

EXECUTIVE SUMMARY

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Title: Southern California Supersite

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Description and Objective of Research

The overall objective of the Southern California Supersite (SCS) was to conduct research and monitoring that contributes to a better understanding of the measurement, sources, size distribution, chemical composition, physical state, spatial and temporal variability, and health effects of suspended particulate matter (PM) in the Los Angeles Basin (LAB). The SCS projects followed the three major research areas called for by the U.S. EPA for the Supersites Program, i.e. *PM Characterization*, *Support of Health Effects and Exposure Research*, and *Methods Testing*. Intensive aerosol measurements, well beyond the traditional PM_{2.5} mass, sulfate and nitrate concentrations, were conducted in several areas of the LAB. These included particle number concentrations, size distributions, and detailed PM chemical composition as a function of particle size. Sampling locations were chosen to provide wide geographical and seasonal coverage, including urban “source” sites and downwind “receptor” sites. The primary sampling facility, a mobile Particle Instrumentation Unit (PIU), was deployed to several locations to conduct a wide range of PM measurements. Sampling in each site lasted for 6-12 months. Intensive PM measurements were also conducted up and downwind of several freeways of the LAB, to characterize near-roadway exposure environments and to support several in vivo and in vitro health studies. In addition a number of existing PM monitoring sites in the LAB supporting the USC Children’s Health Study (CHS), operated by South Coast Air Quality Management District (SCAQMD) and California Air Resources Board (CARB), were used as satellite sites to obtain information on the variability of PM size and composition in the LAB. The SCS activities were integrated with multidisciplinary research in exposure assessment, toxicology, and epidemiology of the U.S. EPA STAR program funded Southern California Particle Center and Supersite (SCPCS). The monitoring activities of the SCS were linked with toxicology studies in the LAB using a mobile PM Concentrator facility to investigate health effects associated with exposures to ultrafine, fine and coarse particles. Finally, the PIU facility was successfully used as a platform to develop, test, and evaluate numerous PM measurement instruments and sampling technologies, including several monitors for semi-continuous size fractionated mass and chemistry, personal PM exposure monitors, particle concentration technologies, and particle counting devices.

In the area of *PM Characterization*, numerous long-term monitoring and shorter-term intensive studies were carried out successfully resulting in several significant findings. Weekly measurements of 24-hour size-fractionated PM chemistry over 3 years of sampling showed interesting seasonal and spatial differences within the LAB. For instance, ultrafine particles (UFP) consisted of mostly organic carbon, with higher wintertime levels at the source sites due to increased organic vapor condensation at lower temperatures from vehicles. Higher ultrafine organic carbon levels were seen at the receptor areas in the summer due to secondary organic

aerosol formation by photochemical reactions and increased advection of polluted air masses from upwind. (Sardar et al., 2005). There has been some debate as to whether a 1.0 μm or a 2.5 μm cut-point provides good separation between accumulation mode particles (combustion emissions and photochemical formation and processing) and coarse mode, mechanically-generated particles (such as soil and road dust). SCS analysis showed that inter-modal PM (1.0-2.5 μm) consists of a significant portion of particles that are more similar in chemical composition to PM₁ particles, suggesting that a PM₁ standard would not constitute an unambiguous separation of coarse and fine mode PM in this urban air shed. (Geller et al., 2004). Measurement of daily size-fractionated ambient PM₁₀ metals was conducted at source (Downey) and receptor (Riverside) sites. The main source of crustal metals (e.g., Al, Si, K, Ca, Fe and Ti) can be attributed to the re-suspension of dust at both source and receptor sites, and were predominantly present in super-micron particles. At the source site, potentially toxic metals (e.g., Pb, Sn, Ni, Cr, V, and Ba) are predominantly found (70–85%, by mass) in the sub-micron particles. Coarse PM metal concentration trends were governed by variations in wind speed, whereas the diurnal trends in the fine PM metal concentrations are found to be a function both meteorological conditions and proximity to sources. (Singh et al., 2002). The very small mass of UFP has posed a great challenge in determining their size-dependent chemical composition using conventional aerosol sampling technologies. Implementing two technologies in series (the USC Ultrafine Concentrator and the MSP NanoMOUDI) allowed for shorter-term size-fractionated ultrafine chemistry measurements. UFP were measured in source and receptor sites during three consecutive 3-hour time intervals (i.e., morning, midday and afternoon). The results indicate a distinct mode in the 32-56 nm size range that is most pronounced in the morning and decreases throughout the day. While the mass concentrations at the source site decrease with time, the levels measured at Riverside (receptor site) are highest in the afternoon with a minimum at midday. These results indicate that in this area, UFP is generated by primary emissions during the morning hours, whereas secondary aerosol formation processes become more important as they day progresses. (Geller et al., 2002). Another study with the Nano-MOUDI explored the size-fractionated ultrafine (10-180 nm) chemical composition at urban source sites (USC and Long Beach) and inland receptor sites (Riverside and Upland) in the LAB over three different seasons. Two week composite samples showed a distinct OC mode was observed between 18 and 56 nm in the summer, likely due to photochemical secondary organic aerosol formation. Collocated continuous measurements of particle size distributions and gaseous pollutants helped to differentiate UFP sources at each site (Sardar et al., 2004).

Other SCS studies looked at the organic carbon component of PM in more detail. Size-fractionated samples analyzed for 15 priority pollutant polycyclic aromatic hydrocarbons (PAH) showed that the fraction of PAH mass in the coarse mode increased with increasing PAH vapor pressure in the warmer months. PAH size distributions peaked in the 0–0.18 μm size range in samples collected in central Los Angeles (source) and in the 0.18–2.5 μm size range at downwind receptor sites, consistent with long range transport and advection (Eiguren-Fernandez et al., 2003; Miguel et al. 2004). In addition to PAHs, ambient measurements of nitro-PAHs were carried out in source and receptor sites. Atmospheric concentrations of nitro-PAHs are of interest because they are known to be potent mutagens and carcinogens. Nitro-PAH levels were highest in Riverside (receptor) during summer, as a result of enhanced summertime photochemistry. (Arey et al., 2005; Reisen et al., 2003). Other individual organic compounds can be used as tracers for primary sources of ambient PM in chemical mass balance receptor models. By examining the seasonal, temporal, geographical, size-fractionation, and inter-correlations of individual organic compounds, the sources and atmospheric fate of these tracers can be better understood and their utility as molecular markers can be assessed. A high-flow rate, low pressure-drop UFP separator collected sufficient mass for organic speciation of ultrafine and accumulation mode aerosol on a diurnal basis. Sampling was conducted at two sites (source and receptor) over two seasons (summer and winter). Hopanes, used as organic markers for vehicular emissions were found to exist primarily in the ultrafine mode, while levoglucosan, an indicator of wood combustion, was more prevalent in the accumulation mode particles. An indicator of photochemical secondary organic aerosol formation, 1,2 benzenedicarboxylic acids, was found primarily in the accumulation mode and varied with site, season and time of day as one would expect for a photochemical product (Fine et al., 2004b).

Continuous and semi-continuous instrumentation showed that diurnal trends of total particle number concentration and elemental carbon (EC) are almost identical throughout the day and irrespective of season at source sites, demonstrating a primary vehicular source. This agreement between EC and particle

number was not observed in receptor sites during the warmer months of the year, again suggesting photochemical particle production (Kim et al., 2002). The size distribution of UFP in source sites was generally unimodal with a mode diameter of 30–40 nm and without significant monthly variations, while the number-based particle size distributions in receptor sites were bimodal, with a significant increase in the larger particles (modes near 100 nm) in the summer (Fine et al., 2004). As part of our evaluation of sampling artifacts from both continuous and time integrated PM measurement methods, we conducted a study to obtain insights into the dynamics of volatilization from a heated TEOM filter. Our experiments showed that under ambient conditions, the mass lost from the Differential TEOM tracks well the ambient particulate nitrate concentration (Hering et al., 2004).

In the area of *Supporting Health Effects and Exposure Research*, numerous atmospheric measurements were made to characterize exposure parameters for toxicological and epidemiological studies. Very highly-publicized studies showed that the exposure to UFP decrease in a quasi-exponential manner with distance from freeways, due to atmospheric dilution and evaporation of particle-bound labile species. Total particle number concentration is directly related to traffic density (Zhu et al., 2002a). The major differences between diesel vs. gasoline traffic freeways was seen in the higher number concentrations and elemental carbon (EC) corresponding to diesel traffic (Zhu et al., 2002b). The decay rates of particle numbers, CO and BC with distance from freeways is greater in summer than in winter due to high dilution rates. Particle number concentrations in the size range of 6–12 nm is significantly higher in winter than in summer, suggesting that wintertime conditions favor greater particle formation, possibly due to increased condensation of organic vapors near the tailpipe. (Zhu et al., 2004). The data from these freeway measurement studies were simulated with a multi-component sectional aerosol dynamic model. Condensation / evaporation and dilution were demonstrated to be the major mechanisms in altering aerosol size distribution, while coagulation and deposition play only minor roles (Zhang et al., 2004).

In collaboration with CARB, we developed a mobile platform designed to characterize UFP and associated pollutants inside vehicles, during commute in Los Angeles freeways and residential streets. Freeway exposure regimes were frequently an order of magnitude higher than on residential streets for UFP, NO_x, BC, and CO, with higher NO_x and BC values observed in diesel traffic freeways than those with mostly light duty vehicle traffic (Westerdahl et al., 2005). Another study to evaluate contributions of vehicle generated ultrafine particles to indoor environments showed particle number concentration I/O ratios to be strongly dependent on particle size and ventilation conditions. Highest I/O ratios (0.6–0.9) were usually observed for larger ultrafine particles (70–100 nm), while the lowest I/O ratios (0.1–0.4) occurred typically around 10–20 nm (Zhu et al., 2005). These results were further supported by a study in which we examined volatility of penetrating ultrafine outdoor particles, predominantly from freeway emissions, into indoor environments using a tandem differential mobility analyzer (TDMA) system. Our findings suggest that outdoor particles are more volatile than indoor aerosols. Evaluation of outdoor particle volatility as a function of distance to the freeway revealed that aerosol volatility decreases with increasing distance from the source (Kuhn et al., 2005). Physical and chemical characteristics, including volatility of PM in the proximity of a Light-Duty Vehicle (LDV) freeway were also measured and analyzed. The volatile component ranged from about 65% volume of 120 nm particles heated to 110°C, to 95% volume of 20 nm particles (Kuhn et al., 2005b). Our freeway PM measurements were compared to those performed in two bores of the Caldecott Tunnel in Northern California. One bore (Bore 1) is open to both heavy- and light-duty vehicles (HDV-LDV) while heavy-duty vehicles are prohibited from entering the second bore (Bore 2). A strong association between particle number and normalized vehicle speed ($R^2 = 0.69$) was observed, and heavy-duty diesel vehicles showed higher particle number emissions than light-duty vehicles. Compared to previous studies at the Caldecott tunnel, less particle mass but more particle numbers (by factors of 2–4 fold) are emitted by vehicles than was the case 7 years ago. As the emissions of carbonaceous PM of newer engines decreases, the formation of nucleation mode particles is favored due to the reduction of the available surface for adsorption of the semi-volatile material. The resulting supersaturation of the mostly organic vapor increases the production of nano-particles by nucleation (Geller et al., 2005). It has been hypothesized that since UFP originate from vehicular emissions, the concentrations of gases such as CO, NO, or NO₂ that also originate from traffic sources can be used as a surrogate measures of UFP. Our studies indicated an overall lack of significant associations between hourly and 24-hr PN versus gaseous co-pollutant concentrations, indicating

that gaseous co-pollutant levels are not good surrogates of particle number, a result which can be attributed to the differences in the sources and formation mechanisms of these pollutants (Sardar et al., 2004b).

In late October 2003, 13 large Southern California wildfires burned more than 750,000 acres of land, destroyed over 3500 structures, and displaced approximately 100,000 people. As part of our Supersite program, we examined the actual impact of these wildfires on air quality in urban Los Angeles (LA) using “opportunistic” data from other air pollution studies being conducted at the time of the fires. In general, the wildfires caused the greatest increases in PM₁₀ levels (a factor of 3–4) and lesser increases in CO, NO, and PN (a factor of up to 2). Ozone concentrations dropped during the fire episode. Particle size distributions of air sampled downwind of the fires showed number modes at diameters between 100 and 200 nm, significantly larger than that of typical urban air. The particles in this size range were shown to effectively penetrate indoors, raising questions about the effectiveness of staying indoors to avoid exposure to wildfire emissions (Phuleria et al., 2005).

Our PM physico-chemical measurements were conducted concurrently and thus supported several in vitro studies, which were funded by the SCPCS. In one of these studies, we have developed three assays for PM redox activity. One of the assays utilizes the reduction of oxygen by dithiothreitol, which serves as an electron source. Redox activity was highest in the ultrafine fraction. Comparison of the redox activity with chemical composition showed a reasonable correlation of redox activity with elemental carbon ($r^2 = 0.79$), organic carbon ($r^2 = 0.53$), and with benzo [ghi] perylene ($r^2 = 0.82$), consistent with species typically found in mobile emission sources. (Cho et al., 2005). UFP were most potent toward inducing cellular heme oxygenase-1 (HO-1) expression and depleting intracellular glutathione, which has relevance to induction of oxidative stress. The small size of UFP allows better tissue penetration; we used electron microscopy to study subcellular localization. UFP and, to a lesser extent, fine particles, localize in mitochondria, where they induce major structural damage. (Li et al., 2003; Xia et al., 2004). In vivo studies using concentrated ambient particulates (CAPs) in the vicinity of freeways were also supported in part by the SCS. Virtually all of these studies showed that CAPs exacerbate airway inflammation and allergic airway responses in a sensitized mouse models, with the responses being greater for UFP, especially those in close proximity (within 50 m or less) of a freeway (Kleinman et al., 2004; Campbell et al., 2004). Neurological inflammation and cardiovascular effects were also observed in close proximity to freeways.

In the area of PM measurement *Methods Testing*, numerous novel PM measurement methods were developed and evaluated. Several continuous mass monitors that measure the concentrations of ultrafine, coarse or PM_{2.5} in hourly or more frequent time intervals were shown to compare favorably with filter-based measurements. These include a coarse PM monitor using a concentrating inlet and a standard TEOM (Misra et al., 2001; Misra et al. 2003), a new differential TEOM monitor for PM_{2.5} (Rupprecht and Patashnick Co., NY) that corrects for volatilization effects (Jaques et al., 2004), and an UFP mass monitor using a size-selective inlet coupled to a Beta Attenuation Monitor (BAM) (Chakrabarti et al., 2004). The ability of a coupled Scanning Mobility Particle Sizer and Aerodynamic Particle Sizer tandem (SMPS–APS) to accurately measure PM₁₀ and PM_{2.5} concentrations was tested with encouraging results (Shen et al, 2003). The need for continuous personal monitoring for exposure to PM has been demonstrated by recent health studies showing effects of PM exposure on time scales of less than a few hours. Towards that goal, we evaluated an active-flow personal DataRAM for PM_{2.5} (pDR, Model 1200; MIE Inc., Bedford, MA), designed as a wearable monitor to continuously measure particle exposure (Chakrabarti et al., 2004b). In collaboration with Aerosol Dynamics Inc. (ADI), we developed and evaluated a new size-fractionated continuous fine particle nitrate monitor (Fine et al., 2003). Also in collaboration with ADI, we evaluated a new water-based condensation particle counter (W-CPC), designed by ADI. The results indicated good correlation between the W-CPC and butanol-based CPCs, with R² values ranging from 0.74-0.99 (Biswas et al., 2005). We developed and evaluated a high-volume, multiple rectangular (slit) geometry jet impactor. Operating with a pre-selective inlet that removes particles larger than 2.5 μm in aerodynamic size, the impactor has been designed to separate ultrafine ($< 0.15 \mu\text{m}$) from the accumulation mode range ($0.15 < dp < 2.5 \mu\text{m}$). The impactor operates at a flow rate 550 l min⁻¹ and at a very low pressure drop of 0.020 kPa (Misra et al, 2002). We have used two different particle mass spectrometers, the Aerodyne AMS and the UC Davis RSMS-3, to evaluate the performance of our Versatile Aerosol Concentration Enrichment System (VACES). RSMS-3 hit rates increased and results showed that the VACES introduces insignificant artifacts (Kim et al., 2001a; Kim

et al., 2001b; Zhao et al., 2005). Studies with the AMS also showed that the shape of the sulfate, organics, and ammonium nitrate mass-weighted size distributions were essentially preserved during passage through the concentrator for all the experiments performed, with a mass enhancement factor of about 10 to 20 depending on the experiment (Khlystov et al., 2005).

In conclusion, the SCS has become an invaluable resource to the abundant ongoing and planned PM monitoring, health and modeling studies in Southern California. The project resulted in about 60 peer-reviewed publications in leading aerosol, environmental, and health effects journals.

Supplemental Keywords: Keywords: airborne PM, PM, PM_{2.5}, UFP, size distribution, chemical speciation, freeways, secondary formation, measurement artifact, continuous monitors

Relevant Web Sites: www.scpcs.ucla.edu; www.usc.edu/aerosol

1) Introduction

a) Objectives of Project

The overall objective of the SCS was to conduct research and monitoring that contributes to a better understanding of the measurement, sources, size distribution, chemical composition, physical state, spatial and temporal variability, and health effects of suspended PM in the LAB.

The research objectives of the SCS were:

1. To characterize PM, its constituents and precursors, to better understand sources and atmospheric transport affecting human exposure and to support development of State Implementation Plans (SIPs).
2. To support health effects and exposure research by providing enhanced atmospheric measurements at ambient sites to obtain an improved understanding of the physical and chemical characteristics of aerosols. This information will help to reduce uncertainty in defining and implementing the NAAQS and improve the health-exposure research community's ability to address research questions and scientific uncertainties about PM source-receptor-exposure-effects relationships.
3. To conduct methods testing by way of inter-comparisons and evaluations of different technologies for characterizing PM, including the evaluation of new instrumentation and sampling methods against federal reference methods.

The SCS projects followed the above three major objectives listed by U.S. EPA in the Supersite RFP, i.e. *PM Characterization, Support of Health Effects and Exposure Research, and Methods Testing*. Specific projects and hypotheses under each of these objectives are listed in Table 1.

Intensive aerosol measurements that went beyond the traditional PM_{2.5} mass, sulfate and nitrate concentrations were conducted in several areas of the LAB. These areas were chosen to provide wide geographical coverage, and thus to be as representative as possible of human exposures to these pollutants. A mobile PIU, was deployed to these locations to conduct PM measurements. Sampling in each site lasted for 6-12 months. Intensive measurements were also conducted up-and downwind of several freeways of the LAB, concurrently with and in support of in vivo and in vitro health studies. Physical, chemical and biological PM characteristics were thus determined as a function of distance from freeways impacted by mostly diesel and-or gasoline traffic over different seasons. A number of existing PM sites in the LAB, operated by SCAQMD and CARB, were used as satellite sites, in addition to the central site, to obtain spatial PM variability in the LAB as a function of size and composition. Most of these satellite sites were also sampling locations of the USC CHS.

The SCS activities were integrated with multidisciplinary research in exposure assessment, toxicology, and epidemiology of the EPA STAR program funded SCPCS. The SCS research activities were conducted in a manner that maximizes the use data collected by State and Local Agencies on PM through continuous interactions with the CARB and the SCAQMD. The monitoring activities of our Supersite were also linked with toxicology studies in the LAB using a mobile PM Concentrator facility to investigate health effects associated with exposures to ultrafine, fine and coarse particles. These studies were funded by the SCPCS, the Health Effects Institute (HEI), the CARB and the National Institute of Environmental Health Sciences (NIEHS). In conclusion, the SCS has become an invaluable resource to the abundant ongoing and planned PM monitoring, health and modeling studies in Southern California.

b) Table of Research Projects/Hypotheses:

A	<i>PM Characterization</i>
1	Comprehensive characterization of PM in the LAB and correlations between particle size distribution, chemical composition and gaseous co-pollutants;
2	Determination of the occurrence, frequency and prevalence of PM_{2.5} sub-modes in different locations of the LAB
3	Systematic evaluation of sampling artifacts of the Federal Reference Method (FRM) in measuring PM_{2.5}, PM₁₀ and Coarse PM concentrations
4	Study of PM formation and growth mechanisms in different locations of the LAB
5	Testing of the hypothesis of that 2.5 μm represents a clear cutpoint between coarse and fine PM and does not depend on location or season
6	Determination of the seasonal and spatial variation of ultrafine, accumulation and coarse PM in the LAB and their relation to sources
B	<i>Support of Health Effects and Exposure Research</i>
1	Detailed physico-chemical characterization of concentrated PM used in ongoing toxicity studies currently under way in the LAB
2	Measurement of within-community PM variability for improved dispersion models describing personal exposure indices based on traffic-based emissions for use in ongoing epidemiological investigations of chronic respiratory health effects of ambient particle matter in children
3	Measurement of the size distribution as well as the spatial and seasonal variation of particle bound PAH, oxy-PAH, nitro-PAH, quinones and other polar PAHs in the LAB
4	Determination of the contribution of volatile and semi-volatile species to Total Suspended PM_{2.5} mass and assess any resulting bias in interpreting epidemiological results
5	Physico-chemical measurements of PM properties as a function of distance from freeways
C	<i>Methods Testing</i>
1	Comparison between the actual 24-hour averaged PM₁₀ and PM_{2.5} concentrations with those determined using continuous PM mass monitors
2	Comparison between the real-time size distribution and mass concentration determined with the SMPS and APS with the 24-hour averaged mass-based size distribution measured with the Microorifice Uniform Deposit Impactor
3	Development of a semi-continuous monitor for size-dependent nitrate and carbon measurement
4	Evaluation and comparison of new and emerging measurement methods for physico-chemical measurement of PM properties

2) Scientific Key Findings

Work produced by the investigators of the Southern California Supersite has been published in about 60 refereed journal publications, which are listed in a separate section of this report. Below we have a summary of our key scientific findings of the past 5 years.

PM Characterization

- a) PM size and chemical composition in the LAB depends on locations and season. UFP concentrations (particle diameter $dp < 0.1 \mu\text{m}$) were found to be the highest at the source sites resulting from fresh vehicular emissions. Mass concentrations in the accumulation mode ($0.1 < dp < 2.5 \mu\text{m}$) were lower in winter than in summer, especially at the receptor sites. PM concentrations in the coarse mode ($2.5 < dp < 10 \mu\text{m}$) were lower in winter and were composed mostly of nitrate and crustal elements (iron, calcium, potassium, silicon, and aluminum). Consistent relative levels of these elements indicate a common source of soil and/or road dust. In the accumulation mode, nitrate and organic carbon were predominant with higher nitrate levels found at the receptor sites. The ultrafine mode PM consisted of mostly organic carbon, with higher wintertime levels at the source sites due to increased organic vapor condensation from vehicles at lower temperatures. Conversely, higher ultrafine organic carbon levels at the receptor areas are due to secondary organic aerosol formation by photochemical reactions as well as increased advection of polluted air masses from upwind. (Sardar et al., 2005)
- b) In source sites, the diurnal trends of total particle number concentration and elemental carbon (EC) appear to be almost identical throughout the day and irrespective of season, thereby corroborating the role of primary emissions in the formation of these particles. This agreement between EC and particle number was not observed in receptor sites during the warmer months of the year, while very similar trends to sources sites were observed during the winter months in that area. (Kim et al., 2002)
- c) Similarly, the size distribution of UFP in source sites was generally unimodal with a mode diameter of 30–40 nm and without significant monthly variations. The number-based particle size distributions obtained in receptor sites were bimodal, with a significant increase in accumulation mode as the season progressed from winter to summer. Afternoon periods in the warmer months are characterized by high number counts while mass and EC remain low, suggesting the formation of new particles by photochemistry. Particle mode diameters range from 30 nm up to above 100 nm, a result not seen in most other studies of particle size distributions in other urban or rural areas where mode diameters are generally less than 50 nm. Evidence is presented that the observed UFP concentrations and size distributions are influenced by long range advection and photochemical processes as well as vehicular emissions, which have been previously assumed to dominate day to day UFP levels. (Fine et al, 2004)
- d) Although previous research does suggest a relationship between coarse mode and intermodal (1-2.5 μm) PM, the sites in which these studies were conducted are not representative of all locations. Similar data for coarse, intermodal, and fine PM were collected across four sites in the LAB. While some similarities exist between these results and those of comparable studies, our Supersite studies showed that intermodal PM consists of a significant portion of particles that

are similar in chemical composition to PM₁ particles that are thought to cause the greatest health effects. This study was performed to shed light on the origin and chemical composition of intermodal particles between the coarse and fine PM modes in Los Angeles, a unique city where crustal, oceanic, anthropogenic primary, and secondary sources are responsible for the high observed PM levels. Our results indicate that a PM₁ standard would not constitute an unambiguous separation of coarse and fine mode PM in this urban air shed. (Geller et al., 2004)

e) We have examined the effects of a denuder placed upstream of a MOUDI impactor on the size distributions of 15 priority pollutant PAHs, including both semi-volatile and particle-bound species, collected near Central LA and at a downwind receptor location. Consistent with literature data, the percentage of PAHs found in the particle phase increases with decreasing vapor pressure. The use of a denuder upstream of a MOUDI does not significantly affect the size distribution shape and form of the less volatile PAHs. The fraction of PAH mass in the coarse mode increased with increasing PAH vapor pressure. The strongest effect of the denuder on the size distribution was seen for the more volatile PAHs collected at the downwind site. PAH size distributions measured with either configuration peaked in the 0–0.18 μm size range in samples collected in central Los Angeles and in the 0.18–2.5 μm size range at the downwind receptor site, consistent with long range transport and advection. Moreover, higher concentrations were found in the source site. (Eiguren-Fernandez et al., 2003)

f) The effects of atmospheric transport on the size distribution of PAHs, EC, OC, SO₄-2, and NO₃- are also reported, in receptor sites of the LAB. From October to February, the size distributions of PAHs are similar, but different from March to July, with increasing temperature and atmospheric transport. A significant fraction of the PAH and the NO₃- mass moved towards the coarse mode, as compared with the previous period. The correlation of temperature with the concentration of all PAHs in the less volatile or particle-phase group (BAA-IND) was negative, consistent with increased partitioning from the vapor-phase with decreasing temperature. During all seasons, the form and shape of the EC size distributions did not vary much and are distinguished by prominent mass in the ultrafine and accumulation modes. For the individual modes of the major species measured, the highest correlations were found in the ultrafine mode for sulfate and EC, suggesting increased atmospheric transport of vehicular emissions from the urban downtown Los Angeles region. (Miguel et al. 2004)

g) Ambient measurements of PAHs and nitro-PAHs were carried out in source and receptor sites. Atmospheric concentrations of PAHs and nitro-PAHs are of interest because both of these compound classes include potent mutagens and carcinogens. To augment our current understanding of atmospheric formation of nitro-PAHs, four sampling periods were employed to study the diurnal variations of these compounds. The PAH concentrations were highest in Los Angeles during winter, as a result of traffic input at this source site under tight wintertime atmospheric inversions. In contrast, nitro-PAH levels were highest in Riverside during summer, as a result of enhanced summertime photochemistry. Hydroxyl radical-initiated reactions produced nitro-PAHs in both seasons, while in winter little evidence for nitrate radical chemistry was seen. For the summer samples, nitrate radical-initiated formation of nitro-PAHs is suggested by nitro-PAH isomer profiles not only at the downwind location as anticipated, but also at the source site. In southern California, the contribution of atmospheric formation through gas-phase radical-initiated PAH reactions to the ambient burden of nitro-PAHs is dominant, with the semi-

volatile nitro-PAHs being the most abundant and 2-nitrofluoranthene being the major particle-associated nitro-PAH (Arey et al., 2005; Reisen et al., 2003)

h) Measurement of daily size-fractionated ambient PM₁₀ metals was conducted at source (Downey) and receptor (Riverside) sites within the LAB. The main source of crustal metals (e.g., Al, Si, K, Ca, Fe and Ti) can be attributed to the re-suspension of dust at both source and receptor sites. All the crustals were predominantly present in super micron particles. At the source site, potentially toxic metals (e.g., Pb, Sn, Ni, Cr, V, and Ba) are predominantly partitioned (70–85%, by mass) in the submicron particles. The receptor site exhibited coarser distributions for almost all particle-bound metals. Fine PM metal concentrations in that site seem to be a combination of few local emissions and those transported from urban Los Angeles. The majority of metals associated with fine particles are in much lower concentrations in the receptor compared to the source site. Coarse PM metal concentration trends are governed by variations in the wind speeds in each location, whereas the diurnal trends in the fine PM metal concentrations are found to be a function both of the prevailing meteorological conditions and their upwind sources. (Singh et al., 2002)

i) The very small mass of UFP has posed a great challenge in determining their size-dependent chemical composition using conventional aerosol sampling technologies. Implementing two technologies in series (the USC Ultrafine Concentrator and the MSP NanoMOUDI) has made it possible to overcome these two problems. UFP were measured in source and receptor sites during three consecutive 3-hour time intervals (i.e., morning, midday and afternoon). The results indicate a distinct mode in the 32-56 nm size range that is most pronounced in the morning and decreases throughout the day. While the mass concentrations at the source site decrease with time, the levels measured at Riverside, CA (a “receptor” site) are highest in the afternoon with a minimum at midday. In that site, ultrafine EC and OC concentrations were highly correlated only during the morning period, whereas these correlations collapsed later in the day. These results indicate that in this area, UFP is generated by primary emissions during the morning hours, whereas secondary aerosol formation processes become more important as they day progresses. (Geller et al, 2002)

j) In a separate study we explored the size-fractionated ultrafine (10-180 nm) chemical composition at urban source sites (USC and Long Beach) and inland receptor sites (Riverside and Upland) in the LAB over three different seasons. Size-fractionated UFP were collected by a NanoMOUDI over a period of 2 weeks at each site. The chemical composition of UFP ranged from 32 to 69% for organic carbon (OC), 1-34% for elemental carbon (EC), 0-24% for sulfate, and 0-4% for nitrate. A distinct OC mode was observed between 18 and 56 nm in the summer, likely due to photochemical secondary organic aerosol formation. The EC levels are higher in winter at the source sites due to lower inversion heights and are higher in summer at the receptor sites due to increased long-range transport from upwind source areas. Nitrate and sulfate were measurable only in the larger particle size ranges of UFP. Collocated continuous measurements of particle size distributions and gaseous pollutants helped to differentiate UFP sources at each site. We showed that in addition to primary emissions, photochemical reactions in the atmosphere can form new particles via nucleation. (Sardar et al., 2004)

k) Individual organic compounds can be used as tracers for primary sources of ambient PM in chemical mass balance receptor models. By examining the seasonal, temporal, geographical, size-fractionation, and inter-correlations of individual organic compounds, the sources and atmospheric fate of these tracers can be better understood and their utility as molecular markers can be assessed. We have used a high-flow rate, low pressure-drop UFP separator to collect sufficient mass for organic speciation of ultrafine and accumulation mode aerosol on a diurnal basis. Sampling was conducted at two sites (source and receptor) over two seasons (summer and winter). Hopanes, used as organic markers for vehicular emissions were found to exist primarily in the ultrafine mode. Levoglucosan, an indicator of wood combustion, was quantified in both size ranges but more was present in the accumulation mode particles. An indicator of photochemical secondary organic aerosol formation, 1,2 benzenedicarboxylic acids, was found primarily in the accumulation mode and varied with site, season and time of day as one would expect for a photochemical product. These data will be used to assess the concentration of specific PM sources to personal exposure and ultimately health effects in upcoming epidemiological and toxicological studies in LAB (Fine et al, 2004b)

l) As part of our evaluation of sampling artifacts from continuous and time integrated methods for PM measurement, we conducted a study to obtain insights into the dynamics of volatilization from the heated TEOM filter. The primary hypothesis that tested was that the rate of ammonium nitric acid and ammonia gas volatilization from particle-bound ammonium nitrate that is loaded on the TEOM's fiber filter occurs over time scales that are longer than the five-minute cycle time for the system. This is important to the use of the Differential TEOM, which utilizes the vaporization measured during alternate five-minute periods as a reference baseline value for its particle mass measurements. Our experiments showed that under ambient conditions, the mass lost from the Differential TEOM tracks well the ambient particulate nitrate concentration. The saturation ratio for nitrate vapor immediately downstream of the filter ranged from 0.05 to 0.2. By comparison, for laboratory collection of ammonium nitrate aerosol the vaporization reached a maximum value corresponding to a saturation of nitrate vapor downstream of the heated filter. This difference is due to the relatively higher particle concentrations used for the laboratory experiments. For the ambient measurements the particulate nitrate concentrations were consistently lower than the equilibrium vapor concentration at the TEOM filter temperature. In both cases the nitrate vaporization is driven by the temperature of the TEOM filter and independent of the pressure drop across the filter. (Hering et al, 2004)

Support of Health Effects and Exposure Research

- a. The concentrations of UFP decrease in a quasi-exponential manner with distance from freeways. Measurements show that both atmospheric dilution, evaporation of particle-bound labile species and coagulation play important roles in the rapid decrease of particle number concentration and the change in particle size distribution with distance away from a freeway. Total particle number concentration is directly related to traffic density and decreases significantly during a traffic slowdown. (Zhu et al., 2002a)
- b. The decrease in concentration of UFP with distance did not depend on the type of traffic impacting the freeway (i.e. diesel vs gasoline vehicles). The only major difference between

diesel vs gasoline traffic freeways was observed in the concentration of elemental carbon (EC), with the diesel traffic freeway having much higher concentrations closer to the freeway than those measured in the vicinity of the light duty traffic freeway. (Zhu et al., 2002b)

c. The decay rates of particle numbers, CO and BC with distance from freeways display a strong seasonal trend, with greater decay rates in summer than in winter for both freeways suggesting a weaker atmospheric dilution effect in winter. Particle number concentration in the size range of 6-12 nm is significantly higher in winter than in summer. The associated concentration in that size range decrease at a slower rate in winter than in summer. These results suggest that wintertime conditions favor greater particle formation, possibly due to increased condensation of organic vapors. (Zhu et al., 2004)

d. The data from our freeway measurement studies were analyzed and then simulated by a multi-component sectional aerosol dynamic model. Condensation / evaporation and dilution were demonstrated to be the major mechanisms in altering aerosol size distribution, while coagulation and deposition play minor roles. Seasonal effects were significant with winters generally less dynamic than summers. Particle compositions probably change dramatically as components adapt to decreasing gas-phase concentration due to dilution, so number distribution evolution is also an evolution of composition. As a result, people who live within about 90 m of roadways are exposed to particle sizes and compositions that others are not. (Zhang et al., 2004)

e. In collaboration with CARB, we developed a mobile platform designed to characterize UFP and associated pollutants inside vehicles, during commute in Los Angeles freeways and residential streets. Average concentrations of UFP and related pollutants varied strongly by location, road type, and truck traffic volumes, suggesting a relationship between these concentrations and truck traffic density. Freeway concentrations were frequently an order of magnitude higher than on residential streets for UFP, NO_x, BC, and CO, with higher NO_x and BC values observed in diesel traffic freeways than those with mostly light duty vehicle traffic. (Westerdahl et al., 2005)

f. We conducted a study to evaluate contributions of vehicle generated UFP to indoor environments in close proximity to freeways in the absence of known indoor aerosol sources. Particle number concentration I/O ratios showed a strong dependence on particle sizes and were influenced by different ventilation mechanisms. Highest I/O ratios (0.6–0.9) were usually observed for larger ultrafine particles (70–100 nm), while the lowest I/O ratios (0.1–0.4) occurred typically around 10–20 nm. The size distributions of indoor aerosols showed less variability than those of outdoor freeway aerosols. The penetration factors and deposition rates also varied significantly depending on particle size and agree with literature data and theories for particles greater than 20 nm. For particles less than 20 nm, I/O ratios, penetration factors, and deposition rates did not follow the expected trend based on theoretical prediction, as a result of the unique, semi-volatile, nature of freeway ultrafine particles. Sub-50 nm particles from mobile sources are semivolatile, thus shrink to a smaller size (or evaporate completely) as they infiltrate indoors. (Zhu et al., 2005)

g. These results were further supported by a study in which we examined volatility of penetrating ultrafine outdoor particles, predominantly from freeway emissions, into indoor

environments where other particle sources were minimized and no cooking activities took place, using a tandem differential mobility analyzer (TDMA) system. Our findings suggest that outdoor particles are more volatile than indoor aerosols. Increasing temperature from ambient to 130 °C decreased and broadened indoor and outdoor aerosol mode diameters, however greater mode decreases were observed for outdoor particles. Furthermore, outdoor particles lost more of their volume upon heating than indoor aerosols. No significant particle losses due to volatilization were observed at 60 °C for either indoor or outdoor aerosols. Heated outdoor particles with diameters greater than 45 nm showed bi-modal distributions, indicating that some of the aerosol is composed of primarily non-volatile particles, whereas the remaining particles are composed of mainly volatile material and consequently shrink. Evaluation of outdoor particle volatility as a function of distance to the freeway revealed that aerosol volatility decreases with increasing distance from the source. (Kuhn et al., 2005)

h. Physical and chemical characteristics, including volatility of PM in the proximity of a Light-Duty Vehicle (LDV) freeway were also measured and analyzed. The ultrafine number concentrations next to the freeway were 46000 cm⁻³ on average during the sampling period. The MOUDI ultrafine mass concentration, nitrate, and EC were higher next to the freeway than at the background site, farther from the freeway. The other components analyzed in the ultrafine mode had similar concentrations next to the freeway and at the background site. Volatility ranged from about 65% volume losses of 120 nm particles heated to 110°C, to 95% of 20 nm particles. The 20 nm aerosol was only internally mixed, whereas increasing non-volatile fractions were found for 40 nm (6% next to the freeway), 80 nm (20%), and 120 nm (28%) aerosols. (Kuhn et al., 2005b)

i. Our freeway PM measurements were compared to those performed in two bores of the Caldecott Tunnel in Northern California. One bore (Bore 1) is open to both heavy- and light-duty vehicles (HDV-LDV) while heavy-duty vehicles are prohibited from entering the second bore (Bore 2). A strong association between particle number and normalized vehicle speed ($R^2 = 0.69$) was observed in bore 1 of the tunnel. Heavy-duty diesel vehicles showed higher particle number emissions than light-duty vehicles. The PM₁₀ mass emission factor for heavy-duty vehicles was 14.5 times higher than that of light-duty vehicles. The particles derived from diesel are more abundant in elemental carbon, 70.9% of PM₁₀ emissions, as compared to the light-duty vehicles with a lower EC fraction of 40.5%. Conversely, a greater percentage of OC was found in light-duty emissions than heavy-duty emissions. Compared to previous studies at the Caldecott tunnel, less particle mass but more particle numbers (by factors of 2-4 fold) are emitted by vehicles than was the case 7 years ago. As the emissions of carbonaceous PM of newer engines decreases, the formation of nucleation mode particles is favored due to the reduction of the available surface for adsorption of the semi-volatile material. The resulting supersaturation of the mostly organic vapor increases the production of nano-particles by nucleation. (Geller et al., 2005)

j. Our PM physico-chemical measurements were conducted concurrently and thus supported several in vitro studies, which were funded by the SCPCS. We have developed three assays for the quantitative determination of redox activity. In one of these studies, we have developed an assay for PM redox activity, utilizing the reduction of oxygen by dithiothreitol, which serves as an electron source. We have found that PM will catalyze the reduction of oxygen and have

examined the distribution and chemical characteristics of the redox activity of PM fractions collected in different sites in the LAB. Redox activity was highest in the ultrafine fraction, in agreement with results indicating ultrafines were the most potent toward inducing heme oxygenase expression and depleting intracellular glutathione, which has relevance to induction of oxidative stress. Comparison of the redox activity with chemical composition showed a reasonable correlation of redox activity with elemental carbon ($r^2 = 0.79$), organic carbon ($r^2 = 0.53$), and with benzo [ghi] perylene ($r^2 = 0.82$), consistent with species typically found in mobile emission sources. (Cho et al., 2005). The other redox assays will be reported elsewhere but they enable determination of the role of metals versus organic compounds. These three assays are unique.

k. Our Supersite measurements also supported another series of in vitro studies, seeking to determine whether differences in the size and composition of coarse (2.5–10 μm), fine (< 2.5 μm), and ultrafine (< 0.1 μm) PM are related to their uptake in macrophages and epithelial cells and their ability to induce oxidative stress. UFP were most potent toward inducing cellular heme oxygenase-1 (HO-1) expression and depleting intracellular glutathione. HO-1 expression, a sensitive marker for oxidative stress, is directly correlated with the high organic carbon and PAH content of UFP. The small size of UFP allow better tissue penetration; we used electron microscopy to study subcellular localization. UFP and, to a lesser extent, fine particles, localize in mitochondria, where they induce major structural damage. Furthermore, perturbations in mitochondrial functions were highly dependent on UFP chemical composition, as they could be reproduced by intact diesel particles (DEP) and ambient UFP. In contrast, commercial polystyrene nanoparticles failed to exert a mitochondrial effect. (Li et al., 2003; Xia et al., 2004)

l. In vivo studies using concentrated ambient particulates (CAPs) in the vicinity of freeways were also supported in part by the SCS (the PM characterization part) and by the SCPCS (the biological analysis). Virtually all of these studies showed that CAPs exacerbate airway inflammation and allergic airway responses in a sensitized mouse models, with the responses being far greater for UFP especially those in close proximity (within 50 m or less) of a freeway. (Kleinman et al., 2004; Campbell et al., 2004). The studies also demonstrated neurological inflammation and cardiovascular effects. Human clinical studies using the UFP concentrator showed both cardiovascular and lung function changes.

m. UFP, which dominate the particle number concentration of ambient aerosols, recently have been the focus of several health studies. It has been hypothesized traditionally that these particles originate from vehicular emissions; thus, the concentrations of gases such as CO, NO, or NO₂ that also originate from traffic sources can be used as a surrogate measures of UFP. The advantage of this approach is that concentrations of these gases are monitored routinely in compliance networks and on personal levels by means of relatively simple and easy-to use monitors. The validity of the assumption using gases as surrogates of UFP was tested in this study in five sites of the LAB over the course of one calendar year. Our studies indicated overall lack of significant associations between hourly and 24-hr PN versus gaseous co-pollutant concentrations, which can be attributed to the differences in the sources and formation mechanisms that are responsible for generating these pollutants in the environment of the LAB. These findings also imply that potential confounding effects of co-pollutants will not affect

epidemiologic analysis seeking to link UFP to health effects because of the general lack of associations between PN and co-pollutant concentrations. (Sardar et al., 2004b)

n. In Southern California, dry summers followed by hot and dry westerly wind conditions contribute to the region's autumn fire season. In late October 2003, 13 large Southern California wildfires burned more than 750,000 acres of land, destroyed over 3500 structures, and displaced approximately 100,000 people. The fire episode was declared the deadliest and most devastating in more than a decade. As part of our Supersite program, we examined the actual impact of these wildfires on air quality in urban Los Angeles using "opportunistic" data from other air pollution studies being conducted at the time of the fires. In general, the wildfires caused the greatest increases in PM₁₀ levels (a factor of 3–4) and lesser increases in CO, NO, and PN (a factor of up to 2). NO₂ levels remained essentially unchanged, and ozone concentrations dropped during the fire episode. Particle size distributions of air sampled downwind of the fires showed number modes at diameters between 100 and 200 nm, significantly larger than that of typical urban air. The particles in this size range were shown to effectively penetrate indoors, raising questions about the effectiveness of staying indoors to avoid exposure to wildfire emissions. These data will be used in epidemiological studies to assess the degree to which these fires have impacted public health in the LAB. (Phuleria et al., 2005)

Methods Development and Testing

a. We have developed and evaluated several continuous mass monitors that measure the concentrations of ultrafine, coarse or PM_{2.5} in hourly or more frequent time intervals. The operating principle of the coarse PM monitor is based on enriching particles in the 2.5 – 10 μm range by a factor of about 25 by means of a 2.5 μm cutpoint round nozzle virtual impactor, while maintaining fine mass, i.e., mass of PM_{2.5} (FM) at ambient concentrations. The aerosol mixture is subsequently drawn through a standard TEOM, the response of which is dominated by the contributions of the coarse PM, due to concentration enrichment. Findings from the field study ascertain that a TEOM coupled with a PM₁₀ inlet followed by a 2.5 μm cutpoint round nozzle virtual impactor can be used very successfully for continuous coarse mass concentration measurements. This monitor can provide reliable measurements in time intervals as short as 5 minutes. The simplicity and reliability of this monitor makes it ideal for use in large scale monitoring networks (Misra et al., 2001; Misra et al. 2003)

b. We have evaluated the performance of a new differential TEOM monitor (Rupprecht and Patashnick Co., NY) and its ability to measure the "actual" ambient near-continuous PM_{2.5} mass in an area often high in semivolatile PM. The Differential TEOM monitor has been developed to directly measure ambient PM mass concentrations while accounting for collection artifacts, including loss of semivolatile aerosols and temperature changes. The Differential TEOM monitors used in this study were self-referencing, providing mass concentration measurements at 5 min intervals. To reference the semi continuous mass measured by the Differential TEOM monitor, its 24 h time-integrated mass concentrations were compared to those determined by collocated filter-based samplers. The results show that PM-2.5 mass measurements using the Differential TEOM monitor are very consistent with those of the time integrated samplers, while differences can be generally explained by loss of ammonium nitrate from the reference samplers.

The field results also demonstrate the ability of the Differential TEOM monitor to track adsorption and desorption processes from its sample filter, thereby confirming that the Differential TEOM monitor provides a very good estimate of the “actual” ambient particulate mass present on a near-continuous basis. (Jaques et al., 2004)

c. UFP mass concentrations vary drastically over short time scales in the atmosphere, yet no monitor currently measures UFP mass continuously. The need for monitors that can perform UFP concentration measurements in shorter time intervals is of paramount importance to environmental health, as such a monitor can lead to substantial improvements in population exposure assessment to ambient UFP. We have modified a Beta Attenuation Monitor (BAM) to measure near-ultrafine (i.e., $<0.15\ \mu\text{m}$ or PM_{0.15}) mass. The BAM is preceded by a $0.15\ \mu\text{m}$ cutpoint impactor, which is designed to have very low pressure drop. Results from the lab and field evaluation of this monitor indicate that the PM_{0.15} mass concentrations are in excellent agreement with collocated reference time-integrated samplers. The poor correlation between the PM_{2.5} and PM_{0.15} mass concentrations demonstrates the need of a continuous monitoring requirement for sub-150 nm particles to ensure effective assessment of the short-term variations in their mass concentration. (Chakrabarti et al., 2004)

d. PM_{2.5} mass concentrations were also measured on a short time interval basis using the continuous ambient mass monitor (CAMM) and real-time ambient mass sampler (RAMS) along with a differential tapered element oscillating microbalance (TEOM) in Rubidoux CA, and intercomparison were made among these samplers to evaluate their measurement performance of PM_{2.5} mass. Based on the correlation with particulate nitrate concentrations and particulate mass, the differential TEOM showed better measurement of semivolatile species in PM_{2.5} than either the CAMM or the RAMS. The RAMS typically measured more mass than the CAMM. The results, consistent with our publication on the new version of the TEOM, suggest that the differential TEOM reflected better the PM_{2.5} mass concentrations, with less impact from the loss of semivolatile components. Particulate ammonium nitrate losses estimated from the paired Harvard-EPA annular denuder system (HEADS) and a commercial federal reference method (FRM) sampler were equivalent to the PM_{2.5} mass difference between the paired differential TEOM and FRM samplers. This finding suggests that volatilization of semivolatile ammonium nitrate was substantial and thus, the differential TEOM performed better in the measurement of the actual PM_{2.5} defined by the FRM. (Lee et al., 2005)

e. We have compared the Scanning Mobility Particle Sizer and Aerodynamic Particle Sizer tandem (SMPS-APS) to other continuous PM measurement devices and to time-integrated mass samplers to determine its ability to measure accurately PM₁₀ and PM_{2.5} concentrations. At the full range of PM_{2.5} measurements, the SMPS-APS consistently compares well to gravimetrically determined reference mass concentrations of the Partisol and those of the MOUDI Impactor -- both at separate locations and across different seasons for various time-intervals. However, the association between the SMPS-APS and Partisol is weakened for PM₁₀, which is driven by the poor efficiency of the APS for measuring coarse particle mass. This study corroborates the results of previous studies in that the APS: 1) overestimates coarse PM greater than about 4 to 5.5 μm , likely due to coincidence counting or the “phantom” effect; and 2) underestimates particles larger than about 6 μm due to a continuous increase of settling losses for larger particles in the sampling train of the APS. Comparisons of the SMPS-APS

demonstrate good agreement for particles larger than 0.3 μm . The differences between SMPS and MOUDI for smaller particles may be due to the effect of shape and density of local vehicular emissions on the measurement of particle mobility by the SMPS, resulting in its underestimation of UFP mass relative to the MOUDI. (Shen et al, 2003)

f. In collaboration with Aerosol Dynamics Inc. (ADI), we developed and evaluated a new size-fractionated continuous fine particle nitrate monitor. The new monitor consists of three cascaded integrated collection and vaporization cells (ICVC) and provides ten-minute resolution particulate nitrate measurements in three particle diameter size ranges (0.10 - 0.45, 0.45 - 1.0, and 1.0 - 2.5 μm) corresponding to observed sub-modes in the particle size distribution in Southern California. Side-by side sampling was conducted for approximately six months at two sites, both at downwind receptor locations east of downtown Los Angeles. Both size-resolved and total PM_{2.5} nitrate concentrations were compared among the different sampling techniques. The ADI monitor and HEADS PM 2.5 nitrate measurements, for which nitrate sampling artifacts are expected to be low, are very well correlated ($r^2 = 0.79$) with a geometric mean ADI:HEADS ratio of 0.90. The ADI size-fractionated nitrate data measured consistently more nitrate than the corresponding MOUDI stages due to volatilization of labile ammonium nitrate from the MOUDI impaction substrates. Less disagreement was observed in the 1.0 – 2.5 μm size range in which nitrate is more likely to exist as non-labile sodium nitrate. We believe that the continuous nature of the data generated by the ADI monitor will provide valuable information on the spatial and temporal distribution of particulate nitrate in the atmosphere, and the new dimension of size-fractionation can help to determine the sources and formation mechanisms of atmospheric particulate nitrate as well. (Fine et al., 2003)

g. The need for continuous personal monitoring for exposure to PM has been demonstrated by recent health studies showing effects of PM exposure on time scales of less than a few hours. Towards that goal, we evaluated an active-flow personal DataRAM for PM_{2.5} (pDR, Model 1200; MIE Inc., Bedford, MA), designed as a wearable monitor to continuously measure particle exposure. The results demonstrated the instrument to be of sufficient accuracy and precision to be used in future personal exposure studies. The instrument precision was found to be good (2.1%) and significantly higher than the passive pDR configuration tested previously. A comparison to other proven continuous monitors resulted in good agreement at low relative humidities. Results at higher humidity followed predictable trends and provided an accurate correction scheme for pDR readings. The active flow feature of the pDR allows collection of the sampled particles on a back-up filter. The 24-hour mass measured on this filter was found to compare very well with both a Federal Reference Method for PM_{2.5} mass and the 24-hour average of the electronically generated, RH-corrected, continuous pDR data. (Chakrabarti et al., 2004b)

h. Also in collaboration with ADI, we evaluated a new water-based condensation particle counter (W-CPC), designed by ADI. Evaluation of performance was based on the response of the instruments to varying particle composition, concentrations, and size. The results indicated good correlation between the W-CPC and butanol-based CPCs, with R² values ranging from 0.74-0.99. The W-CPC performed better for laboratory particles with small mobility diameters (10-50 nm) due to the hygroscopic nature of these particles and relative lower detection limits of the W-CPC. The total particle number concentration has a pronounced impact on the

performance of W-CPC. Good agreement was found between the two instruments for particle concentrations between 0 and 40,000 particles/cm³, with W-CPC/TSI 3022A ratios between 0.8 and 1.2. Due to differences in the photometric mode calibration of these instruments, the ratio drops to 0.6-0.8 between 40,000-100,000 particles/cm³. The TSI 3022A-CPC appears to have a lag period of 2-3 seconds behind the W-CPC readings. Results of this evaluation show that the W-CPC is a reliable particle counting technology for particle concentrations encountered downstream of a DMA as well as in some ambient environments (<40,000 particles/cm³). (Biswas et al., 2005)

i. We developed and evaluated a high-volume, multiple rectangular (slit) geometry jet impactor. Operating with a preselective inlet that removes particles larger than 2.5 μm in aerodynamic size, the impactor has been designed to separate ultrafine (< 0.15 μm) from the accumulation mode range (0.15 < dp < 2.5 μm). The impactor operates at a flow rate 550 l min⁻¹ and at a very low pressure drop of 0.020 kPa. Laboratory experiments conducted with monodisperse PSL particles as well as polydisperse ammonium nitrate, sulfate and indoor aerosols corroborated the 50% cutpoint of the impactor at 0.15 μm. Field test comparisons between the high-volume multi-slit impactor and the Microorifice Uniform Deposit Impactor (MOUDI) showed that the accumulation and ultrafine mode concentrations of particulate nitrate sulfate, elemental and organic carbon are in very good agreement (within 10% or less). This impactor has been developed primarily as a separator of ultrafine from accumulation mode particles for high volume collections of UFP for chemical speciation and source apportionment. (Misra et al, 2002)

j. We have used two different particle mass spectrometers, the Aerodyne AMS and the UC Davis RSMS-3, to evaluate the performance of our Versatile Aerosol Concentration Enrichment System (VACES). The Rapid Single-particle Mass Spectrometer (RSMS-3) experiments were conducted as part of the U.S. Environmental Protection Agency (EPA) Supersite program in Pittsburgh during March 2002. RSMS-3 hit rate increases were measured and possible particle composition changes introduced by the VACES were examined in the single particle mass spectra. Both ambient and concentrated carbonaceous and ammonium nitrate composition distributions were indistinguishable with RSMS-3 suggesting that VACES introduces an insignificant artifact for those particles. (Kim et al., 2001a; Kim et al., 2001b; Zhao et al., 2005)

k. The effect of concentrating semi-volatile aerosols using the VACES and the Aerodyne Aerosol Mass Spectrometer (AMS) during measurements of ambient aerosol in Pittsburgh, PA was also investigated. It was found that the shape of the sulfate mass-weighted size distribution was approximately preserved during passage through the concentrator for all the experiments performed, with a mass enhancement factor of about 10 to 20 depending on the experiment. The size distributions of organics, ammonium and nitrate were preserved on a relatively clean day (sulfate concentration around 7 ug/m³), while during more polluted conditions the concentration of these compounds, especially nitrate, was increased at small sizes after passage through the concentrator. The amount of the extra material, however, is rather small in these experiments: between 2.4% and 7.5% of the final concentrated PM mass is due to “artifact” condensation. (Khlystov et al., 2005)

3) Knowledge Gaps

Our Supersite program identified the following knowledge gaps, which should be addressed in future research endeavors:

- To determine the relative toxicity of PM from different PM sources and how variations in exposure to PM characteristics according to source, season, and location influence the eventual human health response.
- To determine the physical and chemical properties of atmospherically processed PM from real-world sources, including secondary formation, to evaluate how exposure to PM and the toxicity of PM vary with respect to location, season, and particle size, and assess their relative toxicity.
- To develop quantitative assays to link PM characterization from varying sources and temporal characteristics with health outcomes.
- To assess the contributions of these outdoor sources to indoor exposure and toxicity.
- To determine the physical, chemical and toxicological characteristics of the volatile and non-volatile particle components that originate from mobile sources.
- To determine the effectiveness of recent advances in PM emission reduction technologies (i.e., oxidizing catalytic filter traps, etc) to also reduce the emission rate of nucleation mode nanoparticles.

4) Technical and Economic Feasibility

Of the different technologies either developed and/or evaluated by our Supersite, those measuring continuously mass, whether PM_{2.5} (Differential TEOM, BAM), or ultrafine (modified BAM) or coarse (modified TEOM) mass concentrations are robust, straight forward to use after some rudimentary training and economically feasible in terms of meeting the requirements of large scale monitoring networks. Some of the technologies developed (i.e., continuous, size fractionated nitrate and carbon monitors), while efficient and robust from a performance standpoint, they are still mostly research tools and require refinement if they are to be used in large scale networks by local and state agency staff. The active continuous personal monitors (active PDR) may be useful in epidemiological or panel studies in providing trends of PM_{2.5} levels, but like any other nephelometer, their response is rather qualitative and may or may not represent the actual PM_{2.5} level, depending mostly on the relative humidity of the sampled air as well as the size distribution of the aerosol. The former can be relatively easily addressed by either experimentally determined corrections or by “conditioning” the incoming air through either gentle heating or diffusion drying. The latter may be a more complicated issue to resolve, particularly if personal exposures to UFP, such as that originating from mobiles, are to be assessed. The water-based condensation particle counter (W-CPC) evaluated by our Supersite is also a reliable instrument and fairly easy to use. A problem may exist in measuring particles above 30,000-40,000 p/cm³, where it did not agree with traditional butanol-bases CPCs presumably due to differences in the photometric calibrations. This instrument, however, still offers the great advantage of using water as opposed to an organic solvent for particle growth prior to detection, a highly desirable feature, not only when considering indoor sampling, but also when placing these instruments in enclosed environments, typical of a monitoring trailer.

One way of circumventing the higher number concentration issue may be to dilute by 2-3 fold the concentrations of the sampled aerosol, for example by mixing it with particle free air at the inlet of the instrument. The rest of the instruments developed and evaluated are mostly for chemical speciation of single particles. While the information provided by these instruments is of great value to atmospheric and potentially health researchers, they will need to be reduced in size, in power consumption and overall complexity of operation in order to become available for broader use by the monitoring community.

One of the major objectives of the Supersites Program was to better understand PM characteristics on urban and regional scales, including links between sources and receptors. By addressing this objective, results from the program should improve our approaches to developing efficient and effective emissions management strategies for reducing ambient levels of PM that may adversely impact human health and welfare. Towards this goal, our Supersite research derived the following policy-relevant findings:

- The aerosol composition is different in different particle size modes (ultrafine, accumulation, and coarse) and the composition within these modes varies considerably spatially and temporally, including between source and receptor sites within the same air basin and on time scales ranging from minutes to days as well as by season. These differences reflect the impact of different sources, atmospheric chemical processing, transport, and meteorological conditions both on local and regional scales. These results suggest that different control strategies may be needed in the future depending on location and time of the year.
- Forest fires can lead to high PM_{2.5} concentrations over large areas. These natural events cannot be controlled and their impact needs to be considered for areas near or out of attainment of the NAAQS for PM. The particles in this size range were shown to effectively penetrate indoors, raising questions about the effectiveness of staying indoors to avoid exposure to wildfire emissions.
- PM and co-pollutants such as CO and NO_x increase dramatically with distance to a freeway or a busy roadway. The measured concentrations of these pollutants may be an order of magnitude higher (depending on season and other meteorological factors) compared to those in an urban background. This finding has important implications in terms of personal exposure during commute, especially in traffic-congested areas like the LAB, to potentially toxic compounds.
- The majority of these particles are not emitted directly, but are the result of condensation of vapors derived from fuel and lube oils as they expand and cool in the ambient atmosphere. This implies that conventional PM control technologies, such as filters, may not by themselves reduce effectively particle number emissions from mobile sources, and effective reduction may only be accomplished by controlling the emissions of the condensing vapors in the vehicular exhaust. This is not an easy problem to address. In fact, evidence from our measurements in roadway tunnels suggest that the reduction of the solid, soot particles from engines may result in an increase in the number-based emissions of these smaller, semivolatile particles.
- In addition to primary emissions, secondary reactions, especially of potentially toxic organic species, are an important particle formation mechanism in LA. This includes

even the UFP mode, a substantial fraction of which may originate from these secondary reactions in the summer months. This observation is also consistent with the findings of other Supersites and constitutes an unexpected finding, given that most of the aerosol formation theories predict that organic vapours that are products of secondary reactions would condense on larger particles rather than form new smaller ones.

5) **Comprehensive Bibliography**

a) **Journal Papers (peer-reviewed)**

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b) Book Chapters

c) Patents

1. Sioutas, C. and Solomon, P. Continuous Coarse Particle Monitor Patent issued to the University of Southern California and the US Environmental Protection. U.S. Patent Number: 6,829,919, 2005.
2. Sioutas, C. Personal Particle Monitor. Patent issued to the University of Southern California . U.S. Patent Number: 6,786,105, 2005.

d) Presentations

Dr. Sioutas and his laboratory associates have been a major influence in aerosol science conferences and workshops around the world. The list is too vast to enumerate here, and contains over 100 conference presentations.