The Gateway Cities Air Quality Action Plan

STATE-OF-THE-SCIENCE FOR MODELING AND MONITORING ULTRAFINE PARTICLES NEAR ROADWAYS AND IMPLICATIONS FOR GATEWAY CITIES REGION

Final

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Glossary of Terms

µm	Micrometer. One millionth of a meter
Background concentration	Concentration of substance in the absence of any significant sources present in the vicinity.
BC	Black Carbon
CARB	California Air Resources Board
CNG	Compressed Natural Gas
CO	Carbon Monoxide
CPC	Condensation Particle Counter
D_a	Aerodynamic diameter
DMA	differential mobility analyzer
D_P	electric mobility diameter
D_s	Stokes diameter
EC	Elemental Carbon
ELPI™	Electrical Low Pressure Impactor
EPA	Environmental Protection Agency
EU	European Union
Exposure	Contact with a substance by swallowing, breathing, or touching the skin or eyes
FMPS™	Fast Mobility Particle Sizer
I/O	Indoor to Outdoor
LA	Los Angeles
LPG	Liquefied Petroleum Gas
Mass Concentration	Total mass of particles in a unit volume of ambient air. Generally expressed as micrograms per m3
MATES	Multiple Air Toxic Exposure Study
nm	Nanometer. One billionth of a meter.
Number Concentration	Number of particles in a unit volume of ambient air. Generally expressed as number per m3 or number per cm3
OM	Organic Matter
Particle Size Distribution	Characterization of particle properties such as number or mass on basis of the particle size

PM	Particulate Matter
PM10	Particles with aerodynamic diameter less than 10 microns
PM2.5	Particles with aerodynamic diameter less than 2.5 microns
PMP	Particle Measurement Program
RIT	Road-induced Turbulence
SCAQMD	South Coast Air Quality Management District
SEM	Scanning Electron Microscopy
SMPS™	Scanning Mobility Particle Sizer
SO_2	Sulfur Dioxide
SULEV	Super Ultra Low Emissions Vehicle
TEM	Transmission Electron Microscopy
UFP	Ultrafine Particles
Ultrafine particle	Particles generally with aerodynamic diameter less than 0.1 micrometer
UNECE	United Nations Economic Commission for Europe
VIT	Vehicle-induced Turbulence

1. Introduction

Ambient particulate matter (PM) has been linked to numerous adverse health outcomes and remains a major environmental challenge. In recent years, there is increasing evidence of particle size playing a major role in determining the extent of adverse health effects. It has been observed that smaller particles have a higher probability of penetrating into and depositing in lower parts of the human lung and entering the bloodstream than larger particles, which are removed more efficiently by the defense mechanism of the mucociliary system (Daigle et al. 2003). Furthermore, on a mass basis, ultrafine particles have larger surface area and higher oxidant capacity than larger particles. This larger surface area likely enhances the toxic properties of the ultrafine particles. Animal studies in particular have shown increased pulmonary inflammation from ultrafine particles rather than larger particles with the same chemical composition. Therefore, there has been growing interest to better understand the particles in size range less than 100 nanometers (nm), commonly referred to as ultrafine particles.

Although the operational definition of ultrafine particles varies in the scientific literature, it is generally accepted that particles with size less than 100 nm (0.1 μm) are labeled as ultrafine particles. Note that the term nanoparticle is also used often in the scientific literature for particles with sizes on the order of few nanometers, especially in toxicological studies. Although this term is applicable to ambient particles with sizes less than 10 nm, it is applied more often to manufactured nanoparticles rather than those emitted due to human activities.

The health effects of concern from inhaling ultrafine particles include pulmonary inflammation, oxidative stress, and induction of exacerbation of cardiovascular disease (Peters, et al. 1997; Utell and Frampton 2000; Frampton 2001; and Murr and Garza 2009). There remains considerable uncertainty on biological mechanisms and the physiochemical components on the ultrafine particles that lead to these health effects. This remains a very active area of epidemiological research.

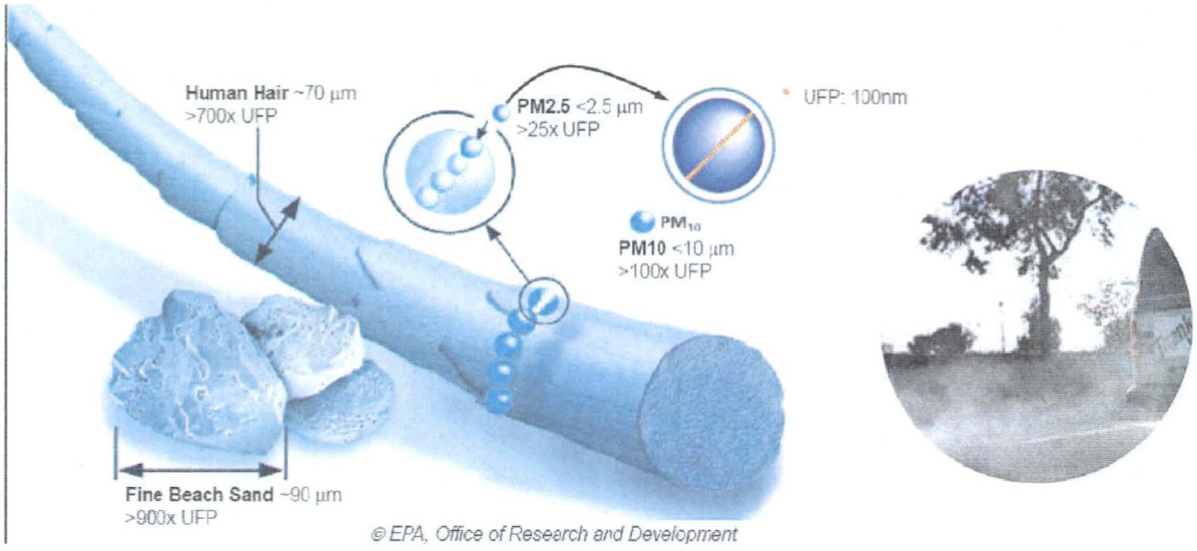
Despite a significant body of suggestive evidence linking ultrafine particles to adverse health effects, there are still many gaps in the knowledge of ultrafine particles to which people are exposed to in the environment. Areas continuing research to improve the understanding of exposure to ultrafine particles include understanding the behavior of the ultrafine particles in the near field and the atmospheric processing of these particles; developing robust measurement techniques and instrumentation; and developing operational protocols to accurately characterize them. The objective is to ultimately advise the public, the environment regulatory community, and other stakeholders in developing effective approaches that reduce exposure to ultrafine particles.

This report is prepared to briefly summarize the latest understanding of ultrafine particles in the context of the Gateway Cities Air Quality Action Plan and is organized as follows. Section 2 describes the characteristics of ultrafine particles and current techniques to measure their ambient concentrations. Section 3 describes emissions and mechanisms of the atmospheric processing of ultrafine particles emitted from vehicle tailpipes. Section 4 summarizes recent monitoring studies performed in the Los Angeles (LA) air basin, identifies major trends, and also presents studies that measure exposure to ultrafine particles in the LA air basin. Section 5 discusses current and prospects for future regulation of ultrafine particles. Finally, Section 6 concludes with a summary of major findings.

2. Characterization of Ultrafine Particles

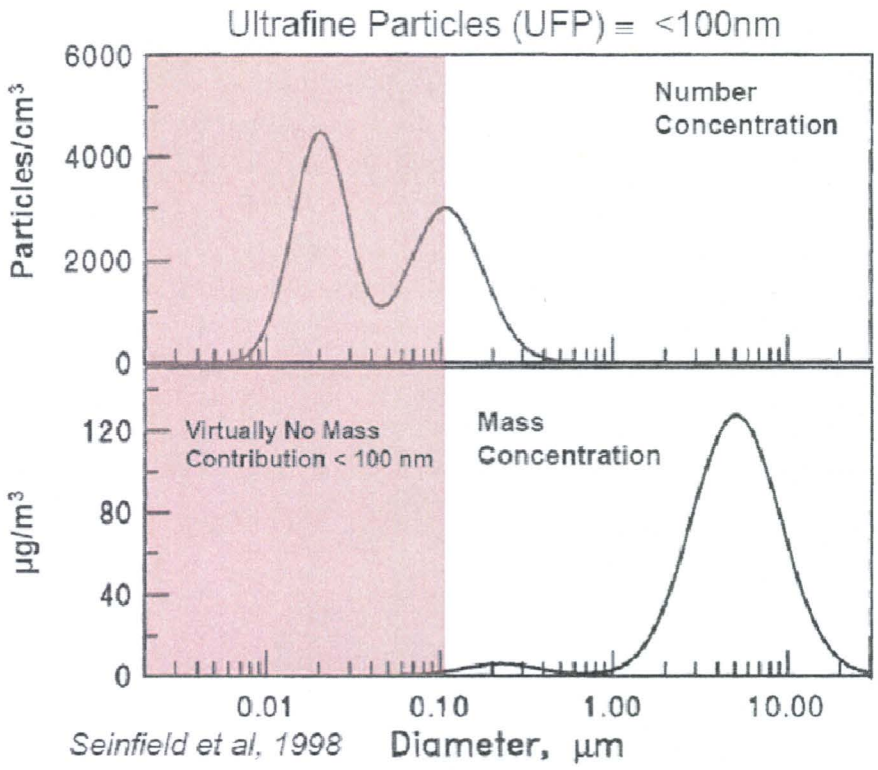
Ambient particles in general are characterized by their size, shape, physical properties (e.g., density, volatility), and chemical composition. For ultrafine particles, the most defining property is their size. As mentioned earlier, ultrafine particles are generally defined as particles with a diameter less than 100 nanometers (nm) or 0.1 micrometers (µm). In contrast, PM2.5 and PM10 consists of particles with diameters less than 2.5 and 10 µm, respectively. Therefore, ultrafine particles are nearly 25 times smaller than PM2.5 and 100 times smaller than PM10 in diameter. Figure 1 illustrates size difference between ultrafine particles, PM2.5, and PM10. Note that PM2.5 and PM10 fractions of the ambient PM also include ultrafine particles based on the above definitions. Currently, the mass concentration of PM2.5 and PM10 are regulated by national and California ambient air quality standards and, for motor vehicles, a mass per mile exhaust emission standard.

Figure 1. Relative Size of Ultrafine Particle (UFP) Relative to Human Hair and Other Particle Sizes



Because of their small size and volume, the aggregate mass of ambient ultrafine particles is not significant compared to larger particles. However, several studies have shown that ultrafine particles dominate the number concentration of ambient PM. For example, Stanier, et al. (2004) showed that 25% of the ambient aerosol particles are less than 10 nm and 75% of the number of particles are less than 50 nm. In contrast, particles larger than 100 nm, i.e. PM2.5 and PM10, primarily contribute to the mass concentration. See Figure 2.

Figure 2. Comparison of Number and Mass Distribution of Ambient Particles



Along with size, chemical composition also influences the toxicity of ultrafine particles. However, the chemical composition of ultrafine particles is even less well understood. This is mainly because a significant variation in chemical composition can be found within various environments, times of the day, and months of the year, depending on the nature and emission strengths of local sources and atmospheric conditions. Organic matter (OM), elemental carbon (EC), and inorganic compounds such as sulfates and nitrates constitute most of the ultrafine particle mass. Other constituents include water, sodium, potassium, and transition metals. EC is considered as a marker of the combustion processes for diesel engines as its major source in urban areas. OM, a mixture of high molecular weight hydrocarbons, is emitted directly from combustion processes and also forms in the atmosphere from gas-phase emissions. OM constitutes a significant fraction of emissions from meat cooking and wood burning. Sulfate in the particles results from sulfur present in the fuel that is oxidized during the combustion process. Similarly, high temperatures during combustion lead to the oxidation of atmospheric nitrogen, which results in presence of nitrates in ambient particles. Some recent studies conducted in southern California region showed that the chemical composition of ultrafine particles ranged from 32 to 69% of OM, 1 to 34% of EC, 0 to 24% of sulfate, and 0 to 4% of nitrate (Sardar et al. 2005; Arhami, et al. 2009).

The United States and California air quality standards for particulate matter are based on mass concentration. Therefore, currently there is no large-scale monitoring network that measures number concentration of ambient particles. Several studies are available in the literature that measured particle number concentration in various outdoor environments. Morawska et al. (Morawska, Ristovski et al. 2008) analyzed 71 measurement studies recently reported in the literature and calculated the mean and median number concentration for eight different environments. As shown in Table 1, the number

concentrations are highest in tunnel environments followed by on road and roadside environments. The mean concentrations for these environments are greater by 64, 27, and 18 times respectively than a clean background.

Table 1. Mean and Median of Particle Number Concentrations in Various Environments

Environment	Mean (10^3 Particles/cm)	Median (10^3 Particles/cm)
Tunnel	167.74 (64)	99.09 (31)
On Road	71.45 (27)	47 (15)
Roadside	48.18 (18)	34.58 (11)
Street Canyon	42.07 (16)	39.13 (12)
Urban	10.76 (4)	8.83 (3)
Urban background	7.29 (3)	8.1 (3)
Rural	4.83 (2)	2.91 (1)
Clean Background	2.61 (1)	3.2 (1)
Data source: Morawska et. al. 2008. The numbers in parenthesis show the ratio of the concentration statistic with that of a clean environment.		

2.1. Measurement of Ultrafine Particles

Ambient particles often have variety of shapes (e.g. tubular, irregular, etc.) rather than ideal spherical shape, which can be characterized by only one size parameter (the diameter). Therefore, this poses some difficulties in their mass and number concentration measurement. Aerodynamic diameter (D_a), Stokes diameter (D_s), and electric mobility diameter (D_P) equivalent are often used in the context of the properties of particles moving in air. Aerodynamic diameter is the diameter of a perfect sphere of unit density with same mobility as the particle being measured. Stokes diameter is similar to the aerodynamic diameter, but uses the true density of the particle. Electric mobility diameter is widely used in instruments and defined as the diameter of a perfect sphere of unit density with same electric mobility as the particle being measured.

A recent review by Kumar, et al. (2010) identified several instruments that are commercially available to measure the size distribution of ambient particles, including ultrafine particles. These instruments include the Scanning Mobility Particle Sizer (SPMS™), Fast Mobility Particle Sizer (FMPS™), and Electrical Low Pressure Impactor (ELPI™) that provide size-segregated number concentration. In addition, a condensation particle counter (CPC) is a standard instrument employed in the study of ultrafines to provide number concentration of the entire aerosol population.

The SMPS system is an instrument that uses electric mobility principles to measure the number and size distribution of an inlet aerosol population Kumar, et al. (2010). This instrument mainly consists of three components: 1) A bipolar radioactive charger to charge the particles, 2) a differential mobility analyzer (DMA) to classify the particles based on their electrical mobility, which in turn depends on their size, and 3) a CPC for detecting the particles and providing the number concentration. Some commercially available SMPS include the 3034 TSI (TSI Inc) that measures D_p between 10 and 487 nm for number concentrations in the range of 10^2 to 10^7 particles/cm^3 and takes 180 seconds to analyze a single scan. More recent models (such as the 3934 TSI) can measure a larger range of particle size and with shorter sampling times (30 seconds).

The FMPS is similar to SMPS in principle. However, instead of a CPC, the FMPS uses multiple, low-noise electrometers for particle detection and count. The main advantage of FMPS is that its sampling frequency can be as high as 1 Hz (i.e., analysis of one sample per second), which is partly accomplished by using a high sample flow rate. Therefore, FMPS can be used to characterize fast-changing particle populations.

The ELPI is another instrument that has high sampling rates and measures the size distribution of particles in the range of 30 to 10,000 nm. This instrument uses aerodynamic size classification of charged particles by a low pressure cascade impactor and subsequent electrical detection for particle count. An added advantage of this instrument is that the cascade system allows for other analysis such as chemical composition and further characterization through scanning electron microscopy (SEM) or transmission electron microscopy (TEM) techniques.

As noted by Kumar et al. (2010) several parameters need to be considered before adopting a particular instrument or detection technique to be used in a regulatory or compliance context. These include portability, time response, detection limits, sampling frequency, reproducibility and capability of unattended operation over long duration, and cost and maintenance requirements. Despite technological advancements in recent years that address many of these issues, reproducibility and consistency across various instruments still remains an area that needs additional research and progress. Asbach et al. (2009) used four different particle sizers (two TSI SMPS, one TSI FMPS, and one Grimm SMPS) to analyze NaCl and diesel soot particles and found that each instrument showed repeatable results. However, there was a considerable difference between instruments. Among these four instruments, FMPS consistently showed lower particle size distributions than SMPS.

3. Ultrafine Particles in Near-Roadway Environment

Several studies have shown that motor vehicles are the dominant source of ultrafine particles in the outdoor environment. Furthermore, the concentrations are highest in the near-roadway locations given their proximity to the emission sources. Among motor vehicles, heavy duty diesel engines are shown to emit significantly higher numbers of ultrafine particles than light-duty gasoline vehicles. In general, a heavy duty diesel truck or a bus may emit one or two orders of magnitude higher than a typical light-duty vehicle (Kirchstetter, et al. 1999; Ristovski, et al. 2005).

The number concentration of ultrafine particles at a near-roadway location is primarily determined by the strength of emissions in the surrounding area and atmospheric processing of the emitted particles as they are transported to the location of interest. These two aspects are discussed further below.

3.1. Emissions of Ultrafine Particles from Vehicles

From emissions perspective, ultrafine particles can be classified into either *primary* or *secondary* particles. Primary particles are formed in the engine and tailpipe and are emitted directly into the ambient atmosphere. These particles, mostly agglomerates of solid-phase carbonaceous material, are generally in the size range 30 to 500 nm. As hot gases are released from the tailpipe, they are cooled rapidly in the ambient environment. Consequently, many of the constituent gases with low-volatility condense to form a large number of new particles. These are known as secondary particles and are generally below 30 nm. Secondary particles are largely composed of sulfate, nitrate, ammonium, and organic material.

The first step in quantifying and addressing the extent of vehicle emission contribution to the ambient concentration is characterization of the tailpipe emissions and how such emissions vary with the nature of fuel composition, operating conditions, and control technologies.

Early studies on particulate emissions and fuel composition focused on sulfur content. Sulfur occurs naturally in crude oil and is mostly removed in the refining process, but a small percentage remains in diesel and gasoline fuel. During the combustion, sulfur is oxidized to sulfuric acid and is emitted through exhaust as a volatile material. After leaving the tailpipe, sulfuric acid undergoes condensation or nucleation and contributes to total particle emissions. Dynamometer and on-road measurements have shown that particle number decreases with the sulfur content both in fuel and lubricants. The introduction of ultra low sulfur fuels in California in 2006 and fully phased in nationwide by 2010 has significantly reduced ultrafine particles forming from sulfur condensation and nucleation.

In recent years, various biofuels (both pure and blended) have begun to be used in heavy duty diesel trucks and buses in order to decrease the reliance on imported fossil fuels and also to mitigate climate change. This transition may affect tailpipe emissions. Studies have shown that most heavy duty vehicles powered by biofuels emit a higher number of particles than those powered by diesel (Kumar, et al. 2010). This increase is attributed to the increased nucleation rates in the exhaust of biofuels due to reduced surface area of pre-existing particles and lower caloric content in biofuels requiring the use of greater quantities of fuel compared to conventional diesel fuel. However, it is important to note that the

use of biofuels leads to overall reduction in particulate mass emissions due to the decrease in the emissions of solid carbonaceous particles.

Among alternative fuels, liquefied petroleum gas (LPG) and compressed natural gas (CNG) are seen as cleaner than conventional diesel or biofuels and are widely used in the South Coast bus fleet, and they have potential as a widespread replacement fuel for heavy duty trucks. A few studies that have examined these fuels have found that LPG vehicles are shown to emit 70% less ultrafine particles than conventional unleaded gasoline vehicles (Ristovski, et al. 2005). CNG vehicles are known to emit considerably less particle mass than diesel vehicles. Although CNG and LNG buses emit lower number of particles at lower loads compared to diesel buses, both dynamometer and on-road studies have shown that the number of particles emitted by CNG at high loads and during acceleration are an order of magnitude greater than those of diesel buses (Jayaratne, et al. 2010). For CNG buses, emissions of particle number vary by an order of magnitude between low-load and high-load conditions.

As with other vehicle emissions, ultrafine particle emissions from tailpipes may be controlled using the after-treatment devices. Two main classes of these devices are oxidation catalysts and particle traps. Oxidation catalysts mainly remove the soluble organic fraction and have no effect on elemental carbon in the exhaust. Further, these catalysts may oxidize sulfur dioxide (SO_2) to sulfate and may increase sulfate in particle mass and number. Therefore, the effectiveness of these devices depends on the relative chemical composition of the exhaust. Application of oxidation catalysts may actually increase the particle number when used in conjunction with high sulfur-content fuels (Vogt, et al. 2003).

Particle traps have proven to be effective in removing the solid-phase particles, including elemental carbon. However, volatile material (in gas or liquid form at high exhaust temperatures), especially sulfate mass, passes through the filter and acts as a precursor to nucleation-mode particles. Furthermore, by retaining solid-phase particles, particle filters may promote nucleation over condensation thus leading to an increase in the formation of new particles.

3.2. Environmental Fate and Transport of Ultrafine Particles

Almost immediately after the ultrafine particles are emitted into the atmosphere, atmospheric processes govern their fate and transport in the ambient environment and dynamically change their physical and chemical characteristics. Consequently, the temporal and spatial evolution of ultrafine particles, especially their size distribution, depend on the extent of atmospheric processing, which in turn depends on ambient meteorological conditions such as temperature, wind speed, relative humidity, and incident solar radiation.

Once particles are emitted from the vehicle tailpipe, transport by wind is the main mechanism by which emissions are initially dispersed into the atmosphere, resulting in dilution of the exhaust plumes. Turbulent transport, both in the horizontal and vertical direction, dominates the dispersion of pollutants. In general, high wind speeds result in efficient transport of pollutants, leading to lower ambient concentration in the vicinity of emissions sources. Atmospheric stability, a measure of

resistance to the vertical motion of air parcels in the atmosphere,[1] also significantly affects the turbulent mixing and resulting dispersion of emissions.

Another distinguishing feature between the dispersion of pollutants from roadway and non-roadway sources is the effect of turbulence generated by vehicles, roadway structures, and noise barriers. The near-roadway turbulence, broadly classified into vehicle-induced turbulence (VIT) and roadway-induced turbulence (RIT), may significantly contribute to the mixing of pollutants, leading to additional dilution of near-roadway emissions (Bäumer, et al. 2005; Kalthoff, et al. 2005). A significant fraction of kinetic energy may be converted into additional turbulence due to the aerodynamic resistance when fast-moving vehicles interact with the ambient air. The turbulence generated strongly depends on the vehicle shape and speed. For example, heavy duty diesel trucks induce more turbulence due to their size and shape than passenger cars. Therefore, reducing congestion and increasing speeds will promote more mixing and lower peak concentration of ultrafine particles.

Road embankments also generate turbulence as they act as the obstacle to the wind flow. The RIT due to embankment depends on the wind speed, direction, and the height and shape of the embankments. Similarly, roadside structures such as noise barriers and vegetation also affect dispersion by creating local turbulence effects (Lidman 1985; Baldauf, Thoma et al. 2008). In addition, the temperature difference between asphalted roadway surfaces and surrounding grassy surfaces creates thermally induced flow, which could affect dispersion of roadway emissions. Finally, in an urban environment, built structures distort the wind field around them and therefore affect the dispersion of pollutants.

Zhang and Wexler (2004); Zhang, et al. (2004); and Zhang, et al. (2005) proposed that the dilution of a vehicle exhaust plume may be considered to occur in two distinct stages. The first is the tailpipe-to-road dilution by the strong turbulence generated by traffic, lasting about 1 to 3 seconds and causing the initial concentration of ultrafine particles to be diluted by up to a factor of 1,000. The second stage is the roadway-to-ambient environment, which further dilutes the concentration of ultrafine particles by about a factor of 10, lasting 3 to 10 minutes. This dilution is due to atmospheric turbulence and advection by the ambient wind. Such large dilution ratios lead to an exponential decrease in the number concentration of ultrafine particles within relatively short distances from a freeway as observed in a number of studies (Zhu, et al. 2002a; Westerdahl, et al. 2005; Morawska, et al. 2008).

As vehicle exhaust plumes are diluted in the atmosphere, three main processes—nucleation, condensation, and coagulation—change the size of the particles in the plume and govern the overall size distribution. Nucleation refers to the formation of new particles with a size generally below 20 nm from super-saturation of low volatility gaseous compounds. However, such super-saturated compounds can also condense onto pre-existing particles, which increases the size of such particles. Thus, nucleation and condensation are two competing processes. The availability of a pre-existing particle surface area

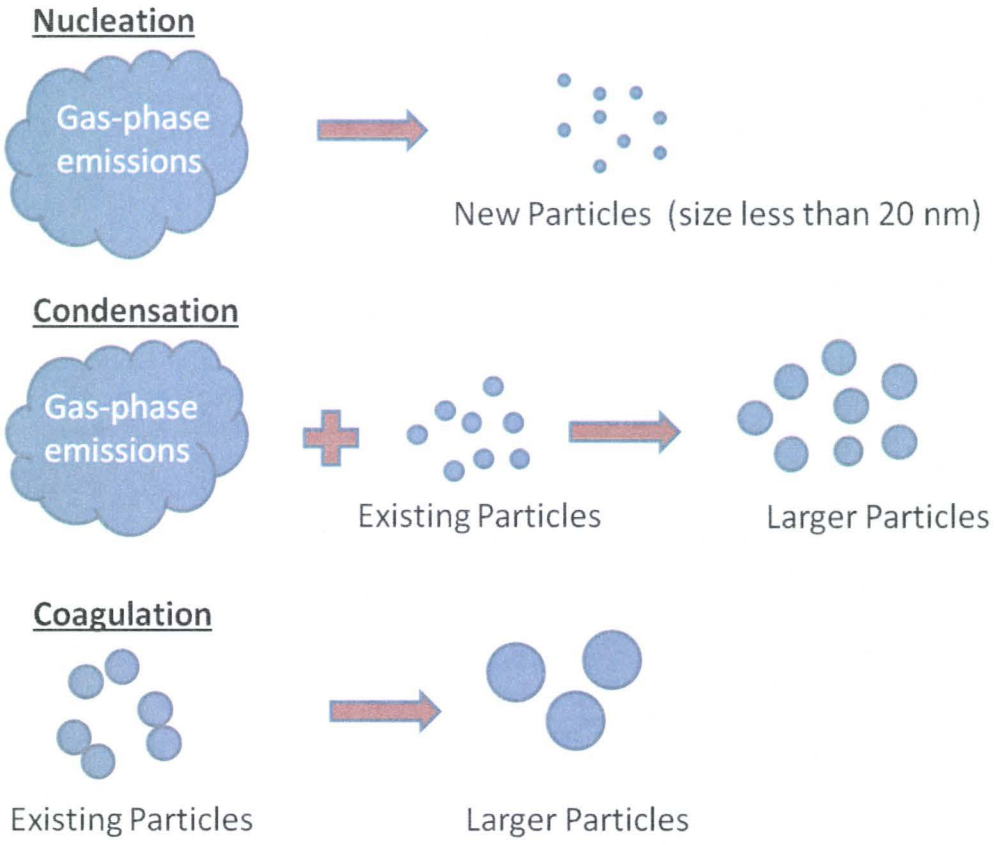
[1] Stability can be broadly classified as stable, neutral, and unstable. Stable atmospheric conditions restrict the vertical movement of air parcels, thus creating conditions conducive for the accumulation of pollutants near the surface. Unstable atmospheric conditions accelerate the vertical movement of air parcels and promote the turbulent mixing of pollutants. Under neutral conditions, the dispersion of pollutants is influenced by both transport and turbulent mixing.

and plume dilution rate determines which of these processes dominates. Polluted urban environments often contain a large number of pre-existing particles to favor condensation over the formation of new particles through nucleation. Coagulation occurs when two or more particles collide and combine to form a new larger particle. Figure 3 graphically illustrates these three processes.

The emissions from the tailpipe of a vehicle containing highly concentrated gas vapors undergoes rapid cooling immediately after leaving the tailpipe, during the tailpipe-to-road dilution phase, and reaches super-saturation, which may result in both the generation of new particles through nucleation and growth of the existing particles through condensation. Such particles, often less than 30 nm in size, have been observed near busy freeways, especially those carrying a large fraction of heavy duty diesel trucks (Zhu, et al. 2002b; Westerdahl, et al. 2005; Ntziachristos, et al. 2007).

The rate of generation of ultrafine particles depends on several factors, including temperature, relative humidity, atmospheric stability, and vapor pressure (Zhang and Wexler 2004; Zhang, et al. 2005). Low ambient air temperatures and high relative humidity favors the formation of new particles as demonstrated in some on-road studies (Ronkko, et al. 2006; Casati et al. 2007). Particles that are generated through nucleation have short atmospheric lifetimes, on the scale of few minutes, because they coagulate with other particles and form new larger particles.

Figure 3. Illustration of Nucleation, Condensation, and Coagulation Processes That Affect the Size Distribution of Ambient Particles



4. Ambient Concentration of Ultrafine Particles in the Los Angeles Area

As the significance of ultrafine particles' possible public health concerns have gained increasing attention in recent years, several monitoring studies have been performed, mostly by academic institutions, to better characterize and understand the ambient concentrations of ultrafine particles, especially in near-roadway environments. This section synthesizes the trends and characteristics of ambient ultrafine particles reported in the scientific literature drawn from studies conducted in the Los Angeles area.

As discussed in the previous section, a distinguishing feature of ultrafine particles is their elevated concentration close to roadways (or emission sources) and their exponential decay with distance from the freeway because of transport and turbulent mixing. This behavior has been confirmed in several field studies conducted in the vicinity of a number of southern California freeways.

Zhu, Hinds et al. (2002a; 2002b) conducted four measurement campaigns to characterize the ultrafine particles in LA area during the summer of 2001 and the winter of 2002. These studies measured the particle number concentration and size distribution in the size range from 6 to 220 nm at various distances from I-405 and I-710. In addition, the mass concentrations of carbon monoxide (CO) and black carbon (BC) were also measured at each sampling location. Black carbon is a close surrogate for diesel PM emissions. Since an identical set of instruments was used and common data analysis methods were applied by the same group of researchers, the bias in configuration and data interpretation is minimized. Therefore, data from these studies can be used to obtain important insights and understanding of ultrafine particle characteristics in the near-roadway environment.

The summertime measurements were conducted 30, 60, 90, 150, and 300 meters downwind and 300 meters upwind for I-405 at the Los Angeles National Cemetery between May 15 and July 18, 2001. For the I-710 study, the measurements were taken at 17, 20, 30, 90, 150, and 300 meters downwind and 200 meters upwind from the freeway in the City of South Gate along Southern Avenue between August 30 and October 27, 2001. Wintertime measurements were taken at the same monitoring sites between January 11 to January 20, 2002, for the I-405 sampling sites and between January 14 and January 25, 2002, for I-710 sampling sites. For all studies, particle measurements were made using a CPC for total number concentration (CPC 3022A; TSI Inc.) and SMPS for size-segregated number concentration (SMPS 3936, TSI Inc.).

The traffic on I-405 is dominated by passenger cars and light-duty vehicles with less than 5% of heavy-duty diesel trucks. In contrast, the heavy-duty diesel traffic on I-710 is often higher than 25%. During the measurement period, the hourly average traffic volume on I-405 was 13,900 vehicles, and more than 93% of these were gasoline-powered passenger cars. A strong correlation was observed between the traffic density and measured total particle concentration for monitoring sites near I-405. For the I-710 study, the hourly average hourly traffic volume was 12, 180 vehicles with heavy trucks constituting nearly 30%. The traffic density during this study has less variation than that of I-405.

As discussed in the previous section, wind plays a major role in the dispersion of emitted ultrafine particles. Due to frequent on-shore winds, the monitoring sites in both studies by Zhu et al. (2002a, 2002b) were downwind of the freeway 80% of the time. Total particle number concentration measured by a CPC located 30 meters downwind from I-405 decreased almost linearly with the wind speed with a correlation coefficient (R^2) value of 0.99. In contrast to this behavior, total particle concentration first increased with the wind speed up to 1.5 m/s and then decreased. This anomaly was attributed to large uncertainty in measurements made when speeds are less than 1 m/s.

The findings from Zhu have important implications for I-710. Prevailing winds along the I-710 corridor in the AQAP study area are from the southeast to east. This will typically lead to the highest ultrafine particle exposure over the Los Angeles River south of State Route 90 (Imperial Highway) where the nearest sensitive receptors on the east side of the I-710 are more than 500 meters from the roadway. North of the State Route 90, where the Los Angeles River is to the west of I-710, the communities of Bell Gardens, Commerce, and East Los Angeles are in much closer proximity to I-710 and are in the prevailing downwind direction.

Despite the difference in the traffic vehicle mix, both studies by Zhu showed some common behaviors. The maximum number concentration that was observed next to I-710 and I-405 is nearly 25 and 30 times that of the background concentration, respectively. However, the number concentration of ultrafine particles near both freeways falls precipitously with the distance and become indistinguishable from an upwind site at 300 meters. In addition, the CO and BC concentrations also decrease at the same rate as particle number in both studies and drop by 60 to 80% within first 100 meters. However, PM mass concentration, measured in the I-405 study decay at a much smaller rate, displays different characteristics from CO, BC and particle number concentrations. This is because direct PM emissions from passenger car emissions contribute a relatively small amount relative to background PM concentration. The CO, BC, and number concentration data is summarized in Tables 2 and 3.

Both studies by Zhu (2002a and 2002b) also analyzed the evolution of size distribution of ultrafine particles in a near-roadway environment using the data collected from SMPS. The size distribution of ultrafine particles sampled at both I-405 and I-710 showed three distinct modes. Zhu (2002b) further examined the number concentration of particles in the following size bins: 6 to 25 nm, 25 to 50 nm, 50 to 100 nm, and 100 to 220 nm. In both studies, the particles in the smallest bin (6 to 25 nm), contribute over 70% to the total number concentration and decrease sharply within first 100 meters from the freeway. This characteristic is attributed to the rate of coagulation and faster diffusion rates of smaller particles.

Table 2. Measured Average Concentrations of Ultrafine Particles, CO and BC at Increasing Distances from I-405

Measurement (m)	30	60	90	150	300
CO (ppm)	2.0 (1.7-2.2)	0.9 (0.7-1.0)	0.6 (0.5-0.7)	0.4 (0.3-0.5)	0.2 (0.1-0.3)
BC ($\mu g/m^3$)	5.4 (3.4-10.0)	3.2 (3.0-3.5)	2.5 (2.4-2.6)	1.6 (1.1-2.0)	1.3 (1.1-1.5)
Number Concentration ($x10^5/cm^3$)	1.5 (1.3-1.7)	0.88 (0.77-0.96)	0.70 (0.61-0.85)	0.50 (0.42-0.58)	0.37 (0.30-0.39)

Data Source: Zhu 2002a, 2002b.

Range given in parenthesis. Note these concentration include background.

Table 3. Measured Average Concentrations of Ultrafine Particles, CO and BC at Increasing Distances from I-710

Measurement (m)	17	20	30	90	150	300
CO (ppm)	2.3 (1.9-2.6)	2.0 (1.5-2.4)	1.7 (1.1-1.9)	0.5 (0.2-0.7)	0.4 (0.1-0.5)	0.2 (0.1-0.3)
BC ($\mu g/m^3$)	21.7 (20.3-24.8)	19.4 (16.5-21.6)	17.1 (12.6-19.3)	7.8 (4.5-9.3)	6.5 (3.9-9.2)	5.5 (3.5-7.7)
Number Concentration ($x10^5/cm^3$)	2.0 (1.8-2.5)	1.8 (1.5-2.5)	1.6 (1.2-1.9)	0.72 (0.42-1.1)	0.61 (0.35-0.98)	0.49 (0.30-0.59)

Data Source: Zhu 2002a, 2002b.

Range given in parenthesis.

Zhu (2002b) also compared the size-resolved number concentrations of ultrafine particles measured 30 meters from I-710 and I-405 freeways. Note that both freeways have nearly the same average traffic density; however, I-710 traffic emits higher levels of PM due to the greater proportion of heavy duty diesel trucks. Both sets of measurements showed three distinct modes. The number concentration close to I-405 for the first mode (10 and 20nm) is slightly higher than corresponding measurement at I-710. However, the concentration for the second mode (around 30 nm) is nearly 30% higher for the I-710 site than that of the I-405 site and is attributed to higher levels of BC particles emitted by diesel vehicles on I-710. The concentration of the last mode (around 70 nm) is similar in both studies and close to the background concentrations.

As discussed in the previous section, meteorological parameters such as temperature and relative humidity play a major role in the processing of ultrafine particles. A comparison of summer and wintertime monitoring data for the I-405 and I-710 sampling sites showed that ultrafine number concentrations in winter are higher than in summer for comparable level of traffic densities. In addition, the decay rates of ultrafine particles, CO, and BC are slightly smaller than those in summer. This decrease in the dilution rate is attributed to the prevailing lower wind speeds during winter. When the size-segregated data was compared, the mode with the smallest diameter (10 to 20 nm) persisted for longer distance during the winter season.

More recent studies have employed mobile measurement platforms to conduct more comprehensive data collection. As opposed to the studies that use a limited number of fixed monitoring locations, mobile platforms measure concentration at all directions from the road from a parallel to perpendicular direction along the roadway, but for generally shorter durations. Further, mobile measurements often also include video and audio recordings of the surroundings in order to infer possible pollution with specific sources, such as passing or following a diesel truck.

Kozawa et al. (Kozawa, Fruin et al. 2009) used an electric vehicle as a mobile measurement platform and collected measurements on two routes of about 30 miles each covering arterial roads in residential areas and freeways in the cities of Carson, San Pedro, Wilmington, and West Long Beach. Among other instruments to measure gas-phase species, the mobile platform also included a portable CPC (TSI Portable CPC, Model 3007) and FMPS (Model 3091, TSI), both having a time resolution of 10 seconds. The CPC was used to measure number concentration of particles in the range 10 nm to 1 μm and the FMPS to provide the size distribution and particles in the range of 5.6 to 560 nm.

Measurements in the Kozawa et al. study were collected by driving twice a day along the two routes, once in the morning between 8:00 and 10:30 AM and again in the afternoon between 14:30 and 17:00, two to three times per week in the summer and winter of 2007 covering a total of 24 sampling days. Since the goal of the study was to measure representative ambient concentrations, individual vehicle influences, such as a passing or trailing a high emitting diesel truck, were removed from the assessment based on the analysis of video recordings made during the measurements. Measured concentrations were analyzed using the "impact zone" and "reference zone" designations based on the location of the measurement and then calculating the ratio of concentration within the impact zone with that of the reference zone. Any location within 150 meters from a major roadway is designated to be within the impact zone. All other locations more than 150 meters from a major roadway are said to be in the reference zone. In addition, meteorological data from the South Coast Air Quality Management District (SCAQMD) monitoring station at North Long Beach were used to determine if the measurement locations are at a downwind or upwind of a major roadway.

Using the above approach, Kozawa et al. observed that the ratio of ultrafine number concentrations in the impact zone to the reference zone in the I-710 area to be at least 2 and as high as 3.7 times higher in the morning hours, and when the impact zone is downwind of the freeway. The impact- to reference-zone ratios were approximately 1 (or no change) when the impact zone was upwind of the freeway.

Near-roadway studies, such as those discussed above, often compare pollutant concentrations close to the freeway with those of "background" or reference concentrations. Background concentration is

typically measured more than 300 meters downwind or upwind of the freeway. For community-scale exposure and impact assessments, the variation in the background concentration also becomes important, especially in order to devise air quality improvement strategies. Insights on neighborhood-scale variation of ultrafine particles, especially in communities that are in the vicinity of the major transportation routes, will provide insight as to whether to focus air quality improvement strategies on a broader or local/neighborhood scale.

Moore et al. (Moore, Krudysz et al. 2009) measured ambient ultrafine particles using a network of 14 closely-spaced monitoring sites that were set up in the Los Angeles area as two clusters—San Pedro/Wilmington and West Long Beach. At these sites, total number concentration of particles greater than 7 nm is measured using a CPC (3022A, TSI Inc.). The measurements were taken from February through December of 2007 in order to assess intra-community variability in the ambient ultrafine particle concentrations. A number of sites within the West Long Beach cluster are in the proximity of I-710 at varying distances. This study showed that the total particle number can vary up to a factor of 10 for sites within a few kilometers. Although the proximity to roadways was a major factor in the variability, the observations emphasize that urban areas have elevated concentrations relative to background levels and that ultrafine concentrations vary widely between communities due to proximity to roadway sources as well as other sources of ultrafine particles.

4.1. Exposure to Ultrafine Particles in Los Angeles Area

Human exposure may generally be defined as any event where a person comes in contact with a contaminant of concern. During an exposure event, the contaminant may enter the human body through one of the exposure pathways (or exposure routes). The main exposure pathways are inhalation, ingestion, and dermal absorption. Exposure to ultrafine particles predominantly occurs through the inhalation of polluted air.

People are exposed to ultrafine particles during the course of their daily activities depending on various microenvironments they spend time in, the amount of time spent, and the concentration of ultrafine particles in those microenvironments. A microenvironment is any space in which human contact with an environmental pollutant takes place and which can be treated as a well-characterized, relatively homogeneous location with respect to pollutant concentrations for a specified time period. Microenvironments are generally grouped into three categories: outdoor, indoor, and in-vehicle. For example, parks, outdoor recreation spaces, and near-roadway locations are some common outdoor microenvironments; residences, schools, and offices are some common indoor microenvironments; and cars, public transit vehicles, and air travel compartments are some common in-vehicle microenvironments.

In the context of near-roadway environment, two classes of populations that are most vulnerable to ultrafine particles exposure: 1) residents or occupants of non-residential buildings within in the vicinity of major roadways, and 2) drivers and passengers of on-road vehicles. The extent of exposure within these two microenvironments (home/office and in-vehicle) mainly depends on the air exchange rate that determines the extent of penetration of outdoor contaminated air into the microenvironment. Air

exchange rates are governed by several factors such as ventilation and meteorological factors (wind speed).

Although exposure to gas-phase pollutants in a near-roadway environment is still a major research area, exposure specifically to ultrafine particles is gaining attention. A recent review published by Knibbs et al. (Knibbs, Cole-Hunter et al. 2011) identified 47 studies that reported numerical exposure values to ultrafine particles across various transportation modes. Based on this data, the trip-weighted mean ultrafine particle concentration near automobile tunnels was highest at 300,000 particles/cm^3 and that in a non-tunnel automobile trip was 45,100 particles/cm^3. By way of comparison with some typical indoor activities, Wilson et al (Wilson, A., O. Karpukhin, L. DeLaura, 2010) found that indoor concentrations of ultrafines during cooking meat, burning candles, and vacuuming ranged between 150,000 to 200,000 particles/cm^3.

In a recent study done by USC researchers (Hudda, Cheung, Moore, and Sioutas 2010), they found that the 2009 outdoor concentration at the University of Southern California site, which was located in an area believed to be representative of a typical urban background site, the highest ultrafine particle concentration occurred in January with an average concentration of 30,000 particles/cm^3 and the lowest ultrafine particle concentration of around 10,000 particles/cm^3 occurred during September. This is comparable to the background concentrations measured by Zhu et al. (2002a), which reported a background concentration of 30,000 particles/cm^3 at 300 meters, but the Zhu study was done in 2001 and ultrafine particle concentrations have shown a region-wide decrease across the South Coast region over this time period.

Two of the recently published studies on exposure to ultrafine particles in near-roadway environment in the LA basin are discussed here. Zhu, et al. 2005 concurrently measured ultrafine particle concentrations for four two-bedroom apartments located within 60 meters of the I-405 freeway. Three of these apartments are located on eastern side of the I-405 on the third floor. The fourth apartment is located on the western side of the I-405 on the second floor. All four apartments are within 50 meters of each other. Measurements were collected under three infiltration conditions: infiltration, mechanical ventilation, and with an open window. Size-segregated number concentration of ultrafine particles were measured using SMPS (3936, TSI Inc.) between October and January of 2003. Total particle number concentration was also measured independently using a particle counters (Model 8525, TSI Inc.) with a lower cut size of 20 nm. The penetration of ultrafine particles into residences is quantified using the ratio of Indoor to Outdoor (I/O) number concentrations of ultrafine particles. The I/O ratios are highest—close to 1.0—across all particle size ranges when the windows are open. Under the infiltration (natural ventilation) condition, the highest ratios (0.6 to 0.9) were observed for larger particles in the size range of 70 to 100 nm, while the lowest I/O ratios (0.1 to 0.4) occurred for particles in the size range of 10 to 20 nm. Finally, the lowest I/O ratios (0.2 to 0.6) occurred when the fan was on (mechanical ventilation) and is attributed partial filtering by the air-handling system in these apartments. The generally high I/O ratios indicate that residents in the vicinity of freeways are potentially exposed to ultrafine particles at levels similar to those in the outdoor air. Similar exposures would be anticipated along I-710 in close proximity and at the same elevation level as the roadway.

In addition to the exposure in indoor environments, in-vehicle exposure while driving on heavily-trafficked freeways is also shown to be significant. Zhu, et al. (2008) measured outdoor and in-vehicle particle number concentrations for three different vehicles (Volkswagen Jetta, Audi A4, and PT Cruiser) while driving on I-405 and I-710 in the LA area. Measurements were taken under three different air circulation conditions: 1) circulation fan off and recirculation off; 2) fan on and recirculation off; and 3) fan on and recirculation on. It is expected that outside air enters the vehicle only in conditions 1 and 2.

Results showed that the particle concentrations inside the vehicle closely tracked outside concentrations under conditions 1 and 2. I-710, with a high proportion of trucks and therefore a large number of ultrafine particle emissions, resulted in the highest in-vehicle ultrafine particle concentrations and therefore the highest exposure as well. The penetration ratio (ratio of indoor to outdoor particle concentrations) was highest when both the fan and recirculation are off and varied between 0.3 and 0.5. These ratios are similar for both the freeways. Overall, maximum protection was obtained when both the fan and recirculation were on. Further, based on the observations from this study, the authors concluded that the exposure to ultrafine particles from a 1-hour commute on busy LA freeways is equivalent to 10 hours of exposure in urban backgrounds away from a freeway.

5. Current and Prospects of Future Regulations on Ultrafine Particles

There are potentially two direct ways of regulating ultrafine particles: 1) by setting ambient standards, similar to that of PM mass per unit volume and other gas-phase pollutant standards, and 2) by regulating the tailpipe emissions, i.e., source-based regulation. Indirect ways of regulating ultrafine particles include restrictions on fuel composition.

Currently, there is no ambient air quality standard any place in the world that regulates the number concentration of airborne particles.

Until recently, there were no emission standards that regulated the particle number concentration from vehicle tailpipes. However, in July 2008, the UK Department of Transportation proposed an emissions standard for light and heavy duty compression (diesel) ignition vehicles at 6×10^{11} particles/km. These limits were agreed for inclusion in the European tailpipe standards for Euro-5 and Euro-6 emission standards. Euro-5 tailpipe particle number standards are being phased in starting in September 2011 and fully phased for all models by January 1, 2013. The test methodologies for the implementation of this standard were developed under the Particulate Measurement Program (PMP) of United Nations Economic Commission for Europe (UNECE). PMP, initiated in 2001, is an inter-governmental research program to develop new vehicle exhaust measurement protocols for regulatory applications. The PMP focuses on development of methodologies to measure solid particles in the range 23 to 2500 nm. Per the PMP test procedure, only solid particles are counted towards the emission standard since any volatile material is removed from the sample. The US Environmental Protection Agency (EPA) has reviewed the PMP testing method and does not see this as an acceptable approach for the US partly because of the missing volatile material.

The EPA has no immediate plans to regulate ultrafine particles. However, there is increasing interest within the EPA in researching investments directed towards the understanding of ultrafine particles and their significance relative to fine particles (PM2.5) and their role in causing adverse health effects. To address these questions, the EPA's priority research goals include investigating ultrafine exposures and the role of different PM size fractions in health outcomes; exploring the potential for alternatives to the current PM mass-based standards; and examining the exposure and health effects of ultrafine components.

In California, the California Air Resources Board (CARB) has authority to set its own standards through an exemption granted under the Clean Air Act if granted by EPA. Recently, CARB has indicated that regulatory actions may be forthcoming to reduce mass and particle number from engines under the LEV III standards. Specifically, they have proposed an optional PM emission standard beginning in 2014 for super ultra low emission light-duty gasoline vehicles (SULEV) to have a solid particle number less than 6×10^{12} particles per mile, approximately equivalent to the European Union (EU) standard. This would allow vehicles to be certified for the EU standard and sold in CA without certification to the CARB PM emission standard. However, CARB has received feedback on several issues regarding testing procedures for this optional standard and may not include in its final recommendation to the state board due in December 2011. They anticipate now to continue pursuing a particle based standard once they have

addressed concerns over testing procedures for the standard. At this time there are no plans for regulating heavy-duty diesel engines as diesel particulate filters appear to be the only lowest achievable technology for reducing DPM emissions and this is the current technology being used in all new diesel engines. CARB will continue to look at issues on how to measure emissions of semi-volatile and volatile ultrafine particles. Technical staff from CARB will publish their final recommendations for California's LEV-III standards to the Air Resources Board in late October 2011.

Local agencies, such as SCAQMD, can also take steps to mitigate ultrafine particle pollution. These can be mainly through mandating requirements on the transportation fleet and also the design of the transportation infrastructure itself. As discussed earlier, heavy-duty engines emit a higher number of particles in high acceleration and low-speed scenarios. Therefore, measures to alleviate congestion and thus promote free-flow of traffic may lead to a reduction in ambient ultrafine particles.

Recently, SCAQMD has announced plans for a new focus under their Multiple Air Toxic Exposure Study (MATES) program – MATES-IV. This study will include looking at measuring ultrafine particles as well as diesel PM near freeways, arterials, intersections, warehouse areas, rail lines, rail yards, and airports. The MATES-IV will examine ultrafine particles from both mobile platforms and fixed locations. Work plan and protocol development began this summer with advisory group meetings starting this fall and board approval by the end of the year. Monitoring for MATES-IV is planned to start in June 2012 and will continue for 1 year. It will be decided in the planning process if and how SCAQMD would conduct modeling of ultrafine particles.

At least one study, Hussein et al (2007), has attempted to model the near-roadway ultrafine particle number. The modeling exercised in the study used both a dispersion model to simulate the transport and turbulent mixing from the roadway to downwind locations and an aerosol dynamic model to simulate particle number size distribution. Two winter periods were simulated in Helsinki, Finland. Even though the model used a very low mixing volume, the model underpredicted concentrations 65 meters downwind by about a factor of 3. This underprediciton was attributed to uncertainty in the emission factors for the ultrafines and the lack of specificity of those emissions factors under varying ambient conditions (e.g., temperature and relative humidity).

In addition to modeling of ultrafine particles, recent efforts have focused on the development of emission inventories for ultrafine particles (Keogh and Sonntag 2011). In this study, the authors developed what they believe is the first published emission inventory of ultrafine particles generated by on-road mobile sources. This emissions inventory was prepared for urban Southeast-Queensland, covering 46,000 square kilometers, which includes all of the Brisbane metropolitan area. The inventory quantified both ultrafine particles (particle number) and particle mass emissions for the motor vehicle fleet, which included light and heavy duty vehicles and buses. The inventory covers a full size range of particles emitted by motor vehicles, and quantified emission rates for different vehicle types and road links. Average particle number and particle mass emission factors were derived using statistical analysis of more than 600 emission factors as identified in the 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. The average particle number was considered suitable for use in other developed countries for quantifying particle inventories. 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). Results from the study showed that for Brisbane, the annual emissions from on-road mobile sources was in aggregate about the same as annual particle number emissions due to artifact operations from Brisbane International Airport. The ultrafine inventory also revealed that while heavy-duty diesel vehicles only contributed 6% of the vehicle miles traveled, they accounted for more than half of the region's ultrafine particles.

6. Summary

Following are some major conclusions based on the current understanding of ultrafine particles.

- Studies have shown that ultrafine particles are at significantly elevated levels in near-roadway environments. These particles, which constitute most of the particle count in the ambient PM, have been linked to adverse health effects.
- Meteorological conditions have a significant effect on the distribution of ultrafine particles from roadways; the highest exposure to populations in the prevailing downwind direction.
- Although there have been major advances in the development of instrumentation to characterize ambient ultrafine particles, especially their number concentration, some challenges still remain. These mainly include the lack of standardized protocols which are essential for monitoring and developing regulations on ultrafine particles.
- Heavy-duty diesel trucks are the largest emitters of ultrafine particles among all vehicle classes. However, currently there are large uncertainties in relation to vehicle emission factors of ultrafine particles. This is partly due to the variability of emissions based on the fuel-type, fuel composition, and engine load and control technologies/after treatment devices.
- Measurement studies have shown that ultrafine particles are at significantly elevated levels in the vicinity of I-710. These high concentrations drop exponentially with the distance from the freeway and reach urban background levels at around 300 meters.
- Studies have shown that ultrafine particles can penetrate efficiently into indoor environments. Therefore, people living and working in the vicinity of major highways are exposed to high levels of ultrafine particles. Furthermore, in-vehicle exposure while driving on highways is also shown to be significant.
- Currently there are no immediate plans to directly regulate ambient levels. However, CARB has proposed a solid particle number standard for tailpipe emissions for SULEVs starting in 2014.

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