CHAPTER 11

ULTRAFINE PARTICLES

INTRODUCTION

In response to the ever-increasing body of research findings pointing to adverse health effects of ultrafine and nanoparticle air pollution that could potentially be significantly greater than the health effects associated with coarse (PM10) and fine particulate (PM2.5), the District Governing Board, in recent years, began to actively monitor scientific developments in the field of ultrafine particulate matter (PM). In December 2004 a representative of the District Governing Board participated in a nanoparticle health effects and technology forum held in Switzerland. In early 2005, staff prepared a report on the key issues associated with the state of knowledge of ultrafine particles, including how AQMD's policies on particulate emissions fit with the CARB current research and regulatory plans. In spring 2006, the District hosted a three-day conference titled Ultrafine Particles: The Science, Technology, and Policy Issues, with several panels of academia, technology experts, and public policy makers, and more than 400 attendees.

This AQMP presents background information on ultrafine particles and the state of current knowledge on the subject. Potential control strategies discussed herein include effectiveness of current controls, improvement of engine combustion systems, use of low-sulfur fuel, reformulation of lubrication oils, and utilization of effective particulate after-treatment devices in conjunction with catalyst technology. A view of on-going and potential research areas that could facilitate the development of control strategies for ultrafine particles is presented. Lastly, recommendations are made regarding future policy direction and actions.

BACKGROUND AND CURRENT KNOWLEDGE

U.S. EPA is mandated to review, and where necessary, revise ambient air quality standards every five years. The current federal standards for particulate matter air pollution are established for annual and 24-hour periods for PM10 and PM2.5. The state also sets ambient air quality standards for annual and 24-hour PM10 and annual PM2.5. Presently, there are no efforts at the federal or state level to consider separate air quality standards for ultrafine particulates.

Particulate matter is broadly classified as "coarse" PM with a diameter of 2.5μm to 10 μm, or "fine" (PM2.5) with a diameter less than 2.5 μm. PM10 includes all particles with diameters less than 10 μm. Ultrafine particles are loosely defined as those with a diameter less than 0.1 μm (or 100 nm). Ultrafines are sometimes alternatively referred to as nanoparticles, often with an upper diameter of 0.05μm (or 50 nm).

Both the federal and California PM ambient air quality standards are based on mass concentrations in air. Due to their small size, ultrafine particles generally make up a very small fraction of the ambient PM2.5 or PM10 mass (less than 10%), but make up the majority of airborne particles by number. As an example, a particle mass concentration of approximately 10 $\mu g/m^3$ is equivalent to a count of one particle per cm^3 for particulates with a diameter of 2.5 μm, but equivalent to a count of more than 2 million particles per cm^3 for particles of a diameter of 0.02 μm (Oberdorster, et al. 1995).

AMBIENT CONCENTRATIONS

Ultrafine particle number and mass concentrations are not routinely measured in the U.S. Thus, there is little data on long-term trends. However, there are a few published reports of ultrafine particle counts and characterization. Recent measurements taken in Southern California show a wide range in particle counts in different environments (Westedahl et al., 2003). The highest counts are found very near mobile sources, with some of the highest concentrations observed on busy roadways. Examples of particle counts found in different areas are shown below in Table 11 - 1.

TABLE 11 - 1

Ultrafine Particle Counts in Southern California

Area	**Particle Number Concentration ($particles/cm^3$)**
Coastal area	600-2000
Office spaces	500-2000
Urban air	10,000 - 40,000
Freeways	40,000 – 1,000,000
Industrial site	Up to 100,000

From Westerdahl, 2003

In the urban environment, motor vehicles are a major source of ultrafine particulates. Other recent studies conducted in Southern California have shown high counts of particulates near freeways. Substantially higher numbers of particles are found near the roadway, while a sharp reduction in particle count has been shown to occur within 100-300 meters downwind of the roadway (Zhu, 2002a, 2002b).

As high particle number concentrations are very localized and dependent on nearby source activity, they exhibit large geographical and temporal variation. Monthly averages for particle number count have been collected at several urban sites in Southern California as part of the Children's Health Study (CHS). Average particle counts tend to be higher in winter compared to spring and summer. The higher number counts during the winter months are likely due to lower temperatures, favoring particle formation by condensable organics freshly emitted from vehicles, as well as a decreased atmospheric mixing height and more stagnant conditions increasing the influence of localized emissions (Sioutas, 2004). The highest ultrafine particle mass measurements also occur during the winter months, with the ultrafine fraction contributing 10% or less of the total average PM10 mass (Sardar, et al. 2005).

Figure 11-1 shows a comparison of monthly average particle counts for the period of October through December 2001. The highest monthly averages were found at monitoring sites in Long Beach, Upland, Mira Loma, and Riverside (Peters, et al. 2004).

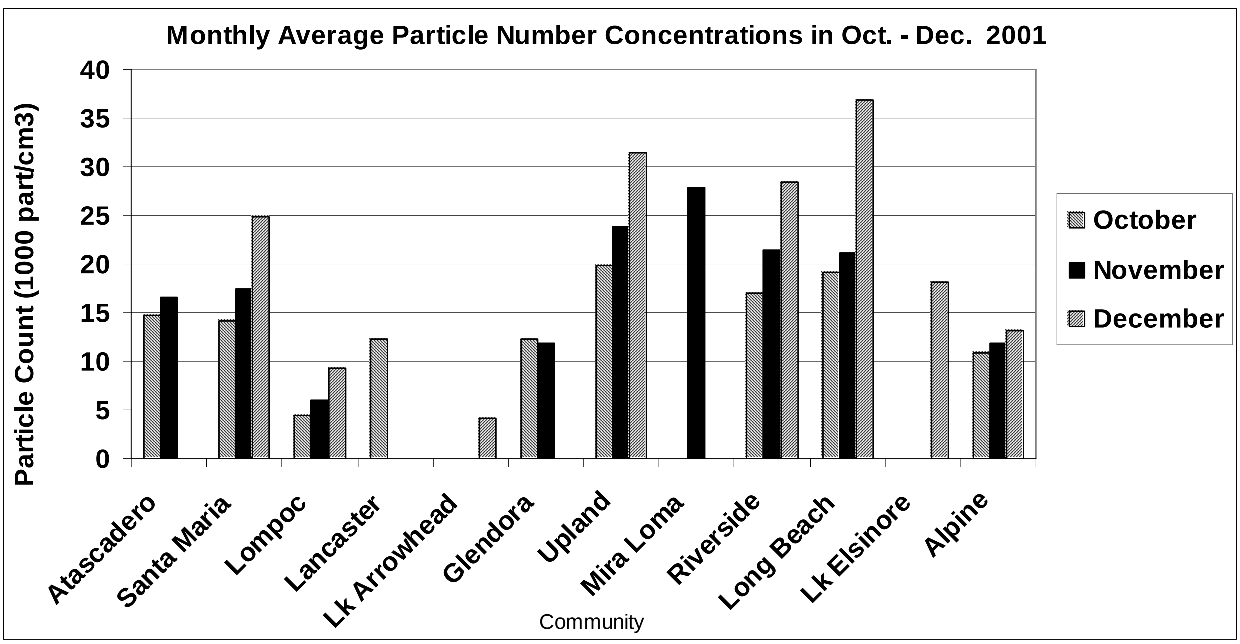

FIGURE 11 - 1

Monthly average particle number concentrations in CHS communities in October–December 2001 (Peters, et al. 2004)

HEALTH EFFECTS

Numerous studies have associated particulate matter levels with adverse health effects, including increased mortality, hospital admissions, and respiratory disease symptoms (U.S. EPA, 2004). Each year, more is known about health effects associated with PM exposure and its mechanisms. The vast majority of these studies used particle mass as the measure of exposure. Some researchers have postulated, however, that ultrafine particles may be responsible for some of the observed associations between particulate matter and health outcomes (Oberdorster, et al. 1995; Seaton, et al. 1995).

Results from several studies and postulated health effects mechanisms suggest that the ultrafine portion of PM may be important in determining the toxicity of ambient particulates. Some of these findings are discussed below.

For a given mass concentration, ultrafine particles have much higher numbers and surface areas compared to larger particles. Particles can act as carriers for other agents, such as trace metals and organic compounds, which can collect on the particles' surfaces; the ultrafine particles with larger surface area may transport more of such toxic agents into the lungs than larger particles. Furthermore, smaller particles can also be inhaled and deposited deeper into the lungs than larger particles. As much as 50% of the particles with 0.02 μm or smaller are estimated to be deposited in the alveolar region of the lung.

In laboratory toxicity studies, a greater inflammatory and oxidative stress response has been elicited from ultrafine particles compared to larger particles at comparable mass doses. Oxidative stress is a term to describe cell, tissue or organ damage caused by reactive oxygen species. Oxidative stress and the biological production of numerous chemicals associated with oxidative processes have been postulated to underlie at least some of the observed effects of particulates. For example, studies using laboratory cell preparations have suggested that the substances adsorbed onto ambient ultrafine particles are responsible for some of the effects observed, rather than the particles themselves (Xia, et al. 2004).

After inhalation, ultrafine particles may penetrate rapidly into lung tissue; and some portions may be translocated to other organs of the body (Oberdoster, et al. 2002; Kreyling, et al. 2002; Nemmar, et al. 2002). A recent study also found evidence that particles may be translocated via neural cells from the nose and pharynx to the olfactory bulb of the brain (Oberdoster, 2004).

Additionally, ultrafine particles were found to penetrate cells and subcellular organelles. In cell cultures exposed to ambient particles, ultrafine particles were found in mitochondria where they induced structural damage (Li, et al. 2003).

Almost all epidemiology studies of particulate effects focus on measurements of particulate mass, either PM10 or PM2.5. However, a few studies have also measured ultrafine particle number counts. For example, in studies conducted in Germany, both the mass and number of particles were assessed in relation to mortality rates (Wichmann, et al. 2000; Stolzel, et al. 2003). Both the mass and number of ultrafine particles were associated with elevations in daily non-accidental mortality. Ultrafine particle number, as well as fine particle mass, has also been found to be associated with impaired lung function and medication use among individuals with asthma (von Klot, et al. 2002; Wichmann, et al. 2000).

European regulations (Euro III, IV, and V) on PM emissions from mobile sources are established on the basis of mass emissions. The Euro IV/V PM emissions limit is 0.02 gram per kilo-watt-hr (g/kWh), an 80 percent reduction in the mass of PM limit required under Euro III (0.10 g/kWh). These regulations lack standards limiting ultrafine particle number emissions because there is currently no widely acceptable test protocol for measuring particle numbers. In recognition of harmful health effects of ultrafine emissions, a Particulate Measurement Program (PMP) was established to assess the appropriateness of a particle number standard, and develop and test a new protocol for measuring particulate emissions. Once PMP work is completed, the European PM standards will be changed to reflect the new test protocol, and a PM number standard may be implemented.

While the information on the health effects of ultrafine particles is limited, these and other studies suggest that ultrafine particles may have significant health effects greater than or independent of the effects due to the larger particles that comprise the majority of ambient PM mass.

SOURCES

PM emissions derive from many natural and man-made activities. This discussion is focused on ultrafine PM emissions formed during engine combustion and in the atmosphere, immediately after leaving the tailpipe as emitted gases condense and rapidly dilute and cool. Internal combustion engines have been identified as significant sources of ultrafine PM. A significant proportion of diesel emission particles have diameters smaller than 100 nm (0.1 μm). Particles emitted from gasoline-powered engines are generally less than 80 nm (0.08 μm) in diameter. Particles from compressed natural gas (CNG) fueled engines are smaller than from diesel emissions, with the majority between 20 and 60 nm (0.02 μm – 0.06 μm). Typically, these particles are a complex mixture of solid and more volatile particles. The solid particles are formed during the combustion process in the engine and are generally larger than the volatile particles. They consist mainly of agglomerated elemental carbon (soot) and act as an absorbent for some of the more volatile organic species formed during combustion. The smaller, more volatile particles are generally spherical. While some of the smaller, spherical particles may be formed in the engine or tailpipe, the majority are formed outside of the engine by the nucleation of hydrocarbon, sulfuric acid, and water vapor as the exhaust undergoes natural processes of dilution and cooling in the atmosphere. The number, size and growth rates of these more-volatile particles depend on variables affecting condensation such as, dilution rate, temperature, residence time, surface area of pre-existing particles, and humidity (Khalek, et al., 1999, 2000). Figure 11-2 shows a typical diesel engine exhaust mass and number -weighted size distributions.

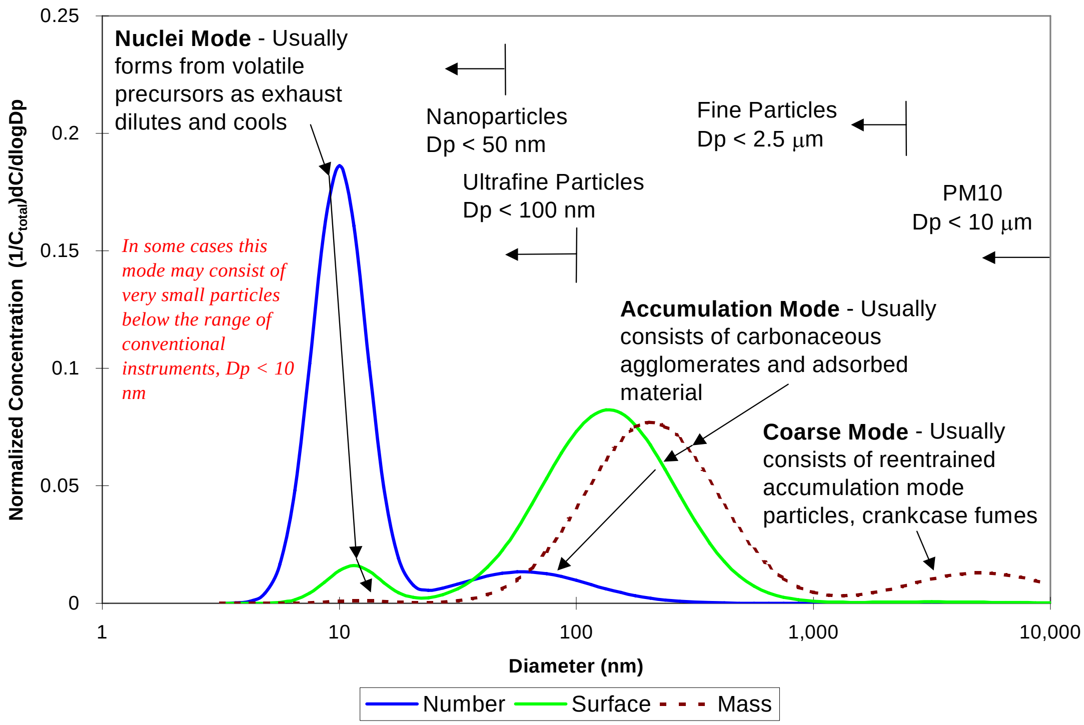

FIGURE 11 - 2:

Typical Mass and Number-weighted Size Distributions of Diesel PM (Kittelson, 1998).

The number of ultrafine particles formed outside the tailpipe is largely influenced by the available surface area of the solid particles. As the total PM mass emissions are reduced by advanced engine technology and effective PM aftertreatment devices, the number, and thus surface area of the larger, solid particles is significantly lowered. With fewer larger particles on which to condense, cooled gas phase species will instead nucleate to form new particles, leading to production of ultrafine numbers as exhaust is diluted and cooled. These particles are formed from condensing gas-phase hydrocarbon precursors. Studies have shown that the hydrocarbon particle precursors are effectively removed by oxidation catalyst technology.

The formation of ultrafine particle numbers in and near the tailpipe is also influenced by the sulfur content of the fuel and the composition of lubricating oil. A fraction of sulfur in fuel is oxidized to sulfur trioxide, SO3. The SO3 binds with water, forming sulfuric acid, one of the gas-phase species that can nucleate to form new smaller particles. Many studies (Kittelson, et al. 2002; Ristovski, et al. 2002a; Ristovski, et al. 2002b; Sakurai, et al. 2001; Wei, et al. 2001) have addressed the influence of fuel sulfur level on ultrafine particle formation from vehicles. In general, most of these studies suggest that a significant reduction of the number of ultrafine particles emitted occurs when fuel sulfur levels are reduced.

Recent studies comparing regulated emissions from diesel and natural gas (CNG) engines show that CNG engines emit a lower level of PM mass emissions than diesel-powered engines. It is probable that lubricating oils used in both diesel and CNG engines produce gas phase ultrafine precursors either due to the sulfur in the oil or components of reformulated oil. In the absence of larger, solid particles, the precursors in lube oil (sulfur, metals and heavy hydrocarbons) undergo nucleation in the vehicles' exhaust systems or immediately after exiting the tailpipe. The exhaust temperatures have been found to decrease from approximately 1,000^{o}F (at the exhaust manifold) to 400^{o}F – 600^{o}F at the outlet of the exhaust. It should be noted that sulfuric acid nucleates to form a mist at temperatures below 620^{o}F. When the sulfuric acid in the exhaust nucleates, the nuclei serve as absorption sites for the semi-volatile and heavier hydrocarbons. Reducing the sulfur and metal content of lubricating oils, as well as using oxidation catalyst technology to reduce hydrocarbon precursors, can reduce the particle numbers from such sources.

CONTROL TECHNOLOGIES

In response to U.S. EPA's and CARB's tighter engine exhaust emissions standards, vehicle and engine manufacturers, emission control manufacturers, and researchers have continued to direct considerable efforts and resources to developing strategies to reduce PM and other criteria pollutant mass emissions. These efforts have resulted in many options available for improving engine design and developing aftertreatment devices to achieve greater emission reductions. Overall, an improved engine combustion system is effective in reducing engine-out total PM mass emissions (mostly accumulation mode particles 0.1 μm to 1 μm), while a well-engineered particulate filter and oxidation catalyst are effective in removing both larger (accumulation/coarse mode) and smaller (ultrafine) particles.

Particulate filters are generally flow-through devices capable of achieving over 90% reduction of the solid portion of the total exhaust particles, particles mostly in the accumulation mode. However, they could be minimally effective or totally ineffective in controlling the gas phase precursors of ultrafine particles unless an oxidation catalyst is used in conjunction with the filter. With most of the solid particles removed, nucleation, rather than condensation, of gas phase species is favored, thereby promoting increased particle number emissions. Specially formulated oxidation catalysts are capable of removing more than 90% of the soluble organic fraction (SOF) as well as ultrafine particles on a number basis. Thus, an effective control technology should be based on a system approach involving both a particulate filter and oxidation catalyst technology. In a recent study to demonstrate the effectiveness of particulate filter technology on reducing particulate emissions from natural gas engines, the research found that total PM emissions were significantly reduced and the filter was capable of reducing ultrafine particles by 99 percent.

Oxidation catalyst technology (OCT) is effective in removing the SOF fraction of total emissions as well as ultrafine particles formed later in the exhaust. Its effectiveness, however, depends on whether the catalyst is formulated to produce little or no sulfate emissions at high temperature. In fact, special catalyst formulations must be employed to hinder the catalytic generation of sulfate particles from sulfur dioxide present in the exhaust gas. While OCT is effective in reducing SOF fraction and smaller particles, it has little effect on larger accumulation or coarse mode particles. Studies have substantiated the effectiveness of OCT in removing ultrafine particles.

Holmen and Ayala (2002) recently studied the effect of particulate filters and oxidation catalyst on the characteristics of particle emissions from heavy-duty CNG and diesel transit buses. The mix of buses included buses equipped with particulate filters (diesel) and oxidation catalysts (CNG). The study showed that particulate filters effectively reduce diesel particles in both in the ultrafine and accumulation modes. In addition, the oxidation catalyst equipped CNG bus showed significant reduction in ultrafine particles.

Gautam, et al. (2004) also measured the particle number emissions from an Orion natural gas fueled transit bus powered by an engine operating at 20 miles per hour under steady state conditions and equipped with OCT. The result of that study showed OCT to be more effective in removing ultrafine particle number at hot versus cold conditions, with the particle count reduced to near background levels. When the same bus was equipped with a catalyzed filter installed upstream of the OCT, the volatile organic species that participate in forming new particles were oxidized by the OCT; and hence this test vehicle showed a near absence of any particles in the exhaust stream.

CURRENT ACTIVITIES

DISTRICT-SPONSORED RESEARCH

Some studies are now showing an increase in the number of ultrafine particles in emissions from engines with low PM mass emissions and engines equipped with currently available aftertreatment devices. The results of these studies and the potential for adverse health effects of particle number concentrations have prompted the District to co-sponsor several projects to investigate ultrafine mass and number of particle emissions from engines. AQMD and West Virginia University recently conducted a study to chemically characterize exhaust emissions from a 40-foot Orion bus powered by a Cummins C8.3G plus CNG engine equipped with a catalyzed particulate filter and an oxidation catalyst.

The District is sponsoring a study on the contribution of lubricating oil to PM emissions from a 40-foot Orion bus with a Cummins C8.3G Plus engine equipped with a catalyzed particulate filter. This study assessed the performance and emission reduction potential of the particulate filter and oxidation catalyst on total PM mass and number. Finally, the District is working to optimize an oxidation catalyst technologies to achieve the maximum reduction possible of benzene, formaldehyde, total PM (ultrafine and nanoparticles), and non-methane hydrocarbon emissions.

Research to assess the health effects of ultrafine particles on elderly individuals is being co-funded by the National Institutes of Health and CARB. Groups of volunteers with heart disease are being followed over time, and any changes in cardiovascular health and particulate exposures are being measured.

CARB ULTRAFINE AND NANOPARTICLE PROGRAM

Over the last few years, CARB has engaged in several programs to measure PM emissions and assess the influence of ultrafine particles on public health. CARB (Holmen and Ayala, 2002) recently collaborated with other public agencies and research institutions to collect emissions data from two late-model heavy-duty transit buses powered by similar engine and fueled by Emission Control Diesel (ECD-1) and CNG. The goals of this project are to: (1) examine the impact of driving cycle on emissions; (2) compare toxicity among new and cleaner heavy duty engine technologies in use in California; and (3) assess total PM and ultrafine particle emissions.

CARB is conducting ambient air measurements at several local freeway and surface street traffic areas in Southern California to collect real-time on-road measurements of pollutants, including black carbon, polycyclic aromatic hydrocarbons (PAH), and particle count and size distribution data of particles between 5 and 600 nm in diameter. A previous study, cited above, deployed condensation particle counters (CPCs) at the 12 Children’s Health Study air monitoring sites in Southern California to provide a continuous record of the ultrafine particle count concentration in ambient air. Mobility particle sizers were periodically deployed at each monitoring station to obtain spatial and temporal information with respect to the particle size distribution between 10 and 450 nm. Finally, CARB is sponsoring a research project to investigate possible links between exposure to freeway-related ultrafine particles and changes in measures of cardiovascular function.

CARB staff does not have a plan at this time to regulate emissions of ultrafine particles on a mass or number basis, but will continue to study unresolved issues relating to ultrafines, such as formation, ambient concentrations, spatial and temporal variability, measurement issues, test protocols, and health impacts.

FUTURE ACTIONS

RESEARCH NEEDS

There are key areas pertaining to ultrafine particulates and their impacts on health and the environment where further research is needed. When developing technologies to reduce the mass of particulate matter, there should also be a focus on technologies to significantly reduce engine-out ultrafine particles and gaseous precursors to ultrafine particles. With the goal of protecting health in mind, the following recommendations are offered for further research and refinement of control strategies:

1. Encourage and support projects that will lead to better understanding of ultrafine particle formation and composition, including further analysis of the relationship between PM mass, surface area, and number concentration with respect to reduction strategies, potential standards, and health impacts.
2. Further support studies into the health effects of ultrafine particles.
3. Develop and finalize measurement methodologies, testing protocols, and on-road emission factors.
4. Further characterize exposures to, and toxicity of, ambient ultrafine particles.
5. Use fuels with reduced sulfur content to minimize formation of sulfate-based ultrafine particles.
6. Develop advanced engine technologies to reduce engine-out ultrafine particles and gas-phase precursors.
7. Develop strategies for the use of both particulate filters and oxidation catalysts in liquid and gaseous powered vehicles with the catalyst specially formulated to reduce and/or prevent creation of gas-phase precursors of particles, to the extent possible.
8. Assess the impact of lubrication oil on engine emissions and develop advanced or improved lubricating oil formulated to reduce oil derived emissions, including the development and demonstration of advanced re-formulated lubricating oil in heavy-duty vehicles.
9. Work with other public agencies and the private sector to establish lubrication oil standards to reduce emissions of ultrafine particles.
10. Conduct studies to account for the existing and aging (legacy) fleet of diesel trucks in the inventory of ultrafine particle emissions.

POLICY FUTURE

Currently, it is recognized that ultrafine particulates are predominately formed through combustion processes and the highest concentrations are associated with mobile sources. Furthermore, ultrafine particles have been implicated in adverse health effects independent of PM mass. Newer generation control technologies have been demonstrated to be cost-effective and are currently available. Current and future regulatory requirements to reduce engine emissions necessitate the use of particulate filters (with oxidation catalyst coatings) and oxidation catalysts in order to meet the current and future emission standards. However, it is necessary to proceed slowly in establishing regulatory requirements in this new area because: additional health studies will be beneficial to fully understanding the impacts of ultrafine particles; further consideration is appropriate relative to the regulation of ultrafine particles on the basis of number versus mass; and the regulatory action to be taken at the local, state, and federal levels, respectively.

It is with this knowledge that the following key recommendations are made:

- Encourage use of after-treatment technologies combined with oxidation catalyst technology to produce concurrent benefit of ultrafine particle reduction.

- Encourage equipment and vehicle manufacturers to develop diesel particulate filters (DPF) with integrated controls for ultrafines since the additional cost may be relatively minor.

- Work with CARB, US EPA, and other stakeholders in conducting research studies and control strategy development efforts.

- When developing control measures for the reduction of PM10 and PM2.5, consideration should be given for reducing any undesired effects on ultrafine number emissions, where feasible.

- Work with CARB and US EPA in developing strategies to reduce ultrafines from mobile and stationary sources.

- Encourage auto manufacturers to include ultrafine particle filters in passenger vehicles to reduce exposure to on-road emissions of particle mass and number.

- Consider ultrafine PM issues in AQMD's PM control and air toxics strategy.

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