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Article

Challenges and Approaches for Developing Ultrafine Particle Emission Inventories for Motor Vehicle and Bus Fleets

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Abstract: Motor vehicles in urban areas are the main source of ultrafine particles (diameters < 0.1 μm). Ultrafine particles are generally measured in terms of particle number because they have little mass and are prolific in terms of their numbers. These sized particles are of particular interest because of their ability to enter deep into the human respiratory system and contribute to negative health effects. Currently ultrafine particles are neither regularly monitored nor regulated by ambient air quality standards. Motor vehicle and bus fleet inventories, epidemiological studies and studies of the chemical composition of ultrafine particles are urgently needed to inform scientific debate and guide development of air quality standards and regulation to control this important pollution source. This article discusses some of the many challenges associated with modelling and quantifying ultrafine particle concentrations and emission rates for developing inventories and microscale modelling of motor vehicles and buses, including the challenge of understanding and quantifying secondary particle formation. Recommendations are made concerning the application of particle emission factors in developing ultrafine particle inventories for motor vehicle fleets. The article presents a précis of the first published inventory of ultrafine particles (particle number) developed for the urban South-East Queensland motor vehicle and bus fleet in Australia, and comments on the applicability of the comprehensive set of average particle emission factors used in this inventory, for

developing ultrafine particle (particle number) and particle mass inventories in other developed countries.

Keywords: buses; emission factors; mobile sources; motor vehicle emission inventories; on-road vehicles; particle mass; particle number; particulate matter; traffic; ultrafine particles

1. Introduction

UFP (ultrafine particles) have diameters less than 0.1 μm and are generally measured in terms of particle number due to their insignificant contributions to particle mass and prolific numbers. They have the ability to enter deep into the human respiratory system and contribute to negative health effects, making them a pollution source of particular interest. As the size of a particle decreases, its biological interaction in the human respiratory tract increases [1] and particles with diameters less than 5 μm which are inhaled can reach a large proportion of alveoli in the human lung [2].

Many studies have shown that motor vehicles are the main source of ultrafine particles in urban areas [3-7] and for this reason ultrafine particles continue to be the subject of ongoing research in urban areas. However, as yet only limited data on long term trends in concentrations of ultrafine particles is available [8].

A pollutant plume of vehicle emissions is highly dynamic and consists of a reactive mixture of particles and hot gases. As the plume is diluted by ambient air its physical characteristics and chemical composition are significantly altered. New particles are formed through nucleation and existing particles grow from the condensation of semi-volatiles within seconds of dilution [8,9]. The particle distribution continues to evolve in the near-road environment by the evaporation and condensation of condensable material [9]. Hence emission factors derived immediately upon particle plume formation are likely to exhibit different characteristics to those measured some period of time after emission, and will also be subject to physical and chemical influences at the measurement site.

Nanoparticles are a subset of UFP. These particles have diameters less than 0.05 μm and, like ultrafine particles, their airborne concentration is generally measured in terms of particle number concentrations per unit volume of air [8]. Close to traffic sources, nanometer sized-particles dominate total particle number concentration and these decrease with distance from the traffic source [10,11].

Although substantial toxicological evidence exists of the harmful effects of exposure to ultrafine particles, current epidemiological evidence is insufficient to enable a conclusion to be reached on an exposure-response relationship [12], impeding development of ambient air quality standards. Hence detailed epidemiological studies are needed of the effect of ultrafine particle exposure on human health, and these studies will be complemented and informed by the development of motor vehicle inventories and measurement campaigns of ultrafine particles.

Most motor vehicle particle emissions are in the ultrafine size range; and ultrafine particles often have a greater biological effect than an equal mass of larger-sized particles comprised of the same substance [13]. Ferin *et al.* [14] in their study of the increased pulmonary toxicity of ultrafine particles found these smaller particles can evoke higher inflammation and overall toxicity. Exposure to fine

particulate matter (particles with aerodynamic diameters less than 2.5 μm) have been found to have adverse effects on cardiopulmonary health, and many population studies have linked exposure to fine particulate matter with increases in hospital admissions and a range of cardiovascular and respiratory diseases and mortality [15].

An association has been implied between particulate matter exposure and effects such as lung cancer [16] and heart attacks [17]. Research has shown that particles can even enter the bloodstream and reach the brain [18]; and there are some indications they can induce inheritable mutations [19].

UFPs are not currently regulated by ambient air quality standards, however modelling and quantification of these particle emissions have been undertaken in a number of microenvironments influenced by traffic emissions, such as near major roads, at busways and in tunnels and street canyons, as well as in laboratories in dynamometer studies. Factors researchers need to consider in these studies range from the structure of local microenvironments and meteorological effects for studies conducted in outdoor locations, to issues such as the dilution of vehicle aerosol in laboratories. Monitoring campaigns of ultrafine particles (particle number) can be complex to carry out, and the equipment needed for measurement is expensive, and so these factors remain major obstacles to monitoring ultrafine particles [1]. Hence such campaigns tend to be short term in nature.

This article discusses a number of current challenges researchers are facing in the development of motor vehicle and bus emission inventories and in measurement campaigns of UFPs, and makes some suggestions concerning the development of ***regional inventories*** (which utilize average emission factors) and ***microscopic inventories*** (which consider speed and vehicle-mode related emission factors and characteristics of microenvironments). The first, and only, ultrafine particle (particle number) inventory of tailpipe particle emissions published in the international literature is also précised. This inventory was prepared for the urban South-East Queensland motor vehicle and bus fleet in Australia [20-22] and derived a comprehensive set of average particle emission factors which have application for use in developing ultrafine particle (particle number) and particle mass inventories in other developed countries.

2. Methods and Models

The analysis in this work comprises two parts. Firstly, relevant findings on modelling and quantifying UFP emissions generated by motor vehicles are discussed; including those identified in an extensive review and analysis of the characteristics, ambient processing and implications on human exposure of ambient nanoparticle and ultrafine particles by Morawska *et al.* [8]. Secondly, issues relating to the development of an UFP inventory for a motor vehicle and bus fleet in Australia are discussed. This inventory combined knowledge from two distinctly different disciplines, from aerosol science and transport modelling [20-22]. Current progress in the development of emission inventories for bus fleets is also discussed. The focus of this work is to relate the findings of the review and inventory development to current issues facing researchers and practitioners developing inventories and conducting measurement of ultrafine particles for vehicles and bus fleets, and to present some possible approaches for developing UFP inventories for consideration.

3. Results and Discussion

This section discusses (i) important factors influencing quantification of UFPs generated by motor vehicles and buses and (ii) issues related to the development of an UFP inventory prepared for an urban vehicle fleet in urban South-East Queensland, Australia, to identify current issues facing the development of UFP inventories and measurement campaigns and to discuss some possible approaches for inventory development.

Three classifications commonly used to describe atmospheric aerosol size distribution in terms of modal diameters are nucleation mode (<0.1 μm), accumulation mode (0.1–1 μm) and coarse particle mode (>1 μm) [23]. These modal classifications are not strictly defined upon particle size, but are based primarily on the production mechanisms [23]. Recent studies with instruments extending the small size limit to 0.003 μm have shown that the nuclei mode needs to be separated into a nucleation mode (<0.01 μm) and an Aitken nuclei mode (0.01–0.1μm) [24]. In this paper, the term nuclei mode will be used to refer to the particle mode size range <0.1 μm.

The term mode can also be used to describe a peak in the lognormal function of the mass or number distribution of an atmospheric aerosol [25]. An extensive worldwide review of modal locations within particle number size distributions in anthropogenic environments revealed that modes in the ≤0.05 μm size ranged from 0.008 μm–0.05 μm for suburban and urban environments, and for the >0.05 μm ≤0.1 μm size range modes were between 0.050 μm–0.065 μm in suburban, traffic and urban-influenced environments, and at around 0.08 μm in urban and traffic-influenced environments [8,20]. Additional modes were found in suburban environments in the range 0.092 μm–0.1 μm [8,20].

The focus of this paper is on ultrafine particles (diameters less than 0.1 μm) which reside in the nuclei mode; and in this mode even greater numbers of particles may be found in the nanoparticle size range (< 0.05 μm). In many cases, the nuclei mode composes over 90% of the total particle number of diesel exhaust, while in other cases they may not be detected [26-28].

3.1. Factors Influencing Development of UFP Inventories and Measurement Campaigns

The primary consideration in developing inventories and measurement campaigns is to quantify or estimate UFP concentrations or emission rates to assess potential human exposure. Research objectives may focus on exposure as it relates to sensitive populations, such as young children at their schools, or the aged or sick housed in facilities located close to busy roads. Alternatively, studies may focus on exposure of the general public, e.g., public transport commuters or residents living close to busy traffic routes. Hence potential population exposed, study location, meteorological conditions, distance from the traffic source, and the temporal and spatial distribution of exposure are major study design considerations. Researchers need to consider many important factors, and a number of current issues with respect to quantification and modelling of UFPs are discussed below.

3.1.1. Lack of consistency of measuring equipment

Currently no standard methods exist for carrying out size-classified measurements of particle number, and a review of differences found between instrumentation measuring ultrafine particles may

be found in Morawska *et al.* [8]. Such lack of standardization has impeded progress in developing particle number standards.

Most on-road studies of vehicle-source ultrafine particles relate to average driving or steady-state conditions [29,30]; and difficulties can be experienced when attempting to replicate real-world emissions in chassis dynamometer studies in laboratories. For example, in laboratories, measured nuclei mode concentrations are usually substantially lower than those measured in real-world conditions, by around a factor of 2–10, and these are partly explained by parameters relating to dilution [31]. These observed differences provide critical challenges for researchers undertaking dynamometer measurements to derive emission factors, and may lead, in some cases, to an underestimation of particle number emissions, particularly in the nanoparticle size range.

Instrumentation can be set with different lower size window settings, based on instrumentation capability and operator choice, which may mean that the full size range of nanoparticles (*i.e.*, 3 nm–50 nm) is not being measured, leading to differing results. For example, an extensive review by Keogh *et al.* [22] which identified 156 emission factors in the international published literature for particle number found statistically significant differences between mean emission factors derived from Scanning Mobility Particle Sizer (SMPS) measurements, compared to Condensation Particle Counter (CPC) measurements (2 and 23 $\times$ 10^{14} particles per vehicle per kilometre, respectively). The SMPS focuses on estimating particle size distribution (as opposed to total particle count) and does not measure the lower size range of the nuclei mode, less than 0.01 μm, its lower size window is generally set higher than it is for the CPC, usually in the range 0.01–0.02 μm, as compared to the CPC which is usually set in the range 0.002–0.01 μm, which means that generally the CPC measures the lower size range of the nuclei mode and the SMPS does not [22].

For example, Rönkkö *et al.* [30] concluded that instruments that only measure down to 0.01 μm could miss or seriously underestimate the lower size range of nuclei mode particles. Studies which have reported particle number emission factors for nanoparticle and ultrafine particle size ranges for heavy and light duty vehicles per vehicle per kilometer using SMPS and Differential Mobility Sizer (DMPS) [32-35] show that particle number emission factors tends to be higher in the size range < 18 nm as compared to emission factors derived for the 18–50 nm and 18–100 nm size ranges, highlighting the importance of measuring particles in the < 18 nm size range [8,20].

These findings referred to above highlight differences in sets of instrumentation commonly used to measure UFPs, and point to the possible underestimation of the lower size range of nuclei mode particles depending on the setting of the lower size window.

The review of particle number emission factors in the international literature by Keogh *et al.* [22] found no statistically significant differences between the means of particle number emissions for studies conducted in Australia, Austria, Germany, Sweden, Switzerland, United Kingdom and the USA. This finding lends weight to the relevance and applicability of particle number emission factors derived in the statistical analysis by Keogh *et al.* [22] for use in developing inventories and air quality assessments in developed countries. The review found few speed-related emission factors for particle number, and derivation of such emission factors are needed for modelling motor vehicle and bus fleets traveling at lower speeds, such as under congested conditions.

Spark ignition gasoline and compression ignition diesel engines have very different combustion environments that yield large differences in the chemical composition, size distribution, and numbers

of ultrafine particles. The particle number emission rates for heavy-duty diesel vehicles are one to two orders of magnitude higher than gasoline-fueled passenger cars [8]. In urban areas heavy duty diesel vehicles, including buses, are major sources of exposure for UFP. However, Keogh *et al.*'s [22] review found no relevant emission factors for buses for particle number where instrumentation had estimated total particle count down to the smaller size range of 3 nm.

Future studies are needed to provide more robust estimates of UFP emission rates from individual vehicle classes and instrumentation methods for measurement.

3.1.2. Particle dynamics and secondary particle formation

Most primary particles generated by the engine range in size from 30–500 nm and reside mainly in the accumulation mode, with secondary particles generally in the nanoparticle size range below 30 nm (in the lower size range of the nuclei mode), and particle number concentration in the accumulation mode tends to be substantially more stable and predictable than in the nuclei mode [8].

Nuclei mode particles are formed from the condensation and nucleation of volatile species upon dilution and cooling of the exhaust, and their formation is highly dependent on dilution conditions [27,36]. Because nuclei mode particles form upon dilution, they can also be referred to as secondary particles. The formation of secondary particles is a complex process that relies on the dynamic interactions with other particles and multiple aspects of dilution conditions, including dilution rate, temperature, residence time, and relative humidity. The nuclei mode particles are composed of semi-volatile organics, sulfates, and in certain conditions can be composed of solid particles such as metallic ash [29]. Regardless, due to the small size of the particles, the nuclei mode only constitutes a minor fraction of the total particulate mass from diesel engines [37]. However, due to the very significant contribution of nuclei mode particles in terms of particle number to ambient airsheds it is vital these sized particles be characterized as part of emission inventories.

Secondary particles in the nanoparticle size range have been found near busy roads, particularly those with large numbers of heavy duty diesel vehicles [38-46], as well as in on-road studies, e.g., mobile on-road laboratories [11,29,47-51]. However, individual UFP measurements from dynamometer tests may differ substantially from road-side and on-road measurements. Secondary particles can sometimes be present, but are not commonly found in dynamometer measurements that use dilution tunnels to dilute and cool exhaust gases [28,52-55] which has implications for the use of dynamometer-derived emission factors in inventories quantifying secondary particles.

Kittelson *et al*. [55] and Rönkkö *et al*. [50] measured UFP emissions from individual diesel vehicles in both on-road emission studies and using chassis dynamometers equipped with artificial dilution tunnels. Both studies consistently measured high concentrations of nuclei mode particles below 0.02 or 0.01 μm that had peak concentrations one to two orders of magnitude higher than the accumulation mode particles. By adjusting the dilution ratio and residence time these researchers were able to replicate on-road particle size distribution measurements. Nevertheless, the lower size range of nuclei mode for particles tended to be underestimated, occasionally by an order of magnitude, when comparing laboratory measurements to on-road measurements [50,55].

The estimation and quantification of secondary particles in motor vehicle emission inventories presents an interesting and complex challenge for researchers, as does the task of measuring secondary

particles in dynamometer studies. To develop a method that is comparable and reproducible in different laboratories located around the world the UNECE-GRPE Particulate Measurement Program (PMP) was formed and, based on their recommendation, a particle number limit has been added to the proposed Euro 5/6 emission standards for light duty vehicles, whereby volatile material is removed from a sample in accordance with the PMP procedure and only solid particles are counted [8]. The PMP removes the volatile fraction of the exhaust, which is responsible for the formation of the nuclei mode particles, in order to maintain repeatable results [8].

Real-world UFP inventories should account for the true magnitude of nuclei mode particles that exist in the exhaust plume from vehicles driving in real-world conditions, however such measurement methods are still evolving. No protocol has been established for measuring nuclei mode particles and measurements can be substantially different based on sampling conditions and the sampling equipment. When using UFP measurements, investigators should be attentive to the details of the experimental design (*i.e.*, sampling equipment, dilution systems, and ambient conditions) which drive the formation mechanisms for secondary particles. It has been suggested that using source emission factors for estimating pollutant concentrations in ambient areas will likely reflect primary particle emissions and that secondary particle formation needs to be predicted [8].

3.1.3. Roadside distance and dilution conditions

Many vehicle-related and meteorological factors can influence the results of on-road particle emission studies. Vehicle emissions consist of hot gases and particles which are highly dynamic, and the prevailing wind is the major determinant in relation to the direction and speed of the plume of pollutants away from the emission source [8]. This can be a very complex phenomenon to model and quantify, particularly given the multiplicity of variations in the micro and macro environment structures of areas, their topography, climatic and weather conditions and vehicle mixes.

For example, under specific night-time and winter inversion conditions, the nuclei mode can persist and grow above 30 nm through vapor condensation [56], while in day-time conditions, the nuclei mode decays rapidly as the plume disperses in the near-road environment [57]. The nuclei mode particles evaporate or coagulate with larger particles leading to short residence time in typical atmospheric conditions [36]. Colder temperatures significantly increase the concentration of UFP in motor vehicle exhaust measured from the roadside [58], on-road [37] and on-board vehicle dilution tunnels [59].

Concentrations of UFPs generated by motor vehicle fleets tend to be higher within 100 metres of the emission source under typical meteorological conditions. Studies have shown that particle concentration decreases with distance from the road up to around 300 metres, beyond which size distributions and concentration levels are similar to local urban background [5,60-62]. A recent study of the evolution of particle size distribution and particle number concentration decay at different distances from a highway in Italy found their results similar to US and Australian studies, despite differences in diesel vehicle percentages and mean speed velocities [63].

The distance from a major roadway and traffic pollution source that children or adults with pre-existing conditions such as asthma, respiratory and cardiovascular disease are housed (or attend school for example) is important, as these populations may be more sensitive to the effects of particulate matter pollution. In the US environmental justice concerns have been raised because

minority and low-income populations are more likely to live in urban areas with high levels of motor vehicle traffic [64]. This situation is likely to be mirrored in other countries throughout the world.

Hitchins *et al.* [61] found when measuring total particle number concentration in the size range 15–697 nm that at around 100–150m from the road (where the wind below directly from the road) concentrations reduced to around half the maximum found at 15 m from the road (the closest point of measurement to the road). Zhu *et al.* [57] in their study of particle number concentration (6–220 nm) using a CPC and SMPS at a site close to a Los Angeles freeway found particle number concentration dropped substantially with increased distance from the road due to atmospheric dilution, dropping to around half the original value at 30 metres at around 90–150 metres, and that downwind of the freeway ultrafine particle concentration was similar to the background location measured 300 metres upwind of the study site.

The importance of distance from traffic source suggests that not only are regional airshed inventories of UFPs important to give a broad picture of particle number emissions in a region, but microscale inventories are also needed. Microscale inventories can be tailored to both sensitive populations and also the general population (particularly considering distance from heavy traffic sources for sensitive populations, especially where there are high concentrations of heavy duty vehicles) to account for the high concentration of particles adjacent to the roadways. Project-level inventories of UFPs for pre and post-new infrastructure construction would also be informative (e.g., construction of tunnels, busways, and transit oriented development sites near public transport) for understanding their public and environmental health impacts. Inventories of UFPs at various scales are also needed to inform scientific debate and development of ambient air quality guidelines and standards and which consider both primary and secondary particle formations.

3.2. Compiling UFP Inventories Using Emission Factors

Methods for compiling particulate matter inventories generated by motor vehicle fleets range from combining performance-related emissions factors with transport data, fuel consumption [65] or remotely sensed data [66]. Choice of approach is very often dependent upon available data, and the scale of inventory required for decision-making such as road-link, local, regional, state or country specific. As mentioned previously, conducting measurement campaigns of UFPs can be very expensive and complex, which can often prevent countries that do not have access to sufficient finding from being able to develop these inventories.

3.2.1. First published UFP motor vehicle emission inventory

Currently only one published inventory of ultrafine particles (particle number) generated by a motor vehicle fleet (which includes buses) exists in the international literature. This inventory was prepared for urban South-East Queensland (the study area was 4,600 square kilometers in the Brisbane metropolitan area), Australia [20-22]. The inventory quantified both ultrafine particles (particle number) and particle mass emissions for the motor vehicle fleet (for light and heavy duty vehicles and buses) for particle number (particles with diameters 3 nm–1 μm) and $PM_{1,}$ $PM_{2.5,}$ PM_{10} (particles with aerodynamic diameters less than 1 μm, 2.5 μm and 10 μm respectively).

The inventory developed for urban SEQ engaged a cost-effective approach for quantifying a comprehensive particle emission inventory covering the full size range of particles emitted by motor vehicles, and quantified emission rates for different vehicle types and road links. It combined relevant average particle emission factors with transport modelling data, and included 95% confidence intervals for each particle metric and vehicle type [21]. To develop the inventory, average particle number and particle mass emission factors (and their 95% confidence intervals) were derived using statistical analysis of more than 600 emission factors identified in the international published literature and combined with travel demand model data for the study region relating to 22,985 model links representing roads in the study region [for more detail [20-22].

To derive the average particle emission factors to use in developing the particle number (UFP) inventory, statistical analysis of published particle number emission factors was undertaken [22]. This statistical analysis included emission factor data from studies of different vehicle types under different measurement conditions (in laboratories on dynamometers, in tunnels and in the vicinity of roads) and studies where particles were measured down to 0.003 μm (the nuclei mode). Hence, the developed inventory quantified both primary and secondary particles. However, the specific focus of the inventory was not on secondary particle formations and it is likely that the inventory quantified a higher proportion of primary particles.

Study characteristics considered in this statistical analysis of published emission factors included country of study, study location (dynamometer, tunnel, vicinity of the road), road type, speed limit on the road, road class (classed according to speed limits of ≤60, >60, <80, ≥80 k/h), vehicle type, fuel type, drive cycle, average vehicle speed, engine power, size range measured and instrumentation used and for fleet emission factors (average number of vehicles per day and percent of heavy duty vehicles traveling in the fleet) [22]. The extensive review identified 156 particle number emission factors published in the international literature and statistical analysis revealed that 85% of the variation in these published emission factors could be explained by the study characteristics vehicle type and instrumentation used in the measurement campaign [22]. This finding reinforces the importance of choice of instrumentation used in measurement studies of ultrafine particles.

Scenario modeling was also undertaken in the urban SEQ inventory study to test the impact of shifts in travel modes in different travel times, average vehicle occupancy rates and anticipated future levels of particle emissions [21].

3.2.2. Relevance of the derived average particle emission factors used in the urban SEQ inventory for developing particle inventories in other developed countries

The average particle number and particle mass emission factors (and their 95% confidence intervals) derived in the statistical analysis by Keogh *et al.* [22] for different vehicle classes for particle number, PM_1, $PM_{2.5}$ and PM_{10} are considered suitable for use in other developed countries for quantifying particle inventories [20,22].

This assumption is based on the finding in the statistical analysis that relatively few statistically significant differences were found between the mean values of published emission factors for different particle metrics for different countries of study and study location (measured on a dynamometer, in a tunnel or in the vicinity of a road) [20,22]. Hence, the Keogh *et al.* [22] published average particle

emission factors for particle number and subsets of particle mass have particular application for regions that do not have particulate matter measurement data, have insufficient funding to undertake measurements, or where experimental data is available but is of insufficient scope.

3.2.3. Implications of the urban SEQ UFP inventory

Annual particle number emissions generated by the urban SEQ fleet were found to be 1.08×10^{25} (with 95% confidence intervals of 0.54–1.97) [21]. This quantification makes an interesting comparison when comparing it to findings of a study by Mazaheri [67] at the Brisbane international and domestic terminals which estimated annual particle number emissions due to aircraft operations (aircraft landing and takeoff cycles and aircraft engine ground running procedures) of 2.43×10^{24} per annum, thereby identifying airports as significant generators of UFPs.

A major finding of the urban SEQ inventory development was that although heavy duty vehicles contributed only 6% of total vehicle kilometers traveled in the study region, this vehicle class contributed more than half the region's particle number (ultrafine particle) and PM_1 emissions due to their high emission rates. This finding is significant as the SEQ region's freight task is expected to double in the next 20 years [68], and highlights the need to carefully monitor and regulate population exposure to busy routes used by heavy duty vehicles, especially where they are close to sensitive populations.

UFP emission inventories have the potential to provide impetus to drive emission reduction policies. For example, particle number emission factors for heavy duty diesel trucks or buses are generally one to two orders of magnitude higher than those for a typical petrol car [28,69-70]; and most heavy duty vehicles (HDVs) are diesel-fuelled. Diesel particle filter after-treatments have been reported to reduce UFP emissions by 90–99% [71-73]. These results are consistent for accumulation mode particles in laboratory and real-world testing. Instances can occur where diesel particle filters can produce higher particle number emissions through the formation of sulfate nuclei mode particles [74], however reductions have been observed both in laboratory [71-73] and on-road studies [74]. Future scenario modelling in terms of the particle number inventory for SEQ could model the estimated impact of future projected rates of diesel retrofits and vehicle replacements for the SEQ fleet.

4. Modeling Motor Vehicle UFP Emissions for Microscale Inventories

The SEQ UFP emission inventory quantified UFP emissions for three general vehicle classes—light-duty gasoline, heavy duty diesel, and buses (Compressed Natural Gas (CNG) and diesel-fuelled), and general operating conditions (in the vicinity of a road, steady state, and driving cycles on chassis dynamometers). To estimate microscale emission inventories to inform construction projects, UFP emission rates need to be modeled from specific vehicle classes and operating conditions. Several recent studies have developed UFP vehicle emission models to quantify the effect of vehicle technologies, vehicle operation, and other factors that influence UFP at the microenvironment, and these are discussed below.

4.1. Modeling UFP Bus Emissions

Conventional diesel buses can be important sources of UFP emissions due to their high UFP emission rates (compared to filter-equipped diesel buses), and their operation in highly congested and urbanized areas. Several studies which have developed particle number emission models for urban buses are discussed.

Holmén *et al.* [75] conducted the first known study that measured particle number emissions in real-world driving conditions using an on-board dilution tunnel. Four Connecticut Transit buses were studied over the course of a year in real-world driving conditions in Hartford, USA. Particle number emissions from the conventional diesel buses were highly dependent on the bus route and the driver [59]. A follow-up study found that three engine parameters (engine load, engine speed, and exhaust temperature) could be used to effectively model UFP emissions across road-way links in a wide-variety of driving conditions, including rural, urban, and restricted highway roads [76]. When engine operation is not known, vehicle speed and vehicle specific power are useful surrogates for engine speed and engine load, and that when only the average vehicle speed on the link is available, information on road grade can significantly improve predictions of particle number. Hence, information on road grade for different road links may substantially improve link-level inventories for areas with substantial elevation changes.

To better estimate the variability of particle number emissions within roadway links for hot-spot analysis, emission models can be developed that are sensitive to instantaneous vehicle emission mode (such as idling and acceleration).

A composite line source emission (CLSE) model developed by Wang *et al.* [77] quantified vehicle emissions in a traffic-influenced microenvironment. The spatial variability of motor vehicle emissions considered queuing, different driving conditions and multirepresentative segments for the capture of emission distribution in terms of real vehicle flow. The researchers applied this model to estimate particle number emissions at a two-directional busway used by CNG and diesel buses. They found acceleration distance was critically important in estimating particle number emissions. Higher particle emissions occurred where buses were accelerating, as compared to idling. By spatially allocating the emissions, the particle emissions were found to be 43 times greater at the front end of the platform as compared to the rear end.

The large influence of acceleration from CNG and diesel buses corroborate findings by Sonntag [78] on diesel Connecticut Transit buses. Sonntag [78] found particle number emission rates (particle number/second) were highest for the buses on high-speed freeway driving conditions. When the bus route was divided into 50-meter segments, the segments in which the bus emitted the most particles occurred near bus stops and intersections where the bus was accelerating from stop. Thus, UFP hot-spots in urban areas are likely occur in areas with high density of pedestrians and travelers. Although, acceleration events were the dominant sources of particle emissions for the bus, idling can become the dominant source if the bus were to idle for more than two minutes [78]. These findings have very important implications for busway design and waiting passenger exposure.

It is important to consider vehicle types in modeling microscopic UFP emissions. For example, CNG buses have significantly lower particulate mass emission rates than conventional diesel buses. Similarly, CNG buses have lower particle number emissions than diesel buses at low loads. However,

in their on-road and dynamometer studies Jayaratne *et al.* [79] found that during acceleration under heavy load, conventional diesel bus particle number emissions were an order of magnitude lower than CNG buses.

4.2. Implications for Future Microscale UFP Inventories

Microscale emission inventories have different data needs than regional emission inventories. For planning local transportation projects the accuracy of the final estimates may be less important than the sensitivity of the inventories to design factors. For example, if inventories are used to compare several roadway designs, the inventory would need to quantify the change in UFP emissions according to varying vehicle volumes and operating conditions. However, accurately estimating the effect of local meteorological conditions on the results may not be necessary. As in any modeling exercise, understanding the objectives in estimating UFP emission inventories is critical for producing achievable and usable results.

Recent research by Buonanno *et al.* [80] of particle number concentration in different urban microenvironments has confirmed that street geometry is the most important factor affecting concentrations, and that spatial variability of particle emissions has a large impact on exposure in microenvironments. Specifically, they found that the path chosen by pedestrians in urban microenvironments strongly influences exposure—those walking kerbside close to the emission source can be exposed to double the average exposure of pedestrians choosing a path close to buildings.

Thus, microscopic inventories can be useful tools to make inferences about the exposure of populations, and can be used to inform policies relating to roadway design (*i.e.*, placement of bus platforms), bus operation (*i.e.*, idling reduction strategies) and bus technologies (fuel technologies).

5. Conclusions

Motor vehicles and buses are a major source of UFPs in urban areas and these particle emissions pose health risks due to their ability to enter deep into the human respiratory system. More information is needed on UFP concentrations and emission rates in urban areas in the form of UFP inventories and long term monitoring data to inform urban planning, scientific debate and to develop relevant ambient air quality guidelines and standards to control this major pollution source.

This article has highlighted some of the complex factors that need to be taken into account in modelling and quantifying UFP inventories at various scales, and measurement studies of motor vehicles and buses.

The issue of specifically quantifying secondary particle formation in inventory development presents a major challenge for researchers and, in particular, for dynamometer studies which frequently underestimate the nuclei size range. Additionally, major differences can occur in measurements even amongst the same vehicle types.

Lack of standardization of measuring equipment for particle number emissions is a global challenge for developing particle number standards, and this study has reported that statistically significant differences have been found between measurements of particle number emissions by the SMPS and CPC instrumentation which require further investigation.

The relevant approach for developing UFP inventories is very dependent upon the objectives of the study, which can range from quantifying emissions at the microscale to regional or country scale, and the data and measurement instrumentation available.

For some inventories it may not be necessary to employ speed-related UFP emission factors and, in fact, few speed-related emission factors are currently available in the international published literature.

The high cost of conducting individual particle number measurement campaigns and acquiring the instrumentation needed in these campaigns pose as substantial impediments, and suggest that more cost-effective approaches are needed for developing UFP inventories, such as the ***regional inventory approach*** outlined below.

Based on the findings in this study, two suggested recommendations for developing UFP inventories in terms of choice of emission factors and approach include:

- The ***regional inventory approach*** (where ***average particle emission factors*** are used). Applications could include quantifying the impact of changes in vehicle fleets, such as retrofits, proposed introduction of new technology and alternate fuels, and changes in emissions resulting from shifts in travel mode choice (such as from passenger cars to buses and other forms of public transport).
- The ***microscopic inventory approach*** (where ***factors such as vehicle operating mode*** e.g., acceleration and ***characteristics of the microenvironment*** e.g., road grade are considered). Applications could include quantifying congested traffic conditions in different microenvironments (e.g., tunnels, freeways, canyons), and the effect of changes in vehicle flow rates and vehicle types as a result of major, new construction in study regions, such as the construction of new busways and highways, and investigating the exposure of specific population groups.

Suggestions for researchers in developing UFP inventories include:

- The ***regional inventory approach*** was used to develop the particle emissions inventory for the urban SEQ fleet to quantify annual fleet emissions for UFP (particle number), PM_1, $PM_{2.5}$ and PM_{10} précised in this article. This inventory highlighted the substantial contribution of HDVs in the region which, although only contributing only 6% of total vehicle kilometers traveled in the region, were found to contribute more than 50% of UFP (particle number) and PM_1 regional emissions. This finding highlights the need to vigilantly monitor HDVs in urban areas due to their very high particle number emission rates and the carcinogenic nature of diesel exhaust.

In the urban SEQ ***inventory average particle emission factors*** were applied to ***traffic count data*** for different vehicle and fuel types (and for PM_{10} average emission factors were also derived for different road types). ***Scenario modeling*** was undertaken to model changes in travel behaviour (travel mode choice), fleet composition, and the possible effect of likely future changes in technology on emission rates in the study region.

Application of the ***regional inventory approach*** for the urban SEQ inventory involved:

- ***Average emission factors*** were derived for different particle metrics, vehicle and fuel types. These were derived based on statistical analysis of more than 600 emission factors in the international published literature. A small number of local bus emission factors were also identified. .
- ***Travel data*** was sourced from an average speed travel demand model that utilized weekday travel data, where vehicle kilometers traveled assigned to each model link (representing different roads) was calculated by multiplying the number of vehicles in each vehicle class by the length of the model link in kilometers for different four travel times.
- Particle emissions in the ***four different travel times*** were calculated by multiplying the relevant average particle emission factor by the relevant VKT and summed to quantify 24 hour emissions, then scaled up to annual emissions using a scaling factor.
- ***Scenario modeling*** included estimating likely regional emissions in 2026 based on anticipated technological advances, and analysis of bus timetable and occupancy rate data to model varying shifts in travel modes (such as shifts from cars to buses) in different travel times, and corresponding average vehicle occupancy rates, to estimate their impact on regional particle emissions.

A second approach, depending on the objectives of the inventory and budget available for the project, is the *microscopic inventory approach* for different vehicle and fuel types. This approach would be particularly useful for quantifying UFP emissions under localized changes to the transportation system, and for estimating exposure of specific populations. Such inventories could consider:

- ***Vehicle operating mode-related UFP emission factors*** are needed for ***different microenvironments*** (e.g., canyons, highways, tunnels *etc.*) at different distances for exposure assessment, that should take into account street or microenvironment geometry, temporal and spatial factors, as well as distance from the source and changing driving conditions.
- UFP inventories are needed that quantify emissions at ***different distances from the traffic source*** to reflect the ageing process of particle size distributions and particle number concentrations, e.g., at <15m, 15–50m, 50–100m, 100–150m, 150–300m (or similar), where background levels are generally expected at 300 m, and which quantify emissions for two distinct groups – for sensitive and general populations.
- Use of ***vehicle operating mode and/or speed-related emission factors***. Particle number emission factors are highly dependent on vehicle operating mode for both gasoline vehicles [81] and heavy-duty diesel engines [78]. Emission factors could be estimated according to engine operating parameters, and/or vehicle operating parameters such as vehicle specific power, vehicle speed, acceleration, and road grade.

- Implementation of ***vehicle-technology specific factors***. As stated, there is substantial variability among particle emission rates across vehicle classes and operating modes. In some cases (*i.e.*, bus terminals, truck terminals, and passenger car expressways) it may only be necessary to quantify emissions from one source. However, in most cases, particle number emission rates should be modeled separately between heavy-duty diesel vehicles and light-duty gasoline vehicles.
- Derivation of ***UFP emission factors*** for different motor vehicle and fuel types using instrumentation capable of measuring down to the lower size range of 3nm, which includes the nucleation mode (<0.01 μm) and Aitken nuclei modes (0.01–0.1μm), such as for subsets in the ranges:

  0.003 μm to <0.01 μm;
  0.01 μm to <0.03 μm;
  0.03 μm to <0.05 μm and
  0.05 μm to 0.1 μm.
- These size ranges take into account the observed higher emission rates in the lower size ranges, and consider the common size ranges for UFP primary particles (0.03 μm to 0.1 μm), and secondary particles (<0.03 μm).
- Factors such as bus ***idling time*** need to be considered as emission hot spots can occur where heavy-duty diesel vehicles idle for extended periods (typically more than several minutes), and how close the ***path chosen*** by (or available to) ***pedestrians*** is to the traffic source or to accelerating buses and acceleration distances.
- Due to measurement difficulties, it is unlikely that it will be possible to quantify the variation of ***secondary particles*** in terms of operating modes. Particle measurements will likely be derived from dynamometer studies, or other studies that employ the use of artificial dilution conditions. The limitations of the particle measurements (including size range measured, dilution conditions, and physical characteristics of the particle size distribution) should be presented together with particle number inventory data.
- UFP exposure studies need to contribute to the development of emission inventories that accurately reflect the ***contribution of secondary particles***, however currently mode-specific emission rates are not available that include these particles (only for general operating conditions). In such cases, it is unlikely that particle emission rates will be sensitive to second-by-second operating modal changes. However, these secondary particle emission rates should be applicable to the vehicle fleet composition, fuel type, meteorological conditions, and general traffic conditions.

The comprehensive set of average UFP (particle number) and particle mass (PM_1, $PM_{2.5}$ and PM_{10}) emission factors derived for the urban SEQ inventory are suitable for use in developing motor vehicle

fleet inventories and air quality assessments in other developed countries, and provide a simple and cost-effective source of emission factor values for modelers and researchers.

The discussion in this paper has highlighted some of the complexities and challenges faced by researchers in developing UFP inventories. The first published inventory of ultrafine particles for a motor vehicle fleet was introduced, and recommendations for developing UFP inventories at varying temporal and spatial scales were presented.

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