APPENDIX VII

MATES IV

FINAL REPORT

Ultrafine Particle Measurements at Fixed Sites

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VII-1 Background

There is continuing concern about the potential health effects caused by exposure to criteria pollutants and air toxics emitted from both gasoline and diesel vehicles (HEI, 2010), especially for people living in urban areas. Motor-vehicle emissions consist of a complex mixture of solid, liquid and gaseous hydrocarbons, metals, black carbon (BC), volatile organic compounds (VOC), sulfates and nitrates that range in size from a few nanometers to several microns in aerodynamic diameter. Over the past decade, regulators in the United States and California have taken major steps to reduce the adverse human health impacts from vehicular emissions. In 1998, the California Air Resources Board (CARB) classified diesel exhaust PM as a toxic air contaminant, citing its potential to cause cancer and other health problems. The U.S. EPA concluded that long-term exposure to diesel engine exhaust is likely to pose a lung cancer hazard to humans and can also contribute to other acute and chronic health effects. The International Agency for Research on Cancer (IARC), part of the World Health Organization (WHO), recently classified diesel exhaust as a human carcinogen (Benbrahim-Tallaa et al., 2012). The MATES studies conducted by the South Coast Air Quality Management District (SCAQMD) are designed to identify and quantify health risks associated with major known toxic air contaminants within the South Coast Air Basin (SCAB). In the MATES III Study, diesel particulate matter (DPM) was identified as the major contributor to carcinogenic risk due to exposure to air toxics, accounting for 84% of the total carcinogenic risk (SCAQMD MATES III Report, 2008). In the current MATES IV assessment, DPM accounts for 68% of the average carcinogenic risk in the SCAB (Executive Summary - Figure ES-2).

Federal, state and local regulatory efforts have been focused on reducing the mass of PM emitted in the ambient air. Current PM regulations are focused on two size fractions: PM_{10} (particles with a diameter less than 10 µm) and $PM_{2.5}$ (diameter < 2.5 µm). Recently, however, there is a growing concern in the public health community about the contribution of the ultrafine particles (UFPs; diameter < 0.1µm) to the overall health impacts of PM. While substantial effort has been made to characterize the health risks associated with exposure to diesel PM, information about the health impacts of UFPs is just recently emerging. These very minute particles (consisting primarily of organic material, soot, and trace elements) have a different chemical composition than the larger PM fractions ($PM_{2.5}$ and PM_{10}). Due to their small size, UFPs are not a major factor in measurements of overall PM mass, but comprise a significant majority (90%) of the number of airborne particles in the atmosphere (Stanier et al., 2004a and Zhang et al., 2004). For this reason, their concentration is usually expressed in terms of total particle count (i.e. # per cubic centimeter of sampled air, or #/cm^3), even though a small fraction of the particles being counted may be above 100 nm. UFPs are emitted from almost every fuel combustion process, including diesel, gasoline, and jet engines. Although there are many sources of UFPs in the atmosphere, vehicle exhaust is the major contributor to UFP concentrations in urban areas, particularly in proximity to major roads. Consequently, there is growing concern that people living in close proximity to highly trafficked roadways and other sources of combustion-related pollutants (e.g. airports, refineries, and railyards) may be exposed to significant levels of UFPs as well as air toxics. In a seminal study conducted in the Los Angeles Basin, the number concentration of UFPs dropped dramatically with increasing distance from busy freeways (Zhu et al., 2002a,b). UFP concentrations were typically highest on or in close proximity to freeways and decreased exponentially to upwind background levels. One type of ultrafine combustion

particles are formed in the engine or tailpipe, and are mostly sub-micrometer agglomerates of carbonaceous material ranging in size from 30 to 500 nm. These particles may also contain metallic ash (from lubricating oil additives and from engine wear), adsorbed or condensed hydrocarbons, and sulfur compounds (Morawska et al., 2008). Another type of ultrafine particle is formed as hot exhaust gases are expelled from the tailpipe. They quickly cool and either condense on existing particles or nucleate to form large numbers of very small particles in the air. They consist mainly of hydrocarbons and hydrated sulfuric acid, are generally 30 nm or less in diameter and are most commonly observed near busy freeways, especially those where a large fraction of heavy-duty diesel vehicles is present (Westerdahl et al., 2005; Ntziachristos et al., 2007; Keskinen and Ronkko, 2010). Once released into the atmosphere, UFPs undergo dilution with ambient air and are subject to chemical reactions and physical processes such as evaporation, condensation, and coagulation. Thus, particles measured away from roadways and other emission sources generally have different characteristics than those measured immediately after formation. Wind speed and direction, precipitation, relative humidity, and temperature are the main meteorological factors affecting UFP transport. In addition to primary UFP emissions, secondary formation of UFPs in the atmosphere through photochemical reactions also contributes to total number concentrations. Particle formation by secondary processes depends strongly on the intensity of solar radiation and is more distinct in summers. Once formed, secondary particles are also transformed by coagulation and condensation in the atmosphere.

VII-2 UFP Measurements During MATES IV

There are very few if any long-term studies of human population exposure to UFPs, as this species is not typically measured in monitoring networks throughout the U.S. Concentrations of UFPs vary geographically, and it is not clear how well central site monitors may capture actual local exposures. Generally there is little or no correlation between ambient particle numbers and mass (Sardar et al., 2004); therefore, measurements of ambient particle number concentrations are necessary to complement the existing PM mass measurements. UFPs have a relatively short lifespan and are strongly dependent on local sources and atmospheric conditions; thus, their number concentrations can vary significantly on short temporal and spatial scales. In order to accurately estimate human exposure and the subsequent health impacts of UFPs, particle number would need to be measured across more spatially resolved monitoring networks.

The purpose of the MATES program is to conduct a series of studies to assess cancer risk from exposure to toxic air contaminants in the SCAB. These studies are comprised of air toxics monitoring and analysis, development of toxic emissions inventories, and regional modeling and evaluations. MATES IV is intended as a follow-up study to MATES II and III; unique to MATES IV is the incorporation of continuous UFP and BC concentration measurements, even though they are not technically specified as air toxics. Details of the sites, their characteristics and sampling protocols are given in MATES IV, Chapter 2. The sampling duration for all fixed stations was one year, ranging from July 1, 2012, to June 30, 2013, excluding Huntington Park, where sampling begun in August 1, 2012, and ended in July 31, 2013. Additionally, local-scale near source monitoring of UFP and BC concentrations was performed in the vicinity of the Los Angeles International Airport (LAX), San Bernardino Railyards, and Mira Loma to assess near-source air toxic emissions. This appendix will focus on the fixed site UFP measurements in the

SCAB as part of MATES IV. Results from the local-scale UFP measurements will be reported separately.

Since there is no "standard" measurement technique or calibration standard by which different instruments can be evaluated and compared, UFP measurements are somewhat operationally defined. The MATES IV UFP continuous real-time minute data was collected at 10 fixed sites utilizing the Teledyne Advanced Pollution Instrumentation (TAPI) Ultrafine Particle Monitor Model 651. This is a continuous laminar flow condensation particle counter (CPC) that uses water to grow UFPs to a larger, detectable size. UFPs are grown through condensation in a controlled super-saturation environment to larger sizes and then measured (counted) using a photodetector. CPCs provide the total number concentration of particles above 7 nm, in real-time. Although CPCs are the most widely used instruments for measuring particle number concentrations, they do not provide any information on the original size of the particles counted. Additional technical details on this CPC model and the results of a test evaluation conducted by SCAQMD and UCLA prior to the beginning of MATES IV are reported elsewhere (Lee et al., 2013). For further information and maintenance instructions, please refer to the Teledyne Advanced Pollution Instrumentation (TAPI) Ultrafine Particle Monitor Model 651 Operation Manual.

VII-3 Data Validation

The particle number count data was downloaded from the instrument using USB drives on a weekly basis. Minute data for each site was validated and examined for anomalies. During the sampling period we experienced minor problems with the USB drives, which led to some data loss and a slightly decreased data recovery. Hourly average particle number concentrations were calculated for each station (i.e., Anaheim, Burbank, Central Los Angeles, Compton, Inland Valley San Bernardino, Huntington Park, North Long Beach, Pico Rivera, Rubidoux, and West Long Beach) from the corresponding one minute data only when the data recovery was 75% or higher (i.e., when more than 45 one minute data within the hour were valid). The hourly data recoveries for each sampling location are provided in Figure VII-1. The overall hourly data recovery for the ten MATES IV sites combined was 82%.

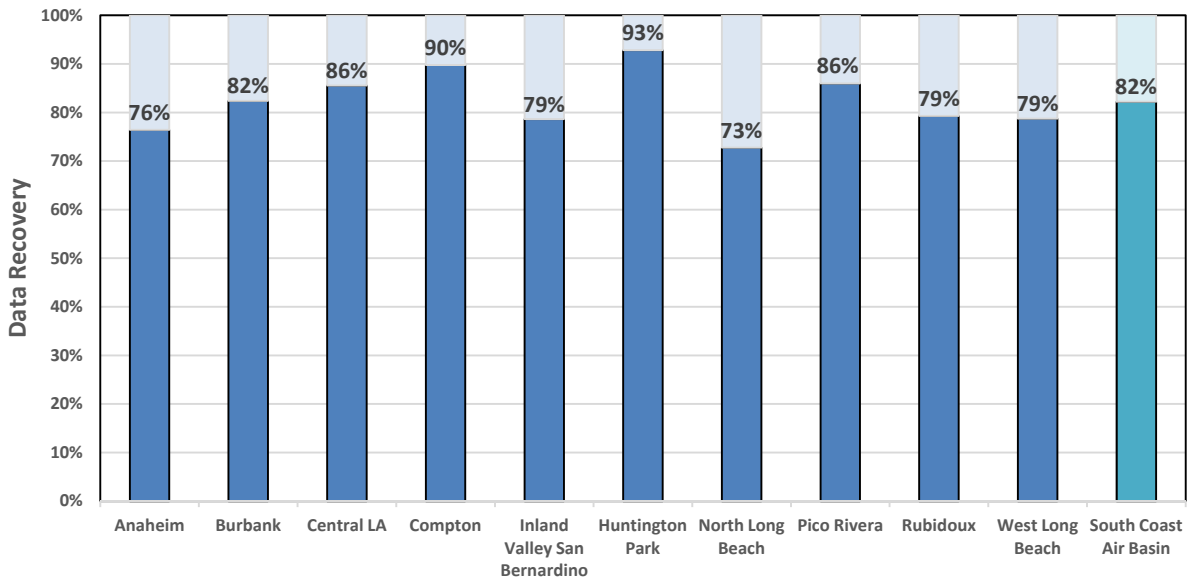

Figure VII-1: Ultrafine particle hourly data completeness for the MATES IV sites.

Three collocation studies were performed against a "Gold Standard" CPC (i.e., a reference instrument that was only used for collocation purposes) as a QA/QC check and to determine if correction factors should have been applied to the data to account for intra-model variations between CPC performances. These studies indicated that all ten site instruments were in good agreement with the "Gold Standard" CPC (i.e., high correlation coefficients; slopes close to one, and small intercepts). Thus, no corrections were applied to the field data.

VII-4 Results and Discussion

Annual Trends

The UFP annual means and standard deviation (error bars) for each site and the SCAB are shown in Figure VII-2. The UFP concentrations varied from site to site, with the highest annual averages measured at West Long Beach and Huntington Park. These sites are near potential emission sources associated with goods movement to and from the San Pedro Bay Ports and other vehicular sources. West Long Beach is located in a mixed residential and industrial area, approximately 2 km inland of the Port of Los Angeles and the Port of Long Beach, the busiest port complex in the USA. It is situated immediately downwind of a railyard and the Terminal Island Freeway 103, where heavy truck traffic consists of 22-25% of the average annual daily traffic (AADT; http://traffic-counts.dot.ca.gov/). Sampling locations with high AADT comprising of a greater percentage of heavy-duty diesel trucks (HDDT) have been shown to have elevated levels of particle number count compared to sites with less traffic and more gasoline vehicles (Zhu et al. 2004). The Huntington Park location is in a residential area, downwind of the Alameda Corridor, a freight rail connecting the downtown Los Angeles rail system to the San Pedro Bay Ports. Although Compton is also located in a residential area downwind from the railroad, it is further east than Huntington Park, potentially resulting in a decreased average annual UFP concentration. The Central L.A. site experienced some construction activity during

the sampling duration, which might have caused increased UFP concentrations. Rubidoux, an inland receptor site, had the lowest annual UFP concentration average.

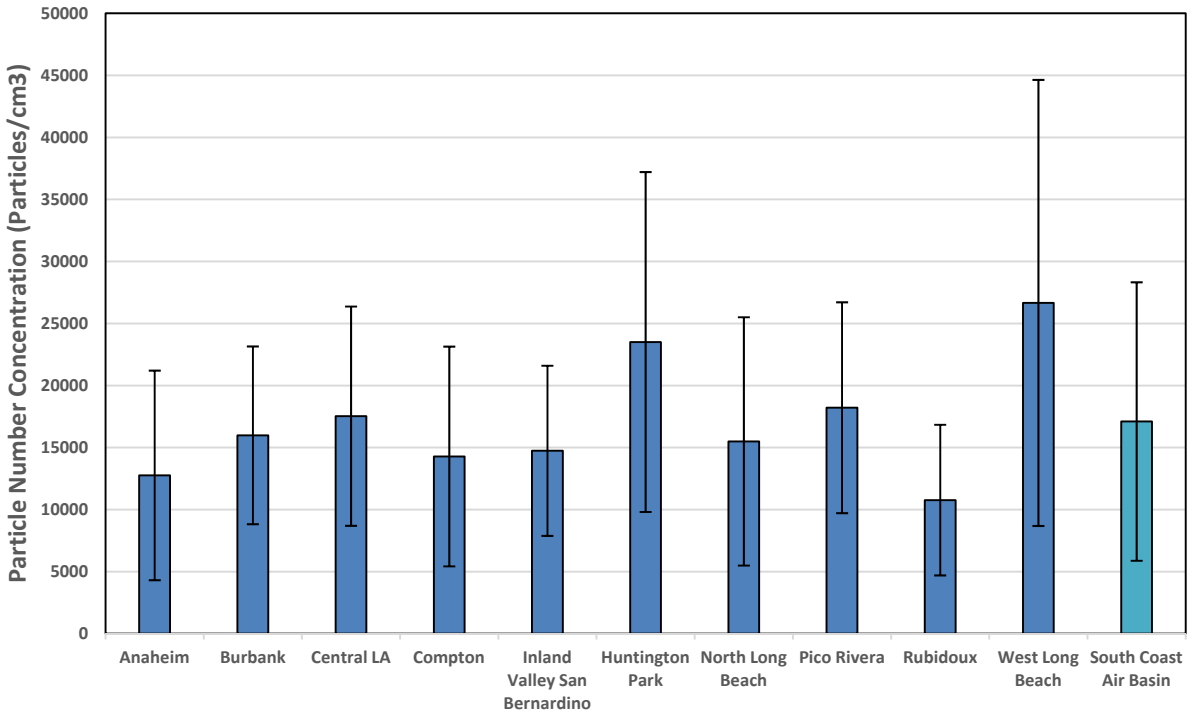

Figure VII-2: Mean and standard deviation for the MATES IV sites.

The box whisker plot in Figure VII-3 summarizes the 10th percentile, first quartile, median, mean, third quartile, and 90th percentile hourly UFP concentrations at each MATES IV site in the SCAB. The plot indicates that the Anaheim, Burbank, Central L.A., Compton, Inland Valley San Bernardino, Pico Rivera, and Rubidoux sites were characterized by a relatively low UFP variability, while the Huntington Park, North Long Beach, and West Long Beach stations had wider UFP ranges and distributions. The relatively high variability among these sites is indicative of their vicinity to one or more emission sources of UFPs (e.g., major roadways).

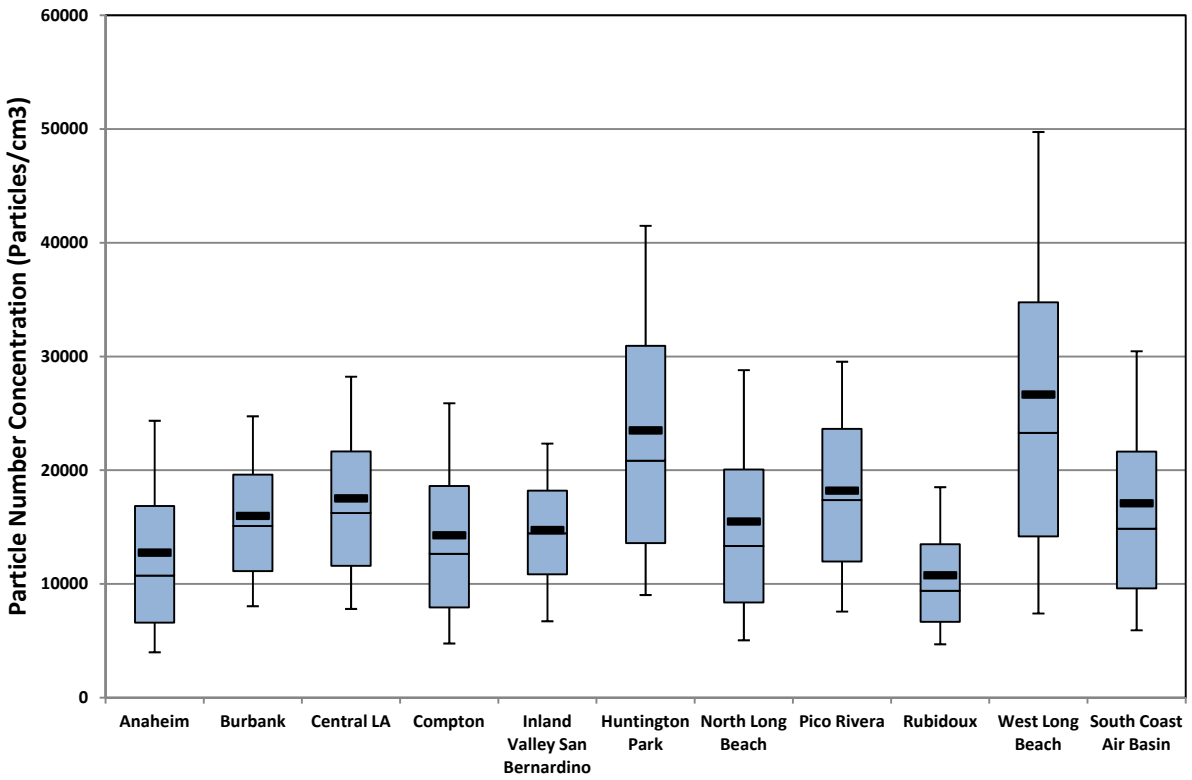

Figure VII-3: Box whisker plot showing the 90% quartile, first quartile, median, mean, third quartile and 10% quartile observed values for the MATES IV sites.

The annual mean SCAB UFP diurnal trend based on data from the 10 fixed MATES IV sites is presented in Figure VII-4. This trend is characterized by a trimodal distribution consisting of a morning peak (05:00 to 09:00), midday peak (10:00 to 16:00), and a less distinct evening peak (17:00 to 02:00). During the early morning, there is a pronounced UFP increase probably due to heavy rush hour traffic and a lower atmospheric mixing height. As the day progresses and the atmosphere is heated, the mixing height rises, leading to a dilution and subsequent decrease of traffic emissions. In the late morning and early afternoon, a second peak emerges due to the formation of secondary UFPs from photochemical processes. The UFP concentration decreases towards the late afternoon, but background levels remain elevated. A third, less pronounced peak due to the trapping of overnight emissions by the nocturnal inversion emerges towards the early evening and persists throughout the night.

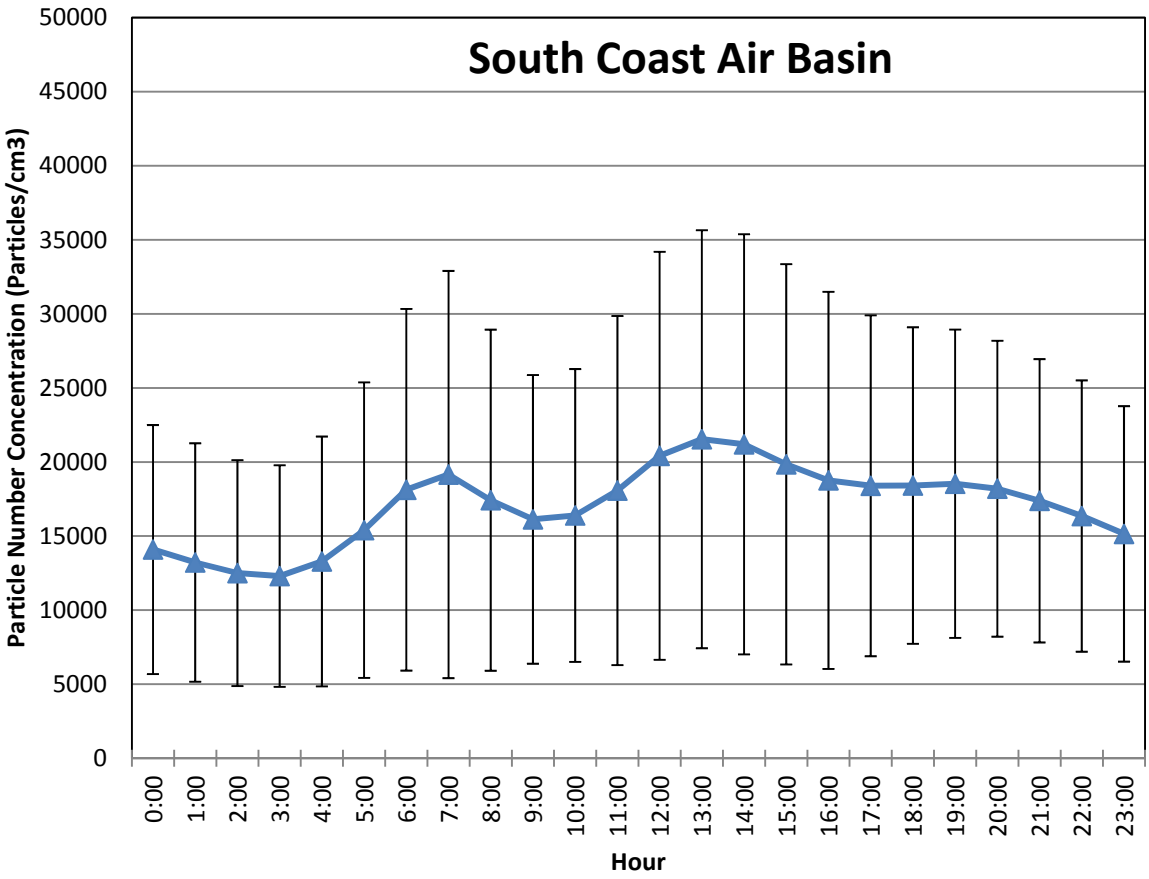

Figure VII-4: Diurnal variation of ultrafine particle concentration in the South Coast Air Basin during MATES IV.

Seasonal Trends

UFP concentrations were averaged by season to characterize seasonal variations. Seasons were divided into fall (September-November), winter (December-February), spring (March-May), and summer (June-August). Figure VII-5 displays the averaged seasonal UFP concentrations at all 10 fixed monitoring sites and for the SCAB. Huntington Park and West Long Beach showed the highest mean seasonal UFP concentrations throughout the entire duration of sampling. The maximum UFP levels observed for all seasons were in West Long Beach, except during winter, when the UFP concentration at Huntington Park was the highest. In most instances, the maximum average particle number concentrations at all sites were observed during winter months with the exception of Inland Valley San Bernardino, where the winter average was the lowest and the summer average the highest. In the wintertime, emissions from primary sources dominate the UFP concentrations due to stagnant atmospheric conditions. In addition, the coastal region experiences surface temperature inversions and weak onshore wind flow during this time of the year, leading to increased UFP levels near the coastal regions, especially near emission sources, such as freeways. During the summertime, increased UFP concentrations inland are influenced by local emission sources and long range advection of upwind sources due

to a strong onshore flow and enhanced photochemical activities. Differences in particle number concentration between the winter and summer seasons are consistent with previous studies that found higher UFP levels in the winter versus the summer months (Kittleson 1998, Kim, et al. 2000, Wang, et al. 2013). In a study conducted to compare seasonal air pollution variations near the I-710 and the I-405 freeways, lower ambient temperatures produced fewer particles in the 50-200nm size range (on the lower size limit of the accumulation mode) and a larger amount of particle number concentrations in the 6-25nm (nuclei mode) size range at both sites (Zhu et al. 2004).

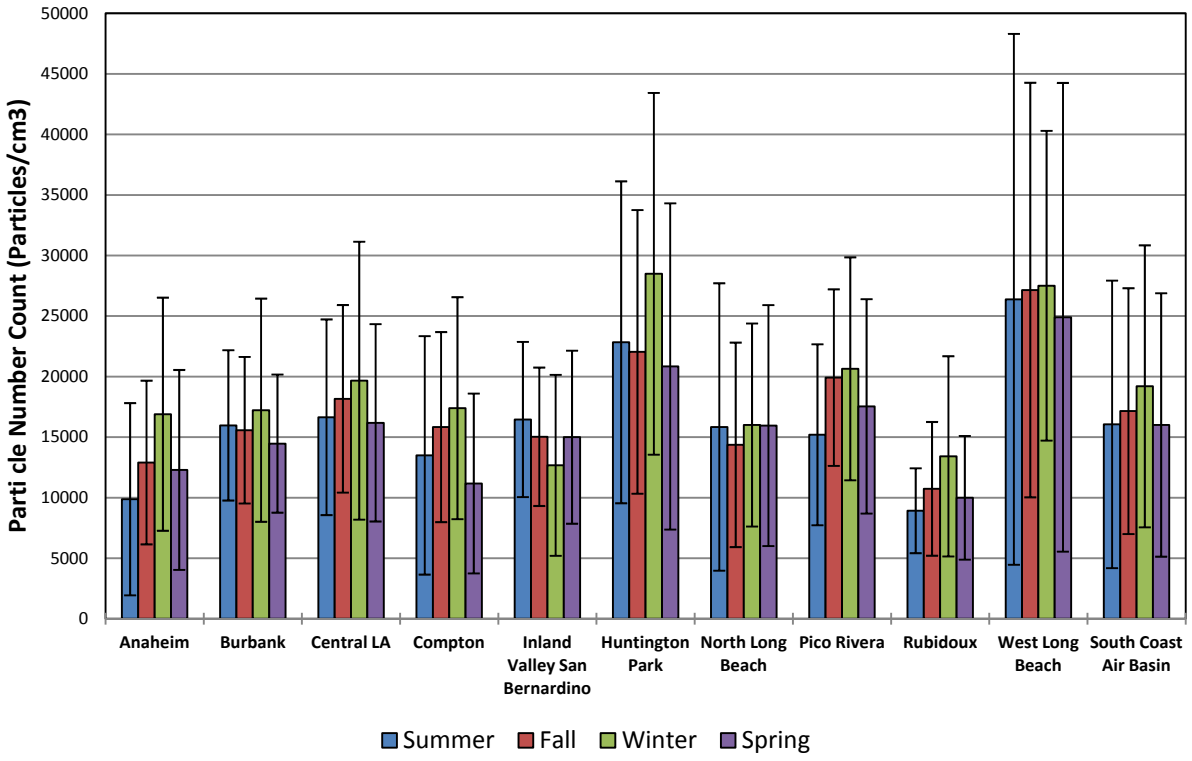

Figure VII-5: Averaged seasonal particle number concentration for the MATES IV sites.

The influence of meteorological conditions on UFP concentrations is further demonstrated in the diurnal trend differences between winter and summer profiles for the SCAB (Figure VII-6) and for each individual MATES site (Figure VII-7). The winter profile is characterized by a bimodal distribution and is distinctly different from that observed in the summer. Traffic emissions generated during the winter morning commute produces a peak during rush-hour that extends until late morning. All sites show a peak during the early morning commute hours (05:00 to 10:00) and evening commute hours (19:00 to 22:00) due to a combination of decreased atmospheric mixing height and enhanced coagulation and nucleation. As the temperature increases in the afternoon, the mixing height rises and the UFP concentrations drop, reaching a minimum in midday. When evening approaches, the nocturnal inversion layer causes an elevation in particle number count, producing a peak that persists throughout the late evening hours. Previous studies by Singh et al. 2006 and Wang, et al. 2012 have found similar winter

diurnal trends. However, these patterns are less pronounced in the spring and fall. During these seasons, especially in the spring, the morning commute peak decreases to near background levels at all stations. This observation was most likely due to warmer overnight temperatures, a higher mixing height, and a subsequent dispersion of air pollutants. Comparable spring and fall diurnal profiles were also observed in previous studies conducted in the SCAB (Sioutas et al. 2011). Throughout the summer, secondary formation of UFP through photochemical reactions generates a midday peak (10:00 to 17:00). Particles smaller than 60nm in aerodynamic diameter have been shown to contribute to this increase in particle number concentration (Singh et al. 2006). This midday photochemical peak is more pronounced in the coastal region and less distinct in the inland sites (Inland Valley San Bernardino, Rubidoux). The Inland Valley San Bernardino location did not reflect the same seasonal trends as Rubidoux. In fact, a large broad peak begins in the early morning commute hours at 04:00, reaches a maximum at 14:00, and remains elevated during the evening. This was the only site where the summer evening particle number concentrations were higher than the winter evening concentrations. The photochemical peak was also in an earlier time frame compared to the other sampling locations.

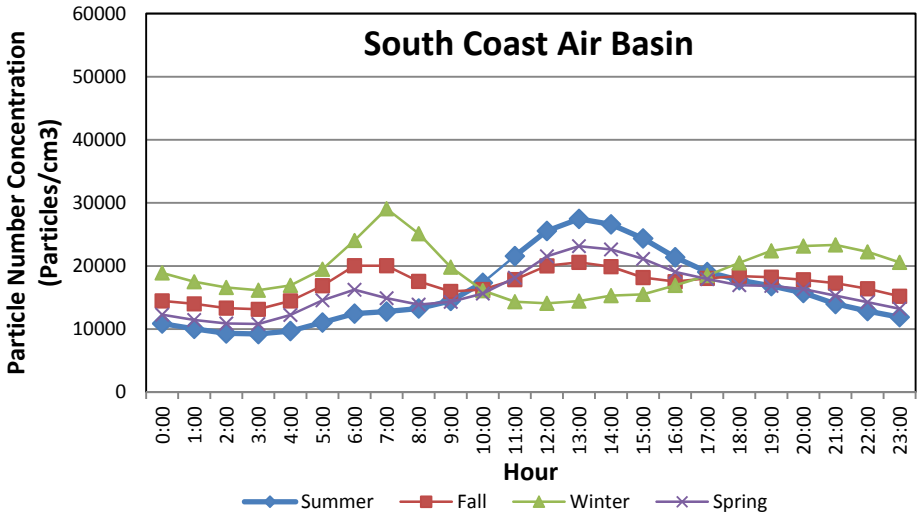

Figure VII-6: Averaged seasonal diurnal particle number concentration for SCAB.

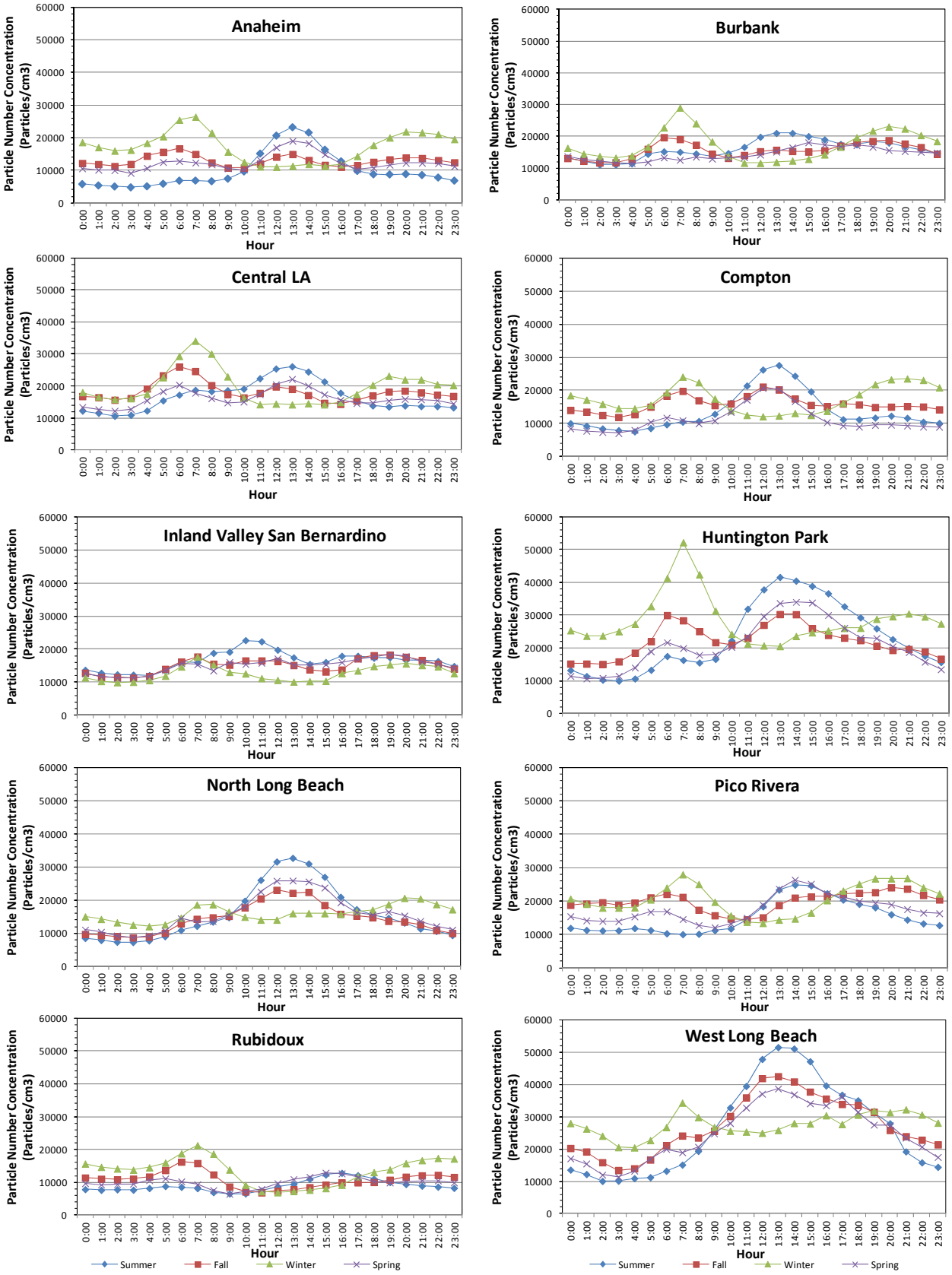

Figure VII-7: Averaged seasonal diurnal particle number concentration at each site.

Weekday/Weekend Diurnal Trends

The effect of traffic emission sources and of meteorological factors is also reflected in the day of the week diurnal UFP distribution plots. Figure VII-8 and Figure VII-9 display seasonal averages for each day of the week for the SCAB and for each individual MATES IV site, respectively. The lowest UFP averages were typically observed on Sundays during all seasons, which is consistent with previous studies (Sabaliauskas et al. 2013, Sioutas 2011, Tiwary et al. 2012). Conversely, the highest UFP levels were observed on Tuesdays and/or Fridays. UFP concentrations were generally higher on weekdays and followed a similar weekly pattern, with the exception of Monday which is associated with lower concentrations than the other weekdays. Similar differences between weekdays and weekends patterns have been observed in various studies (Morawska et al. 2002, Fine et al. 2004). West Long Beach had the highest weekday and weekend average, and the greatest difference between the weekdays and weekends (Figure VII-9). Rubidoux had the lowest weekday and weekend average, with the lowest difference between weekdays and weekends. This weekday/weekend distinction is attributed to vehicular traffic emissions generated during the weekday commute. Sioutas et al. (2011) also observed day of the week differences between sites near the ports versus near Downtown Los Angeles. There was a larger particle number reduction at sampling locations near the vicinity of the ports on weekends versus weekdays when compared to L.A. This greater reduction in UFP concentrations demonstrates that heavy-duty diesel vehicles are important contributors to ambient UFP.

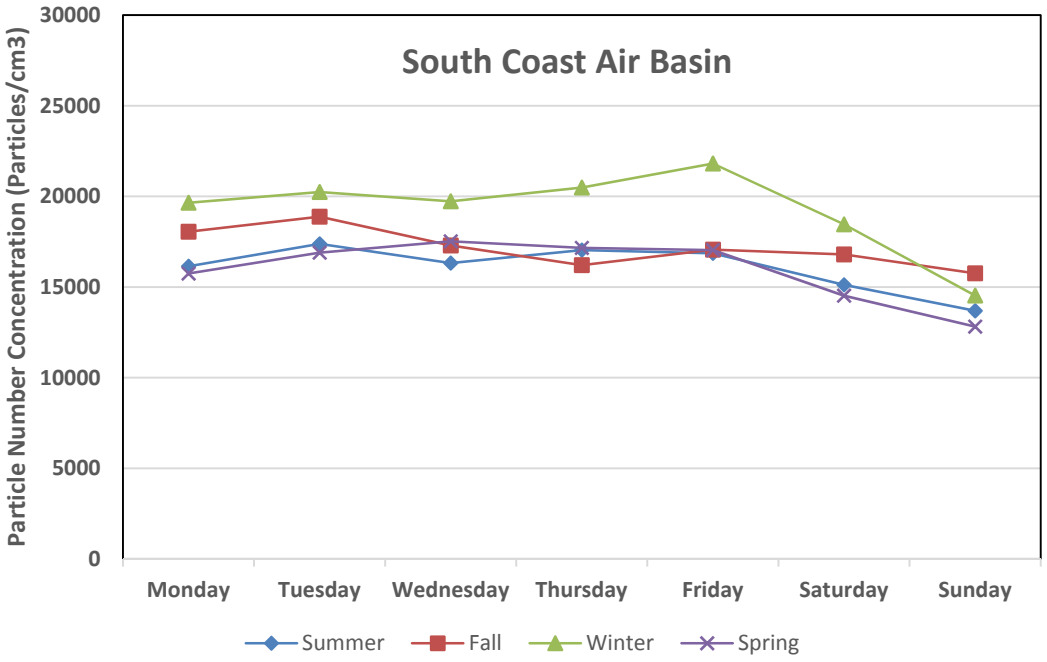

Figure VII-8: Averaged seasonal day of the week particle number concentration for the South Coast Air Basin.

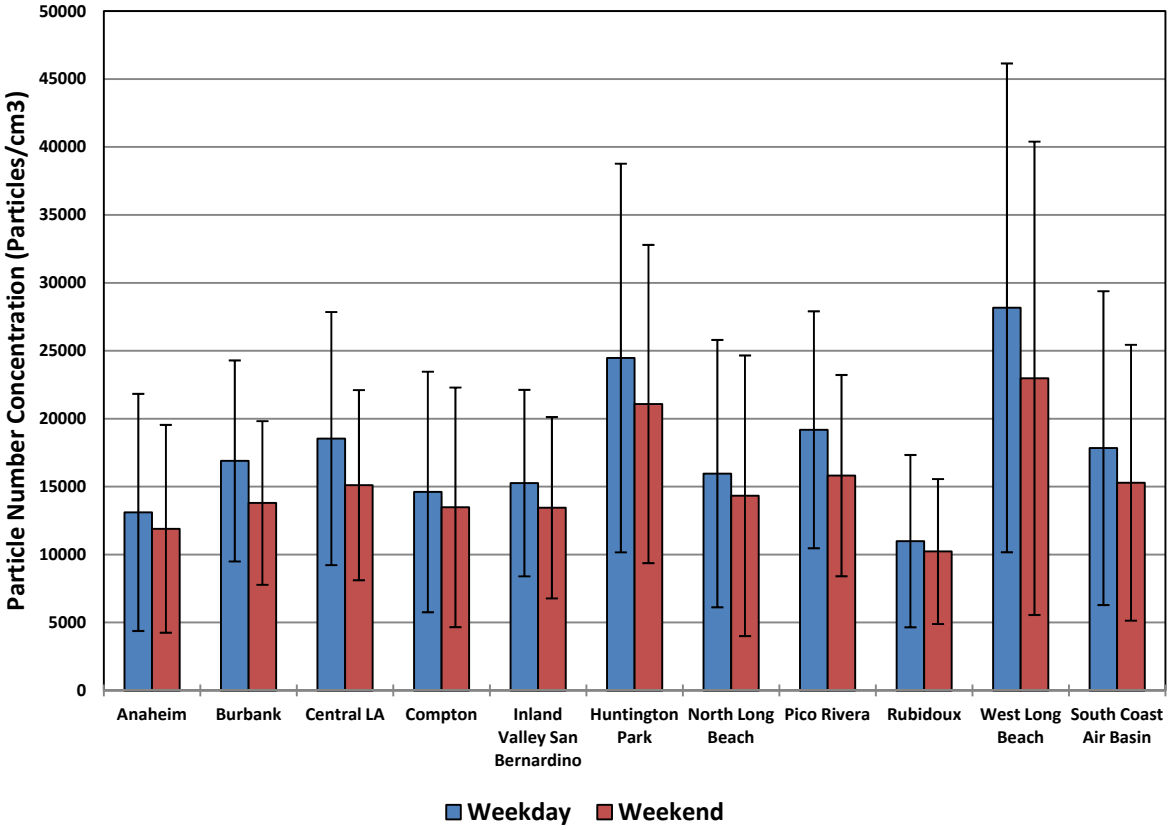

Figure VII-9: Averaged weekday and weekend particle number concentrations for each MATES IV site.

VII-5 Summary

Continuous real-time UFP measurements collected at ten SCAQMD sites during MATES IV showed high temporal and spatial variability. A variety of factors, such as the distance to the nearest emission source, the type of emission source, the traffic volume, wind speed, wind direction, relative humidity, and temperature (among other factors) could all influence the concentration, composition, and dispersion of UFPs. Atmospheric parameters could fluctuate rather rapidly throughout the day, therefore short time scales, particularly on an hourly or less basis, should be used to examine the diurnal trends of UFPs. Despite the high spatial and temporal differences measured across the SCAB, the average diurnal UFP concentrations at most MATES IV sites followed a similar trend, rising and falling throughout the day, with distinct peaks during the early morning commute, midday, and evening commute. As shown here and reported in previous studies, the ambient UFP concentration in urban environments is closely related to the temporal variation in traffic density, with highest levels observed on weekdays during rush hours (Hussein, et al., 2004; Morawska, et al., 2008; AQMD, 2012). Photochemical particle formation also contributes to increasing the afternoon number concentration of UFPs, especially in the summer.

Due to the sharp drop in UFP concentrations over short distances from the emission sources, more detailed local-scale studies are needed to develop a better understanding of the spatial UFP concentrations in the SCAB. For example, in a recent study conducted by the SCAQMD near the Santa Monica Airport (SMO; a general aviation airport), 1-min average UFP levels as high as 2,600,000 #/cm^3 were measured 35 m downwind of the runway during jet aircraft take-off (AQMD, 2011). One-minute maxima between 1,500,000 and 2,000,000 #/cm^3 (also associated with jet aircraft departures) were observed 100 m downwind of the runway in the backyard of a local residence.

Several meteorological factors contribute to the seasonal variability in the concentration of atmospheric PM and UFPs; these include:

- Lower mixing layer height and greater atmospheric stability in winter, which tend to increase particle levels by limiting vertical atmospheric mixing
- Lower winter temperature, which leads to increased nucleation of volatile combustion products, particularly during morning rush hours
- Higher photochemical activity in the summer, which favors photochemical particle formation

In the wintertime most of the factors leading to an increase in particle concentration tend to occur early in the morning (i.e. rush hour traffic, low mixing height, low wind speed and temperature). Summer minima are usually associated with increased ambient temperature (which does not favor the nucleation process), although increased photochemical activity can lead to new UFP formation, which typically occurs midday.

VII-6 References

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