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**A REVIEW OF EMISSION FACTORS AND MODELS FOR ROAD
VEHICLE NON-EXHAUST PARTICULATE MATTER**

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Prepared for: Project Record: **Non-exhaust particulate matter emissions from road traffic**

Client: **Department for the Environment, Food and Rural Affairs, Scottish Executive, Welsh Assembly Government, and the Department of Environment in Northern Ireland**

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Executive summary

TRL Limited, the Division of Environmental Health & Risk Management (DEHRM) at Birmingham University and Cambridge Environmental Research Consultants Limited (CERC) have been commissioned by DEFRA to investigate non-exhaust particulate matter (PM) emissions from road traffic. The main aim of the project is to develop improved prediction methods for emissions and air pollution, primarily for use in the UK National Atmospheric Emissions Inventory (NAEI). The project is divided into five main phases:

- (i) A literature review.
- (ii) Emission model development and application.
- (iii) Initial air quality model development and application.
- (iv) Further air quality model development.
- (v) Discussion of abatement options.

This Report presents the findings of the literature review, which reflects the need to examine the current understanding of non-exhaust particulate matter. The main objectives of the review are to collate the available information on particle emissions from road vehicle non-exhaust sources, to summarise the methodologies currently employed to measure and model emissions from these sources, and to provide general recommendations for model development during the remainder of the project. It includes a substantial amount of information relating to the composition of source materials and particles.

There are a number of non-exhaust processes, involving mechanical abrasion and corrosion, which can result in particulate matter being released directly to the atmosphere. The most important direct emission sources are tyre wear, brake wear, and road surface wear. Clutch wear and corrosion may also contribute to direct PM emissions, but these cannot easily be quantified given the current level of understanding. In addition to direct non-exhaust emissions, material previously deposited on the road surface can be suspended or resuspended in the atmosphere as a result of tyre shear, vehicle-generated turbulence, and the action of the wind. Indeed, of the non-exhaust processes resuspension is likely to be the largest contributor to roadside PM₁₀¹ concentrations. A large number of factors affect the emissions from each source, but these factors have not been fully investigated. Furthermore, the contribution of each source to airborne PM varies considerably, both temporally and spatially.

Non-exhaust particulate matter is important for a number of reasons, including the following:

- (i) There are no EU regulations specifically designed to control non-exhaust particle emissions, though some countries have banned the use of studded tyres.
- (ii) As exhaust emission control technology improves and traffic levels increase, the proportion of total PM emissions originating from the uncontrolled non-exhaust sources will increase.
- (iii) The data relating to the emission rates, physical properties, chemical characteristics, and health impacts of non-exhaust particles are highly uncertain.

¹ PM₁₀ represents the mass concentration of particles passing through a size-selective inlet designed to exclude particles greater than 10 µm aerodynamic diameter.

There is a general lack of consistency in the definitions, terminology and metrics used in the study and reporting of non-exhaust particulate matter. In particular, where emission factors for resuspension are reported, it is not always clear whether they include primary emissions due to abrasion, and a number of modelling methodologies consider abrasion sources but not resuspension. This often renders the incorporation of data into models, and comparisons of model predictions with earlier studies, rather difficult. Ultimately, the various sources need to be considered as independently as possible, and causal relationships need to be identified.

For 'normal' driving conditions, it seems that a wear factor² for tyres of 100 mg/vkm would probably represent the right order of magnitude. For heavy-duty vehicles, the wear factor is likely to be an order of magnitude higher. More information is required to relate tyre wear (and PM emissions) to real-world driving patterns in a quantitative manner. The literature indicates that probably less than 10% of car tyre wear material is emitted as PM₁₀ under 'typical' driving conditions, but the proportion could be as high as 30%. Although it is commonly assumed that airborne tyre wear particles are mainly coarse (larger than 2.5 µm in diameter), finer particles are also emitted directly to the atmosphere.

Total brake wear for light-duty vehicles appears to be around 10 to 20 mg/vkm, and around 50 to 80 mg/vkm for heavy goods vehicles. Typically 50% of brake wear debris from LDVs escapes the vehicle and enters the atmosphere, and more than 80% of airborne brake wear particles can be classified as PM₁₀. A substantial amount of the PM₁₀ can also be present as PM_{2.5}³. There is considerable uncertainty regarding the amount of material which is lost from the brake linings, and the amount which is lost from the disc or drum.

A wide range of road surface wear factors have been reported in a small number of studies, from less than 4 mg/vkm to more than 400 mg/vkm where the surface contains a high proportion of bitumen. However, in areas where there is extensive use of road sanding and studded tyres during the winter, the wear of the road surface is considerably higher - values of between 4 and 24 g/vkm have been reported in some Nordic countries. Very little information on the size distribution of road surface wear particles is available.

Resuspension is probably the single largest vehicle non-exhaust contributor to roadside PM₁₀, particularly where winter maintenance procedures are in place. Although some information on the effects of winter maintenance is available for Nordic countries, the situation in the UK is rather different. In the UK, road de-icing involves the application of rock salt, but little information of the effects of this particular approach on resuspension has been reported. This specific subject requires further investigation.

Models for non-exhaust PM emissions are generally rather crude. The use of average brake wear emission factors (in g/km) in models does not seem particularly logical, as the differences in the extent of braking for different traffic situations cannot be taken into account. Such emission factors are likely to over-estimate brake emissions in areas of low brake usage, and under-estimate emissions in areas of high brake use. More detailed methodologies for estimating brake wear emissions are required.

Emission factors for resuspension in Europe are variable, and most of the available information is derived from Nordic countries where the use of studded tyres presents a significant problem.

² Defined here as the total amount of material lost per vehicle kilometre (vkm).

³ PM_{2.5} represents the mass concentration of particles passing through a size-selective inlet designed to exclude particles greater than 2.5 µm aerodynamic diameter.

There also appear to be some fundamental difference between non-exhaust processes in the US, and those in Europe. Total PM₁₀ emission factors in the US generally appear to be substantially higher than those in Europe. This may be related to the particularly dry and dusty conditions at the locations of US studies. US prediction models, such as the AP-42 are therefore unlikely to be appropriate to the UK.

Some specific recommendations are made for the next phase of the project. These include obtaining more detailed information on the non-exhaust PM models covered in the review, as well as any data relating to UK conditions which are required to run the models. The performance of the selected models should be evaluated for a range of typical UK conditions, using real-world data where possible. Following this comparison, one or more models should be selected for further development. It is expected that rather different modelling approaches will be required for direct emission sources and resuspension, and a number of alternative development methods should be explored.

The review gives no indication of appropriate mechanisms for controlling non-exhaust particles. The effects of different road vehicle/traffic controls upon coarse particle concentrations are not predictable in the absence of tested algorithms for calculating resuspended dust concentrations applicable to European conditions. In particular, the complex inter-relationships between vehicle/traffic operation, road surface dust silt loadings and resuspension have not been determined. Potential abatement options will be reviewed qualitatively (and quantitatively as far as possible) later in the project.

A number of general recommendations are also provided in order to improve the understanding of non-exhaust PM in the UK. These go beyond the scope of the project, and include a survey of tyre, brake and road surface materials in use in the UK, the derivation of component wear factors and vehicle operation effects, more detailed source characterisation, laboratory-based measurements, real-world measurements using instrumented vehicles, and further receptor modelling studies.

1 Introduction

1.1 Overview

TRL Limited, the Division of Environmental Health & Risk Management (DEHRM) at Birmingham University and Cambridge Environmental Research Consultants Limited (CERC) have been commissioned by DEFRA to investigate non-exhaust particulate matter (PM) emissions from road traffic. The main aim of the project is to develop improved prediction methods for emissions and air pollution, primarily for use in the UK National Atmospheric Emissions Inventory (NAEI). The project is divided into five main phases:

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1.2 Airborne particulate matter

It is assumed that the reader is familiar with the general terminology employed to describe airborne particulate matter. Some of the terms and metrics in common use are explained in Table 1.1, and the following paragraphs provide a concise overview. Readers seeking a comprehensive discussion are directed to the reports of APEG (1999) and AQEG (2004).

Airborne particulate matter is a complex mixture of organic and inorganic substances, in solid or liquid form, which undergoes modification or transformation within the atmosphere. It is derived from a wide variety of sources, both natural and anthropogenic, and displays a range of physical and chemical properties. Particles are termed either 'primary', where they are emitted directly into the atmosphere, or 'secondary' where they are formed by reactions between gas-phase components such as sulphur and nitrogen oxides, ammonia, and organic compounds.

Particles in the atmosphere range in size from less than 10 nm to around 100µm. It is common to see them described in terms of three recognised modes relating to sources and particle size: the nucleation mode, the accumulation mode, and the coarse particle mode. The contributions of different sources to mass concentrations in these different modes vary with many factors, including location, season, time of day, and both local and regional weather conditions.

The nucleation mode consists of particles emitted directly from combustion sources, such as road vehicle exhaust, waste incineration, and industrial and domestic burning. Nucleation mode particles typically have a diameter of less than around 0.05 µm. Even though such particles may be present in large numbers, each particle is so small that this mode usually forms only a small proportion of the total aerosol mass. Nucleation mode particles reside in the atmosphere for a few hours, and are transformed by coalescence and condensation into larger accumulation mode particles.

Accumulation mode particles range between around 0.05 μm and 1 μm in diameter, have atmospheric residence times of tens of days, and usually form a significant fraction of the total aerosol mass. They are also efficient light scatterers, and often dominate optical effects such as visibility. As well as being formed via the coagulation of nucleation mode particles, accumulation mode particles originate from primary emission sources and gas-to-particle transformations in the atmosphere.

Particles larger than around 1 μm form the coarse particle mode, and typically include wind-blown crustal matter and material released during abrasion processes. It is clear that the alternative definition of coarse particles given in Table 1.1 (*i.e.* $\text{PM}_{2.5-10}$) is not altogether consistent with this definition. Coarse particles have shorter residence times than accumulation mode particles, although they can contribute substantially to total aerosol mass.

Table 1.1: Explanations of terms used to describe airborne particulate matter

| Term | Definition |
|---|---|
| Primary particles | Particles emitted directly to the atmosphere. |
| Secondary particles | Particles formed within the atmosphere from gas phase precursors. This includes particles originating from atmospheric oxidation of sulphur and nitrogen oxides, and their reaction products with ammonia, and from the oxidation of organic compounds. |
| TSP | Total suspended particulate. |
| PM_{10} | Mass concentration of particles passing through a size-selective inlet designed to exclude particles greater than 10 μm aerodynamic diameter. |
| $\text{PM}_{2.5}$ | Mass concentration of particles passing through a size-selective inlet designed to exclude particles greater than 2.5 μm aerodynamic diameter. These are sometimes referred to as 'fine' particles. |
| $\text{PM}_{2.5-10}$ or $\text{PM}_{\text{COARSE}}$ | Mass concentration of 'coarse' particles, determined as the difference between PM_{10} and $\text{PM}_{2.5}$. |
| PM_1 | Mass concentration of particles passing through a size-selective inlet designed to exclude particles greater than 1 μm aerodynamic diameter. |
| $\text{PM}_{0.1}$ | Mass concentration of particles of diameter smaller than 0.1 μm . These are sometimes referred to as 'ultrafine' particles. |
| Nucleation mode | Particles emitted directly from combustion sources, having a diameter of less than around 50nm and an atmospheric residence time of a few hours. They are transformed by coalescence and condensation into larger accumulation mode particles. |
| Accumulation mode | Particles formed via the coagulation of nucleation mode particles, primary emission sources, and gas-to-particle transformations. Particles range between around 0.05 μm and 1 μm in diameter, and have an atmospheric residence time of tens of days. |
| Coarse particle mode | Particles larger than around 1 μm , including wind-blown crustal matter and material released during abrasion processes. Coarse particles have shorter residence times than accumulation mode particles. This is not consistent with the definition for $\text{PM}_{\text{COARSE}}$ given above. |
| Dustfall | Particles larger than 100 μm , which tend to fall out of the atmosphere within minutes. |

1.3 Definition of non-exhaust particulate matter

The particulate matter generated by road transport activity can be categorised according to its mechanism of formation. It is often assumed that diesel exhaust is the main source of particulate matter from road vehicles, and exhaust emissions have been well characterised in the laboratory under well-defined test conditions. However, there are a number of non-exhaust processes, involving mechanical abrasion and corrosion, which can also result in particulate matter being released directly to the atmosphere. The main abrasion processes leading to the direct emission of particulate matter are tyre wear, brake wear and road surface wear. Other potential sources of direct emissions are clutch wear, engine wear, the abrasion of wheel bearings, and the corrosion of other vehicle components, street furniture and crash barriers. In addition to direct non-exhaust emissions, material previously deposited on the road surface can be suspended or resuspended in the atmosphere as a result of tyre shear, vehicle-generated turbulence, and the action of the wind. In the case of road transport, it is commonly assumed that most primary fine particles ($PM_{2.5}$) are emitted from the exhaust, whereas many of the coarse particles ($PM_{2.5-10}$) are considered to originate from non-exhaust sources. This over-simplifies the situation somewhat; whilst there is a general agreement that exhaust emissions can be classified as $PM_{2.5}$, there is evidence to suggest that non-exhaust particles contribute to both the fine and coarse modes.

1.4 Concerns relating to non-exhaust particulate matter

1.4.1 Lack of regulation

Compliance with air quality standards for PM_{10} requires control of both fine and coarse particles. As the two modes tend to have different sources and formation mechanisms, different types of control are required. Primary fine particles from combustion sources are subject to regulation. For example, all new light-duty vehicle (LDV⁴) models and heavy-duty engine models sold in the UK must be type approved with respect to exhaust emissions in accordance with European Union Directives. Since the first Directives were introduced in the early 1970s, they have been modified to include more pollutants among those controlled, to reduce the emission limit values, and to improve the test procedures. The measurement of total exhaust particulate is only defined in regulation for diesel engines and vehicles. The type approval emission limits for particulate matter are listed for LDVs (from 1988 onwards) in Table 1.2, and for heavy-duty engines (from 1991 onwards) in Table 1.3. Prior to 1988, there was no requirement to measure particulate mass. Light commercial and two-wheel vehicles are subject to slightly different legislation (not shown).

As a result of the changes in legislation, and following the development and application of new technologies, the mass concentration of particles in the exhaust of diesel engines has reduced steadily over the last 20 years. Current and future legislation is reducing diesel and petrol particulate mass emissions towards the current threshold of reliable measurement.

The control of coarse particles is less straightforward (Harrison *et al.*, 2001), as such particles arise from anthropogenic disruption and attrition processes which are difficult to characterise (*e.g.* direct non-exhaust emissions, resuspension, fugitive dust from industrial processes, quarrying), as well as natural processes.

⁴ In this report, the term 'light-duty vehicle' refers to all vehicles with a gross weight of less than 3.5 tonnes, and includes both cars and light goods vehicles. All vehicles having a gross weight of more than 3.5 tonnes are termed heavy-duty vehicles (HDVs).

Table 1.2: Exhaust PM emission legislation for light-duty diesel vehicles (<3.5t GVW)

| Implementation year | Directive | PM limit value (g/km) |
|---------------------|-------------------------------|-----------------------|
| 1988 | 88/76/EEC, 88/436/EEC | 1.1 |
| 1989 | 89/458/EEC | 1.1 |
| 1992 | 91/441/EEC (Euro I) | 0.14 |
| 1993 | 93/59/EEC (Euro I supplement) | 0.14-0.25 |
| 1996 | 94/12/EEC (Euro II) | 0.08-0.10 |
| 2000 | 98/69/EEC (Euro III) | 0.05 |
| 2005 | 98/69/EEC (Euro IV) | 0.025 |

Table 1.3: Exhaust PM emission legislation for engines in heavy-duty vehicles (>3.5t GVW)

| Implementation year | Directive | PM limit value (g/kWh) |
|---------------------|----------------------|------------------------|
| 1992 | 91/542/EEC (Euro I) | 0.36-0.612 |
| 1995 | 91/542/EEC (Euro II) | 0.15 |
| 2000 | 99/96/EEC (Euro III) | 0.10-0.13 |
| 2005 | 99/96/EEC (Euro IV) | 0.02 |
| 2008 | 99/96/EEC (Euro V) | 0.02 |

There are currently no legal requirements for the control of road vehicle non-exhaust particle emissions in the EU. Certain regulations, which are designed for other purposes, could influence non-exhaust PM emissions indirectly. Such regulations include restrictions on the use of studded tyres in certain countries to reduce damage to the road surface, and road/tyre noise standards.

1.4.2 Increasing proportion of total emissions

As exhaust emission control technology improves and traffic levels increase, the proportion of total PM mass emissions originating from the uncontrolled non-exhaust sources will increase.

1.4.3 Uncertainties

Whilst wear of vehicle components is widely recognised, it has received rather little attention as a source of particulate matter in the context of air pollution. Consequently, the data relating to the emission rates, physical properties, chemical characteristics, and health impacts of non-exhaust particles arising from such sources are far from comprehensive. The UK Air Quality Expert Group (AQEG, 2004) has identified the estimation of non-exhaust primary PM emissions from road transport as a priority area of uncertainty, and has stated the need to update and refine the methodologies and estimates.

The uncertainty associated with emission factors for non-exhaust sources can be illustrated by a simple comparison. Provisional results for 2002 from the NAEI indicate that tyre wear was responsible for the emission of 4.6 kt⁵ of PM₁₀ in the UK, compared with total exhaust emissions of 29.8 kt (NETCEN, 2005). However, during the late 1990s PM₁₀ emissions from tyre wear in Germany were estimated to be between 56 kt and 98 kt per annum, a range that was comparable to the amount of PM₁₀ emitted by road vehicles (Rauterberg-Wulff, 1999). Apparent contradictions of this type suggest that there is a need for a re-assessment of tyre wear particle emissions in the UK. The estimates for particle emissions due to brake wear, road surface wear and resuspension are equally uncertain.

Specific epidemiological studies have indicated a link between elevated atmospheric particle concentrations and increases in hospital admissions and death rates (Dockery *et al.*, 1993; Dockery and Pope, 1994; Vedal, 1997; Zanobetti *et al.*, 2000). The increasing weight of evidence for adverse human health effects associated with both short-term and long-term exposure to ambient particulate

⁵ Calculated using the EMEP/CORINAIR method shown in Appendix E.

matter has been comprehensively reviewed elsewhere (COMEAP, 1998; WHO, 2000; EPAQS, 2001; WHO, 2003; USEPA, 2004). In the UK alone, emissions of airborne particles are thought to be responsible for several thousand premature deaths per annum (DETR, 1999). Health concerns have been noted which relate specifically to non-exhaust particles. For example, Miguel *et al.* (1999) found that paved road dust contains at least 20 source materials known to cause or exacerbate allergic reactions in humans, including pollen, animal dander and fungal spores. Resuspension increases allergen concentration levels above those that would prevail without the traffic movement. The study showed that up to 12% of the allergenicity of TSP was attributable to paved road dust emissions. Other specific concerns include tyre constituents such as polycyclic aromatic hydrocarbons (PAHs) and latex, and the heavy metals used in brake linings. More comprehensive information on non-exhaust emissions would help to clarify the mechanisms by which particles affect human health, and would foster the development of pollution abatement strategies.

1.5 Report structure

In this Report the literature is presented from a source perspective. The sources covered in most detail are:

- Tyre wear
- Brake wear
- Road surface wear
- Corrosion
- Resuspension of road dust

There is a general lack of consistency in the definitions, terminology and metrics used in the study and reporting of emission factors for non-exhaust particulate matter. For example, some workers have reported emission factors for the individual non-exhaust sources, whereas others have reported emission factors for various combinations of source, or simply total non-exhaust emissions. Where emission factors for resuspension are reported, it is not always clear whether they include primary emissions due to abrasion, and a number of modelling methodologies consider abrasion sources but not resuspension. In addition, some workers have reported PM according to common metrics (*e.g.* PM₁₀, PM_{2.5}), whereas others have reported less familiar ones (*e.g.* PM₃).

The topics reviewed in relation to each source include:

- The factors affecting material wear and particle generation.
- The physical characteristics and composition of source materials and airborne non-exhaust particles.
- Particle emission factors.
- Contributions to ambient concentrations.

The discussion relates mainly to PM₁₀ and smaller fractions. Data also exist for TSP (*e.g.* Baumann and Ismeier, 1997; Garben *et al.*, 1997), but as the use of this metric is declining, these have not been reported in detail.

There are two general approaches for determining emission factors for non-exhaust sources, and the contributions of different sources to atmospheric particle concentrations. These approaches are direct measurement and receptor modelling. An explanation of the distinction is provided in Chapter 2. The results of studies conducted using both types of approach are considered, with Chapters 3 to 7 of the Report dealing separately with the different sources. Chapter 8 describes a number of emission

models for non-exhaust particulate matter. The Report concentrates on calculation methods which could be incorporated into existing models without the need for a large number of additional input parameters. These models will be critically evaluated later in the project. Chapter 9 provides the summary and discussion, and Chapter 10 the conclusions, some specific recommendations for the next phase of the project, and some general recommendations to improve the overall understanding.

2 Approaches for determining emission factors for non-exhaust sources

There are two general approaches for determining PM emission factors for non-exhaust sources:

- Direct measurement from the sources.
- Receptor modelling.

Emission factors can be derived by direct measurement from specific sources, either in the laboratory or in the field, often combined with a series of assumptions. Receptor models sub-divide ambient pollutant concentration data according to its different sources, and derive emission factors using mass-balance techniques. These methods are described in more detail below.

2.1 Direct measurement (source-oriented) methods

Various techniques have been used to derive emission factors for non-exhaust sources by direct measurement, either under real-world test conditions or in the laboratory. Early field trials were conducted by Dannis (1974), who mounted a filter-based system behind a moving car tyre. Some of the first applications of tyre and road surface abrasion tests to the determination of airborne PM were conducted at the General Motors Research Laboratories (Cadle and Williams, 1978; Williams and Cadle, 1978). More recent work has been conducted in relation to the PM metrics in common use today (e.g. Hildemann *et al.*, 1991; Kupiainen *et al.*, 2005). Garg *et al.* (2000) and Sanders *et al.* (2003) have employed wheel dynamometers to generate and measure airborne brake wear particles.

However, tyre, brake and road surface wear are difficult to simulate in controlled tests. Speed, acceleration/deceleration, tyre material, temperature, and parameters such as the road surface construction and curvature, are all important considerations (Rauterberg-Wulff, 1999). Sampling problems have also been noted in relation to non-exhaust particles. Hildemann *et al.* (1991) collected wear particles from the horizontal surfaces of a rolling resistance testing machine. However, when resuspended, fine tyre wear particles became electrically charged and adhered to the walls of the sampling equipment. As a result, the attempt to resuspend and collect fine tyre debris had to be abandoned.

Resuspension fluxes are also notoriously difficult to measure directly, not least because the space around a vehicle does not form a 'closed' system with obvious air inlets and outlets. A number of studies have recently been conducted using vehicle-based measurement techniques. Several measurement programmes have been undertaken using the TRAKER vehicle, with sensors located in wheel arches (Sanders *et al.*, 1997; Kuhns *et al.*, 2001; Etyemezian *et al.*, 2003a; Kuhns *et al.*, 2003; Etyemezian *et al.*, 2003b), and in the wake of a moving vehicle (Fitz and Bufalino, 2002). The particles sampled in this manner are derived from a combination of tyre wear, brake wear, road surface wear and resuspension processes, with the relative contribution of each source being dependent upon the materials being used, the operational conditions of the vehicle, and the silt loading of the road. The main methods for measuring and characterising the latter are sweeping and vacuuming. Some workers in the US have used a dry method for sample collection. For example, Hildemann *et al.* (1991) and Rogge *et al.* (1993) reported experiments in which a small vacuum sweeper was driven up and down several blocks of Pasadena-area streets to collect the road dust that had accumulated during a two-week dry period. Researchers at the University of Aberdeen have developed a similar wet system (Orr and Deletic, 2000).

2.2 Receptor-oriented methods

Receptor models allocate airborne particulate matter to a range of source categories, and are based on the simple concept of mass conservation (Hopke 1985; 1991). If a number of sources (p) exist, and if there is no interaction between their aerosols to causes mass removal or formation, then the total airborne particulate mass measured at receptor (C), will be linear sum of the contributions of the individual sources (S_j):

$$C = \sum_{j=1}^p S_j \quad (\text{Equation 2.1})$$

There are several reviews on receptor modelling which discuss in detail the theoretical basis, methodology, progress, and the uncertainties associated with the procedures (Gordan 1980a, 1980b and 1988; Hopke 1985, 1986 and 1997; Henry *et al.* 1984; Thurston and Lioy 1987; Currie 1992). At present, are around twenty different procedures are available for identifying and quantifying the sources contributing to the particulate mass, or to its chemical constituents. Among these methods there are two generally recognised classes of source apportionment:

- (i) Chemical mass balance (CMB) techniques
- (ii) Multivariate statistical techniques

The former assigns atmospheric particles to different sources by assuming that each source emits a characteristic set of chemical species in a given ratio. CMB therefore requires knowledge of source composition, and assumes that the sources specified are responsible for the concentrations of the species measured at the receptor. Multivariate techniques, such as multi-linear regression, cluster analysis, discriminant analysis, and principal component analysis, can be used to identify common patterns in atmospheric data (QUARG, 1996).

Rogge *et al.* (1993) have used chemical tracers coupled with CMB methods to apportion sources of organic fine particles. The use of CMB techniques has also been reported by Schauer *et al.* (1996), who successfully identified a range of particle emission sources including road and tyre dust, based upon extensive source profile measurements. Zheng *et al.* (2002) have reported the use of organic tracers and CMB methods to identify particle sources in the southern United States.

The source apportionment and quantification of airborne PM measured in the vicinity of roads is a rather complex task. A number of measurement approaches have been used in the field, including upwind-downwind comparisons, roadside-background comparisons, and tunnel measurements. A number of workers have determined particulate matter emission factors for road vehicles by measuring ambient concentrations at both sides of roads. When the wind direction is perpendicular to the axis of the street, the traffic contribution can be determined from the difference between the downwind and upwind concentrations. Lohmeyer *et al.* (2004) derived emission factors for non-exhaust particulate matter by measuring ambient particulate matter concentrations at both sides of an extra-urban arterial road in Karlsruhe, Germany. Similarly, Gehrig *et al.* (2004) measured PM₁₀, PM₁ and NO_x concentrations at both sides of a number of busy roads in Switzerland. Dilution factors were calculated from the difference in NO_x concentrations, the number of vehicles, and published emission factors NO_x. The emission factors for particulate matter were then calculated from the measured concentration differences, assuming that these undergo the same dilution as NO_x. PM₁ was interpreted as direct exhaust emissions, and PM₁₀ the sum of exhaust emissions and coarse particles. The PM₁₀-PM₁ difference was taken to represent non-exhaust emissions (abrasion and resuspension).

One problem associated with this technique is that the road traffic contribution can be small compared with the background concentration.

Abu-Allaban *et al.* (2003) determined particulate matter emission factors for exhaust, brake wear, tyre wear and re-entrained road dust by measuring ambient concentrations at eight roadside locations in both dry and humid regions of the United States. PM_{2.5} and PM₁₀ concentrations were collected downwind of the roads. Two techniques were used to apportion the airborne particulate matter: scanning electron microscopy to characterise the source material, and receptor modelling by CMB and multi-lag regression.

Measurements in tunnel have also been used to derive emission factors for particulate and gaseous pollutants in a number of studies (*e.g.* Gillies *et al.*, 2001; Staehelin *et al.*, 1997; John *et al.*, 1999; Weingartner *et al.*, 1996; Pierson *et al.*, 1996a, 1996b). Due to the limited dispersion and dilution conditions in the tunnel environment, pollutant concentrations tend to be higher than in normal ambient air. In addition, the influences from meteorology are reduced. Rauterberg-Wulff (1998; 1999) determined PM₁₀ emission factors for tyre and brake wear using receptor modelling in combination with measurements conducted in the Berlin-Tegel tunnel. Dichotomous samplers were used to collect PM₁₀ and PM_{2.5}, which were then analysed for volatile organic compounds (VOC), low volatility organic compounds (LVOC), and elemental carbon (EC). The composition of tyre wear material was determined by analysing samples of used tyre tread. A chemical mass balance approach was employed to calculate the various traffic-related source contributions using a simple box model, and emission factors were obtained using multiple regression analyses. Luhana *et al.* (2004) used PM samples collected at two locations in the Hatfield Tunnel on the A1(M) motorway to determine emission factors for non-exhaust sources. Particles were collected using high-volume samplers with a PM₁₀ size-selective inlet. Induction loops permanently installed in the road surface of the Hatfield Tunnel were used for the purpose of characterising traffic. Ambient airborne particles, as well as source materials, were characterised chemically and physically using a suite of analytical techniques. Emission factors for light-duty and heavy-duty vehicles were determined using principal component analysis and multiple regression analysis. Six main sources were identified as contributing to the PM₁₀ tunnel increment concentration: diesel exhaust, petrol exhaust, resuspended dust, combined brake/tyre wear, road surface wear, and an unidentified component.

3 Tyre wear

Tyre wear, and the associated particulate matter, are discussed in this Chapter of the Report. With the exception of resuspension, tyre wear is probably the largest non-exhaust source of TSP and PM₁₀ emissions from road transport (Lükewille *et al.*, 2001).

Tyre wear itself is a complex physio-chemical process which is driven by the frictional energy developed at the interface between the tread and the road pavement (Veith, 1995). The wear factor (defined here as the total amount of material lost per kilometre), and hence a tyre's lifetime, varies enormously depending on its type and how it is used. The factors affecting tyre wear include:

Tyre characteristics

- Size (radius/width/depth)
- Tread depth
- Construction
- Tyre pressure and temperature
- Contact patch area
- Chemical composition
- Accumulated mileage
- Set-up (*e.g.* tracking, toe-in and camber)

Road surface characteristics

- Material: bitumen/concrete
- Texture pattern
- Texture wavelength - micro/macro/mega
- Porosity
- Condition, including rutting and camber
- Road surface wetness
- Silt loading of road surface
- Surface dressing

Vehicle characteristics

- Vehicle weight and distribution of load
- Location of driving wheels
- Engine power
- Power/unassisted steering
- Electronic braking systems
- Suspension type and condition

Vehicle operation

- Speed
- Linear acceleration
- Radial acceleration
- Frequency and extent of braking and cornering

3.1 Tyre wear factors

In 2000, more than 54 million passenger car tyres were used in the UK, equating to a total weight of almost 430,000 tonnes (Used Tyre Working Group, 2001). A new tyre on an average European car weighs around 8 kg, and loses roughly 1-1.5 kg in weight during its service lifetime, which is typically around 3 years or 50,000 to 60,000 km. Thus, between around 10% and 20% of the rubber which goes into a tyre will disappear before the tyre is ready to scrap (Environment Agency, 1998). Similarly, Ahlbom and Duus (1994) arrived at an average rubber loss figure for Swedish roads of 17%. Based on the upper estimate for rubber loss of 20%, around 85,000 tonnes of tyre material were lost to the UK environment from passenger cars alone during 2000, mainly as a result of in-service wear.

A combination of the tyre usage and material loss statistics given above reveals that an 'average' tyre wear factor would be of the order of 100 mg per vehicle-kilometre (vkm) for a passenger car (assuming four wheels per vehicle). Figure 2.1 shows that a wide range of wear factors have been reported for light-duty vehicle tyres. It incorporates information provided by Councell *et al.* (2004), as well as other values from the literature. These values have either been derived experimentally, or have been estimated from average statistics as above. Figure 2.1 indicates that for 'normal' driving conditions a wear factor close to 100 mg/vkm would probably be appropriate.

Much of the variability in these wear factors can probably be explained by differences in the factors listed. For example, in the studies conducted during the early 1970s cross-ply tyres would have been used. Almost all modern cars are fitted with radial-ply tyres, which have greater rigidity for cornering, have better grip in the wet, and are much less susceptible to wear than the older cross-ply type. Driving behaviour and driving conditions are well-recognised determinants of tyre wear. An aggressive driving style will tend to result in more rapid and uneven tyre wear than a more restrained driving style. The reported driving conditions in the studies cited in Figure 2.1 ranged from ‘gentle’ to ‘severe’⁶. Most tyre rubber is lost during acceleration, braking, and cornering, and the amount of rubber lost will therefore tend to be greatest near busy junctions and on bends. Using a tyre-testing machine, Stalnaker *et al.* (1996) simulated the effects of ‘city’ and ‘motorway’ driving conditions on the wear of tyres. The city conditions included large numbers of turns. It was found that the city driving accounted for 63% of the tyre wear, even though it represented only 5% of the distance driven. Luhana *et al.* (2004) weighed car tyres at two-month intervals, and asked drivers to note the details of each trip undertaken. There was found to be a weak negative correlation between tyre wear and average trip speed, with wear being around 50% higher at an average speed of 40 km/h (dominated by urban driving) than at average speed of 90 km/h (dominated by motorway driving).

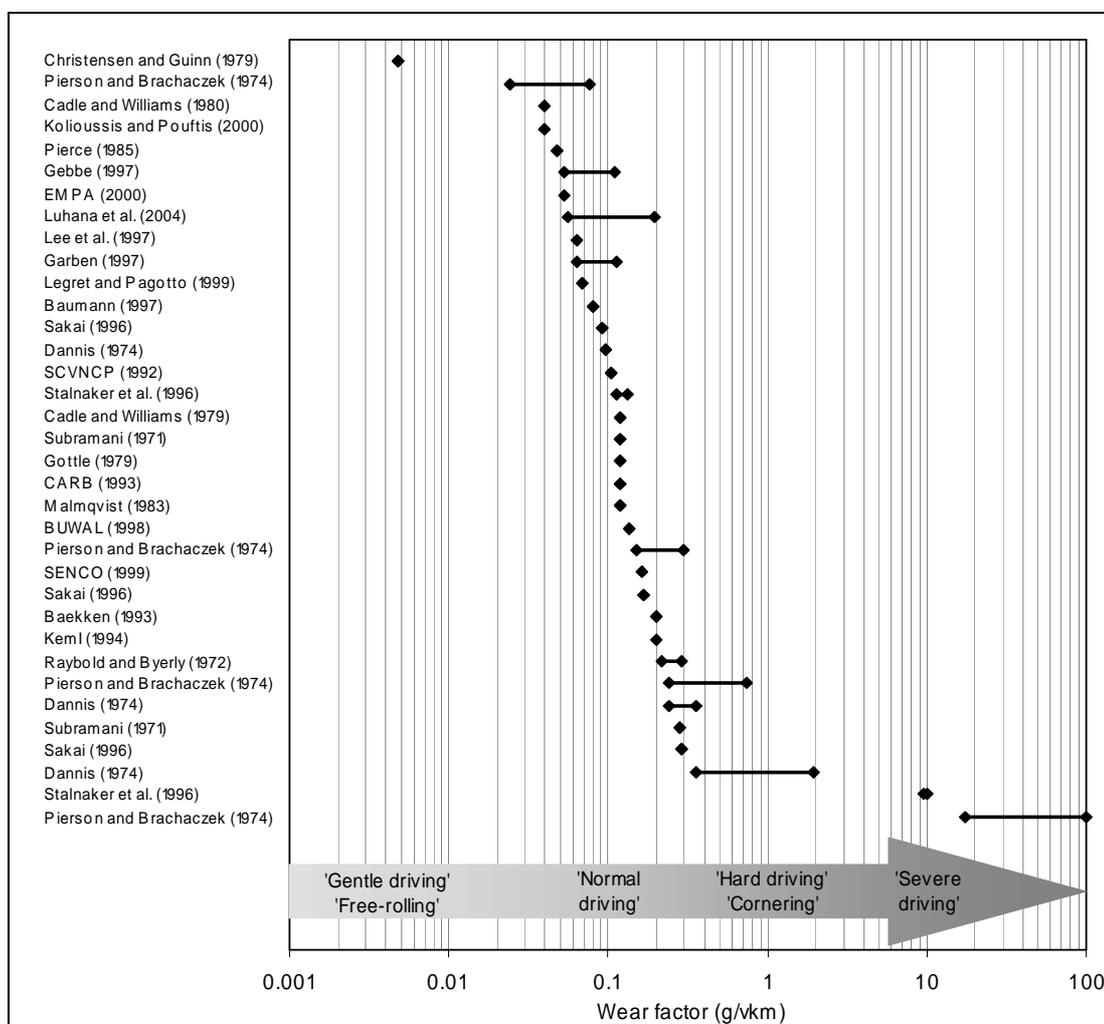


Figure 2.1: Wear factors for light-duty vehicle tyres (adapted from Councill *et al.*, 2004)

⁶ These subjective descriptions have been superimposed on Figure 2.1 by the author, although there is a considerable amount of variation in terms of how driving conditions have been defined in the literature.

Tyre wear factors ought to be substantially higher for HDVs than for LDVs. Legret and Pagotto (1999) assumed that the wear factor for heavy-duty vehicle tyres (at 136 mg/vkm) was double that for light-duty vehicle tyres. However, this appears to be an underestimate. Baumann and Ismeier (1997) give wear factors for 'heavy-duty vehicles', 'articulated lorries' and buses of 189 mg/vkm, 234 mg/vkm, and 192 mg/vkm respectively. Gebbe *et al.* (1997) reports a tyre wear factor for heavy-duty vehicles of 539 mg/vkm. HDV wear factors closer to 800 mg/km have been reported by Garben *et al.* (1997) and EMPA (2000), and SENCO (1999) give a wear factor for HGVs of 1403 mg/vkm. The wear factor per vkm will be dependent on the vehicle configuration, such as the number of axles and the load, and so a wide range of values is to be expected.

3.2 PM size range

Not all worn tyre material will be emitted as airborne PM. Some of the tyre weight loss will be in the form of carbon dioxide and water vapour, although when Cadle and Williams (1978) measured the gases emitted during continuous tyre wear tests, no significant increases in CO₂ could be measured above the background levels. A fraction of the tyre wear material may adhere to vehicle surfaces, and some much coarser solid material, such as tyre shreds, will also be deposited on the road. Camatini *et al.* (2001) collected debris from the road of a tyre proving ground, and observed particles up to a few hundred micrometers in size. Similar observations were also made by Smolders and Degryse (2002), who found that roadside tyre debris (graded using a 100µm sieve) had a mean diameter of 65µm for cars and 80µm for trucks. These findings suggest that most of the material lost during the tyre wear process will not remain airborne for very long.

It is often assumed that any airborne tyre wear particles will be found exclusively in the coarse particle mode. In fact, several studies conducted in the 1970s revealed that airborne wear particles tend to be divided into two distinct size groups: ultrafine particles with an aerodynamic diameter of less than 1 µm, and coarse particles larger than around 7 µm (Cardina, 1974; Dannis, 1974; Pierson and Brachaczek, 1974; Cadle and Williams, 1978). These observations have been confirmed by the more recent work of Fauser (1999). A plausible mechanism for the distinction is the volatilisation (the thermal degradation of tyre polymer and the volatilisation of extender oils) and subsequent condensation of material in the ultrafine particle mode, and normal mechanical wear for larger sizes (Cadle and Williams, 1978).

The relative mass contributions of the fine and coarse modes appear to be rather variable, and dependent on the sampling conditions and the metric used. Pierson and Brachaczek (1974) showed that only 10% by mass of all tyre wear particles were smaller than 3 µm. Cadle and Williams (1978) also observed that the larger particles dominate the total mass, but only during dynamic sampling (*i.e.* with a continuous flow of air through the test chamber). Under low air-flow conditions many of the large particles settled before reaching the sampling point, with the result that sub-micron particles dominated the mass distribution. More recent receptor modelling work by Rauterberg-Wulff (1998; 1999), which indicated that tyre wear PM₁₀ were only present in the coarse fraction (2.5-10 µm), and the work of Moosmüller *et al.* (1998), which indicates that the products of tyre wear are dominated by particles larger than 10µm, appear to confirm the earlier findings. However, Fauser (1999) found that around 90% by mass of tyre wear particles smaller than 20 µm had an aerodynamic diameter of less than 1µm. Other PM₁₀ data from the USEPA (1995) and Berdowski *et al.* (1997) indicate that around 70% by mass of tyre wear PM₁₀ can be classified as PM_{2.5}, 10% as PM₁, and 8% as PM_{0.1}. Dannis (1974) found that mean particle diameter decreases with increasing speed, and this may be one of a number of factors contributing to the differences in the reported findings.

3.3 PM emission factors

Cadle and Williams (1978) found that airborne particles were emitted from car tyres during indoor tests at a typical rate of 1 mg/km (equivalent to 4 mg/vkm). The USEPA (1995) gives a PM₁₀ emission factor for light-duty vehicles of 5 mg/vkm. Lükewille *et al.* (2001) cite research by EMPA (2000) which indicates a PM₁₀ emission factor of 13 mg/vkm for LDVs, and an emission factor for all types of heavy-duty vehicle of 200 mg/vkm.

Abu-Allaban *et al.* (2003) observed no contribution from tyre wear at eight roadside locations in the United States, based on receptor modelling work. However, the authors state that this may have been a result of the electrostatic charge carried by tyre wear particles (as noted by Hildemann *et al.*, 1991), which prevented them from accumulating on filters during sampling. No such problem appears to have been encountered by Rauterberg-Wulff (1998; 1999), who determined PM₁₀ emission factors for tyre wear using receptor modelling in combination with measurements conducted in the Berlin-Tegel tunnel. The author observed PM₁₀ emission factors for LDVs and HDVs of 6.1 mg/vkm and 31 mg/vkm respectively.

By comparing the PM₁₀ emission values given above for light-duty vehicles with the material loss values given in Figure 2.1, it appears likely that less than 10% of car tyre wear material is emitted as PM₁₀ under 'typical' driving conditions, but the proportion could be as high as 30%. Based on the upper estimate of tyre wear PM₁₀ in Germany of 98 kt per annum provided by Rauterberg-Wulff (1999) using the tunnel-derived emission factors, and the tyre arisings of 650 kt in the same country during 2000 (Used Tyre Working Group, 2001), one arrives at a PM₁₀ proportion of approximately 15%. CSTE (2003) estimate a proportion of 5-6%. Using styrene butadiene rubber (SBR) as a tracer, Pierson and Brachaczek (1974) found that 2% to 7% of tread wear material could be classified as airborne PM, but only 0.02 to 0.10% at high-wear locations (*i.e.* near to corners).

3.4 Contribution to ambient PM concentrations

Research has indicated that the tyre rubber content of airborne PM is generally quite low. Cardina (1973, 1974) found that between 2% and 10% of the TSP near a highway was composed of tyre tread rubber, and that dustfall near a highway junction had a tread rubber content of 2%. In Michigan, Pierson and Brachaczek (1974) measured tyre wear contributions to TSP near a road and in a tunnel of 1.2 µg/m³ and 9.6 µg/m³ respectively. Cadle and Williams (1978) observed tyre wear contributions to roadside TSP of less than 1%. In these early studies SBR was used as a tracer for airborne tyre wear particles. Where benzothiazole has been used as a tracer, researchers have also observed relatively low tyre wear contributions. Tyre wear contributions to TSP, PM₁₀, and PM_{3.3} levels at roadside of up to 7 µg/m³ (0.5-3%), 2.2 µg/m³, and 0.2 µg/m³ were noted by Lee and Kim (1989) and Kim *et al.* (1990). Using tracer analysis of benzothiazole derivatives at a number of locations in Switzerland, Huglin (2002) found that tyre wear was only a significant contributor to airborne PM at kerbside monitoring sites. At a kerbside site in Bern, Gehrig *et al.* (2001) reported that tyre wear accounted for 7.5% of PM₁₀, and around 2% of PM₁₀ at an urban background site. The work of Pollard (1997) in Leeds indicated a tyre rubber concentration in roadside TSP of 0.28 µg/m³. In Los Angeles, Miguel *et al.* (1999) reported a PM₁₀ tyre wear contribution of 1µg/m³. Fauser (1999) observed that tyre wear particles constituted around 5% of the TSP (< 20 µm) mass (1 to 10 µg/m³) in roadside air in Copenhagen, and 2.4% (1.4 µg/m³) at a rural site.

Larger tyre wear contributions were recorded by Fukazaki *et al.* (1986), who measured a maximum roadside tyre wear concentration of 24 µg/m³ for PM smaller than 4.7 µm, and Lohmeyer *et al.* (2004), who found that roadside PM₁₀ was comprised of 50% exhaust emissions, 20% tyre wear

particles, 30% road abrasion and resuspension, and less than 1% brake wear, based on the chemical analysis of filters.

Some diverse but relevant information is also available from a number of other studies. Rogge *et al.* (1993) found that no more than 1.6% of road dust comes from tyres. Hildemann *et al.* (1991) reported that tyre wear accounted for 1.4% of the total organic carbon in the air of a heavily trafficked area. Kumata *et al.* (1996) observed a very low tyre wear contribution ($0.001 \mu\text{g}/\text{m}^3$) to TSP concentrations in a forested area approximately 500 metres from a suburban roadway in Tokyo.

3.5 Tyre and PM composition

A wide range of chemical compounds can be found in the tyres of road vehicles. Tyre formulations vary according to required performance standards, such as wet grip, wear rate and drive-by noise, and passenger car tyres differ in composition from those used on heavy-duty vehicles. Different types of tyre formulation result in considerable variation in wear rates. For commercial reasons it is unusual for tyre manufacturers to release exact details of tyre composition. Some general information has been made available, and this has been augmented by a number of scientific studies (*e.g.* Hildemann *et al.*, 1991; Rogge *et al.*, 1993; Kumata *et al.*, 1996 and 2000). The bulk (40 to 60%) of tyre tread is composed of a variety of rubbers, including natural rubber co-polymers, butadiene rubber, SBR, nitride rubber, neoprene rubber, isoprene rubber, and polysulphide rubber. The rubber blend is usually governed by the required tyre characteristics, and for passenger cars Fauser (1999) quoted the following general composition by weight: natural rubber (40%), SBR (30%), butadiene rubber (20%), and butyl and halogenated butyl rubber (10%). A variety of modifiers are added to the rubber mix. For example, vulcanising agents (sulphur, thiazoles, sulphenamides, selenium, tellurium, organic peroxides, nitro compounds and azo compounds) are introduced to improve the durability of tyre rubber. Vulcanising agents typically make up 1% of the tyre rubber mass. Accelerators are also introduced to speed up vulcanisation, with the main additives being sulphur compounds and oxides of calcium, lead, magnesium or zinc. Other chemicals added to the rubber mixture include retardants (terpene-resin acid blends), pigments (carbon black; 20 to 30% of the stock formula), fillers, reinforcing agents, softeners (synthetic organic oils, tars), preservatives (halogenated cyanoalkanes), anti-oxidants (amines, phenols; 1% to 2.5%), anti-ozonants (diamines and waxes), and desiccants (calcium oxide).

Table 3.1 shows metal concentrations in tyre tread and tyre wear debris from several studies, and highlights the presence of zinc in significant quantities in tyres and tyre debris. The typical zinc oxide content of tyres in Europe is around 1-2% (Ahlbom and Duus, 1994; Smolders and Degryse, 2002). Smolders and Degryse (2002) observed zinc concentrations in tyre debris (<100 μm) of 11.4 mg/g for car tyres and 24.1mg/g for truck tyres. Councell *et al.* (2004) report a range for the measured Zn content of tyre tread of 0.04% to 1.55% by weight, based on a review of thirteen separate studies. According to Ahlbom and Duus (1994), the particulate zinc contribution from tyre wear amounted to 10% of the TSP zinc load in Swedish cities. Although there are a number of different sources of zinc in the environment, it has been used as a tyre wear particle tracer. Fauser (1999) identified extractable organic zinc (present in zinc accelerator complexes) as a tyre wear particle tracer since, with the exception of engine lubricants, tyres are the only significant contributor to organic zinc in airborne particles. Extractable organic zinc was found to be present in tyre rubber at a concentration of around 1.2 g/kg.

As far as other metals are concerned, the concentrations are generally several orders of magnitude lower than that of zinc, although significant amounts of aluminium, barium, calcium, copper, iron, potassium, sodium and titanium have been measured. For most metals the concentrations ranges are rather wide, probably reflecting the use of different tyre recipes by different manufacturers. For

example, Brewer (1997) measured the metal concentrations in five tyres from different manufacturers, and found large differences between the concentrations of calcium, cobalt, iron, lead and strontium. Kennedy & Gadd (2000) examined the elemental composition of 12 tyres available in New Zealand. The data suggested that car tyres contained more calcium than LGV and HGV tyres, whereas the HGV tyres contained considerably more zinc.

Table 3.1: Metal concentrations in tyre tread (mg/kg, unless stated).
'N/D' = not detected; '-' = not reported.

| Metal | Car tyres | | | | | LGV ⁷ tyres | | HGV tyres | | | |
|-------|----------------------------|------------------------|--------------------------------|----------------------------|-------------|-------------------------|------------------------------------|----------------------------|--|-----------------------|-----------------------|
| | Christensen & Guinn (1979) | Hewitt & Rashed (1990) | Hildemann <i>et al.</i> (1991) | Brewer (1997) ⁸ | VROM (1997) | Legret & Pagotto (1999) | Kennedy & Gadd (2000) ⁹ | Davis <i>et al.</i> (2001) | Council <i>et al.</i> (2004) ¹⁰ | Kennedy & Gadd (2000) | Kennedy & Gadd (2000) |
| Ag | - | - | - | 0.08 | - | - | N/D | - | - | - | - |
| Al | - | - | 470 | 80.9-181 | - | - | 7-129 | - | - | 9-13 | 51-139 |
| Ba | - | - | 370 | 1.89-4.09 | 0.9 | - | 10.4-166 | - | - | 8.78-39.8 | 13.2-13.6 |
| Be | - | - | - | - | - | - | 0.05-0.7 | - | - | 0.1-0.3 | 0.3-0.4 |
| Ca | - | - | 2,000 | 113-562 | - | - | 100-1680 | - | - | 60 | 150-230 |
| Cd | - | 0.28-4.96 | - | 0.56-1.07 | 1.5 | 2.6 | <0.05-0.34 | 1 | - | <0.05-0.10 | 0.28-0.56 |
| Co | - | - | - | 0.88-12.74 | 1.5 | - | 0.5-4.1 | - | - | 0.4-2.4 | 0.9-1.0 |
| Cr | - | - | 30 | 2.46-6.73 | 3 | 0.4 | <1-2 | - | - | <1 | 1-2 |
| Cu | - | 5.5-29.3 | 490 | 0.04-0.4 | - | 1.8 | <1-2 | 5 | - | 1-3 | 1-2.5 |
| Fe | - | - | 4,600 | 2.12-113.66 | 553 | - | 40-220 | - | - | 100-110 | 195-210 |
| Hg | - | - | - | - | - | - | <0.01-0.03 | - | - | <0.01 | <0.01 |
| K | - | - | 380 | - | 180 | - | 110-410 | - | - | <50-120 | 200-430 |
| La | - | - | - | - | - | - | <0.1-0.4 | - | - | <0.1 | <0.1-0.2 |
| Li | - | - | - | 0.23-2.3 | - | - | <0.2-2.6 | - | - | <0.2 | <0.2 |
| Mg | - | - | - | 39.17-105.5 | 32 | - | <4-37 | - | - | 5-37 | 10-15 |
| Mn | - | - | 100 | - | 2 | - | 0.8-2.5 | - | - | 0.7-1.0 | 3.2-6.1 |
| Mo | - | - | - | - | 2.8 | - | <0.2-1.4 | - | - | 0.3-0.6 | 0.65-1.1 |
| Na | - | - | - | - | 610 | - | 120-530 | - | - | <20-80 | 530-590 |
| Ni | - | - | 50 | - | - | 0.9 | <1-3 | - | - | <1 | <1 |
| Pb | - | - | 160 | 0.99-38.94 | - | 6.3 | 1-5.7 | 17 | - | 0.8-1.5 | 3.2-9.7 |
| Rb | - | - | N/D | - | - | - | <0.2-0.6 | - | - | <0.2 | 0.9-1.5 |
| Sb | - | - | - | - | 2 | - | <0.2-0.9 | - | - | <0.2 | <0.2-1.3 |
| Se | - | - | 20 | - | - | - | N/D | - | - | - | - |
| Sn | - | - | - | - | - | - | <0.5-1 | - | - | <0.5-0.6 | <0.5 |
| Sr | - | - | 40 | 0.16-3.13 | - | - | <0.5-2.6 | - | - | <0.5 | 0.7-1.6 |
| Ti | - | - | 560 | - | 195 | - | - | - | - | - | - |
| V | - | - | N/D | - | 1 | - | - | - | - | - | - |
| Zn | 7,300 | - | 430 | 8,378-13,494 | 10,250 | 10,645 | 5,650-9,640 | 3,400 | 400-15,500 | 1,190-3,640 | 13,800-18,300 |

As already noted, tyre rubber is predominantly composed of organic compounds. Hildemann *et al.* (1991) found that about half of the total mass of a sample of tyre wear particles was composed of organic compounds, with a further sixth being elemental carbon. The same tyre particle samples were analysed by Rogge *et al.* (1993) using GC/MS techniques. The concentrations of more than 100 specific solvent-extractable organic compounds were determined, and these are shown in Tables 3.2 to 3.4 along with the results from other studies. It was noted by Rogge *et al.* (1993) that tyre wear particles contain appreciable amounts of styrene and butadiene polymers, but these cannot be analysed using solvent extraction methods.

⁷ LGV = light goods vehicle.

⁸ May represent underestimates due to incomplete sample digestion.

⁹ Not detected: Au, Ge, In, Pd, Se.

¹⁰ Review of 13 studies.

Table 3.2 lists concentrations of different PAHs. Tyre wear particles were found by Rogge *et al.* (1993) to contain around 200 mg/kg of PAHs, although PAHs in road dust samples were not determined by the PAH content of tyre wear particles. CSTE (2003) estimated the total PAH concentration in tyre tread to be between 17 and 357 mg/kg, and the range in wear particles to be between 13.5 and 112 mg/kg. Rauterberg-Wulff (1998) reported a total PAH concentration in tyre tread of 30-360 mg/kg. VROM (1997) reported concentrations for 10 PAHs in tyre rubber, with concentrations ranging from 0.8 to 34 mg/kg. However, DeMarini *et al.* (1994) obtained much higher PAH concentrations, and this alone highlights the need for improved understanding in this area.

There appears to be no record of specific PAHs being used as tracers for tyre wear. Benzothiazole compounds, on the other hand, have been used to trace tyre wear debris in the urban atmosphere (Kim *et al.*, 1990). These compounds, which are degradation products of the accelerators used in the vulcanisation process, were also used as tracers for tyre material by Kumata *et al.* (1996 and 2000). Concentrations of benzothiazole and a number of derivatives are shown in Table 3.3. These compounds are not easy to analyse, but they are very specific tracers for tyre wear, and the contribution of tyre wear to airborne PM can be estimated directly if the mean content of the tracer in tyres is known (Huglin, 2000). Because benzothiazole is of a semi-volatile nature, according to Rogge *et al.* (1993) it must be trapped as a thiazole-type compound within a tyre wear particle to actually serve as a particle-phase tracer. In a Swiss study, two tracer compounds for tyre material, 2-(4-morpholinyl) benzothiazole and N-cyclohexyl-2-benzothiazolamine (NCBA), were identified from PM₁₀ filters (Schmid, 2000).

Table 3.2: Concentrations of PAHs in tyre tread and wear particles (mg/kg).
'N/D' = not detected; '-' = not reported.

| Compound | Rogge <i>et al.</i> (1993) ^{11,12} | DeMarini <i>et al.</i> (1994) | Reddy & Quinn (1997) | VROM (1997) | Gadd & Kennedy (2000) | Gadd & Kennedy (2000) |
|--|--|-------------------------------------|----------------------------|----------------|-----------------------------|-----------------------------|
| | Car | Car & HGV | Car | Car | Car | HGV |
| Phenanthrene | 11.8 | 238-253 | - | 18 | 6.2-6.3 | <10 |
| Anthracene | N/D | 50-56 | - | 1.6 | - | - |
| Methyl (phenanthrenes, anthracenes) | 23.6 | - | - | - | - | - |
| Dimethyl (phenanthrenes, anthracenes) | 38.5 | - | - | - | - | - |
| Fluorene | - | 187-261 | - | - | - | - |
| Fluoranthene | 11.1 | 339-458 | - | 30 | 9.4-28.5 | <10 |
| Pyrene | 54.1 | 34-452 | 41.4 | - | 10.8-69.7 | 6.3 |
| Indeno[1,2,3- <i>cd</i>]pyrene | - | 52-86 | - | N/D | - | - |
| Napthalene | - | 486-816 | - | - | - | - |
| Acenaphthene | - | 290-2,446 | - | - | - | - |
| Acenaphthylene | - | 562-861 | - | - | 8.2-19.8 | <10 |
| Methyl (fluoranthenes, pyrenes) | 24.4 | - | - | - | - | - |
| Benzo[<i>a</i>]fluorene/benzol[<i>b</i>]fluorene | 1.3 | - | - | - | - | - |
| Benzo[<i>ghi</i>]fluoranthene | 6.3 | - | - | - | - | - |
| Benz[<i>a</i>]anthracene | N/D | 82-102 | - | 3.8 | - | - |
| Chrysene/triphenylene | 8.2 | 71-92 | - | 13.5 | - | - |
| Methyl (benz[<i>a</i>]anthracenes, chrysenes, triphenylenes) | 18.7 | - | - | - | - | - |
| Dimethyl (fluoranthenes, pyrenes) | 19.0 | - | - | - | - | - |
| Benzo[<i>k</i>]fluoranthene | N/D | 74-99 | - | 0.8 | - | - |
| Benzo[<i>b</i>]fluoranthene | N/D | 70-88 | - | - | - | - |
| Benzo[<i>e</i>]pyrene | 5.2 | - | - | - | - | - |
| Benzo[<i>a</i>]pyrene | 3.9 | 85-114 | - | 6.4 | - | - |
| Benzo[<i>ghi</i>]perylene | N/D | 66-159 | - | 34 | <10 | <10 |

¹¹ Tyre tread particles.

¹² Not detected: benzacenaphthylene, 2-phenylnaphthalene, 2-benzyl-naphthalene, indeno[1,2,3-*cd*]fluoranthene, Perylene.

Table 3.3: Concentrations of benzothiazoles in tyre tread and wear particles (mg/kg).
 ‘-’ = not reported.

| Compound | Rogge <i>et al.</i> (1993) ¹³ | Reddy & Quinn (1997) | Gadd & Kennedy (2000) | Gadd & Kennedy (2000) |
|-------------------------------|---|----------------------------|-----------------------------|--------------------------|
| | <i>Car</i> | <i>Car</i> | <i>Car</i> | <i>HGV</i> |
| Benzothiazole | 124.3 | 171 | 23.5-70.4 | 44.7 |
| Methylthiobenzothiazole | - | - | 6.3-6.7 | - |
| Mercaptobenzothiazole | - | - | 242-437 | - |
| 2-hydroxybenzothiazole | - | 80.9 | - | - |
| 2-(4-morpholino)benzothiazole | - | 3.76 | - | - |

The concentrations of various other organic compounds, mainly derived from Rogge *et al.* (1993), are given in Table 3.4. It should be noted that many compounds were below instrument detection limits, and these have not been listed. For these, the original paper should be consulted. The *n*-alkanes, *n*-alkanoic acids and natural resins were found in substantial amounts. It was noted by Rogge *et al.* (1993) that for the *n*-alkanes $\geq C_{35}$ there are few urban emission sources other than tyre wear. The *n*-alkanes $\geq C_{35}$ were therefore considered to be a useful marker for tyre wear, and actually better than benzothiazole.

Other authors have identified the SBR component of synthetic tyre tread as useful molecular tracer for tyre dust (Pierson and Brachaczek, 1974; Cass, 1998; Fauser, 1999). In road dust samples, Pollard (1997) observed that when tyre wear particles were present two specific compounds could be readily identified: di-pentene (limonene, $C_{10}H_{16}$) and styrene (C_8H_8). Di-pentene was also consistently observed by Pollard in airborne particle samples, and arises from natural rubbers or isoprene-based synthetic rubbers used in tyres. Styrene, which appeared irregularly, arises from the SBR used mainly in car tyres. The fraction of tyre rubber in the road dust was found to be 0.61%.

Due to changing operating temperatures, mechanical wear, oxidant and radical attack, and the loss of protective ingredients, tyre rubber is gradually altered during use (Kaidou and Ahagon, 1990; Ahagon and Kaidou, 1990). Cadle and Williams (1978) found that the wear process resulted in the devulcanisation of around 30% of the SBR in large tyre wear particles. In addition to heat effects, some loss of rubber components is likely during contact of tyres with water on the road. This arises as some organic compounds (*e.g.* benzathiozoles) and inorganic constituents (*e.g.* zinc) are soluble in water (Kennedy *et al.*, 2002).

¹³ Tyre tread particles.

Table 3.4: Concentrations of various organic compounds in car tyre tread and wear particles (mg/kg).
'N/D' = not detected; '-' = not reported.

| Compound | Rogge <i>et al.</i> (1993) ¹⁴ | Compound | Rogge <i>et al.</i> (1993) | Gadd & Kennedy (2000) | Compound | Rogge <i>et al.</i> (1993) | Gadd & Kennedy (2000) |
|----------------------------------|--|--|----------------------------------|-----------------------------|---|----------------------------------|-----------------------------|
| n-Alkanes | | n-Alkanoic acids | | | Natural resins | | |
| Nonadecane | 40.7 | Hexanoic acid | 122 | - | Dehydroabiatic acid | 7,970 | - |
| Eicosane | 58.3 | Heptanoic acid | 3.0 | - | Abiatic acid | 69.6 | - |
| Heneicosane | 47.7 | Octanoic acid | 31.1 | - | 7-oxodehydroabiatic acid | 10.1 | - |
| Docosane | 62.5 | Nonanoic acid | 90.9 | - | 13 β -ethyl-13-methylpodocarp-8-en-15-oic acid | 291 | - |
| Tricosane | 70.7 | Decanoic acid | 37.8 | - | 13 α -isopropyl-13-methylpodocarp-8-en-15-oic acid | 256 | - |
| Tetracosane | 114 | Undecanoic acid | 187 | - | 13 β -isopropyl-13-methylpodocarp-8-15-oic acid | 916 | - |
| Pentacosane | 175 | Dodecanoic acid | 137 | - | Chlorinated organics | | |
| Hexacosane | 186 | Tridecanoic acid | 11.6 | - | <i>a</i> -chloroindane | 3.4 | - |
| Heptacosane | 227 | Tetradecanoic acid | 635 | 9.3-33.2 | 2,4,5'-trichlorobiphenyl | 1.9 | - |
| Octocosane | 269 | Pentadecanoic acid | 86.0 | - | Aromatic amines | | |
| Nonacosane | 389 | Hexadecanoic acid | 4,818 | 383-1,480 | 2-phenylmethylbenzenamine | - | 33.4 |
| Triacontane | 546 | Heptadecanoic acid | 151 | - | 2-methyl-N-(2-methylphenyl)-benzenamine | - | 8.95 |
| Hentriacontane | 743 | Octadecanoic acid | 6,009 | 166-1,040 | Diphenylbenzenediamine | - | 300 |
| Dotriacontane | 969 | Nonadecanoic acid | 16.6 | - | Methyl substituted diphenylbenzenediamine | - | 738 |
| Tritriacontane | 1,230 | Eicosanoic acid | 127 | - | Ethyl substituted diphenylbenzenediamine | - | 344 |
| Tetracontane | 1,556 | Polycyc. aromatic ketones/quinones | | | N-(1,3-dimethylbutyl)-N'-phenyl-pphenylenediamine | - | 1170-1790 |
| Pentatriacontane | 2,006 | Xanthone | 0.45 | - | N-isopropyl-N'-phenyl-p-phenylenediamine | - | 44.3 |
| Hexatriacontane | 2,254 | Regular steranes | | | Heterocyclic amines | | |
| Heptatriacontane | 2,302 | Cholestanes | 13.5 | - | Dihydrotrimethylquinoline | - | 14.6-72.5 |
| Octatriacontane | 2,181 | Cholestane | 10.7 | - | Isoindoleione | - | 4.25-27.9 |
| Nonatriacontane | 1,428 | Ergostanes | 20.7 | - | Phenols | | |
| Tetracontane | 1,158 | Sitostanes | 29.3 | - | Di(methylbenzyl) phenols | - | 627 |
| Hentetracontane | 831 | Pentacyclic triterpanes | | | Tri(methylbenzyl) phenols | - | 715 |
| n-Alkenoic acids | | 22,29,30-trisnorneohopane | 29.6 | - | Other compounds | | |
| Oleic acid | 1,116 | 17 β (H),21 β (H)-30-norhopane | 162 | - | Hydroxymethylphthalimide | 54.5 | - |
| Linoleic acid | 164 | 17 β (H),21 β (H)-hopane | 199 | - | <i>p</i> -octyphenol | 17.6 | - |
| Benzoic acids | | 22S-17 α (H),21 β (H) homohopane | 107 | - | Isothiocyanatocyclohexane | - | 31.4-31.8 |
| Benzoic acid | 74.8 | 22R-17 α (H),21 β (H) homohopane | 73.9 | - | Sequiterpenoid compounds | - | 17.45 |
| Substituted benzaldehydes | | 22S-17 α (H),21 β (H) bishomohopane | 69.8 | - | N,N'-diphenylguanidine | - | 135 |
| 2-methylbenzaldehyde | 4.1 | 22R-17 α (H),21 β (H) bishomohopane | 42.9 | - | 1,3-bis(methylethyl)benzene | - | 5.45 |
| 3-methylbenzaldehyde | 1.4 | Amides | | | 1,4-dimethyl-7-(1-methylethyl)azulene | - | 110 |
| 4-methylbenzaldehyde | 2.3 | 4-phenylbenzenamine | 12.9 | - | bis-1,1'-(3-methyl-1-propene-1,3-diyl)benzene | - | 9.60 |

¹⁴ Tyre tread particles.

4 Brake wear

There are two main brake system configurations in current use: disc brakes, in which flat brake pads are forced against a rotating metal disc (usually cast iron or reinforced aluminium), and drum brakes, in which curved brake shoes are forced against the inner surface of a rotating cylinder. Brake pads and brake shoes are composed of friction lining material bound to a metal backing plate. During forced deceleration, vehicle brakes are subject to large frictional heat generation, with the associated wear of linings and rotors. This mechanically-induced wear generates micron-size particles which are subsequently released to the environment.

Cars are usually equipped with front disc brakes and either rear disc or drum brakes, and many modern vehicles are equipped with anti-lock braking systems (ABS). Commercial vehicles tend to be fitted with drum brakes, although disc brakes are being introduced by some manufacturers. Disc brakes have a much better gradual braking efficiency than drum brakes, even though they have a smaller friction lining. The front brakes have to provide around 70% of the braking power (Garg *et al.*, 2000), and it is therefore no surprise that front brake linings have to be replaced more frequently than rear ones.

For many years brake linings were composed of asbestos fibres, but asbestos is no longer incorporated in new brake linings due to health concerns. There are now several basic types of brake lining material available:

- Non-asbestos organic (NAO)
- Low-metallic
- Semi-metallic
- Metallic

As with tyre wear, the composition of the friction material influences the brake wear factor. The first three types of brake lining tend to be used for conventional applications. NAO linings are relatively soft and create less noise, but they generally wear faster and create more dust than the other types. Low-metallic linings are made from an organic formula mixed with small amounts (10 to 30 %) of metal to help with heat transfer and provide better braking. With the added metal, there is more brake dust and they may be slightly noisier. Semi-metallic linings have a metal content of around 30 to 65%. These pads are more durable and have excellent heat transfer, but also wear down rotors faster, have intrusive noise characteristics, and may not perform as well under low-temperature conditions. For high performance requirements, or extreme braking conditions (sports cars, ambulances, police cars), metallic linings are used.

Driving behaviour, in particular the frequency and severity of braking events, is also an important determinant of brake wear. Because brake wear only occurs during forced decelerations, the highest concentrations of brake wear particles should be observed near busy junctions, traffic lights, pedestrian crossings, and corners. However, as Kennedy *et al.* (2002) point out, particles may also be released from the brake mechanism or wheel housing some time after the primary emission event.

4.1 Brake wear factors

The provisional NAEI data for 2002 indicate that brake wear was responsible for 4.5 kt of PM₁₀ emissions in the UK¹⁵, although the value reported in the UK inventory is subject to considerable

¹⁵ Calculated using the EMEP/CORINAIR method shown in Appendix E.

uncertainty. A Swedish study found that around 934 tonnes of brake linings were used in the country during 1993 (Malmo Environmental Health Administration, 1998). Westerlund (2001) calculated that, in Stockholm, cars, goods vehicles and buses released around 45 tonnes, 7.6 tonnes, and 3.3 tonnes respectively of brake lining material to the environment each year.

It has been estimated that front disc brakes last for around 35,000 miles (56,000 km) miles under normal usage, whilst rear brakes can be expected to last around 70,000 miles (112,000 km) (Garg *et al.*, 2000). During a brake's lifetime, normally around 80% of the friction material will have worn away, and on this basis total wear amounts to 11-18 mg/vkm for cars, and for a large pick-up truck it would be 29 mg/vkm (Garg *et al.*, 2000). Based on component size, density, and lifetime, Legret and Pagotto (1999) calculated brake lining wear factors of 20 mg/vkm for cars, 29 mg/vkm for light goods vehicles, and 47 mg/vkm for HGVs. In Stockholm, Westerlund (2001) estimated the amount of material lost from cars, HGVs and buses to be 17 mg/vkm, 84 mg/vkm and 110 mg/vkm respectively. For cars, Luhana *et al.* (2004) determined an average brake lining wear factor of 8.8 mg/vkm, and observed a negative linear dependence of the wear factor on average trip speed. In addition, Luhana *et al.* (2004) noted that a small number of severe braking events appeared to have a large impact on the amount of material lost. When such events were excluded from the analysis, the typical wear factor was around 10 mg/vkm at 40 km/h, and around 2 mg/vkm at 90 km/h. For HGV tractor units in New Zealand, Kennedy *et al.* (2002) calculated a rate of loss of brake lining material of around 54 mg/km.

Although gaseous emissions do occur as a result of the mechanical abrasion of brake linings, they do not appear to be significant. During the tests conducted by Garg *et al.* (2000), increases in the concentrations of CO, CO₂ and hydrocarbons above the background levels in the test chamber could not be detected.

4.2 PM size range

As with tyre wear, not all of the worn brake material will be emitted as airborne PM, although proportionally more it seems in the case of brakes, and a significant proportion as PM_{2.5}. Garg *et al.* (2000) found that, on average, around 35% of brake wear mass is released as airborne PM. However, this does not take into account sampling losses, and if these were to be included in the study by Garg *et al.* (2000) the airborne fraction would increase to around 64% (Sanders *et al.*, 2003). Sanders *et al.* (2003) conducted detailed laboratory tests using state-of-the-art equipment, and observed that, depending on the severity of the braking, between 50% and 90% of the total wear material was in the form of airborne particles. The collection efficiency for wear debris was between 90% and 100% of the wear mass.

Whilst the majority of the fine particulate brake dust from disc brakes is released to the environment, small amounts of brake dust can be retained on the vehicle. According to Lohrer and Mierheim (1983), 10% of brake dust is retained in the drum brake enclosure. This value appears to be slightly low compared with the observations made by Sanders *et al.* (2003), whose test track and wind tunnel measurements revealed that typically 50% of the brake wear debris escapes the vehicle and enters the atmosphere. It was also found that 3-30% of brake debris falls on the road, 16-22% is retained on the wheel, and 8-25% is retained on the brake and steering/suspension equipment, but the exact proportions will vary from vehicle to vehicle depending on the design and operating conditions. When low-metallic brake linings were used, 60% of the wear debris was found to originate from the disc and 40% from the linings, a result which could have implications concerning the interpretation of the results of studies which have only considered the brake linings.

Under controlled laboratory conditions, Cha *et al.* (1983) found that the diameters of airborne particles and deposited dust were generally similar, with a peak in the particle size distribution at 2.1-3.3 μm and around 10% of particles in the sub-micron size range. The percentage of airborne particles was found to increase with vehicle speed. Data from the USEPA (1995) and Berdowski *et al.* (1997) indicated that 98% (by mass) of airborne brake wear particles can be classified as PM_{10} , whilst around 40% of the PM_{10} is $\text{PM}_{2.5}$, 10% is PM_1 , and 8% is $\text{PM}_{0.1}$. Garg *et al.* (2000) recorded airborne brake wear particle mass fractions smaller than 10 μm , 2.5 μm , and 0.1 μm of 88%, 63% and 33% respectively. Sanders *et al.* (2003) give PM_{10} and PM_1 proportions of 80% and 2%. Different size distributions have been obtained elsewhere. Receptor modelling work by Abu-Allaban *et al.* (2003), using PM_{10} and $\text{PM}_{2.5}$ measurements, showed that the brake wear contribution was observed mainly in the PM_{10} fraction, and that the $\text{PM}_{2.5}$ share of PM_{10} was only 5-17%. In any case, it appears that most airborne brake wear particles can be classified as PM_{10} , and a substantial proportion as $\text{PM}_{2.5}$. The mass mean diameter of brake wear debris reported by Garg *et al.* (2000) varied between 0.7 and 2.5 μm . It is possible that the high temperatures generated during braking can vaporise some of the brake pad material, and Garg *et al.* (2000) suggested that the volatile material may condense during measurement and contribute to the fine particle fraction. For three different types of brake lining, Sanders *et al.* (2003) observed a consistent mass-weighted size mean diameter over an urban driving cycle of around 5-6 μm . Under harsh braking conditions, the mass mean diameter was closer to 10 μm , and it was considered possible that a significant proportion of the wear debris could have been larger than 20 μm in diameter.

4.3 PM emission factors

The USEPA PART5 model, as well as the more recent MOBILE6 and EMFAC7g models, uses a brake wear PM_{10} emission factor for light-duty petrol vehicles of 7.8 mg/vkm, although this can be traced back to work conducted during the early 1980s using asbestos brakes (Cha *et al.*, 1983). Since the 1980s, the asbestos in brakes has been replaced by various other materials, and the old EPA emission factor may not be applicable to modern vehicles. More recently, Garg *et al.* (2000) obtained PM_{10} emission factors for light-duty vehicles of 2.9-7.5 mg/vkm, $\text{PM}_{2.5}$ emission factors of 2.1-5.5 mg/vkm, and $\text{PM}_{0.1}$ emission factors of 1.2-3.1 mg/vkm. In fact, the upper limit of the range for PM_{10} emissions is very close to the older USEPA value. Lükewille *et al.* (2001) cite research by Carbotech (1999) which indicated PM_{10} emission factors for LDVs of 1.8-4.9 mg/vkm, and an emission factor for HDVs of 3.5 mg/vkm.

Lower PM_{10} emission factors were obtained in tunnel studies by Rauterberg-Wulff (1998, 1999): 1 mg/vkm for cars, and 24.5 mg/vkm for HGVs. From measurements in the Hatfield Tunnel, Luhana *et al.* (2004) estimated combined tyre and brake wear PM_{10} emission factors for LDVs and HDVs of 6.9 mg/vkm and 49.7 mg/vkm respectively. Given that these latter values represent total tyre and brake wear, they are also towards the lower ends of the ranges of values reported elsewhere in the literature. One reason for these findings may be that vehicles tend to be operated in a 'steady-state' mode inside tunnels. According to Abu-Allaban *et al.* (2003), for vehicles operating in tunnels brake wear emissions are minimal, but this must surely be dependent on the location of the tunnel. In tunnels located in central Gothenburg, Sternbeck *et al.* (2002) observed most vehicles to be braking at some point, with frequent stop-start driving conditions during peak traffic periods. Abu-Allaban *et al.* (2003) conducted PM_{10} measurements at a range of roadside locations. Very high brake wear PM_{10} emission factors (79 mg/vkm for LDVs and 610 mg/vkm for HDVs) were observed at freeway exit sites. Otherwise, brake wear emissions were generally close to zero. It was also noted that under real-world conditions, road dust is contaminated by brake wear particles, and this could lead to higher estimates for brake wear emissions than laboratory-based experiments. However, Malmqvist (1983) estimated that, of all the particles emitted by brake systems, 1.5 mg/vkm are deposited on the road surface, so contamination is probably not a large source of error.

According to Sanders *et al.* (2003), the old USEPA value for LDVs still adequately describes brake emissions during urban driving, but not the variations in emission rate with different brake lining formulations. Over an urban driving cycle on a dynamometer, low-metallic linings were found to result in 2-3 times more wear particles than semi-metallic and NAO linings. PM₁₀ emissions ranged from 1.5 to 1.7 mg/stop for NAO and semi-metallic linings respectively, to 7 mg/stop for low-metallic linings. Under harsh braking conditions, large numbers of sub-micron particles were generated. It was suggested that at the higher temperatures associated with these conditions (500 to 600°C), particles are likely to arise from the combustion of the linings and subsequent nucleation. During the urban driving tests the brake temperatures remained below 200°C, and this effect was not observed. However, the mass remained dominated by the larger wear particles. The mass of wear debris generated during the harsh braking tests was roughly two orders of magnitude higher than during the urban driving programme. Garg *et al.* (2000) found that the mass of friction material lost per stop increased with an increase in brake temperature, although the percent of total wear detected as airborne PM was higher for a brake temperature of 100°C than for a brake temperature of 400°C.

Garg *et al.* (2000) also determined the total numbers of particles emitted during brake tests using an Electrical Low-Pressure Impactor (ELPI) and an Electrical Aerosol Analyser (EAA). With the ELPI, the average number of particles detected per stop ranged from 1.0×10^4 to 5.23×10^6 . The number of particles emitted showed a clear increase with brake temperature. When measured using the EAA, the number of particles per stop ranged from 6.9×10^{10} to 4.7×10^{12} . The higher EAA counts suggested that most of the particles were smaller than the 30 nm minimum size cut-off of the ELPI. However, the numbers of particles observed at the higher brake temperatures may have been due to more frequent braking during the test procedure. As before, the large numbers of particles observed in some wear tests was attributed to condensation occurring as a result of the dilution conditions.

4.4 Brake lining and PM composition

Brake lining material has been reviewed extensively by Kennedy *et al.* (2002). The results of their review, and complementary information from other sources, are summarised in the remainder of this Section.

Linings generally consist of four main components:

Fibres. Fibres provide strength and structure to the friction lining. They can be classified as metallic, mineral, ceramic or organic, and include, copper, steel, brass, potassium titanate, glass, organic material (aramid), and Kevlar.

Binders. Binders hold the other lining components together to form a thermally stable matrix. A variety of modified phenol-formaldehyde resins are used as binders in friction linings, and more than one resin type may be used (Filip *et al.* 1997). The binder may be modified in a number of ways, depending upon the brake pad specifications.

Fillers. Fillers are used to reduce wear and cost, to maintain frictional properties, and to assist in keeping the surface of the friction lining clean (*e.g.* free from molten metal and plastic). They tend to be low-cost materials such as inorganic compounds (*e.g.* barium and antimony sulphate, magnesium oxides and chromium oxides), silicates (kaolinite clays), ground slag, stone, and metal powders.

Abrasives and modifiers. Modifiers are added to influence the wear characteristics of the lining. Friction modifiers can be inorganic, metallic or organic. Graphite is a major modifier used to

influence friction, but other common modifiers include ground rubber, metallic particles, carbon black and cashew nut dust.

The proportions of the different components vary in different types of lining. For organic linings, Howell and Ball (1995) give the following composition: organic fibres (aramid) 20%, inorganic fibres 0.6%, binders 45%, fillers 15%, modifiers 15%, other 4.4%. Eriksson *et al.* (2001), on the other hand, give a much lower proportion of binders (9%), and a much higher proportion of modifiers (38%). For semi-metallic linings, Howell and Ball (1995) give a range of results, with fibres constituting between 20% and 60% of the linings, and fillers between 0% and 52%.

Many different specific brake linings are available on the market, each having a different composition. Concentrations of selected metals in brake lining material and brake dust are summarised for a number of studies in Table 4.1.

Several workers have reported the metal concentrations in new car brake linings, and some of the results from different studies are comparable. However, the concentrations of a number of metals vary by several orders of magnitude, and this clearly shows that compositional information of this type is strongly dependent upon the manufacturer and lining type. The metals which tend to be present in the highest concentrations are iron, copper, lead and zinc. An iron content of new linings of up to 60% has been reported. Copper concentration can be as low as 10 mg/kg, and as high as 25% by mass. Similarly, the reported mass concentration range for lead is 1 mg/kg to 12%, and that for zinc is 15 mg/kg to 19%. Other metals, notably barium, calcium, magnesium, manganese, sodium, nickel, antimony and tin, have also been shown to be prominent. For several metals, Westerlund (2001) found similar concentration ranges in new and replacement car brake linings. Sternbeck *et al.* (2002) concluded that brake wear is a major emission pathway for at copper, antimony and barium, and a Cu:Sb ratio of 4.6 was proposed as a diagnostic criteria for brake wear particles in ambient air. However, this ratio does not appear to be strongly supported by the literature.

Westerlund (2001) also reported the concentrations of several metals in new brake linings for Volvo and Scania heavy goods vehicles and buses. The concentration ranges were generally within those observed for car linings, although large differences were observed between the concentrations of copper and zinc in the linings for the Volvo and Scania vehicles.

During use, the surfaces of brake linings are subject to a range of processes which alter their composition. Inorganic materials will not be altered significantly by temperature (apart from perhaps the oxidation states of some metals). However, the contact between the lining and the discs or brake drum has the potential to alter the composition of the emitted particles relative to the lining material (Kennedy *et al.*, 2002). The chemical composition of brake dust samples has been investigated in a small number of studies. For example, Hildemann *et al.* (1991) found that fine brake dust particles consisted mainly of iron compounds, silicon compounds, organics, magnesium, and barium. Kennedy & Gadd (2000) found that brake dust samples contained higher concentrations of iron and tin when compared with brake linings. This was attributed to wear of the brake drum and disc brake rotor, which are typically made of steel and alloys and may have oxide coatings which are cleaned off by the friction lining contact.

Table 4.1: Metal concentrations in brake linings (concentrations in mg/kg, unless stated). 'N/D' = not detected; '-' = not reported.

| Metal | <i>Brake linings: cars</i> | | | | | | | <i>Brake linings: HDVs</i> | | | | <i>Brake dust: cars</i> | | | |
|--------|----------------------------|------------------|-----------------------------|-------------------------|------------------------------------|---|--------------|--|-------|--------|-------|-------------------------|-----------------------|----------------------------------|--------------|
| | Shaheen (1975) | Armstrong (1994) | Brewer (1997) ¹⁶ | Legret & Pagotto (1999) | Kennedy & Gadd (2000) ² | Westerlund (2001) New linings Old linings ¹⁸ | | Westerlund (2001) HGVs Buses Scania | | | | Hildemann et al. (1991) | Kennedy & Gadd (2000) | Garg et al. (2000) ¹⁷ | |
| Al | - | - | 3,765 | - | - | - | - | - | - | - | - | - | 330 | - | 920-2,500 |
| As | - | - | - | - | <2-18 | - | - | - | - | - | - | - | 10 | <2-11 | - |
| Ba | - | - | 2,638 | - | - | - | - | - | - | - | - | - | 74,400 | - | 5,900-33,900 |
| Ca | - | - | 14,300 | - | - | - | - | - | - | - | - | - | 1,100 | - | 920-8,600 |
| Cd | - | <2.5 | 29.9 | 2.7 | 0.1-1.8 | <1-24.2 | <2-41.4 | <10.3 | <1.99 | <10.1 | <1.98 | - | <0.06-2.6 | - | - |
| Co | - | - | 6.43 | - | 11.6-45.8 | - | - | - | - | - | - | - | - | 12-42.4 | - |
| Cr | 2,200 | 30-160 | 325 | - | 39-411 | 5.73-305 | 31.2-274 | 171 | 157 | <20.1 | 118 | 1,200 | 135-1,320 | - | - |
| Cu | 30,600 | <62.5-205,000 | 15,100 | 142,000 | 11-39,000 | 12.7-234,000 | 43.4-143,000 | 15,000 | 76.9 | 27,300 | 88.3 | 370 | 71-1,980 | 380-39,300 | - |
| Fe (%) | - | - | 39.9 | - | 1.17-63.7 | - | - | - | - | - | - | 11.5 | 8.9-46.8 | 1.1-53.7 | - |
| Hg | - | - | - | - | <0.1-03 | - | - | - | - | - | - | - | <0.06-0.3 | - | - |
| K | - | - | 857 | - | - | - | - | - | - | - | - | 190 | - | 4,120-5,100 | - |
| Li | - | - | 55.6 | - | - | - | - | - | - | - | - | - | - | - | - |
| Mg | - | - | 6,140 | - | - | - | - | - | - | - | - | 83,000 | - | - | - |
| Mn | - | - | 3,220 | - | 181-1,920 | - | - | - | - | - | - | 1,700 | 856-2,870 | 620-5,640 | - |
| Mo | - | - | - | - | 0.4-215 | - | - | - | - | - | - | - | 5-98.9 | 740 | - |
| Na | - | - | 15,400 | - | - | - | - | - | - | - | - | 80 | - | - | - |
| Ni | 7,450 | - | 210 | - | 44-660 | 3.6-382 | 34.7-491 | 118 | 110 | 140 | 178 | 660 | 80-730 | - | - |
| Pb | 1,050 | <100-119,000 | 1,960 | 3,900 | 1.28-873 | <5.84-56,600 | <12.2-62,100 | 656 | 158 | 1,020 | 441 | 50 | 4-1,290 | - | - |
| Rb | - | - | - | - | - | - | - | - | - | - | - | 50 | - | - | - |
| Sb | - | - | - | - | 0.07-201 | - | - | - | - | - | - | - | 4-133 | 16,900 | - |
| Se | - | - | - | - | - | - | - | - | - | - | - | 20 | - | - | - |
| Sn | - | - | - | - | <1-15 | - | - | - | - | - | - | - | 4.5-115 | - | - |
| Sr | - | - | 81.4 | - | - | - | - | - | - | - | - | 740 | - | 300-990 | - |
| Ti | - | - | - | - | - | - | - | - | - | - | - | 3,600 | - | 26,400-33,000 | - |
| V | - | - | - | - | - | - | - | - | - | - | - | 660 | - | 1,180 | - |
| Y | - | - | - | - | - | - | - | - | - | - | - | - | - | - | - |
| Zn | 124 | 100-188,000 | 3,300 | 21,800 | 25-34,500 | 14.5-107,100 | 29.1-51,000 | 14,900 | 127 | 18,500 | 172 | 270 | 360-9,630 | 120-27,300 | - |
| Zr | - | - | - | - | - | - | - | - | - | - | - | - | - | 450-21,400 | - |

¹⁶ May represent underestimates due to incomplete sample digestion.¹⁷ Airborne PM. Based on calculation of the author. Only for cases where metal was detected.¹⁸ Downstream market linings.

According to Kennedy *et al.* (2002) there is little information available from industry on the organic composition of friction linings with regard to particular organic compounds. The chemical composition of organometallic brake wear particles was determined by Hildemann *et al.* (1991) and, based on the same samples, the analysis of the solvent-soluble organic component was reported in greater detail by Rogge *et al.* (1993). The concentrations of a number of specific organic compounds from this latter study are presented in Table 4.2. However, it was found that only a small fraction of the organic fraction of brake dust could be extracted. Many compounds were below instrument detection limits, and have not been listed. For these, the original paper should be consulted. Because brake linings have to withstand excessive mechanical and heat stress and must be resistant to possible leaking brake fluid, organic fibres and binders are used that show high boiling points and resistance to solvents. Hence it is not surprising that only a small fraction of the brake lining dust can be examined using conventional laboratory techniques. The identifiable portion of the brake particle organics measured by Rogge *et al.* (1993) consisted mainly of n-alkanoic acids (34.3%) and polyalkylene glycol ethers (56.9%), the latter being a typical component of brake fluid. By mass, the most abundant solvent-soluble organic compounds were found to be the polyalkylene glycol ethers triethylene glycol monomethyl ether (2-[2-(2-methoxyethoxy)-ethoxy]ethanol) and triethylene glycol monoethyl ether (2-[2-(2-ethoxyethoxy)-ethoxy]ethanol). Both compounds are used mainly in hydraulic fluids. The findings suggest that these compounds are lost from the hydraulic brake systems to the drum assembly, and then find their way onto the brake lining and brake dust (Rogge *et al.*, 1993). Rogge *et al.* (1993) detected a number of PAHs and substituted PAHs, with a total concentration of 16.16 mg/kg, and suggested that the build-up of heat during brake application may be responsible for the additional generation of PAHs. Some compounds present in the brake lining may also be transformed during brake application. A preliminary examination of organic compounds in brake pads used in New Zealand was carried out by Gadd & Kennedy (2000). The composition was dominated by phenols, with concentrations of phenol ranging from 6.9 to 1,310 mg/kg.

The study by Garg *et al.* (2000) revealed that 18% of airborne brake wear PM was carbonaceous material. Elemental analysis indicated that metallic species, together with silicon, phosphorous, sulphur, and chlorine, accounted for most of the remaining mass. It was also found that the analysis of the brake dust samples (seven in-use types were tested) concluded that all had a similar composition to the airborne PM. The analysis showed the presence of a variety of elements that could provide a unique source profile for use in receptor models, but a single source profile was not created since it was not known how representative the tested brake pads were of the in-service vehicle fleet. It was suggested that the analysis of a composite of brake dust samples collected from a large number of vehicles could be used as a practical way of determining an in-service brake PM emission source profile.

Table 4.2: Concentrations of various organic compounds in brake wear particles (concentrations in mg/kg).

‘N/D’ = not detected; ‘-’ = not reported. Data from Rogge *et al.* (1993).

| Compound | Concentration | Compound | Concentration | Compound | Concentration |
|----------------------------------|---------------|---|---------------|--|---------------|
| n-Alkanes | | n-Alkanoic acids | | Pentacyclitriterpanes | |
| Nonadecane | 2.0 | Hexanoic acid | 82.1 | 22,29,30-trisnorhopane | 0.35 |
| Eicosane | 2.4 | Heptanoic acid | 35.4 | 17 β (H),21 β (H)-30-norhopane | 0.69 |
| Heneicosane | 1.7 | Octanoic acid | 60.5 | 17 β (H),21 β (H)-hopane | 1.2 |
| Docosane | 1.4 | Nonanoic acid | 87.4 | 22S-17 α (H),21 β (H) homohopane | 0.41 |
| Tricosane | 3.2 | Decanoic acid | 18.4 | 22R-17 α (H),21 β (H) homohopane | 0.18 |
| Tetracosane | 1.9 | Undecanoic acid | 41.1 | 22S-17 α (H),21 β (H) bishomohopane | 0.13 |
| Pentacosane | 5.7 | Dodecanoic acid | 12.5 | 22R-17 α (H),21 β (H) bishomohopane | 0.1 |
| Hexacosane | 3.3 | Tridecanoic acid | 3.2 | PAH | |
| Heptacosane | 2.1 | Tetradecanoic acid | 8.1 | Phenanthrene | 0.97 |
| Octocosane | 2.0 | Pentadecanoic acid | 3.2 | Methyl (phenanthrenes, anthracenes) | 0.67 |
| Nonacosane | 3.3 | Hexadecanoic acid | 83.3 | Dimethyl (phenanthrenes, anthracenes) | 0.66 |
| Triacontane | 2.1 | Heptadecanoic acid | 4.1 | Fluoranthene | 0.69 |
| Hentriacontane | 2.3 | Octadecanoic acid | 79.5 | Pyrene | 1.10 |
| Dotriacontane | 1.6 | Nonadecanoic acid | 0.83 | Methyl (fluoranthenes, pyrenes) | 0.89 |
| Tritriacontane | 1.6 | Eicosanoic acid | 1.8 | Benzo[a]fluorene/benzol[b]fluorene | 0.26 |
| Tetracontane | 0.8 | Heneicosanoic acid | 0.87 | Benzo[g,h,i]fluoranthene | 0.34 |
| Pentatriacontane | 0.67 | Docosanoic acid | 2.1 | Benz[a]anthracene | 1.50 |
| Hexatriacontane | 0.42 | Tricosanoic acid | 1.9 | Chrysene/triphenylene | 1.70 |
| Substituted benzaldehydes | | Tetracosanoic acid | 1.6 | Methyl(benz[a]anthracenes, chrysenes, triphenylenes) | 2.20 |
| 2-methylbenzaldehyde | 0.48 | Pentacosanoic acid | 0.34 | Benzo[k]fluoranthene | 0.62 |
| 3-methylbenzaldehyde | 0.11 | Hexacosanoic acid | 0.69 | Benzo[b]fluoranthene | 0.42 |
| 4-methylbenzaldehyde | 0.3 | Heptacosanoic acid | 0.36 | Benzo[e]pyrene | 0.84 |
| 3-methoxybenzaldehyde | 13.0 | Octacosanoic acid | 0.55 | Benzo[a]pyrene | 0.74 |
| 3,4-dimethoxybenzaldehyde | 0.45 | Polycyclic aromatic ketones and quinones | | Benzo[g,h,i]perylene | 2.60 |
| 2-hydroxybenzaldehyde | 4.7 | Fluorenone | 0.82 | Polyalkylene glycol ethers | |
| n-Alkenoic acids | | Phenanthrenequinone | 0.31 | 2-(2-butoxyethoxy)ethanol | 45.7 |
| Oleic acid | 10.7 | Anthraquinone | 0.48 | 2-[2-(methoxyethoxy)ethoxy]ethanol | 248 |
| Regular steranes | | Phenanthrone/anthrone | 0.46 | 2-[2-(2-ethoxyethoxy)ethoxy]ethanol | 404 |
| Cholestanes | 0.3 | 7H-benzo[d,e]anthracen-7-one | 0.58 | 2-[2-(2-butoxyethoxy)ethoxy]ethanol | 182 |
| Cholestane | 0.26 | 6H-benzo[c,d]pyren-6-one(benzol[c,d]pyrenone) | 0.36 | Benzoic acids | |
| Ergostanes | 0.99 | Natural resins | | Benzoic acid | 31.9 |
| | | Dehydroabietic acid | 10.1 | 4-methyl-benzoic acid | 2.6 |

5 Road surface wear

There is little information from the UK concerning the effects of road surface wear on PM emissions and ambient concentrations. However, in Nordic countries the situation is rather different on account of the specific problems associated with the use of studded tyres. For example, in Sweden more than 100,000 tonnes of asphalt pavement are worn down every year due to the use of studded tyres and road salt (Gustafsson, 2001). In Norway during the mid-1990s, the use of studded tyres was estimated to be responsible for the wear of around 250,000 tonnes of asphalt per year (NILU, 1996). Calculations of annual mean concentrations of PM₁₀ in the centre of the largest cities in Norway showed that 20% to 30% of the airborne dust was due to studded tyre wear. A significant portion of the tyre and road surface wear literature therefore relates to the use of studded tyres. However, as studded tyres are illegal in the UK and several other European countries, this literature has not been reviewed comprehensively here.

5.1 Road surface wear factors

Asphalt wear has been estimated by Muschack (1990) to be 3.8 mg/vkm. CBS (1998), cited in Lükewille *et al.* (2001), reported wear factors for LDVs and HDVs of 7.9 and 38 mg/vkm respectively, although these values also included tyre and brake wear. For New Zealand, Kennedy *et al.* (2002) calculate a wear factor of 0.44 g/vkm for a road surface containing 50% bitumen. In a situation where the bitumen comprises only 10% of the worn surface, this figure would be reduced to 0.09 g/vkm.

However, in areas where there is extensive use of studded tyres during the winter, the wear of the road surface, and the resulting PM concentrations due to resuspension, are considerably higher. Indeed, the wear when non-studded tyres are used is insignificant compared with studded tyres are used (Sörme and Lagerkvist, 2002). In Sweden, an average of 24 g/vkm of asphalt is worn off during winter (Lindgren, 1996), although it was estimated by Carlsson *et al.* (1995) that the introduction of softer studs and more durable asphalt would have reduced this to 11 g/vkm by 2000. The average wear factor of roads in Stockholm has been estimated to be 4-6 g/vkm (Jacobsson and Hornwall, 1999). Winter maintenance procedures in cold climates, such as traction sanding (the dispersion of sand aggregate on the road surface) and the use of studded tyres, have been associated with high airborne particle concentrations through a formation process known as the 'sandpaper effect' (Kupiainen *et al.*, 2003). The wear of the road surface increases with moisture level, and is 2 to 6 times larger for a wet road than for a dry one (Folkesson, 1992). It also increases after salting of the road, since the surface remains wet for longer periods. Vehicle speed, tyre pressure and air temperature also affect road wear. As the temperature decreases the tyres become less elastic, with the result that the road surface wear rates increase (NTNU, 1997).

5.2 PM size range

Very little information on the size distribution of road surface wear particles can be found in the literature. In the only study identified for this review, Fauser (1999) found that airborne bitumen particles mainly ranged in size between 0.35µm and 2.8µm, with a mean of around 1 µm.

5.3 PM emission factors

Few studies have provided emission factors for road surface wear according to PM₁₀ or any other metric. The abrasion of paved roads is typically included in total non-exhaust emission factors, whereby tyre, brake and road wear, as well as resuspension, are included (Lükewille *et al.*, 2001).

However, it can be possible to disentangle the different sources by consideration of particle composition. Based on the chemical analysis of filters collected in the Hatfield Tunnel, followed by principal component analysis, Luhana *et al.* (2004) determined LDV and HDV emission factors for road surface wear of 3.1 mg/vkm and 29.0 mg/vkm respectively, but these values were considered to be highly uncertain.

Kupiainen *et al.* (2005) tested the 'sandpaper effect' using an indoor road simulator. A range of non-studded (friction) and studded tyres were tested on a bituminous road surface with varying amounts of traction sand (two types of granite and one diabase). In the tests using non-studded tyres and no sand, PM₁₀ emission factors at 15 km/h and 30 km/h were 11 mg/vkm and 9 mg/vkm respectively. Following the addition of between 865 and 1046 g/m² of traction sand, the PM₁₀ emission factor for non-studded tyres increased to between 36 and 108 mg/vkm. In the case of studded tyres, the emission factor without traction sand was 17 mg/vkm at 15 km/h and 40 mg/vkm at 30 km/h. Following the addition of traction sand (865 to 2112 g/m²), the emission factor increased to between 40 and 155 mg/vkm. The traction sand with the lowest resistance to fragmentation resulted in the highest airborne PM concentrations. Analysis of PM₁₀ filters revealed that more than 90% of the particles collected were aluminosilicates, and therefore derived from the road surface and the traction sand. For non-studded tyres in the absence of traction sand, a maximum of 5% of PM₁₀ originated from the tyres. If it is assumed that, for non-studded tyres and no traction sand (*i.e.* conditions which might be more typical of the UK), 95% of PM₁₀ is due to road surface wear and 5% due to tyre wear, this gives road surface and tyre wear emission factors of around 8.5 to 10.5 mg/vkm and around 0.5 to 0.6 mg/vkm at low speeds. These findings contradict the view expressed by Kennedy *et al.* (2002) that in terms of the tyre/road surface wear interactions, any material loss is dominated by the wear material from the tyre treads.

5.4 Road surface and PM composition

A range of concrete-based and asphalt-based road surfacings are used in the UK. Concrete surfacings are composed of coarse aggregate, sand and cement. Asphalts are mixtures of mineral aggregate, sands, filler, and bitumen binder, though the composition can vary widely both from country to country and within countries. Generally, the stone content is around 95% and the bituminous binder around 5%. The properties of asphalt can be modified by additives such as adhesives, polymers, and different types of filler. Van Heystraeten (1996) explained that old rubber tyre material can be used in modified asphalt mixes. The amount of crumb rubber used influences the amount of bitumen added to the mix, and hence the mineral content of the material. This road surface technology was developed in Sweden with the purpose to of using the rubber properties to control snow and ice.

Bitumen is a viscous hydrocarbon material containing many chemical components, and with a mean molecular weight of around 800 (Jennings *et al.*, 1992). It has been described as a colloidal dispersion of solid particles, known as maltenes (which are soluble in *n*-heptane) and asphaltenes (which are not soluble in *n*-heptane), in a medium of saturated hydrocarbons with a molecular weight range of 200-600 (oils) and polyaromatic hydrocarbons with a higher molecular weight (resins). The asphaltenes have a molecular weight of up to 1000 (C₈₀) (Hagen *et al.*, 1989). The complexity of bitumen makes it very difficult to characterise (Lindgren, 1996), and few workers have attempted to identify bitumen particles in the environment. Its precise composition varies according to the source of the crude oil from which the bitumen originates, and any subsequent modification (Whiteoak, 1990). It has been suggested by Fauser (1999) that asphaltenes and maltenes from bitumen, which are amongst the largest molecules present in the atmosphere, can be used as tracers for road surface wear.

Concentrations of specific organic compounds in road surface material have not been reported in detail. Gadd and Kennedy (2000) found that the mass composition of raw bitumen was dominated (86%) by high-molecular weight *n*-alkanes (>C₂₉), with C₁₅-C₂₈ *n*-alkanes contributing 13%, and C₁₀-C₁₄ *n*-alkanes contributing 1%. Elvebakken (1991) used PAHs as bitumen tracers, and noted that more than 300 different PAHs are present in bitumen. However, each compound could only be found in a concentration of less than 0.1%. According to Kennedy *et al.* (2002), only small amounts of individual PAHs are present in bitumen because the distillation process removes most of them. Gadd and Kennedy (2000) reported a total PAH concentration in New Zealand 180/200+P bitumen of 9.2 mg/kg. Brandt and Groot (2001) observed a total PAH concentration in bitumen of 6.4-15.2 mg/kg. Elvebakken (1991) stated that there is no PAH that is unique to bitumen. This lack of specificity probably renders PAHs unsuitable as tracer species for bitumen. He *et al.* (1993) and Rosenmejer (1993) used the polar, aromatic and aliphatic fractions as bitumen tracers, but did not reveal a usable fingerprint profile for bitumen.

Traces of metal such as vanadium, nickel, iron, magnesium, and calcium are also present in bitumen. Lindgren (1996) determined the metal composition of bitumen (B180) and two rock minerals used in asphalt, a gabbro and a porphyry. For the bitumen, chromium, copper, and zinc were below their detection limits. Analyses of raw bitumen and bitumen samples collected from roads in New Zealand were reported by Kennedy & Gadd (2000). The metal concentrations in road surface materials obtained in these studies are given in Table 5.1.

Kennedy & Gadd (2000) found that in the raw bitumen few metals were present in elevated concentrations. A wider range of elements were detected in the road bitumen samples, and concentrations of several metals were much higher. The road bitumen samples will have included some material that was incorporated into the bitumen surface as a result of vehicles driving on the road surface, and some metals which were added to the bitumen when it was milled from the road. Concentrations of cobalt, molybdenum, nickel, tin and zinc were higher in the road bitumen samples than raw bitumen, indicating additional sources of these pollutants.

Table 5.1: Metal concentrations in road surface material (concentrations in mg/kg, unless stated).
 'N/D' = not detected; '-' = not reported.

| Metal | Asphalt constituents | | | | | |
|-------|----------------------|-----------------|-----------------|----------------------|-----------------------|-----------------------|
| | Asphalt | Gabbro | Porphyry | Bitumen (B180) | Raw bitumen | Road bitumen |
| | Alloway (1990) | Lindgren (1996) | Lindgren (1996) | Lindgren (1996) | Kennedy & Gadd (2000) | Kennedy & Gadd (2000) |
| Al | - | - | - | - | <5-7 | 12400-22100 |
| As | - | 11.2 | 2.52 | - | <1 | 0.9-12.6 |
| Ba | - | - | - | - | <0.2-0.2 | 19.2-33 |
| Be | - | - | - | - | <0.1 | 0.46-1.23 |
| Ca | - | - | - | - | <50 | 6310-9520 |
| Cd | 0.09 | 0.127 | 0.136 | - | <0.05 | 0.05-0.15 |
| Co | - | - | - | - | <0.2-0.2 | 17.4-31.6 |
| Cr | 4 | 238 | 63.9 | <35 | <1 | 46.3-102.2 |
| Cu | 13 | 70.5 | 21.9 | <17 | <1 | 40-60.1 |
| Fe | - | - | - | - | <20-100 | 25100-48100 |
| Hg | - | - | - | - | <0.01 | <0.01-0.03 |
| K | - | - | - | - | <50 | 1600-3510 |
| La | - | - | - | - | <0.1 | 0.56-9.97 |
| Li | - | - | - | - | <0.2-2.9 | 3.72-5.03 |
| Mg | - | - | - | - | <4 | 15000-29000 |
| Mn | - | - | - | - | <0.5 | 390-654 |
| Mo | - | - | - | - | <0.2-1.5 | 1.21-1.97 |
| Na | - | - | - | - | <20-20 | 5520-14100 |
| Ni | 0.5 | 110 | 16.2 | 15-100 ¹⁹ | 2-32 | 77.1-153 |
| Nb | - | - | - | - | - | 2.6-3.5 |
| Pb | 24 | 2.75 | 19.8 | - | <0.2 | 1.89-146 |
| Rb | - | - | - | - | <0.2 | 7.14-12.6 |
| Sb | - | - | - | - | <0.2 | <0.2-0.3 |
| Sc | - | - | - | - | - | 2.5-3.6 |
| Se | - | - | - | - | N/D | N/D |
| Sn | - | - | - | - | <0.5-4 | 0.5-1.3 |
| Sr | - | - | - | - | <0.5 | 64.8-119 |
| Th | - | - | - | - | <0.02 | <0.02-0.03 |
| Ti | - | - | - | - | - | 2310-3885 |
| U | - | - | - | - | <0.05 | 0.32-0.67 |
| V | - | 215 | 24.9 | 50-600 ²⁰ | <50-110 | 60-80 |
| Zn | 52 | 149 | 36.3 | <17 | <2-99 | 47.4-79.9 |

¹⁹ Baekken (1993)

²⁰ Baekken (1993)

6 Corrosion

The corrosion of vehicle components, crash barriers, and street furniture can contribute to the dust deposited on the road surface, and hence forms part of resuspended material. Corrosion may also be a direct source of airborne particles, but there is little information relating to its significance.

Fry *et al.* (2005) determined the sources of heavy metal contamination in road dust at two locations in the south of England. The first stage of the work was carried out at the TRL research track, and the second stage on the A243 north of Junction 9 of the M25 motorway. The two roads were similar in their density of road furniture and their surroundings. However, the traffic flow on the test track was estimated to be less than 50 vehicles per day, whereas on the A243 it was around 30,000 vehicles per day. Samples were taken from all potential sources of contamination, including lamp posts, sign posts, drain covers and grills, barriers, and white lining. Surface dust samples were swept from areas adjacent to the base of lamp posts and from similar surfaces (both asphalt and concrete), as far away from the metallic sources as possible. Major elements in the dust were quantitatively determined by inductively coupled plasma techniques. Typical chemical 'fingerprints' for the source materials were also determined from the literature. These were used to assess the potential contributions of contaminants from road surface and street furniture. It was concluded that soil was not a major component of the road dust, and that traffic had a smaller influence than commonly perceived. It therefore appeared that the road surface and the street furniture were significant factors in the contamination of road dust. There were four key sources of heavy metals contamination: asphalt, galvanised metals, iron, and steel but source apportionment was difficult due to the preferential dissolution of some elements during weathering and corrosion. Further work is required to study these weathering effects to ascertain the relative solubility of the heavy metals before a model can be developed to predict the relative inputs from the sources identified.

Legret and Pagotto (1999) conducted a study of the corrosion of safety barriers in galvanised steel, by means of an experimental set-up consisting of a 150m-long barrier equipped with a gutter. Rain water was collected and the heavy metal content was determined over a one year period. The amount of zinc removed was approximately 950 g/km/year. The corresponding fluxes for cadmium, copper and lead were 0.16, 0.23, and 2.1 g/km/year respectively.

Compositional analysis of used car body parts and fresh primer paint by Brewer (1997) revealed that a number of heavy metals are found in a range of concentrations in different vehicle components. Unsurprisingly, in all bodywork and chassis components (except paint) Fe was found in the highest concentrations. Exhaust components also contain high concentrations of Pb, Zn, and Cu. The lead probably originated from the use of leaded fuel. Car paint was found to contain high levels of barium, aluminium, iron, chromium, zinc, and lead.

7 Resuspension of road dust

Several studies in the United States have indicated that the resuspension of paved road dust contributes significantly to atmospheric PM₁₀ and PM_{2.5} concentrations (Chow *et al.*, 1995; Schauer *et al.*, 1996; Kleeman and Cass, 1999). Kleeman and Cass (1999) found that the entrainment of road dust from paved and unpaved roads was responsible for 34% of all PM₁₀ and 20% of all PM_{2.5} in Los Angeles. The fact that the same study showed that exhaust emissions from road vehicles were responsible for 5% of all PM₁₀ and 14% of all PM_{2.5} illustrates the potential importance of resuspension processes. It was also found that particles emitted from paved road dust sources, and crustal material other than paved road dust, dominated the 24-hr size distribution for particles larger than 1µm. Similarly, Gaffney *et al.* (1995) and Zimmer *et al.* (1992) estimated that the contribution of emissions from paved roads to total PM₁₀ might be as high as 30% in California and 40% to 70% in the Denver Metropolitan area respectively. An inventory of fine particulate organic carbon emissions to the Los Angeles area atmosphere compiled by Hildemann *et al.* (1991) indicated that paved road dust was the second largest source of fine aerosol organic carbon particles to the urban atmosphere. Brake linings and tyre wear were found to be the eleventh and thirteenth largest sources respectively.

A study in France showed that resuspension may be three to seven times higher than exhaust emissions from road transport (Jaeger-Voirol and Pelt, 2000). At a range of sites in Germany, PM₁₀ emissions due to resuspension have also been found to be up to several times larger in magnitude than those from vehicle exhaust, but with a large range of variation between locations (Pregger and Friedrich, 2002; Düring and Lohmeyer, 2003). Investigations in Berlin by Rauterberg-Wulff (1998) showed that more than 50% of traffic-derived PM was in the coarse mode, with this mode consisting of 22% carbonaceous material (elemental and organic carbon), and 78% minerals. This indicated that most of the coarse mode particles are due to resuspension of road dust and tyre and brake wear. Even the fine PM mode contained a considerable proportion of mineral matter.

As already stated in Chapter 5, resuspension is also a major concern in Nordic countries, especially in relation to the use of studded tyres and road sanding. Johansson (2002) derived emission factors for non-exhaust emissions for PM_{2.5} and PM₁₀ which were respectively equal to, and 9 times larger than, the exhaust emission factors. The resuspension of road dust is the most important local source in urban areas of Sweden, contributing 17 to 22 µg/m³ to annual mean concentrations at kerbside sites (Areskoug *et al.*, 2004). At sites where traffic is the major source, the coarse particles mass is about 6 times larger than the fine particle mass, although PM_{2.5} can contain a considerable fraction of resuspended particles (Areskoug *et al.*, 2004).

In the UK, Harrison *et al.*, (2001) concluded that vehicle-induced resuspension has a source strength approximately equal to that of exhaust emissions. However, UK emission inventories and models cannot adequately include the resuspension of road dust as a particle source due to the absence of appropriate emission factors.

7.1 The road dust complex: sources and composition

Road surfaces act as repositories for various forms of particulate matter. The dust which collects on the road surface is an agglomeration of particles from a wide range of sources; virtually any anthropogenic and biogenic sources can contribute to the road dust complex. In addition to the particles deposited as a result of abrasion and corrosion processes, road dust may also contain exhaust particles, de-icing salt and grit deposited during winter maintenance operations, crustal and vegetative material carried onto the road by vehicles, and matter from a range of nearby sources which are not related to road transport (*e.g.* crustal and vegetative material, and material from

industrial, commercial and domestic activity). Wind-borne particles derived from the above sources at more distant locations are also deposited on the road surface.

Clearly the composition of road dust is going to vary substantially with location and season, as well as many other parameters. Legret and Pagotto (1999) labelled road dust semi-quantitatively as 60% 'sand', 20% 'fine sand', and 20% 'silt', and found it to be heavily contaminated by lead, copper, cadmium and zinc, probably originating from tyre and brake wear. Concentrations of chromium and nickel were found to be low. Hildemann *et al.* (1991) found that paved road dust samples consisted of 27% SiO₂, 17% organics, 11% Al₂O₃, 9% Fe₂O₃, and 4% calcium. The detailed composition of the solvent-soluble organic component of the same road dust samples was determined by Rogge *et al.* (1993). Orr and Deletic (2000) observed high heavy metal concentrations in the finest particle fraction analysed (2 to 63 µm). Miguel *et al.* (1999) found that the relative abundance of iron, copper, zinc, lead and sulphur increased with decreasing particle size.

The amount of PM deposited on the road surface is dependent upon many factors, the most important being the amount of traffic, the quality of the road surface, to what extent studded tyres are used, and the duration of wet periods, since the deposit on the surface increases when the surface is wet. Orr and Deletic (2000) found that the road sediment loading was at its greatest during the winter months, particularly when snow was present on the road surface. Also, the road cleaning regime is of importance, especially for the resuspension of coarse particles, but the exact effects of this are still uncertain (CAFE Working Group on Particulate Matter, 2004).

7.2 Resuspension processes

Road dust may be suspended or resuspended in the atmosphere as a result of vehicle-generated turbulence during dry periods as air is squeezed from beneath the tyre, by tyre shear caused by rotation, and by the action of the wind (Sehmel, 1973; 1976). The concentrations of PM derived from vehicle-induced resuspension will be greatest in the vicinity of roads, whereas wind-driven resuspension will be a more widespread phenomena (Harrison *et al.*, 2001). The induced turbulence is much larger for large trucks than for passenger cars, so the fraction of heavy goods vehicles in traffic is an important parameter. During wet conditions, tyre spray can be an important resuspension mechanism (Smith, 1970), though this may result in more particles being deposited on the road verge than suspended in the air. The wind can also be responsible for the advection of PM derived from these processes at other locations. Road dust therefore bears a dynamic relationship to airborne PM, and may also be removed in run-off water during periods of rainfall, subsequently contributing to toxin levels in watercourses.

The factors affecting resuspension, and resuspension rates, have already been reviewed extensively elsewhere (Nicholson, 1988). There is no intention to repeat this work here, suffice to say that there are a multitude of factors affecting resuspension, and resuspension rates remain uncertain. Some of the more important parameters are briefly summarised in the following paragraphs.

Data presented by APEG (1999) and Harrison *et al.* (2001) showed that, within individual seasons PM₁₀ and PM_{2.5} concentrations were strongly correlated, but the percentage of PM₁₀ comprised by PM_{2.5} showed a strong seasonal dependence. The proportion of coarse particles was greatest in the spring and summer, and least in the wetter autumn and winter seasons. This was interpreted as being most probably the result of better dispersion during the hotter months, leading to lower PM_{2.5} concentrations, and more effective wind-driven suspension of coarse particles during dry weather. Harrison *et al.* (2001) observed that inter-annual variability in the weather can have a strong influence on PM_{2.5}.

A comparison between particle concentrations near a busy street canyon (Marylebone Road) and at an urban background location (Bloomsbury) in London revealed that, at the former site, the concentration of coarse particles (PM_{COARSE}) was substantially higher (Harrison *et al.*, 2001). This was attributed to traffic-induced resuspension at the roadside locations. In mass terms, the elevation in coarse particles due to resuspension was of a similar magnitude to the elevation in fine particles due to exhaust emissions. The association of fine particles with exhaust emissions and coarse particles with resuspension was based on the observed correlations between particle concentrations and NO_x concentrations. Comparisons between particle size distributions at urban and rural locations showed that, at the rural locations, levels of both fine and coarse particles were lower. The higher level of fine particles at urban sites was attributed to vehicle exhaust emissions. The rural air was also depleted in the larger particles which corresponded to the effects of human activity, including road dust raised by traffic, construction, and industry. At other sites where PM_{10} and $PM_{2.5}$ concentrations were monitored simultaneously, particle concentrations were examined in an attempt to disaggregate the wind-driven component of coarse particles from the component resuspended by traffic. The results indicated the presence of two coarse particle components, one which was diluted with increasing wind speed, the other which increased with increasing wind speed. By adjusting for the effects of dilution, the effect of wind speed on particle suspension was clearly observed, with concentrations increasing exponentially with wind speed. The non-wind-suspended component of coarse particles corresponded with emissions from industry, construction activity, traffic-induced resuspension and biological processes.

Another factor to consider is the silt loading of the road. This is the mass of silt (material less than $75\mu m$) in a square metre of road surface. A review of resuspension studies by Nicholson (1988) showed that resuspension factors (defined as the ratio of the airborne PM concentration to the surface loading) for wind-induced processes in urban locations are typically between $10^{-8}/m$ and $10^{-5}/m$. Few silt loading measurements are available for the UK. A study carried out by Ball and Caswell (1983) in London showed data that would equate to around $0.02g/m^2$, but such a silt loading would equate to very low airborne PM concentrations (Harrison *et al.*, 2001). In addition, Harrison *et al.* (2001) observed a substantial difference between weekday and weekend concentrations of coarse particles, and very little resuspended mass was observed at night-time, when both traffic is light and wind speeds and turbulence are lower. Harrison *et al.* (2001) therefore proposed that resuspension requires two effects. The first may relate to traffic or other activity to suspend the material in the first place, the other relating to a need for wind to maintain the particles airborne. Analyses of the influence of wind speed on coarse particle concentrations and of weekday/weekend and daytime/night-time concentrations indicated that it was human activity rather than the action of the wind which was primarily responsible for the presence of coarse particles in the atmosphere at the study sites.

Nicholson and Branson (1990) examined the importance of traffic-generated resuspension by measuring surface concentrations of spherical silica particles of known sizes. Four nominal particle sizes were used ($5\mu m$, $10\mu m$, $12\mu m$, and $20\mu m$), and a fluorescent dye was attached to the particles to enable their detection. The particles were dispersed over the road surface and allowed to settle in an enclosed volume. The surface amounts of fluorescent particles were determined shortly after their deposition, and additional measurements were made in a period which included several passes by a vehicle. The vehicle was driven so that its wheels passed either side of the test areas, thus giving an indication of the effects of its turbulent wake. It was found that turbulence-induced resuspension became less efficient with decreasing particle size, and that large fractions of the deposited material were removed after the single passage of a vehicle. Experiments with a single particle size revealed that there was a clear dependence of resuspension on vehicle speed. A proportion of the fluorescent particles remained on the road after each experiment, and it was concluded that this was associated with depressions in the road surface. A further experiment, in

which the wheels passed through the test area, was used to indicate the combined effects of turbulence and tyre shear on resuspension. There was little difference in the resuspension curves to those found for vehicle-induced turbulence alone. For a busy road with fast-moving traffic, it appeared that resuspension is likely to be controlled by the rate of incorporation of deposited material. Such incorporation may be most rapid when the surface is moist, such that resuspension factors resulting from turbulence of tyre shear might be greatest immediately after a road becomes dry.

Direct, real-time measurement of particle concentrations inside the wheel arch of a moving vehicle (called TRAKER) by Kuhns *et al.* (2001) showed that aerosol concentrations behind the vehicle tyre increased exponentially with speed, and this could therefore be an important factor in the modelling of resuspended particulate matter. However, the authors also state that the speed effect could be an artefact associated with the air flow around the instrument inlet. The vehicle was used to derive silt loading values for a range of different roads in Las Vegas.

As stated earlier, the road surface wear associated with studded tyres is at least one order of magnitude larger than that associated with non-studded tyres. During winter in cold climates, a large number of pollutants can accumulate in snow and then be released when the snow melts (Baekken, 1993). When the roadways are dry, the resulting resuspension is important and may be 10 times larger than the direct emission from the exhaust pipe (Johansson, 2002). On cold, dry winter days near major arterial roads in Norway, the concentrations measured by NILU (1996) were often higher than the recommended air quality guideline by a factor of three to four. The highest concentrations measured were more than $600 \mu\text{g}/\text{m}^3$ as a daily mean. In these situations, an estimated 80% of the PM_{10} originates from resuspended road dust.

A number of additional processes relating to resuspension appear to have received relatively little attention. These include tyre spray as a resuspension mechanism during wet conditions, and the resuspension of particles from surfaces other than the road, such as building surfaces and pedestrian pavements in enclosed urban environments.

7.3 Emission factors

As emissions from resuspension depend on a number of different parameters, including traffic density, road surface material, state of the road, maintenance of the road, driving patterns, fleet composition and climatological factors, and there are large differences in all these parameters, it has not been possible to derive commonly accepted emission factors which are applicable at a large range of different situations (CAFE Working Group on Particulate Matter, 2004). Gustafsson (2003) has summarised the literature of studies of resuspension PM emission factors related to roads. The studies and emission models available give emission factors varying over several orders of magnitude, from less than 100 mg/vkm to several thousand mg/vkm for passenger cars, and up to several tens of thousand mg/vkm for heavy-duty vehicles, dependent upon which PM fraction is considered, the traffic speed, type of road, use of studded tyres, *etc.*

It is interesting to note that Gehrig *et al.* (2004) state that the concept of an emission factor for abrasion and resuspension processes is, in itself, questionable. In the case of a road location with wind blowing across the street, the concept is plausible because the mechanically produced particles are continuously being transported off the street, and limited accumulation of these particles on the road surface is to be expected. Therefore, resuspension should be of minor importance. However, without wind or in a street canyon, sedimentation of the abrasion particles on the road surface with resuspension is possible. It seems plausible that this portion of the particles can be kept in suspension by a small number of passing vehicles and is not increased by a higher number of vehicles.

Resuspension emissions would not then be proportional to the traffic volume. Abrasion emissions are also highly dependent on the condition of the pavement. Nevertheless, attempts have been made, and some of the results for various studies in different countries or regions are presented below.

United Kingdom

Nicholson (2000) developed a model which considered the net input and output from the road surface. Deposition and other inputs (such as deposition from cars, transfer on tyres, and wind blown material) were equilibrated with resuspension and surface run-off. The first estimate of resuspension was based on two assumptions: (i) all inputs other than atmospheric deposition were equal to removal in runoff, and (ii) material deposited in rain was not available for resuspension. Consequently, resuspension by traffic was equal to the dry deposition on the road. Using the model, an estimated net PM₁₀ value for resuspended material in the UK of 0.04 g/km was obtained for the vehicle fleet composition at the time. The emission factor was based on resuspension under dry conditions, not making any distinction between vehicle-induced turbulence and tyre shear, or between different vehicle types.

Nicholson's emission factor for PM₁₀ of 40 mg/vkm is quoted in the UK NAEI to aid the understanding of roadside pollution measurements, but resuspension is not included in official reported estimates to avoid double counting. Double counting could be an issue given that particles that are re-entrained in the air have already been emitted and deposited. In 2000, the estimate for PM₁₀ in the United Kingdom for this source sector was 19.4 ktonnes. Other emissions from road transport were estimated to be 31 ktonnes (Goodwin *et al.*, 2002). Clearly it is not adequate that the emission factor is to be applied to all vehicles, locations and time periods. The USEPA have derived a formula for calculating the contribution of resuspension (see Appendix 1), but it is based on old measurements near dusty roads. Such conditions are not thought to be relevant to the UK (APEG, 1999). Until further resuspension measurements are made in the UK, this source remains a major area of uncertainty in the UK particulate emission inventory.

Based on measurements in the Hatfield Tunnel, Luhana *et al.* (2004) recorded much lower PM₁₀ emission factors for resuspension: 0.8 mg/vkm for LDVs and 14.4 mg/vkm for HDVs. It is noteworthy that Düring *et al.* (2002) reported that measurements in tunnels in Germany have shown that PM₁₀ emissions due to resuspension are lower than in open streets, and according to Abu-Allaban *et al.* (2003) resuspended road dust emissions are minimal for vehicles operating in tunnels. However, according to Sternbeck *et al.* (2000), disregarding resuspension in tunnels leads to considerably overestimated vehicle exhaust emissions.

Central Europe

Gehrig *et al.* (2004) measured PM₁₀, PM₁ and NO_x concentrations at both sides of a number of busy roads in Switzerland, obtaining total non-exhaust PM₁₀ emission factors for LDV and HDV of 22 to 47 mg/vkm and 74 to 144 mg/vkm respectively. A review of studies of roadside measurements in Central Europe by Lohmeyer *et al.* (2004) indicated total non-exhaust emission factors for PM₁₀ of 4 to 70 mg/vkm for passenger cars, and 70 to 1,270 mg/vkm for HGVs. The quotient of the total PM₁₀ and the exhaust PM₁₀ varied between 1.1 and 4.6, again highlighting the importance of non-exhaust sources. It should be noted that in these cases, where the non-exhaust emission factor is derived from the total emission factor minus the exhaust emission factor, the result is dependent on the accuracy of the emission factors used for exhaust emissions. In these examples, the values used were from the Handbook of Emission Factors used in Austria, Germany and Switzerland (UBA, 2004).

Northern Europe

Emission factors for resuspension in Nordic countries have been summarised by the CAFE Working Group on Particulate Matter (2004). For Sweden, an average winter PM₁₀ resuspension emission factor of 209 mg/vkm has been determined from measurements in a Stockholm street (Johansson, 2002; Foltescu *et al.*, 2001). A model has been developed for estimating resuspension emissions in Norway (Tønnesen, 2000). When applied to the whole of the Norwegian traffic activity, this model gives an average winter PM₁₀ resuspension emission factor of 130 mg/vkm. The increased resuspension emission factors in Sweden and Norway clearly show the effects of studded tyres, which are used widely in both countries during winter months. The dominant part of the mass of the particles from the resuspension source fall in the coarse fraction (PM₁₀-PM_{2.5}), although an estimate of the resuspension emission factor for PM_{2.5} has also been calculated in the Swedish study (Johansson, 2002). The estimate is 25 mg/vkm, about one tenth of the PM₁₀ emission factor for resuspension, and is valid for a situation with extensive use of studded tyres.

Based on comparisons of roadside and urban background (roof level) concentrations of NO_x, PM₁₀ and PM_{2.5} at a street canyon site in central Stockholm, Areskoug *et al.* (2004) determined average total PM₁₀ and PM_{2.5} emission factors of 223 mg/vkm and 48 mg/vkm respectively. Assuming that the average vehicle exhaust emission of particles is approximately equal to the minimum value of the calculated PM_{2.5} emission (23 mg/vkm), the non-exhaust emissions of PM₁₀ and PM_{2.5} were estimated. For PM₁₀ the non-exhaust emissions would have thus contributed 200 mg/vkm, and 85% of the locally generated PM₁₀ concentrations would have been due to non-exhaust particle emissions. For PM_{2.5} non-exhaust emissions of PM contributed 25 mg/vkm, which meant that 45% of the locally generated PM_{2.5} concentration were due to non-exhaust emissions.

United States

Claiborn *et al.* (1995) reported PM₁₀ emission factors for resuspended dust from mixed fleets of 1000 to 6700 mg/vkm. Venkatram *et al.* (1999) observed a range for mixed fleets of 120-10,000 mg/vkm. It has been noted by Moosmüller *et al.* (1998) that large vehicles are responsible for most dust resuspension from unpaved shoulders of paved roads. The resuspension mechanism proposed by Moosmüller *et al.* (1998) was high turbulent kinetic energy in vehicle wakes, and Abu-Allaban *et al.* (2003) suggest that this should also be valid for paved road resuspension.

Based on roadside measurements at a number of locations, Abu-Allaban *et al.* (2003) determined PM₁₀ emission factors for resuspended road dust ranging from 40 to 780 mg/vkm for LDVs, and from 230 to 7800 mg/vkm for HDVs. The resuspension of road dust was found to be dependent upon vehicle speed and silt loading, and heavy-duty vehicles were found to contribute around eight times more resuspended dust than light-duty vehicles. However, ambient background PM levels were not recorded, and the authors suggested that the resuspended road dust components might therefore be overestimated.

Fitz and Bufalino (2002) measured total PM₁₀ emissions directly behind moving vehicles and characterized the emissions under a wide variety of driving conditions. Observations of vehicles travelling on unpaved roads showed that the plume does not appreciably disperse for several car lengths. PM₁₀ measurements were made at several positions 4.3m behind the rear bumper. To calculate emission factors it was assumed that a wake of the dimensions of the vehicle's frontal area was swept out, and that the PM emissions remained within this volume until the point of measurement. The emission rate was calculated by multiplying the frontal area by the concentration.

Total PM₁₀ emission factors for paved roads ranged from 64 to 124 mg/km, somewhat lower than in the other US studies.

8 Emission models for non-exhaust PM

This Chapter provides an overview of models which treat non-exhaust particulate matter from a source perspective (*i.e.* they describe emissions), as opposed to models which describe it from a receptor perspective (ambient concentrations). Only models which incorporate a distinct approach or a distinct set of emission factors have been considered. Only a few models are available, and as might be expected from the earlier discussion, the models do not always consider the sources in the same manner. These models are not evaluated in this report, as this will form the next stage of the project.

8.1 USEPA AP-42 model

The calculation methods presented in the USEPA's AP-42 compilation of emission factors (USEPA, 1995), are probably the most widely-used models for predicting non-exhaust emissions. The AP-42 provides methods to calculate emissions from brake wear, tyre wear and 'fugitive dust' emissions from unpaved and paved roads, as described in Appendix A.

The fugitive dust methods include all traffic PM sources, including exhaust emissions, abrasion products and resuspension, but have been subjected to a great deal of scrutiny. An incisive critique of the AP-42 method for paved roads by Venkatram (2000) found a number of shortcomings with the method. He concluded that the AP-42 model shows that it is not likely to provide adequate estimates of PM₁₀ emissions from paved roads. Because the model has little mechanistic basis, it relies on an input variable, the silt loading, that cannot be measured unambiguously. An analysis of those data used to develop the empirical model showed that different but equally plausible empirical models could be developed by using different subsets of the data set, and that these models provided mean emission factor estimates that could differ by a factor of two. Venkatram argued that the conclusions drawn from the study of the paved road emissions model apply to most empirical models that lack a mechanistic foundation. The results from independent evaluations of the AP-42 method (Zimmer *et al.*, 1992; Kantamaneni *et al.*, 1996) have highlighted the problem of basing a model on a particular data set. According to Fitz and Bufalino (2002), the AP-42 states that the silt loading reaches an equilibrium value without the addition of fresh material. If equilibrium is attained, then the emission rate should go to zero, although this is not what the paved road equation predicts. Therefore, it is difficult to understand how this equation could be universally applicable unless the material is continuously replaced, a phenomenon which for most public roads is not likely. Gustafsson (2003) has also identified a number of problems with the approach, including the fact that the amount of silt deposited is much less than the amount emitted due to low deposition speed compared with vehicle-induced turbulence and the wind, the silt loading is not proven to be related to the PM₁₀ (or PM_{2.5}) concentrations, and the silt loading is hard to generalise. These factors have led to very variable levels of agreement with measurements.

Winiwarter *et al.* (2002) have estimated that PM₁₀ emissions from Austrian traffic caused by resuspension exceed those from tail-pipe emissions by a factor larger than ten, when using the US EPA AP-42 method. According to APEG (1999), the AP-42 method is based on old measurements near dusty roads. Such conditions are not thought to be relevant to the UK.

8.2 Modified AP-42 for use in Germany

The USEPA AP-42 model was modified by Rauterberg-Wulff (2000) for use in Berlin (see Appendix B), but the modification only involved adjustments to the values of constants. More detailed modifications, involving the separation of the exhaust and non-exhaust contributions, have been made by Gamez *et al.* (2001) (Appendix B). The method has been shown to predict PM₁₀

concentrations which agree reasonably well with measurements, although the measurements were those used in the development of the model (Düring *et al.*, 2002). A validation exercise conducted by Lohmeyer *et al.* (2004) alongside an arterial road in Karlsruhe indicated that the modified method over-predicted roadside non-exhaust PM₁₀ concentrations. Over-predictions were also observed at motorway sites in Germany (Lohmeyer *et al.*, 2003). By 2004, the modified AP-42 method had been abandoned in favour of allocating emission factors to ‘traffic situations’ (Lohmeyer *et al.*, 2004) – see Appendix B. This approach was found to give a better performance.

8.3 SMHI model

A model was developed by the Swedish Environmental and Health Protection Administration for resuspension, proposed by Johansson *et al.* (1998) (cited in Rauterberg-Wulff, 2000 and Gustafsson, 2003) – see Appendix C. As with the USEPA AP-42 method, the equation relates to emissions of particles from all non-exhaust sources. Resuspension is calculated using the formula proposed by Sehmel (1984). Rauterberg-Wulff (2000) found that the model predicted much lower emissions than the AP-42 method, and resulted in unrealistic roadside PM concentrations. It is also unclear whether the model relates to PM₁₀ or TSP, or whether it includes exhaust emissions. Gustafsson (2003) expressed concerns regarding the validity of Sehmel’s function under all conditions, the difficulties associated with collecting the required input data, and the lack of consideration of the state of the pavement.

8.4 VLUFT model

NILU’s VLUFT model (Tønnesen, 2000) incorporates a resuspension module. The general form of the model is shown in Appendix D, but there is little supporting documentation in English. Gustafsson (2003) highlighted the potential problems of this approach as being the dependency of resuspension on fine particle emissions, the limited consideration of meteorology, and the limited consideration of differences in the pavement.

8.5 EMEP/CORINAIR method

The EMEP/CORINAIR Emissions Inventory Guidebook (EEA, 2004) presents a methodology for determining primary non-exhaust PM emissions. It has been used in the latest revision to the NAEI (NETCEN, 2005). The methodology covers tyre, brake and road surface wear; resuspension is not included. Two methods are described, a simple method for PM₁₀, and a detailed method for various size metrics which includes corrections for speed and heavy-duty vehicles. The methods are presented in Appendix E.

The detailed EMEP/CORINAIR method has been applied by Winther and Berkowicz (2004) to calculate non-exhaust particulate emissions in Denmark. The primary total non-exhaust emissions were compared with emission factors derived from roadside air pollution measurements. It was found that the emission factors for non-exhaust PM₁₀ derived from the measurements were much higher than what could be expected from the model. For PM_{2.5} (exhaust plus non-exhaust) the level of agreement between the two approaches was good, it was concluded that the underestimation of PM₁₀ was due to a much higher contribution from the coarse mode to the measurements than accounted for in the model, and the reasons for this were likely to be the exclusion of resuspension from the EMEP/CORINAIR method, and/or lack of accuracy in the emission factors for road surface wear.

8.6 CEPMEIP database

The Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance (CEPMEIP)²¹ is part of the activities aimed at supporting national experts in reporting particulate matter emission inventories to the EMEP programme under the UNECE Convention on Long Range Transport of Air Pollutants (CLRTAP). Within this work programme, TNO has compiled an overview of particulate emission estimation methods and applied these in a European emission inventory for particulates for the base year 1995. The emission factors used in CEPMEIP for tyre, brake and road surface wear are derived from the international literature (Berdowski *et al.*, 2001), and are shown in Appendix F.

8.7 RAINS database

The European Commission has established the 'Clean Air For Europe' (CAFE) programme to conduct a systematic and holistic review of all EU legislation related to air quality. The Commission has decided that the RAINS model (Alcamo *et al.* 1990), developed by IIASA²², will serve as the central integrated assessment tool for CAFE. The emission factors for non-exhaust sources used in RAINS are tabulated in Appendix G.

²¹ This site gives access to the main results of the project:

http://www.mep.tno.nl/wie_we_zijn_eng/organisatie/kenniscentra/University_projects/applied1_EM_eng.html

²² <http://www.iiasa.ac.at/rains/>

9 Summary and discussion

This Report has described the findings of a literature review of emission factors and models for non-exhaust sources of particulate matter, and has included a substantial amount of information relating to the composition of source materials and particles. It follows on from, and updates, a previous review for the EU 5th Framework PARTICULATES project (Luhana *et al.*, 2004). A number of similar reviews have also recently been conducted, notably by Kennedy *et al.* (2002) in New Zealand, and by the Swedish National Road Transport Research Institute (VTI) (Gustafsson, 2001; 2003). Although the former provides a great deal of information relating to the composition of source materials, it does not deal specifically with airborne particles. The latter reports are only available in Swedish; translations into English would be beneficial to the understanding of the subject in the UK, although much of the detail will relate to the particular conditions in Nordic countries (*e.g.* extensive use of road gritting and studded tyres). The findings of the current review are described below.

9.1 Non-exhaust PM sources

There are a number of non-exhaust processes, involving mechanical abrasion and corrosion, which can result in particulate matter being released directly to the atmosphere. The main direct emission sources are:

- Tyre wear
- Brake wear
- Road surface wear

Clutch wear and corrosion may also contribute to direct PM emissions, but cannot easily be quantified given the current level of understanding.

In addition to direct non-exhaust emissions, material previously deposited on the road surface can be suspended or resuspended in the atmosphere as a result of tyre shear, vehicle-generated turbulence, and the action of the wind. Indeed, of the non-exhaust processes, resuspension is likely to be the largest contributor to roadside PM₁₀ concentrations.

Non-exhaust particulate matter is important for a number of reasons, including the following:

- There are no EU regulations specifically designed to control non-exhaust particle emissions, though some countries have banned the use of studded tyres.
- As exhaust emission control technology improves and traffic levels increase, the proportion of total PM emissions originating from the uncontrolled non-exhaust sources will increase.
- The data relating to the emission rates, physical properties, chemical characteristics, and health impacts of non-exhaust particles are highly uncertain.

There is also a general lack of consistency in the definitions, terminology and metrics used in the study and reporting of non-exhaust particulate matter. For example, some workers have reported emission factors for the individual non-exhaust sources, whereas others have reported emission factors for various combinations of source, or simply total non-exhaust emissions. This renders the development of comprehensive empirical models from the available data rather difficult.

9.2 Tyre, brake and road surface wear

9.2.1 Wear factors

The wear factors (total material lost per km) of tyres, brakes and road surfaces have been summarised from the literature to give a broad indication of maximum possible values for airborne particle emissions (*i.e.* to provide a ‘reality check’ on airborne PM emission factors).

For ‘normal’ driving conditions, it seems that a value for total tyre wear of 100 mg/vkm would probably represent the right order of magnitude. For heavy-duty vehicles, the wear factor is likely to be an order of magnitude higher. More information is required to relate tyre wear (and PM emissions) to real-world driving patterns in a quantitative manner.

Total brake wear for light-duty vehicles appears to be around 10 to 20 mg/vkm, and around 50 to 80 mg/vkm for heavy goods vehicles. One study has estimated a brake wear factor for buses of 110 mg/vkm. For light-duty vehicles, the wear factor has been shown to decrease with an increase in average speed. Studies have also revealed that a significant proportion of the brake wear debris can arise from the disc or drum rather than the linings, a finding which could have implications concerning the interpretation of the results of studies which have only considered the brake linings. In the UK certain metals, such as iron, are enriched in roadside airborne PM, and the wear of metallic brake parts could be one of the sources. However, much more information is required on the types of brake linings in use in the UK, and the brake wear characteristics of UK vehicles.

A wide range of road surface wear factors have been reported in a small number of studies, from less than 4 mg/vkm to more than 400 mg/vkm where the surface contains a high proportion of bitumen. However, in areas where there is extensive use of studded tyres during the winter, the wear of the road surface is considerably higher - values of between 4 and 24 g/vkm have been reported. Winter maintenance procedures in cold climates, such as traction sanding (the dispersion of sand aggregate on the road surface) and the use of studded tyres have also been associated with high airborne particle concentrations, through a formation process known as the ‘sandpaper effect’. In the UK, road de-icing involves the application of rock salt, but little information of the effects of this particular approach on airborne PM has been reported.

9.2.2 PM size and airborne proportion

Not all worn material from tyres, brakes and road surfaces will be emitted as airborne PM. Some of the weight loss will be in the form of carbon dioxide and water vapour, although it seems that gaseous emissions are not significant. A fraction of the wear material may adhere to vehicle surfaces, and some much coarser solid material will be deposited on the road.

The literature indicates that probably less than 10% of car tyre wear material is emitted as PM₁₀ under ‘typical’ driving conditions, but the proportion could be as high as 30%. Both fine and coarse particles are emitted directly to the atmosphere as a result of tyre wear. A plausible mechanism for the distinction is the volatilisation (the thermal degradation of tyre polymer and the volatilisation of extender oils) and subsequent condensation of material in the ultrafine particle mode, and normal mechanical wear for larger sizes. The relative mass contributions of the fine and coarse modes appear to be rather variable. Although it is commonly assumed that airborne tyre wear particles are mainly present as PM_{COARSE}, this does not appear to be fully supported by the literature.

One study found that typically 50% of brake wear debris from LDVs escapes the vehicle and enters the atmosphere, and data generally indicate that more than 80% of airborne brake wear particles can

be classified as PM₁₀. A substantial amount of the PM₁₀ can be present as PM_{2.5} and smaller fractions, but the proportions appear to be highly variable (results indicate between 5% and 75% for PM_{2.5}).

Very little information on the size distribution of road surface wear particles could be found in the literature. In the only study identified, airborne bitumen particles mainly ranged in size between 0.35µm and 2.8µm, with a mean of around 1 µm.

9.2.3 Contribution to airborne PM concentrations

The contributions of tyre, brake and road surface wear to airborne particle concentrations have been investigated in a small number of studies. These studies have been conducted in different locations, at different types of site, and near to roads with different traffic characteristics. One could not therefore expect a great deal of consistency in the results, but a broad estimate is available for some sources. Contributions of tyre wear to TSP at roadside sites in the range of 0.5 to 10% , and 1 to 10 µg/m³) have been observed. For PM₁₀, the equivalent percentage range is 7.5 to 20%. Little equivalent information is available for brake and road surface wear, although one study found that roadside PM₁₀ was comprised of less than 1% brake wear, but 30% 'road abrasion and resuspension'. Indeed, road abrasion and resuspension are often inseparable in the literature, and hence contributions of road abrasion to airborne PM are considered in the discussion relating to resuspension.

9.3 Corrosion

The corrosion of vehicle components, crash barriers, and street furniture can contribute to the dust deposited on the road surface, and hence forms part of resuspended material. Corrosion may also be a direct source of airborne particles, but there is little information relating to its significance. It may not be a rapid enough process to represent a significant air quality concern. Further work is required to study weathering effects.

9.4 Resuspension

Several studies in the United States and Europe have indicated that the resuspension of paved road dust contributes significantly to atmospheric PM₁₀ and PM_{2.5} concentrations. In the UK, it appears that vehicle-induced resuspension has a source strength approximately equal to that of exhaust emissions. As already stated, resuspension is also a major concern in Nordic countries, especially in relation to the use of studded tyres and road sanding during winter. In some locations it may represent a source strength several times higher than that of exhaust emissions.

Road dust may be suspended or resuspended in the atmosphere as a result of vehicle-generated turbulence as air is squeezed from beneath the tyre, by tyre shear caused by rotation, and by the action of the wind. The concentrations of PM due to vehicle-induced resuspension will be greatest in the vicinity of roads, whereas wind-driven resuspension will be a more widespread phenomena (Harrison *et al.*, 2001). Harrison *et al.* (2001) proposed that resuspension requires two components. The first may relate to traffic or other activity to suspend the material in the first place, the other relates to a need for wind to maintain the particles airborne. Traffic activity, rather than the action of the wind, appears to be primarily responsible for the presence of coarse particles in the atmosphere, and in the case of vehicle-induced resuspension, turbulence appears to be more effective than tyre shear, although the effect of turbulence appears to become more difficult with decreasing particle size. The induced turbulence is much larger for large heavy-duty trucks than for passenger cars, and there is a strong dependence of resuspension on vehicle speed. Another factor to consider is the silt loading of the road, but as the silt loading is reduced by the passage of each vehicle there ought to be a time dependence of resuspension. However, it has been shown that large fractions of the deposited

material can be removed after the passage of just a few vehicles. This implies that the silt must be replenished, but the processes are poorly understood. The road surface wear associated with studded tyres is at least one order of magnitude larger than from non-studded tyres. During winter a great number of pollutants can be accumulated in the snow and suspended in the air by traffic when the snow melts, and the road dries. Wind-driven suspension of coarse particles also becomes more effective during dry weather.

A number of additional processes relating to resuspension appear to have received relatively little attention. These include tyre spray as a resuspension mechanism during wet conditions, and the resuspension of particles from surfaces other than the road, such as building surfaces and pedestrian pavements in enclosed urban environments.

Further information on resuspension in the UK is being derived in the EPSRC DAPPLE (Dispersion of Air Pollution & Penetration into the Local Environment) project²³, and the results of this work are expected shortly.

9.5 Emission factors for airborne PM

For road transport, it is commonly assumed that most primary fine particles (PM_{2.5}) are emitted from the exhaust, whereas many of the coarse particles (PM_{2.5-10}) are considered to originate from non-exhaust sources. The literature review covered PM emission factors for non-exhaust sources derived from a range of direct measurement and receptor modelling studies. Table 9.1 shows a summary of the PM₁₀ non-exhaust emission factors, with exhaust emission factors included for comparison where appropriate.

In the results of Abu-Allaban *et al.* (2003), no tyre wear emissions at all were detected, and brake wear emissions appeared to relate to two types of condition: no braking at all, and harsh braking at motorway exits. The highest brake wear HDV emission factor reported by Abu-Allaban *et al.* (2003) (610 mg/vkm) is somewhat higher than the total brake wear factors reported in a number of other studies. It could be argued that these conditions are not representative of typical driving. For the purposes of summarising the data, these results have been excluded.

Considering only the PM₁₀ emission factors derived for all non-exhaust sources in European studies, for LDVs values for combined non-exhaust sources range between 4 and 92 mg/vkm, whereas the emission factors for HDVs range between 70 and 1,270 mg/vkm. These are still obviously very wide ranges. Emission factors for mixed fleets range from 12 to 209 mg/vkm (the values at the high end of the range include locations where studded tyres are in use), and assuming that the proportion of HDVs in the fleet is typically 10 to 20%, this range seems broadly consistent with the individual ranges for LDVs and HDVs.

For LDVs, PM₁₀ emission factors for tyre wear range from less than 1 mg/vkm to 13 mg/vkm. For HDVs, tyre wear emission factors of between 7.5 and 200 mg/vkm have been reported. Reported emission factors for motorcycles are 2.5 to 7 mg/vkm. Emissions for LDV brake wear range from 1 to 8 mg/vkm, and HDV Brake wear emission factors range from 3.5 to 41 mg/vkm. According to Sanders *et al.* (2003), the old USEPA brake wear PM₁₀ value for LDVs of around 8 mg/vkm still adequately describes brake emissions during urban driving. The combined tyre and brake wear emission factors for LDVs and HDVs from the Hatfield Tunnel study of 6.9 and 49.7 mg/vkm fall within the range of values reported elsewhere, if perhaps a little on the low side. Values for road

²³ <http://midsummer.mech.surrey.ac.uk/~DAPPLE/>

surface wear due to LDVs range from 3.1 to 10.5 mg/vkm. Only one emission factor for road surface wear due to HDVs could be found in the literature (29 mg/vkm).

The literature has tended to place a large emphasis on resuspension, partly because the emission factors for brake and tyre wear do not account for very large airborne concentrations. A number of workers have attempted to derive emission factors total PM₁₀ emissions, or total non-exhaust PM₁₀ emissions, with resuspension being included in both. However, few studies have attempted to derive emission factors solely for resuspension. The two studies that have attempted this by measurement (Abu-Allaban *et al.* 2003 and Luhana *et al.*, 2004) have produced emission factors which differ by up to three orders of magnitude.

Some of these differences may be due to the use of a range of methodological approaches. Düring *et al.* (2002) reported that measurements in tunnels in Germany have shown that PM₁₀ emissions due to resuspension (all non-exhaust) are lower than in open streets. According to Abu-Allaban *et al.* (2003), for vehicles operating in tunnels (the measurements of Luhana *et al.*, 2004 were conducted in the UK Hatfield Tunnel) the resuspended road dust emissions are minimal. However, according to Sternbeck *et al.* (2000), disregarding resuspension in tunnels leads to considerably overestimated vehicle exhaust emissions. The more theoretical mass balance approach of Nicholson (2000) yielded a mixed fleet emission factor between those of the other two studies. Whilst some of the factors determining particle resuspension such as tyre shear are likely to be similar inside and outside tunnels, other processes may be very different. The containment provided by the tunnel may influence the scale and intensity of vehicle-induced turbulence, possibly influencing the efficiency of this factor in the resuspension process. Perhaps most importantly, the sources and loadings of particles on the road surface within a tunnel are likely to differ from those externally, and therefore the amount of material on the road surface available for resuspension will be different.

It is clear that the resuspension source is very important for PM concentrations in hot spot situations, such as close to streets with large traffic flows during dry road conditions, especially when studded tyres are used extensively. Differences between locations may also be due to large differences in climate. There do appear to be some fundamental differences between non-exhaust processes in the US and those in Europe. In particular, the total emission and resuspension emission factors derived in US studies are extremely variable, and often rather high. For mixed fleets, the resuspension results from the US studies appear to be consistently higher than total emissions from European studies, even those including studded tyres. More research is needed to clarify the strength of the resuspension source of PM, for various traffic, meteorological and road conditions.

Interestingly, Gehrig *et al.* (2004) state that the concept of an traffic-related emission factor for abrasion and resuspension processes is, in itself, questionable. It seems plausible that particles from road dust can be kept in suspension by a small number of passing vehicles, and is not increased by a higher number of vehicles. Resuspension emissions would not then be proportional to the traffic volume. However, this view is only valid if the resuspension process is very efficient. If the various resuspension processes are inefficient, then traffic volumes will be influential although the source strength might not scale directly with traffic volume. UK field measurement data do not support the idea of a relatively constant concentration irrespective of traffic volumes. Coarse particle mass at roadside and urban background locations in the UK is heavily influenced by traffic activity, but in the absence of a clear disaggregation between resuspension and other coarse particle sources, it is very hard to provide data which seriously challenge Gehrig's hypothesis. In the absence of wholly specific chemical tracers, it is going to prove extremely difficult to disentangle the various source contributions.

Table 9.1: summary of PM₁₀ emission factors for non-exhaust sources

| Study | Notes on methodology | PM ₁₀ emission factor (mg/vkm) | | | | | | | | |
|----------------------------------|---|---|------------|-----------------------|------------------------|-------------------------------------|--------------|-------------------|---------|-------------|
| | | Tyre wear | Brake wear | Combined tyre & brake | Road surface wear | Combined tyre, brake & road surface | Resuspension | All non-exhaust | Exhaust | All sources |
| Light-duty vehicles | | | | | | | | | | |
| Cha <i>et al.</i> , 1983 | Asbestos brakes | - | 7.8 | - | - | - | - | - | - | - |
| USEPA, 1995 | | 5 | - | - | - | - | - | - | - | - |
| Berdowski <i>et al.</i> , 1997 | N/A | - | - | - | - | - | - | 70 | - | - |
| CBS, 1998 | N/A | - | - | - | - | 7-9 | - | - | - | - |
| Rauterberg-Wulff, 1998 | Tunnel measurements | 6.1 | 1.0 | - | - | - | - | 32 | - | - |
| Carbotech, 1999 | N/A | - | 1.8-4.9 | - | - | - | - | - | - | - |
| EMPA, 2000 | N/A | 13 | - | - | - | - | - | 30 | - | - |
| Garg <i>et al.</i> , 2000 | Wheel dynamometer | - | 2.9 – 7.5 | - | - | - | - | - | - | - |
| Abu-Allaban <i>et al.</i> , 2003 | Roadside measurements, CMB | 0 | 0/79 | - | - | - | 41-780 | - | 7-65 | - |
| Luhana <i>et al.</i> , 2004 | Tunnel measurements, PCA | - | - | 6.9 | 3.1 | 10 | 0.8 | 10.8 | 13.9 | - |
| Gehrig <i>et al.</i> , 2004 | Upwind-downwind comparison | - | - | - | - | - | - | 22-47 | 11-16 | - |
| Gehrig <i>et al.</i> , 2004 | Roadside-background comparison | - | - | - | - | - | - | 17-92 | 10-13 | - |
| Lohmeyer <i>et al.</i> , 2004 | Upwind-downwind comparison | - | - | - | - | - | - | 4-70 | - | - |
| Kupiainen <i>et al.</i> , 2005 | Road simulator, low speeds | 0.5-0.6 ²⁴ | - | - | 8.5-10.5 ²⁵ | - | - | - | - | - |
| Heavy-duty vehicles | | | | | | | | | | |
| USEPA, 1995 | | 7.5-22.5 | - | - | - | - | - | - | - | - |
| Berdowski <i>et al.</i> , 1997 | N/A | - | - | - | - | - | - | 1170 | - | - |
| CBS, 1998 | N/A | - | - | - | - | 38 | - | - | - | - |
| Rauterberg-Wulff, 1998 | Tunnel measurements | 31 | 24.5 | - | - | - | - | 834 | - | - |
| Carbotech, 1999 | M/A | - | 3.5 | - | - | - | - | - | - | - |
| EMPA, 2000 | N/A | 200 | - | - | - | - | - | 450 | - | - |
| Luhana <i>et al.</i> , 2004 | Tunnel measurements, PCA | - | - | 49.7 | 29 | 78.7 | 14.4 | 93.1 | 79.3 | - |
| Abu-Allaban <i>et al.</i> , 2003 | Roadside measurements, CMB | 0 | 0/610 | - | - | - | 230-7800 | - | 57-570 | - |
| Gehrig <i>et al.</i> , 2004 | Upwind-downwind comparison | - | - | - | - | - | - | 74-144 | 193-200 | - |
| Gehrig <i>et al.</i> , 2004 | Roadside-background comparison | - | - | - | - | - | - | 115-819 | 187-449 | - |
| Lohmeyer <i>et al.</i> , 2004 | Upwind-downwind comparison | - | - | - | - | - | - | 70-1270 | - | - |
| Motorcycles | | | | | | | | | | |
| Berdowski <i>et al.</i> , 1997 | N/A | - | - | - | - | - | - | 23 | - | - |
| CBS, 1998 | N/A | - | - | - | - | 2-4 | - | - | - | - |
| Carbotech, 1999 | N/A | - | 0.9 | - | - | - | - | - | - | - |
| Environment Australia, 2000 | N/A | 2.5 | - | - | - | - | - | - | - | - |
| EMPA, 2000 | N/A | 7 | - | - | - | - | - | - | - | - |
| Mixed fleets | | | | | | | | | | |
| Muschack, 1990 | N/A | - | - | - | 3.8 ²⁶ | - | - | - | - | - |
| Claiborn <i>et al.</i> , 1995 | N/A | - | - | - | - | - | - | - | - | 1000-6700 |
| Venkatram <i>et al.</i> , 1999 | N/A | - | - | - | - | - | - | - | - | 120-10000 |
| Keuken <i>et al.</i> , 1999 | | - | - | - | - | - | - | 12-19.7 | - | - |
| Nicholson, 2000 | | - | - | - | - | - | 40 | - | - | - |
| Tønnesen, 2000 | | - | - | - | - | - | - | 130 ²⁷ | - | - |
| Fitz and Bufalino, 2002 | On-road measurements | - | - | - | - | - | - | - | - | 64-124 |
| Johansson, 2002 | | - | - | - | - | - | - | 209 ²⁸ | - | - |
| Lough <i>et al.</i> , 2005 | Tunnel measurements, PM _{COARSE} ²⁹ | - | - | - | - | - | - | 58.5 | - | - |
| Areskoug <i>et al.</i> , 2005 | Roadside-background comparison | - | - | - | - | - | - | 200 ³⁰ | 23 | - |

²⁴ Calculation by the author.²⁵ Calculation by the author.²⁶ Total material lost.²⁷ With use of studded tyres.²⁸ With use of studded tyres.²⁹ Calculated by the author.³⁰ With use of studded tyres.

Clutch wear is also a potential emission source. The friction materials used in clutch linings are similar to those used in brake linings (*i.e.* a combination of mineral fibres, including asbestos in older vehicles, and binders). However, although some studies of the wear of asbestos-based clutches were conducted in the 1970s, there appears to be no recent information relating to the amount of material lost to the environment, or emissions of particulate matter, from this source. Clutch wear therefore represents an area of uncertainty, but the enclosed nature of the clutch mechanism probably means that much of the wear material is retained inside the housing. In any case, clutch wear as a source of particles warrants further investigation. Similarly, the quantification of PM arising from corrosion, engine wear and the abrasion of wheel bearings is hampered by a lack of research.

9.6 Models

Several different modelling procedures have been briefly reviewed. Some of these models treat sources individually, whereas others treat them collectively. Models which predict total non-exhaust PM emissions, and do not discriminate between the relative contributions of the different source paths, offer the simplest means of prediction. The most commonly used model of this type is the USEPA's AP-42 (although separate tyre and brake wear emission factors are also presented in AP-42). Even so, the AP-42 method has been subject to some criticism on scientific grounds, and there appear to have been few attempts to apply the AP-42 method to the UK. There are a number of other shortcomings associated with treating all non-exhaust particles in terms of a single source, including:

- It does not fit in with the current nomenclature for reporting to international conventions.
- It offers little insight into possible control policies and technologies.
- Different factors affect the different sources – sometimes act together, sometimes against (*e.g.* speed might increase resuspension, but decrease tyre wear).

Ultimately, the various sources need to be considered as independently as possible, and causal relationships need to be identified. The following have been identified as weak aspects of the current methodologies, and therefore areas for improvement:

- More detailed information is required on the relative effects of different tyre and road surface combinations.
- Emission factor values for road surface wear are highly uncertain. Again, additional experimental information is necessary to establish more precise value
- A significant weak area is the contribution from the resuspension of road dust. Models have typically been derived from studies conducted on dry days with dry road conditions, and few models allow for consideration of the road condition.
- Few models take into account vehicle operation, such as speed, gradient, cornering, and the number of braking events. Average brake wear does not adequately represent the wear factors on roads with significantly different driving conditions. As the amount worn is related to the total brake use, then the primary factor to be considered is the number of brake applications made and the force of the applications. The number of brake applications made is dependent upon the route geometry and the traffic conditions.

9.7 Model development

The next stage of the project will be the development of a refined method for estimating non-exhaust emissions of particulate matter from road transport in the UK, and thereby improve upon the estimates currently provided in the NAEI. It is likely that rather different methodologies will be required to model emissions arising from abrasion processes and those due to resuspension.

The use of fixed brake wear emission factors (in g/km) in models does not seem particularly logical, as the differences in the extent of braking for different traffic situations cannot be taken into account. Recent US studies have yielded limited emission factors in terms of g/stop, and for stops of differing severity. TRL holds a large database of driving patterns for different traffic situations. The possibility of combining these two types of information should be considered as part of this project.

The new approach will also include a calculation method for emissions due to resuspension. A number of alternative methods will be explored to this end. There are a limited number of high-quality published UK estimates of emission factors for total non-exhaust particles disaggregated according to heavy-duty and light-duty vehicles (*e.g.* AQEG, 2004). Available estimates from the published literature will be collated and used to calculate emission factors for resuspended particles. Using incremental $PM_{2.5-10}$ as a surrogate for non-exhaust particles, the best estimate of the total non-exhaust emission factor from light duty and heavy-duty vehicles will be derived. For comparison, estimates of resuspension will be made using the USEPA AP42 algorithm although the applicability of this to the UK (and even to North America) must be in doubt.

10 Conclusions and recommendations

This review has summarised the current understanding of road vehicle non-exhaust particulate matter, based on a large number of studies in various countries. The next phase of the project will be to evaluate the existing models for non-exhaust PM, and to develop improved modelling approaches for use in the NAEI. The conclusions drawn from this review, and some specific recommendations for this next phase, and some general recommendations for improving the overall understanding, are given below.

10.1 Conclusions

1. The most important non-exhaust sources of airborne PM are likely to be tyre wear, brake wear, road surface wear and resuspension. A large number of factors affect the emissions from each source, but these factors have not been fully investigated. Furthermore, the contribution of each source to airborne PM varies considerably, both temporally and spatially, and for the UK the relative importance of the sources is highly uncertain.
2. There is a general lack of consistency in the definitions, terminology and metrics used in the study and reporting of non-exhaust particulate matter. In particular, where emission factors for resuspension are reported, it is not always clear whether they include primary emissions due to abrasion, and a number of modelling methodologies consider abrasion sources but not resuspension. This often renders the incorporation of data into models, and comparisons of model predictions with earlier studies, rather difficult. Ultimately, the various sources need to be considered as independently as possible, and causal relationships need to be identified.
3. For 'normal' driving conditions, it seems that a value for total tyre wear of 100 mg/vkm would probably represent the right order of magnitude. For heavy-duty vehicles, the wear factor is likely to be an order of magnitude higher. More information is required to relate tyre wear (and PM emissions) to real-world driving patterns in a quantitative manner. The literature indicates that probably less than 10% of car tyre wear material is emitted as PM₁₀ under 'typical' driving conditions, but the proportion could be as high as 30%. Although it is commonly assumed that airborne tyre wear particles are mainly present as PM_{COARSE}, both fine and coarse particles are emitted directly to the atmosphere as a result of tyre wear.
4. Total brake wear for light-duty vehicles appears to be around 10 to 20 mg/vkm, and around 50 to 80 mg/vkm for heavy goods vehicles. Typically 50% of brake wear debris from LDVs escapes the vehicle and enters the atmosphere, and more than 80% of airborne brake wear particles can be classified as PM₁₀. A substantial amount of the PM₁₀ can also be present as PM_{2.5}. There is considerable uncertainty regarding the amount of material which is lost from the brake linings, and the amount which is lost from the disc or drum.
5. A wide range of road surface wear factors have been reported in a small number of studies, from less than 4 mg/vkm to more than 400 mg/vkm where the surface contains a high proportion of bitumen. However, in areas where there is extensive use of road sanding and studded tyres during the winter, the wear of the road surface is considerably higher - values of between 4 and 24 g/vkm have been reported. Very little information on the size distribution of road surface wear particles is available.
6. Resuspension is probably the single largest vehicle non-exhaust contributor to roadside PM₁₀, particularly where winter maintenance procedures are in place. Although some information on

the effects of winter maintenance is available for Nordic countries, the situation in the UK is rather different. In the UK, road de-icing involves the application of rock salt, but little information of the effects of this particular approach on resuspension has been reported. This specific subject requires further investigation.

7. Models for non-exhaust PM emissions are generally rather crude. The use of average brake wear emission factors (in g/km) in models does not seem particularly logical, as the differences in the extent of braking for different traffic situations cannot be taken into account. Such emission factors are likely to over-estimate brake emissions in areas of low brake usage, and underestimate emissions in areas of high brake use. More detailed methodologies for estimating brake wear emissions are required.
8. Emission factors for resuspension in Europe are variable, and most of the available information is derived from Nordic countries where the use of studded tyres presents a significant problem. There also appear to be some fundamental difference between non-exhaust processes in the US, and those in Europe. Total PM₁₀ emission factors in the US generally appear to be substantially higher than those in Europe. This may be related to the particularly dry and dusty conditions at the locations of US studies. US prediction models, such as the AP-42 are therefore unlikely to be appropriate to the UK.
9. The review gives no indication of appropriate mechanisms for controlling non-exhaust particles. The effects of different road vehicle/traffic controls upon coarse particle concentrations are not predictable in the absence of tested algorithms for calculating resuspended dust concentrations applicable to European conditions. In particular, the complex inter-relationships between vehicle/traffic operation, road surface dust silt loadings and resuspension have not been determined. Potential abatement options will be reviewed qualitatively (and quantitatively as far as possible) later in the project.

10.2 Recommendations for the next phase of the project

The following recommendations are made for the next phase of the project:

1. Further information on the non-exhaust PM models covered in this review should be obtained, along with information on any models which may have been overlooked. Any data relating to UK conditions which are required to run the models should be obtained.
2. The performance of the selected models should be evaluated for a range of typical UK conditions, using real-world data where possible.
3. Following this comparison, one or more models should be selected for further development. It is expected that rather different modelling approaches will be required for direct emission sources and resuspension. This development may therefore take a range of forms, but most attention will be paid to resuspension and a number of alternative methods will be explored. A refined method for predicting resuspension might take account of location and season, for example.
4. Recent US studies have yielded limited emission factors in terms of g/stop, and for stops of differing severity. TRL holds a large database of driving patterns for different traffic situations. The possibility of combining these two types of information to derive new emission factors should be considered as part of this project.

10.3 General recommendations

In order to improve the understanding of non-exhaust PM in the UK, a number of general recommendations are provided below.

1. *Tyre, brake and road surface materials in use.* Many different brake and tyre types are in use in the UK, but little information on the structure of the UK market, the performance of different materials, and the use of different types by location, has been reported in the scientific literature. Similarly, there is a need to understand the different road surfaces used in the UK by road type and geographic location. A survey of materials in use would assist in the design of future experimental work and modelling approaches.
2. *Component wear factors.* Although some wear factors for tyres and brake linings are available in the literature, there is little information specifically for the UK, and the relationships between wear and real-world vehicle operation are not well understood. Furthermore, the relative wear factors for brake linings and brake rotors are not well documented. Further information of this type is required for modelling purposes.
3. *Source characterisation.* The compositions of the various tyre and brake lining formulations used in the UK have not been reported in detail. An extensive examination of the composition of the tyres and brakes available on the UK market is required. It would also be useful to examine the extent to which tyre tread and brake lining material are altered during use, and to analyse the tyres, the brake lining, and the brake dust from same in-service vehicles at regular intervals.
4. *Laboratory-based experiments.* Further sampling of tyre, brake and road surface wear particles is required under controlled laboratory conditions, and using analytical equipment which cannot easily be deployed in the field.
5. *Real-world measurements using instrumented vehicles.* The measurement of non-exhaust particles could be conducted *in situ* under a range of real-world vehicle operating conditions. Some experiments of this type have already been conducted in the United States.
6. *Further receptor modelling studies.* Receptor modelling studies at a varied range of locations, and based on new source profiles, would contribute significantly to the understanding of non-exhaust PM in the UK. Comparisons between receptor modelling studies inside and outside of road tunnels could assist in the understanding of resuspension processes.

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Appendix A. USEPA AP-42 model

Tyre wear

The tyre wear emission factor per vehicle is calculated as:

$$EF_{TYRE(v)} = 0.002 * F_{TYRE} * IVEHWL(v) \quad (\text{Equation A1})$$

Where:

- $EF_{TYRE(v)}$ = Tyre wear emission factor for a vehicle in class v (g/mile)
0.002 = Emission rate of airborne particulates from tyre wear for LDVs
 F_{TYRE} = The fraction of particles less than or equal to the particle size cut-off
 $IVEHWL(v)$ = The average number of wheels on a vehicle of class v (Table A.1)

Table A1: Number of wheels per vehicle

| Vehicle category (note: US) | Average number of wheels |
|-----------------------------|--------------------------|
| LDV | 4 |
| Small HGV | 6 |
| Medium HGV | 6 |
| Large HGV | 18 ³¹ |
| Bus | 4 |
| Motorcycle | 2 |

The particle size cut-off is the maximum aerodynamic diameter of the particles in the emission factor. For tyre wear (all vehicles), the value of F_{TYRE} for a cut-off of 10 μm is 1.0, and 0.01 for a cut-off of 0.1 μm .

Brake wear

The brake wear emission factor, which is applied to all vehicle categories, is calculated as:

$$EF_{BRAKE} = 0.0128 * F_{BRAKE} \quad (\text{Equation A2})$$

Where:

- EF_{BRAKE} = Brake wear emission factor (g/mile)
0.0128 = Emission factor for airborne particles
 F_{BRAKE} = The fraction of particles less than or equal to the particle size cut-off (Table A2).

³¹ A more typical maximum value for Europe would be 16.

Table A2: Fraction of particles for each cut-off

| Particle size cut-off (μm) | F_{BRAKE} |
|---|-------------|
| 10 | 0.98 |
| 7 | 0.90 |
| 4.7 | 0.82 |
| 1.1 | 0.16 |
| 0.43 | 0.09 |

Sanders *et al.* (2003) suggest that the following values for F_{BRAKE} are more appropriate for urban driving: 0.8 at 10 μm , 0.6 at 7 μm , 0.35 at 4.7 μm , 0.02 at 1.1 μm , and <0.01 at 0.43 μm .

'Fugitive dust' (resuspension)

Re-entrained road dust emission factors are calculated for both unpaved and paved roads include all PM sources (exhaust, tyre wear, brake wear, road surface wear and resuspension). The models did not formerly apply to days with rain, although in a memorandum the USEPA (2003) indicated that a 25% reduction in emissions should be assumed for such days.

For unpaved roads, the following equation is used:

$$EF_{UNPAVED} = F_{UNPAVED} \times 5.9 \times \frac{sL}{12} \times \frac{V}{30} \times \left[\frac{W}{3} \right]^{0.7} \times \left[\frac{N_w}{4} \right]^{0.5} \times \frac{(365 - d)}{365} \times 453.592 \quad (\text{Equation A3})$$

Where:

| | | |
|----------------|---|--|
| $EF_{UNPAVED}$ | = | Fleet average unpaved road dust emission factor (g/mile) |
| $F_{UNPAVED}$ | = | The fraction of particles less than or equal to the particle size cut-off (Table A3) |
| sL | = | Silt content (particles <75 μm diameter) of the surface material (%) |
| V | = | Fleet average vehicle speed (mph) |
| W | = | Fleet average vehicle weight (imperial tons) |
| N_w | = | Fleet average number of wheels per vehicle |
| d | = | Average number of days per year with more than 0.01 inches of rain |

Table A3: Fraction of particles for each cut-off

| Particle size cut-off (μm) | $F_{UNPAVED}$ |
|---|---------------|
| 10 | 0.36 |
| 5 | 0.20 |
| 2.5 | 0.095 |

For paved roads, the following equation is used:

$$EF_{PAVED} = e_{PAVED} \times \left[\frac{sL}{2} \right]^{0.65} \times \left[\frac{W}{3} \right]^{1.5} \quad (\text{Equation A4})$$

Where:

| | | |
|--------------------------------|---|--|
| EF_{PAVED} | = | Fleet average paved road dust emission factor (g/mile) |
| e_{PAVED} | = | The base emission factor for the particle size cut-off |
| sL | = | The road surface silt loading (g/m ²) |
| W | = | Fleet average vehicle weight (tons) |

For size cut-off of 10 μm , e_{PAVED} is 7.3 g/mile, and for a cut off of 2.5 μm e_{PAVED} is 3.3 g/mile.

Appendix B. Modified AP-42 for use in Germany

An alternative version of the USEPA model for paved roads was developed by Rauterberg-Wulff (2000) for use in Berlin:

$$EF_{PAVED} = c \times sL^{0.65} \times W^{1.5} \quad (\text{Equation B1})$$

Where :

$$\begin{aligned} EF_{PAVED} &= \text{Fleet average paved road dust emission factor (g/km)} \\ c &= \text{Constant (0.26 for PM}_{2.5}\text{, 0.56 for PM}_{10}\text{, 0.68 for PM}_{15}\text{, 2.92 for PM}_{30}\text{)} \end{aligned}$$

The further modification to the AP-42 method proposed by Gamez *et al.* (2001) involves the separation of the exhaust and non-exhaust contributions:

$$E_{total} = e_{exhaust} + e_{non-exhaust} \quad (\text{Equation B2})$$

Where $e_{non-exhaust}$ includes tyre, brake and road surface wear, as well as resuspension :

$$E_{non-exhaust} = e_{road} + e_{tyre+brake} + e_{resuspension} \quad (\text{Equation B3})$$

Combining this with the AP-42 method yielded the following equation for PM₁₀:

$$e_{non-exhaust} = a \times j \times sL^{0.52} \times W^{2.14} \times \left[\frac{1}{0.85} (1 - 0.5r) \right] - (e_{exhaust})_{2000} \quad (\text{Equation B4})$$

Where:

$$\begin{aligned} a &= \text{Correction factor for use in Germany} \\ k &= \text{Basic USEPA emission factor for PM}_{10} \\ sL &= \text{Silt loading of the road (g/m}^2\text{)} \\ W &= \text{Mean vehicle weight (t)} \\ r &= \text{Fraction of rainy days during the year} \\ (e_{exhaust})_{2000} &= \text{Fleet-weighted exhaust emission for the year 2000 (formula requires} \\ &\quad \text{modification for use in other years)} \end{aligned}$$

The values for a , S and W shown in Table B1 were proposed by Düring *et al.* (2002).

By 2004, the modified AP-42 method had been abandoned in favour of allocating emission factors to 'traffic situations' (Lohmeyer *et al.*, 2004) – see Appendix B. Examples of these emission factors are

given in the Table B2. The traffic situations are taken from the Handbook of emission factors (UBA, 2004).

Table B1: values of a , S and W used in the modified AP-42 formula

| Location | a | S | W |
|----------------------|---|---|--------------------------------------|
| City roads | 'Good' road surfaces: 0.8 'Bad' road surfaces: 2.0 | 'Good' road surfaces: 0.2 g/m ² 'Bad' road surfaces: 0.4 g/m ² | Car: 1.1t LGV: 1.9t HGV: 9.0t |
| Roads outside cities | 'Good' road surfaces: 0.8 'Bad' road surfaces: 2.0 | 0.1 g/m ² | Car: 1.2t LGV: 2.0t HGV: 11.0t |
| Motorways | 'Good' road surfaces: 0.8 'Bad' road surfaces: 2.0 | 0.1 g/m ² | Car: 1.3t LGV: 2.1t HGV: 13.0t |

Table B2: Emission factors by traffic situation

| Traffic situation | Speed limit (km/h) | Percentage of constant-speed driving (%) | Percentage no traffic flow (%) | Emission factor (mg/vkm) | |
|----------------------------------|--------------------|--|--------------------------------|--------------------------|-----|
| | | | | Cars or LGV | HGV |
| Motorway>120 | None | | | 22 | 200 |
| Motorway120 | 120 | | | 22 | 200 |
| Motorway100 | 100 | | | 22 | 200 |
| Motorway80 | 80 | | | 22 | 200 |
| Motorway60 | 60 | | | 22 | 200 |
| Motorway stop-and-go | None | | | 22 | 200 |
| Rural1 | 100 | 60 | 1 | 22 | 200 |
| Rural2 | 100 | 53 | 1 | 22 | 200 |
| Rural3 | 100 | 28 | 1 | 22 | 200 |
| City>50 | 60 | 46 | 1 | 22 | 200 |
| Tunnel, motorway100 | 100 | | | 10 | 200 |
| Tunnel, motorway 80 | 80 | | | 10 | 200 |
| Tunnel, motorway 60 | 60 | | | 10 | 200 |
| Tunnel, city | 60 | 46 | 1 | 10 | 200 |
| HVS1 | 50 | 46 | 1 | 22 | 200 |
| HVS2 | 50 | 52 | 1 | 30 | 300 |
| HVS3 | 50 | 44 | 7 | 40 | 380 |
| LSA [†] , minimum delay | 50 | 44 | 7 | 40 | 380 |
| HVS4 | 50 | 37 | 14 | 50 | 450 |
| LSA, medium delay | 50 | 32 | 20 | 60 | 600 |
| LSA, heavy delay | 50 | 28 | 26 | 90 | 800 |
| Inner city1 | 50 | 23 | 33 | 90 | 800 |
| Inner city2 | 50 | 3 | 5 | 90 | 800 |

[†] LSA = Urban main road with traffic lights

Appendix C. SMHI model

The model developed by the Swedish Environmental and Health Protection Administration (SMHI) for resuspension, proposed by Bringfelt *et al.* (1997) (cited in Gustafsson, 2003), is:

$$e_f^{tot} = e_f^{direct} + F_{qe} (e_f^{dust}) \quad (\text{Equation C1})$$

Where: e_f^{tot} = Total PM emission factor
 e_f^{direct} = Emission factor for exhaust particles
 e_f^{dust} = Emission factor for resuspended particles
 F_{qe} = Reduction factor

e_f^{dust} is given by (Sehmel, 1984):

$$e_f^{dust} = \left[\frac{\%LDV}{100} \times (e_f^{dust})_{LDV} \times \left(\frac{V}{V_0} \right)^2 \right] + \left[\frac{\%HDV}{100} \times (e_f^{dust})_{HDV} \times \left(\frac{V}{V_0} \right)^{0.5} \right] \quad (\text{Equation C2})$$

Where: $e_f^{dust}(LDV)$ = Reference emission factor for light-duty vehicles
 $e_f^{dust}(HDV)$ = Reference emission factor for heavy-duty vehicles
 $\%LDV$ = Percentage of light-duty vehicles
 $\%HDV$ = Percentage of heavy-duty vehicles
 V = Vehicle speed in km/h
 V_0 = Reference vehicle speed in km/h

In this expression resuspension for cars is proportional to the square of the vehicle speed (v in km/h) and for trucks to the square root of the speed. At 110 km/h both cars and trucks emit 6.7 mg/vkm. The form of the equation means that passenger cars have a large influence on emissions at speeds greater than 110 km/h, and heavy goods vehicles have a larger influence at lower speeds.

F_{qe} depends on factors such as the amount of moisture in the dust and on the road surface, meteorological parameters, and the level of winter maintenance operations.

As with the USEPA AP-42 method, the equation relates to emissions of particles from all non-exhaust sources.

Appendix D. VLUFT model

There is little documentation in English describing the VLUFT model. This form of the model was presented by Gustafsson (2003):

$$Q_{10-2.5} = QR_{2.5} \cdot (aTT+b) \cdot (V_T/V_R)^2 \cdot RP \cdot RW \quad (\text{Equation D1})$$

Where:

| | | |
|--------------|---|---|
| $Q_{10-2.5}$ | = | Emission factor for coarse particles |
| $QR_{2.5}$ | = | Emission factor for fine particles |
| aTT | = | Heavy vehicles as % of traffic flow |
| V_T | = | Mean speed |
| V_R | = | Reference speed |
| RP | = | Reduction factor relating to use of studded tyres |
| RW | = | Reduction factor for road surface humidity |

Appendix E. EMEP/CORINAIR method

E.1 Simple methodology for PM₁₀

Equation E1 is presented to calculate PM₁₀ emissions from brake wear and tyre wear emissions from a given vehicle category, spatial area and time period by selecting appropriate values for the fleet size and the activity rate (mileage). Total traffic-generated emissions can be estimated by summing the emissions from individual vehicle classes.

$$E_{s,j} = N_j \times M_j \times e_{s,j} \quad (\text{Equation E1})$$

Where:

- $E_{s,j}$ = Total PM₁₀ Emissions (g) for the defined time period and spatial boundary
- N_j = Number of vehicles in defined class within the defined spatial boundary
- M_j = Average mileage driven (km) per vehicle in defined class during the defined time period
- $e_{s,j}$ = Mass emission factor (g/km)
- s = Non-exhaust emission source (tyre, brake, road surface)
- j = vehicle category (two-wheel vehicle, passenger car, LGV, HDV)

The user of this simple methodology need only determine activity rates in the form of the vehicle population (by class) and mileage driven per vehicle (by class) for the requested temporal and spatial resolution. The relevant emission factors for use in Equation (1) are given in Table E1.

Table E1: Non-exhaust PM₁₀ emission factors to be used with the simple methodology and comparison with aggregated exhaust emission factors

| Vehicle class (j) | Particle source and emission factor (g/km) | | |
|-----------------------|--|------------|-------------------|
| | Tyre wear | Brake wear | Road surface wear |
| Two-wheel vehicles | 0.0028 | 0.0037 | 0.0030 |
| Cars | 0.0064 | 0.0073 | 0.0075 |
| LGVs | 0.0101 | 0.0115 | 0.0075 |
| HDVs | 0.0270 | 0.0320 | 0.0380 |

E.2 Detailed methodology

Tyre wear particle emissions

In order to estimate particle emissions from tyre wear, Equation E2 can be used. This equation refers to a single vehicle category for a defined temporal and spatial resolution. Also, different particle size classes are considered.

$$E_{TYRE,i,j} = N_j \times M_j \times e_{TYRE,TSP,j} \times f_{TYRE,i} \times S_{T(v)} \quad (\text{Equation E2})$$

Where:

| | | |
|------------------|---|--|
| $E_{TYRE,ij}$ | = | Total emissions (g) for the defined time period and spatial boundary |
| N_j | = | Number of vehicles in the defined class within the defined spatial boundary |
| M_j | = | Mileage driven (km) by vehicles in the defined class during the defined time period |
| $e_{TYRE,TSP,j}$ | = | TSP mass emission factor from tyre wear (g/km) |
| $f_{TYRE,i}$ | = | Mass fraction of tyre-wear TSP that can be attributed to particle size class i |
| $S_{T(V)}$ | = | Tyre-wear correction factor for a mean vehicle travelling speed V |
| i | = | Size fraction (TSP, PM ₁₀ , PM _{2.5} , PM ₁ and PM _{0.1}) |
| j | = | vehicle category (two-wheel vehicle, passenger car, LGV, HDV) |

TSP emission factors for different vehicle classes are given in Table E2. All emission factors are based on available experimental data. It should be noted that the TSP emission rates do not assume that all tyre wear material is transformed into suspended particulate, as a large fraction of tyre rubber may be produced as dustfall particles or larger shreds (*e.g.* under heavy braking). A value of 0.6 has been selected as the PM₁₀/TSP ratio for tyre wear in order to derived TSP values where PM₁₀ emission rates are available in the literature.

Table E2: TSP emission factors from tyre wear

| Vehicle class (j) | Emission factor (g/km) |
|--------------------|------------------------|
| | $e_{TYRE,TSP}$ |
| Two-wheel vehicles | 0.0046 |
| Cars | 0.0107 |
| LGV | 0.0169 |
| HDV | Equation E3 |

For the heavy-duty vehicle case, emission factor needs to take vehicle size into account. This is introduced by the equation:

$$(E_{TYRE})_{HDV} = \frac{N_{axle}}{2} \cdot LCF_{TYRE} \cdot (E_{TYRE})_{PC} \quad (\text{Equation E3})$$

Where:

| | | |
|-------------------|---|----------------------------------|
| N_{axle} | = | Number of truck axles |
| LCF_{TYRE} | = | Load correction factor |
| $(E_{TYRE})_{PC}$ | = | The TSP emission factor for cars |

For heavy-duty vehicles, the number of axles is a parameter which can be used to differentiate size. An additional parameter is a load correction factor, which accounts for the load carried by the truck or bus. The load correction factor can be estimated on the basis of Equation E4 which has been derived by linear regression on experimental data:

$$LCF_T = 1.41 + (1.38 \times LF) \quad (\text{Equation E4})$$

Where LF is the load factor for the truck, ranging from 0 for an empty truck to 1 for a fully laden one. The same equations can be used for urban busses and coaches.

Typical size profiles for TSP emitted by tyre wear have been obtained by combining information from the literature. Based on this information, the mass fraction of TSP in the different particle size classes is shown in Table E3.

Table E3: Size distribution of tyre wear emitted particles

| Particle size class (i) | Mass fraction (f_{TYRE}) of TSP |
|-------------------------|-------------------------------------|
| TSP | 1.000 |
| PM ₁₀ | 0.600 |
| PM _{2.5} | 0.420 |
| PM ₁ | 0.060 |
| PM _{0.1} | 0.048 |

A speed correction is required to account for the different wear factor of the tyre depending on the vehicle speed. Figure E1 shows the speed correction, based on the findings of Luhana *et al.* (2002). It should be noted that, as in the case of exhaust emission factors, vehicle speed corresponds to mean trip speed and not constant travelling speed. There is a decreasing pattern of emissions with increasing speed. This is in contrast to the usual perception that airborne particles increase in the wake of a vehicle as speed increases, because Figure E1 corresponds to primary particle emissions from the tyre and not resuspended dust. Tyre wear decreases as mean trip speed increases, because braking and cornering are more frequent in urban driving than in motorway driving. Note that $S_{T(V)} = 1$ when the mean trip speed is 80 km/h, and stabilizes below 40 km/h and above 90 km/h due to the absence of any experimental data. Also, although the proposed equation has been obtained from measurements on passenger cars, it is to be used for all vehicle categories.

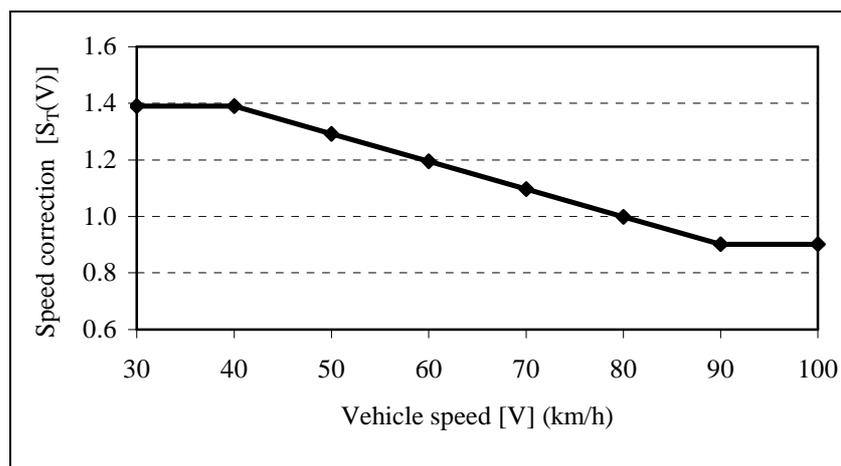


Figure E1: Speed correction factor ($S_{T(V)}$) for tyre wear particle emissions

Brake wear particle emissions

Similarly to tyre wear, brake wear emissions can be calculated by:

$$E_{BRAKE,ij} = N_j \times M_j \times e_{BRAKE,TSPj} \times f_{BRAKEi} \times S_{B(V)} \quad (\text{Equation E5})$$

Where the nomenclature is similar to that of Equation E2.

TSP emission factors for brake wear particles are given in Table E4, together with the range and a quality code for the emission factor.

Table E4: TSP emission factors from brake wear

| Vehicle class (j) | Emission factor (g/km) |
|--------------------|------------------------|
| | $e_{BRAKE,TSP}$ |
| Two-wheel vehicles | 0.0037 |
| Cars | 0.0075 |
| LGV | 0.0117 |
| HDV | Equation E6 |

The heavy-duty emission factor is calculated by adjusting the passenger car emission factor to fit heavy-duty vehicle experimental data:

$$(E_{BRAKE})_{HDV} = 3.13 \cdot LCF_{BRAKE} \cdot (E_{BRAKE})_{PC} \quad (\text{Equation E6})$$

In Equation E6, 3.13 is an empirical factor derived from experimental data and LCF_B is defined in a similar way to LCF_{TYRE} and can be determined again by linear regression on experimental data by the equation:

$$LCF_{BRAKE} = 1 + 0.79 \times LF \quad (\text{Equation E7})$$

LF again has the value of 0 for an empty truck and 1 for a fully laden one. Equations E6 and E7 are also used for urban buses and coaches. The mass fraction of TSP in the different particle size classes is shown in Table E5.

Table E5: Size distribution of brake wear emitted particles

| Particle size class (i) | Mass fraction (f_{BRAKE}) of TSP |
|-------------------------|--------------------------------------|
| TSP | 1.000 |
| PM ₁₀ | 0.980 |
| PM _{2.5} | 0.390 |
| PM ₁ | 0.100 |
| PM _{0.1} | 0.080 |

The speed correction factor for the case of brake wear is given in Figure E2.

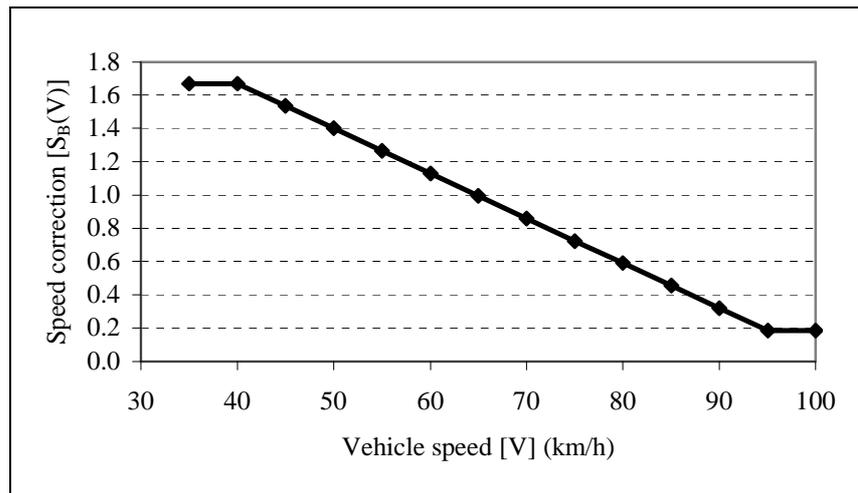


Figure E2: Speed correction factor ($S_{B(V)}$) for brake wear particle emissions

In this case, the speed correction is normalised for a speed of 65 km/h, and the slope is generally larger than for tyre wear because brake wear is negligible at high motorway speeds when limited braking occurs. Again, although the proposed equation has been obtained from measurements on passenger cars, it is to be used for all vehicle categories.

Road surface wear emissions

There is very little information on airborne emission rates from asphalt wear, and therefore the quality of the detailed methodology does not differ from the quality of the simple one. The detailed methodology only provides a mass-weighted size classification of road surface wear particles based on the work of Lükewille *et al.* (2002), according to the equation:

$$E_{ROAD,ij} = N_j \times M_j \times (e_{ROAD})_j \times f_{ROAD,i} \quad (\text{Equation E8})$$

Where the nomenclature is similar to Equation E2, but **ROAD** corresponds to road surface wear. The TSP emission factors for particles of road surface wear are presented in Table E1. The mass fraction of TSP in the different particle size classes is shown in Table E6.

Table E6: Size distribution of road surface wear emitted particles

| Particle size class (i) | Mass fraction (f_{ROAD}) of TSP |
|-------------------------|-------------------------------------|
| TSP | 1.00 |
| PM ₁₀ | 0.50 |
| PM _{2.5} | 0.27 |

Due to the lack of appropriate experimental data, no emission factors are included for road surface wear associated with the use of studded tyres, although it is recognised that in some countries this may be an important particle source. Preliminary values for road surface wear TSP emissions are shown in Table E7. These TSP values should correspond to primary particles from road surface wear but they are based on limited information and are highly uncertain.

Table E7: TSP emission factors from road surface wear

| Vehicle class (j) | Emission factor (g/km) <i>e_{ROAD,TSP}</i> |
|--------------------|---|
| Two-wheel vehicles | 0.0060 |
| Cars | 0.0150 |
| LGV | 0.0150 |
| HDV | 0.0760 |

Appendix F. CEPMEIP database

The emission factors for tyre, brake and road surface wear used in the CEPMEIP database (Berdowski *et al.*, 2001) are shown in Table F1. For tyre wear it is assumed that all PM₁₀ is in the coarse mode (PM_{10-2.5}), but most of the airborne mass is larger than PM₁₀. For brake wear, it is assumed that all particles are emitted in the PM_{2.5} fraction. For road surface wear, it assumed that no particles are emitted in the PM_{2.5} fraction, and that 5% of TSP is emitted as PM₁₀ (3.6% for HDVs).

Table F1: CEPMEIP emission factors for tyre, brake and road surface wear

| Source | Vehicle category | Emission factor (mg/km) | | |
|-------------------|---------------------|-------------------------|------------------|-------------------|
| | | TSP | PM ₁₀ | PM _{2.5} |
| Tyre wear | Motorcycles | 34.5 | 1.72 | 0 |
| | Passenger cars | 69 | 3.45 | 0 |
| | Light-duty vehicles | 90 | 4.5 | 0 |
| | Heavy-duty vehicles | 371.25 | 18.56 | 0 |
| Brake wear | Motorcycles | 3 | 3 | 3 |
| | Passenger cars | 6 | 6 | 6 |
| | Light-duty vehicles | 7.5 | 7.5 | 7.5 |
| | Heavy-duty vehicles | 32.25 | 32.25 | 32.25 |
| Road surface wear | Motorcycles | 73 | 3.65 | 0 |
| | Passenger cars | 145 | 7.25 | 0 |
| | Light-duty vehicles | 190 | 9.5 | 0 |
| | Heavy-duty vehicles | 738 | 26.9 | 0 |

Appendix G. RAINS database

The emission factors in the RAINS database are given below.

Tyre wear

The model assumes that tyre wear $PM_{2.5}$ is 5% of PM_{10} .

Table G1: RAINS emission factors for tyre wear

| | Emission factor (g/km) | | |
|------------------|------------------------|-----------|------------|
| | TSP | PM_{10} | $PM_{2.5}$ |
| LDV | 0.0661 | 0.0065 | 0.0003 |
| 2-wheel vehicles | 0.0282 | 0.0032 | 0.0001 |
| HDV | 0.4208 | 0.0400 | 0.002 |

Brake lining wear

Emission factors for motorcycles are assumed to be 15% of those for cars.

Table G2: RAINS emission factors for brake wear

| | Emission factor (g/km) | | |
|------------------|------------------------|-----------|------------|
| | TSP | PM_{10} | $PM_{2.5}$ |
| LDV | 0.0044 | 0.0036 | 0.0022 |
| 2-wheel vehicles | 0.0006 | 0.0005 | 0.0003 |
| HDV | 0.0275 | 0.0228 | 0.0071 |

Road surface wear

Table G3: RAINS emission factors for road surface wear

| | Emission factor (g/km) | | |
|------------------|------------------------|-----------|------------|
| | TSP | PM_{10} | $PM_{2.5}$ |
| LDV | 0.015 | 0.0075 | 0.0042 |
| 2-wheel vehicles | 0.006 | 0.003 | 0.0016 |
| HDV | 0.076 | 0.038 | 0.0209 |