REGULATIONS AND POLICY MEASURES RELATED TO THE REDUCTION OF AMBIENT PARTICULATE MATTER

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Abstract

The aim of this paper was to review the existing regulations and policy measures related to the reduction of airborne particulate matter, which is perhaps the most complex airborne pollutant to understand and manage. The complexity arises from the multi-factorial nature of particulate matter, and is amplified by difficulties associated with the measurement of various particle properties, as well as the lack of clarity regarding which properties are the most significant in terms of their impact on human health and the environment, and, therefore, which should be the prime target of regulations. Consequently, only a few standard techniques exist with which to measure certain particle characteristics.

Health guidelines, the most important tool in developing national regulations, exist only in relation to one type of particle matter characteristic, since the current state of knowledge on exposure-response relationships only provides a basis for particle mass concentration health guidelines - currently two size ranges: $PM_{2.5}$ and PM_{10}. It has been shown that within the current range of concentrations studied in epidemiological studies there are no threshold levels and that there is a linear exposure-health response relationship. Based on this, in the most recent review the World Health Organization Air Quality Guidelines, a new set of guidelines for particulate matter was introduced, with the guideline values representing the lowest end of the range over which significant effects on survival have been observed. These values are not much higher than the concentration levels encountered commonly in natural environments. While there is a large body of toxicological evidence of the potential detrimental health impacts of ultrafine (UF) particles measured in terms of number concentration, epidemiological studies have not yet provided any quantification of exposure-response relationships. Therefore, there is no scientific basis for setting particle number or surface area health guidelines. However, the current levels of UF particle concentrations in environments affected by vehicle emissions are often up to two orders of magnitude higher than in the natural environments.

Motor vehicle emission standards are derived on the basis of factors such as technological capabilities of engine design and after-treatment devices, as well as the impact of emissions on the environment and ambient air quality standards. Emission standards and test procedures are well stipulated for both heavy-duty and light-duty vehicles in the US and in Europe. These standards are generally described in terms of total particle mass emission, rather than PM_{10}, but in 2009 the European Union introduced a particle number limit to its Euro 5/6 emission standard for light-duty diesel vehicles. In response to these increasingly stringent standards, manufacturers have been forced to consistently improve engine design and fit more sophisticated emission control devices and systems to vehicles. Another important group of emission standards are those related to industrial emissions, which commonly regulate total emissions of particulate matter, and not size specific emissions (such as $PM_{2.5}$ or PM_{10}). This constitutes a clear disparity between emission and air quality regulations, with the later being size specific. There are many other sources of airborne particulate matter, some of which include aviation, shipping, biomass combustion, waste incineration, construction, agricultural processes, cigarette smoke, fugitive emissions, cooking, non-vehicle diesel combustion, and two- and four-stroke combustion. None of these sources are directly regulated in terms of particle emissions.

1. Introduction

A process leading to the development of regulations and introduction of policy measures to reduce concentrations of a pollutant in the environment in order to protect human health is a very complex one. Such a process is normally initiated when evidence emerges that exposure to the pollutant constitutes risk to human health. Some of the key subsequent steps of the process include quantification of both the exposure response relationship and the impact of the proposed regulations or policy measures on the actual reduction of the exposures. The entire process of risk assessment for the purpose of risk management encompasses several more steps; in particular, establishing of the exposure-dose and dose-response relationships (these two not always conducted), life time individual risk, and risk to exposed population (Naugle and Pierson, 1991; Pierson et al., 1991). In relation to air quality, guidelines for the protection of human health against adverse effects of pollution are developed first based on public health studies (epidemiological, toxicological, and clinical), which in turn serve as a foundation for setting national air quality standards as the main means to enforce policies related to air quality management.

Quantification of risk due to an environmental exposure to a particular pollutant is always a complex process, since the multiplicity of other pollutants and factors have an impact on the individual at the same time, and the particular exposure investigated is only one of the factors. Risk assessment of airborne particulate matter is even more complex; in addition to the aforementioned challenges is the complexity arising from the multifactorial nature of particulate matter. While some pollutants, such as carbon monoxide, have only one characteristic (i.e. their concentration) averaged over relevant periods of exposure, there are many characteristics of particulate matter which can be considered. One of the fundamental properties is size: airborne particles range in size over five orders of magnitude, from molecular dimensions to the sizes that are distinguishable with the naked eye (from about 0.001μm to about 100 μm; Baron and Willeke, 2001). Other characteristics include: number concentration, number size distribution, mass concentration mass size distribution, surface area, shape, electrical charge or light scattering properties. Usually only some of these properties are measured, and almost never all of them, simultaneously.

In relation to some of these metrics, not only hazard has been established, but epidemiologic studies have quantified exposure – response relationships. This is particularly the case for mass concentration fractions including $PM_{2.5}$ and PM_{10} (mass concentration of particles with aerodynamic diameters smaller than 2.5 and 10 μm, respectively; more precisely the definitions specify the inlet cut-offs for which 50% efficiency is obtained for these sizes) and TSP (Total Suspended Particulate Matter). However, in relation to other metrics, for example number concentration of ultrafine particles (< 0.1 μm), while there is ample evidence of hazard provided by toxicological studies, the epidemiology has not yet established an exposure – response relationship, and therefore health guidelines could not be proposed (as discussed in more detail below). In relation to other metrics, including particle surface area, there is even less quantitative evidence of effects, yet mechanisms have been proposed by which exposure to this and to other metrics can lead to health effects. However, whether the effects are independent, and whether each of the metrics should be regulated, or whether they are dependent and therefore only some of them need to be regulated, are open questions at present.

An additional issue is related to the nature of particle formation: primary versus secondary particles. A primary particle is a particle introduced from the source into the air in solid or

liquid form, while a secondary particle is formed in the air by gas to particle conversion. Particles in the ultrafine and more generally in the submicrometer ranges are typically generated by combustion, gas to particle conversion, nucleation or photochemical processes with some of them primary and some secondary in nature. In contrast, supermicrometer particles are generated by a variety of mechanical processes, and as such, are primary in nature. While management of primary particles is somewhat easier, secondary particles present a far greater challenge, particularly since the scientific understanding of the secondary particle formation process is far from complete.

Quantitative assessment of individual source contribution to specific particle metrics presents another challenge. Again, there is a reasonably good understanding and quantification in relation to some of the metrics, such as $PM_{2.5}$ or PM_{10}. However, there is little information available regarding particle number (further complicated by the issue of secondary particle formation), and it is almost non-existent in relation to particle surface area. Without source apportionment it is not possible to manage the emissions in an informed way. Subsequently, the link between both source emission and air quality standards is often lacking (this will be further discussed below in relation to vehicle emissions). In general, anthropogenic combustion sources significantly contribute to ambient particulate matter, with vehicle combustion contributing approximately 90% and 65% of ultrafine and nanoparticle count, respectively, in the urban environment (Mejia et al., 2008). These particles are generally measured in terms of number concentration because their mass is very small and very little or no information about them can be gained from $PM_{2.5}$ or PM_{10} measurements.

Instrumentation and measurement techniques are an intrinsic part of the discussion of particulate matter management. Sampling of particles and choice of appropriate instrumentation and methodology is primarily based on particle physical properties. Due to different physical properties, different methods need to be used for measuring very small particles (mostly affected by diffusion) compared to those employed for large particles (mostly affected by inertia). While there are generally accepted instrumental techniques applied to some particle metrics, especially mass concentration measurements, there are no standard methods in relation to others (particle number for example). This further complicates the matter, as introduction of any regulation in regard to concentrations or emission levels has to be accompanied by setting a method by which compliance with the regulations be measured and monitored.

With the above complexities in mind, the aim of this work was to review the existing health guidelines and national standards in relation to the ambient concentrations of particulate matter, as well as national source emission standards. The focus of the review was on particle physical properties of mass and number, without, however, including chemistry: this could not have been discussed without including the topic of phase distribution (between the vapour and particle phases), and as such would require a significant broadening of this work, beyond its scope. In addition, occupational environments, where specific production processes lead to the generation of specific types of particles (such as mining or mineral processing) are not considered here, since these are usually managed by occupational regulations.

2. Particulate Matter - Mass

2.1 Health Guidelines

Health guidelines in relation to ambient particulate matter as well as other environmental pollutants are published by the World Health Organization (WHO) and adopted based on expert panel agreements, following consideration of evidence from medical and public health studies. As stressed in the latest edition of the guidelines, they are written "for worldwide use, intended to support actions aiming for air quality at the optimal achievable level of public health protection in different contexts" (WHO, 2005).

Review of the existing evidence by the WHO panel showed that a broad range of effects have been documented after both short and long-term exposures, in particularly affecting the respiratory and cardiovascular systems of children and adults, as well as some susceptible groups within the general urban population of developed and developing countries. Importantly, there is no evidence of an existence of a threshold below which no adverse health effects would be anticipated, and, in fact, the lower range of concentrations at which effects has been demonstrated is not significantly greater than the background $PM_{2.5}$ concentration (estimated at 3-5 $\mu g/m^3$) in the United States and Western Europe.

Since the majority of monitoring data as well as epidemiological evidence is based on measurement of PM_{10} as opposed to other particulate matter metrics, PM_{10} was the most obvious indicator to chose in relation to the guidelines. However, the numerical guideline value itself is based on studies measuring $PM_{2.5}$, and on a $PM_{2.5}/PM_{10}$ ratio of 0.5 (typical in developing country urban areas and at the bottom of the range in such areas in developed countries). PM_{10} includes both the coarse (PM_{10}-$PM_{2.5}$) and fine ($PM_{2,5}$) fractions and if justified by local conditions, the ratio may be changed accordingly when setting the local standards.

The composition of coarse and fine particles is likely to vary substantially between cities around the world depending in particular on the specific local sources. A major contribution to the fine particles is combustion of wood and other biomass. Since similar effect estimates have been reported across cities in developed and developing countries, despite likely differences in fractional contribution from these two sources of combustion, it was concluded that it is reasonable to assume generally similar effects of $PM_{2.5}$ from these different sources.

The values chosen for the WHO air quality guidelines, which apply to both outdoor and indoor air are:

$PM_{2.5}$: 10 $\mu g/m^3$ annual mean, 25 $\mu g/m^3$ 24-hour mean
PM_{10}: 20 $\mu g/m^3$ annual mean, 50 $\mu g/m^3$ 24-hour mean

The rationale for the choice of the annual average guideline value of 10 $\mu g/ m^3$ for $PM_{2,5}$ was that it represents the lower end of the range over which significant effects on survival have been observed in the American Cancer Society Study (ACS) (Pope et al., 2002). Further discussion on this can be found in the document (WHO 2005), which also concludes that although adverse effects on health cannot be entirely ruled out even below that level, it is a level achievable in urban areas of developed countries, and therefore attainment to it is expected to effectively reduce the health risks. However, the experts emphasized the need to

reduce exposure to non-threshold pollutants such as particulate matter, even where current concentrations are close to or below the proposed guidelines.

In addition to the guideline values, three interim targets (IT) were defined, which are achievable with successive and sustained abatement measures and helpful in gauging progress over time in the process of steadily reducing population exposures to particulate matter.

2.2 National Standards

National air quality standards are based upon the potential for health and economic effects of identified pollutants. As discussed above, the relationship between some size fractions of particulate matter and adverse health effects has been well documented. The present indicator of particulate matter is PM_{10}. Consequently, while most monitoring studies report particulate mass as PM_{10}, the majority of epidemiological studies use it as the exposure indicator. In many cities, especially in the developing world, the average annual levels of PM_{10} exceed 70 $\mu g\ m^{-3}$; a level that is known to result in adverse health effects. It has been estimated that reducing PM_{10} levels from 70 to 20 $\mu g\ m^{-3}$ can reduce mortality rates attributed to air quality by 15% (WHO, 2005). However, there is no level below which damage to health has not been documented. Therefore, in specifying national standards, the aim is to achieve the lowest concentrations possible.

Particulate matter standards were first introduced in the US in 1971. In 1987, the indicator of the standards was changed from TSP to PM_{10}. In 2006, it was decided to retain the existing 24-hour PM_{10} standard of 150 $\mu g\ m^{-3}$ and, "*due to a lack of evidence linking health effects to long-term exposure to coarse particle pollution*", the annual PM_{10} standard of 50 $\mu g\ m^{-3}$ was revoked (USEPA, 2006).

Fig 1 presents the 24-hour mean PM_{10} standards currently adopted by various countries. It should be noted that some countries like China and India specify different standards for industrial, urban and rural areas and also for sensitive areas that require special protection. The values shown in the chart for China refer to "typical urban and rural areas". Several other countries that are not shown in the chart use indicators based on total particulate matter (TPM) or TSP and not PM_{10}. While the adoption of particulate matter standards by developing nations is to be encouraged, it is also important to ensure that the resources and facilities to make the required measurements are available.

As stated earlier, the WHO guidelines for annual and 24-hour mean ambient fine particle ($PM_{2.5}$) levels are 10 and 25 $\mu g\ m^{-3}$, respectively. At present, with the exception of the US and Canada, no countries have adopted a fine particle standard. The US EPA first issued fine particle standards in 1997, when the annual and 24-hour standards were set at 15 and 65 $\mu g\ m^{-3}$, respectively. In 2006, the 24-hour standard was tightened to 35 $\mu g\ m^{-3}$ while the annual standard was retained at 15 $\mu g\ m^{-3}$. Note that these two current values are still well away from the WHO guidelines. Recently, the European Union (EU) has introduced a $PM_{2.5}$ standard target value of 25 $\mu g\ m^{-3}$, averaged over one year, that enters into force in 2010 (EU, 2008). A target value is to be attained as far as possible by the attainment date and so is less strict than a limit value. The corresponding limit value has been set to 20 $\mu g\ m^{-3}$ to become legally binding in 2015.

The US EPA is in the process of revising the particulate matter standards, with one of the proposals being the introduction of a coarse particle standard for particulate matter between 2.5 and 10 μm (USEPA, 2008b). This process and schedule of this review is based on two key components in the national ambient air quality standards review process: an Integrated Science Assessment (ISA) and a Risk and Exposure Assessment (REA), both of which are expected to be completed by July 2010 (USEPA, 2009). The assessment will address concerns related to human health risk and exposure, visibility impairment as well as other welfare-related effects.

The Australian Environment Protection (Ambient Air Quality) Measure was varied in 2003 to include particles as $PM_{2.5}$, in the form of advisory reporting standards (NEPC, 2003). The present annual and 24-hour mean ambient $PM_{2.5}$ reporting advisory standards are 8 and 25 μg m^{-3}, respectively. This annual reporting standard is in fact more stringent than the corresponding WHO guideline value.

For completeness of this review it should be mentioned that there has also been discussion about introducing a PM_1 standard, as it would provide pertinent information about contributions from combustion processes and would afford improved delineation between combustion and mechanically generated aerosols compared to that provided by $PM_{2.5}$ measurements (Morawska et al, 2008a). It would thus appear that PM_1 and PM_{10} mass standards would be most desirable from the legislation point of view. The main problems, however, are that PM_1 is not routinely monitored and there are some technical challenges in relation to such measurements, and that there have been too few epidemiological studies conducted to quantify exposure-relationship in relation to PM_1.

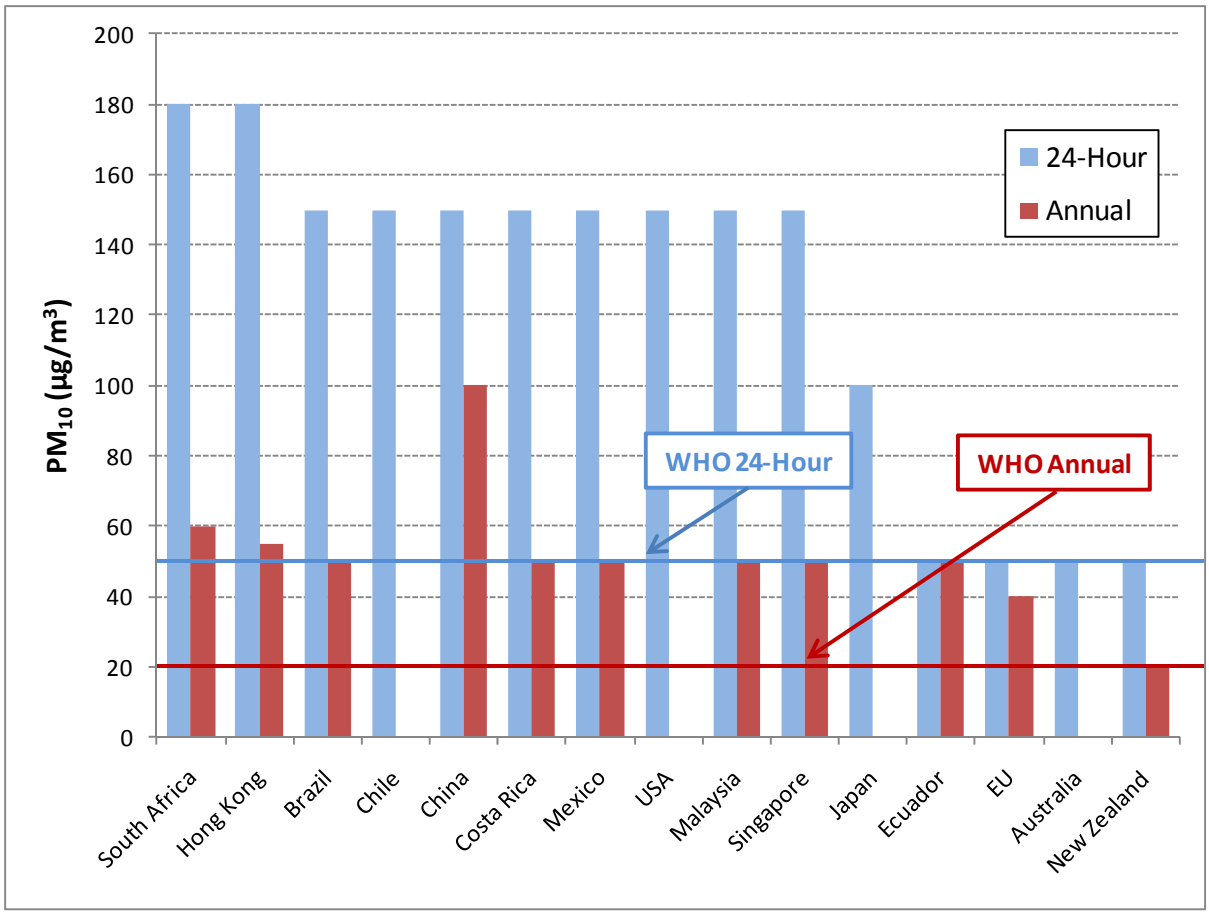

Fig 1: 24-hour and annual mean PM_{10} standards currently adopted by various countries. The two horizontal lines show the corresponding WHO guidelines. Chile, USA, Japan and Australia do not enforce a 24-hour standard at present. Sources: South Africa: SAGG (2005); Hong Kong: HKEPA (2006); Brazil: CONAMA (2005); Chile: ; China: GB (1996); Costa Rica: ; Mexico: DOF (1994); USA: NAAQS (2009); Malaysia: DOE (2009); Singapore: Yong et al (2002); Japan: MOE (2009); Ecuador: ; EU: EU (2008); Australia: NEPC (2003); New Zealand: NZMOE (2004).

3. Particle Number

3.1 National Standards

Unlike for particle mass concentrations ($PM_{2.5}$, PM_{10}), there are no standard methods for conducting size-classified particle number measurements. Furthermore, the impact of ultrafine particles on human health and the environment is still not fully quantified so that there is a lack of information on exposure-response relationships from epidemiological studies. These factors have prevented the development of particle number based health guidelines and any subsequent national regulations.

The main source of ultrafine particles in urban locations is motor vehicles. While particle number concentrations at clean background sites are of the order of 2000-3000 cm^{-3}, levels at urban sites are about 4 times higher. Levels at sites that are closely affected by motor vehicle emissions are even higher. Morawska et al (2008b), in an extensive review of the literature,

showed that particle number concentrations at road, roadside, street canyon and road tunnel sites are respectively 16, 18, 27 and 64 times higher than typical background values. Thus, unlike for particle mass, the range of concentrations between clean and vehicle effected environments is relatively large, varying over two orders of magnitude. This large variation across different environments has profound significance in relation to human exposure assessment and epidemiological studies, and implies that unless exposure assessment is conducted where the exposures occur; it is unlikely that epidemiological studies will provide meaningful answers based on monitoring in central locations alone. In other words, central monitoring underestimates exposures and may lead to inappropriate guidelines and standards for the management of public health risks.

3.2 Cleanroom Particle Number Concentration Standards

Cleanrooms serve various functions during the production of many items that share a common need for a highly controlled and pure environment during their manufacture. Additionally, cleanrooms or isolation rooms are often required in hospitals to prevent the spread of pathogens during surgical procedures, and to facilitate containment of infectious persons and protection of the immune-compromised. Cleanroom classes are defined on the basis of size-based particle number concentrations that are not to be exceeded within them. There are myriad cleanroom standards and guidelines around the world, although measures to rationalise the number of standards are in place (Möller, 1999). The regulation, control and attendant design aspects related to particle number concentration in cleanrooms are topics discussed elsewhere, and the reader is directed to Whyte (1999) and chapter 15 of the ASHRAE Handbook of HVAC Applications (1999) for an overview of these topics. Within the context of this chapter, the key point to consider is that particle number concentrations within cleanrooms are tightly regulated by virtue of the specific criteria with which they must comply to secure and maintain a given classification. For example, to comply with Class 5 criteria under ISO 146114-1, no more than 1.0×10^{5}, 2.37×10^{4}, 1.02×10^{4}, 3.52×10^{3}, 8.32×10^{2} and 2.9×10^{1} particles m^{-3} may be present in room air for particle sizes equal to and greater than 0.1, 0.2, 0.3, 0.5, 1 and 5μm, respectively (Möller, 1999). The large difference between acceptable concentrations of the smallest (i.e. 0.1 μm) and the largest (i.e. 5 μm) particles within the aforementioned cleanroom class thus considers typical particle size distributions present in many indoor and outdoor environments (i.e. dominated, in terms of number, by smaller particles). Different activities necessitate varying degrees of air cleanliness, and the several cleanroom classes are designated in order to minimise the possibility of negative effects caused by airborne particles, and other parameters, on the process(es) undertaken within the cleanroom.

4. Source Emissions of Particulate Matter

4.1 Particle Mass

There are many sources that contribute to particulate matter in the environment. Fig 2 shows a breakdown of PM_{10} emissions from various sectors in the US in 2002. The total emission is 1.87 x 10^7 tonne, of which 50% comes from road dust. Industrial processes together with electricity generation account for about 1.6 x 10^6 tonne or 9%. The corresponding $PM_{2.5}$ emissions show a 27% contribution from fires, followed by 22% from industrial processes and electricity generation and 19% from road dust. Nowadays, most industrial stacks in the developing world are equipped with highly effective dust removing facilities, such as electrostatic precipitators and fabric filters, so that the total particle mass emissions are often reduced by over 99% to give concentrations of less than 1-2 mg m^{-3}. The PM_{10} component is higher than 90% when the sources are combustion or evaporative processes (Dreiseidler et al, 2000).

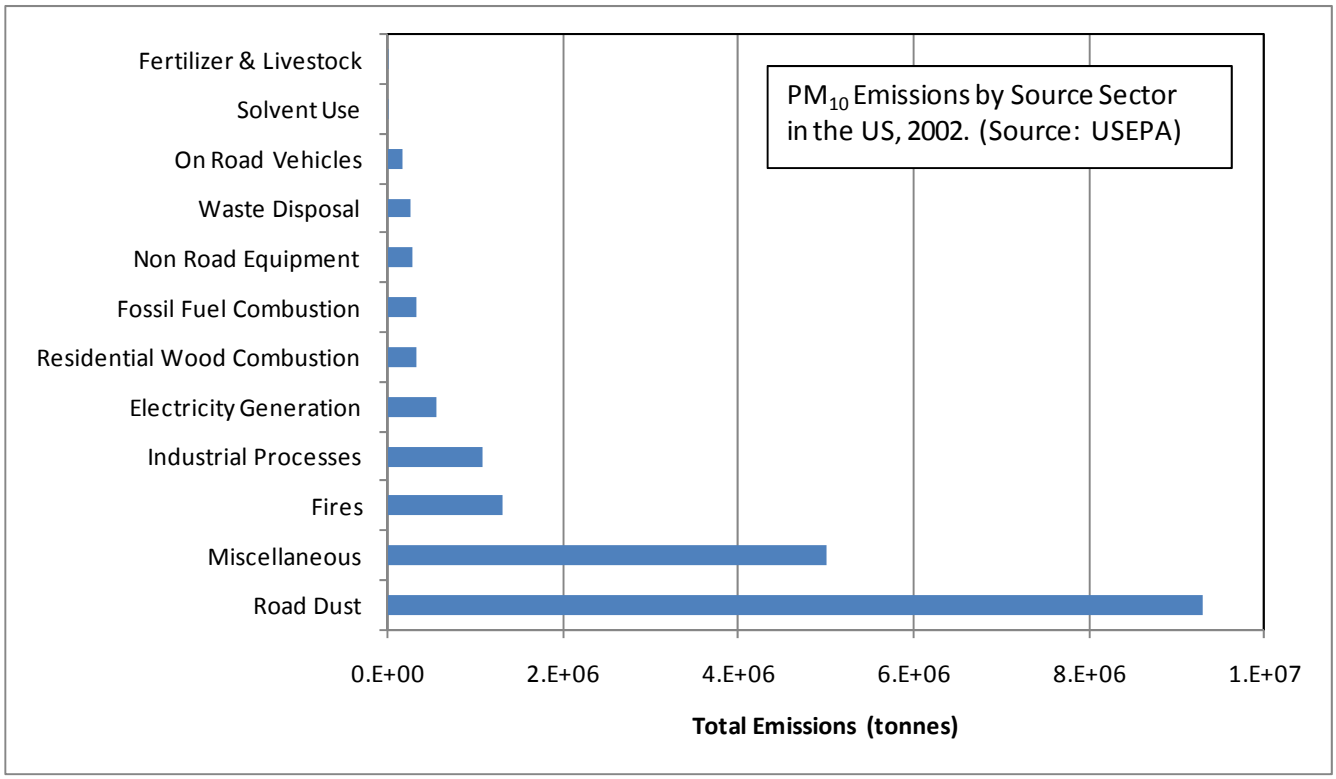

Fig 2: PM_{10} Emissions by Source Sector in the US, 2002. (Source: USEPA (2008)).

4.1.1 Industry

A major industrial source of particulate matter in the environment is coal-fired combustion for electricity generation. Source performance standards in the US restrict total particulate emissions from new coal-fired power plants to 6.4 ng J^{-1} (USEPA, 2005). The EU standards are specified for three rated thermal input ranges of 50-100, 100-500 and >500 MW and are

100, 100 and 50 mg m^{-3} for existing plants and 50, 30 and 30 mg m^{-3} for new plants, respectively. Australian standards are also based on the age of the plant, and range from 400 mg m^{-3} for plants licensed before January 1972, to 50 mg m^{-3} for plants licensed after September 2005.

Industrial sources include asphalt and cement manufacture, rock processing, fertilizer, chipboard, steel and other metal production plants. Most of these industries have their own emission standards that vary from country to country, and may be specified for individual stages in the manufacturing processes.

4.1.2 Motor Vehicles

Motor vehicle emission standards specify the maximum amount of pollutants in engine emissions. First introduced in California in 1959 to control carbon monoxide and hydrocarbon emissions from gasoline engines, they are usually regulated through government ministries and agencies responsible for the protection of the environment. While the standards dictate how much pollution vehicles may emit, it is left to the manufacturers to determine how these limits are to be achieved. In response to tighter standards, vehicle manufacturers have consistently improved engine design and equipped more sophisticated emission control devices and systems over the years.

Today, emissions, including particulate matter, are regulated in many countries and specified separately for the various emission parameters and vehicle and fuel types. Particulate mass emissions are measured by gravimetric methods over engine or vehicle test cycles, which consist of a sequence of speed and load conditions conducted on an engine or chassis dynamometer. Test cycles are designed to closely mimic the typical driving patterns of vehicles in various situations. Emissions measured on a vehicle or chassis dynamometer are usually expressed in mass per distance, such as g km^{-1} while when measured on an engine dynamometer in accordance with a test cycle, they are expressed in terms of mass per mechanical energy output, such as g kWh^{-1}. Emission test procedures are not the same across different countries and many types of drive cycles are used. Therefore, even when expressed in the same units of measure, engine emissions measured on different test cycles may not be comparable. This should be borne in mind when comparing emission standards from different countries.

4.1.2.1 Cars and Light-Duty Trucks

The US presently adopts the Tier 2 Standards as specified in the National Clean Air Act that were phased in between 2004 and 2009. Emissions are measured over the Federal Test Procedure (FTP-75) and expressed in g $mile^{-1}$. These standards apply to all passenger cars, minivans, light-duty trucks and SUV's irrespective of fuel type (such as gasoline, diesel or alternative fuels). However, they are structured into 11 certification levels of different stringency called 'certification bins' and, within certain restrictions related to type of vehicle and weight, vehicle manufacturers are given the choice of assigning vehicles to the available bins. The particulate mass emission standards for cars and light-duty vehicles range from 10 to 20 mg $mile^{-1}$.

The EU adopted emission regulations for new light duty vehicles (cars and light commercial vehicles) as specified in Directive 70/220/EEC. This basic directive has been amended a number of times from Euro 1 to the most recent Euro 6 standard, which was applied in 2009.

These regulations include different emission limits for diesel and gasoline powered vehicles and are in general more stringent than the corresponding US standards. The Euro 4 standard introduced in 2005 limited the sulfur content in both fuels to 50 ppm, with the proviso that 'sulfur-free' (≤ 10 ppm) be made available from 2005 and mandatory from 2009. Emissions are determined over the European 13-mode test cycle and expressed in g km^{-1}. The EU particulate matter emission standards for passenger cars and light commercial vehicles are summarised in Table 1. There were no European regulations for petrol cars until Euro 5 in 2009. Light commercial vehicles are classified into three classes according to vehicle mass. The Euro 5 and 6 standards of 0.005 g km^{-1} are to be changed to 0.003 g km^{-1} using the particle measurement programme (PMP) procedure.

		Passenger Cars		Light Commercial Vehicles					
		Petrol	Diesel	Petrol			Diesel		
Tier	Date			N1	N2	N3	N1	N2	N3
Euro I	Sep 1992	-	0.14	-	-	-	0.14	0.19	0.25-
Euro 2 IDI	Jan 1996	-	0.08	-	-	-	0.08	0.12	0.17
Euro 2 DI	Sep 1999	-	0.10	-	-	-	0.10	0.14	0.20
Euro 3	Jan 2000	-	0.05	-	-	-	0.05	0.07	0.10
Euro 4	Jan 2005	-	0.025	-	-	-	0.025	0.04	0.06
Euro 5	Sep 2009	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005
Euro 6	Sep 2014	0.005	0.005	0.005	0.005	0.005	0.005	0.005	0.005

Table 1: The European Union particulate matter emission standards for passenger cars and light commercial vehicles (N1: ≤1305 kg; N2: 1305-1760 kg; N3: >1760 kg).

Australian Design Rules (ADR) to regulate vehicle emissions are based on European standards. The current standards for all vehicles are equivalent to Euro 4. Japan introduced a regulation based on a new JC08 cycle in 2009 that limits particle mass emissions from cars and light duty vehicles to 0.005 g km^{-1}, to be fully phased-in by 2011. China presently stipulates Euro 4 standards for the Beijing region and Euro 3 for the rest of the nation.

4.1.2.2 Heavy-Duty Trucks and Buses

In the US, heavy-duty vehicles are defined as vehicles with gross weight over 8,500 lbs. USEPA emission standards for heavy duty diesel truck and bus engines controlled particle mass emissions from 0.60 g bhp^{-1} h^{-1} in 1990 to 0.10 g bhp^{-1} h^{-1} in 1998. A new stringent mass emission standard that took effect with the 2007 heavy-duty engine model year limits particle mass emissions to 0.01 g bhp^{-1} h^{-1}. This included a diesel fuel regulation that currently limits sulphur content to 15 ppm.

Emission regulations for heavy-duty vehicles in Europe were originally introduced by Directive 88/77/EEC which was followed by a number of amendments. As a result, mass emission standards for diesel heavy-duty engines were reduced from 0.36 g kWh^{-1} in 1992 (Euro 1) to 0.02 g kWh^{-1} in 2008 (Euro 5). Fig 3 shows the improvement over this time. It is further expected to be reduced to 0.01 g kWh^{-1} in 2013 (Euro 6).

Emission standards for Australia are generally based on European standards. Starting from Euro 1 standards in 1995/96, the present standard (2009) is Euro 4.

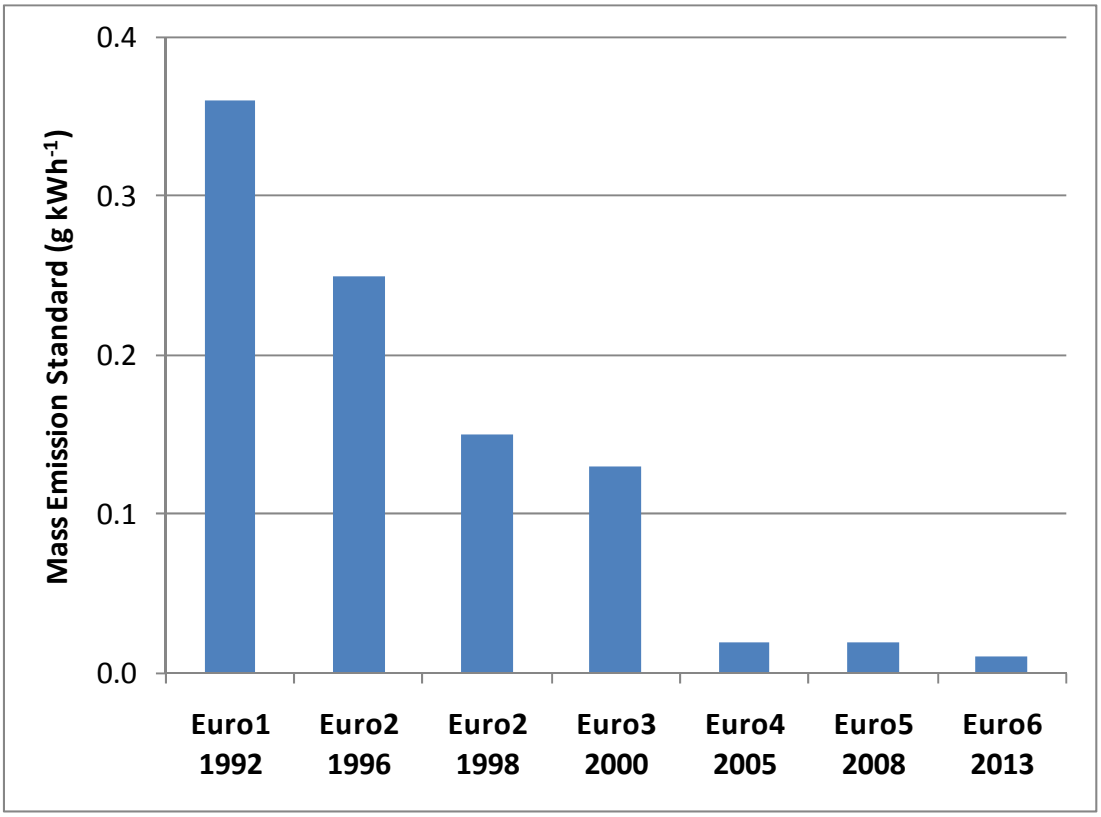

Fig 3: European mass emission standards for heavy-duty diesel engines (data derived from Dieselnet, 2009)

4.2 Particle Number

4.2.1 Industry

There is very little information in the literature on particle number emissions from industrial facilities. Consequently, there are no standards controlling particle number emissions from industrial sources. Dreiseidler et al (2000) used an Anderson cascade impactor to collect particulate samples in industrial stacks in Germany. The samples were analysed gravimetrically and the peak particle number concentrations were found at an aerodynamic diameter of 0.66 µm from a hard coal combustion plant and 7.15 µm from a rock processing crusher plant, clearly illustrating that the emitted particles from combustion processes are finer than those from mechanical processes. Ohlstrom et al (2000) measured fine particle ($PM_{2.5}$) emissions from several types of boiler plants in Finland using an electrical low pressure impactor (ELPI) and showed that the particle size distributions varied with boiler size and particle separation devices. They reported a peak particle concentration at a size of about 0.1 µm. Morawska et al (2006) used a scanning mobility particle sizer (SMPS) to monitor particle emissions in the stack of a shale facility in Australia and showed that the particle size distributions were bimodal with average modal count median diameters of 24

and 52 nm (Fig 4). This study also demonstrated that particle size distribution measurements have the potential to be used as source signatures of industrial emissions.

4.2.2 Motor Vehicles

In 2009, the European Union introduced a particle number limit to its Euro 5/6 emission standard for light-duty diesel vehicles. This is the first particle number standard introduced anywhere in the world. Particle number concentrations will be measured according to the PMP procedure, and emission factors will be limited to 5 x 10^{11} km^{-1}. Motor vehicle emissions often contain large numbers of volatile and semivolatile nanoparticles that are produced after emission from the tailpipe. The formation mechanisms of these nanoparticles, their associated health effects and concentrations are topics that are not well-understoof; as a result, the issue of whether these nanoparticles should be included in particle number standards has been the subject of much controversy in the recent past. In order to avoid this issue, the PMP procedure eliminates all volatile material from the particles, such that only solid particles are counted.

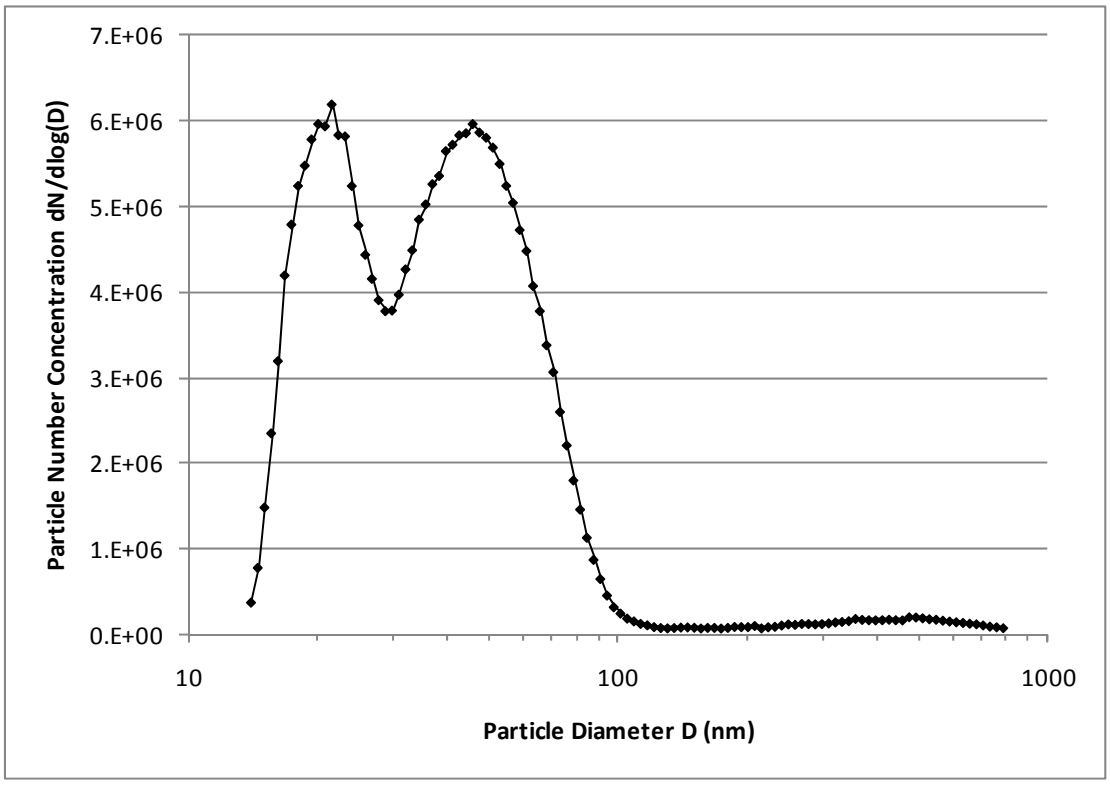

Fig 4: Particle size distribution measured in a shale facility stack by Morawska et al (2006).

4.3 Other Source Emissions of Particulate Matter

Whilst the contributions of vehicular combustion and industrial processes to overall particle emissions are significant, the majority of global emissions are ascribed to natural sources (Hinds, 1999). Obviously, it is not possible to directly regulate the latter; only those

anthropogenic emissions with which they may interact. Natural particle sources are distributed relatively evenly around the world, whilst anthropogenic emissions are largely confined to, and thus affect, populated areas (Hinds, 1999). Identifying the sources of particulate matter present in ambient air in these locations is not a straightforward process. Source apportionment of particulate matter seeks to identify individual source signatures amongst what Eastwood (2008) referred to as the "almost incomprehensible anarchy" that characterises the dynamics and origin of many aerosols. Chemical mass-balance approaches are typically implemented to achieve this, and these can be divided into source and receptor-oriented techniques. In their oft-cited study, Schauer et al. (1996) used organic compounds to trace the origins of fine particulate matter collected at various urban sites in the Los Angeles area during 1982. The authors found that the bulk of primary fine particles measured at their monitoring sites were due to diesel and gasoline combustion exhaust, meat cooking, wood combustion and dust emitted from paved roads (Schauer et al., 1996). These sources also contributed substantially to overall fine ($PM_{2.5}$) particle mass (i.e. primary and secondary); although their relative importance was different to that measured in terms of primary fine particles, and road dust, wood combustion and meat cooking often made a larger combined contribution to fine particle mass than diesel and gasoline exhaust (Schauer et al., 1996). Evidently, emission sources other than automobiles and trucks can constitute a significant proportion of urban aerosols, and some of the most common of these are introduced below.

4.3.1 Roads Dust and Wear Products

Particles emitted from the surface of both paved and unpaved roads can have diverse origins, and are attributable to the mechanical wear of both vehicle tyres and the road surface at their interface in addition to products of the wear-and-tear of vehicle components (e.g. brakes, clutch, catalytic converters and exhaust systems), natural materials (e.g. mineral dust and leaf litter) and deposited combustion particles. The particles released from road/tyre interactions may thus be primary, secondary or resuspended. The characteristics of particles released via such processes, and their relationship with vehicle (Gillies et al., 2005), tyre (Dahl et al., 2006; Hussein et al., 2008) and road (Claiborn et al., 1995) parameters, in addition to the development of emission factors, is gathering momentum as a research topic. Given the potentially large contribution of road dusts (especially for unpaved roads) to ambient particle mass concentrations and their potential effects on human health (Miguel et al., 1999), such investigations are critical for emission assessment, modelling and control. Ultrafine particle release from certain road/tyre interactions have been shown to be substantial (Dahl et al., 2006), and as noted by Morawska et al. (2008), this is a topic requiring further systematic research.

4.3.2 Aviation and Shipping

Non-road transportation modes, of which aviation and shipping are the most dominant in most locations, have come under increasing scrutiny with respect to their particulate emissions in recent times. Both involve the use of large combustion engines operating over extended periods at varying loads during transit and when at air or sea ports. This can have an appreciable impact on ambient particle concentration. For example, based on a study in the Mediterranean, Viana et al. (2009) estimated that proximate shipping activities contribute 14% of annual average ambient $PM_{2.5}$ mass concentration in the port city of Melilla. Global annual particle mass emissions caused by shipping have been estimated to lie between 0.4 and 3.4 Tg (Eyring et al., in press), which marks them as a significant contributor to global

anthropogenic particle emissions. Particle number emissions from shipping activities may also have a significant effect, and Sinha et al. (2003) estimated that global annual emissions of particles greater than 3nm attributable to ocean-going ships were approximately 25% of those caused by biomass burning; which is itself a major source of particles. Notwithstanding the often substantial uncertainties inherent in emission estimates, shipping activities have the potential to have a measureable impact on particle mass and number concentration at a range of spatial scales. It is important to note, however, that in areas with multiple strong particulate sources (i.e. many urban areas), the relative contribution of shipping activities is likely to be relatively minor (Minguillón et al., 2008). It is equally important to consider, that, despite this, the mobile nature of the emission source coupled with the fact that the overwhelming majority of ship emissions occur within 400km of land, gives rise to a scenario where shipping may affect air quality at a multitude of coastal locations (Eyring et al., in press). Particulate emissions from ships are not directly regulated; although progressively more stringent fuel sulphur content restrictions will be adopted over the next decade, and these are expect to substantially reduce particle emissions (Lack et al., 2009).

Emissions due to aviation (which primarily relies on kerosene fuels) can have various impacts depending upon the spatial and temporal scale of interest. There are numerous research foci regarding the effects of aviation particle emissions on planetary radiative forcing and climate phenomena (Lee et al., in press), and given the ever-increasing popularity and volume of civil aviation flights, this topic is very likely to come under even greater scientific scrutiny in the future. However, from the perspective of human health impacts, it is ground based aircraft activities (idling, taxiing and take-off) that are of paramount significance. Recent studies have indicated that particulate emissions due to ground-level such procedures can cause particle concentrations substantially elevated above background levels, especially in terms of UFPs (Westerdahl et al., 2008; Mazaheri et al., 2009; Hu et al., 2009). Aircraft particulate emissions are not directly regulated.

4.3.3 Miscellaneous Fossil Fuel Combustion

There are many non-vehicle fossil fuel combustion sources that can potentially impact upon ambient particulate concentration. Two and four-stroke garden implements (lawn mowers, edge trimmers, leaf blowers, chainsaws etc.), generators (both diesel and petrol) and non-road recreational sources such as outboard and inboard marine engines and snowmobiles, to name but a few, are all sources of particulate emissions. Additionally, in some developing countries, two-stroke road vehicles dominate transport fleets in urban areas (Eastwood, 2008). Two-stroke combustion has traditionally been regarded as an emission-intensive process; owing to the large fraction of fuel and lubricant that escapes complete combustion, and despite recent technological developments that allow newly-built two-stoke engines to conform to increasingly stringent HC emission standards in developed countries, the developing regions where two-stroke combustion predominates are unlikely to utilise engines fitted with such control mechanisms (Eastwood, 2008). Particulate emissions from non-road two and four-stroke engines are not regulated, although HC and NO_x emissions are in many developed countries, and it follows that regulation of these parameters will lead to associated reductions in primary and secondary particulate emissions from these sources.

In addition to its widespread use in power generation, coal is also used for heat generation in residential settings across many countries, as too are liquid and gas-phase fossil fuels (Lighty et al., 2000). Such practices can affect ambient air quality, and Junninen et al. (2009)

estimated that up to 58% of PM_{10} concentrations measured in the Polish town of Zakopane were due to residential heating.

4.3.4 Cooking

Although primarily conducted indoors, certain cooking processes leave an observable signature in outdoor air and contribute to ambient fine particle mass (Schauer et al., 1996). Frying, grilling and barbecuing are the techniques most likely to generate significant particle emissions, given the high temperatures often involved. There are numerous factors affecting particle emissions due to cooking, such as food type, cooking technique, temperature, the presence and amount of oils and type of heat source (i.e. electric, gas or other); although under many permutations of these factors, the overwhelming majority of emitted particle numbers fall within the ultrafine size range (Wallace et al., 2004; Buonanno et al., 2009). The bulk of solid fuel use (fossil and biomass) for cooking and heating occurs primarily in developing world, and there are numerous attendant health implications of this practice (Smith et al., 2000). Although a significant source of ambient particles, the ethical and logistical issues associated with implementing any meaningful regulations regarding how people and businesses cook food within their own properties are numerous, such that regulation of this source seems highly unlikely in the short term.

4.3.5 Biomass Combustion

Anthropogenic biomass combustion, whether for the reduction of seasonal fire fuel loads or residential purposes, can be a major contributor to ambient particle concentration (Wardoyo et al., 2006). Under certain conditions, a substantial proportion of PM_{10} and $PM_{2.5}$ can be ascribed to wood-fired domestic heating in some cooler climates (Luhar et al., 2006; Ries et al., 2009). The emissions of fine and ultrafine particles from biomass combustion are not well-characterised, however, somewhat similarly to emissions from cooking, appear to be strongly dependent on biomass type, section (e.g. wood or leaves/branches) and combustion temperature (Wardoyo et al., 2006). Wood heater and fuel manufacturers in some countries are required to conform to allowable levels of particulate mass emission per unit mass of fuel burnt. Particle mass emissions from other types of biomass combustion are regulated indirectly in some locations via qualitative guidelines, such as limiting the number or extent of fires or not permitting biomass burning under certain meteorological conditions. Particle number emissions from biomass combustion are unregulated.

4.3.6 Miscellaneous Sources

The scope of this section has been limited to an introduction to the most widespread anthropogenic particle emission sources. There are multiple additional sources of primary and secondary particulate sources, including, but not limited to: waste incineration, construction, agricultural processes, cigarette smoke and fugitive emissions.

5. Other policy measures

The reduction of human exposures to particles as a preventative health care measure can have diverse applications in numerous environments. However, for the purpose of providing an example of wide relevance, the indoor environment of residential and commercial buildings located proximate to major roadways will be used as a case in point.

It is clear that, in terms of number, the overwhelming majority of freshly emitted particles from vehicular combustion reside in the ultrafine size range. The instability of many of these particles results in their relatively rapid transformation into a larger size fraction with an appreciable and readily-measured mass. How the variable time and spatial scales involved in these processes relate to human exposure to particles near this source was poorly understood until the first few years of the 21st century, when two separate but equally comprehensive studies were reported, based on measurements of the relationship between particle concentration and distance from roadways under various wind conditions (Sioutas et al., 2005). These studies, performed in Australia (Hitchins et al., 2000) and the USA (Zhu et al, 2002a,b), highlighted that ultrafine particle concentrations do not reach concentrations comparable to background until a distance of at least 300m cross or downwind of major roads is reached. By comparison, these studies also noted that fine particle mass concentrations exhibited relatively small decay with distance from roadways.

The work of Hitchins et al. (2000) and Zhu et al. (2002a,b) have clear implications for ultrafine and fine particle exposure reduction as they relate to persons living and working near major roads. Depending on geographic location, residential dwellings employ either natural ventilation, or a combination of natural and mechanical ventilation. In the former case, particle ingress from outdoors to indoors via open windows, doors and through infiltration pathways occurs relatively unimpeded for accumulation mode particles, while ultrafine and coarse particles enter with reduced efficiency due to their increased susceptibility to removal mechanisms. However, where ventilation pathways are large, as is the case in most naturally ventilated buildings, the reduction in concentration of particles is typically minimal (Liu and Nazaroff, 2003). Accordingly, constructing naturally ventilated dwellings proximate to major roads and freeways (especially within ~150m), or vice-versa, is likely to lead to significantly increased particle exposure for the occupants. In the case of mechanically ventilated offices and dwellings, the factors influencing exposure to outdoor particles are the particle removal efficiency of the heating, ventilation and air conditioning (HVAC) system and its associated filtration devices, and the rate at which outdoor air is delivered to the building. Most standard filtration media exhibit minimum capture efficiency for accumulation mode particles for the reasons outlined previously, and a variety of operational factors, such as dust loading and air flow rate, can further influence their efficiency (Hanley et al., 1994). The ability of the HVAC system and standard filters to reduce particle number concentrations in air delivered to commercial buildings located near major roads was shown by Jamriska et al. (2000) to be fair at best. Additionally, the height at which air is drawn into the building from outdoors can be an important determinant of particle concentrations contained in incoming outdoor air (Hitchins et al., 2002). Although occupants of mechanically ventilated buildings located near busy roads will be afforded greater exposure reduction than those in naturally ventilated buildings, the best exposure reduction policy is to separate, at the planning stage, buildings and major roads by as large a distance as practicable, preferably by 150m or more.

6. Summary and Conclusions

The aim of this work was to review the existing regulations and policy measures related to reduction of airborne particulate matter, which is perhaps the most complex airborne pollutant to understand and manage. This is because there are many different physical and chemical (as well as microbiological) properties of the particles. This complexity is

amplified by difficulties associated with simultaneous measurement of these properties and the lack of clarity regarding which are the most significant in terms of their impact on human health and the environment, and, therefore, which should be the prime target of regulations. The complexity starts when we attempt to measure one of the basic property of the particles, their size, to find out that it spans over five orders of magnitude, which is a too large range for any single measuring technique; consequently, there are only a few existing standard techniques with which to measure certain particle characteristics.

Health guidelines, the most important tool in developing national regulations, exist only in relation to one type of particle matter characteristic; its mass concentration in two size ranges: $PM_{2.5}$ and PM_{10}. Over the last few decades the studies have shown that it is the smaller particles, which potentially have higher health impact, and therefore there has been a trend in lowering the size of regulated particles. A good example of this trend is the US, where initially TSP was regulated, and was eventually replaced by the PM_{10}, with $PM_{2.5}$ regulations introduced later. It has been shown that within the current range of concentrations studied in epidemiological studies there are no threshold levels and that there is a linear exposure-health response relationship. Based on this, in the most recent review the World Health Organization Air Quality Guidelines, a new set of guidelines for particulate matter was introduced, with annual mean values for $PM_{2.5}$ and PM_{10} of 10 and 20 $\mu g\ m^{-3}$, respectively. This was based on an American Cancer Society study (Pope et al. 2002) and represents the lowest end of the range over which significant effects on survival have been observed (WHO 2005). It is important to note that these values are not much higher than the concentration levels encountered commonly in natural environments (while it should be acknowledged that, in some locations and under some circumstances, concentrations in natural environments may be well below or above those cited). If future epidemiological studies report response at lower concentration levels $PM_{2.5}$ and PM_{10}, it is likely that the guideline values will be lowered even further.

While there is a large body of toxicological evidence of the potential detrimental health impacts of UF particles measured in terms of number concentration and surface area, epidemiological studies have not yet provided any quantification of exposure – response relationships. Therefore, there are no scientific bases for setting particle number or surface area health guidelines. It has been shown in this review that vehicles are significant sources of UF particles, and it is the vehicle emissions that are commonly the most significant source of air pollution in most populated urban areas. It is therefore of particular significance to understand the magnitude and characteristics of vehicle-generated UF particles in urban air, as it is this environment which is the most likely to be considered as a target for future air quality regulations in relation to particle number. Industrial and power plant emissions have a significant impact on the environment and climate, but as they often (but not always) occur outside the most populated urban settings, their direct impact on human exposure is lower than the impact of vehicle emissions.

Despite the limitations in epidemiological studies, from atmospheric studies a picture starts emerging regarding particle number concentration levels and their spatial variation; both of significance when considering human exposure. It has been shown that the clean background levels are on average of the order of $2.67 \pm 1.79 \times 10^3\ cm^{-3}$, while levels at urban sites are 4 times higher and levels at road, roadside, street canyon and road tunnel sites are 16, 18, 27 and 64 times higher, respectively. Thus, the range of concentrations between clean and vehicle-affected environments spans over two orders of magnitude. This is very different from particle mass; for example a review by Morawska (2003) showed the decrease in mass

concentration between a busy road and urban background ranges only from 0 to about 25–30%. This large variation in particle number concentration across different environments has profound significance in relation to human exposure assessment and epidemiological studies. This means that unless exposure assessment is conducted where exposures occur and at time scales that elucidate the temporal nature of the exposure, it is unlikely that epidemiological studies would provide answers based only on monitoring in central locations. In other words, central monitoring alone underestimates exposures and may lead to inappropriate management of public health risks.

As discussed above, the current state of knowledge on exposure-response relationships provides basis only for particle mass concentration health guidelines, and as a result, there are currently standards for mass only.. PM_{10} includes $PM_{2.5}$, currently discussed in the US is the combination of $PM_{2.5}$ and $PM_{10-2.5}$ (fine and coarse) standards, which enable more efficient regulations of particles generated from different sources. Even more efficient would be a combination of PM_1 and PM_{10} standards in controlling combustion related ambient particles, as well as those originating from mechanical processes, however, the very limited body of epidemiological studies related to PM_1, coupled with the technical challenges of PM_1 measurements, make this combination unlikely to be considered in a foreseeable future.

While lack of UF particles exposure response relationship makes it impossible to propose health guideline for UF particles, it is important to point out that as discussed above, the current levels in environments affected by vehicle emissions are over an order of magnitude higher than in the natural environments. Thus, if there is also no threshold level in response to exposure to UF particles, as it does not relate to $PM_{2.5}$ or PM_{10}, future control and management strategies should target a decrease of these particles in urban environments to bring them closer to the background levels. This means that in many urban areas the concentrations should be lowered by one two orders order of magnitude to achieve this.

It is important to keep in mind, that, unlike those relating to particle mass concentration ($PM_{2.5}$, PM_{10}), there are no standard methods for conducting size classified particle number measurements. Additionally, the number concentrations reported depend on the instrument used and its setting. This indicates the need for the development and utilization of standardised measurement procedures, thus enabling meaningful comparisons between the results of different studies. This is of particular significance for human exposure and epidemiological studies, and also regarding any future regulations of UF particles.

One additional issue in relation to UF particles is that secondary particle formation results in rapid increase of particle number concentrations by one to two orders of magnitude to concentration levels of the order of 10^5 particles cm^{-3}. This would present a challenge if there were future regulations considered based on particle number. Issues to resolve would include: (i) whether the regulations should be set around the base line concentrations without the peak concentrations, or whether they should include the peaks; and (ii) how the peaks are to be defined. Developing a much better picture of particle formation dynamics in different environments, including those which are influenced by traffic, would greatly assist the formulation of such regulations.

In contrast to the above, there are cleanroom particle number concentration standards. Within the context of this review, the key point to consider is that particle number concentrations within cleanrooms are tightly regulated by virtue of the specific criteria with which they must comply to secure and maintain a given classification. This stands in great contrast to outdoor

environments worldwide, where no regulation policies are in effect. Many questions are raised by this apparent discontinuity, and the casual observer is permitted to wonder why such well-defined measures are in place to quantify and assess particle number concentration in the specialised indoor environment that cleanrooms represent, yet any discussion of applying similar approaches to outdoor environments is often subject to consternation and contention. The answer to this is not simple, and a topic unto itself. However, the complex and variable nature of particle emission sources outdoors, coupled with the vicissitudes of particle generation processes and behaviour (especially towards the smaller end of the measureable size spectrum), can be held partly accountable. .

Motor vehicle emission standards are derived on the basis of factors such as technological capabilities of engine design and after-treatment devices and the impact of emissions on the environment and ambient air quality standards. Emission standards and test procedures are well stipulated for both heavy-duty and light-duty vehicles in the US and in Europe. These standards are generally described in terms of total particle emission, rather than PM_{10}. Emission test procedures are not the same across different countries, and many types of drive cycles are used. Therefore, even when expressed in the same units of measure, engine emissions measured on different test cycles may not be comparable. Several countries, such as Australia and India, have adopted standards based on the Euro standards. In general, Euro standards are more stringent than enforced in the USA.

With improvements in technology and more rigorous restrictions on air quality, motor vehicle emission standards have been getting tighter over the years. For example, between 1992 and 2008, European particulate mass emission standards for heavy-duty diesel engines have been reduced by an order of magnitude. But the most significant change occurred in 2009, when the European Union introduced a particle number limit of 5×10^{11} km^{-1} to its Euro 5/6 emission standard for light-duty diesel vehicles. In response to these increasingly stringent standards, manufacturers have been forced to consistently improve engine design and fit more sophisticated emission control devices and systems to vehicles.

Another important group of emission standards are those related to industrial emissions. There are standards in place restricting particle emissions from coal-fired power plants targeting different rated thermal inputs, as well as different age of the plants. Other industrial sources such as asphalt and cement manufacture, rock processing, fertilizer, chipboard, steel and other metal production plants usually are regulated by their specific emission standards, vary from country to country, and are sometimes specified for individual stages in the manufacturing processes. It is important to note that all these standards refer to total emissions of particulate matter, and not size specific emissions (such as $PM_{2.5}$ or PM_{10}). This constitutes a clear disparity between emission and air quality regulations, with the later being size specific. In relation to particle number emission from industrial sources, there is very little knowledge on this, and in turn, no regulations.

There are many other sources of airborne particulate matter; some of the most significant of these include aviation, shipping, biomass combustion, waste incineration, construction, agricultural processes, cigarette smoke, fugitive emissions, cooking and non-vehicle diesel, two, and four-stroke combustion None of these sources are directly regulated in terms of particle emissions.

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