NON-EXHAUST PARTICLES IN THE ROAD ENVIRONMENT – A LITERATURE REVIEW

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1. Abstract

Non-exhaust particles in the road environment originate from wear of asphalt road pavement, mainly caused by the use of studded tyres, and corrosion of vehicle components such as tyres and brakes. Other sources are road maintenance, road equipment and particles originating in the road surroundings. This literature survey aims at giving an overview of the current knowledge about airborne particles from these different sources in the context of characteristics and emissions as well as health and environmental effects.

2. Introduction

Particles related to road traffic have over the last few decades become an important issue among scientists, the reason being the often-recurrent indications of relationships between airborne particles and effects on health and the environment. A recently published study shows that particles related to road traffic are responsible for about 3 % of the total mortality in France, Austria and Switzerland (Kunzli et al., 2000).

Due to their small size and their ability to adsorb toxic components in the exhaust gas, exhaust particles have been the main focus for research. In the road environment though, particles from several other sources occur. Particles generated through wear of vehicles and pavement, added through maintenance measures or transported to the road environment from the surroundings, make up a large fraction of road dust. All these particles accumulated on the road surface can be re-suspended by the action of passing vehicles (fig.1.).

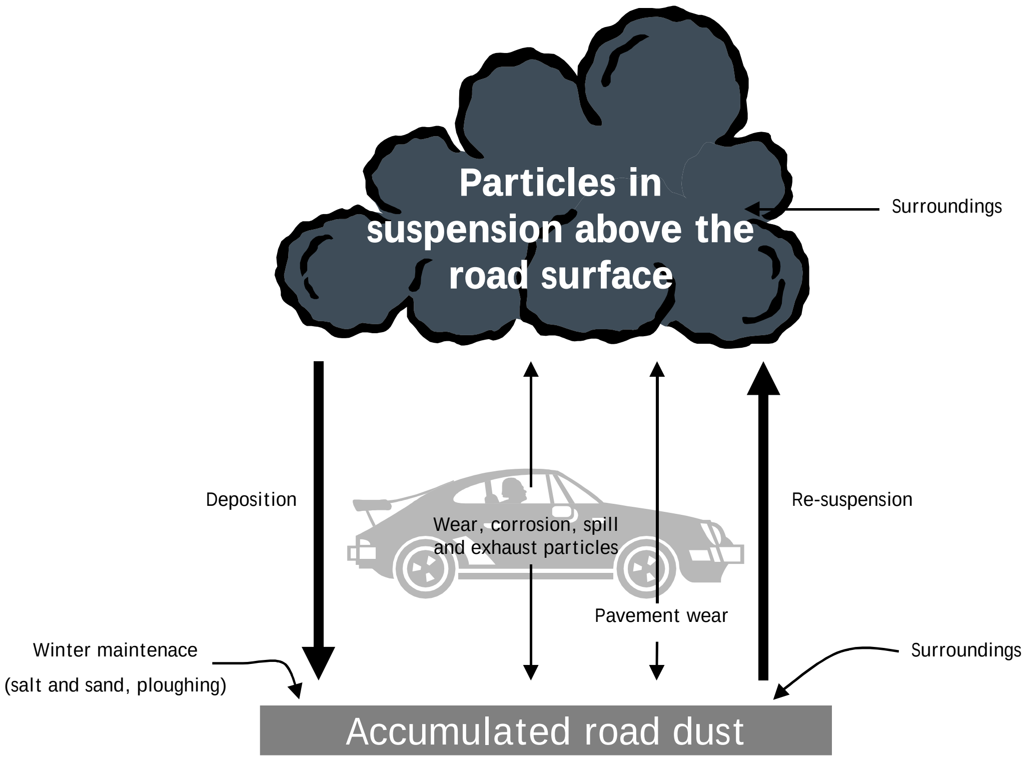

Figure 1. Schematic illustration of sources and fluxes of particles in the road environment.

The concentration of particles in the air is usually measured with respect to mass or number per volume of air. Mass distribution, chemistry and physical properties are also important aspects when describing the characteristics of particles. When the importance of particle size became apparent a standardised measure for inhalable particles was constructed called PM_{10}. Roughly, this is the fraction of particles smaller than 10 µm in diameter. This standard is commonly used throughout the world and also in the relatively few Swedish cities that measure particles on a regular basis. As interest has shifted towards even smaller particles, the standard has been supplemented with $PM_{2.5}$, to measure particles smaller than 2.5 µm.

This literature survey aims to summarise current knowledge of sources, emissions and health and environmental effects of non-exhaust particles in the road environment. These are generally larger than exhaust particles, but a significant fraction occur within PM_{10} and also $PM_{2.5}$.

3. Particle sources

Wear particles in the road environment have mainly three sources; tyres, brakes and pavement, but also incorporate wear from other movable parts in vehicles. For Swedish conditions, pavement wear during the winter months, when studded tyres are used, is the main source of wear particles.

3.1 Tyres

Depending on quality demands and area of use, tyres consist of a variety of mixtures of rubber polymers. Latex, as well as synthetic rubber, such as e.g. isoprene rubber, is used to obtain the desired properties of elasticity, heat resistance and friction (Ahlbom and Duus, 1994). More latex is used in bus and truck tyres, due to higher friction demands. Rubber mixtures vary greatly between manufacturers and also between summer, winter and studded tyres, making it difficult to generalise the chemistry of tyre wear particles (Johansson, 2000).

Tyres also consist of a large number of chemicals added during manufacture. Reinforcing agents, vulcanisers, accelerators, activators, colour pigments, softeners, dispersing agents, anti-oxidants, anti-ozonants, stabilisers etc are used during production (Rogge et al., 1993). In literature, tyres are often mentioned as a main source of zinc in the road environment (Rogge et al., 1993). This is due to the relatively large amount of zinc oxide used as an activator to make the accelerators more efficient during manufacture.

Also, PAH (polycyclic aromatic hydrocarbons) occurs in relatively large amounts in tyres. Very different information on the concentration of substituted and non-substituted PAH in tyres is reported in literature. (Ahlbom and Duus, 1994) report 7000 µg g^{-1}, while in a Swedish survey, (Lindgren, 1998) has measured between 33 and 93 µg g^{-1} in three ordinary tyres. Measured in wear particles, (Takada et al., 1991) reports 31-71 µg g^{-1}.

The size distribution of tyre wear particles is difficult to generalise. (Kobriger and Geinopolos, 1984) and (Noll et al., 1987) claims a mean diameter of 20-25 µm, while (Kumata et al., 1997) reports a bimodal distribution with peaks at 0,4-0,5 µm and 5-7 µm. These results imply that a considerable proportion of the wear particles are airborne and inhalable.

Studded tyres, commonly used in Sweden during winter, are made of the same types of rubber mixtures as other tyres. The studs were initially made of steel, but due to the large wear they caused to road pavements, they are nowadays made of light metals or plastic. The number of studs and the stud force have also been decreased (Jacobson and Hornvall, 1999a).

3.2 Brake linings

Similarly to tyres, brake linings are very heterogeneous in composition and manufacturer-dependent. The friction materials contain binders, fillers, fibres of glass, plastic, steel, organic or inorganic material or metals. Metals are also used as heat conductors (Rogge et al., 1993). Table X shows the metal content of the most common brake linings for cars making up approx. 60% of the Swedish car fleet (Westerlund, 1998).

An important feature of brake lining particles is their small size. Using a brake dynamometer (Garg et al., 2000) showed that 63% of the wear particles were smaller than 2.5 µm ($PM_{2.5}$), i.e. respirable.

3.3 Pavement

The most important wear associated with the use of studded tyres is pavement wear. This depends on the weight, number and composition of the studs, the flow, composition and speed of the traffic, climatic conditions, road geometry, pavement composition to mention a few factors. The percentage and quality of the stone material and the properties of the asphalt itself are of great importance. In Sweden, high quality pavements with a high percentage of very hard porphyry and quartzite (about 95% in the surface) have gradually replaced pavements with less resistant local rock material on the heavily trafficked roads (Jacobson and Hornvall, 1999b).

The high cost of maintenance related to pavement wear has caused many countries to prohibit the use of studded tyres. In Japan and Norway, regulations have also been based upon the health aspects of the road dust. In Japan studs are prohibited. Before the restrictions, the concentration of airborne dust could vary between 30 µg m^{-3} in summer and 400 µg m^{-3} in winter (Takishima et al., 1987). The current restrictions are questioned though, since the climate of Hokkaido involves very icy roads, which has affected the number of traffic accidents negatively (Norem, 1998). In Norway, the “Road grip project” (Krokeborg, 1997; Larssen and Haugsbakk, 1996) has so far resulted in stud restrictions in Oslo.

The particles formed through pavement wear reflect the asphalt composition. In Sweden about 95% is rock material and 5% bitumen. This oil product contains asphaltenes (5-25%), saturates (5-20%), cyclic compounds (45-60%) and resins (15-25%) (Gonzàles Arrojo, 2000). The PAH content is very small and not considered a main source of PAH in the road environment (Lindgren, 1998). The size of the pavement wear particles varies, but is generally regarded as being fairly large. According to (Bækken, 1993) only about 2% are smaller than 36 µm. Japanese studies on the other hand state a size interval of about 5-50 µm (Amemiya et al., 1984). This particle size depends mainly upon the properties of the rock material in the pavement, so a large variation is to be expected between countries and regions.

3.4 Salt and sand

About 200,000 – 400,000 tons of de-icing salt was added to Swedish roads annually between 1991/92 and 1995/96. The salt is re-suspended as wet or dry aerosol by passing vehicles and can be transported hundreds of meters from the road. The effects of these salt droplets and particles have been described exhaustively in literature (Blomqvist, 1999), but information about their characteristics is unfortunately very rare.

3.5 The road as source for PM_{10} and $PM_{2.5}$

Many studies make no attempt to distinguish between particle sources, but concentrate on describing concentrations of the PM_{10} and/or $PM_{2.5}$ fractions.

In Norway, before the regulation of studded tyre use, (Larssen, 1987) measured PM_{10} concentration to 55 µg m^{-3} in dry conditions and 10 µg m^{-3} in wet and therefore concluded that road wear particles contributed 45 µg m^{-3} in dry conditions. In a later study, (Larssen and Haugsbakk, 1996) found that in dry conditions, the road dust depot does not grow due to a balance between produced and re-suspended particles. The contribution of road dust to mean annual PM_{10} and $PM_{2.5}$ concentrations in Norwegian cities has been shown to be high along roads and streets. The contribution to highest mean diurnal concentration is significant both in city centres and along roads and streets (tab. X) (Larssen and Hagen, 1997).

Results from countries where studded tyres are not allowed shows that in spite of this, road dust often contributes a substantial proportion of PM_{10} or $PM_{2.5}$. (Schauer and Cass, 2000) determined the concentration of road pavement dust to 0.5-1 g m^{-3} during severe air pollution episodes in California, USA. Also in the USA, (Noll et al., 1987) found that tyre rubber particles made up approximately 35%, limestone 54% and silicates 10%, both important road building materials, of road dust collected in business districts in Argonne and Chicago. (Chow et al., 1996) estimated that the road dust contributed to 25-27% of urban PM_{10} concentrations as compared to a 30-42% contribution from exhaust particles.

4. Emissions

4.1 Non-exhaust Particles

Emission factors for tyres in literature range from 0.006 to 0.36 g km^{-1}. Using a road simulator (Rogge et al., 1993) estimated the wear to 0.006-0.09 g km^{-1} and tyre. Swedish investigations in co-operation with the police authorities, a local traffic company and tyre manufacturers calculated the emissions to 0.09 g km^{-1} for a car and 1.0 g km^{-1} for a bus (Table 1) (Lindström and Rossipal, 1987).

Table 1. Emissions from tyres *(Lindström and Rossipal, 1987).*

Component	*Car*	*Bus*
	g km^{-1}	*g pbkm^{-1}*
Rubber	0,05	0,7
Carbon black	0,03	0,3
Process chemicals, Activators, Accelerators	0,011	0,1
Sulphur	0,002	0,02
Total	**0,09**	**1,0**

Total emissions from tyres in Sweden has been calculated by (Ahlbom and Duus, 1994), see Table X). These figures has been criticised by STRO (Scandinavian Tyre and Rim Organisation) who claim that the calculations do not account for tyre tread and therefore overestimates the emissions (Johansson, 2000). (Ahlbom and Duus, 1994) also calculated the PAH emissions connected to tyre wear and concluded that these, 28 μg km^{-1}, are about six times as high as the contribution from exhaust from a car with a catalytic converter, 5 μg km^{-1}. For Sweden the total amount is 14 tonnes PAH per year. STRO on the other hand, have calculated the emissions to 284-470 kg y^{-1}.

Table 2. Emissions from tyres (Ahlbom and Duus, 1994)

Component	*Total annual emissions in Sweden (tonnes)*
Polymeres	5 000
Black carbon	2 500
Oil	2 000
Zinc oxide	150
Stearin acid	70
Sulphur	100
Accelerators	50
Anti oxidants	100
Other	30
Totalt	**10 000**

In a recently published brake dynamometer investigation (Garg et al., 2000), brake lining emissions were calculated to 3.2-8.8 mg km^{-1}. Early work by (Cha et al., 1984) supports these figures. (Westerlund, 1998) estimated the contribution from brake lining wear to metals in the Stockholm environment and found that about 3,900 kg of copper, 900 kg of zinc, 560 kg of lead as well as a few kg of chromium and nickel were added each year from cars, buses and trucks. About 80% of the brake lining wear could be attributed to cars.

Emissions of airborne, inhalable particles from pavements are difficult to handle from literature. The pavement source contribution is often hidden in terms like "road dust". Even in Norway, where great efforts are made to measure PM_{10} concentrations in i.e. Oslo, the contributions from long range transport and local wood burning disturbs the possibilities to calculate pavement emissions (Larssen, 2000). The pavement wear caused by studded tyres has in Sweden decreased from about 30 g vkm^{-1} in the 80ies to about 10 g vkm^{-1} today. This sums up to about 110 000 tonnes each year.

4.2 Exhaust Particles

The main particle emissions related to vehicle exhaust come from diesel engines. As the attention paid to the health effects of these particles has increased, the development of cleaner diesel engines has accelerated. (Lenner and Karlsson, 1998) compiled particle emission figures from 19 different sources to be used in a quantitative model (Table 3). The figures presented here might therefore be somewhat out of date due to engine development.

Table 3. Emissions of exhaust particles (mg km^{-1}) (Lenner and Karlsson, 1998). W cat. = without catalytic converter (figures in brackets are standard deviations).

Car				***Heavy truck***		
W cat.	**Cat.**	**New cat.**	**Diesel**	**< 16 t**	**> 16 t**	**Bus**
16 (2)	2,4 (0,5)	1,4 (0,3)	279 (56)	630 (227)	1080 (430)	830 (274)

5. Health and Environmental Effects

A very extensive literature deals with the relationships between airborne particle concentrations and public health. This study mainly concentrates on literature that discusses the coarser fraction of inhalable particles (> 2.5 μm), but some information about smaller fractions has been included for comparison. Of special importance for human health is the inhalable fraction. Both their ability to enter more or less deep into the respiratory system and their ability to adsorb toxic substances, such as PAH and heavy metals, to their surface make them highly interesting to health scientists.

During the late 1990s, American studies have shown that latex particles from tyre wear contain allergens which might be coupled to an increased risk of latex allergy and asthma (Miguel et al., 1996; Williams et al., 1995). The studies have their background in the increasing over-sensitivity to latex in society, the causes of which are not yet clear. (Williams et al., 1995) found that 53% of the latex particles found in Denver air were inhalable and that their mean size was 6-7 μm. Chemical analyses suggested the particles originate from tyre wear. (Glovsky et al., 1997; Miguel et al., 1996) extracted latex allergens from tyre wear particles in Los Angeles and suggested the particles were a potentially important factor for latex allergy and asthmatic symptoms associated to air pollution.

The health effects of particles related to the use of studded tyres have mainly been studied in Japan. As early as the mid-1980s, (Morikawa, 1985) related the road dust concentrations to respiratory symptoms among asthmatic children and (Ikeda et al., 1986) to the frequency of upper respiratory symptoms. (Watanabe et al., 1990) studied the concentration of elements in the lungs of feral pigeons and found significantly higher concentrations of Si, Al, Pb and Ti in pigeons living in cities where studded tyres were used.

Many highly resistant Swedish pavements are based upon rocks containing high concentrations of quartz (mainly quartzite and porphyry). Quartz dust is well known to induce silicosis among for example miners, and quartz is regarded as one of the most toxic minerals. In Norway, ongoing research on PM_{10} particles from road tunnels shows that particles containing the minerals quartz, amphibole, chlorite and epidote induced a much higher production of interleukin-6 and –8 in human lung epitel than did particles containing plagioclase (Hetland et al., 2000). (Murphy et al., 1998) compared particles of crystalline quartz, amorphous quartz, from diesel exhaust and black carbon and their impact on rats' lungs. Somewhat surprisingly, he found more damages from crystalline quartz and amorphous quartz no effects from diesel or black carbon particles. This should imply a surface structure or chemical effect. As opposed to this, many studies rather imply a particle size effect, i.e. the chemistry or structure is not important (Camner, 2000).

Epidemiological studies relate inhalable particle concentrations to mortality, morbidity, lung cancer, asthma, respiratory symptoms and coughs, usually in urban areas and to a greater extent among sensitive populations such as children, asthmatics and elderly people. Extensive compilations of current knowledge has been made by e.g. (Vedal, 1997) and (Areskoug, 2000).

Commonly, literature implies that fine and ultra-fine particles (< 1 μm) show stronger relationships to health effects than do coarser fractions. The increase in mortality is generally 0.5-1.0% per 10 μg m^{-3} increase in PM_{10} concentration. Hospital admissions due to short term exposure increases by 0.5-3.0%, which confirms a relationship to particle concentration. The relationship is often stronger for symptoms in the lower respiratory tract than in the upper and also stronger for elderly people but also for children (Areskoug, 2000).

Despite the fact that most studies stress the importance of the fine fractions, quite a large number of exceptions exist. (Pekkanen et al., 1997) found that ultra-fine particles were not more strongly related to variations in peak expiratory flow rate (PEFR) than were PM_{10} or black smoke particles. A study made in Cochella valley in USA where coarse particles with a geologic origin contribute to a very large fraction of PM_{10} show that PM_{10} was significantly associated to all used measures of mortality (Ostro et al., 1999). On the other hand, (Schwartz et al., 1999) saw no such signs in a similar study. In Mexico City, (Castillejos et al., 2000) found that $PM_{10-2.5}$ had a stronger effect on mortality (4.07% associated with a 10 μg m^{-3} concentration increase) than PM_{10} (1.83%) and $PM_{2.5}$ (1.48%). A probable explanation might be the presence of biogenetic material in the $PM_{10-2.5}$ fraction.

Except for the health effects, non-exhaust particles affect public comfort by dirtying cars, sidewalks, house fronts, windows and even insides of houses.

Literature on the environmental effects of road dust particles as such is scarce. Most of it deals with the road as a source of pollution for the roadside environment (Bækken, 1993; Bækken and Jörgensen, 1994; Bjelkås and Lindmark, 1994; Gjessing et al., 1984; Kobriger and Geinopolos, 1984; Lygren and Gjessing, 1984; Sansalone et al., 1995). Particles from pavement wear contribute to the structure and composition of the roadside soils. The accumulation of material might amount to as much as 1.5 cm y^{-1}. Roadside soils diverge strongly from adjacent soils both regarding size distribution and chemical properties. High pH, high content of base cat-ions and heavy metals are characteristic features of these soils (Bækken, 1993).

Some studies concern the effects of particles on vegetation. Particles on the surface of leaves and needles have been shown to cause stress and therefore reduced growth due to increased temperature, blocked stomata and the hygroscopic properties of some particles (Farmer, 1993; Flückiger et al., 1978). Effects on limnic systems include high, and sometimes toxic, concentrations of PAH and heavy metals in lake sediments (Bækken and Jörgensen, 1994), but also first flush effects, where the particle depot accumulated on a road causes high concentrations of toxic compounds in streams during rainfall.

De-icing salt, which can be transported to the roadside environment as an aerosol or as dry dust, also affects vegetation negatively, which is a very visible problem in Sweden as Norway spruce and Scots pine along salted roads often turn brownish in spring due to the salt (Blomqvist, 2001). Salt deposited on leaves and needles causes osmotic stress causing dessication. The salt has also been shown to accumulate in ground water reservoirs with a hydrologic connection to road environments (Thunqvist, 2000).

6. Discussion

The sources for non-exhaust particles, judging from this literature survey, are many and their interplay complicated. Information about emissions and characteristics of more diffuse sources, like corrosion and biogenic material deposited on the road, has not been found in literature.

A critical report on tyre wear in Sweden (Ahlbom and Duus, 1994) has caused a debate, where the STRO claim the report to be incorrect and exaggerated in many respects. Nevertheless the debate has led many tyre manufactures to develop winter tyres without HA-oils, which is used as an argument in commercials. In summer tyres, the HA oils are more difficult to omit, since they are responsible for a large part of the grip properties (Johansson, 2000). Very varying information is reported about the properties of tyre particles. Both small quantities of inhalable particles as well as rather large proportions of respirable particles are reported, which might be a result of the large variation in materials, wear conditions and measurement methods.

Studded tyres and the related pavement wear are the particle source most thoroughly investigated in conditions similar to those in Sweden. In our neighbouring country, Norway, studded tyres have been the subject of much debate and major research for two decades. The focus has been on health effects, and an economic investigation during the “Veggreppsprosjektet” (Road grip project) showed that restrictions in the four largest cities in Norway were profitable to society (Krokeborg, 1997). So far, only Oslo has introduced the restrictions since 1999. Studded tyre use is now about 30% as compared to 70% before the restrictions. Due to large variations in seasonal weather it is too early yet to determine any effect on PM_{10} concentrations (Hagen and Haugsbakk, 2000).

There are a few studies on brake linings and their contribution to pollution. Most of these show that particulate heavy metals are the main cause of concern. An important aspect from a health point of view is that brake lining particles are very small and therefore potentially more dangerous to health.

For Swedish conditions the three most important emission sources for non-exhaust particles are, in order of magnitude, pavement wear (about 110,000 tonnes), tyre wear (about 10,000 tonnes) and brake lining wear (about 1,000 tonnes). The total amount of these particles emitted during a year is in the same order of magnitude as that of exhaust particles, but seasonal effects of climate, local sources, maintenance actions etc make the emissions very uneven both temporally and spatially.

Health surveys dealing with particle effects are common and usually based upon measurement of PM_{10} and/or $PM_{2.5}$, which are measures produced for this specific purpose. The results of these studies show a rather scattered picture of how PM relates to toxicological or epidemiological effects. There is a consensus that particle size matters and that there is a specific particle effect, but what this effect really involves is still not clear. A part of the problem definitely lies in the PM methodology. PM_{10} or $PM_{2.5}$ says nothing about size distribution below 10 or 2.5 µm and nothing about chemical or physical properties, i.e. surface area, of the particles. According to (Camner, 2000), there is a gap between toxicology and epidemiology, since the effects shown in epidemiological studies, with effects on populations at rather low PM concentrations can not be verified in toxicological experiments where it takes much higher concentrations to cause the same medical symptoms. Regarding environmental effects of particles, these are seldom related to particles as such, but rather to pollution from PAH or heavy metals.

7. Conclusions and research needs

The international literature on non-exhaust particles and their effects is quite extensive. The information about particle emissions and particle characteristics displays a very large variation though, depending on investigation quality, methods and extent as well as geographical variations. The material is often based on short-term measurements seldom valid for other geographic locations or during other time intervals. For Swedish conditions, the following approximations might be considered:

Wear

Total in Sweden

- Pavement 110 000 t y^{-1}
- Tyre 10 000 t y^{-1}
- Brake linings 1 000 t y^{-1}

Emissions

- Pavement wear from studded tyre use <10 g km^{-1}
- Tyre car 0,006 - 0,36 g km^{-1}
  truck, bus approx. 1 g km^{-1}
- Brake lining wear 0,0032 - 0.0088 g km^{-1}
- Re-suspension 0,13 - 6 g km^{-1}
- Exhaust car with catalytic converter 0.0014 - 0.0024 g km^{-1}
  car (diesel) 0,279 g km^{-1}
  heavy truck (>16 tonnes, diesel) 1,08 g km^{-1}

Health effects

Toxicological aspects

- Tyres — allergy and asthma from latex particles?
  relatively high PAH content
  relatively large percentage of PM_{10}
- Pavement wear — important source for PM_{10} in the road environment
  mineral composition and surface properties?
- Brake lining wear — large percentage of PM_{10}
  especially important in cities?

Epidemiological aspects

- Few other measures than PM_{10} and $PM_{2.5}$ are studied
- Wear particles mainly in the coarse fraction $PM_{10-2.5}$
- Many studies show a higher correlation of health effects to $PM_{2.5}$ but there are also studies indicating higher correlation to $PM_{10-2.5}$
- Not only a particle effect. Chemistry and surface properties probably also important.

To be able to thoroughly investigate the relations between non-exhaust, as well as exhaust-, particles and health- and environmental effects it is essential to complete PM measurements with measurements giving more information on chemical characteristics, particle size distribution and maybe also surface characteristics. Characterisation should be made on source specific particles to be able to make reliable source apportionments in field measurements as well as risk assessments. More field data covering a greater temporal and spatial variation are needed to improve knowledge about variations in particle concentration, composition and characteristics.

At VTI (National Swedish Road and Transport Research Institute) efforts to characterise particles from pavement and tyre wear using the VTI road simulator are being made. The road simulator offers the possibility to study “pure” wear particles since it is situated inside a building. Ongoing projects deal with emission factors for inhalable wear and re-suspension particles to be used in emission models and an inventory of road cleaning methods effective for particle removal. Future particle research efforts at VTI are planned to include spatial and temporal variations and model validation studies in the field.

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