Response to the comments of Anonymous Referee #1

General Comments

The manuscript discusses the spatial variability in particle number concentration (PNC) in the urban area of Los Angeles air basin (LAB). Contemporary measurements of PNC were taken at seven sites in LAB for about one year. The concepts of ultrafine particles in urban areas, both in terms of particle number and size distribution, compared to PM mass, and the difficulty to assess human exposure to their sharp number concentration spatial gradients, is well introduced, and clearly motivated the manuscript.
The work is original, but not completely new, and authors should probably give more credit to previous related works. One critical point is about the methodology used to assess aerosol variability: If authors fully discussed particle number size distribution data instead of total PNC, they would probably get more robust conclusions.
Overall, it is a well-written manuscript addressing a relevant scientific topic within the scope of ACP, and substantial and interesting conclusions are reached. I believe the paper merits publication in ACP after considering the following specific comments, which should be addressed before publication.

Response

We have added Particle Number Size Distribution (PNSD) analysis (in three size ranges, as suggested by the reviewer in the following comment) for each of the three sites that particle size distribution data was collected at; in the forms of inset figures in Fig 2: b,c, 3: b,c and 4: b,c along with relevant text in the discussion.

Specific comments

- Pag.13916, lines 20-21, and throughout the manuscript: I wonder whether the overall similarity in total PNC data in seven sites of LA basin can be an evidence for a well dispersed regional-scale aerosol. I would conversely say that aerosol populations - i.e. nucleation mode, Aitken mode, accumulation mode particles, etc., have to be analyzed to get similar conclusions. I suggest to fully discuss particle number size distribution (PNSD) data, to make the conclusions on aerosol spatial variability more robust. (In this regard, PNSD measurements operated in parallel should be presented in a clearer way.) It is worth noting that when authors analyse aerosol particle spatial variability with the PNSD data (pag.13918, lines 10–25), overall CODs (Coefficients of Divergence) increase up to 0.67 and exhibit an inverse relationship with particle size. (Similar results were discussed by Costabile et al., Atmos. Chem. Phys., 9, 3163–3195, 2009.)

Response
The suggested comments are incorporated in the new section added, in which we conducted analysis on the three size ranges, as suggested by the reviewer. Though only a sub section of data is presented, Sep and Dec (the warmer and cooler periods, respectively), the data presented was collected concurrently with very good recovery rates and reinforces the points made about monthly averages. The PNSD analysis of this section shows consistency across the LAB as well as at the specific sites. The results

indicate that even if PNC look similar, there is a substantial variation in the size range distribution at each site. The analysis further bolsters the earlier conclusions of the paper.

- Paragraph 3.2 and Figg. 2a, 3a, 4a: Interestingly, the plots of total particle number concentration (PNC) show a three-peak structure. Further insights to the interpretation given by the authors are likely to be provided by the analysis of the weekly PNC variability, e.g. week-end vs. week-day. More specifically, it would be interesting to add typical traffic flow profiles of the nearest freeways in order to compare trends and peaks. (E.g., this could help to understand the reason why, in Fig.2a, the hourly average PNC at the USC urban background site shows a morning peak from 5:00 to 10:00 a.m., whereas in July, the same morning peak starts at 4:00 a.m.) I believe that this could also aid in evaluating the contribution of traffic volume particle emissions to the third evening peak of PNC. This contribution can be as relevant as the contribution during the morning rush hours, the lower total PNC being due to a more intense vertical mixing after midday than during the early morning. (Similar results were discussed by Birmili et al., Atmos. Chem. Phys., 9, 2355-2374, 2009.)

Response

Traffic flow patterns were added to the inset figures to enunciate the points made by the reviewer as well as our observations.

The one-hour shift in peak as pointed by the reviewer is because of time shift from PST to PDT. In the last version submitted, we presented all our data in PST, which explains why it looks like 4 am when it is actually 5 am local time. In this version all data is present in the local time at the site. We hope that this correction clarifies this point

- Paragraph 3.2: I suggest the authors to describe here more clearly the spatial representativeness of the selected sites with the aim to enhance text comprehension and readability. (This information should also be included in Table 1 by adding a column for site representativeness - e.g.: “USC = urban background” - and a column for the prevailing aerosol sources - e.g.: nearest freeway, regional transport, etc.) Besides, authors should explain why they decided to discuss three sites only - USC, UPL, and AGO. It is not clear if and why SMPS measurements were operated only at those three sites. (Perhaps, those sites are the most representative ones in terms of urban background, downwind regional background, and remote regional background.) Finally, I suggest to name the sites according to their representativeness , e.g.: “the urban background site” instead of “USC”. (The frequent referring to acronyms, which are apparently un-meaningful. g. UPL, AGO, USC, makes it hard for the reader to follow through the text in some parts of the manuscript.)

Response

A table (1b) has been added to summarize the sources around the sites.

Table 1 b: Site information regarding dominant local sources

Site ID	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**

We would prefer to use the acronyms, but concise reference terms, such as "urban background" or "regional" have been added at the beginning of sections where these sites are discussed to provide the reader with perspective. A concise discussion has been added in the site descriptions section on why the three sites were chosen to measure PNSD.

Relevant text from manuscript:

> "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."

Additionally, the section discussing Site Locations was appended with relevant texts to incorporate reviewer's comments.

- The early afternoon peak at the USC urban background site has been apportioned to secondary particle formation. It is even more evident in the summer profiles of PNC at the USC site, but the corresponding PNC values in winter are as high as in summer, or even more. (PNC is up to 16000 cm□3 in January, and up to 13000 cm□3 in September.) A similar early afternoon peak is also evident in winter months at the other two sites, AGO and UPL. The findings give evidence for a significant contribution to PNC due to secondary photochemical formation in urban areas. This issue should probably be better discussed in the revised manuscript, including the related PNSD analysis.

It is difficult to conclude the effect of photochemistry at USC site during winter given the size range we are measuring, even though the concentration in winter afternoons is comparable to that of summer.

The contribution of secondary formation during summer is however quite evident and discussed in the PNSD analysis section added to the manuscript.

- Pag. 13913, line 29: Fresh emissions are associated to the 14-25 nm size range. The 25-100 nm size range is contrarily associated with coagulation and/or growth of the pre-existing particles. However, where both these size ranges increase together - e.g., evenings in September at the UPL site, I think a soot-mode due to freshly emitted particles has also to be discussed.

An analysis of PNSD at UPL during September shows that these two size ranges do not increase together. Fig 3c inset shows that point clearly.

- Pag 13914, lines 4-6: Authors conclude that "there could be significant distinction in the size distribution profiles observed at sites due to seasonal variation". I would suggest to provide more conclusions about the findings, including a categorization of results according to: site (spatial) representativeness, diurnal and seasonal profiles, and prevailing local emissions.

This particular sentence has been deleted from the manuscript, but the idea that seasonal variation is present in particle size distributions is discussed when sites USC, UPL and AGO are discussed individually. The PNSD analysis provided context for this.

- Paragraph 3.2, figg.2, 3, 4: The modality of particle number size distribution is only qualitatively discussed, in terms of graphical visualization of the mode. As it is, the discussion can be strongly misleading since it depends on the y-axis scale of the graphs. (In this regard, it is not clear to me why the y-axes have a different - linear - scale for each plot.) I suggest to either mathematically quantify the modality or to provide a more detailed - and standardized- discussion. (I also suggest authors to re-plot the figg. 2b, 2c, 3b, 3c, 4b, 4c using a standard y-axis scale in order to enhance their inter-comparison.)

We can change the scale of graphs to be the same, but then the AGO is dwarfed in comparison to USC/UPL and we loose the ability to discern any diurnal variations in AGO. We have made the scale same for USC and UPL.

• Pag. 13912, lines 12-13: I suggest to use "not clearly evident" instead of "not significant". ("The seasonal variation of the diurnal patterns for particles > 100 nm is not clearly evident".) I believe that in order to conclude that it is "not significant", one should provide a quantification of PNC variability instead of a graphical analysis only.

Changed.

- Pag.13913, lines 19-28, and Figg.3b and 3c: I believe that the difference of PNSD observed in the early afternoon from 15:00 to 17:00 at the UPL site (downwind of LA) between September and December is interesting and probably lines 27-29 should discuss the point with more details.

The revised text discussing Fig 3 b & c discusses this.

• Pag.13915, lines 10-20: The transport of PM downwind of LA during afternoon and evening hours is a relevant topic. It is particularly interesting with respect to the transformation of the particle number size distribution (PNSD) from the particle and gas emission point to the downwind receptor sites. It would be interesting to include time/particle diameter/PNSD plots of some selected typical days, and discuss the findings.

PNSD data and accompanying text have been added to the manuscript

• Pag.13920, line 19: I think that authors should also mention here that sub-30 nm particles can also be associated to previous new particle formation events.

Added.

All technical corrections suggested by the reviewer have been made.

Inter-community variability in total particle number concentrations in the eastern Los Angeles air basin

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Abstract

Ultrafine Particles (UFP) can display sharp gradients in their number concentrations in urban environment due to their transient nature and rapid atmospheric processing. The ability of using air pollution data generated at a central monitoring station to assess exposure relies on our understanding of the spatial variability of a specific pollutant associated with a region. High spatial variation in the concentrations of air pollutants has been reported at scales of 10s of km for areas affected by primary 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 assumption of homogeneity in both of these metrics. This study was conducted at six sites, one in 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) and Scanning Mobility Particle Sizer (SMPS). The seasonal and diurnal variations of PNC implied that PNC might vary significantly with meteorological conditions, even though the general patterns at the sites may remain generally similar across the year due to consistency of sources around them. Regionally transported particulate matter (PM) from upwind urban areas of Los Angeles lowered

spatial variation by acting as a “homogenizing” factor during favorable meteorological conditions. 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, CODs), averaged 0.3, which was generally lower than that based on specific size ranges. Results showed an inverse relationship of COD with particles size, with fairly uniform values in the particle range which is associated with regional transport. Our results suggest that spatial variability, even in the receptor regions of Los Angeles Basin, should be assessed for both PNC and size distributions, and should be interpreted in context of seasonal and diurnal influences, and suitably factored if values for exposure are ascertained using a central monitoring station.

1. Introduction

Numerous recent epidemiological and toxicological studies investigating associations between 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 al. 2006; Delfino et al., 2005 & 2009) compared to particles of greater diameters. In vitro toxicological studies have also shown that ultrafine particles have higher oxidative potential and can penetrate and destroy mitochondria within epithelial cells (Li et al., 2003). Penttinen et al. (2001) found that daily mean number concentration and peak expiratory flow (PEF) are negatively associated and that the effect is most prominent for particles in the ultrafine range. A study by Peters et al. (1997) also found associations between number concentrations of ultrafine PM and lowered PEF among asthmatic adults.

Although current federal standards for particulate matter (PM) are mass-based, there is increasing 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 (Hinds 1999). Current standards are based on PM2.5 and PM10, , although poor correlation has been reported between PNC (dominated by UFP) and PM2.5 (dominated by accumulation mode particles) (Sardar et al., 2004). Not only is PM2.5 not an adequate surrogate measure, but also such data is often used from central monitoring stations to ascertain exposure values that might lead to exposure misclassification due to spatial variability in UFP concentrations (Delfino et al., 2005). Urban environments are often characterized by a complex set of factors (sources, meteorology, solar 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., 2009). 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). In urban areas, a dominant source of UFP is primary emissions from vehicular sources (Shi et al., 2001; Phuleria et al., 2005; Fine et al., 2004) and as much as 80% particles can be in the UFP size range (Morawska et al., 1998; Shi et al., 2001; Sioutas et al., 2005). Other combustion sources, such as food cooking and wood burning, can also be sources of ultrafine particles to the atmosphere (Kleeman et al., 1999; Schauer et al., 2001). Shi et al. (2001) have shown that nanoparticles from fresh emissions can be 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. Consequently, given their short lifetimes, the gradient of UFP concentration in atmosphere can be strong (Sioutas et al., 2005).

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. (2004) reviewed particle formation by secondary processes and showed that such particle formation events are more distinct in summer. Particle formation rates depend strongly on the intensity of solar radiation, but the exact mechanism by which the process occurs is not fully understood (Zhang et al., 2002). Once formed, particles are transformed in the atmosphere, by coagulation and condensation of semivolatile vapors on their surface as they are advected downwind. This long-range transport as well as photochemical particle formation in the atmosphere can lead to increased particle number observations downwind of urban areas (Kim et al., 2002; Fine et al., 2004; Verma et al., 2009; Ning et al., 2007).

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

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

2. Experimental Methods

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).

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

2.1 Site Descriptions

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.

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 surrounded by low trafficked streets. The site is located close the base of the San Gabriel Mountains, which act as a barrier for further transport of aerosols in northern direction The site is therefore influenced mostly by the aged aerosol advected eastwards from urban Los Angeles (Fine et al., 2004)

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 the nearest freeway and the major roadways next to the site have low traffic load. A substantial component of PM at this site can be attributed to the PM advected from the west to this area after hours of aging. This site has also been discussed in (Pakbin et al., 2010; Moore at al., 2010)

The Rubidoux site, 'RUB,' is located about 100 km inland from the ocean and 80 km east of USC (and 8 km east of VBR) at an AQMD sampling station. It is in vicinity of CA-60, situated about 200 m to its south. This site is impacted by similar sources as VBR, with the additional influence of the neighboring 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.

~~The San Bernardino site, 'SBR,' is located at another AQMD monitoring station in San Bernardino area. The major roadways next to the site have moderate traffic. It is the farthest inland site, about 115 km 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.

2.2 Instrumentation

Total particle number concentrations were measured at all sites using Condensation Particle Counters (CPC, Model 3022A, TSI, Inc., Shoreview, MN). A Scanning Mobility Particle Sizer (SMPS, Model 3936, 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 for a diameter of 7 nm. The upper size range for detection is 3 µm. The CPC recorded data at one-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 (DMA< Model 3081, TSI, Inc., Shoreview, MN) and CPC 3022A (operating at 0.3 ± 0.03 liters per minute, sheath air was not pre-conditioned), set to 5 minute scans covering the size range 14-736 nm. TSI software Aerosol Instrumentation Manager was used to collect data from both the CPC and the SMPS. Weekly site visits were made to ensure proper equipment operation and perform maintenance. Flow rates were checked weekly and maintained within the range indicated in this section. All inlets used to sample ambient aerosols were copper tubes of 1 cm diameter.

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.

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.

2.3 Data Processing and Validation

Given the high temporal resolution of the data (i.e., 1-minute particle number concentration, 5-minute size distribution scans, wind speed, wind direction, temperature, relative humidity, and other parameters collected up to a year at 7 sites) it was not practical to provide detailed description and interpretation of all data. Therefore, in this paper we present data as hourly averages and for 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 included in averages if the counts reported were below 1000 particles/cm^3 or exceeded 10^6 particles/cm^3, which were associated with electronic errors in CPC. The data recovery rates are reported in the Table 1. The lowest data recovery was reported for June 2009, when we experienced excessive water condensation inside the CPC butanol reservoirs. Data from site VBR are not reported 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 campaign to ascertain consistency. Data analysis indicated that the average slope of a CPC against the '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 up to run side-by-side for over 48 hours and each CPC concentration was compared to the 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

205 factory-calibrated CPC. We elected to compare CPCs with a unit calibrated by the factory instead of
206 the mean of the CPC values because the CPCs had been operating in field continuously for over two
207 years, and several units used in earlier studies by our group had shown performance deterioration with
208 prolonged field use. The data were corrected (assuming a linear deterioration in performance over the
209 span of operating period) to compensate for the inconsistency between the CPCs. No corrections were
210 made for diffusion losses, due to different inlet lengths, because our earlier characterization showed
211 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
213 al., 2009 and Krudysz et al., 2009.) The paper reports coefficients of divergence (COD) to analyze the
214 relationship between sites. While parameters such as the correlation coefficient are often used to
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.,
220 2009). It is defined as:

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

221

222 Where j,k are two sites, n is the number of simultaneous observations. The value of COD varies from 0
223 (the concentration being identical at the two sites) to 1 (the concentration being different). A low
224 value of COD represents a high level of homogeneity between sites and a value of COD above roughly
225 0.2 is considered to be generally heterogeneous (Wilson et al., 2005).

3. Results and Discussion

3.1 Meteorology

Meteorological conditions can influence ultrafine particle concentrations significantly, but the Los Angeles area exhibits relatively limited diurnal and seasonal variation, as was the case during the study 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 the morning is transported over the course of several hours of aging toward the eastern portion of the 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 the vertical dispersion and westerly sea breeze, which sets in during the afternoons, transports this pollution further inland. This is also evident from inset plots in Figure 2 (a), 3 (a), 4(a) showing vector 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 the day. As the mixing layer stabilizes during evenings, the trapped pollutants can linger overnight and then be re-entrained to the surface during early morning hours in east LA (Lu et al. 1994, 1995). The 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 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) 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 much across sites and the seasonal trend across sites is quite similar, with slightly lower temperatures 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 relative humidity at all sites was consistent during sampling period, except during Santa Ana winds that brought in dry winds from the desert, due to a synoptic high-pressure system, also typical of this time 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,

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

3.2 Diurnal and Seasonal Variations

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.

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 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 eventually flattens, as higher temperatures during the early mornings increase mixing heights, thus enhancing dispersion, and also lead to possible volatilization of semi-volatile organics bound to PM from traffic emissions (Biswas et al., 2007; Ning et al., 2007). However, another peak emerges, which has its crest in early afternoon, and in summer months is associated with the highest diurnal values for PNC. This peak has been identified with the secondary particle formation, and is consistent with the work of Moore et al., (2009), Moore et al., (2007), Ning et al., (2007) and Verma et al., (2009). The presence of this peak implies that secondary photochemical formation can contribute to PNC in some 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 observed in the evenings and early night, which is weaker during summers, possibly related to particle formation by condensation of semivolatile vapors emitted by traffic during preceding hours. The depression of the atmospheric mixing layer during later hours of the day in cooler months can further 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/cm^3. Biswas et al. (2007) have previously reported a similar data pattern. The observations at USC suggest that PNC can

vary significantly at a site across seasons (morning commute peak in winters ~40,000/cm^3 and in summers ~15,000/cm^3), 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.

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-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 progressively through the fall and into December. Insets in Fig 2 b & c further elucidate this point by comparing the particle size distribution during 10:00-14:00 hours between September and December, i.e, the warmer and cooler months of our sampling campaign. In September, a simultaneous rise in total particle numbers and the sub-25nm particles can be seen and is attributed to photochemical formation. The possibility of these particles being associated with fresh (traffic) emissions is unlikely because analysis of traffic trends of the neighboring freeway, (the major source of fresh emissions at USC), confirms no significant changes either during the hours associated with photochemical activity, 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 Sep and Dec) are also shown in the inset figures. The tri-modal diurnal profile observed at USC during warmer months in Figure 2(a) is limited to sub-50nm particles, while the seasonal variation of the diurnal patterns for particles >100 nm is not clearly evident. This is a distinctly different pattern than that observed at the inland sites, and illustrates a size distribution that is characteristically associated with urban sites in proximity to primary emissions from vehicles (Morawska et al., 2007; Ronkko et al., 2006 & 2007).

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

winter peak is a compounded effect of vehicular emissions and lower mixing height in winter mornings. (This is clearly evident in the inset in Figure 3 c). A gradual increase in concentration is observed as the winter progresses. The formation of strong surface-based temperature inversions that can lead to 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, when PNC plateau overnight. Concentrations as high at 15,000/cm^3 can be observed during winter nights compared to only ca. 10,000/cm^3 during summer. The nighttime peak is flatter, broader, and persists longer than the morning traffic peak, and has concentrations that are comparable if not higher than the morning peak, thus producing maximum diurnal concentrations during the night, when local emissions are at their lowest. In comparison, the maximum concentration at USC in the evenings is about half of the morning maximum. Other inland sites exhibit a similar pattern, with nighttime maxima being comparable to morning maxima and the highest PNC being observed during winter months. This concentration pattern may lead to a longer period of exposure to higher PNC in inland areas than in areas with greater local emissions nearer the coast.

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 afternoon peak in concentrations associated with photochemical activity, as observed at USC and later 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 by a rise in total PNC, as is observed at USC and AGO. A possible explanation is that the contribution of photochemical activity to the total PNC is obscured (and thus not as distinguishable) by the contribution of the advected aerosols from the upwind urban areas of LAB to the overall PNC. Further, since UPL is distant from major freeways, the concentrations of gaseous and semi-volatile organic 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. Analysis of particle concentrations less than 25 nm and total particle concentrations, as reported by the SMPS, during September further corroborate this hypothesis (shown as an inset in Figure 3 b). No significant differences are observed in PNC < 25nm during 10:00-16:00. The results plotted in Figure 3 a

show that during 15:00-17:00 hours, when the highest wind speeds of the day are observed, the particle concentrations in the range of 25-100 nm increase (while the 14-25 nm range remains stable). This particle range is typically associated with coagulation and-or growth of preexisting particles via condensation of semi-volatile organics on pre-existing PM (Rodriguez et al., 2007). The increase in that 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 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 with distinctly higher mode diameter during winter. The size range of 14-25 nm, associated with fresh emissions, shows a sharp increase during morning as well as in evening, as evident in the inset (total 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 temperature (increasing the partitioning of semivolatile organic emissions towards the particulate phase) and mixing height (which reduces dispersion), all of which lead to a more pronounced effect of local emissions than that observed during summers. These comparisons suggest that there could be significant distinction in the size distribution profiles observed at sites due to seasonal variation.

Figure 4 (a) shows data for AGO, one of the eastern most regional receptor sites of the study. Diurnal averages are shown for late fall (Nov), winter (Jan), spring (Mar) and summer (May, July and September). The morning peak in the plot can be explained by the morning commute (as this site is near a freeway). However, this morning peak subsides as the year progresses into warmer months when there is greater dispersion of fresh traffic emissions. Similar to UPL, during colder months, there is an evening and early nighttime rise in concentrations, leading to PNCs comparable to that in mornings. This peak diminishes in the summer and returns in September. Figure 4 (b & c) contrasts 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 irradiance and the mode diameter of the distribution decreases from ~30 nm at 11:00 to about 16-17 nm between 11:00-14:00. This decrease in mode diameter along with an increase in overall particle 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
371 particle numbers, occurs consistently through the months of September to December for particles >25
372 nm in late afternoon. For reasons similar to UPL, the effect of local emissions is more pronounced in
373 winter mornings at AGO, as evident in the morning peaks in sub 25 nm concentrations and the effect
374 of dilution as the day progresses can be seen in the inset in Figure 3 c.

375 The diurnal pattern in particle concentrations across these sites, (i.e., USC, AGO and UPL) is dominated
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 ### 3.3 Spatial Distribution of Particle Number Concentrations
384
385 Figures 5 (a & b) compare PNC at all sites for two months (during the warmer and cooler periods of the
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
388 month.

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

emissions and the arrival of advected PM from urban Los Angeles) that persist for a far longer period than the morning peak does. This is an important observation since it suggests that, in the receptor areas of the LAB, PM transported from central and west Los Angeles can contribute to higher and more sustained concentration levels even during the hours when local sources have minimum contributions. These results are also consistent with the findings of Zhu et al. (2006) and Hu et al. (2009) both conducted in the LAB. The highest of morning concentrations were observed at USC and RUB, the two 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 PNC during nighttime, when a stable stratification predominates the area. Though UPL (which is closer 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 of 2009 (counting particles down to 3 nm compared to 7 nm of the 3022, and thus reporting higher numbers) follows the diurnal pattern of inland sites.~~ The degree of variability based on PNC was examined using the Coefficient of Divergence, and the median value of COD is plotted for all site pairs ~~except SBR (since PNC for the latter are reported using a different instrument).~~ The highest CODs, or the maximum spatial variability, are observed during the hours of morning commute. The overall COD range was 0.17-0.28, indicating that PNC are only moderately heterogeneous.

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 comparable to morning traffic-related peak, as discussed earlier. Nighttime PNC become comparable 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 are not as pronounced as those in winter (December, Figure 5 a) as the primary emissions are quickly dispersed in summer and the higher ambient temperatures may be shifting the partitioning of semi-volatile organics emitted by primary sources to the gas phase (Miracolo et al., 2010). Particle number concentrations at all sites were generally lower in summer than in spring or winter. Post midday, there is a steady rise in PNC concentrations in all receptor sites, which is due to the combined effects of 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 regional-scale 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.

The hourly concentrations observed during this campaign varied across seasons, though the diurnal variations were more consistent. The maximum diurnal change in PNCs across seasons was observed at USC, along with the highest average concentrations. This was expected, as USC is located in the 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. (2006). In comparison to our earlier study (Moore at al., 2009), which reported concentrations comparable to USC at several sites in the Wilmington and West Long Beach area of Los Angeles, the receptor sites had lower concentrations due to lower impact of heavy traffic emissions in the immediate vicinity. During site selection, preference was given to sites not in the immediate vicinity of a source, to differentiate between local and regional contributions to the measured PNC in these sites. 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 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 somewhat lower, which could be interpreted (with some caution) as an encouraging outcome of the implementation of effective emission control technologies and the replacement of older heavy and light duty vehicles by newer vehicles in the LAB. The seasonal patterns identified in this study are 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
455 units), the values reported in this study are lower. This implies that in LAB, the *inter-community*
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
458 homogeneity at the inter-community level among receptor sites in LAB can be attributed to the effect
459 regional transport and meteorology that appear to override the contributions of local primary
460 emissions. The effects of local traffic sources were also observed at the sites in this study, but were
461 restricted to morning and (only during winter) evening commute hours.

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
465 composition. Wongphatarakul et al., 1998 showed that only moderately heterogeneous COD values
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
468 for different PM sizes. Overall CODs varied from 0.40-0.67, and exhibited a roughly inverse relationship
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
474 divergence in COD values observed for different site pairs. The data in Figure 7 reinforce the
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).

4. Conclusions

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 driven by a specific site pair, (site pair varied by hour and season), but the range of upper and lower quartile of COD vales was mostly within 0.1 units, implying that Particle Number Concentration (PNC) in these sites were homogeneous-to-moderately heterogeneous. Although, there were differences in the spatial variability through different seasons, the temporal patterns were consistent, and exhibited 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 variability in size distributions (reflection of the source composition) was higher than that of total 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 spatial variability based on particle size distributions support the notion of relative homogeneity in receptor areas in LAB, where concentrations are dominated by aged aerosols, advected eastwards from the source regions of urban Los Angeles, since the lowest variability was observed for particles in the size range of 40-100 nm, associated with long-range transport, compared to sub-30 nm particles associated with fresh emissions or new particle formation events. The largest differences in PNC were 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 spatial homogeneity, when phenomena that are regional in nature (i.e, summertime photochemical processes, long range transport, and higher degree of mixing) are active.

Even though our results suggest that PNC are moderately heterogeneous in the polluted receptor areas of the LAB, concerns related to population exposure assessment based on monitoring from a central station are still valid, especially in relation to urban areas impacted by a multitude of local and highly variable sources. Moreover, despite the moderate heterogeneity in total PNC at the inter-community level of receptor sites in LAB, particle size distributions may be significantly variable, resulting in 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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List of Tables and Figures

Tables

Figures

Table 1 a: Site information including the designation code, geographic co-ordinates, site and equipment elevations, sampling period and (CPC) data recovery[a]

Site ID	Latitude	Longitude	Site elevation (m)	Inlet elevation (m)	Distance from nearest Freeway (m), [Average Vehicles/day]	Sampling period	Data recovery (%)
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**

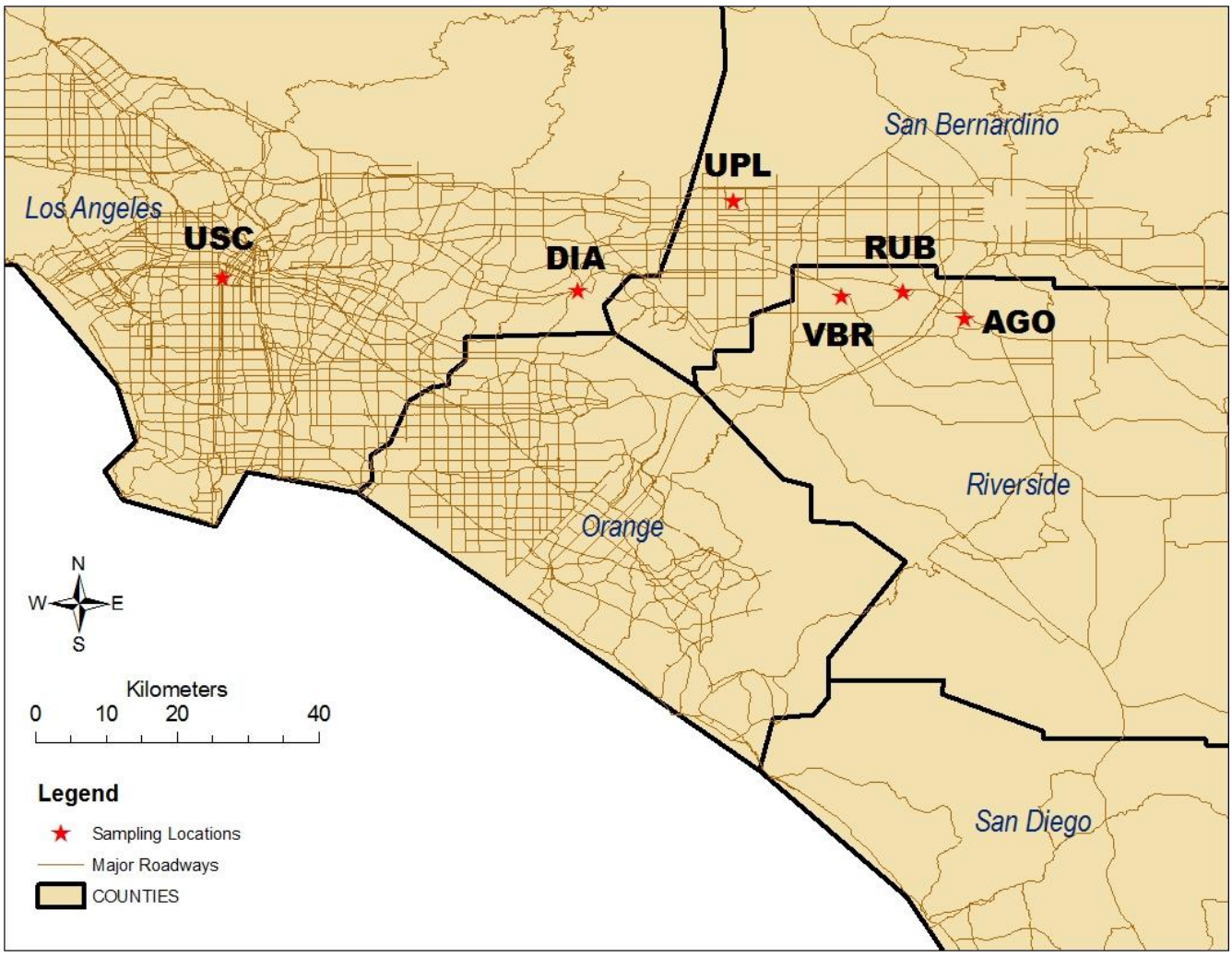
Table 2: Prevailing Wind Direction and Speed at sampling sites

Dominant Wind Direction and Wind Speed													
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
AGO	2008		2009										
WD (deg)	SW	E	E	E	W	W	W	W	W	W	W	W	E
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				2009									
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	2008		2009										
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)	NW	N	N	N	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	2008		2009										
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)	W	N	N	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	2008		2009										
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)	W	W	N	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 3: Temperature (^{O}C) and Relative Humidity (%) at sites during sampling period

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

Sites	UPL		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



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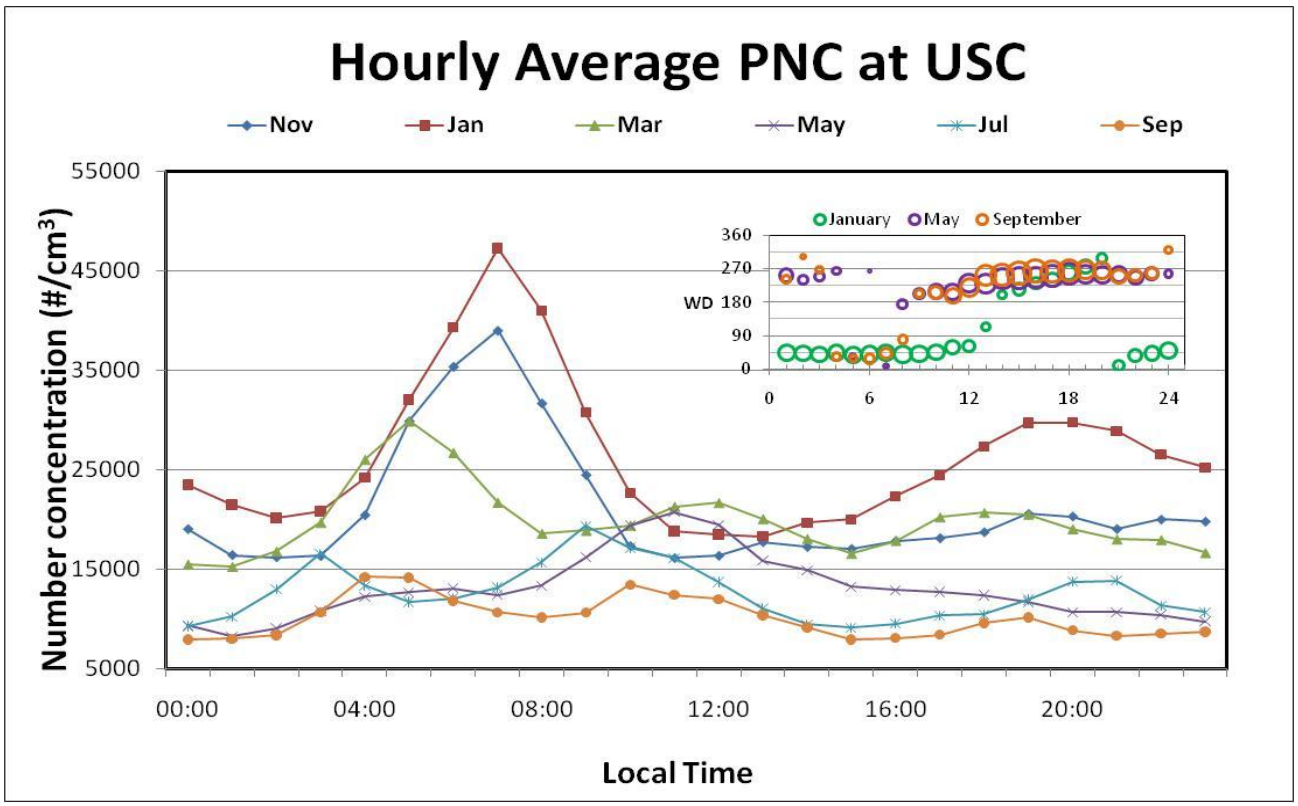
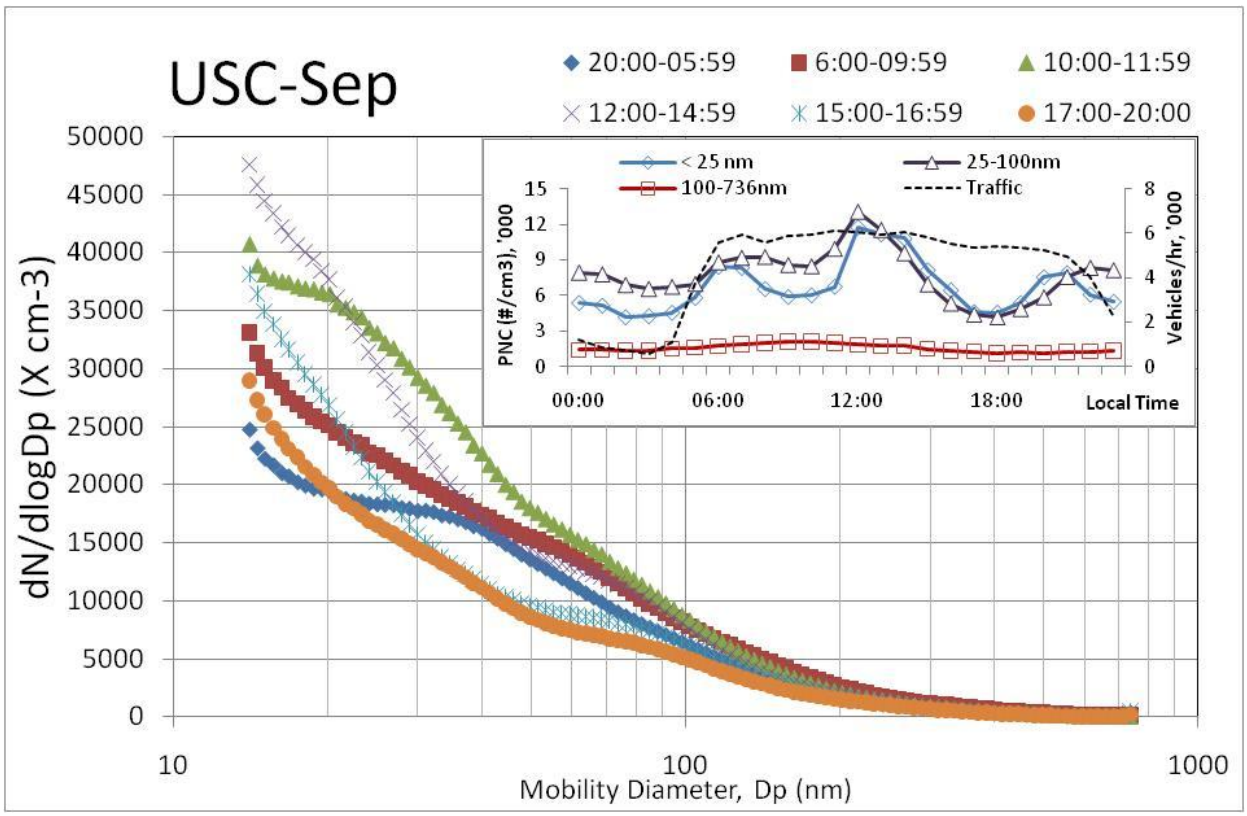
791 **Figure 1: Location of sampling sites in Los Angeles air basin.**



792

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



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798 **Figure 2b: Average Size Distribution of Particles during six time periods (local time) of the day at USC during September**
799 **2009.**

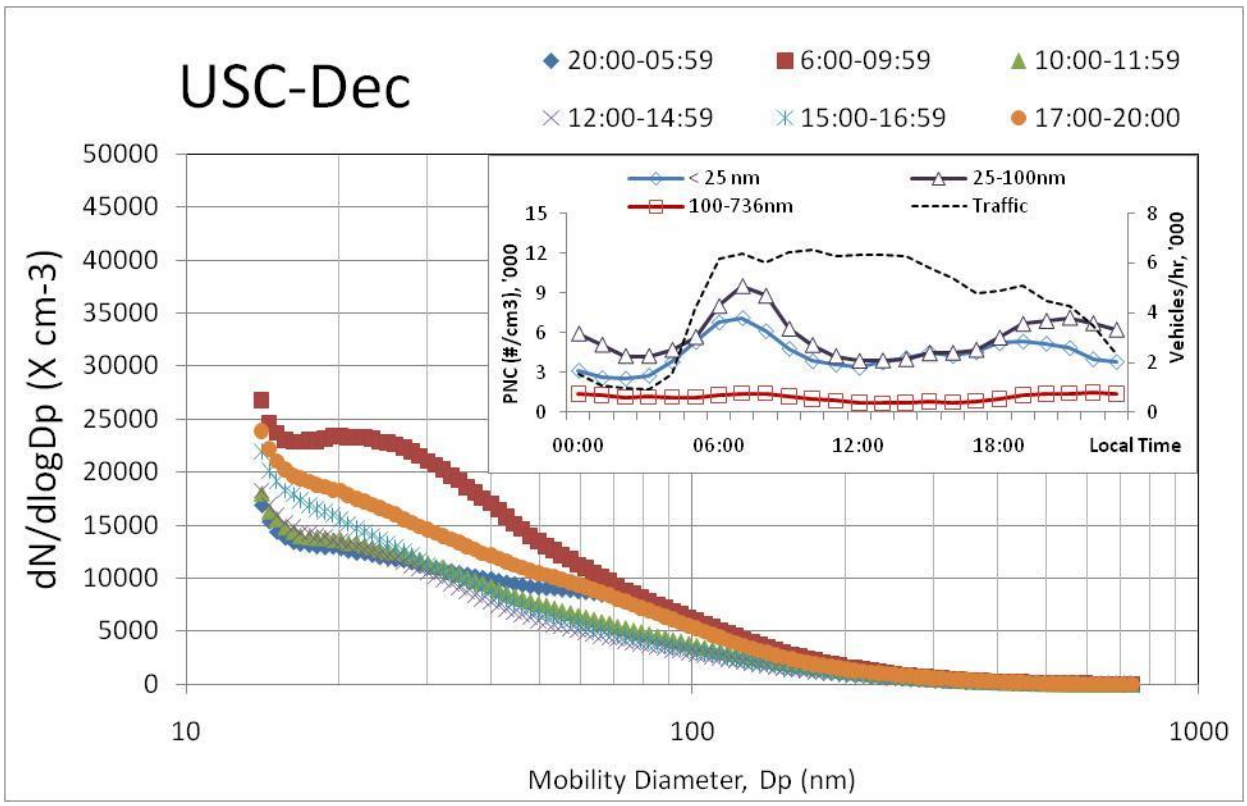
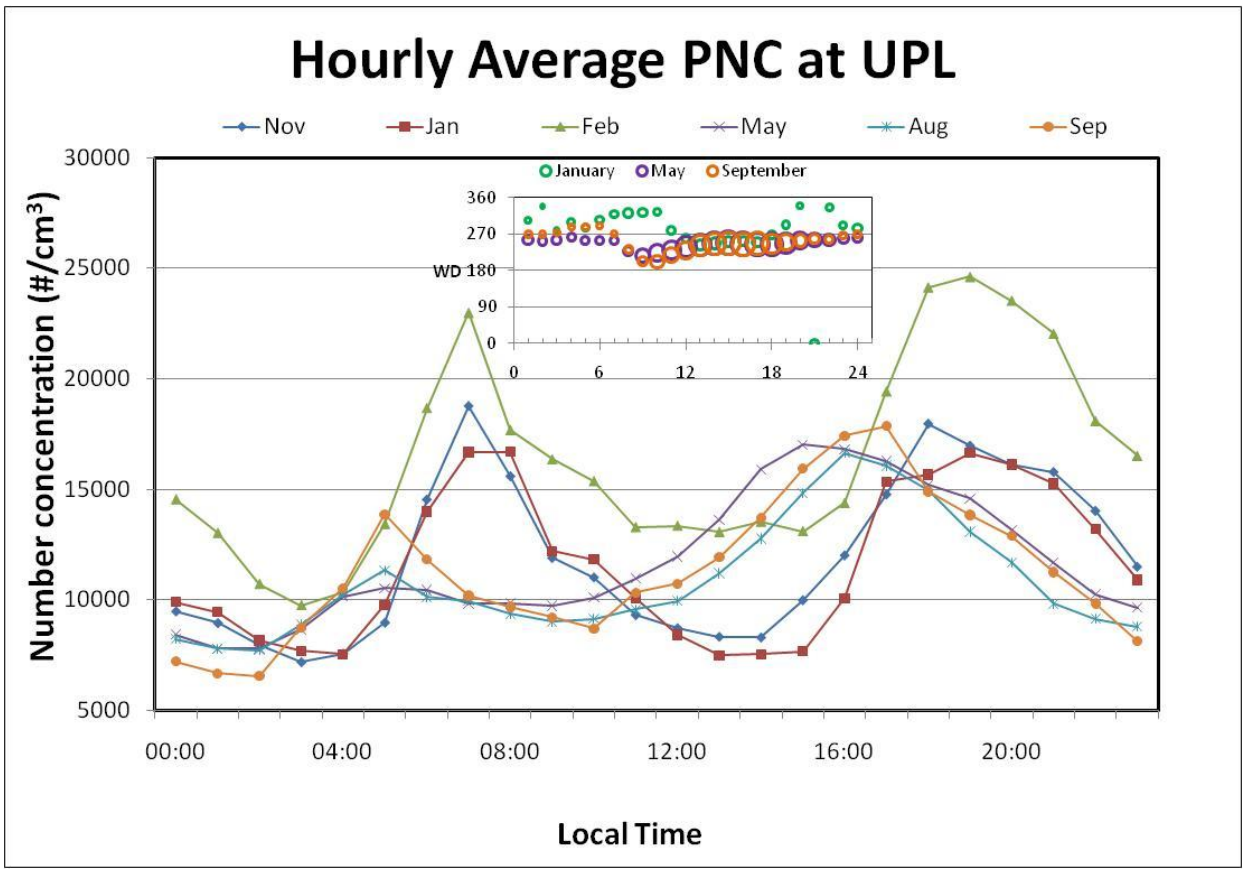


Figure 2c: Average Size Distribution of Particles during six time periods (local time) of the day at USC during December 2009.



804

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

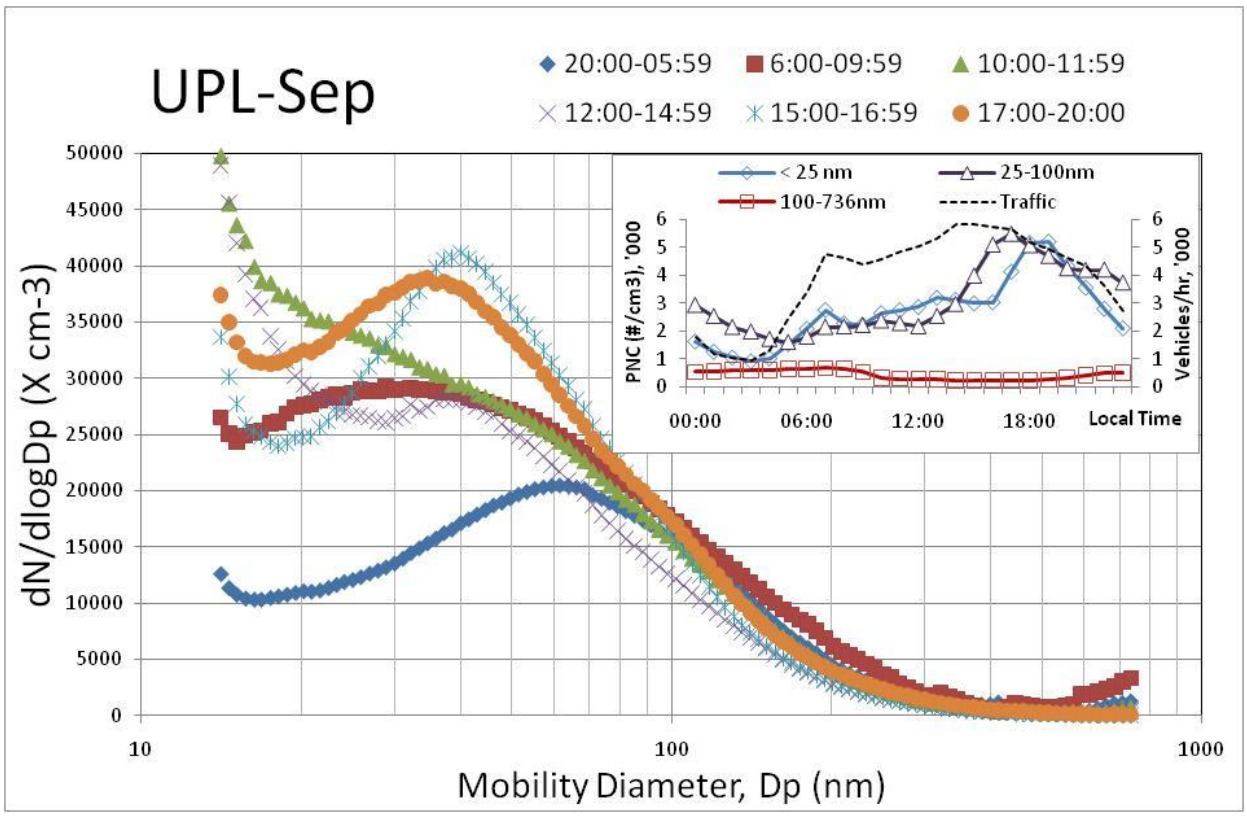


Figure 3b: Average Size Distribution of Particles during six time periods (local time) of the day at UPL during September 2009.

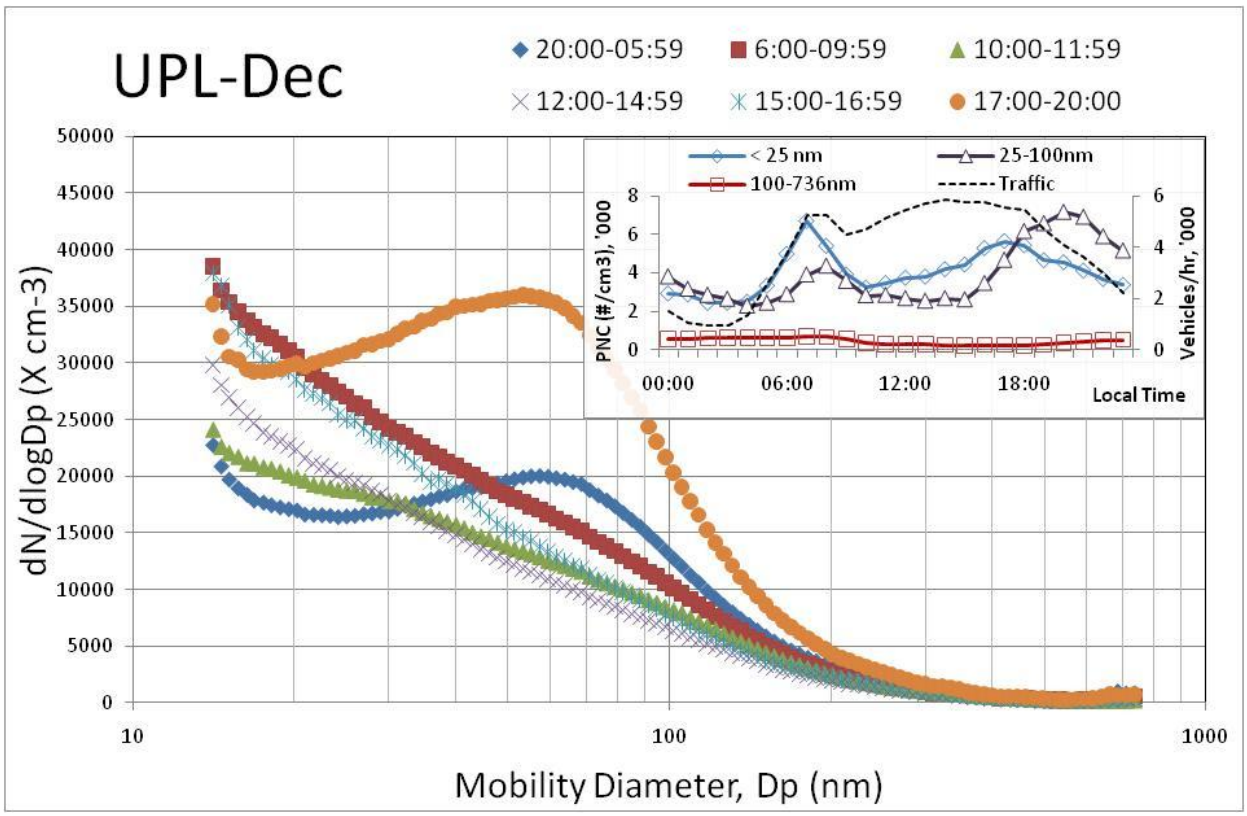
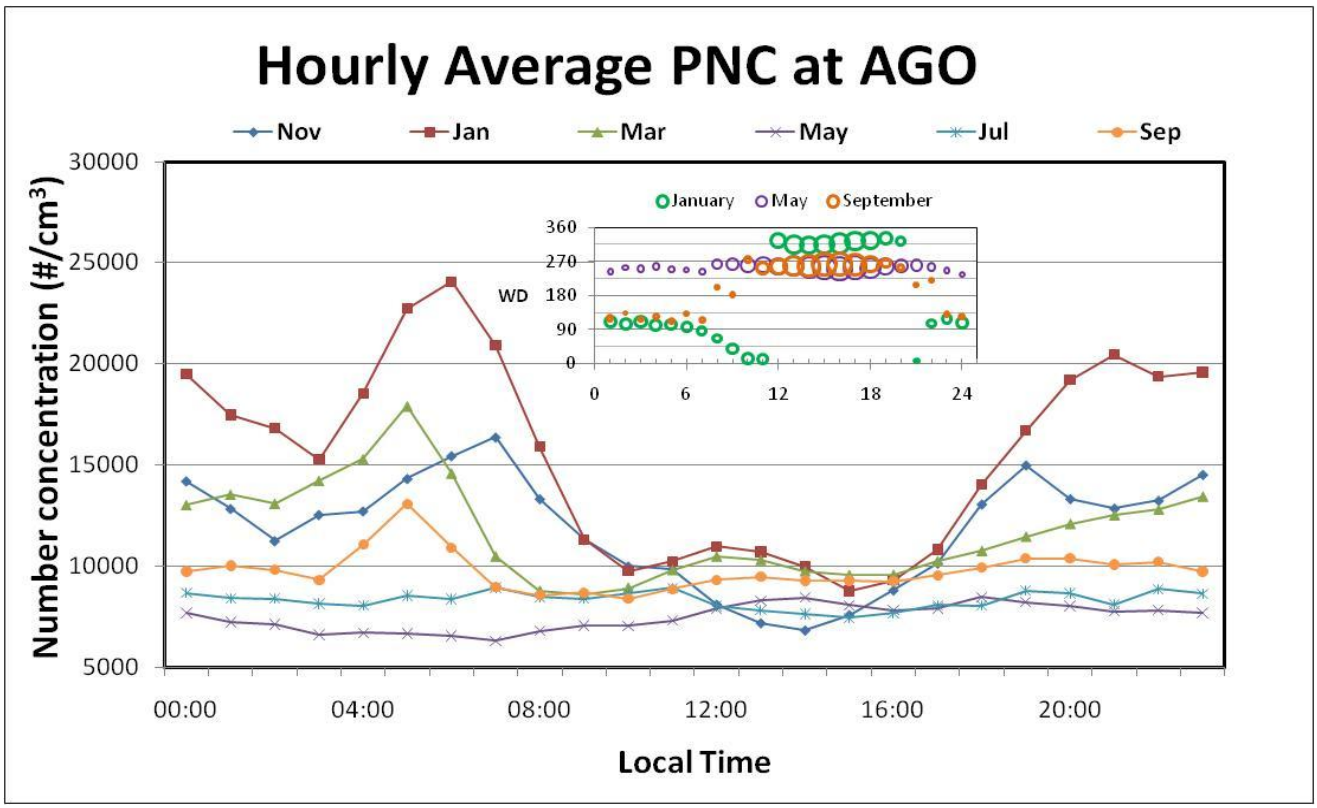


Figure 3c: Average Size Distribution of Particles during six time periods (local time) of the day at UPL during December 2009.



817

818 **Figure 4a: Hourly average particle number concentration at AGO for hours of the day in local time. The relative standard**
819 **error for the hourly averages reported above was less than 3%. The inset is a plot of vector averaged wind direction (WD)**
820 **with the bubble area weighed to wind speed plotted for hours of the day in local time.**

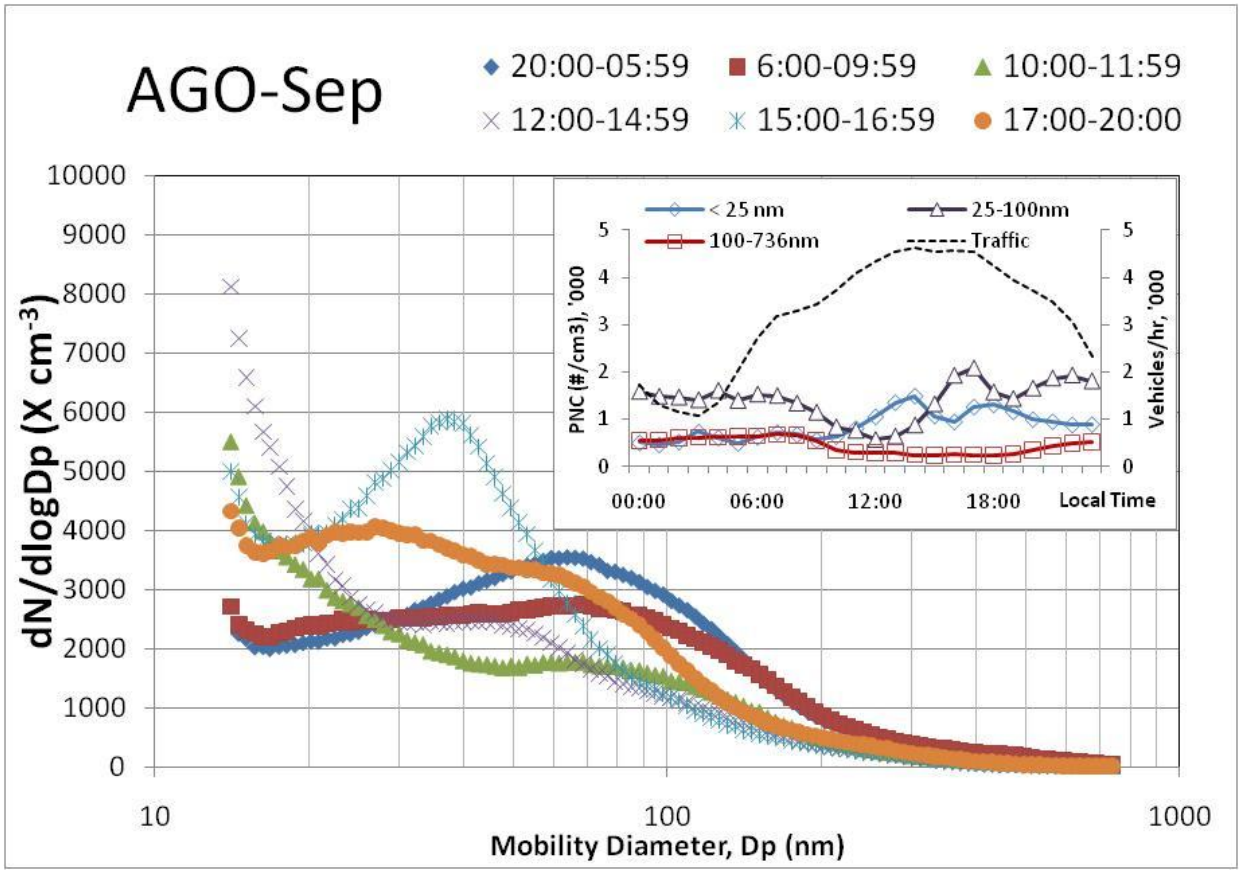


Figure 4b: Average Size Distribution of Particles during six time periods (local time) of the day at AGO during September 2009.

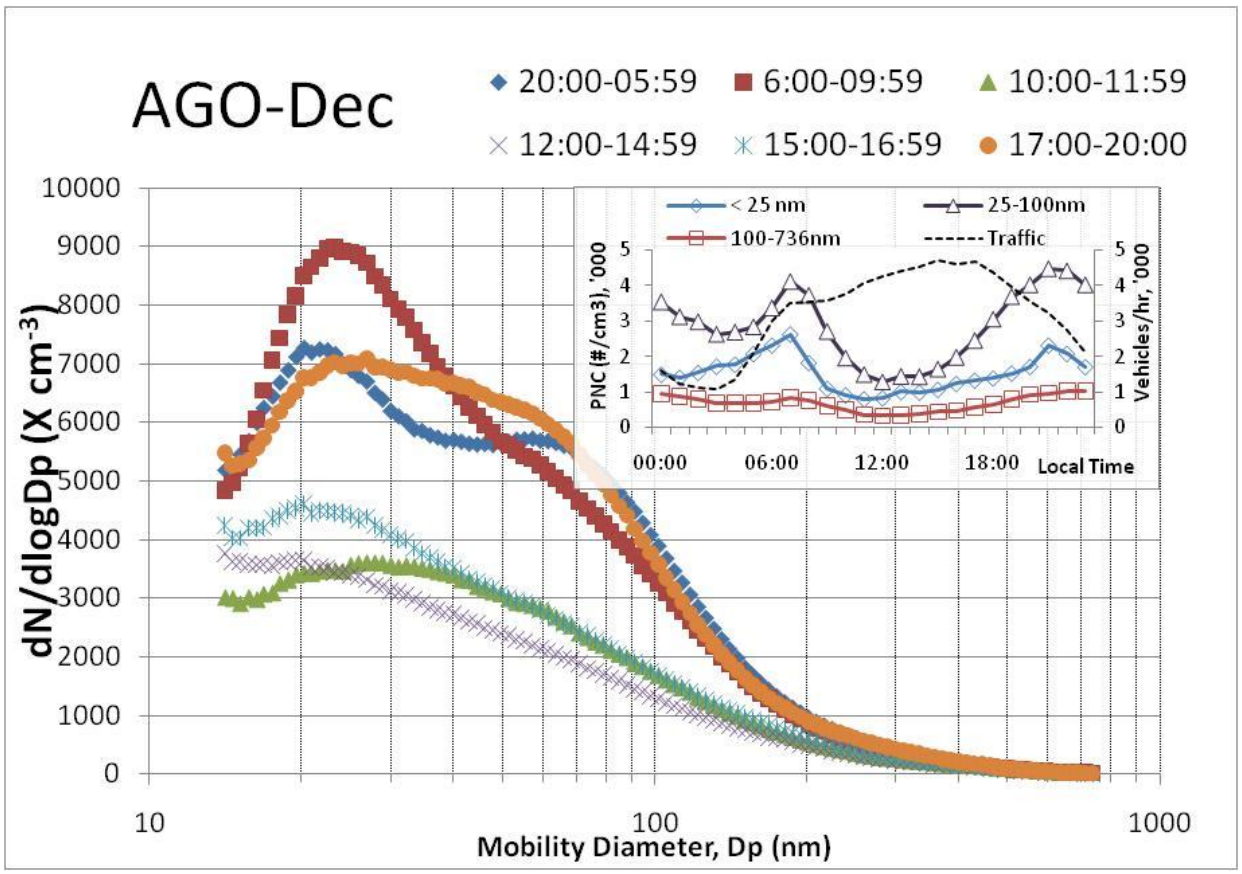
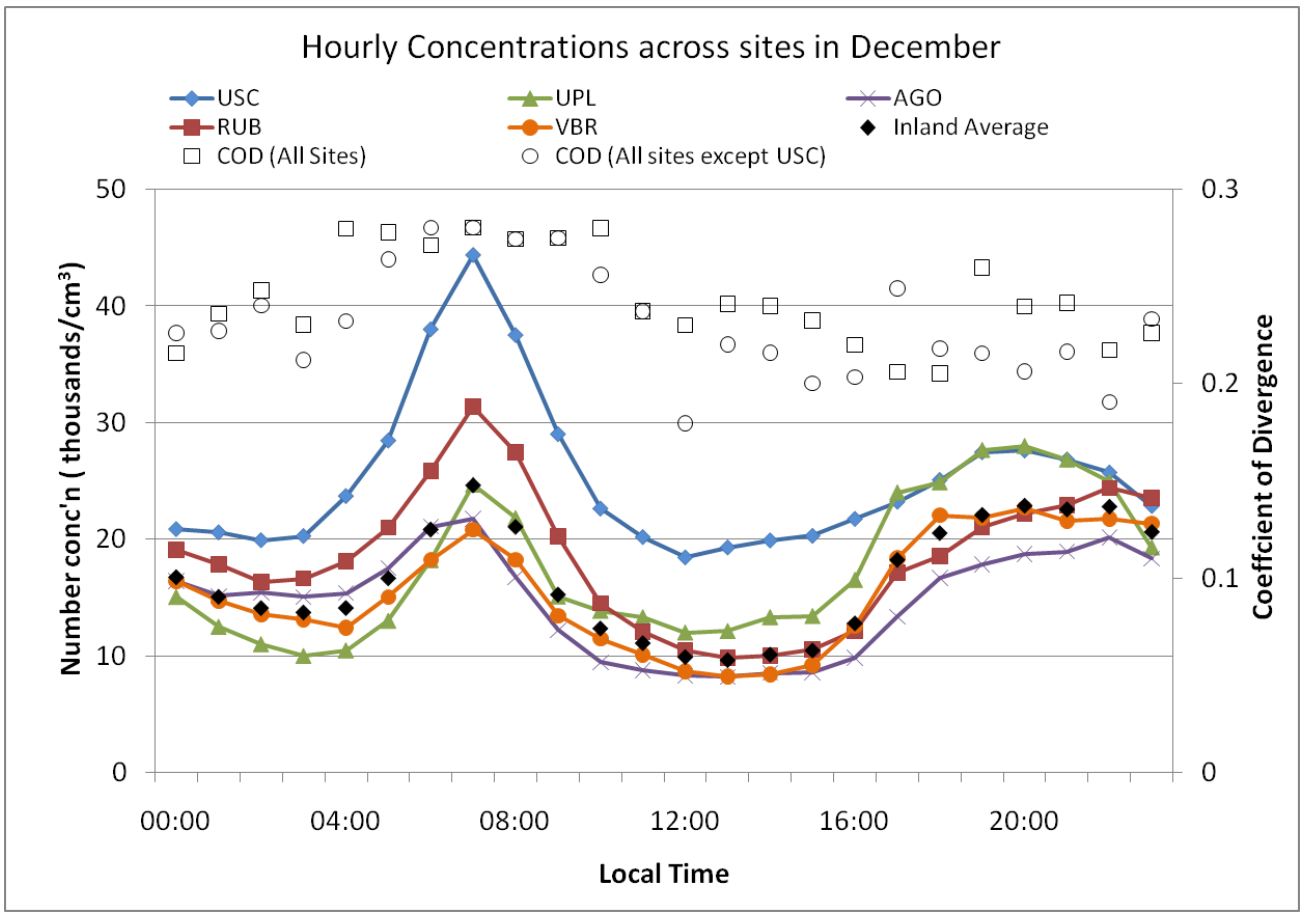
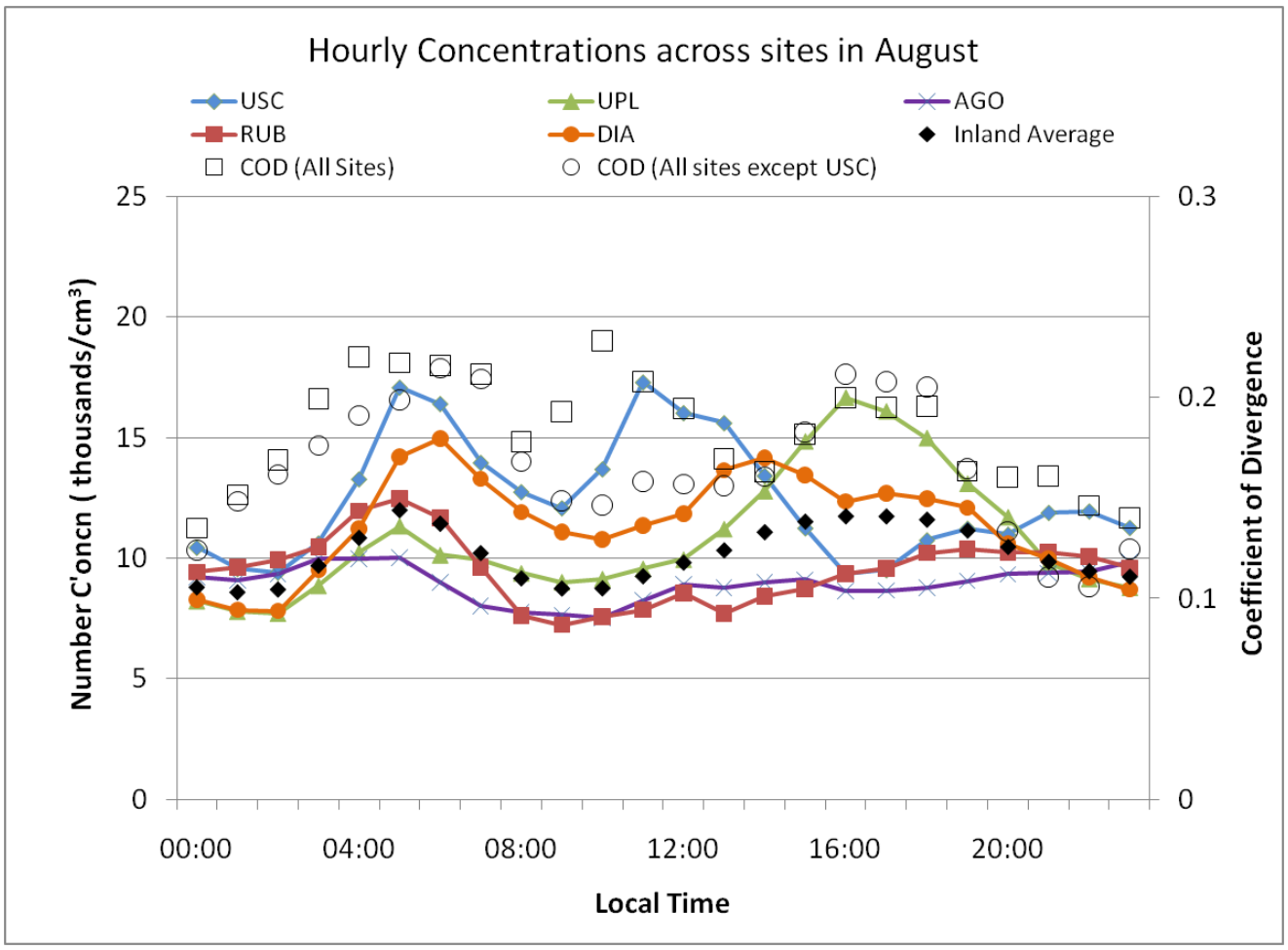


Figure 4c: Average Size Distribution of Particles during six time periods (local time) of the day at AGO during December 2009.



829

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



831

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

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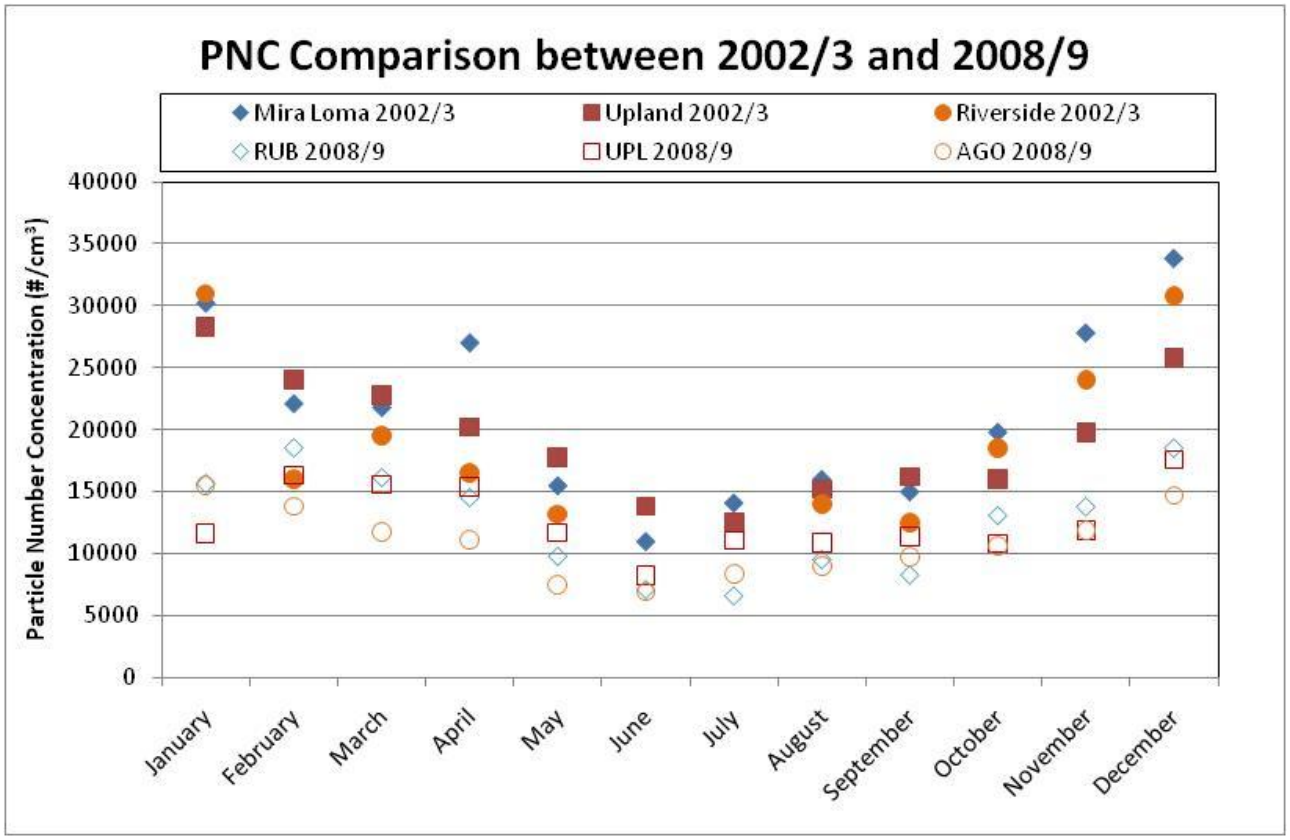
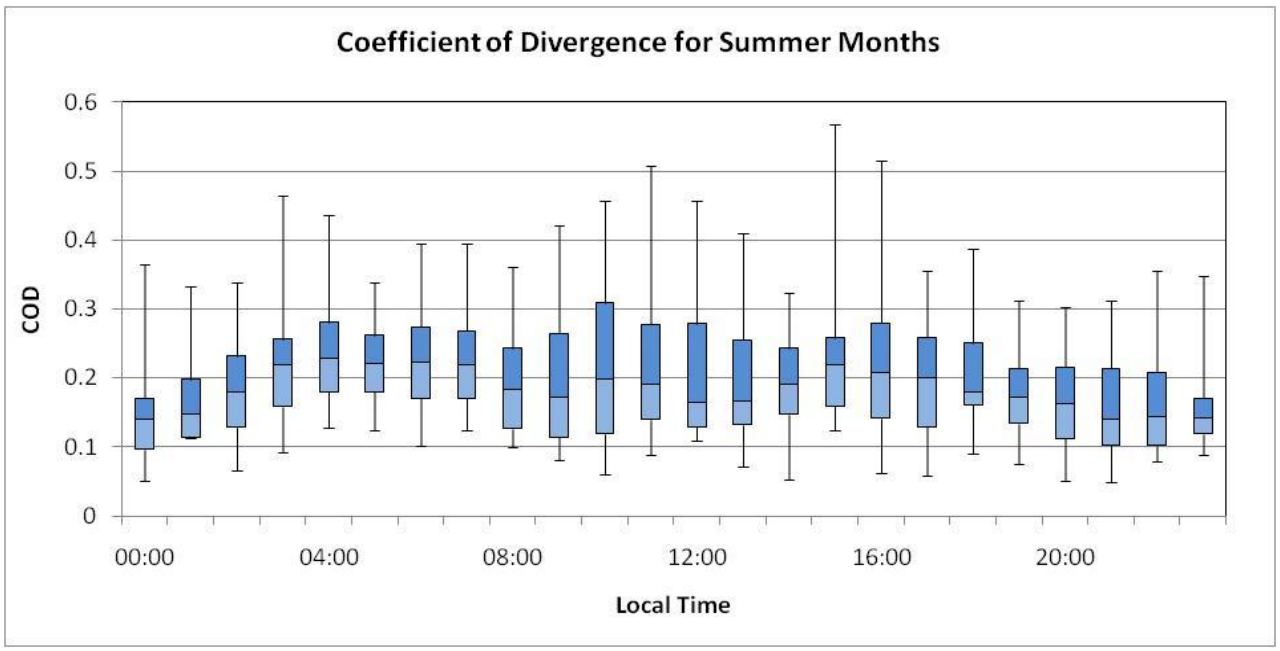


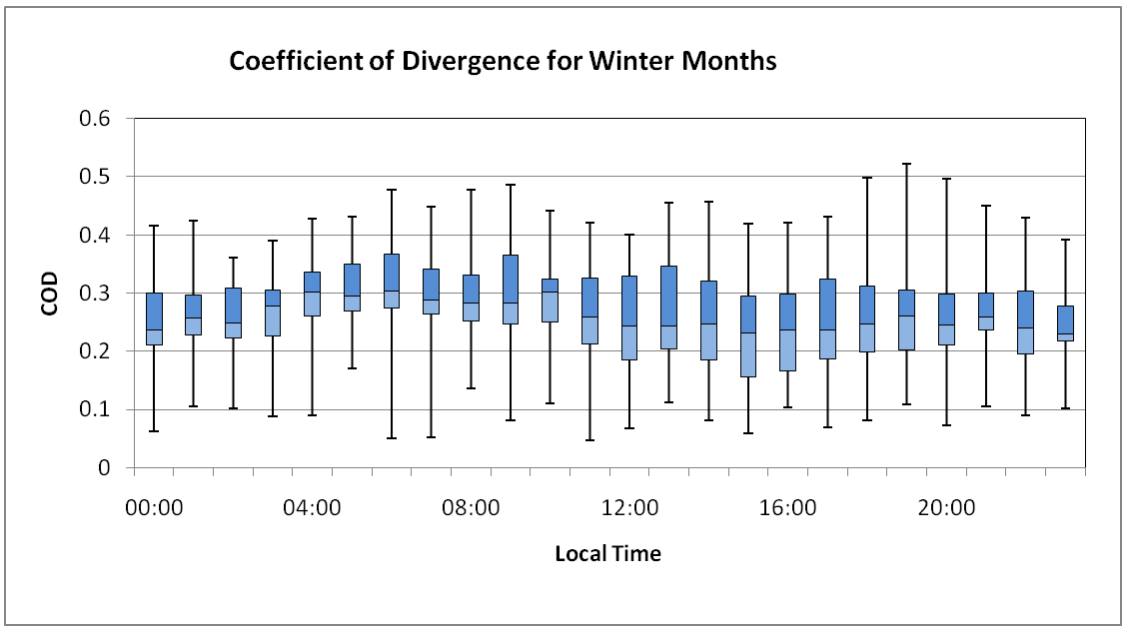
835

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



837

838 **Figure 7a: Coefficients of divergence during the summer months of May-Aug, 2009.**



839

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