH and other semi-volatile organic compound emissions from a low-metallic brake lining tested using the ISO 26867 friction evaluation standard procedure. Rogge et al. (1993) also reported PAHs in brake dust, with their most abundant class of organic compounds being polyalkylene glycol ethers and n-alkanoic acids. However, brake liner formulations will have changed since these earlier measurements were made. In a recent study, Alves et al. (2021) report the presence of aliphatic and aromatic hydrocarbons, alcohols, glycol/glycerol derivatives, plasticisers, sugars, sterols and various phenolic constituents in particles derived from brake wear. Total carbon accounted for 5 – 74 % of PM_{10} mass in tests on a range of pad formulations and braking cycles. A useful organic marker of brake-wear particles is considered unlikely (Grigoratos and Martini, 2015; Thorpe and Harrison, 2008). Non-frictional, regenerative braking systems significantly reduce this source of non-exhaust particle emissions.

4.2 Tyre Wear Particles

In contrast to brake systems, whose composition is dominated by metals and metal-containing inorganic compounds (such as silicates, sulphides, oxides), tyre composition is dominated by organic materials. However, similar to brake systems, the specific composition of tyres varies widely between vehicle type (e.g. passenger car, heavy-duty vehicle) and manufacturer. The main categories of materials in a tyre by mass are as follows (Baensch-Baltruschat et al., 2020; Grigoratos and Martini, 2014; Sommer et al., 2018).

- Rubber hydrocarbons (40-60%): natural rubber (polyisoprene $[C_5H_8]_n$) and synthetic rubber, e.g. styrene butadiene rubber (SBR) or butadiene rubber (BR).
- Filler (20-35%): typically soot/carbon black, silica (SiO_2) and chalk ($CaCO_3$).
- Softener (15%): hydrocarbon oils and resin.
- Textile and metal reinforcement (5-10%).
- Vulcanization agents (2-5%): sulphur and zinc oxide (ZnO).
- Other additives (5-10%): preservatives (halogenated cyanoalkanes), anti-oxidants (amines, phenols), desiccants (calcium oxides), and plasticizers (aromatic and aliphatic esters).

Although tyre wear is primarily carbonaceous, a number of inorganic materials are used in tyre manufacture. In particular, ZnO is used as an activator for the vulcanisation process and Zn can account for around 1% of tyre tread material (Smolders and Degryse, 2002; Davis et al., 2001). Tyre wear is therefore a significant source of Zn in the environment (Councell et al., 2004; Adachi and Tainosho, 2004; Blok, 2005). In the UK, the NAEI estimates that 23% of Zn emissions to air in the UK in 2016 derived from tyre wear. This proportion will be considerably higher near roads. Enrichment of Cd in tyre wear has also been observed (Fabretti et al., 2009; Hjortenkrans et al., 2007; Lanzerstorfer, 2018). However, Cd is also a constituent of brake wear (Song and Gao, 2011), although emissions from this latter source may be declining (Hjortenkrans et al., 2007), no Cd was detected in tyre-wear particles by Kreider et al. (2010).

The significant enrichment of Zn in tyre-wear particles compared with crustal dust (e.g. Gustafsson et al., 2008) has led to its routine use as a marker for tyre-wear emissions in the same way as Cu, Sb and Ba are used as makers for brake wear in ambient PM source apportionment studies such as those described above. For example, Hicks et al. (2021) applied a scaling factor of 50 to their roadside increment in Zn concentration (taken from Harrison et al. (2012)) to derive the contribution to PM_{10} from tyre wear. However, as noted above, these markers are not unambiguous for their respective sources and the suitability of Zn, in particular, has been discussed (Thorpe and Harrison, 2008). Zn also has a significant anthropogenic source from wear of Zn-galvanised materials such as fencing, and a source in biomass burning emissions.

In respect of other inorganic components in tyres, SiO_2 is commonly used as a reinforcing filler (Rattanasom et al., 2007) and a prevalence of Si-rich particles has been observed from tyre-wear driving simulators (Kim and Lee, 2018; Park et al., 2018). These particles are particularly prevalent in the ultrafine fraction (Park et al., 2018), whose production is substantially enhanced when tyre-tread surface temperatures reached 160°C (Park et al., 2017). Sulphur was identified in the larger particles (Park et al., 2018), but S is not useful as an elemental marker of tyre-wear in ambient PM because of the prevalence of sulphate (SO_4^{2-}) from the oxidation of SO_2 emissions. Black carbon is also a significant component of tyre-wear emissions – the EMEP/EEA Emissions Inventory Guidebook applies a $BC/PM_{2.5}$ ratio of 0.15 for tyre wear (EMEP/EEA, 2016) – but the BC emission is both highly uncertain and not a unique source marker for tyre wear.

Information on the organic compounds in tyre wear particles is sparse because it is laborious to extract, separate and quantify individual organic species by GC-MS, LC-MS and similar off-line techniques (Rogge et al., 1993; Nolte et al., 2002). The focus in this review is identification of potential organic markers for a tyre-wear source. One such class of compounds is benzothiazole (BT) and its derivatives (Rogge et al., 1993; Lin et al., 2010; Zhang et al., 2018), which are used as vulcanization accelerators and UV-light stabilisers for rubbers (Liao et al., 2018). Zhang et al. (2018) quantified benzothiazole and six of its derivatives in tyre-wear particles using solid-liquid extraction and ultra-high performance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS). All seven BTs were found in 17 tyre-wear particle samples from different tyre brands. The mass fractions of all seven BTs summed ranged from 46.93 to 215 μg g^{-1} with an average concentration of 99.32 μg g^{-1}. Benzothiazole and 2-hydroxybenzothiazole (2-OH-BT) were the two major compounds, accounting for 56-89% of the total.

A second class of potential organic markers for tyre wear emissions are hydrogenated resin acids. Resin acids are natural products found in pine tars, which are additives used by manufacturers to give tyres certain desirable properties (Nolte et al., 2002). Some of these resin acids are chemically hydrogenated before being added during the manufacture process, which differentiates them from the natural resin acids that may be present in ambient PM from sources such as wood smoke. Kumata et al. (2011) proposed the following four dihydroresin acids as molecular markers for tyre-wear emissions: 8-isopimaren-18-oic acid (I), 8-pimaren-18-oic acid (II), 13β(H)-abieten-18-oic acid (III), and 13α(H)-abiet-8-en-18-oic acid (IV). The absence of I–IV in natural sources and the linear correlations between dihydroresin acids with different skeletons in tyres and in environmental samples demonstrated that I–IV are specific markers for synthetic rubbers. These workers further suggest from the results of photolysis experiments that I–IV can set lower limits for tyre-wear contributions to environmental loads of PM and polycyclic aromatic hydrocarbons (PAHs) with molecular weight ≥202. By comparing III + IV concentrations or (III+IV)/pyrene or (III+IV)/benzo[a]pyrene ratios in tyres and those in environmental matrices, Kumata et al. (2011) estimated the contributions of tyre-wear emissions to airborne PM, pyrene, and benzo[a]pyrene as, respectively, 0.68 ± 0.54%, 6.9 ± 4.8%, and 0.37 ± 0.18% in roadside PM and 0.83 ± 0.21%, 0.88 ± 0.52%, and 0.08 ± 0.06% in rooftop PM.

Other studies have also highlighted PAHs in tyre-wear emissions. In an experimental simulation of tyre-road interaction, Aatmeeyata and Sharma (2010) observed a linear increase in cumulative emissions of PAH (in particles of diameter > 10 μm) over a 14,500 km run of the tyre. The most abundant PAH was pyrene (30 ± 4 mg kg^{-1}), followed by benzo[ghi]perylene (17 ± 2 mg kg^{-1}). Sadiktsis et al. (2012) analysed for 15 high molecular weight PAH in 8 different tyres, albeit not in tyre-wear emissions. Whilst there was a factor 20 difference in PAH concentration between tyres, the relative abundance of PAHs was quite similar. Almost all (92%) of the total extractable PAH content was attributed to five PAHs: benzo[ghi]perylene, coronene, indeno[1,2,3-cd]pyrene, benzo[e]pyrene, and benzo[a]pyrene. The four dibenzopyrene isomers analysed (dibenzo[a,l]pyrene, dibenzo[a,e]pyrene, dibenzo[a,i]pyrene, dibenzo[a,h]pyrene) constituted <2% of the sum of the 15 PAH determined but represent a potential hazard as they are carcinogenic. In the EU, the concentrations of PAH in tyre-wear emissions should have declined markedly from 2010 due to implementation of EU Directive 2005/69/EC that prohibits the use of 'PAH-rich' extender oils in tyres produced after January 2010 (AQEG, 2019). It is also important to note that PAH are emitted to the air from multiple sources other than via tyre-wear particles, in particular for the road environment, in diesel exhaust emissions.

ATOFMS presents the opportunity to seek *m*/*z* distributions unique to a particular source in the mass spectra of individual particles (Dall'Osto et al., 2014). ATOFMS mass spectra of tyre-wear particles derived in a laboratory simulator showed peaks due to sulphur compounds, nitrate, Zn and *m/z* values >100 attributed to organic polymers for particles in the size range 200–3000 nm. Examination of large datasets of ATOFMS spectra for road dust and PM in ambient air revealed that the majority (95%) of tyre-wear particles present in the road dust samples and atmospheric samples were internally mixed with metals (Li, Na, Ca, Fe, Ti), as well as phosphate.

The extent to which tyre-wear (and road-wear) particles contribute to the microscopic plastic particles (microplastics) entering the environment depends on the definition of plastic. There is some disagreement on which polymers are "plastics". The ISO 472:2013 definition of plastic is "material which contains as an essential ingredient a high molecular weight polymer and which, at some stage in its processing into finished products, can be shaped by flow" (Hartmann et al., 2019). Rubbers (and some other elastomers) are excluded from this definition of plastic. However, this definition reflects the historic industrial landscape rather than perspectives about the behaviour of rubber fragments in the environment. Other authors include rubber within their definition of plastics and conclude that tyre-wear is a substantial, and under-reported, source of microplastics into the environment (e.g. Kole et al., 2017; Knight et al., 2020; Sommer et al., 2018). The annual global rubber production of ~27 million tonnes in 2015 was ~9% the total plastic production (Kole et al., 2017). However, estimating the fluxes of these plastic and rubber materials in the form of microparticles into the environment is a challenge. A proportion of tyre wear will not be released to air but remain on the road surface or enter the environment in run-off. A further consideration in terming tyre wear as plastic arises from the way in which tyre wear particles may be substantially (50%) internally mixed with road-wear fragments (Kreider et al., 2010; Panko et al., 2013; Panko et al., 2019).

4.3 Road Surface Wear Particles

It is not possible from field measurements to distinguish unambiguously between direct emissions of road surface wear particles and resuspension of such particles already on the road surface since the chemical composition is often very similar (Bukowiecki et al., 2010). Using mobile load simulators to compare the two contributions, Gehrig et al. (2010) concluded that road abrasion fluxes are normally lower than re-suspension fluxes, and that the latter increase with the dust loading on the road, as expected. However, road-surface wear emissions are substantially enhanced in locations with sub-zero temperature winters where studded tyres are routinely used for additional traction (Denby et al., 2018; Gustafsson, 2018).

Road surfaces are broadly composed of concrete or asphalt/tarmac. The former is a mixture of mineral aggregate, sand, and cement. The latter comprises ~95% mineral aggregates and ~5% bituminous binder. Since the dominant component in both concrete and asphalt road surfaces is the aggregate, road-wear particles mainly consists of small mineral fragments whose composition varies with the vast array of geological sources of the aggregate. The dominant crustal elements in these minerals are Si, Ca, K, Fe, and Al. Whilst a grouping of these elements is useful to quantify road dust contribution to airborne PM (see next section), it is nearly impossible to separate primary road wear from other mineral dust deposited on the road (Denier van der Gon et al., 2013).

The bituminous binder in asphalt/tarmac road surfaces comprise many thousands of high molecular weight aliphatic and aromatic hydrocarbons, which may also contain heteroatom functional groups with O, N and S. Smaller amounts of polymers, epoxy resins and metal complexes are also present to enhance binder-aggregate bonding (NIOSH, 2000; Thorpe and Harrison, 2008). There is a paucity of detailed chemical information on road surface composition, but it is unlikely that there is a definitive molecular (or elemental) marker for road surface wear (Boulter, 2005; Thorpe and Harrison, 2008). Many studies have reported elevated concentrations (0.08–149 μg/g) of PAHs in urban road dusts (Hwang et al., 2019), and the much greater prevalence of PAHs in road dust from coal-tar-based compared with asphalt-based road surfaces indicates their predominant source is road-surface wear (Metre et al., 2009) (coal-based bitumen containing substantially greater PAH content than oil-based bitumen). However, coal-based tars are now infrequently used.

4.4 Resuspension Particles

The traffic-related resuspension source refers to the uplift into the air by vehicle movement of particles already present on the road surface, mediated for example via tyre shear stress or vehicle wake turbulence. Resuspended particles therefore comprise particles from all three of the sources already described – brake wear, tyre wear, road surface wear – as well as particles from other sources that have also deposited onto the road surface, for example exhaust emission particles, particles from de-icing and gritting, wind-blown dust and biogenic particles. Consequently, the composition of resuspension particles comprise all the chemical components already described, and many more. Chemical analysis of road dust has received particular attention because of its contribution to potential toxicity to near-roadside ecosystems and to aquatic systems via run-off as well as to inhaled airborne PM. The very wide range of elemental and organic components in road dust is highlighted in a number of recent reviews (Haynes et al., 2020; Hwang et al., 2016; Hwang et al., 2019; Khan and Strand, 2018; Lanzerstorfer, 2020; Maeaba et al., 2020). Individual studies identify mutual correlation and/or enrichment in road dust particles of the 'crustal' elements associated with road surface mineral aggregates and wind-blown dust, for example Si, Al, Ca, Fe, Ti and Mg (Alves et al., 2020; Amato et al., 2009a; Amato et al., 2011; Amato et al., 2012a; Dall'Osto et al., 2013; Fabretti et al., 2009; Ondráček et al., 2011; Padoan et al., 2017).

Road dust is also highly enriched and contaminated by elements from tyre and brake wear that have deposited onto the surface, e.g. Sb, Cu, Zn, Sn, Cr (Adamiec et al., 2016; Alves et al., 2020; Cowan et al., 2021; De Silva et al., 2021; Lanzerstorfer, 2018; Padoan et al., 2017; Pipalatkar et al., 2014). A class of elements in road dust particles that has received particular attention due to their toxicity (and potential for recovery) are the platinum group elements Pt, Pd, Rh derived from wear of exhaust after-treatment catalysts (De Silva et al., 2021; Ladonin, 2018; Wang and Li, 2012; Zereini et al., 2012). Whilst the ultimate source of these elements is exhaust emissions, when particles containing these elements deposit on the road surface and are resuspended they contribute to the chemical composition of traffic-derived non-exhaust particles.

5. MEASUREMENTS OF NEE IN THE ATMOSPHERE

5.1 Principles of Measurements

NEE particles are, by their nature, refractory and insoluble in water. This means that many of the common tools for analysing particulate composition (both offline and in situ) such as ion chromatography, thermal desorption gas chromatography and thermal aerosol mass spectrometry such as the aerosol chemical speciation monitor (ACMS), are not well suited for analysis. Instead, offline analysis methods usually focus on the analysis of the metallic fraction. A common method is the digestion of a sample in acid (usually nitric acid) followed by analysis using Inductively Coupled Plasma (ICP) analysis with Optical Emission Spectroscopy (OES, also known as Atomic Emission Spectroscopy or AES) or Mass Spectrometry (MS) (Chow and Watson, 2013; Chow, 1995). It is also common to analyse metals in place on a substrate using x-ray spectroscopic methods such as X-Ray Fluorescence (XRF). To increase sensitivity, this may use a synchrotron source (Bukowiecki et al., 2005). The particles can also be analysed using electron microscopy and classified according to morphology and composition (Weinbruch et al., 2014).

The selective analysis of the carbonaceous component of tyre wear that makes up the majority of its composition is analytically challenging because it is (by design) not volatile or soluble, so cannot generally be analysed using thermal desorption or liquid chromatography (Noziere et al., 2015). However, it is possible to detect certain organic tracers and toxins such as PAHs and plasticisers (Avagyan et al., 2014; Alves et al., 2018). Another method to study the organic fraction of tyre material is pyrolysis, followed by Gas Chromatography (py-GC), a technique that has been successfully used to detect the presence of tyre wear particles in groundwater and soils (Unice et al., 2012). It is also possible to use FTIR to study the polymers present (Mengistu et al., 2019).

Another feature of NEE particles that is distinct from many other atmospheric aerosols is their size, owing to the fact they are produced mainly from abrasion rather than combustion or chemical processes. This means that in terms of mass, they tend to occupy predominantly the coarse $PM_{2.5-10}$ fraction rather than the $PM_{2.5}$ fraction. This is evident when samples are obtained using size-segregated methods such as cascade impactors (e.g. Kwak et al., 2013). NEE particles can also be associated with modes identified in the data from aerosol size spectrometers that are optimised for micron sized particles, such as the Aerodynamic Particle Sizer (APS) and optical particle counters (Harrison et al., 2012). By way of example, Harrison et al. (2011) reported a volume concentration size mode at 2 μm for brake dust and at >10 μm for resuspended road dust. By contrast, the mode corresponding to exhaust emissions existed at 0.2 μm. Also, Kreider et al. (2010) reported volume modes of around 70 μm for roadway particles and 100 μm for tyre wear particles.

It is known that NEE can be a source of ultrafine particles, i.e. those smaller than 100 nm (Kwak et al., 2014; Nosko et al., 2017). These contribute little to the PM mass concentrations, but can dominate number concentrations. While it is possible to measure these in the ambient atmosphere using mobility size spectrometers and condensation particle counters (Wiedensohler et al., 2012), exhaust emissions are known to be a major source of these, so disentangling the NEE contribution is challenging.

Semi-continuous analysis of these particles is possible using in situ XRF analysis, as used in the XACT monitor (Tremper et al., 2018; Furger et al., 2017). This has the advantage over offline analysis in that it produces data in near real time and can produce data on an hourly basis. Real-time analysis is also offered by single particle mass spectrometers such as the ATOFMS and related instruments (Murphy, 2007; Pratt and Prather, 2012). These use pulsed lasers to vaporise and ionise the components of individual particles and analyse the resultant ions using mass spectrometry. This can identify particle types through numerical clustering of the mass spectrometric data (Rebotier and Prather, 2007). This can assess the relative contributions of NEE particles to ambient number concentrations (Beddows et al., 2016; Dall'Osto et al., 2014). Because these instruments are usually operated using optical detection to trigger the desorption later, the data is limited to particles of around 200 nm or greater.

5.2 Receptor Modelling Techniques

The influence of individual sources of particulate matter at a receptor site can be qualitatively inferred from the inspection of tracers (Gietl et al., 2010). This works particularly well if a tracer is unique to a particular source, so in the case of NEE, tracers such as Sb and Cu can be taken as indicative of brake wear, but depending on the environment, it is harder associating tracers such as zinc with tyre wear when it is also associated with wood burning. These can be compared with data from other sources to estimate contributions to the overall particulate budget (Harrison et al., 2012). The ISO/TS 20593:2017 standard covers the estimation of tyre and road wear contributions based on analysis of samples using pyrolysis.

A common method of aerosol receptor modelling for the purposes of quantitative source apportionment as applied to other aerosol types is the Chemical Mass Balance (CMB) technique, which involves estimating a source’s contribution to PM through inspection of tracers and application of knowledge of the emissions from a source (Chow and Watson, 2002). However, this does not work particularly well for NEE because the source profiles are known to vary greatly according to formulation in the case of brake wear and the local composition in the case of road dust.

As an alternative, non-negative factorisation techniques are often applied, in particular PMF (Paatero, 1997; Paatero and Tapper, 1994). This method is able to deconstruct a multivariate dataset into a specified number of ‘factors’ using error-weighted residual minimisation without any a priori assumptions regarding the nature of the factors. This method works very well in source apportionment when a given contribution's profile is invariant (has a fixed source profile that is not modified in the atmosphere) and can be applied to a composite of observations, such as the time series of multiple metals derived using ICP-MS analysis. The technique is further improved when data is obtained as a function of time of day, thereby capturing variations in traffic density, so this technique is especially powerful when applied to high frequency sampling and semicontinuous analysis. PMF can also be applied to size distribution data, which can also be used to resolve different sources (Harrison et al., 2011; Vu et al., 2015).

A limitation of XRF is that it struggles to separate sources that are covariant at a given receptor site, which can frequently be the case with NEE, so sometimes these are lumped together as a single 'traffic related' factor. Moreover, there is a conceptual problem with this data model when it comes to separating road dust from brake and tyre wear based on chemical measurements, because much of the road dust is contaminated with the chemical components of the other sources (Alves et al., 2018). Likewise, tyre wear particles are also known to contain small road dust particles (Kreider et al., 2010). The only receptor measurements theoretically capable of distinguishing these are the single particle methods such as electron microscopy and single particle mass spectrometry, however these are still not able to explicitly tell whether a given particle was emitted directly to the atmosphere, or initially settled to be subsequently resuspended.

A further variation of PMF is the application of the Multilinear Engine (ME2) algorithm, which through adding additional penalty functions to the weighted residuals, enables the user to invoke a degree of a priori knowledge, by assuming 'target' profiles or 'pulling' time series towards a desired solution (Amato and Hopke, 2012; Paatero, 1999; Amato et al., 2009b). Another expansion of the PMF technique includes bringing in data from multiple sources; if (say) metal data alone were to be used, the derived factors would only concern the contributions to the available metal data in isolation, but by including other chemical analyses, size spectrometer and/or overall particulate mass data, the contributions to the PM mass budget can be estimated.

5.3 Measured Contributions and Size Distributions

A common method of capturing NEE data is by assessing its contribution to the roadside increment, i.e. comparing roadside concentrations with equivalent background measurements for a given conurbation. This can be done for mass concentrations of particulate components and size resolved data. NEE has been seen as a responsible for a general roadside or kerbside increment in $PM_{2.5-10}$ and this can be linked to metals associated with NEE (Hopke et al., 2018; Lenschow et al., 2001; Querol et al., 2008; Lough et al., 2005; Gietl et al., 2010). Road dust can also be sampled directly using suction and aerodynamic separation, to obtain a sample representative of what could contribute to ambient PM_{10} if resuspended by traffic (Amato et al., 2016b; Alves et al., 2020). This has the advantage of providing a larger volume of material for analysis.

As shown in Table 2, various metals can be associated with different NEE sources based on ambient measurements. The associations with specific sources can be based on knowledge of emissions, analysing the roadside increment and tunnel studies. Some elements can be associated with multiple sources such as iron present in both brake wear and mineral dust. A common method of inspecting the brake wear contribution is through evaluation of the Cu:Sb ratio, and values between approximately 1 and 10 have been reported (Pant and Harrison, 2013, and references therein).

Table 2: Metal profiles of various NEE and mineral dusts based on selected ambient, tunnel and road dust studies. *Metals detected at a subset of sites in parenthesis.

Reference	Brake wear	Tyre wear	Combined vehicular	Mineral dust
Tanner et al. (2008)	Cu, Cd	Zn		
Fabretti et al. (2009)			Cu, Zn, Sb, Sn	Rb, Sr, Mn, Fe, As
Gietl et al. (2010)			Ba, Fe, Cu, Sb	
Pérez et al. (2010)			Sb, Cu, Ni, Sn	Fe

Amato et al. (2011)	Fe, Cu, Zn, Cr, Sn, Sb	Zn		Al, Ca, Fe, V
Harrison et al. (2012)	Ba, Cu, Fe, Sb	Zn		Si, Al
Visser et al. (2015)	Cu, Zr, Sb, Ba		Fe	Si, Ca
Amato et al. (2016b)	Cu, Sn, Sb, Cr, Ba, Fe, Al, Ca	Ca		Al, Ca, K, Ti, Fe, Mg
Crilley et al. (2017)			Ti, Cr, Mn, Fe, Ni, Cu, Rb, Zr, Mo	Al, Si, Ca, Ti, Mn, Ni, Zn
Alves et al. (2018)			Pb, An, Fe, Cu, Sn, Sb	Al
Jeong et al. (2019)	Cu, Ba			Ca
Alves et al. (2020)			Cu, Zn, Sn, Sb, As, Se, Bi, Cd, Pb	Si, Al, Fe, Ca, K
Daellenbach et al. (2020)			Cu, Fe, Mo, Sb	Al, Se, Ti
Rai et al. (2020)			Fe, Cr, Cu, Mn, Zn, Ba	Ca, Si
Zhang et al. (2020)	Fe, Ba, Zr, Cu, Ti	Si, Zn		Si, Ca
Rai et al. (2021)			Ba, Ni, Mn (Rb, Cr, Fe, Zr, Sr)	Ca, Si, Ti (Sr, Fe, Zr)

Mineral dust can be associated with Si, Al and Ca, but it is can be harder to unambiguously identify because its composition varies with local mineralogy, nearby sources (e.g. construction) and contamination from exhaust and non-exhaust emissions (Adamiec et al., 2016). In some works, the contribution of traffic to sampled material is expressed as an ‘enrichment’ relative to the local continental crust (using an element such as Al as a reference), thereby capturing all the non-natural components (Alves et al., 2018). Note that this does by no means imply that airborne mineral dust is inherently ‘natural’ as the vehicles are still responsible for its resuspension. In addition, a component of road salt is sometimes reported in the form of Na, Mg and Cl (not shown in table).

Other components of the brake and tyre particles include elemental and organic carbon as measured using thermal-optical analysis (Amato et al., 2016b), however in isolation, these are difficult to distinguish from exhaust emissions, which are also predominantly carbonaceous in nature. The polymer markers associated with tyre wear, as analysed using py-GC, are relatively unambiguous and can be used to assess its contribution (Panko et al., 2013).

The application of PMF to urban data has allowed the emissions to be profiled beyond individual elements and complete profiles for the different components of NEE as observed at a receptor site, although as explained above, it is often difficult to separate the brake and tyre wear components. Examples include Fabretti et al. (2009), who were able to divide $PM_{2.5}$ into three categories, specifically resuspension, a combined 'vehicle abrasion' factor and exhaust emissions. Another example is the work of Rai et al. (2020), who were able to separate 'road dust' and 'traffic related' factors from background sources and fireworks in XACT data. Further insight can also be gained through the comparison of data collected at multiple sites simultaneously. Through PMF analysis of size-resolved metal data from roadside, urban background and rural measurements in London, Visser et al. (2015) were able to profile and assess the urban and roadside increments due to resuspended dust, brake wear, road salt and other traffic related aerosols. This again found that NEE could be most associated with PM_{10}, but an increment in $PM_{2.5}$ was also detected.

Some studies have presented a combined factorisation analysis from multiple data sources. For instance, Crilley et al. (2017) were able to use metal data combined with size distributions. Contributions of vehicle wear and road dust to $PM_{2.5\text{-}10}$ particles were found to be 13 and 31% respectively at a roadside site, although the contributions to the fine fraction and a background site were not disentangled from general traffic emissions. In a similar vein, Jeong et al. (2019) presented a combined factorisation of $PM_{2.5}$ data obtained in Toronto, using a variety of online techniques including an XACT, ACSM and Aethalometer, and offline analysis. This showed that NEE represented around a third of traffic-related $PM_{2.5}$.

While many factorisation papers (e.g. Rai et al., 2020) report NEE as a percentage of the total elements measured using a given analysis, these do not strictly represent contributions to the total PM budget because of other components that are not measured during XRF or ICP analysis (e.g. carbon, oxygen and hydrogen). Instead, tracers or a combined factorisation must be used.

The papers that do estimate absolute contributions (summarised in Table 3) have presented a significant contribution of NEE to coarse aerosol in urban environments. Based on tracer-derived data originally presented by Harrison et al. (2012), Beddows and Harrison (2021) reported the kerbside PM_{10} increment at the London Marylebone Road site due to brake wear, tyre wear and resuspension to be 2.8, 0.5 and 1.5 µg m^{-3} respectively. During a later study at the same site, Crilley et al. (2017) used combined factorisation to estimate the total contribution of vehicle wear and resuspension to $PM_{2.5-10}$ to be 1.3 and 3.1 µg m^{-3} respectively. Through the analysis and classification of samples analysed using electron microscopy, Weinbruch et al. (2014) estimated a PM_{10} kerbside increment in Ruhr, Germany due to abrasion and resuspension of 0.8 and 4.2 µg m^{-3} respectively.

In contrast, estimates of the contribution to $PM_{2.5}$ tend to be more minor. In Toronto, Jeong et al. (2019) used combined factorisation to estimate the average brake wear and resuspension contributions to be 0.2 µg m^{-3} each at a downtown site and 0.6 and 0.3 µg m^{-3} respectively at a highway site. While concentrations were low, it was stressed that these still represented the majority of some metals such as Ba, Cu and Mn. Using a tracer method applied to samples collected in London, Tokyo and Los Angeles, Panko et al. (2019) estimated that tyre wear particles represented only 0.1-0.68% of $PM_{2.5}$ mass. Absolute concentrations varied, but the highest concentration of 0.29 µg m^{-3} was noted at the Blackwall Tunnel approach in London, where significant braking occurs.

Table 3: Summary of quantitative mass apportionment studies of NEE in polluted urban environments. Notes: (a) Kerbside increment rather than overall concentration reported; (b) Downtown site; (c) Highway site.

Reference	**Brake Wear**	**Tyre Wear**	**Combined wear**	**Resuspension**
PM_{10} (µg m^{-3})				
Beddows and Harrison (2021)	2.8 (a)	0.5 (a)		1.5 (a)
Crilley et al. (2017)			1.3	3.1
Weinbruch et al. (2014)			0.8 (a)	4.2 (a)
$PM_{2.5}$ (µg m^{-3})				
Jeong et al. (2019)	0.2 (b)			0.2 (b)

	0.6 (c)			0.3 (c)
Panko et al. (2019)		<= 0.29		

6. MODELLED CONCENTRATIONS OF NEE

6.1 Modelling Approaches and Data

The usual purpose of modelling particulate concentrations is to predict annual-mean or daily-mean total mass concentrations in locations where measurements have not been made, or under alternative scenarios. Modelling thus refines estimates of exposure and allows interventions to be tested. Local-scale models provide high output resolution (metres) close to roads (e.g. Hood et al., 2014). Regional models provide coarser output resolution but cover much larger areas (e.g. Simpson et al., 2003). Some models have multi-scale capabilities (e.g. USEPA, 2020b) and different types of models may also be nested (e.g. Vieno et al., 2016). Regardless of the spatial scale, emissions of NEE are usually calculated using the same information which underpins the national inventories, as described in Section 2. These emissions are then aggregated with those from vehicle exhausts for input to the model, meaning that their spatial distribution across road links is closely aligned with that of exhaust emissions. This is not always appropriate, for example the largest braking emissions may occur on road segments where accelerations and hence exhaust emissions are relatively low. Most models of NEE concentrations are mechanistically separated from the emissions calculations. For example, the treatment of local weather conditions for dispersion is independent of any consideration of the impact of rainfall when calculating emissions. Similarly, while traffic-induced turbulence can be varied in some models, this is not used to calculate emissions. Where particle size distributions are defined (e.g. Archer-Nicholls et al., 2014) they do not usually take explicit account of relative source-strengths of the different emission types: exhaust, brake, tyre and road wear or resuspension. The relative contribution to concentrations made by different traffic sources thus mirrors that defined by the emissions inventories.

Concentration modelling presents an opportunity to verify against ambient measurements and thus test the effective performance of NEE emissions factors. Such comparisons are necessarily confounded by uncertainties in the model to which the calculated emissions are input, and made more difficult because of the relatively large contribution of non-traffic sources to roadside concentrations when compared with NO_x. A common approach is to compare modelled and measured concentrations while varying the basis of emissions calculation (e.g. de la Paz et al., 2015).

Denby et al. (2013) used concentration modelling to test the performance of NORTRIP emissions calculations, which take specific account of winter salting and sanding, as well as road wear from studded tyres used in northern Europe. The model successfully predicted the effect on concentrations of a reduction in studded tyre use in Stockholm, but performed less well in Copenhagen, where studded tyres are not used at all. Inclusion of road salt within the emissions calculations led to a 6-16% increase in mean total concentrations and improved overall model performance.

Pay et al. (2011) used the CALIOPE model for Spain to compare EEA emissions factors with a bespoke routine for resuspension, which they derived from ambient measurements made elsewhere in Europe, adjusted for local rainfall. The bespoke resuspension calculations reduced, but did not remove, negative bias in the modelled concentrations. Resuspension was predicted to contribute up to 7 $\mu g/m^3$ to annual mean PM_{10} in urban areas. Amato et al. (2016c) predicted concentrations in Barcelona using a Gaussian line-source model, exhaust and wear emissions from TREMOVE 2.7, and an experimentally-derived fleet-average resuspension emission rate for Barcelona of 82 mg vkm^{-1} which was reduced for periods during and after rainfall using a response curve determined from local measurements (Amato et al., 2012b). Applying this curvilinear relationship to rainfall improved model performance compared with turning resuspension on/off or assuming a constant emission. The bespoke resuspension emission routine removed most negative bias in predicted concentrations, but the spatially-uniform resuspension emission factor could not account for large inter-site variations. At traffic sites, resuspension contributed 23-44% of PM_{10}, with other traffic-related emissions contributing 8-14%. The importance of resuspension was highest in the most densely-populated parts of the city. Most modelling studies do not include rain frequency as part of the emissions calculations.

Hood et al. (2018) predicted annual mean PM_{10} and $PM_{2.5}$ concentrations across London using ADMS-Urban coupled with EMEP4UK. EEA emissions factors were adjusted based on measurements made in London (Harrison et al., 2012). AQEG (2019) used these results to show that NEE contributed more than exhaust emissions to annual mean PM_{10} and $PM_{2.5}$ and that NEE thus strongly impacted the predicted spatial variation in annual mean $PM_{2.5}$ across central London.

Daellenbach et al. (2020) used a regional chemical transport model to calculate exposure across Europe to both PM mass and to its oxidative potential (a measure of the ability to deplete anti-oxidant defences in the body). NEE emissions were not explicitly represented, but were included as a function of NO_x emissions, with size distributions subsequently parameterised from ambient measurements. The model results compared well with measurements and were used to inform a wider discussion of the oxidative potential of PM.

Machine learning methods may also be used to predict total PM mass concentrations at individual monitoring sites under alternative traffic management scenarios (e.g. Suileman et. al, 2019). These approaches have similarities to source receptor modelling and, while not widely used in this context, have the potential to determine the site-specific dependence of total concentrations on individual causal factors, such as rainfall or traffic volume (Carslaw and Taylor, 2009); potentially allowing alternative scenarios to be tested without resolving the NEE contribution and without reliance on mechanistic emissions factors.

7. MITIGATION MEASURES

7.1 Engineering Control and Material Reformulation

There has been consistent effort by automotive manufacturers and aftermarket parts manufacturers to develop brake systems which have good wear characteristics and good characteristics with regards fouling of nearby surfaces. The drivers have historically related to cleanliness during operation (e.g., Gaylard et al., 2010) and controlling operating costs, however, the compromise with braking performance (including noise, friction characteristics and thermal behaviour) has meant that brake particulates have not had the focus they are now receiving.

Attention was drawn around 2006 to the contribution of brake pads on the aquatic ecosystem in the San Francisco Bay area, US as a result of emissions of copper containing particles (e.g., Rosselot, 2006). This attention brought into focus the need for reduction of copper emissions from brake systems and led to a marked increase in commercial interest in reducing particulate emissions from brakes evident in the proliferation of related patents at this time. A memorandum of understanding between US EPA and the automotive industry was signed in 2015 with the aim of phasing out the use of copper in brake pads (to less than 0.5% by weight) by 2025 (Memorandum of Understanding, 2015), which may itself resolve the copper issue, but contributed to the awareness of brake systems as sources of vehicular particulate emissions. The studies investigating technological approaches to emission reduction or control therefore remain of interest.

There are two technological approaches evident in the literature for reducing airborne particulate emissions from brakes, namely changing the formulation of the brake friction materials and adding technology to trap the particles before they are released into the surroundings. It can be inferred also that vehicle design decisions beyond the friction brake system will influence particle emissions.

7.2 Friction Material Formulation

New materials have been evaluated thoroughly in respect of friction characteristics, noise and wear rates, but only recently has significant attention focused on particle emissions. Low copper formulations developed in light of the MoU have often focused on wear rather than emissions limiting the amount of data on the effects of replacing copper fibres in brake pads with alternative metallic materials (commonly steels), and non-metallic materials (such as natural fibres and ceramics). Despite the expectation of mass balance, wear rates typically show a weak correlation with particle number emissions at lower brake material temperatures (Tarasiuk et al., 2020) and are therefore not a reliable indicator.

There are a myriad of options available to manufacturers when developing new friction materials such as the materials used to create the primary plateaus and the binders and other materials which contribute to the secondary plateau formation. Despite only a fraction of this design space having been explored in the literature, it is clear that brake material formulation has a significant effect on particle emissions both in laboratory environments (e.g. Lyu et al., 2020) and on road (Farwick zum Hagen et al., 2019a).

Ceramic fibre and steel fibre based friction materials have been shown to produce higher particle number emissions than copper based materials at lower operating temperatures (<100°C) (Wei et al., 2020) raising concerns about the risk of particle emissions increasing as a result of reducing copper content. It is well recognised that a transition temperature exists at which the resins in the formulation start to decompose creating a large increase in fine particle emissions (Perricone et al., 2018). This transition temperature is affected by resin choice, with higher resin molecular weight

(Joo et al., 2020) and higher heat resistance of binders (Joo et al., 2019) correlating well with low particle emission rates, particularly under higher temperature testing.

The location at which the emissions are formed is that of the friction contact. The dissipation of energy at this contact results in locally high temperatures, particularly on the pads (which explains the impact of fibre choice on particle emissions).

The temperature achieved, which relates to the particulate emissions is influenced by the thermal behaviour of both the brake pad and disc. Thermal conductivity of the brake disc has consequently been shown to influence the particle emissions, with particle emissions increasing as disc thermal conductivity reduces, even when the lower thermal conductivity is only at the surface through a thin oxynitride layer (Seo et al., 2021). Hardening of the disc has been demonstrated to reduce particle emissions as a result of the harder material behaviour as part of the friction surface (Perricone et al., 2018.

7.3 Vehicle Design Decisions

The importance of brake material temperatures highlights the influence of vehicle design decisions on brake particle emissions. Frictional power has been shown to be a significant metric determining the particle emission rates (Alemani et al., 2018), disproportionately increasing emissions when considered on a per unit energy dissipated basis likely as a combined effect of contact temperatures and contact area. This suggests that vehicle mass and regenerative braking systems, both of which reduce the frictional power demand for a given deceleration event, will significantly influence particle emission rates. Although regenerative braking will reduce frictional power demand, there is a trend of increasing vehicle weight (EPA, 2021) which will somewhat counter this. Vehicle aerodynamic design, specifically with respect to brake cooling and airflows will also affect brake contact temperatures and therefore emissions.

Studies which have compared enclosed and open brake systems (i.e. drum brakes versus disc brakes) have shown that the emitted airborne particulates can be ~10 times lower when the brake system is enclosed (Hagino et al., 2016). Drum brakes seem to have fallen out of favour in many markets, however, the drop in particle emissions is somewhat attributed to the enclosing of the friction surfaces which may give a technological opportunity to reduce particle emissions not dissimilar in some ways to a passive particle filter system.

7.4 Brake Particle Filters/Traps

Filtration systems have been developed for many industrial applications and are now common in automotive internal combustion engine exhaust systems. This technology is translating across to brake systems with examples of forced induction filtration systems (developed by Tallano) (Hascoet and Adamczak, 2020; Gramstat et al., 2020) and passive filtration systems (developed by Mann Hummel) (Bock et al., 2019).

The Tallano system uses a grooved pad with an extraction system drawing air out of the groove through a filter using an electric motor powered from the vehicle electrical system. The system has been tested on brake dynamometer rigs and on vehicle. Mass based measurement was primarily used to determine the trapping effectiveness by comparing the weight loss of the friction materials to the weight gain of the filter, supported by particle measurements. The results indicate that filtration efficiencies between 40 and 90% are achievable depending on the braking behaviour, particle size range and filter condition (Hascoet and Adamczak, 2020). The filtration efficiency appeared to degrade to the lower levels when brake temperatures exceeded the decomposition temperatures – suggesting potential difficulties in this particular design in trapping finer particles. Comments on filter capacity suggest that limited life of the filter (consumable) may still need to be resolved as well as understanding its behaviour as a function of weather (Gramstat et al., 2020).

The Mann-Hummel system consists of a passive filter element located downstream of the brake pads and caliper with the intention of using the induced flows to transport the airborne particles through a filter layer. This has the advantage of being retrofittable due to the minimal changes to the existing braking system (Bock et al., 2019). It is reported to have been demonstrated on many trial vehicles under many conditions but limited supporting data is currently published. They claim filter life comparable with pad life and filtration efficiencies as high as 80%. The flow rate through the filter will depend on the filter permeability which will change over time, thereby changing the bypass ratios and filtration efficiencies. Conceptually similar filters have reported unevidenced filtration efficiencies of up to 92% (Fieldhouse and Gelb, 2016). Partial flow filters used in exhaust systems exhibit similar behaviour with filter loading and flow condition, achieving filtration efficiencies between 30 and 70% (Schrewe et al., 2012). This comparison should clearly be taken with caution, but does indicate the degree of variation that is typical of partial flow systems.

7.5 Road Surface Cleaning

Road maintenance has been implemented for many generations to reduce the debris and loose matter on the streets. Technologies to achieve this include sweeping systems, vacuum systems (often combined with sweeping) and street washing.

There have been a number of studies attempting to elucidate the effect of street sweeping, vacuuming and washing on airborne particulate number, the majority of which have struggled to identify a statistically significant effect due to the influence of, for example meteorological effects. Daily averages were highlighted to be a problematic metric for identifying effects of road maintenance. The majority of the older studies have already been reviewed well (Amato et al., 2010b) to which the reader is directed. The difficulties in measuring PM change amongst other external effects meant that many studies were inconclusive. The minority identified improvements in PM_{10} of the order of 2-3 $\mu g.m^{-3}$ and 6-30% reduction. The most significant differences arose in the hours immediately after the road maintenance, with almost twice the reduction in coarse particles compared to fine particles (Amato et al., 2010a).

Since that review (Amato et al., 2010b), there have been few focused studies on the effects of street maintenance on airborne PM. A longer duration study in Nevada US, overcame many of the issues with short term studies through analysis of a year's worth of data alongside street maintenance data (Zhu et al., 2012). They concluded that winter road treatments have an extended impact on emissions throughout subsequent months through the transport and degradation of traction additives (e.g. sand, gravel and salt), with the recommendation to perform street maintenance (sweeping and vacuuming) after winter storms to remove the remaining traction materials having recognised statistically significant improvements in roads with similar maintenance schedules. Other analysis focused studies indicate benefits of ~2 $\mu g.m^{-3}$ and 2-15% reduction in PM_{10} (Karanasiou et al., 2011).

The importance of transport of emission material from secondary and tertiary roads was highlighted by Zhu et al. (2012) with similar results in a more recent study reported by Monks et al. (2019). In this latter 2017 study a road which was heavily loaded with suspendable matter due to industrial use was subject to street maintenance. Combined sweeping and washing contributed to a measurable benefit in PM_{10} whereas sweeping alone was reported not to, supporting earlier conclusions from Amato et al. (2010b).

7.6 Dust Suppressants

For unpaved roads (particularly those in mining industries), dust suppressants are being widely used to help bind the surface road matter and reduce the amount of transfer to airborne PM. Water can have a short-term benefit widely recognised through meteorological effects in other studies and industrial applications of water sprays, however, dust suppressants aim to have a longer lasting effect. Typical formulations include salts, polymers and resins.

Results, like those for street sweeping, vary depending on the location, with focused reviews on this area showing benefits in areas of high surface loading of suspendable materials (e.g. areas with significant use of winter road traction additives, industrial areas or unpaved roads), with benefits of typically 25-50% in PM_{10} (Memorandum of Understanding, 2015). The effect of dust suppressants on industrial roads in Spain identified an ~8% reduction in kerbside PM_{10} compared to ~18% for street washing (Amato et al., 2016d). There is a distinct lack of available recent literature for more lightly loaded road surfaces.

Research communities, particularly for the mining industries continue to develop dust suppressants with improved durability and effectiveness through improved mechanical properties and ability to be robust to wetting events, for example through the careful use of hydrophilic and hydrophobic components in a polymer dust suppressant to improve water durability (Lo et al., 2020). However, the application is focused on centimetres of depth of loose material and therefore offers little insight into the effects on paved roads where the resuspendable layer is substantially thinner.

8. BATTERY ELECTRIC AND AUTONOMOUS VEHICLES

Many countries have now set deadline dates for terminating the sale of light duty internal combustion engine vehicles (ICEVs), after which new passenger car sales are expected to be restricted to battery electric vehicles. The average battery electric vehicle is heavier than the nearest equivalent ICEV, and Timmers and Achten (2016) raised the possibility that non-exhaust particle emissions from such a vehicle would exceed all emissions, including exhaust, from an ICEV. Their estimates were based upon assumed increases in tyre and road surface wear, and greater resuspension due to increased vehicle mass, and included much reduced brake wear due to regenerative braking (a system in which vehicle inertia is used to rotate the electric motor, which then acts as a generator) on electric vehicles. A scenario in which battery electric vehicles became mandatory in Ireland (Alam et al., 2018) showed an increase in $PM_{2.5}$ emissions based upon the emission factors assumed by Timmers and Achten (2016).

Beddows and Harrison (2021) used the vehicle weight dependence of emission factors to estimate the consequences of a change from an ICEV to a battery electric passenger car. The outcomes differed for gasoline and diesel vehicles, primarily due to a weight difference, and between rural, urban and motorway driving due to differing levels of wear emissions according to driving mode. However, the largest influence was the reduction in brake wear due to regenerative braking. A review by OECD (2020) found estimates of the reduction in brake wear ranging from 25-95%. Based upon an estimate of 90% reduction due to regenerative braking, Beddows and Harrison (2021) report reductions of up to 23% in total emissions of $PM_{2.5}$ (including exhaust for the ICEV) for a battery electric vehicle during urban driving, with smaller improvements in rural and motorway driving. OECD (2020) used similar methods of calculation, but assumed identical emissions from gasoline and diesel vehicles, and a 75% reduction in brake wear due to regenerative braking. They calculated non-exhaust emission factors for passenger cars, SUV and light commercial vehicles with ranges of 100 and 300km, finding net reductions in $PM_{2.5}$ emissions of 11.2 – 13.3% for the 100 km range vehicles and an increase of 2.6 – 7.8% for the 300 km range vehicles. Exhaust emissions were not however included. Both studies therefore show little impact of electrification of the vehicle fleet upon $PM_{2.5}$ emissions, with broadly similar conclusions for emissions of PM_{10}.

There are, as yet few hard data regarding autonomous (self-driving) vehicles. Current opinion is that the additional weight of batteries required for the computerisation of operation would detrimentally affect emissions compared to a conventional BEV, while the reduced braking due to smoother operation would be beneficial. The balance between the two has yet to be tested in practice.

9. NON-EXHAUST EMISSIONS OF VOLATILE ORGANIC COMPOUNDS

Whilst particulate emissions are the most frequently cited atmospheric emission arising from vehicle non-exhaust sources, and currently the largest by mass, some vehicles are associated with non-exhaust emissions of VOCs. VOC emissions from road transport have reduced dramatically over the last thirty years, both from the tailpipe and from the evaporative loss of fuels. Emissions from the tailpipe (all vehicle types) in the United Kingdom reduced from 594 kt pa in 1990 to ~16 kt in 2017 (NAEI, 2020)[1], driven by the universal adoption of the highly effective three-way catalytic convertor. Tailpipe emissions of VOCs typically comprise a mixture of unburned fuel (most significantly, toluene, xylenes and a range of C4-C10 alkanes), and products from partial combustion conditions (such as ethene, formaldehyde, acetaldehyde, propene) (NAEI, 2020).

There are non-exhaust emissions of VOCs from ICE vehicles linked to the evaporative loss of fuel (predominantly gasoline vapour but also some emissions of intermediate volatility VOCs from diesel fuel). The speciation of those emissions is unsurprisingly linked to fuel formulation, in the UK the dominant VOCs released from evaporative fuel loss are C6 and C5 alkanes, xylenes, ethanol, butane, propane and toluene along with smaller amounts of a range of other alkanes and alkenes (NAEI, 2020). Technical controls on non-exhaust fuel evaporative losses have also been highly effective, reducing from 261 kt pa in the UK in 1990 to 13 kt in 2017. In combination, the emission from tailpipe and fuel loss can be considered as emissions that are intrinsic to ICE vehicles, and that over time are likely to be eliminated as these are replaced by EV/ low carbon equivalents.

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[1] The UK reports emissions from road transport separately on a 'fuel sold' and on a 'fuel used' basis. For tracking compliance with the UK's emissions ceilings, the 'fuel used' figures are used derived from kilometres driven. The UK figures shown in this report are on a fuel used basis.

An often-overlooked component of non-exhaust VOC emissions are those associated with solvents used in ancillary systems and for car care. Most notable are screen-wash and windscreen de-icer. These emissions are agnostic to the propulsion system and will likely persist even with low carbon vehicle fleets. The non-exhaust emissions from this class fall into two types, non-aerosol and aerosol (referred to in EMEP reporting for inventories as: Non-aerosol product – automotive products NFR Code 2D3a 258 and Aerosols - car care NFR Code 2D3a 274). In combination, these non-exhaust VOC emissions from vehicles are estimated in the UK at ~55 kt pa, around 3.4 times the amount of VOC emitted from vehicle tailpipes in that year.   Considered in the broader context, non-exhaust emissions of VOCs (and excluding fuel evaporation) represents around 6.8% of total UK anthropogenic VOC emissions. Non-aerosol non-exhaust emissions from screen-wash are dominated by VOCs such as ethanol, ethylene glycol, methanol and 2-methoxy ethanol. Methanol is particularly notable because of its direct toxicity and EU regulations introduced using the REACH Directive in 2018 limited its use in screen-wash to 0.6% v/v, now replaced most commonly by ethanol. Use of aerosol care car products (predominantly for de-icer, but other sources such as cleaners and polishes contribute also) releases *n*- and *iso*-butane and propane (as the propellant) and ethanol / 2 propanol as the active ingredient for de-icing. The trends since 1995 in the UK exhaust and fuel evaporative and NEE VOCs associated with all road transport for the UK are show in Figure 3.

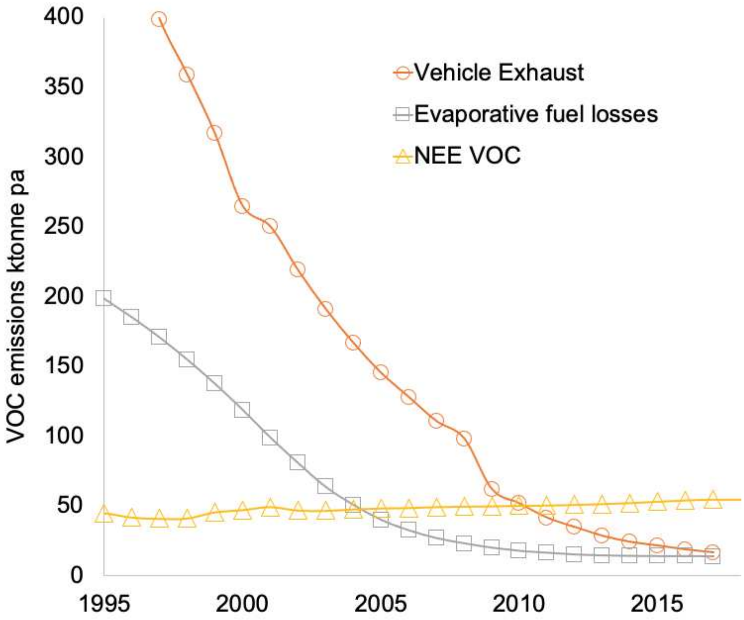

Figure 3: UK emissions of NMVOCs from all road transport by sources according to the United Kingdom National Atmospheric Emissions Inventory, summary data available at https://naei.beis.gov.uk. Methodology for the construction of the VOC element of the NAEI is described in Passant et al., 2002.

Whilst the total mass of emissions of VOC from non-exhaust sources is now dominated by ancillary emissions from care car products, the differences in speciation between the various non-exhaust sources are worth noting. Considering the Photochemical Ozone Creation Potentials (POCP) of the two different NEE VOC sources, evaporative fuels are a more significant ozone source on a tonne-for-tonne emission basis, since these contain some highly OH-reactive species such as monoaromatics and alkenes. The alcohols which make up a significant component of VOCs from aerosols and de-icers, whilst still capable of forming ozone, are somewhat lower reactivity. The exact ozone formation potentials from different NEE VOC mixtures will depend on the wider photochemical environment and is beyond this review.

In summary, whilst the total mass of VOCs emitted from vehicles has reduced dramatically over the last three decades, there remains a significant non-exhaust source of emissions, through the release of solvents used as consumable products in screen-wash and windscreen de-icing. NEE solvent emissions from vehicles substantially exceed the total currently being emitted from the tailpipe and from evaporative fuel losses and would likely persist with future decarbonised fleets.

10. FUTURE TRENDS

With traffic levels predicted to grow in many countries, non-exhaust emissions of PM are expected to further increase if no actions are taken to control them. The UK's emissions inventory predicts emissions from tyre and brake wear and road abrasion will increase by 13% by 2030, relative to 2018 levels. With exhaust emissions, and emissions from other sources, expected to decline over this time period as new vehicles meeting tighter exhaust emission standards enter the fleet, non-exhaust emissions are predicted to be responsible for 90% of all road transport emissions and 10% of all UK primary emissions of $PM_{2.5}$ by 2030. Emission projections for all countries reported in this section are taken from official inventory reporting to EMEP Centre on Emission Inventories and Projections (EMEP, 2021)

The future trend is expected to be similar elsewhere in the world where traffic is predicted to increase. Emission inventory projections reported by countries in Europe for $PM_{2.5}$, all excluding resuspension, are not always complete or comparable making it difficult to compare predicted trends in non-exhaust emissions between countries. Many countries do predict a growth in non-exhaust emissions or for the trend to remain roughly flat, although some countries do consider a reduction in emissions will occur without any clear explanation why. According to the latest available inventory data, France and Germany each predict a 2% and 4% increase in non-exhaust emissions of $PM_{2.5}$ from 2018 to 2030, respectively, with the share in these emissions to overall road transport emissions increasing from 42% to 74% in France and from 62% to 86% in Germany, a trend similar to the UK situation (EMEP, 2021). Based on inventories for all those countries in Europe which have made national inventory predictions of non-exhaust emissions, the contribution made by these sources to total road transport emissions of $PM_{2.5}$ increases, on average, from 50% in

2018 to 71% by 2030. Therefore, the overall picture in Europe is clearly one in which non-exhaust emissions are expected to become the dominant source of overall traffic emissions of PM by 2030.

Data on future trends in emissions are sparse for other countries and regions of the world. The California Air Resources Board (CARB) has a comprehensive set of inventory and projections data showing a similar trend in non-exhaust emissions as in Europe (CARB, 2016; 2017). As described earlier, the California inventory includes emissions from tyre and brake wear and resuspension and these are predicted to increase by 11% and 12%, respectively, from 2018 to 2030. To compare with the scope of $PM_{2.5}$ emissions shown in Table 1 for 2018, tyre and brake wear emissions combined as a share of total road transport emissions in California (i.e. excluding resuspension) are expected to increase from 71% in 2018 to 89% in 2030. Including resuspension further increases the contribution of non-exhaust emission in future years.

Future trends in non-exhaust emissions according to emission inventories are largely based on predictions in traffic growth, changes in the vehicle fleet and emission factors, but the general assumption is made that emission factors for non-exhaust sources on a g/km basis remain unchanged. A major uncertainty in these predictions is whether this is the case and whether PM factors will change with changes in vehicle design, weight and tyre and brake wear materials and technologies. A particular area of uncertainty is the effect the future fleet penetration of hybrid and battery electric vehicles will have on non-exhaust emissions. As discussed in previous chapters, emissions may be impacted in either direction through the effect of increased vehicle mass which may increase emissions and the effect of regenerative braking which may reduce emissions. There is an urgent need for measurements to confirm emission factors for these vehicle types as there are currently no confirmed or widely accepted emission factors for use in inventories that reflect the effect these technologies have on tyre and brake wear emissions under real-world conditions.

The report by the OECD on non-exhaust emissions estimated that there would be a 53.5% increase in non-exhaust emissions of PM from passenger vehicles worldwide, from 0.85 million tonnes in 2017 to 1.3 million tonnes in 2030 based on the predicted growth in the global vehicle stock (OECD, 2020). This was for a Business-as-Usual scenario that assumed an uptake of lightweight electric vehicles (EVs) to 4% of the vehicle stock by 2030 and the emission estimates considered the effect of vehicle weight and regenerative braking on emissions. The study concluded that a doubling of the EV numbers to 8% of the vehicle stock would have a minimal effect on the rate of increase in non-exhaust emissions. However, higher emissions would occur if the uptake were with heavier, larger range EVs.

Beddows and Harrison (2021) recently considered the dependence of non-exhaust emissions from battery electric vehicles on vehicle mass. The study considered the issue on a g/km emission factor basis for tyre and brake wear, road abrasion and resuspension under different driving conditions. It concluded that overall there would only be a modest change in total PM emissions from a passenger car built to current Euro 6 standards as gains made by removal of the exhaust emissions are offset by the likely increase in non-exhaust emissions due to the increased weight of EVs. However, the outcome is critically dependent upon the extent of regenerative braking which is essential if there is to be a net benefit of EVs relative to conventional internal combustion engine vehicles. Studies like this highlight the importance of gathering relevant emission factors for each aspect of non-exhaust emissions from EVs if accurate predictions are to be made on future trends in PM emissions from the global vehicle fleet as increasing numbers of EVs enter service.

11. CONCLUSIONS

Nearly all non-exhaust vehicle emissions (brake wear, tyre wear, road wear and resuspension particles, and vehicle-care VOCs) arise regardless of the type of vehicle and its mode of power. No legislation is currently in place specifically to target non-exhaust emissions, in contrast to progressively stringent legislation that has been effective in reducing exhaust emissions. Consequently, in most developed countries national emissions inventories indicate that non-exhaust particle and VOC emissions now exceed their respective exhaust emissions. Future projections are for non-exhaust emissions to be an increasingly dominant proportion of road transport-related particle and VOC emissions. From an air quality perspective it is therefore essential to recognise that further reductions in $PM_{2.5}$ and PM_{10} in relation to road sources requires attention to non-exhaust as well as to exhaust emissions.

Comparison between inventories for non-exhaust particle emissions in different regions of the world is complicated by the different scope in the sources that are covered, for instance with resuspension being included in inventories reported in the U.S. and Latin America, but not in Europe. Nevertheless, there is a consistent trend showing at least 50% of $PM_{2.5}$ emissions from road transport come from tyre, brake wear and road abrasion in much of Europe and the U.S. while estimates of resuspension in the U.S. suggest around a further 50% occur from this source meaning a total dominance in all non-exhaust source emissions.

Non-exhaust emission factors used in inventories are sparse, have not been updated since the 1990s and span a wide range. Therefore, the exact contribution of non-exhaust emissions to air quality locally and nationally is subject to considerable uncertainty. It is strongly recommended that efforts continue on development of consistent approaches internationally for measurement of non-exhaust emission factors. Methods must strive to capture real-world variation in non-exhaust emissions arising from different materials used, type of road, surface wetness, different cycles of braking, cornering, etc.

Measurement evidence is clear that non-exhaust emissions increase concentrations of PM_{10} and $PM_{2.5}$ and some metals at roadside. Studies suggest contributions (including resuspension) of several (4-5) µg m^{-3} of PM_{10} at busy roadsides, mostly in $PM_{2.5-10}$, and in the region 1-2 µg m^{-3} for the urban background in central London.

Emission inventories for non-exhaust PM sources are largely based on old measurements and do not account for contemporary vehicle designs, technologies, tyre and brake systems and materials. Emission factors are held constant at current values and assumed not to change over time. The effect of electrification of the vehicle fleet on non-exhaust PM emissions is highly uncertain. Important future determinants of non-exhaust particle emissions are the extent of regenerative braking in the vehicle fleet and vehicle mass. The former eliminates direct brake-wear emissions but where batteries, sensors and computer hardware increases vehicle mass, the result may be greater tyre, road and resuspension emissions. A limited number of recent studies show little impact of electrification of the vehicle fleet upon $PM_{2.5}$ emissions, with broadly similar conclusions for emissions of PM_{10}.

Other non-exhaust emission mitigation approaches (in addition to driving less) include: managing driving patterns towards lower speeds and less braking; on-vehicle brake-wear capture; development of low-wear tyres and road surfaces; and road sweeping/washing and application of dust suppressants to road surfaces, although there is little evidence at present that the latter measures are effective at mitigating non-exhaust particle emissions in the long term.

Technological innovation on reducing non-exhaust emissions is encouraged, including via regulation, but the net effect of all on-vehicle changes (e.g. regenerative braking, particle capture, autonomous driving, vehicle mass) must be evaluated holistically across all sources of non-exhaust emissions, including any trade-offs between particle mass and particle number emissions.

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CONFLICT OF INTERESTS

The authors declare no competing financial interest.

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TABLE LEGENDS:

Table 1: Contribution of non-exhaust sources to road transport and all primary sources of PM emissions in Europe and the U.S. These are according to current estimates from emission inventories for the respective regions, with comparisons done on a like-for-like basis, as far as is possible. Data are derived for the UK from NAEI (2020), for EU28 from EEA (2021), for California from CARB (2016) and CARB (2017) and for the USA from USEPA (2020a).

Table 2: Metal profiles of various NEE and mineral dusts based on selected ambient, tunnel and road dust studies. *Metals detected at a subset of sites in parenthesis.

Table 3: Summary of quantitative mass apportionment studies of NEE in polluted urban environments. Notes: (a) Kerbside increment rather than overall concentration reported; (b) Downtown site; (c) Highway site.

FIGURE LEGENDS:

Figure 1: Total exhaust and non-exhaust road transport emissions of $PM_{2.5}$ from EU28 countries from emission inventories reported by Parties under the Convention on Long-range Transboundary Air Pollution in 2020, covering years up to 2018 (EEA, 2021).

Figure 2: Percentage share of total non-exhaust emissions of $PM_{2.5}$ and PM_{10} for EU28 countries combined, as a percentage share of total road transport and of all national primary emissions (shown in the figure marked ‘Total’). Based on data from EEA (2021).

Figure 3: UK emissions of NMVOCs from all road transport by sources according to the United Kingdom National Atmospheric Emissions Inventory, summary data available at https://naei.beis.gov.uk. Methodology for the construction of the VOC element of the NAEI is described in Passant et al., 2002.