Anonymous Referee 2

Anonymous Referee #2

We have appended the comments with our responses in the red font.

General comments:

The paper examines the spatial and diurnal variation of PNC in the greater Los Angeles Area. The use of CODs is a helpful approach to compare measurements between different stations. The data collected at seven locations during two measurement campaigns is presented in a scientifically but rather conventional way. However, I believe that the paper could be improves with regard to sufficient physical interpretation. The authors fail in providing the reader with a clear message of their findings. It is an interesting work because it discuss number concentrations rather than mass concentration of ultrafine particles. Number concentration is a useful parameter for epidemiological studies. Exposure assessment to UFPs is still in its initial stage compared to exposure assessment for fine particle PM2.5 and PM10. One of the major questions is how many sample points distributed in urban area are required to represent human exposure to particle number concentration. Due to their different physical properties, UFPs are supposed to have larger spatial and temporal variability than fine particles. Furthermore, there is a limited number of experimental studies for ultrafine particles in the international bibliography.

However, the contribution of this paper to exposure issues for epidemiological studies is limited. Beside the spatial variation (which is important for long term epidemiological studies) the temporal variation is very important for short-term studies. Unfortunately, the analysis presented in this paper focus on the spatial variability. The analysis of temporal variability (correlations between the monitoring sites on hourly and daily base) was not conducted.

We have added Particle Number Size Distribution analysis for each of the three sites; particle size distribution data were measured and analyzed for two seasons in the forms of inset figures for Fig 2: b,c, 3: b,c and 4: b,c along with relevant text in the discussion. We believe this addresses the issues raised by the reviewer about physical interpretation. Although we do not conduct correlation analysis, Figure 5 a, b do present hourly averages across sites, and the diurnal profiles can be interpreted in that context. We do discuss (briefly and at multiple places in the paper) how concentrations differ across sites at the same time of day/period.

The authors should give some information on the air quality status of the study area. How is the air quality status in the greater Los Angeles Area, what is the relationship between air quality and wind field (or other meteorological parameters), what are the main emission sources? The authors should provide a description on the seasonal distribution of the wind direction/intensity, the local flows, possible interaction with background wind. This information is necessary to be reported - before proceeding to the analysis of the measurements - to help the reader to understand the background air quality conditions and mechanisms responsible for the formation of these conditions. Meteorology:

The following text has been added to the Meteorology Section with references to studies that discuss the meteorology of LA and pollutant transport in greater detail.

"Meteorology of the Los Angeles Basin and its effects on air pollutant's movements have been discussed in greater detail by Blumenthal et al. (1978), Lu et al., (1994, 1995), McElroy et al., (1986, 1983), Shultz et al., (1982), Ulrickson et al., (1990), Wakimoto et al., (1986)."

Table 2 and 3 which were present in the version of the paper submitted earlier show the "the seasonal distribution of the wind direction/intensity" in terms of monthly averages across the year.

Specific comments:

Introduction:

The authors stated in the introduction (line 54) that PM2.5 mass concentrations are often used as a surrogate for UFP concentration. I don't agree with this statement and I couldn't find it in the cited reference (Wilson et al., 2006).

We have deleted this sentence.

To my knowledge Jerret et al. (2005) analyzed the associations between PM2.5 and mortality and not between UFP and health effects (line 62). Moreover, Jerret and colleagues didn't assume a homogeneous distribution of particles. In contrast, in this study PM2.5 exposure surface was developed by use of interpolation technique. It means that this reference is inadequate and the authors should exchange it. In the same sentence the authors state that UFP have pronounced spatial variation at local and regional scale with some references.

Authors quote from Jerret et al., 2005

"The assessment of air pollution exposure using only community average concentrations likely underestimates the health burden attributable to elevated concentrations in the vicinity of sources^{8,9} Health effects may be larger around sources, and these effects are diminished when using average concentrations for the entire community. Previous ACS studies have relied on between-community exposure contrasts at the scale of a metropolitan area giving all residents of a city the same exposure concentrations. Exposure to air pollution, however, may vary spatially within a city, ¹⁰⁻¹⁴..... The spatial correspondence between high exposure and potentially susceptible populations within cities may further bias estimates that rely on central monitors to proxy exposure over wide areas."

We wanted to reference Jerret et al., to convey that exposure may vary in a metropolitan area and use of central monitoring station can lead to a bias. Further, from Monn et al.:

"Recent studies in outdoor air show that ultrafine particle number counts have large spatial variations and that they are not well correlated to mass data."

We have modified the sentence is our manuscript to

Using a central monitoring station assumes a homogeneous distribution of UFP over large spatial scales, but recent works of Kim et al., 2002, Zhu et al., 2002, Tuch et al., 2006, Puustinen et al. 2007, Krudysz et al., 2009 and Moore et al, 2009 all suggest that UFP vary spatially at local as well as regional scales and use of central monitoring stations can lead to a bias in exposure assessment given the variability (Cyrys et al., 2008, Puustinen et al. 2007, Jerrett et al., 2005, Monn et al., 2001).

The references are not chosen very well: -Pinto et al. (2004) described the spatial variation of PM2.5 and not of UFP –

We agree. This reference has been removed from that context.

Zhu et al. (2002) described the decrease of UFP concentration depending on the distance to a major road (what is on a very local scale) –

It is important in context of the paper since some of our sites are in vicinities of roadways and at a spatial scale where the effects are relevant.

Krudysz et al. (2009) is still submitted (since 2008)? - Moore et al. (2009) is not in the reference list.

Krudysz is present in reference list as Line 28, Page 13921 and is published in ACP. If the presence of doi link was causing confusion, then it has been removed. Moore et al., 2009 is present in the reference list at Line 17, Pg 13922.

I miss some very relevant European papers published on this topic: Buzorius et al., (1999), Tuch et al., (2006), Puustinen et al. (2007) and Cyrys et al. (2008).

They have been incorporated in the sentence quoted above from the manuscript.

Site description:

Concerning the monitoring sites, what are their characteristics related to any particle sources, apart from their distance from traffic sources? For example, are they close to an industrial area, or another combustion source? This material will greatly help the discussion of the results. The site type (urban background, regional background, traffic influenced, industrial: : :) for each location should be added either to the text in this section or to Table 1.

Table 1b has been added to the manuscript that meets the expectations outlined by the reviewer. The site description has also been appended in the text where appropriate.

Site ID	Relevant Information on Potential PM Sources
USC	Located next to a gasoline dominated freeway in a parking lot, urban background site
DIA	Located on a hill in a parking lot, at over 100 m elevation from the neighboring freeway
UPL	Located at the foothills of mountain range, limited local sources, regional site in terms of northern extent of LAB
VBR	Located in a residential area, rural regional site
RUB	Located behind an office building near a low trafficked street and a freeway
AGO	Located in an agricultural research facility near a university, freeways nearby, most inland, regional background site

Table 1 b: Site information regarding dominant local sources

More details are required for the meteorological station: What is the distance of the meteorological station from the other measuring sites? Is it installed in an open area? Are there influences by other buildings? How representative is the station of the wind flow prevailing in the whole examined area? At which height is the wind recorded?

The meteorological station was located at the site itself with other instruments. The anemometer was placed at the height of 5 m above ground. The meteorological data was compared with standardized AQMD station data and were shown to making similar observations.

The following text is added in the manuscript.

Meteorological data, i.e., temperature, relative humidity, wind speed and direction amongst other parameters were collected using Vantage Pro 2 Weather Stations (Davis Instruments, Hayward, CA). The meteorological station was placed above the enclosure and the wind vane sampled at a height of 5 m from the ground surface. The meteorological data were compared with neighboring AQMD stations with more standardized meteorological equipments, and only for wind speed and direction slight differences were observed. This was because of lower height of our equipment. Even then, the diurnal patterns of these parameters were consistent with those reported by AQMD.

Data processing and validation: What is PSD (line 167)?

We could not find PSD, however if reviewer meant PNC or PNSD, then these two acronyms have been detailed at their first use in the manuscript.

PNC: Particle Number Concentration PNSD: Particle Number Size Distribution

It is well known that CPCs have an uncertainty of about 20%. Therefore the quality assurance procedures are very important, especially for this study. The authors report some quality assurance aspects, especially with regard to the CPCs. However, it is not clear to me why the CPCs at the beginning of the study were compared to the 'mean' CPC and at the end of the study to the concentration measured by one (even freshly calibrated) CPC. The comparison should be conducted in the same way. For each CPC the regression equation and R2 to the same "standard" (either to the 'mean' CPC or to the factory-calibrated CPC) at the beginning and the end of the study should be stated. It is not clear how the CPC data were corrected. The same is valid for the SMPS instruments.

The following text was added to manuscript.

We elected to compare CPCs with a unit calibrated by the factory instead of the mean of the CPC values because the CPCs had been operating in field continuously for over two years, and several units used in earlier studies by our group had shown performance deterioration with prolonged field use. The data were corrected (assuming a linear deterioration in performance over the span of operating period) to compensate for the inconsistency between the CPCs.

3.2 Diurnal and Seasonal Variation:

For Figures 2b&c (3b&c, 4b&c and so on) please use the same scale on the y-axis (for better directly comparison).

The new figures reflect this change.

3.2 Spatial distribution of PNC:

The discussion of Figure 5a is imprecise. The highest morning concentrations were observed for USC (45 000 1/cm3) followed by RUB and SBR (about 30 000 1/cm3 for both sites). The authors suggest similar concentrations for USC and RUB (as the two sites were located closest to freeways), but this is not true.

The statement has been modified to avoid this misinterpretation.

"The highest of morning concentrations were observed at USC and RUB, the two sites closest to freeways."

If the higher concentrations at SBR were caused only by the CPC 3025 (reporting higher numbers of particles) the curve for SBR should be corrected or removed from the figure.

SBR data has been excluded from the paper.

Inter-community variability in total particle

number concentrations in the eastern Los Angeles air basin

- 4 5
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14 Abstract

Ultrafine Particles (UFP) can display sharp gradients in their number concentrations in urban 15 environment due to their transient nature and rapid atmospheric processing. The ability of using air 16 pollution data generated at a central monitoring station to assess exposure relies on our understanding 17 of the spatial variability of a specific pollutant associated with a region. High spatial variation in the 18 19 concentrations of air pollutants has been reported at scales of 10s of km for areas affected by primary 20 emissions. Spatial variability in particle number concentrations (PNC) and size distributions needs to be investigated, as the representativeness of a monitoring station in a region is premised on the 21 assumption of homogeneity in both of these metrics. This study was conducted at six sites, one in 22 23 downtown Los Angeles and five located about 40 - 115 km downwind in the receptor areas of Los Angeles air basin. PNC and size distribution were measured using Condensation Particle Counters (CPC) 24 25 and Scanning Mobility Particle Sizer (SMPS). The seasonal and diurnal variations of PNC implied that 26 PNC might vary significantly with meteorological conditions, even though the general patterns at the 27 sites may remain generally similar across the year due to consistency of sources around them. 28 Regionally transported particulate matter (PM) from upwind urban areas of Los Angeles lowered

29 spatial variation by acting as a "homogenizing" factor during favorable meteorological conditions. 30 Spatial variability also increased during hours of the day during which the effects of local sources predominate. The spatial variability associated with PNC (quantified using Coefficients of Divergence, 31 CODs), averaged 0.3, which was generally lower than that based on specific size ranges. Results 32 showed an inverse relationship of COD with particles size, with fairly uniform values in the particle 33 range which is associated with regional transport. Our results suggest that spatial variability, even in 34 the receptor regions of Los Angeles Basin, should be assessed for both PNC and size distributions, and 35 should be interpreted in context of seasonal and diurnal influences, and suitably factored if values for 36 37 exposure are ascertained using a central monitoring station.

39 1. Introduction

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41 Numerous recent epidemiological and toxicological studies investigating associations between 42 particulate pollution and health effects have attributed greater risk to ultrafine particles (UFP, diameter less than ~ 100nm) (Oberdörster et al. 1995; Donaldson et al. 1998; Gong et al. 2007; Xia et 43 al. 2006; Delfino et al., 2005 & 2009) compared to particles of greater diameters. In vitro toxicological 44 45 studies have also shown that ultrafine particles have higher oxidative potential and can penetrate and 46 destroy mitochondria within epithelial cells (Li et al., 2003). Penttinen et al. (2001) found that daily 47 mean number concentration and peak expiratory flow (PEF) are negatively associated and that the 48 effect is most prominent for particles in the ultrafine range. A study by Peters et al. (1997) also found 49 associations between number concentrations of ultrafine PM and lowered PEF among asthmatic adults. 50

Although current federal standards for particulate matter (PM) are mass-based, there is increasing 51 52 evidence that a number-based standard might be better suited for UFP concentrations and the associated risks (Englert et al. 2004), since UFP are more numerous and contribute little to PM mass 53 (Hinds 1999). Current standards are based on PM2.5 and PM10, , although poor correlation has been 54 reported between PNC (dominated by UFP) and PM2.5 (dominated by accumulation mode particles) 55 (Sardar et al., 2004). Not only is PM2.5 not an adequate surrogate measure, but also such data is often 56 57 used from central monitoring stations to ascertain exposure values that might lead to exposure 58 misclassification due to spatial variability in UFP concentrations (Delfino et al., 2005). Urban 59 environments are often characterized by a complex set of factors (sources, meteorology, solar 60 radiation, mixing height, and topography amongst others) that can influence not only the particulate matter (especially ultrafine particles) concentration, but also its spatial variability (Costabile et al., 61 2009). Using a central monitoring station assumes a homogeneous distribution of UFP over large 62 spatial scales, but recent works of Kim et al. (2002), Zhu et al. (2002), Tuch et al. (2006), Puustinen et 63 al. (2007), Krudysz et al. (2009) and Moore et al. (2009) all suggest that UFP vary spatially at local as 64 well as regional scales and use of central monitoring stations can lead to a bias in exposure assessment 65

66 given the variability (Cyrys et al., 2008; Puustinen et al., 2007; Jerrett et al., 2005; Monn et al., 2001). In urban areas, a dominant source of UFP is primary emissions from vehicular sources (Shi et al., 2001; 67 Phuleria et al., 2005; Fine et al., 2004) and as much as 80% particles can be in the UFP size range 68 69 (Morawska et al., 1998; Shi et al., 2001; Sioutas et al., 2005). Other combustion sources, such as food 70 cooking and wood burning, can also be sources of ultrafine particles to the atmosphere (Kleeman et al., 71 1999; Schauer et al., 2001). Shi et al. (2001) have shown that nanoparticles from fresh emissions can be 72 persistent in urban atmospheres, given the multiplicity of sources. Zhu et al. (2002 a,b; 2005; 2006) have shown that UFP concentrations can decay exponentially with distance from the freeways. 73 Consequently, given their short lifetimes, the gradient of UFP concentration in atmosphere can be 74 75 strong (Sioutas et al., 2005).

76 In addition to primary, or direct, ultrafine particle emissions, photochemical reactions in the atmosphere may also be responsible for the formation of secondary ultrafine particles. Kulmala et al. 77 78 (2004) reviewed particle formation by secondary processes and showed that such particle formation 79 events are more distinct in summer. Particle formation rates depend strongly on the intensity of solar 80 radiation, but the exact mechanism by which the process occurs is not fully understood (Zhang et al., 81 2002). Once formed, particles are transformed in the atmosphere, by coagulation and condensation of 82 semivolatile vapors on their surface as they are advected downwind. This long-range transport as well 83 as photochemical particle formation in the atmosphere can lead to increased particle number 84 observations downwind of urban areas (Kim et al., 2002; Fine et al., 2004; Verma et al., 2009; Ning et 85 al., 2007).

In large urban areas like the Los Angeles air basin (LAB), both primary direct emissions and also 86 transported aged aerosols from locations upwind (some potentially distant) contribute to the observed 87 PM levels. This spatial transport of PM, coupled with local factors like the micrometeorology of a site 88 and its exposure to local sources, can produce distinct diurnal patterns, which vary spatially over scales 89 at which inter-community variability can be assessed. It has been suggested (Turner et al., 2008) that 90 91 secondary formation during regional transport can be a homogenizing factor on spatial variability. 92 However, in 2002 and 2003, investigators in the USC Children's Health Study (Sardar et al., 2004; Singh 93 et al., 2006) made measurements at several areas in LAB and found that, although some sites may

exhibit similar diurnal patterns, PNC may still vary considerably, and have only a modest correlation
among even proximate sites. Lianou et al. (2007) found that the spatial variation in PNC might far
exceed that in particulate mass concentrations. Fine et al. (2004) have also shown that sites in the
receptor areas of LAB can have different particle size distribution patterns as well as different PNC
diurnal patterns.

99 Thus, in order to better quantify the risk that ultrafine PM (UFP) poses to human health, it is necessary
100 to characterize its spatial variability to better assess the potentially different population exposure to
101 UFP, both in terms of particle numbers as well as the size distribution, compared to PM mass.

102 2. Experimental Methods

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This study is a second phase of an investigation of the intra- and inter-community variability of PNC in
the greater Los Angeles Area. The earlier phase focused exclusively on the area of the Los Angeles –
Long Beach Harbor and has been reported in Moore at al. (2009) and Krudysz at al. (2009).

107 This study was conducted at five sites in eastern Los Angeles air basin and another site in downtown 108 Los Angeles during November 2008 - December 2009. Site Information is provided in Table 1 and the 109 actual locations of these sites are shown in Figure 1. Highways and major arterials, common sources of 110 ultrafine particles, are identified in Figure 1. The distances to freeways are also tabulated in Table 1. 111 The sites in the receptor area were within 50 kilometers of each other in the E-W direction and 20 112 kilometers in the N-S direction. Sampling sites were located in areas where there were no known 113 major contributors to UFP, except for local traffic (e.g., residential neighborhoods).

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2.1 Site Descriptions

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Site 'USC' is located in downtown Los Angeles and is the Southern California Particle Center Supersite located at the University of Southern California (USC) where extensive air quality measurements have previously been reported (Sardar et al., 2005; Moore et al., 2007). It is an urban background site

influenced by traffic from the I-110 freeway located approximately 120 m to the west. This site was

chosen to represent urbanized areas of Los Angeles, which are heavily impacted by traffic. USC site is
classified as the 'source' or the 'urban background' site in this paper as it is representative of the
prevailing conditions in western part of Los Angeles basin where the urban center is located in the
basin. It is contrasted against the effect of transport and aging observed at 'receptor' or 'regional
background' sites, which are downwind in the eastern region of the basin towards which the
meteorology of the basin transports the pollutants from the source region.

The Diamond Bar site 'DIA' is located about 60 km inland from the Pacific Ocean and 40 km east of USC site. The site is located 200 m south of the CA-60 freeway. It is the first site in eastern LAB cluster along a typical trajectory over which primary aerosols emitted in the west and central parts of LA are being transported during atmospheric aging (Pandis et al., 1992). However, it is located on a hill in the prominent upwind direction and is therefore not directly impacted by traffic emissions throughout the day.

132 The Upland site 'UPL' is located in a mobile home park in Upland that is about 80 km inland from the ocean and about 60 km east of USC site. It is over 2 km away from the neighboring freeways and 133 surrounded by low trafficked streets. The site is located close the base of the San Gabriel Mountains, 134 which act as a barrier for further transport of aerosols in northern direction The site is therefore 135 136 influenced mostly by the aged aerosol advected eastwards from urban Los Angeles (Fine et al., 2004) 137 The Van Buren site, 'VBR,' is located 97 km inland and 57 km east of USC at a South Coast Air Quality Monitoring District (AQMD) sampling station in a rural residential neighborhood. It is 3 km (south) from 138 the nearest freeway and the major roadways next to the site have low traffic load. A substantial 139 component of PM at this site can be attributed to the PM advected from the west to this area after 140 hours of aging. This site has also been discussed in (Pakbin et al., 2010; Moore at al., 2010) 141 The Rubidoux site, 'RUB,' is located about 100 km inland from the ocean and 80 km east of USC (and 8 142 143 km east of VBR) at an AQMD sampling station. It is in vicinity of CA-60, situated about 200 m to its 144 south. This site is impacted by similar sources as VBR, with the additional influence of the neighboring 145 freeway.

The site 'AGO' is located in Riverside within the premises of the Citrus Research Center and the
Agricultural Experiment Station of the University of California, Riverside. It is 8 km further east of RUB,
and similarly influenced in terms of PM sources. It is about 750 m southwest of CA-60/I-215. This was

the furthermost inland site, located at the periphery of what can be termed as the populous region of
the LAB, and represents a regional background in terms of the eastern extent of LAB. Except for the
neighboring freeway, it has no other primary emission sources nearby.

152 The San Bernardino site, 'SBR,' is located at another AQMD monitoring station in San Bernardino area.

153 The major roadways next to the site have moderate traffic. It is the farthest inland site, about 115 km

154 inland and 95 km east of USC, located 6 km from the base of San Gabriel Mountains.

Particle number size distributions (PNSD) were measured at USC, the urban background site located in
the source region of the LAB as well as at UPL and AGO, both in the receptor region of the basin, with
UPL being at its northern edge and AGO at its eastern.

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2.2 Instrumentation

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Total particle number concentrations were measured at all sites using Condensation Particle Counters 161 162 (CPC, Model 3022A, TSI, Inc., Shoreview, MN). A Scanning Mobility Particle Sizer (SMPS, Model 3936, 163 TSI, Inc., Shoreview, MN) was used at select sites to measure the particle size distributions. The CPC used can measure with about 100% efficiency particles above 20 nm and has 50% detection efficiency 164 165 for a diameter of 7 nm. The upper size range for detection is 3 µm. The CPC recorded data at one-166 minute interval. The sampling rate was maintained at 1.5 ± 0.2 liters per minute and the air stream was not conditioned prior to sampling. The SMPS system consists of a long Differential Mobility Analyzer 167 168 (DMA< Model 3081, TSI, Inc., Shoreview, MN) and CPC 3022A (operating at 0.3 ± 0.03 liters per minute, 169 sheath air was not pre-conditioned), set to 5 minute scans covering the size range 14-736 nm. TSI 170 software Aerosol Instrumentation Manager was used to collect data from both the CPC and the SMPS. 171 Weekly site visits were made to ensure proper equipment operation and perform maintenance. Flow 172 rates were checked weekly and maintained within the range indicated in this section. All inlets used to 173 sample ambient aerosols were copper tubes of 1 cm diameter.

174 Meteorological data, i.e., temperature, relative humidity, wind speed and direction amongst other

parameters were collected using Vantage Pro 2 Weather Stations (Davis Instruments, Hayward, CA).

176 The meteorological station was placed above the enclosure and the wind vane sampled at a height of 5

m from the ground surface. The meteorological data were compared with neighboring AQMD stations
with more standardized meteorological equipments, and only for wind speed and direction slight
differences were observed. This was because of lower height of our equipment. Even then, the diurnal
patterns of these parameters were consistent with those reported by AQMD.

All particle equipment was placed in an air-conditioned enclosure, but there were instances in summer when temperatures exceeded the optimum operation temperature for the equipment (~ 35°C) and the data were screened out for such instances. At times during summer, water condensation was observed in the CPC. The CPC reservoirs were drained and the data for such events has been excluded from analysis.

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2.3 Data Processing and Validation

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188 Given the high temporal resolution of the data (i.e., 1-minute particle number concentration, 5-minute 189 size distribution scans, wind speed, wind direction, temperature, relative humidity, and other 190 parameters collected up to a year at 7 sites) it was not practical to provide detailed description and 191 interpretation of all data. Therefore, in this paper we present data as hourly averages and for 192 consistency, the hourly averages are reported in local time for the entire year. All collected data were thoroughly reviewed for irregularities, similar to the work of Puustinen et al. (2007). Data were not 193 included in averages if the counts reported were below 1000 particles/cm³ or exceeded 10⁶ 194 particles/cm³, which were associated with electronic errors in CPC. The data recovery rates are 195 196 reported in the Table 1. The lowest data recovery was reported for June 2009, when we experienced 197 excessive water condensation inside the CPC butanol reservoirs. Data from site VBR are not reported 198 after April 2009, as the measured concentrations were unreliable due to CPC malfunction. CPCs were operated side-by-side at USC for a 24-hour period before the commencement of the sampling 199 200 campaign to ascertain consistency. Data analysis indicated that the average slope of a CPC against the 201 'mean' CPC was 0.98 ± 0.16 and the range was 0.72-1.26. At the end of the study, the CPCs were set 202 up to run side-by-side for over 48 hours and each CPC concentration was compared to the 203 concentrations measured by a factory-calibrated CPC. The correlation coefficient between all the CPCs was in the range 0.86-0.99, even though two CPC reported an average slope less than 0.7 against a 204

factory-calibrated CPC. We elected to compare CPCs with a unit calibrated by the factory instead of the mean of the CPC values because the CPCs had been operating in field continuously for over two years, and several units used in earlier studies by our group had shown performance deterioration with prolonged field use. The data were corrected (assuming a linear deterioration in performance over the span of operating period) to compensate for the inconsistency between the CPCs. No corrections were made for diffusion losses, due to different inlet lengths, because our earlier characterization showed that they are insignificant (Moore et al., 2009).

212 Statistical methods used for analysis in the present study are discussed in our earlier work (Moore et al., 2009 and Krudysz et al., 2009.) The paper reports coefficients of divergence (COD) to analyze the 213 relationship between sites. While parameters such as the correlation coefficient are often used to 214 215 quantify a linear relationship between data sets, and in this context would quantify a fraction of 216 observations at a particular site that can be explained in terms of simultaneous observations made at 217 another sites, a high correlation between paired sites would only imply uniform temporal variation 218 (Lianou et al., 2007), but not the variability in itself amongst sites. The COD is in this context more 219 suitable to characterize this spatial variability (Wilson et al., 2005; Krudysz et al., 2009; Moore et al., 2009). It is defined as: 220

$$COD_{jk} = \sqrt{\frac{1}{n} \sum_{i=1}^{n} \left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}}\right)^2}$$

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Where j,k are two sites, n is the number of simultaneous observations. The value of COD varies from 0 (the concentration being identical at the two sites) to 1 (the concentration being different). A low value of COD represents a high level of homogeneity between sites and a value of COD above roughly 0.2 is considered to be generally heterogeneous (Wilson et al., 2005).

3. **Results and Discussion**

227 3.1 Meteorology

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Meteorological conditions can influence ultrafine particle concentrations significantly, but the Los 229 Angeles area exhibits relatively limited diurnal and seasonal variation, as was the case during the study 230 231 period. The mesoscale meteorology of the area that is most relevant in context of this study is the interaction of coastal winds with the San Gabriel Mountains. The pollution generated in west LA during 232 233 the morning is transported over the course of several hours of aging toward the eastern portion of the 234 Los Angeles Basin and up the southern flanks of the San Gabriel Mountains. The strong subsidence inversion layer, frequently present over the area in the winter and almost always in the summer, limits 235 the vertical dispersion and westerly sea breeze, which sets in during the afternoons, transports this 236 237 pollution further inland. This is also evident from inset plots in Figure 2 (a), 3 (a), 4(a) showing vector 238 average wind direction during three months (January, May and September) of 2009. Across the sites, winds were observed from the west during afternoons, at relatively higher speeds than most hours of 239 240 the day. As the mixing layer stabilizes during evenings, the trapped pollutants can linger overnight and 241 then be re-entrained to the surface during early morning hours in east LA (Lu et al. 1994, 1995). The 242 particle number concentrations and the size distributions will be discussed in this context.

Meteorology of the Los Angeles Basin and its effects on air pollutant's movements has been discussed 243 in greater detail by Blumenthal et al., (1978), Lu et al., (1994, 1995), McElroy et al., (1986, 1983), Shultz 244 245 et al., (1982), Ulrickson et al., (1990) and Wakimoto et al., (1986). Table 2 and 3 present an overview of select data for the stable meteorological conditions at sampling sites. Air temperatures do not vary 246 much across sites and the seasonal trend across sites is quite similar, with slightly lower temperatures 247 248 observed at sites further inland during winter. January was warmer than February, and September across sites was at least as warm as or warmer than August, which is quite typical of the area. The 249 250 relative humidity at all sites was consistent during sampling period, except during Santa Ana winds that 251 brought in dry winds from the desert, due to a synoptic high-pressure system, also typical of this time 252 of the year in southwest Unites States. The predominant wind direction at the sites, except for winter months (Dec-Feb), was from the west, with stronger winds from the west recorded during afternoons, 253

and nighttime stagnation being the most dominant winds speed characteristics in the basin.

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3.2 Diurnal and Seasonal Variations

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In this section, particle number concentrations (PNC) for different sites are discussed as diurnal, hourly
averaged, data for selected months. Alternate months of the year were chosen (unless another
particular month was more relevant) to maintain clarity in graphs and to illustrate the
similarities/differences across the diurnal, seasonal and spatial trends observed at these sites. The
relative standard error was less than 5%. The hourly average data presented is an arithmetic mean.
Further, the CODs are discussed in context of the spatial variability.

263 Figure 2 (a) shows the PNC hourly averages across the odd months of the year at USC. This site is regarded as a typical urban background site in Los Angeles. In the cooler months of late spring and late 264 265 fall, a characteristic early morning peak, associated with mostly light-duty gasoline vehicle morning commute, is observed from 5-10 am. Advancing into summer months, this peak is not as robust and 266 267 eventually flattens, as higher temperatures during the early mornings increase mixing heights, thus 268 enhancing dispersion, and also lead to possible volatilization of semi-volatile organics bound to PM 269 from traffic emissions (Biswas et al., 2007; Ning et al., 2007). However, another peak emerges, which 270 has its crest in early afternoon, and in summer months is associated with the highest diurnal values for 271 PNC. This peak has been identified with the secondary particle formation, and is consistent with the 272 work of Moore et al., (2009), Moore et al., (2007), Ning et al., (2007) and Verma et al., (2009). The 273 presence of this peak implies that secondary photochemical formation can contribute to PNC in some 274 months as significantly as primary emissions from local sources. Similar results have been reported by Costabile et al., 2009 and Wehner et al., 2007. During the cooler months of the year, another peak is 275 276 observed in the evenings and early night, which is weaker during summers, possibly related to particle 277 formation by condensation of semivolatile vapors emitted by traffic during preceding hours. The 278 depression of the atmospheric mixing layer during later hours of the day in cooler months can further 279 enhance the production of these elevated particle concentrations, and its effect is most pronounced in peak winter months (Dec-Feb), when night time concentrations can reach ca. 30,000/cm³. Biswas et al. 280 (2007) have previously reported a similar data pattern. The observations at USC suggest that PNC can 281

vary significantly at a site across seasons (morning commute peak in winters ~40,000/cm³ and in
summers ~15,000/cm³), even though they may be associated with consistent local emission sources, all
due to different meteorological conditions. Thus, when considering exposure to UFP, especially using a
number-based metric, meteorological conditions and secondary sources can be of as much
consequence as direct emissions from local sources.

287 Figures 2 (b & c) compare the average particle size distribution of during different time periods of the day at USC during September and December of 2009. As discussed above, the photochemical activity-288 289 related peak (observed during the afternoon period 12:00-14:00 hours, by when previously formed particles grow to a size range that is measurable by SMPS) is very robust in September and weakens 290 progressively through the fall and into December. Insets in Fig 2 b & c further elucidate this point by 291 292 comparing the particle size distribution during 10:00-14:00 hours between September and December, 293 i.e, the warmer and cooler months of our sampling campaign. In September, a simultaneous rise in 294 total particle numbers and the sub-25nm particles can be seen and is attributed to photochemical 295 formation. The possibility of these particles being associated with fresh (traffic) emissions is unlikely 296 because analysis of traffic trends of the neighboring freeway, (the major source of fresh emissions at 297 USC), confirms no significant changes either during the hours associated with photochemical activity, 298 or across seasons. Further, the increase in atmospheric mixing height during this time of day would decrease the concentrations of PM of primary origin. Traffic profiles (vehicle count/hr for the month of 299 Sep and Dec) are also shown in the inset figures. The tri-modal diurnal profile observed at USC during 300 warmer months in Figure 2(a) is limited to sub-50nm particles, while the seasonal variation of the 301 302 diurnal patterns for particles >100 nm is not clearly evident. This is a distinctly different pattern than 303 that observed at the inland sites, and illustrates a size distribution that is characteristically associated 304 with urban sites in proximity to primary emissions from vehicles (Morawska et al., 2007; Ronkko et al., 2006 & 2007). 305

Figure 3 (a) shows monthly-averaged diurnal particle number concentrations across six months of the
year at UPL, the northern receptor site (i.e. Nov, Jan, Feb, May, Aug & Sep). A bi-modal diurnal
distribution is observed at this site, with a morning time peak, similar to USC, corresponding to
morning commute during 6:00-10:00 hours in winter months that is not as robust during summer. This

310 winter peak is a compounded effect of vehicular emissions and lower mixing height in winter mornings. 311 (This is clearly evident in the inset in Figure 3 c). A gradual increase in concentration is observed as the 312 winter progresses. The formation of strong surface-based temperature inversions that can lead to 313 almost no vertical mixing (during winters) of the transported PM load, coupled with condensational growth of particles, is responsible for the extended late evening and early night peaks observed at UPL, 314 when PNC plateau overnight. Concentrations as high at 15,000/cm³ can be observed during winter 315 nights compared to only ca. 10,000/cm³ during summer. The nighttime peak is flatter, broader, and 316 persists longer than the morning traffic peak, and has concentrations that are comparable if not higher 317 than the morning peak, thus producing maximum diurnal concentrations during the night, when local 318 319 emissions are at their lowest. In comparison, the maximum concentration at USC in the evenings is 320 about half of the morning maximum. Other inland sites exhibit a similar pattern, with nighttime 321 maxima being comparable to morning maxima and the highest PNC being observed during winter 322 months. This concentration pattern may lead to a longer period of exposure to higher PNC in inland 323 areas than in areas with greater local emissions nearer the coast.

324 Figures 3 (b & c) compare the PNC in various size ranges at UPL. Between the warm September and cool December months there is a marked change in the diurnal pattern for different size ranges. The 325 326 afternoon peak in concentrations associated with photochemical activity, as observed at USC and later 327 at AGO, is not as prominent at UPL. Even though the PNSD during 10:00-14:00 hours indicates the presence of particles of sizes that could be attributed to photochemical activity, it is not accompanied 328 by a rise in total PNC, as is observed at USC and AGO. A possible explanation is that the contribution of 329 330 photochemical activity to the total PNC is obscured (and thus not as distinguishable) by the 331 contribution of the advected aerosols from the upwind urban areas of LAB to the overall PNC. Further, 332 since UPL is distant from major freeways, the concentrations of gaseous and semi-volatile organic 333 vapor precursors that participate in secondary particle formation are lower compared to those at USC (or in general in central LAB), which may decrease the degree of PM formation through this pathway. 334 Analysis of particle concentrations less than 25 nm and total particle concentrations, as reported by 335 the SMPS, during September further corroborate this hypothesis (shown as an inset in Figure 3 b). No 336 337 significant differences are observed in PNC < 25nm during 10:00-16:00. The results plotted in Figure 3 a

338 show that during 15:00-17:00 hours, when the highest wind speeds of the day are observed, the 339 particle concentrations in the range of 25-100 nm increase (while the 14-25 nm range remains stable). 340 This particle range is typically associated with coagulation and or growth of preexisting particles via 341 condensation of semi-volatile organics on pre-existing PM (Rodriguez et al., 2007). The increase in that 342 size range later in the afternoon (during other hours of summer days, the concentrations within this size range remain stable) could be due to the arrival of the polluted air mass from Los Angeles. Similar 343 344 observations have been made by Kim et al, 2002 and Fine et al. 2004. However, during winters (Figure 3 c) the distribution is uni-modal and the bi-modal distribution is only observed during evening/night 345 with distinctly higher mode diameter during winter. The size range of 14-25 nm, associated with fresh 346 emissions, shows a sharp increase during morning as well as in evening, as evident in the inset (total 347 348 PNC increases and the mode particle diameter decreases, shifting the distribution towards freshly emitted PM). This is due to the combined effects of local traffic, coupled with the decreasing 349 350 temperature (increasing the partitioning of semivolatile organic emissions towards the particulate 351 phase) and mixing height (which reduces dispersion), all of which lead to a more pronounced effect of 352 local emissions than that observed during summers. These comparisons suggest that there could be 353 significant distinction in the size distribution profiles observed at sites due to seasonal variation.

354 Figure 4 (a) shows data for AGO, one of the eastern most regional receptor sites of the study. Diurnal 355 averages are shown for late fall (Nov), winter (Jan), spring (Mar) and summer (May, July and 356 September). The morning peak in the plot can be explained by the morning commute (as this site is 357 near a freeway). However, this morning peak subsides as the year progresses into warmer months 358 when there is greater dispersion of fresh traffic emissions. Similar to UPL, during colder months, there 359 is an evening and early nighttime rise in concentrations, leading to PNCs comparable to that in 360 mornings. This peak diminishes in the summer and returns in September. Figure 4 (b & c) contrasts 361 particle size distributions during different time periods of the day. During September we observed a rise in the concentrations of smaller particles (<25nm) during the hours coincident with strong solar 362 363 irradiance and the mode diameter of the distribution decreases from ~30 nm at 11:00 to about 16-17 364 nm between 11:00-14:00. This decrease in mode diameter along with an increase in overall particle 365 numbers, indicates the possibility of new particle formation in the absence of significant changes in

366 traffic during this period. Further, this increase in midday PNC concentration is not observed in 367 December, and the peak declines steadily through the fall. Similar observations in that area have been 368 made previously by Fine et al. in 2004. The inset in Figure 4 b corroborates this argument by showing 369 an increase in particle concentrations in the 25-100 nm range in the evenings, similar to UPL, which is 370 attributed to the arrival of aged aerosol from the LAB. An increase in mode diameter, along with particle numbers, occurs consistently through the months of September to December for particles >25 371 372 nm in late afternoon. For reasons similar to UPL, the effect of local emissions is more pronounced in winter mornings at AGO, as evident in the morning peaks in sub 25 nm concentrations and the effect 373 of dilution as the day progresses can be seen in the inset in Figure 3 c. 374

The diurnal pattern in particle concentrations across these sites, (i.e., USC, AGO and UPL) is dominated 375 376 by a bi-modal distribution, except for summers at USC. The overall particle concentrations decrease 377 due to dispersion as the air parcels move inland (eastwards). The increase in nighttime concentrations 378 (at hours when there are limited fresh emissions) at AGO (easternmost receptor) are lower than at UPL 379 (northern receptor). PNCs at the RUB and VBR sites, which are further inland than UPL, are also lower 380 than at UPL, but higher than at AGO, which is further east of these sites. A similar pattern is observed 381 in the morning peaks corresponding to commute hours, because the traffic volume decreases as one 382 moves farther inland from Downtown Los Angeles.

383

Spatial Distribution of Particle Number Concentrations

384

3.3

Figures 5 (a & b) compare PNC at all sites for two months (during the warmer and cooler periods of the 385 386 year) to contrast spatial variation in the concentrations across the basin. A representative month from 387 each season was chosen and data have been plotted as the diurnal averages over the span of the month. 388

389 Figure 5 (a) shows a winter month data across sites. The all-hour average December 2008 390 temperatures across the inland sites ranged from 10.7 to 12.7 degrees Celsius while the relative 391 humidity ranged from 59 to 67%. The wind data in Table 2 shows the predominant wind direction 392 based on hourly vector averages for different sites. At all inland sites, the morning peak concentrations 393 during winter seem to be comparable to those of the nighttime peak (a mix of local evening commute

394 emissions and the arrival of advected PM from urban Los Angeles) that persist for a far longer period 395 than the morning peak does. This is an important observation since it suggests that, in the receptor 396 areas of the LAB, PM transported from central and west Los Angeles can contribute to higher and more 397 sustained concentration levels even during the hours when local sources have minimum contributions. 398 These results are also consistent with the findings of Zhu et al. (2006) and Hu et al. (2009) both 399 conducted in the LAB. The highest of morning concentrations were observed at USC and RUB, the two 400 sites closest to freeways. VBR, which is close to RUB, but farther away from any freeways, had lower concentrations during the morning commute. However, VBR and RUB show excellent agreement in 401 PNC during nighttime, when a stable stratification predominates the area. Though UPL (which is closer 402 403 to USC) and AGO (which is farther east) show higher and lower night time concentrations, respectively, compared to RUB and VBR. PNC at SBR the were measured by means of the TSI CPC 3025 in December 404 of 2009 (counting particles down to 3 nm compared to 7 nm of the 3022, and thus reporting higher 405 numbers) follows the diurnal pattern of inland sites. The degree of variability based on PNC was 406 examined using the Coefficient of Divergence, and the median value of COD is plotted for all site pairs 407 except SBR (since PNC for the latter are reported using a different instrument). The highest CODs, or 408 409 the maximum spatial variability, are observed during the hours of morning commute. The overall COD 410 range was 0.17-0.28, indicating that PNC are only moderately heterogeneous.

411 Figure 5 (b) shows the hourly averages at all sites during August, 2009. USC not only has the highest PNC, but also a very sharp midday peak (related to photochemical particle formation), which is 412 413 comparable to morning traffic-related peak, as discussed earlier. Nighttime PNC become comparable 414 to those at inland sites. The increased PNC pattern during morning commute is observed across all sites even though the numeric values of PNC differ significantly. The morning commute peaks however 415 416 are not as pronounced as those in winter (December, Figure 5 a) as the primary emissions are quickly 417 dispersed in summer and the higher ambient temperatures may be shifting the partitioning of semivolatile organics emitted by primary sources to the gas phase (Miracolo et al., 2010). Particle number 418 419 concentrations at all sites were generally lower in summer than in spring or winter. Post midday, there 420 is a steady rise in PNC concentrations in all receptor sites, which is due to the combined effects of 421 photochemical activity along with the contribution of advected PM from western Los Angeles. The

overall similarity in PNC data in all sites during overnight hours illustrates a well-dispersed regionalscale aerosol during summer nights. The lowest CODs were observed during summer, with the range
for August being 0.13-0.23. These data corroborate the effect of dispersion and advection on regional
scales as homogenizing factors leading to low variability at the inter-community level.

426 The hourly concentrations observed during this campaign varied across seasons, though the diurnal 427 variations were more consistent. The maximum diurnal change in PNCs across seasons was observed at 428 USC, along with the highest average concentrations. This was expected, as USC is located in the 429 immediate vicinity of a freeway (about 120 m downwind) and in the source region of the LAB. The higher concentrations observed in the fall/winter months were consistent with the work of Singh et al. 430 431 (2006). In comparison to our earlier study (Moore at al., 2009), which reported concentrations 432 comparable to USC at several sites in the Wilmington and West Long Beach area of Los Angeles, the 433 receptor sites had lower concentrations due to lower impact of heavy traffic emissions in the 434 immediate vicinity. During site selection, preference was given to sites not in the immediate vicinity of 435 a source, to differentiate between local and regional contributions to the measured PNC in these sites. 436 Figure 6 compares the concentrations observed during this study with earlier observations made by Singh et al. (2006), who reported PNC data 6-7 years earlier, using identical instrumentation at similar 437 438 sites. The sites AGO and UPL are referred to as Riverside and Upland by Singh et al., (2006). The Mira Loma site is about 8 km west of RUB. In general, the observed concentrations in the present study are 439 440 somewhat lower, which could be interpreted (with some caution) as an encouraging outcome of the 441 implementation of effective emission control technologies and the replacement of older heavy and 442 light duty vehicles by newer vehicles in the LAB. The seasonal patterns identified in this study are 443 consistent with the earlier observations by Singh et al. (2006).

Figures 7 (a & b) compare the CODs across summer and winter periods. Summer seems to be the season with lowest spatial variability; in fact, for the majority of the day, COD values were mostly below 0.2, indicating remarkable spatial homogeneity for a metropolitan area of this size and complexity in PM sources. The values are generally higher in winter, but still below 0.3, indicating only moderate heterogeneity. The deviation in CODs for all site pairs was highest for the hours in which primary local sources are predominant, implying that one or more sites with a heavy local influence

450 (which in most cases would be traffic) is increasing the COD. This was further ascertained by inspecting 451 individual site pair values. During both summer and winter, homogeneity is observed in late night and 452 early morning concentrations, indicating the presence of a regional aerosol. In comparison to our 453 previous study (Moore et al., 2009 and Krudysz et al., 2009) that reports median COD values of about 454 0.3-0.5 in source regions of the LAB (the range between first and third quartiles was on the order of 0.2 units), the values reported in this study are lower. This implies that in LAB, the *inter-community* 455 456 variability in PNC is lower than the *intra-community* variability of areas like the LA harbor, impacted by 457 a multitude of traffic, ship and industrial emissions in a much shorter spatial scale. The relative homogeneity at the inter-community level among receptor sites in LAB can be attributed to the effect 458 459 regional transport and meteorology that appear to override the contributions of local primary emissions. The effects of local traffic sources were also observed at the sites in this study, but were 460 restricted to morning and (only during winter) evening commute hours. 461

462 The spatial complexity of the PNC was further resolved with the size distribution data. Synergistic 463 effects of multiple factors can lead to similar particle number concentrations at two sites; however, the 464 shape in size distributions may be distinctly different at the two locations due to particle source composition. Wongphatarakul et al., 1998 showed that only moderately heterogeneous COD values 465 466 can be observed for chemical composition of particles even when the sources are different. Since 467 particle size distribution is as important for exposure classification, the spatial variability was assessed for different PM sizes. Overall CODs varied from 0.40-0.67, and exhibited a roughly inverse relationship 468 469 with particle size. This can in part be accounted for by the difference in sources and their magnitude 470 between USC and the inland sites as well as the PM size range, which would affect. This observation is 471 further supported by the lower COD values between the inland sites of AGO-UPL 0.35 (range 0.34-0.36) 472 compared to 0.55 (range 0.53-0.57) for USC-AGO (source and inland site). Even though the degree of 473 spatial heterogeneity is moderate for particles in bigger size ranges, this is the size range with minimal divergence in COD values observed for different site pairs. The data in Figure 7 reinforce the 474 475 observation that sites appear to be more homogeneous when the local sources (which contribute to 476 the smaller size spectrum of the particle size distribution more than the bigger size) are not dominant. 477 Similar observations were made by Turner et al. (2002) and Costabile et al. (2009).

478 4. Conclusions

479

480 Moderate inter-community variability in total particle number concentrations was observed across the sites of the eastern Los Angeles Basin. The extreme Coefficient of Divergence (COD) values were often 481 driven by a specific site pair, (site pair varied by hour and season), but the range of upper and lower 482 483 quartile of COD vales was mostly within 0.1 units, implying that Particle Number Concentration (PNC) 484 in these sites were homogeneous-to-moderately heterogeneous. Although, there were differences in 485 the spatial variability through different seasons, the temporal patterns were consistent, and exhibited 486 least variability in hours when local sources were not dominant. Comparable PNC can be observed in sites separated by several tens of kilometers overnight during stable stratification conditions. The 487 variability in size distributions (reflection of the source composition) was higher than that of total 488 489 particle number concentrations. Overall the spatial variability in PNC was lower than the values reported by Moore at al. 2009 for intra-community variability in urban "source" areas of the LAB. The 490 491 spatial variability based on particle size distributions support the notion of relative homogeneity in 492 receptor areas in LAB, where concentrations are dominated by aged aerosols, advected eastwards 493 from the source regions of urban Los Angeles, since the lowest variability was observed for particles in 494 the size range of 40-100 nm, associated with long-range transport, compared to sub-30 nm particles 495 associated with fresh emissions or new particle formation events. The largest differences in PNC were 496 observed between receptor sites and the source site at USC, while PNC were relatively homogeneous among the receptor sites. Further, the data suggest that meteorological conditions can contribute to 497 spatial homogeneity, when phenomena that are regional in nature (i.e, summertime photochemical 498 processes, long range transport, and higher degree of mixing) are active. 499

500 Even though our results suggest that PNC are moderately heterogeneous in the polluted receptor areas 501 of the LAB, concerns related to population exposure assessment based on monitoring from a central 502 station are still valid, especially in relation to urban areas impacted by a multitude of local and highly 503 variable sources. Moreover, despite the moderate heterogeneity in total PNC at the inter-community 504 level of receptor sites in LAB, particle size distributions may be significantly variable, resulting in 505 differences in the overall inhaled dose of PM mass. Efforts should be made to characterize the seasonal

nature of the variability in both size distributions and number concentrations, because meteorological
factors can influence both even when PM sources are similar.

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509

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775 Table 1 a: Site information including the designation code, geographic co-ordinates, site and equipment elevations, sampling

period and (CPC) data recovery^a

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Site ID	Latitude	Longitude	Site	Inlet	Distance from nearest	Sampling period	Data recovery
			elevation	elevation	Freeway (m), [Average		(%)
			(m)	(m)	Vehicles/day]		
USC	34°1'9" N	118° 16' 39" W	61	4.6	120, [112000]	11/17/2008 - 12/21/2009	91%
DIA	34° 0' 1" N	117° 49' 54" W	223	2.0	200, [99000]	02/25/2009 - 12/21/2009	96%
UPL	34° 6' 14"N	117° 37' 45" W	386	1.9	2000,[96000]	11/17/2008 - 12/21/2009	90%
VBR	33°59' 45"N	117° 29' 31" W	220	1.9	3000,[85000]	11/17/2008 - 04/30/2009	95%
RUB	33°59' 58"N	117° 24' 58" W	248	2.0	200,[72000]	11/17/2008 - 12/21/2009	93%
AGO	33°57' 41"N	117° 20' 0" W	323	2.1	750,[81000]	11/17/2008 - 12/21/2009	98%

a: The SMSP were operated at sites USC, UPL and AGO from 09/04/2009 - 12/21/2009 at greater than 90% data recovery.

Table 1 b: Site information regarding dominant local sources

Site ID	Relevant Information on Potential PM Sources
USC	Located next to a gasoline dominated freeway in a parking lot, urban background site
DIA	Located on a hill in a parking lot, at over 100 m elevation from the neighboring freeway
UPL	Located at the foothills of mountain range, limited local sources, regional site in terms of northern extent of LAB
VBR	Located in a residential area, rural regional site
RUB	Located behind an office building near a low trafficked street and a freeway
AGO	Located in an agricultural research facility near a university, freeways nearby, most inland, regional background site

Dominant Wind Direction and Wind Speed													
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
AGO	20	08		2009									
WD (deg)	SW	Е	Е	Е	W	W	W	W	W	W	W	W	Е
WS (m/s)	0.70	0.89	1.21	0.93	1.21	1.31	1.17	1.23	1.13	1.04	0.96	1.37	0.93
SD (m/s)	0.70	0.81	1.35	0.82	1.22	1.21	1.10	1.16	1.21	1.13	1.23	1.55	1.04
DIA								20	09				
Month				Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)				S	S	SW	S	W	W	W	S	W	S
WS (m/s)				0.62	0.47	0.53	0.50	0.41	0.40	0.38	0.31	0.39	0.35
SD (m/s)				0.81	0.48	0.48	0.40	0.30	0.33	0.36	0.32	0.56	0.55
RUB	20	08					20	09					
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)	NW	Ν	Ν	Ν	W	W	W	W	W	W	W	W	NW
WS (m/s)	0.53	0.96	2.38	0.78	0.97	0.84	0.84	0.71	0.70	0.62	0.62	1.00	1.28
SD (m/s)	0.75	1.49	2.66	0.98	1.20	0.63	0.56	0.57	0.60	0.54	0.67	1.48	0.37
UPL	20	08	2009										
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)	W	Ν	Ν	W	SW	SW	W	W	W	W	W	W	W
WS (m/s)	0.39	0.48	0.52	0.56	0.86	1.14	1.15	1.19	1.11	1.01	0.90	0.80	0.65
SD (m/s)	0.37	0.46	0.47	0.53	0.69	0.94	0.92	0.89	1.05	0.93	0.87	0.70	0.51
VBR	20	08						2009					
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)	W	W	Ν	W	SW	W	SW	W	W	SW	W	W	W
WS (m/s)	0.45	0.64	2.04	0.67	0.92	1.01	1.03	1.09	1.01	0.92	0.88	1.02	0.43
SD (m/s)	0.81	1.18	2.27	0.92	1.08	1.03	0.89	0.92	1.00	0.90	1.02	1.19	0.56
USC								2009					
Month			Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)			NE	NE	W	W	W	W	W	W	W	NE	NE
WS (m/s)		2.23	2.41	2.44	2.71	2.50	2.53	2.66	2.74	2.45	2.58	2.34	
SD (m/s)			0.86	0.97	1.04	1.16	0.96	1.04	1.05	1.12	1.02	1.05	0.77

Table 2: Prevailing Wind Direction and Speed at sampling sites

Sites	AGO		DIA		RUB		SBR	
Months	RH	Temp	RH	Temp	RH	Temp	RH	Temp
Dec '08	64 ± 22	11.3 ± 5.0			62 ± 23	12.1 ± 5.6	59 ± 20	11.9±6.1
Jan '09	44 ± 23	15.4 ± 5.0			42 ± 26	16.2 ± 6.1	47 ± 23	14.5 ± 6.6
Feb '09	63 ± 23	12.1 ± 5.3	67 ± 22	67.3 ± 21.7	64 ± 24	12.8 ± 5.7	62 ± 20	12.1 ± 5.9
Mar '09	58 ± 21	14.0 ± 5.2	64 ± 19	63.5 ± 19.4	58 ± 21	15.1 ± 5.7	55 ± 18	14.8±5.7
April '09	55 ± 21	16.0 ± 6.3	58 ± 21	58.2 ± 21.0	54 ±21	17.2 ± 6.6	53 ± 19	16.7 ± 6.5
May '09	65 ± 17	19.9 ± 5.3	69 ± 15	68.7 ± 14.8	62 ± 18	21.4 ± 5.8	58 ± 16	21.5 ± 5.5
June '09	66 ± 16	19.7 ± 5.1	69 ± 14	69.0 ± 13.8	65 ± 16	20.8 ± 5.2	60 ± 15	21.0 ± 5.3
July' 09	52 ± 18	25.9 ± 6.0	58 ± 18	58.4 ± 18.4	52 ± 19	28.0 ± 6.4	47 ± 16	27.7 ± 5.9
Aug '09	53 ± 22	24.9 ± 6.4	55 ± 22	55.2 ± 21.7	52 ± 21	26.2 ± 6.9	49 ± 18	26.3±6.4
Sep '09	47 ± 22	26.0 ± 6.6	53 ± 22	52.8 ± 21.7	52 ± 21	26.2 ± 7.0	47 ± 18	26.5 ± 6.7
Oct '09	52 ± 23	18.8 ± 5.8	56 ± 24	55.8 ± 23.9	52 ± 24	19.7 ± 6.4	49 ± 20	19.3 ± 6.5
Nov'09	47 ± 24	16.6 ± 5.5	51 ± 25	51.3 ± 24.8	49 ± 25	17.1±6.3	48 ± 21	16.2 ± 6.8
Dec '09	67 ± 18	12.6 ± 2.9	68 ± 16	68.4 ± 16.4	67 ± 18	13.6±3.8	65 ± 16	12.8 ± 4.0

787 Table 3: Temperature (^oC) and Relative Humidity (%) at sites during sampling period

Sites	U	PL	VBR		USC	
Months	RH	Temp	RH	Temp	RH	Temp
Dec '08	67 ± 21	10.7 ± 5.4	62 ± 26	12.7 ± 5.8		
Jan '09	52 ± 25	14.2 ± 5.9	44 ± 29	16.3 ± 6.7	73 ± 16	14.0 ± 3.4
Feb '09	64 ± 24	11.9 ± 5.6	68 ± 25	12.1 ± 5.7	83 ± 08	12.2 ± 1.8
Mar '09	67 ± 18	13.3 ± 5.3	64 ± 22	14.0 ± 5.4	72 ± 19	13.6±3.1
April '09	59 ± 22	16.0 ± 6.6	59 ± 21	16.0 ± 6.3	68 ± 14	14.6±4.0
May '09	66 ± 17	19.4 ± 5.4	68 ± 17	19.9 ± 5.1	78 ± 08	17.5 ± 1.7
June '09	67 ± 16	20.2 ± 4.8	66 ± 17	21.0 ± 5.1	76±07	17.7 ± 1.1
July' 09	60 ± 19	24.2 ± 5.5	62 ± 19	24.7±6.1	69 ± 11	22.7 ± 3.4
Aug '09	61 ± 20	23.4±5.7	59 ± 22	24.2±6.4	64 ± 18	22.4 ± 4.4
Sep '09	56 ± 22	24.3±6.4	57 ± 23	24.8±6.8	66 ± 17	23.1 ± 4.2
Oct '09	57 ± 23	18.2 ± 5.9	57 ± 25	18.5 ± 6.0	60 ± 21	19.1 ± 3.9
Nov'09	54 ± 23	15.7 ± 5.9	55 ± 27	16.2 ± 6.4	52 ± 22	16.7 ± 4.3
Dec '09	72 ± 19	12.0 ± 3.6			58 ± 20	13.8 ± 3.8



791 Figure 1: Location of sampling sites in Los Angeles air basin.



792

Figure 2a: Hourly average particle number concentration at USC plotted for hours of the day in local time. The relative
 standard error for the hourly averages reported above was less than 2%. The inset is a plot of vector averaged wind

direction (WD) with the bubble area weighed to wind speed plotted for hours of the day in local time.



Figure 2b: Average Size Distribution of Particles during six time periods (local time) of the day at USC during September
 2009.



Figure 2c: Average Size Distribution of Particles during six time periods (local time) of the day at USC during December 2009.



Figure 3a: Hourly average particle number concentration at UPL for hours of the day in local time. The relative standard
 error for the hourly averages reported above was less than 2%. The inset is a plot of vector averaged wind direction (WD)
 with the bubble area weighed to wind speed plotted for hours of the day in local time.



Figure 3b: Average Size Distribution of Particles during six time periods (local time) of the day at UPL during September
 2009.



Figure 3c: Average Size Distribution of Particles during six time periods (local time) of the day at UPL during December
 2009.



818 Figure 4a: Hourly average particle number concentration at AGO for hours of the day in local time. The relative standard

error for the hourly averages reported above was less than 3%. The inset is a plot of vector averaged wind direction (WD)
with the bubble area weighed to wind speed plotted for hours of the day in local time.



Figure 4b: Average Size Distribution of Particles during six time periods (local time) of the day at AGO during September
2009.



Figure 4c: Average Size Distribution of Particles during six time periods (local time) of the day at AGO during December
2009.



830 Figure 5a: PNC and Coefficients of Divergence across sites for December 2008.



832 Figure 5b: PNC and Coefficients of Divergence across sites for August 2009.



Figure 6: Comparison of PNC at select sites measured during 2008/09 with Singh et al. (2006) measured during 2002/03.







840 Figure 7b: Coefficients of divergence during the winter months of Dec 2008-Feb 2009.