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

N. Hudda, K. Cheung, K. F. Moore, and C. Sioutas

University of Southern California, Sonny Astani Department of Civil and Environmental Engineering, 3620 S Vermont Ave., Los Angeles, CA, 90089, USA

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Correspondence to: C. Sioutas (sioutas@usc.edu)

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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 seven sites, one in downtown Los Angeles and six 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 predominated. 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 above 40–50 nm, 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.

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1 Introduction

Numerous recent epidemiological and toxicological studies investigating associations between particulate pollution and health effects have attributed greater risk to ultrafine particles (UFP, dp less than ~100 nm) (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 $PM_{2.5}$ and PM_{10}, and $PM_{2.5}$ concentrations are often used as a surrogate for UFP concentrations (Wilson et al., 2006), although poor correlation has been reported between PNC (dominated by UFP) and $PM_{2.5}$ (dominated by accumulation mode particles) (Sardar et al., 2004). Not only is $PM_{2.5}$ 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 depth, 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 (Jerrett et al., 2005) over large spatial scales, but recent works of Kim et al. (2002) Pinto et al. (2004), Zhu

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et al. (2002), Krudysz et al. (2009), and Moore et al. (2009) all suggest that UFP vary spatially at local as well as regional scales.

In urban areas, a dominant source of UFP is primary emissions from vehicular sources (Shi et al., 1999; 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. (2002a, 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; Zhi 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 intercommunity variability can be assessed. It has been suggested (Turner and Allen, 2008)

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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 six 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 Fig. 1. Highways and major arterials, common sources of ultrafine particles, are identified in Fig. 1. The distances to freeways are also tabulated in Table 1. The sites were within 60 km of each other in the E-W direction and 20 km in the N-S direction. Sampling sites were generally located in areas where there were no known major contributors to UFP, except for local traffic (e.g., residential neighborhoods).

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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 150 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" 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" sites which are downwind in the eastern region of the basin towards which the meterology 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).

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. The site is also influenced by the aged aerosol and is located close the base of San Gabriel Mountains. 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. It is 3 km (south) from the nearest freeway and the major roadways next to the site have moderate 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

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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.

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.

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 min 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 aerosol were copper tubes of 1 cm diameter. Meteorological data, i.e., temperature, relative humidity, wind speed and direction amongst other parameters was collected using Vantage Pro

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2 Weather Stations (Davis Instruments, Hayward, CA). All 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-min particle number concentration, 5-min 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 PST 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. The data from site VBR is not reported after April 2009 as the reported concentrations were unreasonable due to CPC malfunction. The data at SBR was collected from May 2009–December 2009, except for August 2009 when a malfunction occurred. CPCs were operated side-by-side at USC for a 24-h 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 h and each CPCs concentrations were 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

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against a factory-calibrated CPC. The data were corrected 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).

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

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

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

3.1 Meteorology

Meteorological conditions can influence ultrafine particle concentrations significantly, but the Los Angeles area exhibits relatively limited diurnal and seasonal variation, as

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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 Fig. 1a, 2a, 3a 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., 1995). The particle number concentrations and the size distributions will be discussed in this context. Tables 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, 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 (December–February), 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.

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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 erro, 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 2a 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 a.m. Advancing into summer months, this peak is not as robust and eventually disappears, 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. During the cooler months of the year, another peak is observed in the evenings and early night, possibly related to particle formation by condensation of semivolatile vapors emitted by traffic during preceding hours. A shallower mixing layer in this season and time period also leads to these elevated concentrations and its effect is most pronounced in peak winter months (December–February) when night time concentrations can reach ca. 30 000/cm^3. Biswas et al. (2007) have previously reported a similar data pattern. The

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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 very 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.

Figure 2b and c compares the average size distribution of particles 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 h) is very robust in September which weakens progressively through the fall and into December. The tri-modal diurnal profile observed at USC during warmer months in Fig. 1a is limited to sub-50 nm particles, while the seasonal variation of the diurnal patterns for particles >100 nm is not significant. This is a distinctly different pattern than 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 3a shows monthly average diurnal particle number data across six months of the year at UPL (i.e. November, January, February, May, August and September). A bi-modal diurnal distribution is observed at this site, with a morning time peak, similar to USC, corresponding to morning commute during 06:00–10:00 h in winter months that is not as robust during summer. This winter peak is a compounded effect of greater vehicular emissions and lower mixing height in winter mornings. 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

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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.

Figure 3b and c compares 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. A possible explanation is that the contribution of photochemical activity to 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 a distance away from major freeways, the concentrations of gaseous and semi-volatile organic vapor precursors that participate in secondary particle formation are smaller compared to those at USC (or in general in central LAB), which may decrease the degree of PM formation via that pathway. The results plotted in Fig. 2b shows that during 15:00–17:00 h when the highest wind speeds of the day are observed, the concentrations for particles in the range of 25–100 nm increase (while 14–25 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 of could be due to the arrival of the polluted air mass from Los Angeles. Further, during other hours of the day during summer months the concentrations within this size range remained stable. Similar observations have been made by Kim et al. (2002) and Fine et al. (2004). However, during winters (Fig. 2c) 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 associated with fresh emissions 14–25 nm shows a sharp increase during

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evening (total PNC increases and the mode particle diameter decreases, shifting the distribution towards what can be characterized as fresh local emission sources), which may be due to the combined effects of local traffic, coupled with the decreasing temperature and mixing depth with increasing relative humidity. These comparisons suggest that there could be significant distinction in the size distribution profiles observed at sites due to seasonal variation.

Figure 4a shows data for AGO, one of the eastern most sites of the study. Diurnal averages are shown for late fall (November), winter (January), spring (March) 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. 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 4b and c contrasts the size distributions during different time periods of the day. During September we observed a rise in the concentrations of smaller particles (<25 nm) 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 to 14:00. This decrease in mode diameter along with an increase in overall particle numbers, along with the timing, indicates the possibility of new particle formation. Further, this increase is not observed in December, and the peak declines steadily through the fall. Similar observations in that area have been made previously by Fine et al. in 2004. An increase in mode diameter along with particle numbers occurs consistently through the months September to December for particles >25 nm in late afternoon.

The diurnal pattern in particle concentrations across these sites, (i.e., USC, AGO and UPL) is dominated by a bi-modal distribution, except for summers at USC. But there is a decrease in overall particle concentrations due to dispersion of the air parcels moving inland (eastwards). The flat peak in nighttime concentrations (at hours when there are limited fresh emissions) at AGO (further inland) are lower than at UPL. PNCs

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at the RUB and VBR sites, which are further inland than UPL, also are lower than at UPL, but higher than at AGO, which is further east of these sites. A similar pattern is observed in the morning peaks corresponding to commute hours, because the traffic volume decreases as one moves farther inland from Downtown Los Angeles.

3.3 Spatial distribution of particle number concentrations

Figure 5a, b compares all sites across two months to contrast spatial variation in the concentrations across the basin. A representative month from each season was chosen and data have been plotted as the diurnal averages over the span of the month.

Figure 5a shows a winter month data across sites. The all-hour average December 2008 temperatures across the inland sites ranged from 10.7 to 12.7 degrees Celsius while the relative humidity ranged from 59 to 67%. The wind data in Table 2 shows the predominant wind direction based on hourly vector averages for different sites. At all inland sites, the morning peak concentrations 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 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

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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 5b 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, Fig. 5a) 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; Pinto et al., 2005). 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. This is discussed further in the following section.

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

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(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 5 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).

Figure 7a and b compares 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 (which in most cases would be traffic) is increasing the COD. This was further ascertained by inspecting individual site pair values. During both summer and winter, homogeneity is observed in late night and early morning concentrations, indicating the presence of a regional aerosol. In comparison to our previous study (Moore et al., 2009; Krudysz et al., 2009) that reports median COD values of about 0.3–0.5 in source regions of

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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 variability in PNC is lower than the intra-community variability of areas like the LA harbor, impacted by 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 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 restricted to morning and (only during winter) evening commute hours.

The spatial complexity of the PNC was further resolved with the size distribution data. Synergistic effects of multiple factors can lead to similar particle number concentrations at two sites; however, the 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 can be observed for chemical composition of particles even when the sources are different. Since 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 exhibit a rough inverse relationship with particle size. This can in part be accounted for by the difference in sources between USC and the inland sites. This observation is supported by the argument that the average COD values (range) was 0.35 (0.34–0.36) for AGO-UPL, 0.55 (0.53–0.57) for USC-AGO. Even though the degree of 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 Fig. 7 reinforce the observation that sites appear to be more homogeneous when the local sources (which contribute to the smaller size spectrum of the particle size distribution more than the bigger size) are not dominant. Similar observations were made by Turner et al. (2002).

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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. 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

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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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Table 1. 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	150, [112 000]	17 Nov 2008–21 Dec 2009	91%
DIA	34°0′1″ N	117°49′54″ W	223	2	200, [99 000]	25 Feb 2009–21 Dec 2009	96%
UPL	34°6′14″ N	117°37′45″ W	386	1.85	2000, [96 000]	17 Nov 2008–21 Dec 2009	90%
VBR	33°59′45″ N	117°29′31″ W	220	1.9	3000, [85 000]	17 Nov 2008–30 Apr 2009	95%
RUB	33°59′58″ N	117°24′58″ W	248	2	200, [72 000]	17 Nov 2008–21 Dec 2009	93%
AGO	33°57′41″ N	117°20′0″ W	323	2.1	750, [81 000]	17 Nov 2008–21 Dec 2009	98%
SBR	34°6′24″ N	117°16′27″ W	317	1.8	2000, [65 000]	18 May–30 Jun, 15 Jul–30 Jul, 4 Sep–17 Dec 2009	87%

[a] The SMSP were operated at sites USC, UPL and AGO from 4 Sep 2009–21 Dec 2009 at greater than 90% data recovery.

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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
SBR	2008						2009						
Month	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov
WD (deg)	NE	NE	NE	SE	SW	W	W	W	W	W	W	W	NE
WS (m/s)	0.47	0.56	0.94	0.63	1.09	1.38	1.50	1.47	1.40	1.26	1.08	1.12	0.66
SD (m/s)	0.64	0.78	1.29	0.80	1.22	1.45	1.22	1.14	1.27	1.24	1.21	1.33	0.89
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

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Table 3. Temperature (˚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	12±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±2	312.1±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
Apr 09	55±21	16.0±6.3	58±21	58.2±21.0	54±20	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
Jun 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
Jul 09	52±18	25.9±6.0	58±18	58.4±18.4	52±19	28±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±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	69±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	44.3±29	16.3±6.7	73±15	14.0±3.4
Feb 09	64±24	11.9±5.6	68±25	12.1±5.7	83±8	12.2±1.8
Mar 09	67±18	13.3±5.3	64±22	14.0±5.4	72±19	13.6±3.1
Apr 09	59±22	16.0±6.6	59±21	16.0±6.3	68±14	14.6±4.0
May 09	68±17	19.4±5.4	68±17	19.9±5.1	78±7.7	17.5±1.7
Jun 09	67±16	20.2±4.8	66±17	21.0±5.1	76±7.4	17.7±1.1
Jul 09	60±20	24.2±5.5	61±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±3.6			58±20	13.8±3.8

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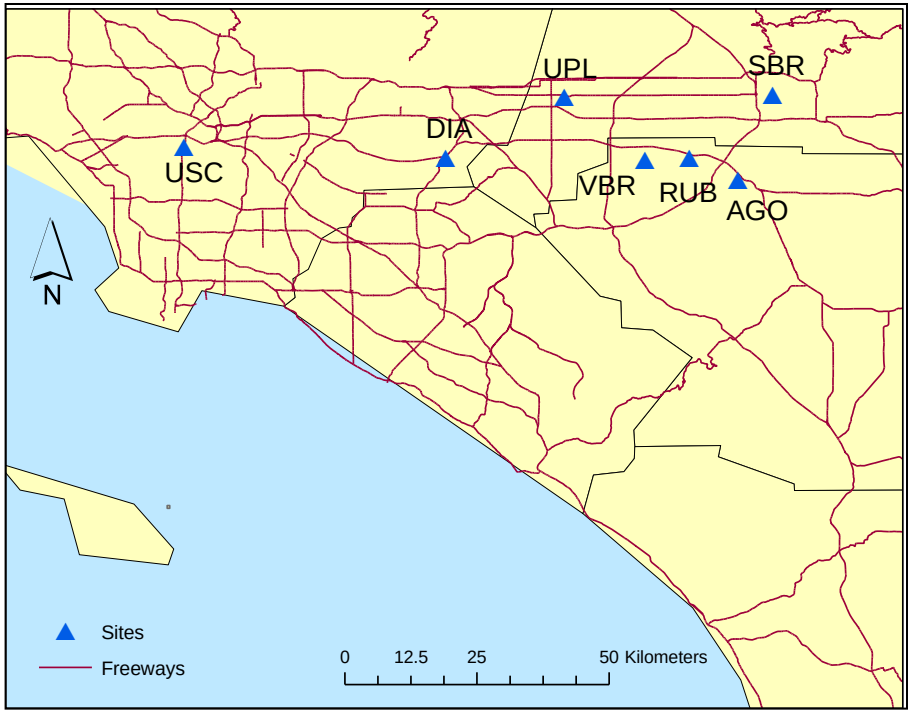

Fig. 1. Location of sampling sites in Los Angeles air basin.

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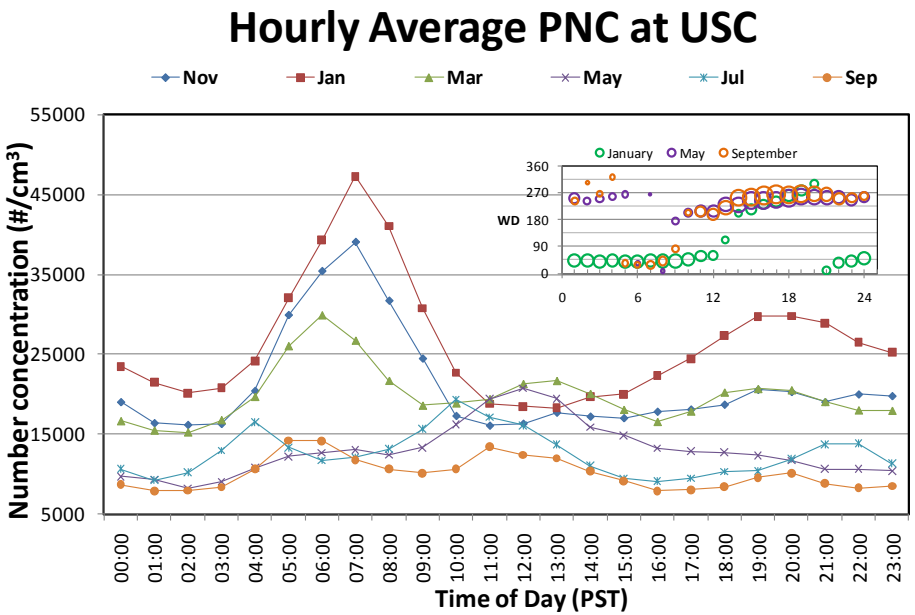

Fig. 2a. Hourly average particle number concentration at USC plotted for hours of the day in Pacific Standard Time (PST). The relative standard error for the hourly averages reported above was less than 2%. The inset is a plot of vector averaged wind direction (WD) with the bubble area weighed to wind speed plotted for hours of the day in PST.

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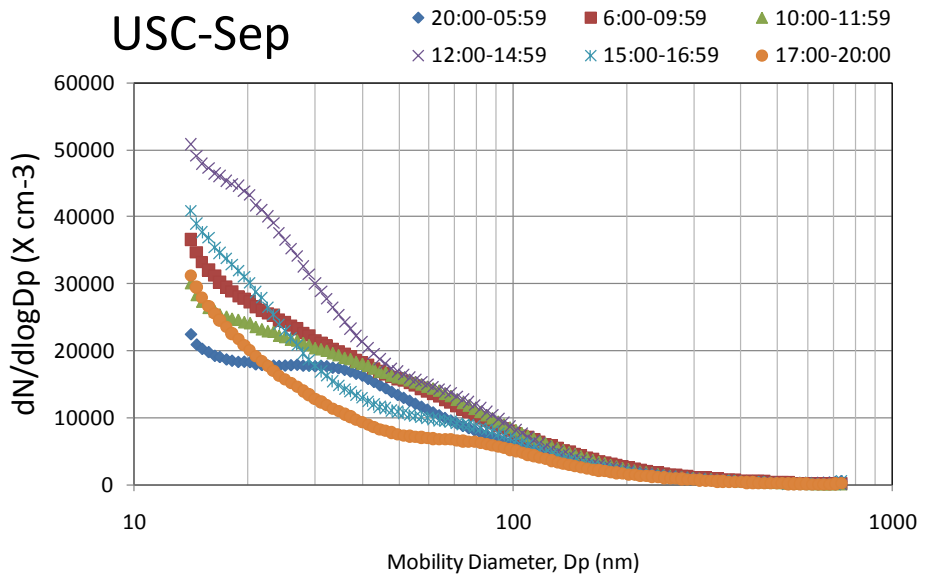

Fig. 2b. Average size distribution of particles during six time periods (PST) of the day at USC during September 2009.

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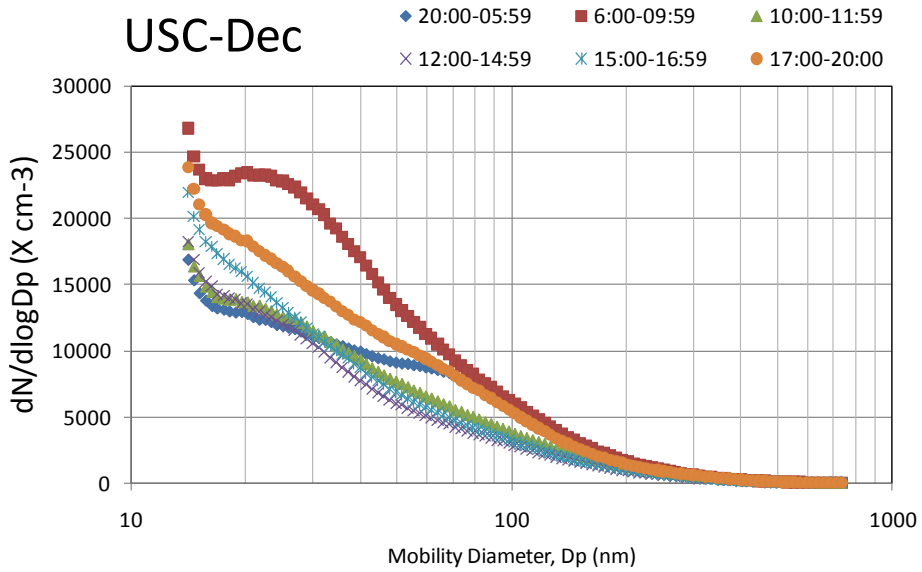

Fig. 2c. Average size distribution of particles during six time periods (PST) of the day at USC during December 2009.

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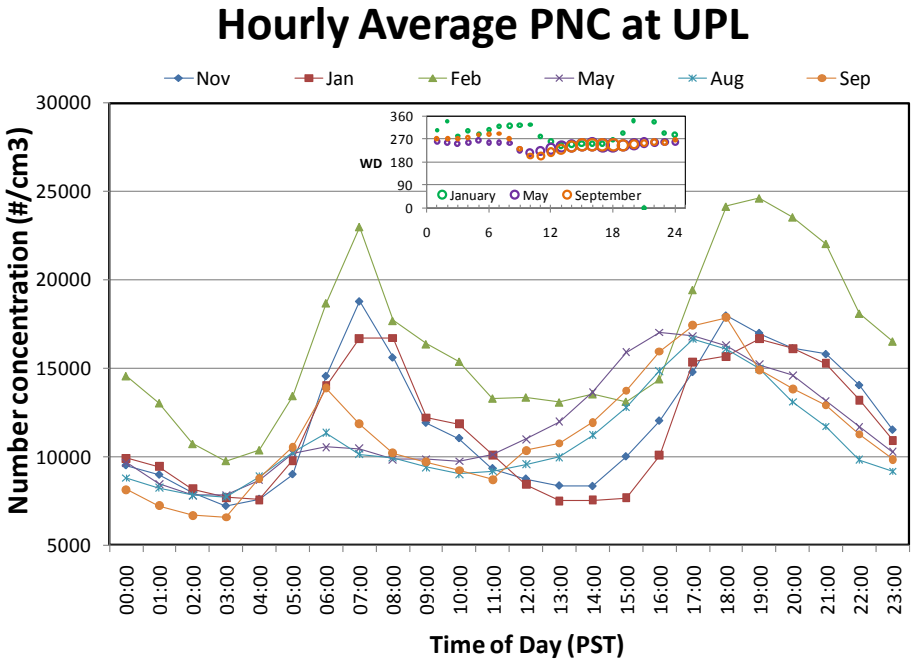

Fig. 3a. Hourly average particle number concentration at UPL for hours of the day in Pacific Standard Time (PST). The relative standard error for the hourly averages reported above was less than 2%. The inset is a plot of vector averaged wind direction (WD) with the bubble area weighed to wind speed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

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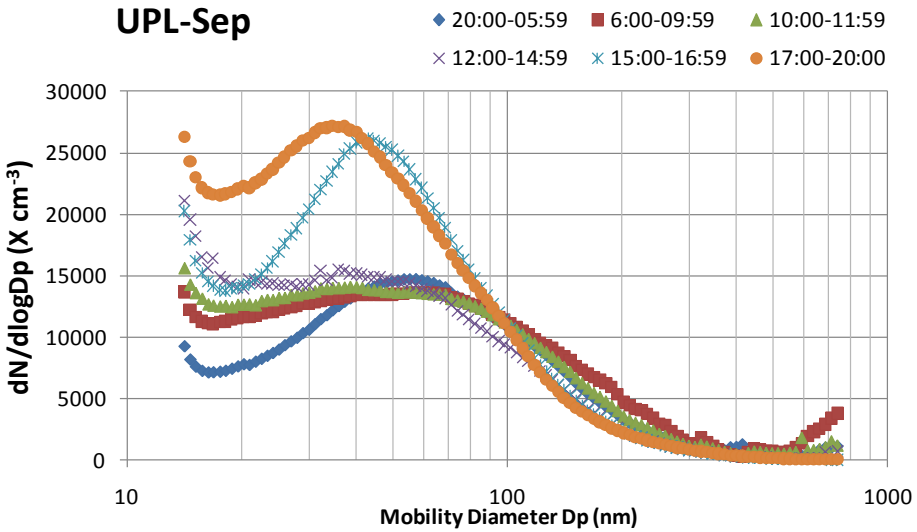

Fig. 3b. Average size distribution of particles during six time periods (PST) of the day at UPL during September 2009.

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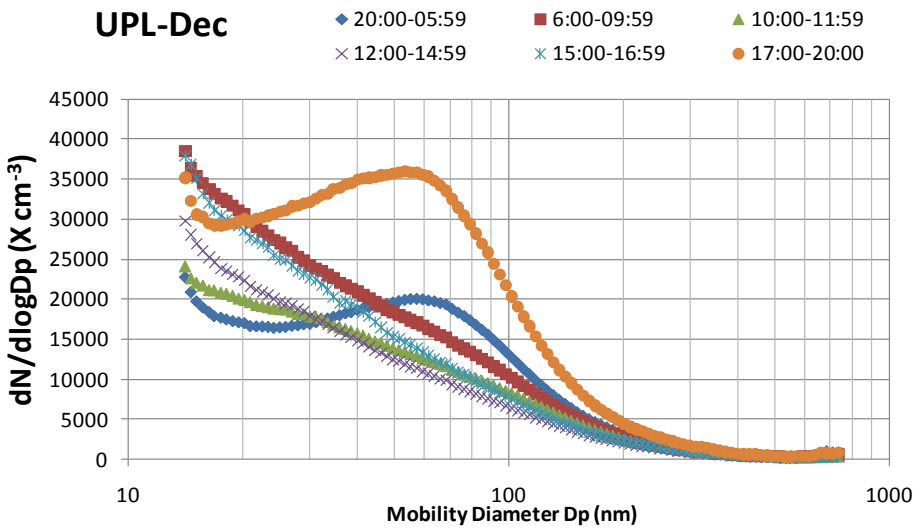

Fig. 3c. Average size distribution of particles during six time periods (PST) of the day at UPL during December 2009.

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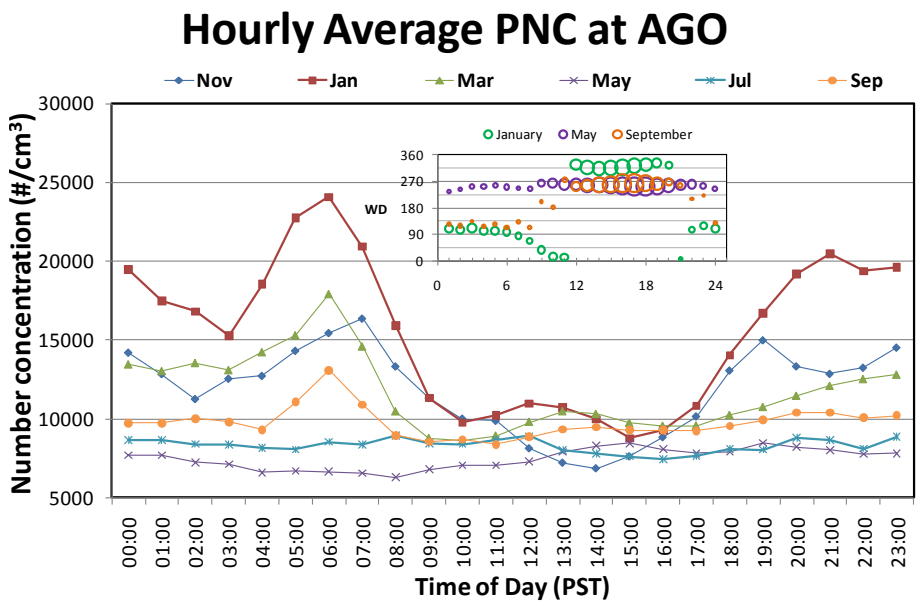

Fig. 4a. Hourly average particle number concentration at AGO for hours of the day in Pacific Standard Time (PST). The relative standard error for the hourly averages reported above was less than 3%. The inset is a plot of vector averaged wind direction (WD) with the bubble area weighed to wind speed plotted for hours of the day in PST.

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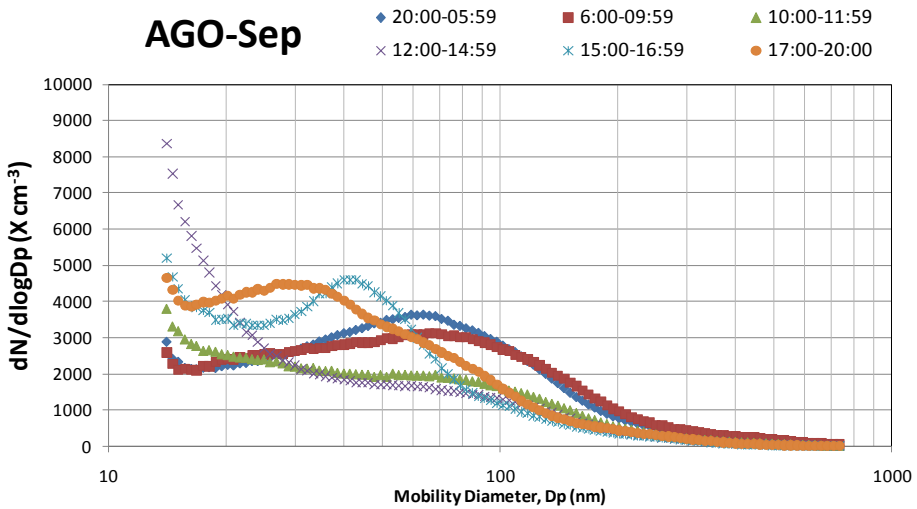

Fig. 4b. Average size distribution of particles during six time periods (PST) of the day at AGO during September 2009.

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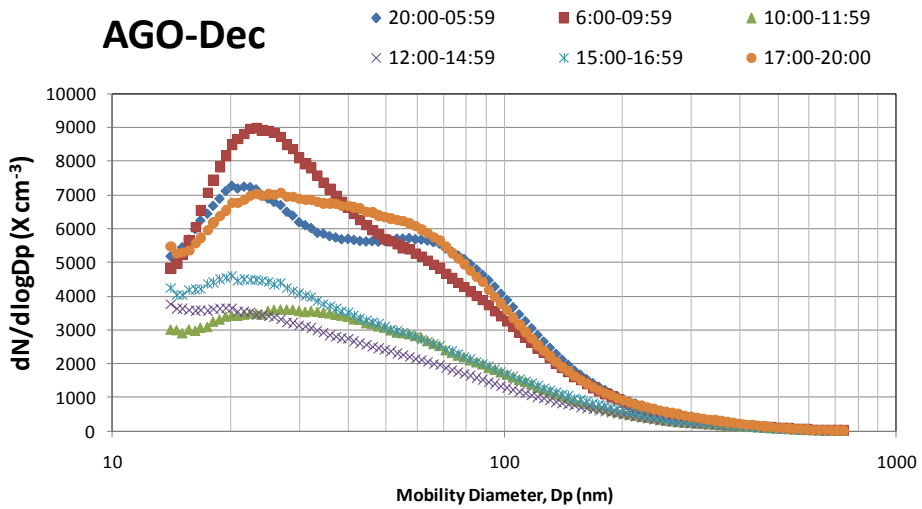

Fig. 4c. Average size distribution of particles during six time periods (PST) of the day at AGO during December 2009.

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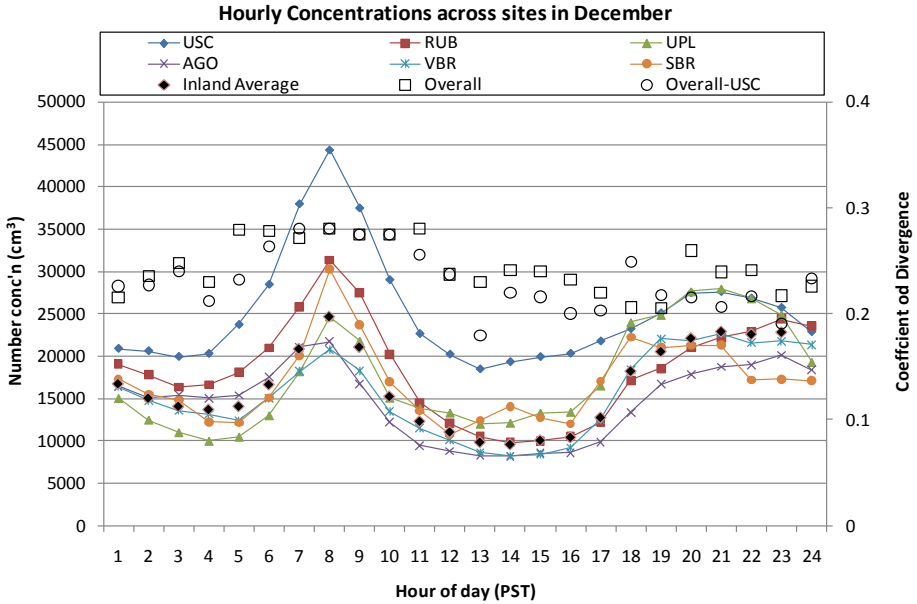

Fig. 5a. PNC and coefficients of divergence across sites for December 2008 (except SBR which is December 2009).

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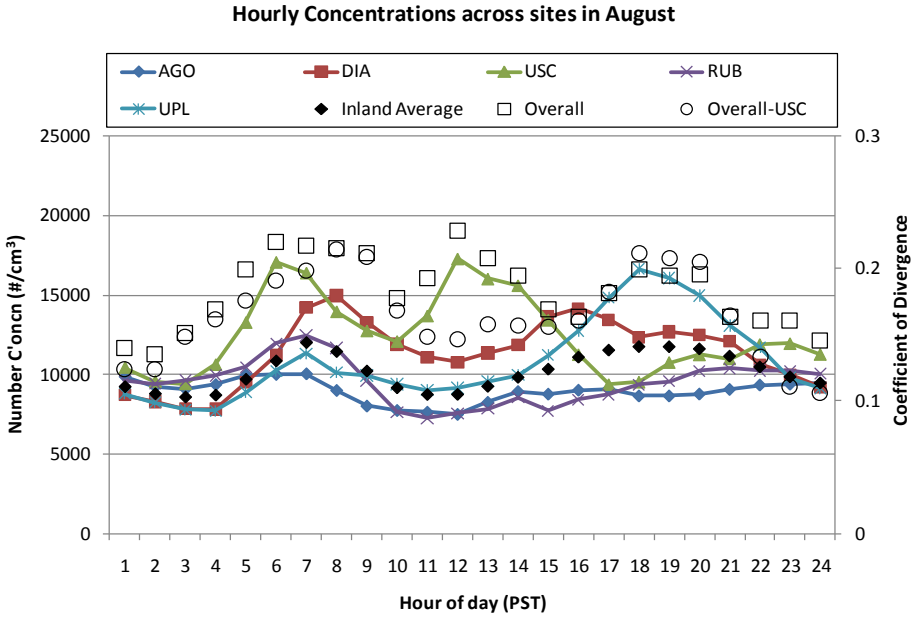

Fig. 5b. PNC and coefficients of divergence across sites for August 2009.

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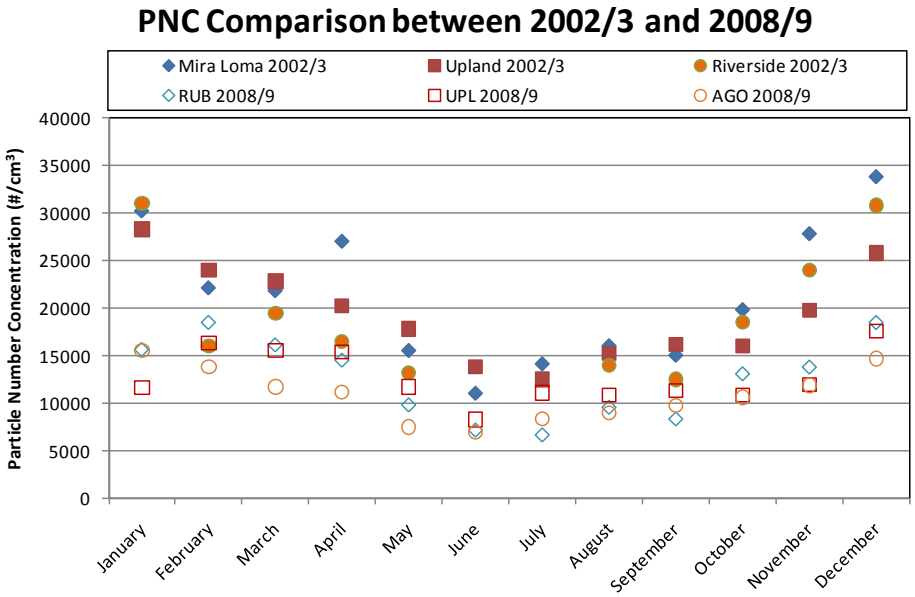

Fig. 6. Comparison of PNC at select sites measured during 2008/09 with Singh et al. (2006) measured during 2002/03.

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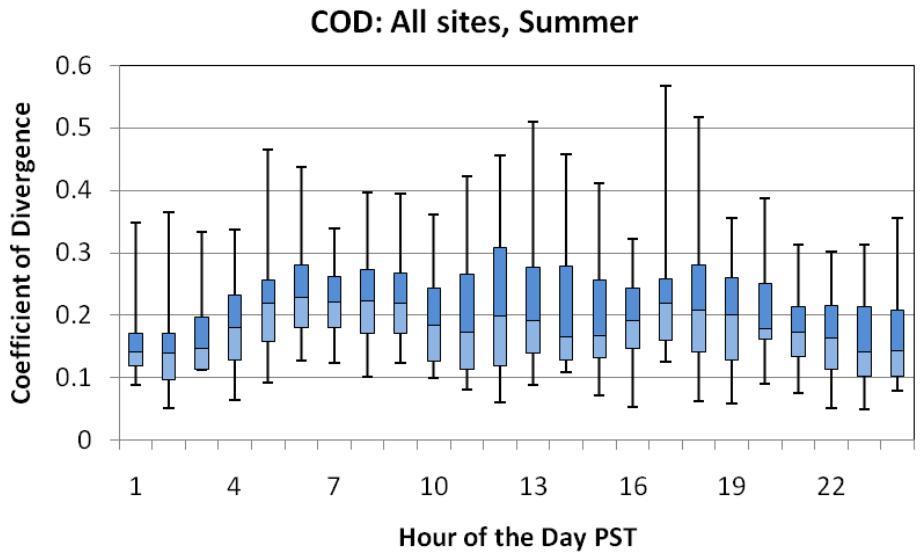

Fig. 7a. Coefficients of divergence during the summer months of May–August 2009.

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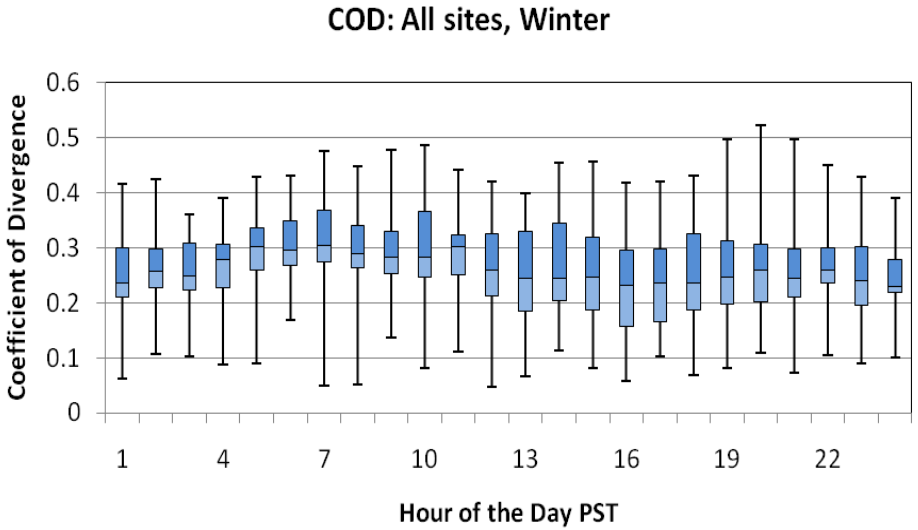

Fig. 7b. Coefficients of divergence during the winter months of December 2008–February 2009.

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