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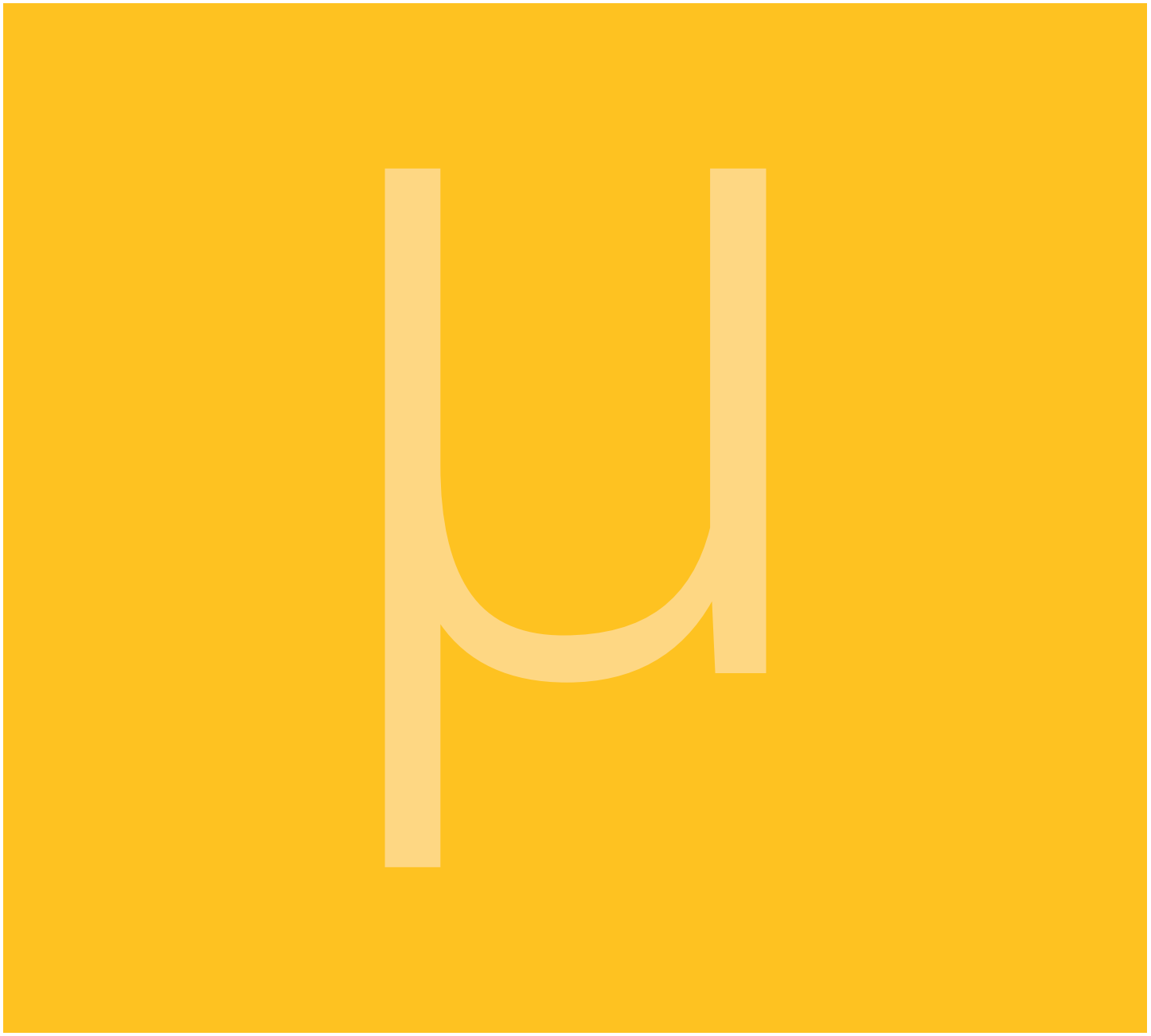
Review of non-exhaust particle emissions from road vehicles



Review of non-exhaust particle emissions from road vehicles

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EXECUTIVE SUMMARY

Non-exhaust emissions of particles from road vehicles arise from wear of the brakes, tyres and road surface, and resuspension of road surface dusts by passing traffic. These now widely exceed exhaust emissions of particles in countries where the fitting of diesel particle filters has been mandatory for several years. There have been multiple studies of the creation of particles from the abrasion of vehicle brakes and tyres. Particle size distributions have been measured both in the laboratory and the field, and can include an ultrafine fraction generated at higher temperatures. Brake linings, and hence brake wear particles show huge chemical diversity, but Cu, Ba and Sb can be used as environmental tracers. Fe which arises from both the pad and disc is emitted in greater quantities, but has other substantial environmental sources. Zn has been used as a tracer for tyre wear, although it is also emitted from brake wear and other sources. A number of organic compounds have been used as tracers for tyre wear particles, which are typically emitted with embedded mineral material. There has been much less work on road surface wear, and although emission factors are available, there have been few environmental measurements. Its composition is likely to bear some similarity to that of the other main source, resuspension of road surface dust, which is dominated by crustal elements such as Si, Al, Ca and Fe. Resuspension emissions are dependent upon the road surface dust loadings, which are highly variable and rather few measurements exist. The USEPA algorithm widely used to calculate resuspension emissions is empirical, but has been subject to some criticism. While most brake wear particles are <10μm diameter, both tyre wear and resuspended dust extend to much larger sizes, but much coarser material remains on the road surface. Ultrafine particles (<100nm diameter) can be emitted from both brake and tyre wear, but require high temperatures for their creation which are rarely achieved during normal vehicle operation. There are relatively few published measurements of non-exhaust particle concentrations in the atmosphere, and not all estimate the split between the different source types. This sub-division varies from site to site and with season, so no general pattern can be described. The total mass is divided between the fine and coarse particle fractions, with typically a mass median at 3-4 μm, and hence less than half is in the fine ($PM_{2.5}$) fraction. Total concentrations (as PM_{10}) in urban air are typically in the range of 1 to 5 μg m^{-3}, the higher concentrations occurring in street canyon locations. A study designed to predict emissions from Battery

Electric Vehicles suggests little overall difference in total particle emissions relative to Euro 6 diesel and gasoline vehicles.

There have been a number of *in vitro* studies of the toxicity and Oxidative Potential of non-exhaust particles, either as brake dust, tyre dust or particles sampled in roadside air, as well as both inhalation and installation *in vivo* studies in rodent models. These show toxic effects, typically only at higher concentrations, but are inconsistent in their findings. They establish a probable hazard from inhalation of non-exhaust particles, but not the risk to humans. There have also been population-based epidemiological studies. Such studies require a pre-processing of the air quality data to identify one or more "factors" which represent non-exhaust particle sources, which are subsequently used to seek associations with adverse health outcomes. In many of the studies, there appears to be an incomplete separation of source-related factors, or an erroneous or ambiguous assignment of source to a factor. The studies cover a wide range of health outcomes, and no consistent pattern of effects related to a "non-exhaust" particle source emerges from the results of such studies, and hence there are no acceptable exposure-response functions either for total non-exhaust particles, or for any of the individual non-exhaust sources. These are likely to prove difficult to determine due to uncertainties in the source attribution stage of the studies, but also because of the likely strong spatial and temporal correlation of non-exhaust and exhaust particles in the atmosphere which challenges the ability of epidemiological models to achieve reliable separation of effect attribution. The recently achieved dominance of non-exhaust over exhaust particles in urban air may simplify this problem when a traffic factor can be taken to represent largely non-exhaust emissions. In the absence of exposure-response functions specific to non-exhaust particles, and considering the lack of adequate coherent information on the differential toxicity of particles from different sources (as concluded by a number of authoritative bodies) , the only viable approach to estimating the public health impact of non-exhaust particles is to view them simply as contributors to $PM_{2.5}$ and PM_{10} and to apply exposure-response functions derived for those metrics.

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INTRODUCTION

As controls on exhaust emissions of particulate matter from road vehicles have taken effect over the past decade, non-exhaust emissions arising from abrasion of brakes, tyres and the road surface, and from resuspension of dust have become more prominent, and now exceed exhaust emissions. Figure 1 shows the estimates and future projections from the United Kingdom and German inventories for the quantified exhaust and non-exhaust sources which do not include resuspension emissions. Although not as yet subject to regulation, these are now attracting the attention of regulators and may well be a target for controls in future, especially since electrification of the road vehicle fleet will not eliminate emissions. Although all road vehicle types emit non-exhaust particles, passenger cars make much the greatest contribution (Figure 2). This review seeks to summarise the current state of knowledge of the emissions and their possible consequences for human health. It is based upon an overview of the voluminous literature, building upon earlier reviews such as those by Thorpe and Harrison (2008), Grigoratos and Martini (2015), Amato et al. (2014a) and AQEG (2019), without seeking to be exhaustive. It first considers the properties and controlling factors for the emissions, and then the possible consequences for human health.

1. REVIEW OF CHEMICAL AND PHYSICAL PROPERTIES OF NON-EXHAUST PARTICLES

1.1 Brake Wear Particles

Brake dust is the best characterised component of non-exhaust emissions, but exhibits huge complexity, creating problems for environmental measurements. This complexity arises in part from the use of three major types of brake lining, referred to as non-asbestos organic, low metallic and semi-metallic (Sanders et al., 2003), which are each favoured in different markets. Ceramic linings are also used on some of the more expensive vehicles (Ostermeyer and Muller, 2008). Kim et al. (2020) show that the composition of the friction materials can have an appreciable effect upon the rate of particle emissions. The lining itself comprises four main components: binders, fibres, fillers and friction modifiers and more than 3000 materials are reportedly used across brands (Roubicek et al., 2008). This complexity is demonstrated by data reported by Hulskotte et al. (2014) who collected 50 used brake pads and 15 discs removed from

passenger cars in maintenance workshops. They found huge variations in the composition of brake linings, with Fe and Cu as the most abundant metals, but in widely varying ratios. Overall, Fe, Cu, Zn and Sn comprised 80-90% of the metallic content of the liners, but the remaining 10-20% contained a very wide range of metallic components (Figure 3). A further 50% of the liner comprised non-metallic elements, mostly carbon and sulphur. In comparison, the discs showed only minor variations in composition, being comprised of >95% Fe, 2-3% Si, and no other metal comprising as much as 1% (Figure 4). Based upon environmental data, Hulskotte et al. (2014) estimated that the total wear derived from the pads and discs in a 30:70 ratio. While these data are likely to be representative of Europe, the composition of brake pads used in other parts of the world may differ substantially. However, in a study of tunnels in the United States, Lough et al. (2005) found elevated concentrations of Cu, Zn, Sb and Ba which they associated with brake wear emissions, which is similar to findings in Europe. The presence of Cu is being phased out in California due to concerns over aquatic toxicity.

Although Fe is the major component of brake wear particles, this element has many other sources in the environment and except at roadside is not a reliable tracer of brake dust. On the other hand, some of the less abundant components of brake wear, although present at much lower concentrations, have fewer other significant sources and have been used either individually or in combination as brake wear tracers. Those components are typically Cu, Ba and Sb, and are summarised in Table 1. Thus, for example, Gietl et al. (2010) demonstrated that Fe, Cu, Ba and Sb had similar particle size distributions in roadside air and in a nearby park, despite much reduced concentrations in the park (Figure 5), and recommended Ba as a suitable atmospheric tracer. Cu could also be used as emissions inventories show that brake wear is a dominant source of this element (AQEG, 2019). In order to use the tracer to estimate the concentration of brake wear particles, its concentration in the emitted particles as well as in the atmosphere needs to be known. Given the diversity in brake pad composition referred to above, chemical analysis of brake pads is unlikely to represent the fleet average, and would not allow for any chemical fractionation during the abrasion process. The approach of Gietl et al. (2010) was to estimate brake wear particle emissions using emission factors from EMEP/CORINAIR and determine a ratio of elemental concentration to estimated total

concentration, which may be a source of error if the emission factors are incorrect. Receptor modelling studies using multivariate statistical methods such as Positive Matrix Factorization can identify emission sources from their chemical profiles, but are typically unable to separate a brake wear factor, but report brake wear within generic non-exhaust traffic, road dust or even road traffic factors. There has been far less interest in the organic compound emissions from brakes, but Placha et al. (2017) identified more than 100 compounds, mostly oxidation products, but including the BTEX and PAH groups of compounds, although the temperature used in the experiments (200^{o}C) exceeded that in normal on-road braking.

Brake wear particles cover a wide range of sizes, and not all become airborne, with some remaining on the vehicle and others falling on the road surface. According to Budai and Clement (2018), around 70% of brake wear particles deposited to the road surface and not washed off the road are subsequently resuspended after a rain event. Sanders et al. (2003) reported that 50% by mass becomes airborne, while Klein et al. (2013) estimated 49% of particles to be in the PM_{10} size range, and hence liable to become airborne with a significant atmospheric lifetime (minutes to days, dependent upon size). Studies with brake dynamometers have shown 80-89% (Garg et al., 2000) and 80-91% of airborne particle mass to be within the PM_{10} (Sanders et al., 2003) size range. The ratio of fine to coarse particles as reflected in the $PM_{2.5}$:PM_{10} ratio appears to be quite variable. Klein et al. (2013) report 15% of emissions to be in the $PM_{2.5}$ range, while Garg et al. (2000) indicate 58-68% of airborne particles to be within this range. The results of Iijima et al. (2008) imply 67% of PM_{10} to be in the fine particle range. Nielsen et al. (2003) measured emission factors for a number of vehicle types finding no more than 3% of suspended particles larger than PM_{10}, and 41% of PM_{10} as $PM_{2.5}$. Such variability between studies can arise for a number of reasons including different braking conditions, brake liner characteristics, and ventilation conditions during the sampling.

Brake wear particle size spectra

In dynamometer tests, Garg et al. (2000) found highly varied particle mass size spectra between the different brake lining types that they tested. Sanders et al. (2003) suggest that this result may have been influenced by particle losses in the sampling system. Sanders et al. (2003) found particle mass size spectra with modes around 3 - 4 μm despite the use of different braking scenarios. Iijima et al. (2008) identified

brake dust spectra with a mass concentration mode at 3 – 6 μm. Pun et al. (2006) provide brake pad wear mass size spectra with a corrected modal diameter of 3.1 μm and assume a nominal density of 3 g cm^{-3} for brake wear particles. On the other hand, Nosko et al. (2017) report brake wear particles to have an effective density far lower than the bulk density of the constituent material and concluded that the particles are porous.

Various brake pad and disc materials were tested by Wahlstrom et al. (2009; 2010) in a laboratory rig, who found a mode in the mass distribution around 3 μm. Wahlin et al. (2006) found that particles created by brake abrasion had aerodynamic diameters in the inhalable size range around 2.8 μm, and that this particle diameter was a common mass median for a long list of heavy metals apportioned to the brakes source: chromium, iron, copper, zinc, zirconium, molybdenum, tin, antimony, barium and lead. In a PMF study of 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 barium, used as a tracer of brake wear, seen in Figure 6.

Sanders et al. (2003) in dynamometer and test track studies found brake wear to have a mode in the mass spectra at 3-4 μm for all brake types, although there was considerable difference in total wear rates with low metallic pads emitting 3-4 times the material of semi-metallic and non-asbestos organic (NAO) linings, although with a consistent presence of iron, copper and barium. Lough et al. (2005) measured mass size spectra of brake associated metals (Cu, Ba, Fe, Sb) with modes at 3-6 μm diameter. A dominant mass mode of iron, copper, barium and antimony found at Marylebone Road was between 1.2 and 7.2 μm aerodynamic diameter (Gietl et al., 2010). Iijima et al. (2009) identified a bimodal structure to the antimony mass spectra with a mode at 3.6-5.2 μm being due to brake dust. The consequence of brake wear particulate emissions having a mode in the mass size spectrum at a diameter close to 2.5 μm is that dividing particulate mass into $PM_{2.5}$ and coarse ($PM_{2.5-10}$) fractions results in significant quantities of brake wear

particulate matter being 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 above (Garg et al., 2000; Iijima et al., 2008; Klein et al., 2013).

When particle number concentration spectra are measured, as opposed to the particle mass size spectra as reported above, maximum concentrations will be seen at smaller, possibly sub-micrometre diameters. Roubicek et al. (2008) examined the wear of brake parts and identified the emission of nanoparticles, and associated brake wear, with the emission of CO_2 from the brake pad. Wahlstrom et al. (2009, 2010) tested various brake pad and disc materials finding similar spectra for the emitted particles with modes in the number distribution around 350 nm and 550 nm. It is likely that the sub-micrometre particles arise from vaporisation-condensation of metals, rather than abrasion. Recent research shows a need for a critical temperature (160-190^{o}C) to be reached to create a large emission of nanoparticles (Mathissen et al., 2018; Gonet and Maher, 2019), and this temperature is not normally achieved during on-road vehicle useage (Farwick zum Hagen, 2019a,b). Such particles can be as small as 1.3-10nm diameter (Nosko et al., 2017). Mathissen et al (2011) did not observe ultrafine particles under normal braking. Under extreme full stop braking condition with visible smoke formation behind the tyre, Mathissen et al. (2011) identified a unimodal particle number size distribution near the disc brake, with a maximum at 11 nm. Such small particles would, however, have little mass associated.

Lough et al. (2005) noted a sub-micron mode in atmospheric measurements of copper, iron, lead and calcium, while a secondary mass mode of copper and antimony was observed in roadside tests at a diameter of 0.2 – 0.4 μm by Gietl et al., (2010). Iijima et al. (2008) identified a number concentration mode at 0.8 μm in brake dust spectra, but Iijima et al. (2009) suggest that a mode in the antimony size spectrum at 0.5 – 0.7 μm may have been due to fly ash from waste incineration.

Atmospheric concentrations

Few studies have been successful in measuring airborne concentrations of brake wear particles alone, although a number of receptor modelling studies have included them in generic categories such as non-exhaust particles without the ability to disaggregate. Although Gietl et al. (2010) did not report a brake dust concentration, it may be estimated from the data in their paper as 1.6 +/- 1.2 μg m^{-3} on Marylebone Road and 0.4 +/- 0.2 μg m^{-3} in nearby Regent's Park. This is consistent with a concentration of 0.8 μg m^{-3} reported for Copenhagen from a tracer study (Wahlin et al., 2006) and a working group estimate of 1-3 μg m^{-3} of total wear emissions reported by Denier van der Gon et al. (2013). Modelled concentrations of a similar magnitude are reported by AQEG (2019) for major roads in London.

Emission factors for particulate matter

The United Kingdom National Atmospheric Emissions Inventory lists PM_{10} brake wear emission factors derived from the EMEP/EEA Emissions Inventory Guidebook for urban and rural roads and motorways. These appear as Table 2, taken from Wakeling et al. (2017). These are derived using speed dependence shown in Figure 7. It is important to recognise that these use average speeds, and hence the higher emission at lower speeds reflects the frequent braking in slow urban driving conditions. In the UK inventory, $PM_{2.5}$ emissions are calculated from the PM_{10} data using a $PM_{2.5}$:PM_{10} ratio of 0.4, which appears broadly in line with the data reviewed above. Wakeling et al. (2017) present projections of UK and German emissions to 2030, demonstrating the expected increase in all non-exhaust sources included in the inventories (brake, tyre and road abrasion) while exhaust emissions decline. At the current time, non-exhaust emissions well exceed exhaust emissions in most developed countries. Although it has been suggested that adoption of battery-electric vehicles may lead to increased non-exhaust emissions due to greater vehicle weight (Timmers and Achten, 2016), recent work by Beddows and Harrison (2020) indicates this not to be the case, suggesting little overall change (see Section 1.6).

1.2 *Tyre Wear Particles*

In comparison to brake wear, there has been much less research on tyre wear particle emissions. Particulate emissions occur at the contact point between the vehicle wheel and the road surface as a result of abrasion damage to either the tyre or the surface. As a consequence, fragments of road surface material may be embedded in tyre wear particles, forming Tyre Road Surface Wear Particles (TRWP), elaborated upon later. Differing degrees of road surface roughness may affect the wear rate of tyres by a factor of two to three (Kennedy et al., 2002). Large emissions may occur when the road surface is unpaved, or when studded tyres are in use (Gustafsson et al., 2009). Within most developed countries, unpaved roads carry only small amounts of traffic, and the use of studded tyres may not be permitted (Nicholls et al., 2006), but in many countries one or the other factor may be an important consideration. In the USA, where resuspension is a significant contributor to airborne particulate matter in the arid south-western states, extensive emission factors have been derived over several decades for both unpaved and paved roads (EPA, 2011b). More recently, 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.

Chemical composition of tyres

Vehicle tyre treads are predominantly organic matter (Kennedy et al., 2002) with 40 to 60 % of mass comprising a blend of rubbers, along with naphthenic and aromatic extender oils that are added to tyres to improve stress resistance, pigments, vulcanising agents and accelerators such as zinc, to speed up vulcanisation (Boulter, 2005). Zinc may comprise up to 1% by weight of tyre tread material (Pant and Harrison, 2013), resulting in zinc being the dominant metallic element in tyre tread, with the zinc content being largely independent of tyre type (Weckwerth, 2001). Total emissions of zinc from tyres were calculated to be around four times the mass of zinc emissions from brake linings in Stockholm (Hjortenkrans et al., 2007). From analysis of 21 tyre samples, Klockner et al. (2019) found a mean zinc concentration of 8.7 +/- 2.0 mg g^{-1} (8700 +/- 2000 ppm), and regarded Zn as the best elemental marker for tyre wear. Similarly, Kreider et al. (2010) report a Zn concentration of 9000 ppm in tyre tread. From

measurements of zinc concentrations on a road wear simulator, Gehrig et al. (2010) estimated that PM_{10} emissions from tyre wear were 0.8 mg veh^{-1} km^{-1} for LDVs and 17 mg veh^{-1} km^{-1} for HDVs. Dong et al. (2017) measured the isotopic composition of zinc in PM_{10} in London, concluding a significant contribution arose from tyre wear. Environmental concerns have resulted in research to find ways of reducing zinc oxide content in tyres (Heideman et al., 2006), although the presence of zinc oxide improves the dissipation of heat due to the deformation of the tyre while the vehicle is in motion and may improve abrasion resistance (Moezzi et al., 2012).

Benzothiazole is used as a vulcanising agent in rubber production and together with its derivatives is widespread in the environment (Liao et al., 2018). Zhang et al. (2018) measured benzothiazole and its derivatives in debris from 17 major brands of tyres from 8 countries. Benzothiazole and 2-hydroxybenzothiazole were found to be the dominant species within this class in both tyres and road dust samples. Lin et al. (2010) reviewed the use of compounds in complex organic mixtures as tracers including those in tyre dust. N-cyclohexyl-2-benzothiazolamine (NCBA) and 2-(4-morpholinyl) benzothiazole (24MoBT) were used as markers for tyre tread wear by Allen et al. (2006) and by Kumata et al. (2002). However, Unice et al. (2013) suggest that these tracers are not specific to tyre tread polymer and their use may result in an overestimation of tyre wear particles in the environment, and that a better method is to pyrolyse samples and to analyse for rubber polymers by GC/MS against polybutadiene, polystyrene and polyisoprene standards as described by Unice et al. (2012).

Four dihydroresin acids (pimar-8-en-18-oic acid; isopimar-8-en-18-oic acid; 13α(H)-abiet-8-en-18-oic acid; 13β(H)-abiet-8-en-18-oic acid), which are not natural products but are chemically hydrogenated before being added during the tyre manufacturing process, were identified by Nolte et al. (2002) as possible markers to distinguish tyre debris from natural materials. Kumata et al. (2011) used these as molecular markers for tyre-wear emissions, and concluded that tyre debris made up 0.68% of PM_{10} at a roadside site. Similarly, Panko et al. (2013) used chemical markers for rubber polymer (Unice et al., 2012) in air quality

samples obtained from France, USA and Japan and found a mean concentration of tyre and road surface wear particles of 0.16 µg m^{-3} within the PM_{10} fraction, making up 0.84% of the PM_{10}.

According to Valle et al. (2007) the naphthenic and aromatic extender oils that are added to tyres to improve stress resistance, and contribute to tyre tread grip, wear, and endurance qualities, contain PAHs, and tyre tread can contain 17 to 357 mg of PAHs for each kilogramme of tyre tread. Sadiktsis et al. (2012) examined the high molecular weight PAH content of a number of types of tyre, and found a factor of 22.6 between the highest and lowest total concentrations of PAH, although the relative abundances of the 5 dominant PAHs (benzo[ghi]perylene, coronene, indeno[1,2,3-cd]pyrene, benzo[e]pyrene, and benzo[a]pyrene), which made up around 92.3% of the total extractable content, were similar between different types of tyre. Four carcinogenic dibenzopyrene isomers were identified, which made up less than 2% of the analysed high molecular weight PAHs. Only low concentrations of PAHs were found in tyre wear particulate matter by Kreider et al. (2010) who question the suitability of PAHs as markers for tyre debris, and suggest that in addition PAHs are not easily extracted from tyre wear particulate matter. Larnesjo (1999) considered PAH concentrations to be too variable to be used as a marker for tyre emissions. Kumata et al. (2011) demonstrated that tyre wear particles accounted for only a very small proportion of pyrene and benzo[a}pyrene in roadside and rooftop air. From January 2010, the European Union has restricted the permissible PAH content of tyres in Europe by statute (Directive 2005/69/EC) (EU, 2005), with a specific requirement that benzo[a]pyrene (BaP) does not exceed 1 mg kg^{-1}, and that the sum of benzo[a]pyrene (BaP), benzo[e]pyrene (BeP), benzo[a]anthracene, chrysene (CHR), benzo[b]fluoranthrene (BbFA), benzo[j]fluoranthrene (BjFA), benzo[k]fluoranthrene (BkFA) and dibenzo[a,h]anthracene (DBAhA) does not exceed 10 mg kg^{-1}.

The noise regulations introduced in recent years may have influenced the tread pattern and formulation of tyre rubber. The impact upon generation of tyre dust and differences between summer and winter tyres do not appear to have been studied in detail. Grigoratos et al. (2018) compared the wear properties of a

number of tyre brands and tread patterns. They report that the tyre tread mass loss showed no significant relationship to PM_{10}, $PM_{2.5}$ or particle number emissions.

Experimental rigs

Several different types of experimental rig have been used to study wear at the tyre-road surface contact point (Zhang et al., 2020a). A laboratory rig designed to examine the emissions from studded winter tyres running on an asphalt surface was used by Gustafsson et al. (2009) to examine the emissions from summer tyres. Wear from summer tyres was found to be negligible in comparison to that from the winter tyres. This rig had a tyre on a wheel (on a horizontal axis) running on a 3.8 metre diameter surface which turns about a vertical axis. This arrangement will result in a difference in the relative speed of the tyre to the surface across the width of the tyre, and is equivalent to cornering by a vehicle on a road.

Laboratory tests using a small car tyre running on a concrete drum rotating on parallel axes are described by Aatmeeyata et al. (2009) and Aatmeeyata and Sharma (2010). These authors measured the emissions of organic and elemental carbon, PAHs and mass from the predominately large (>10 μm) tyre wear particles, finding 0.02% of the total mass to be within the PM_{10} fraction (equivalent to 6.4 mg $tyre^{-1}$ km^{-1}). The concrete drum was constructed in a similar manner to that used to make concrete pipes. Their attempts to construct an asphalt drum in a similar manner were unsuccessful.

Kreider et al. (2010) obtained tyre wear particulate matter for examination and chemical analysis using a drum with an internal diameter of 3.8 metres where the tyre ran on a parallel axis on the internal drum surface made up of 12 sections of asphalt concrete surface.

Association of tyre wear and road surface wear particles

Single particle analysis can reveal the composition and morphology of tyre dust particles. Studies by Kreider et al. (2010) using electron microscopy and Panko et al. (2013) have shown that tyre dust collected from on-road studies contain tyre rubber internally mixed with other constituents such as road surface

wear or other traffic-related sources, leading to the concept of Tyre Road Wear Particles (TRWP). Similarly, particles characterised individually by an Aerosol Time-of-Flight Mass Spectrometer (ATOFMS) also showed an association of crustal elements with particles derived from tyre dust (Dall'Osto et al., 2014).

Airborne emissions size fractions

Only a fraction of the total material lost in tyre and road surface wear will become airborne. Tyre wear particles are relatively large - microscopic examination of tyre debris has shown (Adachi and Tainosho, 2004; Kreider et al., 2010) the presence of elongated particles in excess of 100 µm in size, with metal and mineral particles (presumably from road and brake dust) 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). Pischinger and Rauterberg-Wulff (1999) analysed PM_{10} and $PM_{2.5}$ samples collected at a highway tunnel for elemental carbon and low volatility organic carbon - to distinguish between diesel exhaust and tyre debris emissions. They only found tyre wear particles in the coarse mode ($PM_{2.5-10}$), where the emission rate was 6.1 ± 1.1 mg veh^{-1} km^{-1}, while ten Broeke et al. (2008) found 5% of tyre wear to be in the PM_{10} fraction.

Tyre wear and road abrasion emission rates in the various size fractions measured for cars, and estimated for other vehicle types, given by Nielsen et al. (2003) indicate a much greater contribution of larger particle sizes than for brake wear, with no road abrasion products expected in the $PM_{2.5}$ fraction. The UK National Atmospheric Emissions Inventory uses a $PM_{2.5}$:PM_{10} ratio for tyre wear particles of 0.7, although many other countries use a lower ratio (AQEG, 2019).

Particle size spectra

Dominant modal diameters in the mass size spectrum 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 mass size spectrum was not dependent on asphalt or aggregate mineralogy. In tests on a rig where tyre and road surface rotated on parallel axes, and in on-road tests (where it was estimated that 15% of particulate matter was due to tyre wear as opposed to resuspension), Kreider et al. (2010) found monomodal particulate volume concentration 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 report the fraction of particles below 10 μm or 2.5 μm, but the volume size spectra show it to be very small. Park et al. (2018) report studies on a tyre wear simulator where a mass modal diameter of around 2 μm was observed, which broadened under harsh friction conditions. The same team (Kim and Lee, 2018) found tyre wear modal diameters of 3 and 4 μm in unimodal distributions. This is similar to the mode in Zn concentrations attributed primarily to tyre wear measured on Marylebone Road, London by Harrison et al. (2012) and seen in Figure 6:.

In measurements of particle number concentration in the 6 – 562 nm size range near the contact point on the road surface of a car travelling on a test track, Mathissen et al. (2011) identified a mode in the number concentration spectra at 30 – 60 nm (mobility diameter) during high acceleration, braking and extreme cornering, but not during normal driving. When smoke was visible behind the tyre during full lock braking the modal diameter increased to 60 – 70 nm. Near the tyre contact point there was generally no relationship between particle number concentration and speed. Other workers reporting an ultrafine mode in the tyre wear size spectrum include Kwak et al. (2014), Kim and Lee (2018), Foitzik et al. (2018) and Park et al. (2017). Dahl et al. (2006), measuring ultrafine particle number emissions of studded and winter tyres on a laboratory rig, found that particle number emissions in the 15 – 700 nm size range increased with vehicle speed, while the smallest particles are volatile. It seems probable that the nanoparticle fraction arises from a vaporisation/ condensation pathway of oils used in the tyre, as opposed to the larger particles which arise from abrasion (Kwak et al., 2014). This is consistent with the observations of Park et al. (2017) who found a threshold tyre tread temperature of 160^{o}C for generation of this mode, with increased nanoparticle generation as temperatures increased. Such a temperature is likely to be reached only during extreme acceleration, braking, or very hard cornering.

Effects on tyre emissions of vehicle usage and driving behaviour

In studies of tyre tracer components in the exhaust air from a road tunnel, Allen et al. (2006) found that there was a reduction by a factor of 1.4 to 2 (from 0.17 – 0.35 mg veh^{-1} km^{-1} to 0.12 – 0.17 mg veh^{-1} km^{-1} dependent upon the tracer used) in tyre wear emissions after the Portland cement concrete road surface in the tunnel had been replaced by an asphalt rubber surface. They attributed the improvement to a reduction in the roughness of the new surface causing less wear to tyres.

In work with a test rig where the summer tyre was effectively performing a turn, Sjodin et al. (2010) found PM_{10} increased as speed increased, and as tyre temperature increased. In on-road tests Kwak et al. (2013) observed an increase in the particle mass concentration spectra at particle diameters of 4 μm and above when the vehicle was cornering.

The predicted emission rates for tyre abrasion (Table 2 and Figure 7, calculated from the typical road speeds of different highway types given in the EMEP/EEA Emissions Inventory Guidebook (EEA, 2013), show a reduction when moving from urban, to rural, to motorway locations, but at a lesser rate than for brake emissions. This is associated with less acceleration and braking causing lower tyre wear in free moving traffic.

Contribution of tyre wear particles to airborne concentrations

A table reproduced from the review by Grigoratos and Martini (2014) appears as Table 3 and shows the contribution of tyre wear particles to PM mass as a percentage. For an urban PM_{10} concentration of 20 μg m^{-3}, these imply an airborne concentration of 1-2 μg m^{-3} with some studies showing markedly less. From samples collected in Europe, the United States and Japan, Panko et al. (2013) estimate airborne concentrations within PM_{10} of only 0.05 – 0.70 μg m^{-3}. Although they did not report a tyre wear particle concentration, a value of 0.5 +/- 0.1 μg m^{-3} for Marylebone Road, London may be estimated from the data presented by Harrison et al. (2012).

1.3 Road Surface Wear Particles

This is the least well studied of the three abrasion sources of particles, despite according to the emissions inventory being comparable in magnitude to the tyre and brake wear sources (see Table 2). As noted above, road surface materials are incorporated in TRWP, and the relative quantities of purely road surface abrasion particles and TRWP, in which they are in a tyre rubber matrix, is unclear. The UK National Atmospheric Emissions Inventory uses a $PM_{2.5}$:PM_{10} ratio for road abrasion of 0.54 (AQEG, 2019), while the Danish inventory assumes zero particle mass in the fine fraction (Nielsen et al., 2003).

Gehrig et al. (2010) used mobile load simulators, where tyres were run linearly along specially prepared asphalt concrete and porous asphalt surfaces, and state that emissions of PM_{10} due to road surface abrasion was low and that, provided that the surface was in good condition, was in the range of a few mg veh^{-1} km^{-1}, if quantifiable at all. They considered that high particulate emissions at the start of the tests may have been due to the resuspension of contaminants on the surface, although given the surfaces were newly prepared, this may have been caused by the wear of additional binder on the new surface.

Composition – road surface

A range of different road surface types have been used in tyre and surface wear testing. These include: Portland cement (Aatmeeyata and Sharma, 2010; Allen et al., 2006), asphalt concrete and porous asphalt (Gehrig et al., 2010). Porous asphalt has a porosity of about 20% by volume compared to that of 5% for asphalt concrete, the greater porosity being achieved with a lower quantity of small aggregate and sand filler (Gehrig et al., 2010).

Portland cement highway road surfaces are not widely used but calcium may be expected to be present in any abrasion products from such a surface. On an asphalt road surface, it is only when studded tyres are in use that damage to the aggregate by grinding may occur (Kennedy et al., 2002). The bitumen binder of an asphalt road surface generally contains only small amounts of individual PAHs compared to the crude oil from which it is manufactured (Kennedy et al., 2002), although recycled tyre rubber has been used in some asphalt mixes (Allen et al., 2006).

Since road surface materials are of variable composition, and most of the major elements present (Ca, Si, Al, C) are present in crustal materials, including soil, which comprise the major part of road surface dusts, and C is a major component of vehicle exhaust emissions, there are no unique chemical tracers of road surface abrasion particles, and these are not easily distinguished from road dust, some of which may derive from road surface abrasion. Consequently, there are no measurements of airborne concentrations which can be unequivocally attributed to road surface abrasion particles, although Amato et al. (2014a) quantify the percentage contribution of a road wear category of particles to "road dust".

1.4 Particle Resuspension Emissions

Particulate material that is resting on the road surface can be resuspended by vehicles moving over the surface. Three processes may be involved: high speed airflows as air is expelled from the space in front of the tyre as the tyre closes on the road surface and returns behind the rear of the tyre; turbulent eddies due to the movement of the body of the vehicle through the air; and the adhesion of material from the road surface to the tyre and its subsequent detachment. The latter process may involve material in liquid droplets when the road is wet, with additional material being washed out of recesses in the road surface.

Some authors (e.g. Dall'Osto et al., 2013) identify a traffic vortex as being a major contributor to resuspended dust. However, it has been found in California (Moosmuller et al., 1998) that PM_{10} dust entrainment from an unpaved (and unvegetated) road shoulder under dry conditions by vehicle body turbulence was almost exclusively limited to large vehicles moving in excess of 80 km hr^{-1}, resulting in an emission rate of 8 g veh^{-1} km^{-1} from this type of vehicle. Particulate matter may also accumulate on the road surface despite the presence of passing vehicles. After applying tracer grit to the road surface, Patra et al. (2008) observed an accumulation of material close to the kerb which was only resuspended by larger vehicles where the tyres track closer to the kerb. Etyemezian et al. (2003b) observed dust and debris tending to concentrate near kerbs and along centre dividers and suggest that when a vehicle accidentally travels outside a lane some of the accumulating dust may be shifted back onto the road. Pirjola et al.

(2010) noted a substantial difference between resuspension due to summer and winter tyres (both studded and non-studded), the latter causing greater resuspension.

Since 1968 the US Environmental Protection Agency has published a document "Compilation of Air Pollutant Emission Factors" (AP-42), which includes a section (13.2.1) on the emission of resuspended dust from paved roads (EPA, 2011a). This contains an empirical equation to estimate the particulate emission factor (E) of resuspended loose material from a surface of a dry paved road which, in its most recent formulation (EPA, 2011b), takes the form;

$$E = k \cdot (sL)^{0.91} \cdot (W)^{1.02}$$

where k is a factor dependent upon the particle size range (and units) of interest, sL is the silt loading per unit area of road, and W is the average weight of vehicles on the road. Silt is defined as material less than 75 µm physical diameter, and the loading can be measured by sweeping and vacuuming a unit area of the road, although in practice standard default values have been identified for four categories of road, which are inversely related to the average daily traffic flow of each category (Cowherd, 2009a). An emission factor may be calculated for a number (N) of days by multiplying the above equation by (1-P/4N), where P is the number of days when precipitation exceeded 0.254 mm during those N days (EPA, 2011b).

The difficulties of predicting emissions of resuspended material by vehicles using the AP-42 method have been considered by Venkatram (2000) and Nicholson (2001). They note that silt loading cannot be a stable explanatory variable, because it has to be continuously reduced by moving vehicles (Venkatram, 2000), and that unless the input of material to the carriageway surface is directly related to traffic density, then the amount of resuspended material cannot be proportional to the vehicle distances travelled (Nicholson, 2001). It is notable that the AP42 formulation makes no allowance for vehicle speed. However, Amato et al. (2017) found such a dependence with resuspension increasing with velocity to the power 1.5.

The consequences of the difficulties in specifying resuspension emissions are illustrated by Claiborn et al. (1995) who, when making measurements of PM_{10} alongside a tracer (SF_6), found that the measured PM_{10} emission factor was 6.7 g veh^{-1} km^{-1} for a two lane road with less than 10000 vehicles per day (approximately 80% higher than predicted by AP-42) while for a major highway (4 + lanes and over 10000 vehicles per day) the emission factor was 1.0 g veh^{-1} km^{-1} (approximately 40% less than predicted by AP-42). This is consistent with the observation of Amato et al. (2013) that road dust loadings reduce with daily traffic volumes. However, the benefits of including road dust resuspension in air quality modelling was demonstrated by Pay et al. (2011) who included the resuspension of road dust in an air quality model in Spain, achieving an improvement in PM_{10} predictions while reducing biases and errors by 15% to 18%.

A further complication is that emissions generated by brake wear, tyre wear and road surface attrition can deposit upon the road surface, to be subsequently resuspended. This gives a problem of classification, as they have both primary (component wear) and secondary (resuspension) sources.

Carriageway silt

Road dust samples taken from 28 Chinese roads by Chen et al. (2012) had an average dust loading 3.82 g m^{-2}, and when resuspended in the laboratory organic carbon (OC) comprised 11.5% and 12.5%, and elemental carbon (EC) 2.0% and 2.1% respectively of the PM_{10} and $PM_{2.5}$ fractions, while calcium, aluminium, iron and magnesium formed between 1.4% and 8% of both fractions. Amato et al. (2009a) found resuspendable PM_{10} concentrations on the surfaces of inner city streets in Barcelona of 3 to 23 mg m^{-2}, with the main components having mean values of 1363 µg Ca m^{-2}, 816 µg OC m^{-2}, 239 µg EC m^{-2}, 13 µg Cu m^{-2}, 12 µg Zn m^{-2}, 1.9 µg Sb m^{-2} and 2.0 µg Pb m^{-2}. It is concluded by Kristensson et al. (2004) that emissions due to resuspension of brake and tyre wear particles are about 100 times less than the soil part of the resuspension.

McDonald et al. (2013) have reported chemical analyses of both the $PM_{2.5}$ and $PM_{2.5-10}$ fractions of paved road dust, which was collected in bulk and aerosolised before collection of the relevant size fractions. The main constituents were crustal elements and organic carbon. The overall composition was similar to that

of local soil, but with an increased proportion of organic matter and trace metals. Chemical signatures bore more relation to region than site type.

Measurements of resuspension have been made by laying tracer material on the road surface and measuring its removal (Nicholson et al., 1989; Patra et al., 2008). Road gritting salt was applied to a carriageway in Central London to act as a tracer, and was rapidly moved along the road in the direction of traffic flow (Patra et al., 2008). Coarse particles were resuspended more rapidly than fine ones, with build-up near the kerb and the reservoir of fine particles apparently being controlled by grinding. Resuspension accounted for 40% of the material removed from a road segment, and 70% of the material removed in total.

The quantity of silt on the road surface may depend upon the local environment. Amato et al. (2012b) observed that construction dust, urban unpaved parking areas and low-vegetated gardens are likely to be responsible for an extra supply of mineral dust on urban roads in Spain. Winter maintenance can result in additional material being added to the road surface. Zhu et al. (2009) found road dust emission rates were five times higher in winter. Gertler et al. (2006) measured road surface emission factors by masts on either side of the road over several days including a snow storm, and found that pre-storm application of NaCl solution resulted in a 30% increase in both $PM_{2.5}$ and PM_{10} when the solution dried, with further increases (particularly in PM_{10}) when an abrasive was added to the road surface to improve traction. An additional increase in $PM_{2.5}$ and PM_{10} was noted following wet brushing to remove the abrasive.

Increasing the texture depth of the road surface may reduce emissions caused by an applied airflow (China and James, 2012). Langston et al. (2008) hypothesise the existence of a “differential silt mobilisation” and that on busier roads, a greater proportion of the silt is located within recesses on the road surface, and is therefore more difficult to suspend by traffic. They suggest that the standard method of sampling silt by vacuum cleaner results in an aerodynamic shear one to three orders of magnitude greater than that generated by traffic, resulting in higher estimations of silt loading on a road surface when localised

sampling is carried out for an AP-42 assessment, than when a moving vehicle raises particulate matter. Jonsson et al. (2008) describe an alternative (washing) system for sampling particulate matter on a road surface.

Resuspension particle size spectra

The particle mass size spectra behind a tyre on a moving vehicle were found to have 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) measured the particulate matter behind the tyre of a moving vehicle and found a number mode at 800 nm with 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 by Lough et al. (2005) for the geological elements magnesium and calcium. Harrison et al. (2011) in a PMF study of particle spectra identified a factor with a mode at diameter >10 µm which occurred at traffic influenced wind directions and during the morning rush hour when HDV 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, as seen in Figure 6.

Resuspension – measured by moving vehicles

Several systems of measuring resuspension by and from a moving vehicle have been reported. These include the "TRACKER" and "SCAMPER" systems in the south western United States (Etyemezian et al., 2003a; Fitz et al., 2005), the "SNIFFER" system in Finland (Pirjola et al., 2004), the "ΔDust" system in Korea (Han and Jung, 2012) and a car - with an instrumented trailer with the exhaust of the towing vehicle ducted to the rear - in Germany (Mathissen et al., 2012). Cowherd (2009a and b) has reported in a review supporting the use of mobile monitoring technologies as an alternative to the use of standardised emission factors for paved roads in predicting resuspension.

Etyemezian et al. (2003b) found that the "emission potential" – a measure of the "dirtiness" of the road (a linear function of the cubic root of the difference in PM_{10} behind the tyre to that in front of the vehicle, divided by the vehicle speed) is lower on higher speed roads, and suggest that traffic volume has less importance than traffic speed. Emission factors were less dependent on typical road speed, and high speed roads contribute more to the overall PM_{10} than low speed roads. Etyemezian et al. (2003b) suggest that while the residence time of dust on active travel lanes may only be in the order of a few hours, the debris found near kerbs may form a reservoir of material for resuspension. The emission factor may vary significantly depending on where the vehicle tyres are in a specific lane, but over a long route, movement of the vehicle across traffic lanes may give a more representative measurement of resuspended PM_{10} than sampling small areas of road surface by vacuuming (Etyemezian et al., 2005).

Han and Jung (2012) used an instrumented vehicle to measure silt loading on representative paved roads on seasonal, daily and three-hour time scales, and in relation to rainfall. The emission rates of mobile ("SCAMPER" and "TRACKER") systems and the AP 42 silt loading assessment were compared with emissions measured by masts on either side of a test road on which silt had been laid, and found good agreement with earlier data from Langston et al. (2008). The emission rates calculated by AP 42 were slightly less than those obtained with the mobile systems. When "TRACKER" and AP 42 results were compared on a public road it was found that the "TRACKER" calculated emission rate was much lower than that calculated by AP 42 which Langston et al. (2008) attributed to the "differential silt mobilisation" described above. Since AP 42 is an empirical formulation which has been updated more recently, this difference may need re-evaluation.

Mathissen et al. (2012) measured particulate matter behind a light duty vehicle (where exhaust gases had been ducted away) and found an emission factor for PM_{10} on paved roads of 26 mg veh^{-1} km^{-1}, which they noted as being large compared to the permissible exhaust emission factor of 4.5 mg veh^{-1} km^{-1} for EURO-5 vehicles. Emission factors were enhanced when the vehicle was braking or accelerating, with the lowest emissions occurring on high speed motorways, and high emissions being associated with hot spots.

In response to a consultation on the reformulation of the AP-42 equation for the emission factor of resuspended material, it was acknowledged (EPA, 2011a) that the use of mobile monitoring to estimate silt loading or the emission factor provides an advance in characterising system-wide emissions and the variation between different roads, and provides significant improvements in the estimation of road dust emissions by vehicle traffic.

Effects of road type, driving and weather conditions

Static assessments of resuspension emission rates confirm the importance of road type, traffic and driving and weather conditions. Passive measurements of resuspended dust made beside a freeway allowed Amato et al. (2012b) to derived emission factors for PM_{10} that were between 12 and 47 mg veh^{-1} km^{-1}, The authors note that these are generally lower that those measured for urban roads. In their PMF study of traffic generated PM_{10} using calcium as a tracer of resuspended road dust, Bukowiecki et al. (2010) identified 38% (LDV: 1 mg veh^{-1} km^{-1}; HDV: 262 mg veh^{-1} km^{-1}) and 56% (LDV: 28 mg veh^{-1} km^{-1}; HDV: 160 mg veh^{-1} km^{-1}) of PM_{10} to be resuspended road dust at street canyon and interurban freeway sites respectively, which compares with 38.1 +/- 9.7% of the roadside coarse particle increment at Marylebone Road (Harrison et al., 2012). They noted that road dust resuspension showed no linear relationship with traffic counts, probably as a result of variability in the availability of road dust for resuspension. Ning et al. (2008) found good agreement between emission factors obtained in roadside, chassis dynamometer and tunnel studies for brake associated metals (Cu and Ba) but poor agreement for metals associated with both traffic and non-traffic sources (Fe, Ca and Mg) which varied considerably between locations. Escrig et al. (2011) found relatively high resuspension rates at 10 – 20 g veh^{-1} km^{-1} on a lightly trafficked road in an industrial area and noted no change in resuspension over time after street cleaning. Zhu et al. (2009) found that the emission rate reduced with average speed on the road, due to material being removed from the road at a rate which increases exponentially with vehicle speed.

The wetness of the carriageway surface will affect resuspension. Precipitation can lead to wash off of the particulate matter (Omstedt et al., 2005), with the removal of salt from a road surface by spray or blow-off found to depend upon the wetness of the road by Lysbakken and Norem (2008) who concluded that more material is retained on the road when it is dry.

However, Omstedt et al. (2005) in modelling non-tailpipe emissions on Swedish roads, also noted that during wet conditions, particulate matter built up on paved roads, and was removed by resuspension when the roads were dry. Wallin (1998) observed that resuspension will increase the longer a dry period is, with as little as 0.2 mm of rain being enough to stop resuspension, and that the number of precipitation events is of greater importance that the amount. Increasing humidity will also decrease the emission rates for resuspended PM_{10} from a paved road (Kantamaneni et al., 1996). Amato et al. (2012a) measured the rate of increase in PM_{10} in vacuumed samples of the deposited dust on two roads in Utrecht and Barcelona after rain on a limited number of occasions, finding that the dust loading reached 90% of its final value after 90 hours and 27 hours respectively at the two sites. They suggest that the more rapid build-up at Barcelona is due to higher solar radiation. Berger and Denby (2011), while describing a generalised model for road dust emissions, noted that resuspension was greatest immediately after the road surface has dried up after rain, due to the accumulation of additional material during the period of rainfall when damp conditions reduce resuspension (Amato et al., 2012a).

Ball et al. (1998) found that the daily build-up of pollutant constituents on a road surface after rainfall could be represented by a hyperbolic or a power function, preferring the hyperbolic function because it leads to an asymptote. When fitting power functions of the form $B = aD^{-b}$ to the build-up (B) of metals on the road surface in the days (D) after rain, Egodawatta et al. (2013) found the values of b for geogenic metals (aluminium, calcium, iron and manganese) in the range 0.90 to 1.05, while for the anthropogenic metals (copper and zinc) and lead, which were assumed to be from contaminated roadside soils, the value of b was between 1.24 and 1.25.

The effect of precipitation may be replicated by cleaning. Amato et al. (2010) measured a 19% reduction in the concentrations of (resuspended) mineral dusts on a well trafficked urban road between 12:00 and 18:00 hours on days after the pavement had been cleaned between 23:00 and 02:00 hours, despite other airborne particulate components increasing due to meteorological and traffic congestion factors. A small increase in mineral dust occurred during the early morning hours on days when cleaning took place. Sternbeck et al. (2002) found a greater difference between the emission rates in two tunnels of PM_{10} than in metallic particulate matter – the measurements occurring at different times of year with one tunnel newly cleaned, implying that cleaning had an effect on total resuspended particulate matter, but not on metallic particulate matter from brake wear.

Emission factors were calculated by Thorpe et al. (2007) at a central London road by measuring the roadside increment of coarse particulate matter ($PM_{2.5-10}$) above the local background, and subtracting brake and tyre wear estimated from inventory-based emission factors. From changes in the vehicle fleet composition on different days of the week, they estimated emission factors for resuspension and road wear for heavy duty vehicles of 156 to 200 mg km^{-1}, far greater than the 2.9 to 8.6 mg km^{-1} that they obtained for light duty vehicles by a ratio not dissimilar to the ratio in vehicle weights. Holmen et al. (2001) measured the variation in the lidar signal 20 metres above a busy highway noting that HDVs were three times as big as LDVs, and found that there was no significant difference in the delay from the vehicle passing, concluding that the effect was due to resuspension rather than exhaust, which would be released at a higher point on the vehicle. In studies in a highway tunnel, Grieshop et al. (2006) noted that there were higher emissions of crustal elements (Fe, Ca, Mg, Li) than Zn and Mn in the early morning when there was more heavy truck traffic.

Omstedt et al. (2005) concluded that using a constant value for PM_{10} emission, or relating PM_{10} emissions to NO_x emissions, could not be justified. The NORTRIP model (described by Denby et al., 2013 and Johansson et al., 2012) predicted 76 – 91% of variability in daily mean concentrations where studded tyres

are in use, and 44 – 51% of variability where studded tyres are not used. Resuspension rates are derived for the model from on road measurements behind vehicle wheels, and a knowledge of the surface moisture.

Chemical characteristics of road dust and resuspended particles

McDonald et al. (2013) have reported chemical analyses of both the $PM_{2.5}$ and $PM_{2.5-10}$ fractions of paved road dust, which was collected in bulk, and aerosolised before collection of the relevant size fractions. The main constituents were crustal elements and organic carbon. The overall composition was similar to that of local soil, but with an increased proportion of organic matter and trace metals. Chemical signatures bore more relation to region than site type. Wang et al. (2005) also found a dominance of crustal, soil-derived elements in bulk road dust, but that these diminished in abundance, accompanied by a progressive increase in trace element concentration in smaller size fractions. Similar findings were reported by Lanzerstorfer (2018) and Lanzerstorfer and Logiewa (2019). Alves et al. (2018; 2020) report analyses of the PM_{10} fraction of road dust, finding a predominance of crustal elements accompanied by trace metals associated with brake and tyre wear. Padoan et al. (2017) compared the composition of road dust and soil samples, finding a depletion of crustal elements and an enrichment of trace elements in road dust relative to the soil. Amato et al. (2013) found a relationship between emission factors for trace metals and the surface loading of the metal in the PM_{10} size fraction of road dust.

There are no specific chemical tracers for resuspended road dust. Harrison et al. (2012) used Si and Al on the assumption that road dust is comprised mostly of crustal material. They observed a ratio of Si/Al consistent with such an assumption, which has support from the more recent work of McDonald et al. (2013), referred to above. Such an approach will work when carrying out apportionment of a traffic increment (roadside minus background), but will not distinguish road dust from other crustal material (e.g. soil) when the background is not subtracted. Pant et al. (2015) used elemental tracers to apportion road dust collected in Birmingham (UK) and Delhi (India) to crustal, brake wear and tyre wear sources, finding a predominance (71-89%) of crustal dust accompanied by brake dust (4-7%) and tyre dust (3-16%) with a very small contribution (up to 1%) estimated to arise from exhaust emissions. If such results are typical, direct

emissions of brake and tyre dust are likely to exceed substantially contributions from resuspension to these source components in the atmosphere.

Estimates of resuspended particle concentrations are rarely reported. Although not reported in their paper, the roadside increment data of Harrison et al. (2012) allow calculation of a resuspended particle concentration of 1.9 +/- 0.5 µg m^{-3} on Marylebone Road, London. This may include road surface wear due to the use of crustal element tracers. Singh et al. (2020) calculated traffic emissions estimated for Delhi, including resuspension using the USEPA algorithm. Their estimates were: exhaust, 14%; brake wear, 3%; tyre wear, 2%; road wear, 2%; and resuspension, 79% for the PM_{10} size fraction. Unfortunately, this was not translated into airborne concentrations. Modelling PM_{10} concentrations in Barcelona, Amato et al. (2016a) report a road dust contribution of 9-15% at background sites (total PM_{10}, 22-29 µg m^{-3}) and 23-44% at traffic sites (total PM_{10}, 32-41µg m^{-3}). Jeong et al. (2019) report that traffic emissions accounted for half of $PM_{2.5}$ concentrations during morning rush hours. Source apportionment revealed exhaust emissions accounting for an average 9-19% of $PM_{2.5}$, with brake wear and resuspension accounting for 2-6% and 3-4% respectively at sites in downtown Toronto. A table of relevant studies appears in Table 4. Because of uncertainties in their estimation, most national emissions inventories do not estimate PM emissions arising from resuspension.

1.5 Receptor Modelling

In recent years a number of authors have conducted receptor model studies in the vicinity of roads (Pant and Harrison, 2013), although a more limited number have sought to quantify the different sources of vehicle emissions. Positive matrix factorization (PMF) studies were carried out on data from measurements in road tunnels by Fabretti et al. (2009) and PCA/MLRA by Lawrence et al. (2013) analysing metallic trace elements in $PM_{2.5}$, and metals and PAHs in PM_{10}, respectively. Both were able to identify particulate matter due to vehicle wear, resuspension of material from the carriageway surface and exhaust emissions. Bukowiecki et al. (2010) conducted PMF studies on incremental trace metal and black carbon data collected within a street canyon and at an urban background site, and on either side of a freeway, identifying a much

greater contribution of brake wear emissions at the more congested urban traffic site. In a comparison between a Multilinear Engine (ME-2) method and PMF2, Amato et al. (2009b) identified components of PM_{10}, $PM_{2.5}$ and PM_1 associated with resuspended road dust and brake wear respectively. Size-fractionated measurements of tracer elements at urban road canyon and background sites allowed Harrison et al. (2012) to distinguish between brake, tyre and resuspended material within the roadside increment of the coarse fraction (> 0.9 µm aerodynamic diameter) particulate matter. Zhang et al. (2020b) applied receptor modelling to PM sampled in Chinese road tunnels, attributing 26-40% of PM_{10} to tailpipe exhaust, 1-3% to brake wear and 7-9% to tyre wear. For $PM_{2.5}$, the respective percentages were 59-80% (exhaust), 1-5% (brake wear) and 4-10% (tyre wear). Fleet composition, road gradient and pavement roughness were determining factors. The remaining PM was attributed to cement dust and soil.

Few studies report actual airborne concentrations. Amato et al. (2016b) report measurements from four southern European cities. The PMF factor attributed to non-exhaust emissions had concentrations of 1.8 – 5.8 µg m^{-3} representing 8 -14% of PM_{10}, and 0.2 – 2.5 µg m^{-3}, or 1-8% of $PM_{2.5}$. Source apportionment data for the Milan urban background site appear in Figure 8. The data reported by Harrison et al. (2012) from campaign measurements on Marylebone Road, London give a total average concentration of 5.2 µg m^{-3}, comprising 2.8 µg m^{-3} for brake wear, 0.5 µg m^{-3} for tyre wear and 1.9 µg m^{-3} for resuspension, with likely inclusion of road surface wear in the latter. In a study of cities in southern Spain, Amato et al. (2014b) report "road dust" contributions to airborne particulate matter. These vary by site type, but typically account for 29-34% of PM_{10} and 11-31% of $PM_{2.5}$ at urban sites. Contributions were higher in the dry season and average contributions to the "road dust" were 52% carbonaceous (tyre wear and bitumen), 35% road wear/mineral, and 13% brake wear. A number of papers in Section 2.3 on epidemiological studies have also separated a non-exhaust particle component by receptor modelling.

1.6 Battery-Electric Vehicles (BEV)

Timmers and Achten (2016a,b) suggested that as BEV are heavier than internal combustion engine vehicles (ICEV), they may emit a greater mass of non-exhaust emissions than equivalent ICEV, even if no brake wear

particles were emitted by the BEV due to regenerative braking. Beddows and Harrison (2020) used the vehicle mass dependence of emissions from brake wear, tyre wear, road surface wear and resuspension to evaluate the relative emissions of BEV and ICEV, for regenerative braking contributions of 0, 90 and 100%. The overall conclusion is that at a realistic contribution of regenerative braking of 90%, on urban and rural roads and motorways there is little difference between the emissions of the two vehicle types when including the exhaust emissions from Euro 6 passenger cars.

2. TOXICITY AND HEALTH EFFECTS OF NON-EXHAUST PARTICLES

2.1 In Vitro Toxicity Testing

The studies considered appear in Table 5. There have been a few studies of particles sampled from roadside air, or sampled from a non-exhaust source in the laboratory. Godri et al. (2011) compared the Oxidative Potential (OP) of size fractionated particles collected at roadside and an urban background site using assays based on the oxidation of ascorbic acid and glutathione in a synthetic lung lining fluid. The results showed that Oxidative Potential, a measure of the ability of particles to deplete the body's anti-oxidant defences, was not associated with the size fraction of the particles, but correlated with the content of trace metals. This is consistent with the finding of Gao et al. (2020) that OP assays are highly responsive to metal ions (especially Fe and Cu), and are most probably influenced most highly by the brake wear particles present. Shirmohammadi et al. (2016) used an alternative assay based upon dithiothreitol (DTT), also in a synthetic respiratory tract lining fluid. Samples from different distances from a highway showed responses related to the concentration of elemental carbon, consistent with the known responsiveness of this reagent to organic oxidants, and especially quinones. In a recent paper, Daellenbach et al. (2020) carried out a source apportionment of Oxidative Potential measured by three separate assays, showing that non-exhaust traffic emissions make a much larger contribution to Oxidative Potential than their contribution to PM_{10} mass. Rajhelova et al. (2016, 2019) used a lipid peroxidation assay to evaluate the toxicity of brake wear debris, reporting slight toxicity.

Gaultieri et al. (2005a,b; 2008) exposed A549 cells (model alveolar epithelial cells) to a dichloromethane extract of tyre particles finding cytotoxicity (MTT and LDH assays), DNA damage (Comet assay) and changes in cell morphology, significant only at higher concentrations. Wessels et al (2010) carried out a similar set of tests on A549 cells with size-segregated airborne particles collected from various sites - remote rural, urban background, urban roadside and road tunnel. They found an effect per unit mass of particles which increased with the degree of local pollution, but showed little difference between size fractions, exemplified for the LDH assay in Figure 9. This suggests that cytotoxicity per unit mass of particles is insensitive to the particle source, as the exhaust particle would be predominantly in the <0.5µm fraction, and non-exhaust particles in the 1.5 - 3µm and 3 - 7µm size fractions. Gasser et al. (2009) exposed A549 cells to brake wear particles finding only a low response in relation to cytotoxicity and oxidative stress. Barosova et al. (2018) exposed A549 cells and macrophages to brake wear particles, and found no significant effect on cell viability, morphology, oxidative stress or inflammation. Jalava et al. (2015) exposed a macrophage cell line to four size ranges of urban particulate matter. The Spring samples which were reported to be enriched in road dust showed greatest toxicity, manifested as loss of cell viability and highest release of nitric oxide and IL-6 (an inflammatory mediator). Particulate matter from a road simulator using studded tyres was used to expose human blood monocyte macrophages, causing enhanced release of the inflammatory markers IL6, IL8 and TNF-alpha relative to diesel exhaust particles (Gustafsson et al., 2008). Brake wear material was also used to expose human primary lymphocytes (Kazimirova et al., 2016), human peripheral blood cells, green algae (Peikertova et al., 2017), bacteria, and a mutagenicity assay (Malachova et al., 2016), and Calu-3 bronchial epithelial cells (Puisney et al., 2018). Although the level of consistency is not high, in general the higher exposures induced cytotoxicity and release of inflammatory cytokines, and generated oxidative damage. There is however no clear evidence of toxicity substantially different from that seen with airborne particulate matter more generally. Selley et al. (2020) compared brake wear particles with diesel exhaust particles using a range of tests and concluded that each exerts similar adverse effects on human macrophage cell function. It should also be noted that extrapolation of such test data to whole organisms is difficult, and hence such studies are useful primarily

in indicating potential mechanisms of toxicity rather than being predictive of effects in human populations, and especially the magnitude of effects.

2.2 In Vivo Toxicity Studies

There are a number of relevant studies in animal models, summarised in Table 6. Gottipolu et al. (2008) and Mantecca et al. (2009) found lung inflammation in rats and mice respectively as a result of installation of tyre wear particles. Kreider et al. (2012) exposed rats to tyre and road wear particles (TRWP) by inhalation, finding no major effect, but occasional lung pathology in the group with the highest dose. They concluded a No Observable Adverse Effect Level (NOAEL) of 112 µg m^{-3}. Bernstein et al. (2014, 2015, 2018) exposed rats by inhalation to brake dust with 30% chrysotile (no amphibole-rod shaped asbestos). Some lung accumulation of particle-laden macrophages was observed, but lung damage was very minor. Some lung inflammation, but no cytotoxicity with inhaled brake dust was reported by Gerlofs-Nijland et al. (2019) in an inhalation study of mice. In some, but not all cases, brake wear had a greater inflammatory effect than TRWP, diesel exhaust or wood smoke, but particulate matter from a poultry farm had the greatest effect, probably attributable to endotoxin (a bacterial cell wall component well known to induce inflammatory reactions). The exposures eliciting an effect were well in excess of normal human exposures. In an earlier study, Gerlofs-Nijland et al. (2007) exposed spontaneously hypertensive rats to roadside particulate matter by installation and elicited effects upon lung inflammation and blood fibrinogen at high exposure concentrations which were greater in coarse particles than fine. Lung inflammation was also observed in a study of mice dosed with urban particulate matter by installation, also greater with coarser particle fractions (Happo et al., 2010).

Taken together, the rodent exposure studies do not give a very consistent picture, with outcomes which are positive in some studies proving negative in others. Significant effects are generally seen only at higher concentrations, well above human exposures from ambient air. Several studies have reported higher toxicity associated with coarse fraction particles, suggesting a possible association with the non-exhaust particle component of ambient air, although the presence of endotoxin may be significant. There is

insufficient evidence to attribute effects to the nanoparticle fraction specifically. There is some evidence of differential toxicity of particles from different sources, but this is insufficient to conclude definitively that non-exhaust particles are of greater or lesser toxicity than diesel exhaust. Kreider et al. (2019) have applied temporal extrapolation and allowance for differential respiratory tract deposition efficiencies to their studies of rat inhalation exposure to TRWP to propose a NOAEL in humans of 55 μg m^{-3}, but this takes no account of subtle effects, sensitive individuals or inter-species extrapolation and cannot be considered as an appropriate guideline for human populations.

2.3 Results of Epidemiological Studies

A number of population-based epidemiological studies, summarised in Table 7, have sought to quantify the human health impacts of a non-exhaust fraction of road traffic emissions, although careful attention needs to be given to the methods used to identify and quantify the non-exhaust component.

Rich et al. (2019) report a time series study of cardiovascular hospital admissions using source-specific particle concentrations. The source apportionment was carried out by Positive Matrix Factorization by one of the leading groups in this field. The chemical profiles of the sources are in a companion paper and allowed the separate identification of diesel exhaust, gasoline exhaust and road dust sources. Five separate health outcomes were evaluated against 12 particle source categories. Significant associations ($p < 0.017$) were for gasoline exhaust with ischemic stroke at lag day zero, diesel exhaust with congestive heart failure and ischemic heart disease, both at lag day zero and road dust with ischemic heart disease at lag day zero. Since particles from gasoline engines, diesel engines and road dust are likely to be quite strongly correlated, attributing any of the effects unequivocally to road dust would be inappropriate. The road dust was noted as containing a number of transition metals including copper, iron and zinc which suggest contributions from both brake and tyre wear.

In a panel study of in-vehicle environmental exposures, Golan et al. (2018) measured changes in acute pulmonary and inflammatory responses. Organic carbon and several elements including Cu, Fe, S and Sb

were negatively associated with FEV1 (a measure of lung function) at a confidence level of $p < 0.05$. There were also marginal negative associations between FEV1 and elemental carbon particle number count, particle bound PAH and total $PM_{2.5}$. The elements Cu, Fe and Sb are strongly linked with brake wear, but organic carbon would more probably be linked with vehicle exhaust emissions or other non-traffic sources.

In a panel study of commuters, Krall et al. (2018) related source specific pollution exposures to pulmonary responses. Positive Matrix Factorization was used to identify four categories of particles, crustal non-tailpipe, primary tailpipe and secondary. The non-tailpipe particles were characterised by a high contribution to concentrations of Ba, Fe, Mn and S suggesting a strong contribution from brake wear, whereas the primary tailpipe particles were responsible for a large proportion of the organic carbon. Amongst asthmatic subjects, changes in lung function were associated with the crustal and secondary pollution factors whilst amongst non-asthmatic subjects, non-tailpipe pollution was associated with a pulmonary response only at 2.5 hours post-commute and no significant associations were found with primary tailpipe pollution.

Associations between right ventricular function and five year residential concentrations of coarse particle mass, copper, zinc, phosphorus, silicon and endotoxin were examined by D'Sousa et al. (2017), but no particle source apportionment was carried out. Positive associations of right ventricular mass, and to a lesser extent diastolic volume with coarse particle mass were found amongst susceptible populations. There was no clear pattern of associations with any of the chemical constituents analysed.

In a time series design, associations between resuspended mineral dust with emergency hospitalisations for three major respiratory causes were investigated in Hong Kong by Pun et al. (2017). Positive Matrix Factorization was used to source apportion PM_{10}, and a mineral dust factor was identified using aluminium, calcium and iron as tracer elements. Since such elements are associated with natural dusts and soils, this could not be considered as a surrogate for non-exhaust emissions, although there may be some contained within it.

The effects of a number of air pollutants and noise pollution on birth weight were studied by Smith et al. (2017) within a cohort in London. Air pollutant concentrations were estimated as monthly means by numerical modelling which included estimation of a $PM_{2.5}$ traffic exhaust and $PM_{2.5}$ traffic non-exhaust concentration. Only $PM_{2.5}$ mass and $PM_{2.5}$ (traffic exhaust) were consistently associated with increased risk of term low birth weight after adjustment for other pollutants.

Carey et al. (2016) report concentrations of a range of air pollutants including particle mass and $PM_{2.5}$ attributable to exhaust and non-exhaust sources estimated by numerical modelling in a study of cardiorespiratory outcomes in an adult cohort in London. The largest observed associations were between traffic related air pollution and heart failure. The strongest predictors were NO_x and percent of $PM_{2.5}$ due to traffic, but separate results for $PM_{2.5}$ from exhaust and non-exhaust sources are not presented.

Exposures of children to Cu, Fe, K, Ni, S, Si, V and Zn in both the $PM_{2.5}$ and PM_{10} size fractions were estimated by land use regression modelling and related to blood pressure measurements performed at age 12 in a study by Bilenko et al. (2015). Significant associations after control for potential confounders were found between diastolic blood pressure and Fe, Si and K in PM_{10}. Marginally significant positive associations were also found between Fe and Si in $PM_{2.5}$ and diastolic blood pressure. Effects were also associated with NO_2 exposure. These elements are associated with natural dusts and soils and cannot be taken as indicative of exposure to non-exhaust particles.

Associations were sought between local pollution averaged over the period 2005-2012 and the incidence of stroke for the population of south London (Crichton et al., 2016). Significant associations were found between total anterior circulation infarcts, but not other stroke types, and concentrations of PM_{10}, $PM_{2.5}$ and exhaust $PM_{2.5}$, but not non-exhaust $PM_{2.5}$. Both the exhaust and non-exhaust fractions were estimated using a dispersion modelling approach. In a subsequent paper (Desikan et al., 2016), the same group studied associations between air pollutant concentrations and mortality in the five years following a stroke.

The only pollutant metric showing significant positive associations was $PM_{2.5}$, with neither the exhaust or non-exhaust fractions significant.

Dadvand et al. (2014) report a study of particulate matter exposure and preeclampsia using PM_{10} and $PM_{2.5}$ mass and sources as exposure categories. Source apportionment of particulate matter was carried out by Positive Matrix Factorization which revealed eight factors of which vehicle exhaust was characterised by a high concentration of total carbon while brake wear showed high contributions from Ca, Fe, S, Cu and nitrate. Unfortunately, the paper does not show the explained variations for these components which would allow a more confident attribution to sources. In addition to the separate source categories, a combined traffic-related source category was obtained from the sum of the vehicle exhaust and brake dust and 70% of secondary nitrates/organics. The only strongly positive associations between risk of preeclampsia and exposure during the entire pregnancy were with PM_{10} brake dust (44% increase (95% CI, 7%-94%)) and combined traffic-related sources (80% increase (95% CI, 4%-211%)). The results for $PM_{2.5}$ mass and other sources, and also for exposure during trimester one were inconclusive.

A study of nine healthy young non-smoking male patrol officers with measurements of in-vehicle $PM_{2.5}$ was reported by Riediker et al. (2004). A principal factor analysis method was used to identify up to four source related factors described as soils, automotive steel wear, gasoline combustion and speed-changing traffic, the latter being characterised by high concentrations of copper, sulphur and aldehydes suggesting a possible brake wear component. Source factor scores were compared to cardiac and blood parameters measured 10 and 15 hours after each shift. Significant associations of a range of cardiovascular symptoms and markers were associated with the speed-change factor and a lesser number with a crustal factor. It was concluded that $PM_{2.5}$ associated with speed-changing traffic led to effects upon the autonomic control of heart rhythm, and the frequency of premature supraventricular beats as well as eliciting pro-inflammatory and pro-thrombotic responses. However, the link to non-exhaust particles is extremely tenuous because of a very limited range of chemical analyses conducted on the particulate matter and the relatively crude method used for source apportionment.

These studies, all identified by search terms relating to non-exhaust emissions, are highly diverse in terms of design, health outcome and means of identification/characterisation of non-exhaust emissions. Some report positive findings in relation to exposure to a "non-exhaust" component, although this is only one of many tests conducted and may have arisen by chance. The strong correlation of non-exhaust emissions with exhaust emissions causes difficulties in cleanly separating the two components, meaning that some effects due to exhaust emissions may be inaccurately attributed to non-exhaust emissions, and *vice versa*. Moreover, some studies use crustal elements to identify the non-exhaust particle component, leading to likely confusion with wind-blown soil or road dust. Viewed together, these epidemiological studies do not have the consistency and coherence that would give confidence in attributing any adverse effects to exposure to non-exhaust particulate matter.

2.4 Carcinogenicity of Antimony

Antimony is a component of many brake pad formulations, and is hence also present in brake wear particles. Although not prohibited, there appears to be have been a reduction in antimony use in brake pads over recent years, reflected in smaller Sb/Cu ratios in brake wear particles. According to the International Agency for Research on Cancer (IARC), Sb is classified as 2B: possibly carcinogenic in humans. A review of occupational exposure studies (Saerens et al, 2019) found the evidence for occupational carcinogenicity to be inadequate, but on the basis of the human studies available and effects in experimental animals recommended classification as 2A; probably carcinogenic in humans. However, they were unable to define a range of concentrations over which carcinogenicity occurs, or to recommend exposure-response functions. It therefore seems likely that the cancer risk associated with exposure to the very small antimony concentrations in ambient air is so low as to be negligible.

2.5 Synthesis of Health-Related Studies

As discussed above, the in vitro and in vivo studies are suggestive of toxicity, but do not provide a basis from which to predict effects on human health. The population-based epidemiology could allow the establishment of exposure-response functions for human populations, but currently the studies performed

to date have insufficient coherence to do so, as well as there being doubts over the reliability of their identification of non-exhaust particles in many cases.

The quantification of differential toxicity of airborne particles on the basis of chemical composition, particle size, or source would be of great value in the development of cost-effective abatement strategies. However, the large published literature which has addressed this issue has attributed toxic effects to a very wide range of constituents and sources. It is not clear whether this is a consequence of the imprecise nature of the epidemiological methods, or of toxicity residing in all species irrespective of composition and size (within a given fraction such as $PM_{2.5}$). In this context, it is important to recognise that classical mechanisms of toxicity are not relevant – concentrations of trace metals, for example in urban air are well below those necessary to elicit the effects upon cardiorespiratory health associated with $PM_{2.5}$ exposure. There are a number of mechanisms by which airborne particles are believed to exert toxic effects, the main one being through oxidative stress. However, it seems unlikely that all particles are of equal toxicity irrespective of composition, as, for example, the small number of human challenge studies suggest that some particle types, such as diesel exhaust elicit significant biochemical and physiological effects, while others such as ammonium sulphate appear far less toxic in challenge studies, but this is not reflected in the outcomes of epidemiological studies. As a consequence, many authoritative bodies including the USEPA (2009), WHO (2013), UK expert committee (COMEAP, 2015), and French expert panel (ANSES, 2019) who have examined the evidence have been unable to identify any clear pattern of relative toxicity. Resolving this conundrum is currently a major focus of research, and recognising the contradictions, the UK Committee on the Medical Effects of Air Pollutants (COMEAP, 2015) recommends that all particle types within a size fraction such as $PM_{2.5}$ are treated as if they contribute equally per unit mass to toxicity. Although this is not stated policy in all other jurisdictions, the implementation of air quality policy based upon PM mass as practised worldwide is an implicit acceptance of this concept. The consequence is that concentrations of non-exhaust particles, irrespective of source should be viewed in the light of their contribution to overall concentrations of $PM_{2.5}$ and PM_{10}, in the context of the air quality guidelines and standards relating to those metrics.

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Table 1: Overview of most common key tracers used for brake wear emissions (based upon Grigoratis and Martini, 2015).

Reference	Tracer	Reference	Tracer
Sternbeck et al., 2002	Ba, Cu, Sb	Gietl et al., 2010	Ba, Cu, Fe, Sb
Adachi and Tainosho, 2004	Ba, Ce, Cu, Fe, La, Sb, Ti, Y, Zr	Keuken et al., 2010	Cu
Schauer et al., 2006	Ba, Cu, Fe, Sb, Si, Zn	Amado et al., 2011	Cu, Cr, Fe, Sb, Sn, Zn
Hjortenkrans et al., 2007	Cd, Cu, Pb, Sb, Zn	Apeagyei et al., 2011	Cu, Ba, Fe, Mo, Ti, Zr
Harrison, 2009	Ba, Cu	Duong and Lee, 2011	Cu, Ni
Iijima et al., 2008	Sb	Song and Gao, 2011	Sb, Cu, Fe, Pb
Tanner et al., 2008	Cu, Cd	Harrison et al., 2012	Sb, Cu, Fe, Pb
Bukowiecki et al., 2009	Cu, Fe, Mo, Sb, Sn, Zn, Zr	Lawrence et al., 2013	Ba, Cu, Fe, Mn, Ni, Pb, Sb
Dongarra et al., 2009	Cu, Mo, Sb	Varrica et al., 2013	Sb
Lough et al., 2005	Cu, Zn, Sb, Ba	Dong et al. (2017)	Fe, Ba, Sb
Charron et al. (2019)	Cu, Fe, Sn		

Table 2: Emission factors from PM_{10} from tyre and brake wear (from AQEG, 2019).

mg PM_{10} / km		**Tyre**	**Brake**
Cars	Urban	8.7	11.7
	Rural	6.8	5.5
	Motorway	5.8	1.4
LGVs	Urban	13.8	18.2
	Rural	10.7	8.6
	Motorway	9.2	2.1
Rigid HGVs	Urban	20.7	51.0
	Rural	17.4	27.1
	Motorway	14.0	8.4
Articulated HGVs	Urban	47.1	51.0
	Rural	38.2	27.1
	Motorway	31.5	8.4
Buses	Urban	21.2	53.6
	Rural	17.4	27.1
	Motorway	14.0	8.4
Motorcycles	Urban	3.7	5.8
	Rural	2.9	2.8
	Motorway	2.5	0.7

Table 3: Literature estimations of the contribution of tyre wear particles to airborne PM (%) (from Grigoratos and Martini, 2014).

Reference	**PM_{10}**	**$PM_{2.5}$**
Annema et al., 1996	5.0	-
Israël et al., 1994	10	-
Rauterberg-Wulff, 1999	5.0	-
Fauser, 1999	5.0	-
Hüeglin and Gehrig, 2000	1.0-7.5	-
Gaultieri et al., 2005a,b	3.0-7.0	-
BLIC, 2005	6.0	3.0
Sjodin et al., 2010	0.1-3.9	-
Panko et al., 2013	0.84	-
Kwak et al., 2013	3-4	4.7

Table 4: Percentage contributions of road dust sources to airborne PM concentrations (based upon Gulia et al., 2019).

Authors	Study location	Study period	Contribution from non-exhaust PM emissions
Gupta et al. (2007)	Kolkata, India	November 2003-November 2004	PM_{10} (paved road in residential area): 21%
Amato et al. (2009a)	Barcelona, Spain	15-22 June 2007	PM_{10} (urban environment) City centre (8.9%), ring road (34.5%) Harbour (1.65%)
Amato et al. (2011)	Zurich, Girona, Barcelona	Zurich: February 2008 Girona: July 2008 Barcelona: June 2007	Barcelona and Girona: 30-60% (road wear) Zurich: 15%-30% (road wear)
Karanasiou et al. (2011)	Madrid, Spain	22-24 September 2010	Road dust contribute 29% of ambient PM_{10} in urban area
Gummeneni et al. (2011)	Hyderabad, India	January 2004–May 2005	Contribution of road dust at urban location in PM_{10} (40%) and $PM_{2.5}$ (26%)
Gupta et al. (2012)	Mumbai, India	April 2007-March 2008	Contribution of road dust in PM_{10} level Industrial: 27%; Urban residential area: 18%; Slum residential area: 15%
Pipalatkar et al. (2014)	Nagpur, India	Residential: September-November 2009 Commercial: December 2009-January 2010 Industrial: January-February 2010	Contribution of road dust in $PM_{2.5}$ level Residential: 6%; Commercial: 10%; Industrial: 7% Contribution of road dust in ambient PM_{10} level
Amato et al. (2016a,b)	Barcelona, Spain	February-March 2009	Background site: 9-15%; Kerbside: 23-44%

Table 5: In vitro tests of toxicity of source samples and/or airborne particles.

Reference	Material tested	Test methods
Godri et al. (2011)	Size-fractionated airborne particles	Oxidative potential (AA and GSH)
Shirmohammadi et al. (2016)	Roadside particles	Oxidative potential (DTT)
Daellenbach et al. (2020)	Airborne particles	Oxidative potential (DTT, OCFH, AA)
Gualtieri et al. (2005a)	Tyre particles	Trypan blue, Comet, Cell morphology
Gualtieri et al. (2005b)	Tyre particles	MTT, LDH, Comet, Cell morphology
Gualtieri et al. (2008)	Tyre particles	DCFH, Hsp70 expression
Wessels et al. (2010)	Size fractioned airborne particles (4 sites)	Hydroxyl generation, LDH, IL8, Comet
Gasser et al. (2009)	Brake wear	LDH, IL8, Oxidative stress
Barosova et al. (2018)	Brake wear	GSH, IL8, TNF- α
Jalava et al. (2015)	Size fractionated airborne particles	NO, IL6 release, cell viability
Gustafsson et al. (2008)	Road wear; airborne particles	IL6, IL8, TNF- α
Kazimirova et al. (2016)	Brake wear	Genotoxicity
Peikertova et al. (2017)	Brake wear	Phagocytosis, micronuclear breaks
Malachova et al. (2016)	Brake pads	Bioilum, Ames
Puisney et al. (2018)	Brake wear	WST-1, DCFH, IL6, IL8, TNF-α
Selley et al. (2020)	Brake wear and diesel exhaust particles	IL8, IL10, TNF-α, phagocytosis

Table 6: In vivo tests of toxicity source materials and/or airborne particles.

Reference	Material tested	Test methods
Gottipolu et al. (2008)	Tyre particles	Lung inflammation (rats)
Mantecca et al (2009)	Tyre particles	Lung inflammation (rats), LDH, BALF, GSH, TNF-α, MIP2
Kreider et al. (2012)	Tyre and road wear particles	Lung pathology (rats)
Bernstein et al. (2014)	Brake dust with 30% chrysotile	Lung macrophages (rats)
Bernstein et al. (2015)	Brake dust with 30% chrysotile	Lung macrophages (rats)
Bernstein et al. (2018)	Brake dust with 30% chrysotile	Lung macrophages (rats), BALF cells, LDH, cytokines
Gerlofs-Nijland et al. (2007)	Roadside particulate matter	BALF, blood fibrinogen (rats)
Gerlofs-Nijland et al. (2019)	Brake, tyre, road wear and airborne particles	Lung inflammation (mice), LDH, GSH, BALF, blood leucocytes and fibrinogen
Happo et al. (2010)	Urban airborne particles	Lung inflammation (mouse), BALF, TNF-α, IL6, KC

Table 7: Epidemiological studies with a measure of non-exhaust particles.

Reference	Measure of non-exhaust particles	Health outcome investigated
Rich et al. (2019)	Road dust (from PMF)	Cardiovascular hospital admissions
Golan et al. (2018)	Brake wear (elemental tracers)	Respiratory symptoms
Krall et al. (2018)	Crustal non-tailpipe (from PMF)	Lung function changes
D'Sousa et al. (2017)	Elemental tracers	Cardiac function
Pun et al. (2017)	Mineral dust (from PMF)	Respiratory emergency admissions
Smith et al. (2017)	Non-exhaust particles (from model)	Birth weight
Carey et al. (2016)	Non-exhaust particles (from model)	Cardiorespiratory health
Bilenko et al. (2015)	Elemental tracers	Blood pressure
Crichton et al. (2016)	Non-exhaust particles (from model)	Incidence of stroke
Desikan et al. (2016)	Non-exhaust particles (from model)	Mortality from stroke
Dadvand et al. (2014)	Brake dust (from PMF)	Preeclampsia
Riediker et al. (2004)	Brake wear (from Factor Analysis)	Cardiovascular symptoms

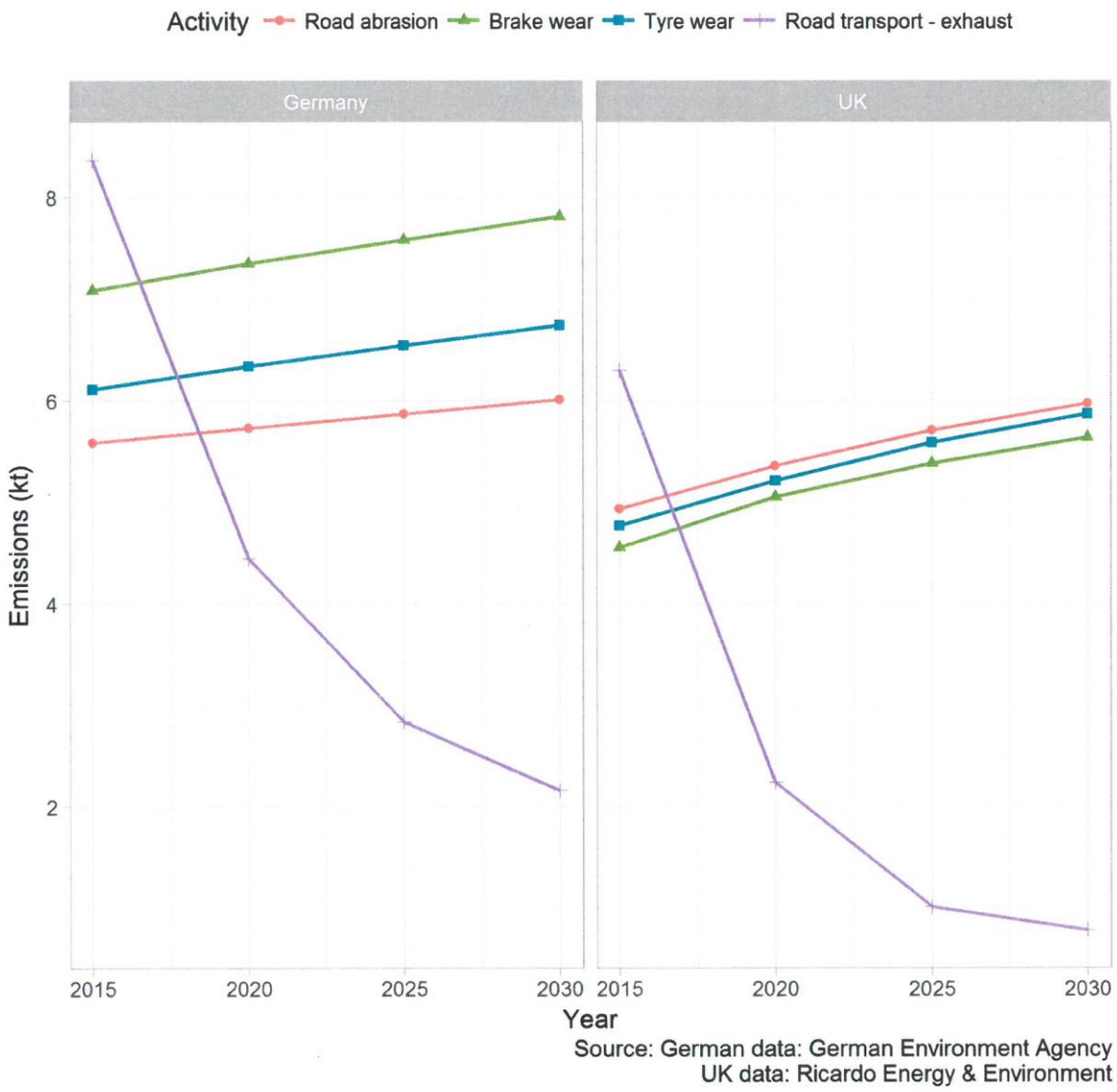

Figure 1: PM_{10} emissions from road transport sources according to the UK and German Inventories. The scale on the y-axis applies to both countries (from Wakeling et al.,2017).

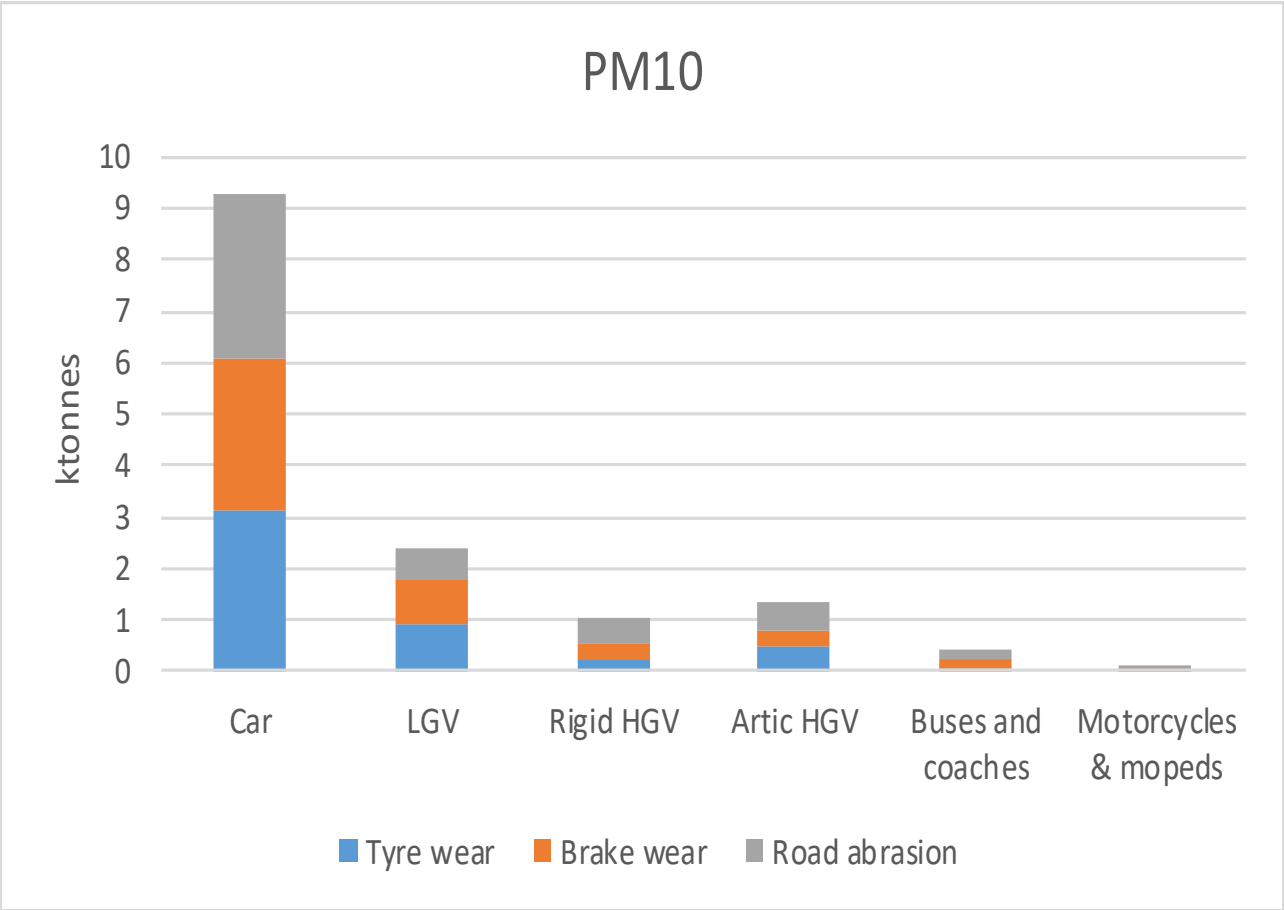

Figure 2: UK non-exhaust emissions (tonnes/year) for different vehicle classes for 2016 from the UK National Atmospheric Emissions Inventory (from AQEG, 2019).

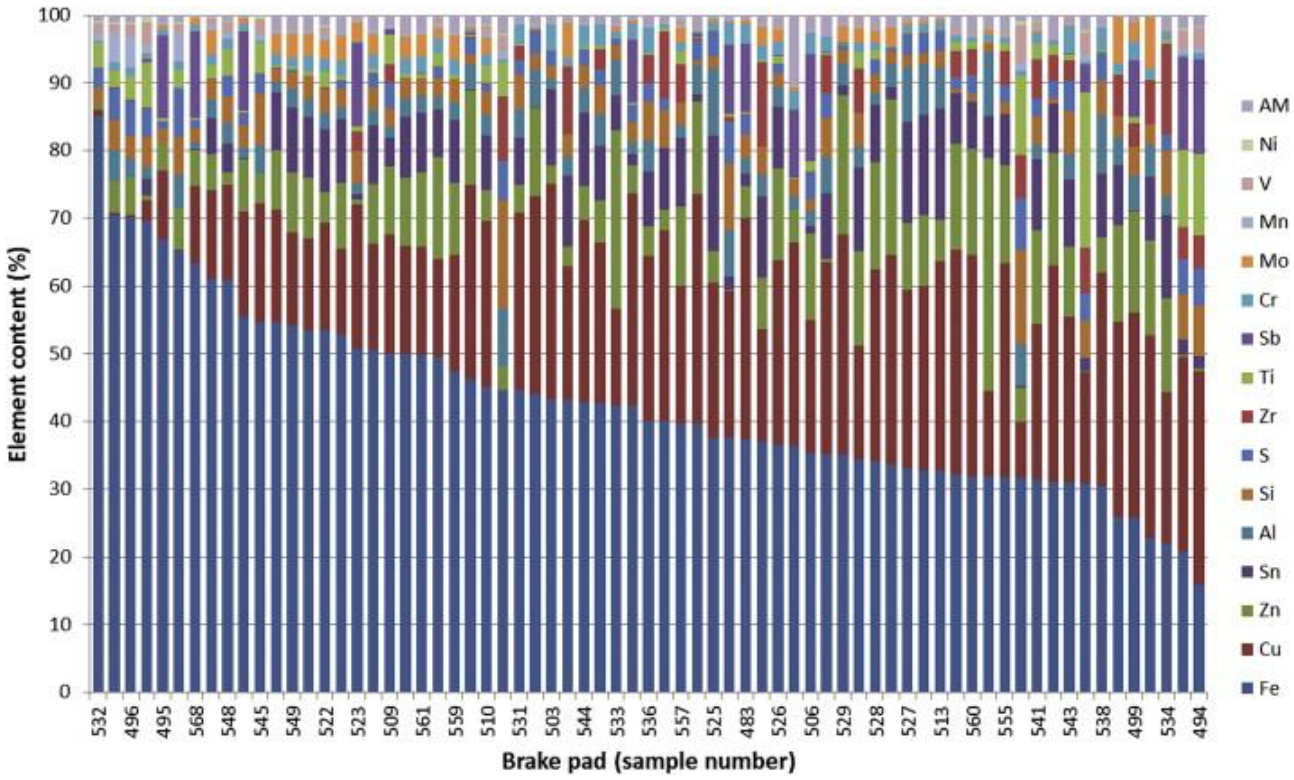

Figure 3: Elemental content of metals in brake pads, as measured by Hulskotte et al. (2014).

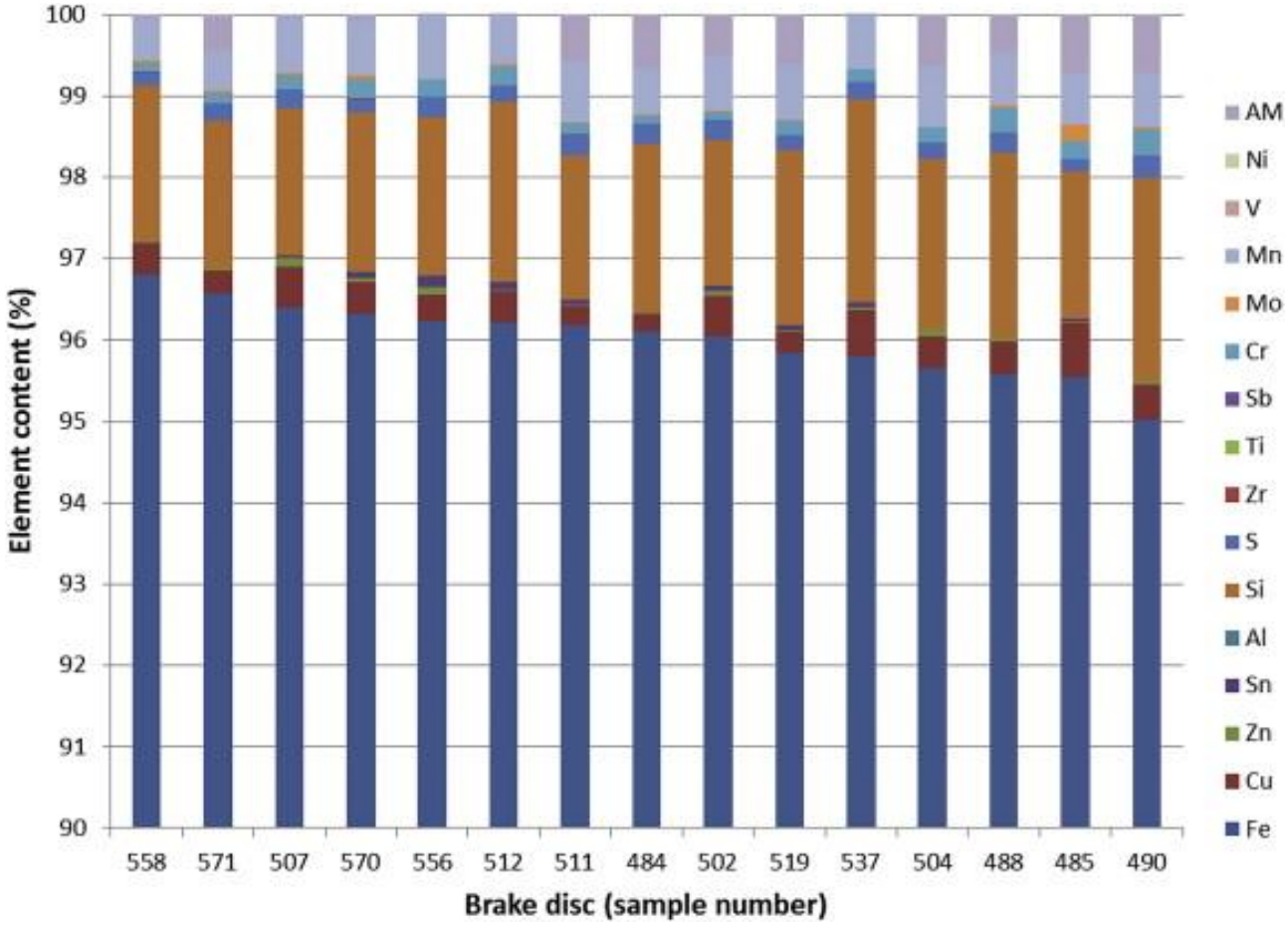

Figure 4: Elemental content of metals in brake discs, as measured by Hulskotte et al. (2014).

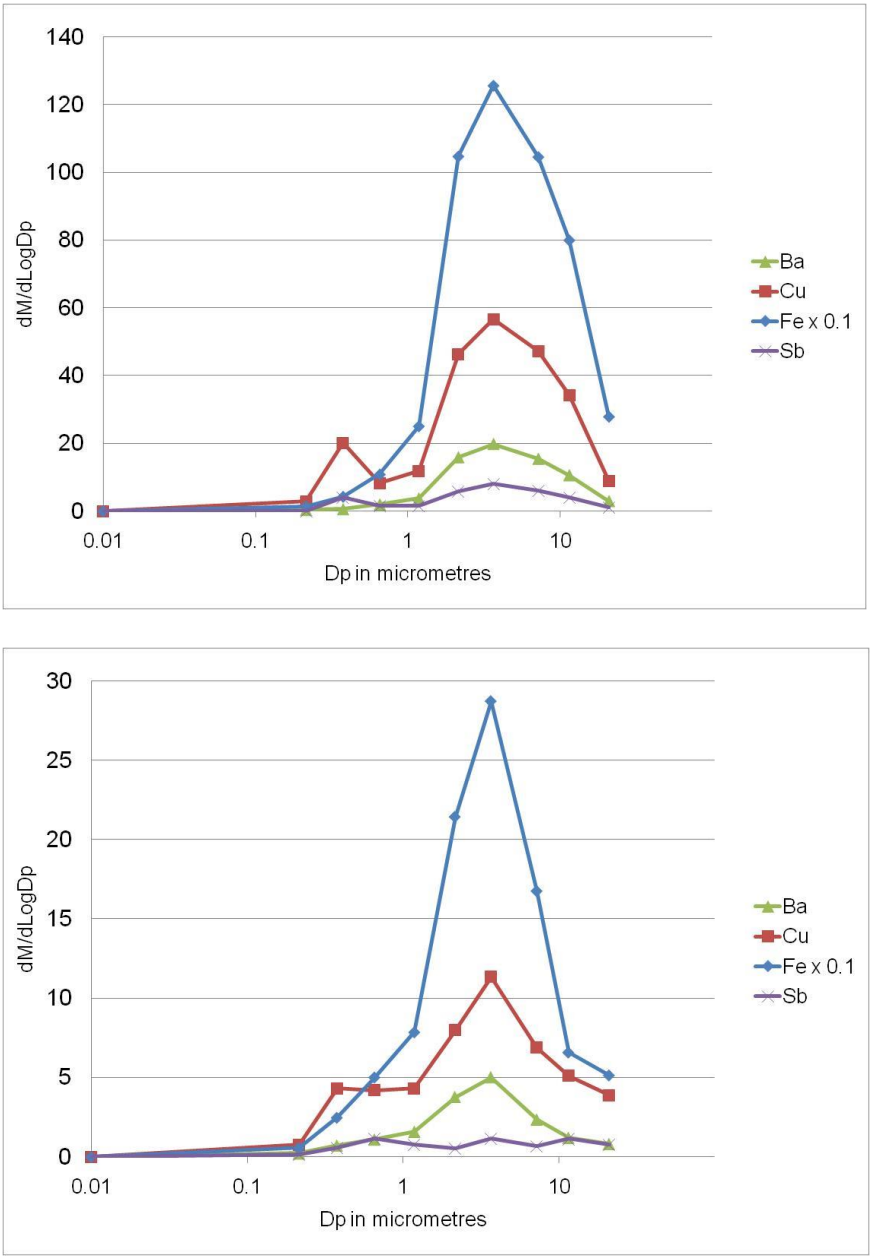

Figure 5: Size Distribution of Ba, Cu, Fe, and Sb at (a) Marylebone Road and (b) Regent's Park (ng m^{-3}) (from Gietl et al., 2010).

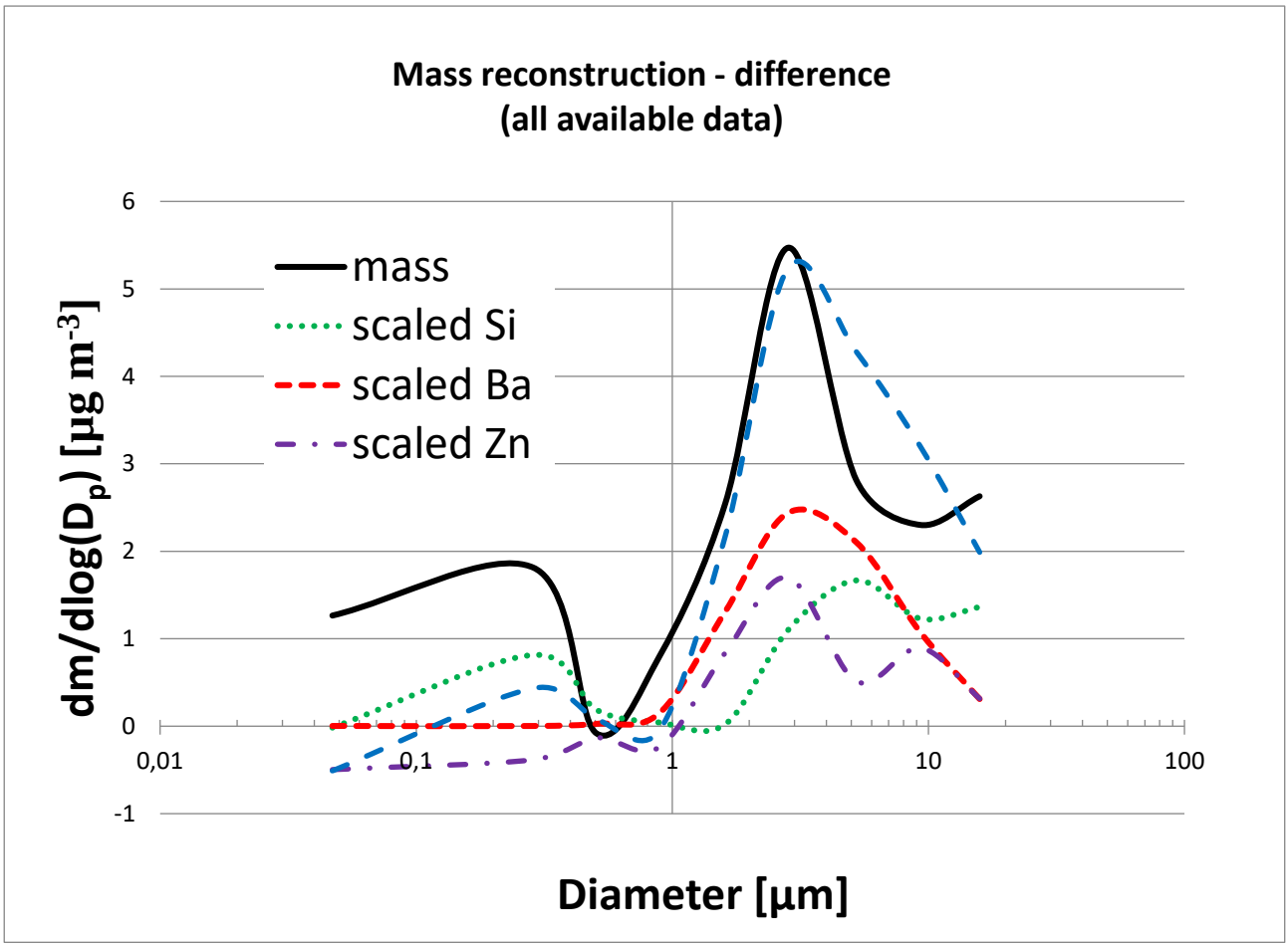

Figure 6: Particle mass size distributions of Ba, Si and Zn scaled to represent the masses of brake wear, resuspension and tyre wear respectively, compared to measured total mass in the roadside increment at London, Marylebone Road (from Harrison et al., 2012).

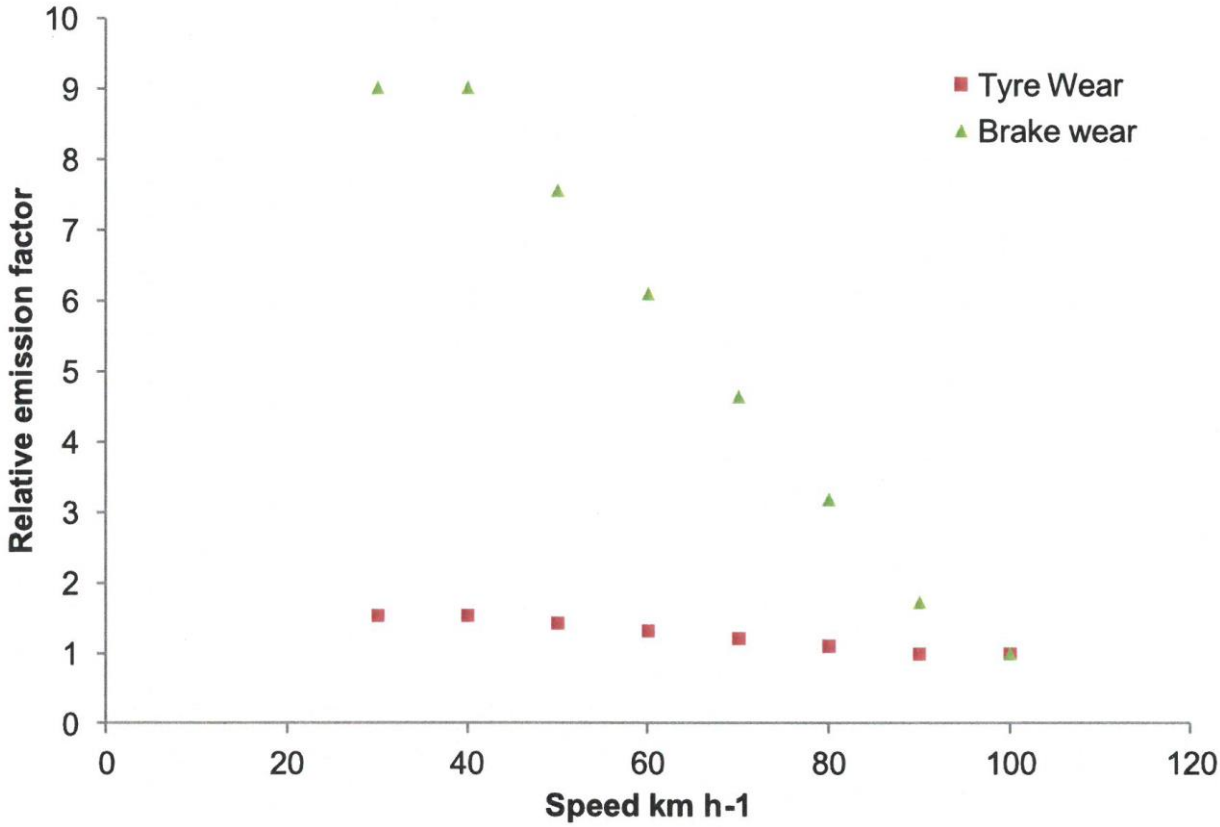

Figure 7: Speed dependence of emissions factors for brake and tyre wear. Each are normalised to their respective emissions rate at 100 km h^{-1}. In this plot speed refers to the average traffic speed and not the instantaneous speed of any vehicle (from AQEG, 2019).

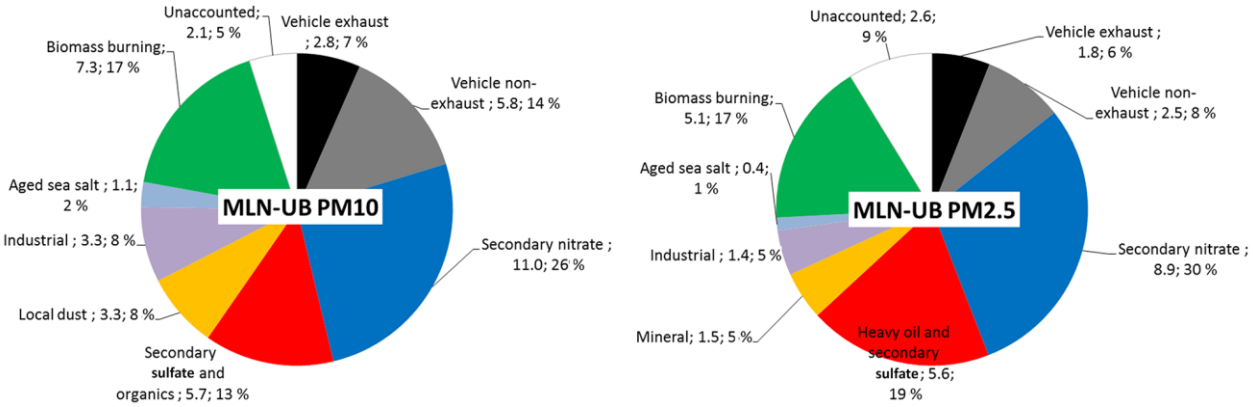

Figure 8: Source apportionment of $PM_{2.5}$ and PM_{10} in air samples from a Milan urban background site (from Amato et al., 2016c).

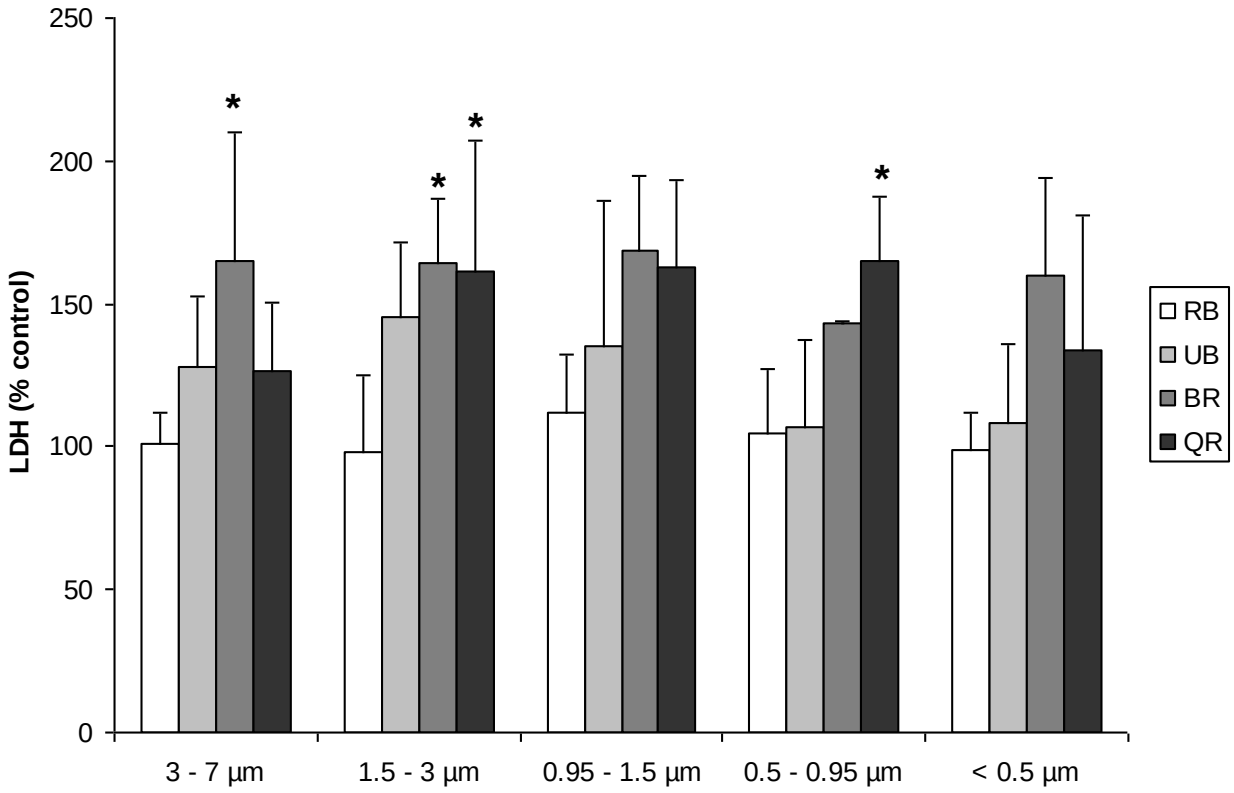

Figure 9: Cytotoxicity measured as the release of lactate dehydrogenase (LDH) in A549 cells after treatment with the different PM samplers for 24 h at a [C] or 100 µg/cm^2. Data is shown as percentage of control (untreated A549 cells) for constant mass. *P < 0.05 vs RB within the same size fraction (ANOVA with Dunnett). Two factor ANOVA analysis of the full dataset, revealed no significant contrasts related to particle size, upon adjustment for sampling location (from Wessels et al., 2010).

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