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Associations between Particle Number and Gaseous Co-Pollutant Concentrations in the Los Angeles Basin

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ABSTRACT

Continuous measurements of particle number (PN), particle mass (PM_{10}), and gaseous pollutants [carbon monoxide (CO), nitric oxide (NO), oxides of nitrogen (NO_x), and ozone (O_3)] were performed at five urban sites in the Los Angeles Basin to support the University of Southern California Children's Health Study in 2002. The degree of correlation between hourly PN and concentrations of CO, NO, and nitrogen dioxide (NO_2) at each site over the entire year was generally low to moderate (*r* values in the range of 0.1–0.5), with a few notable exceptions. In general, associations between PN and O_3 were either negative or insignificant. Similar analyses of seasonal data resulted in levels of correlation with large variation, ranging from 0.0 to 0.94 depending on site and season. Summertime data showed a generally higher correlation between the 24-hr average PN concentrations and CO, NO, and NO_2 than corresponding hourly concentrations. Hourly correlations between PN and both CO and NO were strengthened during morning rush-hour periods, indicating a common vehicular source. Comparing hourly particle number concentrations between sites also showed low to moderate spatial correlations, with most correlation coefficients below 0.4. Given the low to moderate associations found in this study, gaseous co-pollutants should not be used as surrogates to assess human exposure to airborne particle number concentrations.

IMPLICATIONS

The overall lack of significant associations between hourly and 24-hr PN versus gaseous co-pollutant concentrations can be attributed to the differences in the sources and formation mechanisms that are responsible for generating these pollutants in the environment of the Los Angeles Basin. These associations may become stronger for specific pollutants and time periods, for example, between PN, CO, and NO during traffic rush hours. However, if examined on a yearly or even on a seasonal basis, the results from this study suggest that co-pollutants such as CO, O_3, or NO_x cannot be used as surrogates to assess human exposure to PN in epidemiologic studies. These findings also imply that potential confounding effects of co-pollutants will not affect epidemiologic analysis seeking to link ultrafine particles to health effects because of the general lack of associations between PN and co-pollutant concentrations.

INTRODUCTION

Recent research has demonstrated that numerous adverse health outcomes are associated with atmospheric particulate matter (PM). Epidemiologic studies have shown significant relationships between ambient PM and respiratory-related mortality and morbidity.[1,2] The observed effects are even more significant in susceptible populations, such as the elderly, with pre-existing respiratory and cardiovascular diseases.[3] Toxicological studies by Oberdorster[4] and Donaldson et al.[5] have concluded that ultrafine particles (particles with diameters less than ~100 nm) are comparatively more toxic than larger particles with identical chemical composition and mass. Because of their small size, ultrafine particles contribute very little to the overall PM mass but comprise a significant majority of the number of airborne particles in the atmosphere.[3,6]

It is still unknown whether the observed PM health effects are related to particle number (PN), particle surface area, particle mass, or particle chemical composition. Studies on rodents show that inflammatory response is more prominent when ultrafine particles are administered compared with larger particles,[3] suggesting either a particle number or surface area effect. Toxicological studies also have shown that ultrafine particles have higher oxidative stress potential and can penetrate and destroy mitochondria within epithelial cells.[7] Penttinen et al.[8] tested the hypothesis that high numbers of ultrafine particles in the atmosphere can induce alveolar inflammation and exacerbation of pre-existing cardiopulmonary diseases. They found that daily mean number concentration and peak expiratory flow (PEF) are negatively associated and that the effect is most prominent with particles in the ultrafine range. Another study by Peters et al.[9] found associations between number concentrations of ultrafine PM and lowered PEF among asthmatic adults.

As one of many sources contributing to urban air pollution in general, the combustion of fossil fuel in motor vehicles is one of the major primary emission sources

of ultrafine particles in urban atmospheres.[10,11] Recent studies have demonstrated how ultrafine number concentrations drop dramatically with increasing distance from busy freeways in the Los Angeles Basin, confirming that vehicular pollution is the major source of ultrafine particles near and on the freeways and that high particle number counts can be a very local phenomenon (on scales of 100–500 m).[12,13] Source tests also have demonstrated that vehicles emit significant numbers of particle in the size range of 40–100 nm.[14–16] Particles with diameters less than 30 nm emitted from vehicles consist primarily of condensed semi-volatile material such as hydrocarbons derived from lubricating oils.[17] Other combustion sources, such as food cooking and wood burning, also can be sources of ultrafine particles to the atmosphere.[18,19]

In addition to primary, or direct, ultrafine particle emissions, photochemical secondary formation in the atmosphere also is responsible for the formation of ultrafine particles in the atmosphere.[20–26] Kulmala et al.[20] investigated particle formation by secondary processes and showed that such particle formation events are more distinct in the summer. Evidence of summertime secondary particle formation also is given by the studies of Shi and Qian[22] and Birmili et al.,[23] where they observed enhanced particle growth rates during that season. Particle formation rates depend strongly on the intensity of solar radiation,[20] but the exact mechanism by which the process occurs is not fully understood.[27] Once formed, particles are transformed by coagulation and condensation in the atmosphere 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.[24,28]

Ultrafine particles and co-pollutants, such as nitrogen oxide (NO) and carbon monoxide (CO), are emitted by both diesel and gasoline engines.[11,24] A previous study found significant correlation at street level in central Copenhagen between CO, NO_x, and particle number.[29] Such results may lead epidemiologists to believe that these vehicular co-pollutants will serve as surrogates for particle number. However, co-pollutant associations measured at a roadside may not necessarily reflect potential associations further downwind, where most of the population is exposed.

Strong correlations between various co-pollutants and PM also can make it difficult to estimate the contribution of each pollutant to the observed health outcome because of confounding effects.[30] Additional health studies have shown that ultrafine particles in the presence of a gaseous co-pollutant, ozone (O_3), can have a significantly enhanced adverse health effect, greater than the sum of the effects from the co-pollutants alone.[3] For these reasons, it is essential to fully understand the relationships among co-pollutants not just near vehicles or roadways but at all locations and times of human exposure.

Because of the different nature of sources, formation mechanisms, and atmospheric processes associated with each co-pollutant, one might expect the extent of correlation among co-pollutants to diminish as distance from the sources increases. For example, CO is largely unreactive on the time scales of urban transport and, thus, atmospheric concentrations are determined by emissions and dispersion alone.[31] NO, however, reacts to form nitrogen dioxide (NO_2), which can subsequently react to form nitric acid (HNO_3).[32] Particle number concentrations are influenced in the atmosphere by coagulation and by photochemical secondary processes.[20] Poor correlation among co-pollutants also might occur at or very near the source (i.e., tailpipe) because different combustion conditions will affect the emissions of different pollutants in different ways. Ultrafine particles from vehicles are formed in the engines, in the exhaust pipe, or immediately after exhaust to the atmosphere.[29] An appreciable portion of ultrafine particles from vehicular emissions may originate from uncombusted lubricating oil.[17] Thus, particle formation will depend strongly on the engine load, engine temperature, ambient temperature, and ambient relative humidity.[33,34] Hence, given the various combustion conditions at the sources and the different atmospheric behaviors, significant associations among co-pollutants may not be expected. The current study demonstrates these results by exploring the relationship between continuous and time-integrated particle number concentrations and those of various co-pollutants [i.e., NO, NO_2, O_3, particulate matter with aerodynamic diameter less than 10 μm (PM_{10}), and CO] at five sites in the Los Angeles Basin for the entire 2002 calendar year.

METHODS

Measurements of particle number, CO, O_3, PM_{10}, NO, and NO_x were conducted as part of the routine sampling protocol of the South Coast Air Quality Management District. Continuous data were collected concurrently over the calendar year 2002 at five different sites in the Los Angeles Basin (Figure 1) in support of the University of Southern California Children's Health Study (CHS). The data from this sampling campaign, which started in 1993, are currently being used by CHS epidemiologists to investigate the association between air pollution and the incidence, prevalence, and severity of childhood asthma and lung function.[35]

The five sites selected for the current analysis are Long Beach, Glendora, Mira Loma, Upland, and Riverside. The choice of sampling sites considered in this study is based on their location within the Los Angeles Basin as well as the adverse health outcomes observed at these

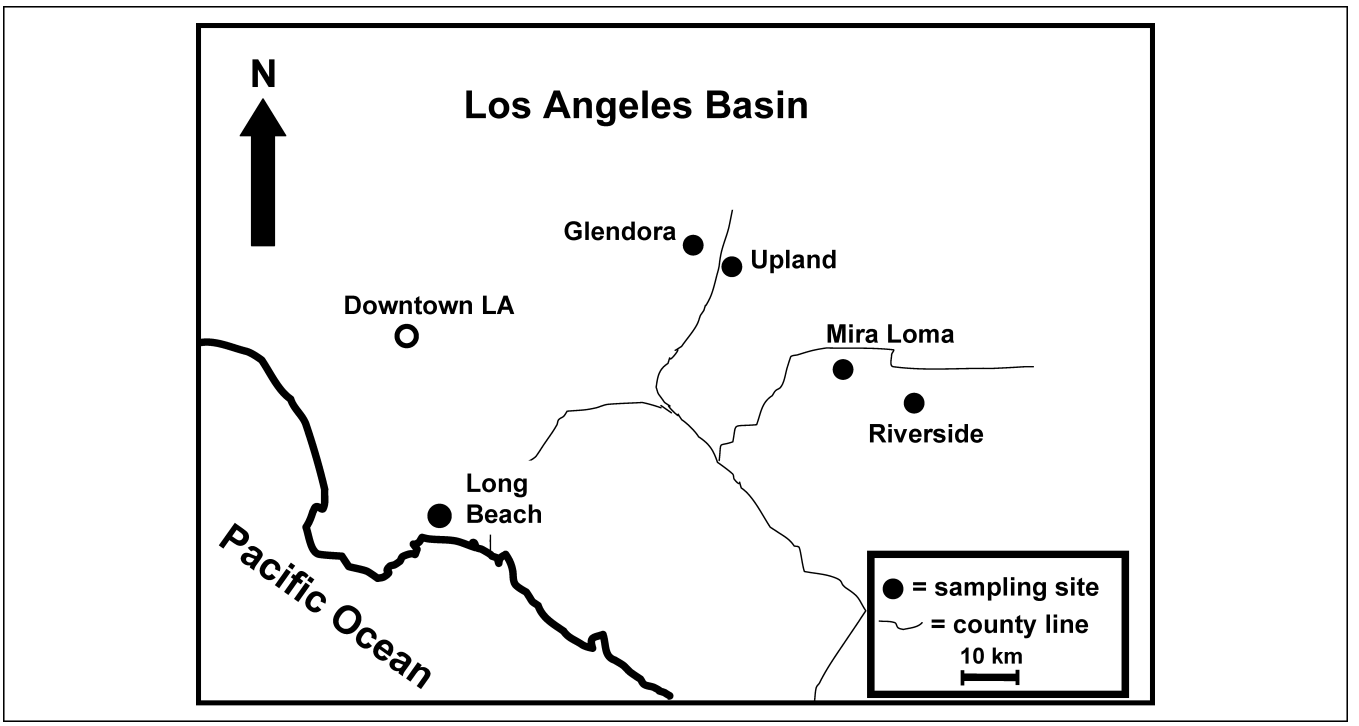


Figure 1. Locations of the source site (Long Beach) and the receptor sites (Mira Loma, Riverside, Upland, and Glendora) in the Los Angeles Basin.

sites[36] attributed to air pollution. Kunzli et al.[36] have studied these sites as part of the CHS investigating the long-term consequences of air pollution on the health of children. These sites generally are considered typical polluted urban areas of the Los Angeles Basin and have shown high prevalence of early childhood asthma among the residents. The high incidence of adverse effects of air pollution at these sites makes them appropriate locations to investigate the associations between PN and various co-pollutants.

The Long Beach station is located on one of the busiest streets in Long Beach and is ~0.8 km northeast of a major freeway. The Glendora station is located in a residential area, at least 3 km away from major roadways. The Upland site is less than 1 km north of a freeway and is located in a residential area ~6 km downwind of the Glendora site. The Mira Loma site is situated inside Jurupa Valley High School. It is directly east of a major freeway interchange (~3 km), is surrounded by several major warehouse facilities and residential communities, and is located ~80 km east of downtown Los Angeles. The sampling location at Riverside is at the Citrus Research Center and Agricultural Experiment Station (CRS–AES), a part of the University of California–Riverside. It is ~10 km southeast of the Mira Loma site and is situated upwind of surrounding freeways and major roads.

The five sites also are differentiated based on the characteristics of the air pollution in the Los Angeles Basin. Because of the relatively western location and the surrounding urban environment, Long Beach is considered a "source" site, where fresh particles are emitted primarily from vehicular and industrial sources, and thus represents well-mixed urban air. The other sites, Riverside, Upland, Glendora, and Mira Loma, are designated "receptor" sites, which have comparatively less traffic density and experience advected, aged, and photochemically processed air masses from the central Los Angeles area.[37] The transport time of air masses from source to receptor sites can vary from a few hours to more than a day. It is important to note that a receptor site may be close to a freeway, for example, the site at Upland. The proximity of Upland to a freeway alters some of the characteristics typical of a receptor site, adding the influence of local traffic to the regional effects. Glendora and Upland are on what has been termed the vehicular trajectory, while Mira Loma and Riverside are on the so-called nitrate (NO_3^-) trajectory, influenced by the Chino area dairy farms, a strong ammonia (NH_3) source leading to high concentrations of ammonium nitrate (NH_4NO_3).[32,38]

Particle number (particle diameter >15 nm) concentrations were measured continuously by a CPC (condensation particle counter, Model 3022/A, TSI Inc.). The CPC was set at a flow rate of 1.5 L/min. PM_{10} mass was measured using a tapered element oscillating microbalance (TEOM 1400A, R&P Inc.). The concentrations of CO were measured near-continuously by means of a Thermo Environmental Inc. Model 48C trace-level CO monitor. Concentrations of NO and NO_2 were measured by a continuous chemiluminescence analyzer (Monitor Labs Model 8840), and O_3 concentrations were measured using a UV photometer (Dasibi Model 1003 AH). Continuous particle

number and gaseous co-pollutant concentrations were averaged to form 1- and 24-hr average values for the subsequent analysis. All instruments were calibrated every 3 months by AQMD (Air Quality Management District) and ARB (Air Resource Board). The sampling, which is part of a compliance measurement network, was subjected to rigorous quality assurance procedures.

The analysis presented here is limited to assessing the associations among co-pollutants, presenting explanations for the presence or lack of associations, and discussing the implications for epidemiologic studies using such data. A thorough examination of the diurnal, geographical, and seasonal patterns of these six co-pollutants will be presented in a future publication.

RESULTS AND DISCUSSION

The Pearson correlation coefficient (r) between the hourly and 24-hr average PN concentration data and the co-pollutant concentrations (CO, NO, O_3, NO_2, and PM_{10}) are shown in Tables 1 and 2, respectively. Given the large number of data, even low values of r can indicate significance.[39] For example, hourly correlations in most cases were based on over 2000 data points, for which r values as low as 0.05 are significant at a $p = 0.05$ level. Nevertheless, these small values of r should be viewed cautiously in the context of exposure and epidemiology and as to whether one of these pollutants can be substituted as a predictive surrogate of another. The results shown in Tables 1 and 2 are based on the entire data set from the calendar year 2002. The r values for the hourly concentrations ranged from a high of 0.66 for PN versus CO in Upland down to practically 0.00. Interestingly, associations between PN and O_3 resulted in negative correlations in most cases, based on both hourly and 24-hr data. Similarly, the r for the 24-hr average concentrations ranged from 0.00 to 0.68 (PN vs. NO_2 in Long Beach). The values shown in Table 1 and Table 2 indicate generally low to moderate (i.e., $r < 0.5$) levels of association between PN and co-pollutants, and in only few cases, moderate to high (r between 0.5 and 0.68) levels of correlation on both an hourly and 24-hr basis. Based on these results, which were highly variable among sites, using any of these co-pollutants as a surrogate for ultrafine particles over a prolonged time period, such as an entire year, in Los Angeles is likely to introduce a high level of error in PN predictions.

Table 1. Hourly Pearson correlation coefficient, r, of PN vs. co-pollutant concentrations for the entire calendar year 2002, all sites.

	Glendora/TD	Long Beach	Mira Loma	Riverside	Upland
CO	0.13	0.46	0.47	0.52	0.66
NO	0.06	0.44	0.60	0.59	0.65
NO_2	0.21	0.50	0.24	0.32	0.17
PM_{10}	0.18	0.27	0.00	−0.16	0.14
O_3	0.30	−0.22	−0.34	−0.04	−0.26

Table 2. 24-hr average Pearson correlation coefficient, r, of PN vs. co-pollutant concentrations for the entire calendar year 2002, all sites.

	Glendora	Long Beach	Mira Loma	Riverside	Upland
CO	0.00	0.50	0.44	0.39	0.63
NO	0.30	0.48	0.34	0.32	0.66
NO_2	0.07	0.68	0.11	0.23	0.08
PM_{10}	−0.18	0.10	−0.17	−0.32	−0.19
O_3	−0.31	−0.63	−0.33	−0.26	−0.54

Figures 2a-e display the scatter plots of the hourly concentrations of PN versus the five measured co-pollutants at the Upland site, where the highest correlations were observed. The relatively high correlations of PN with CO and NO are driven by the higher concentration values, indicating the occasional influence of vehicular sources emitting these three pollutants. The other four sites show even lower levels of correlation between PN and co-pollutants (as shown in Tables 1 and 2) and are not graphically represented.

It is possible that seasonal differences in source strengths, ambient temperatures, sunlight, and weather patterns, which might affect different pollutants differently, could account for the lack of correlation over an entire year. Thus, the data were separated by season to determine whether the associations among co-pollutants improved. The seasons were defined as winter (December–February); spring (March–May); summer (June–August); and fall (September–November). December 2002 was combined with January and February 2002 to form a winter period. Table 3 shows the Pearson correlation coefficients between PN and the other co-pollutant (both hourly and 24-hr average) concentrations for the five sites and the four seasons. The lowest levels of correlation ($r < 0.59$) between PN and co-pollutants, for any season and for both averaging times, were observed in Glendora. The apparent lack of correlation with NO and CO may be caused by the minimal traffic influence at that site, which is located in a residential area and away from major roadways. However, at the Upland site, which is located only 6 km to the east, the hourly correlation between PN and the other primary vehicular emissions (CO and NO) showed higher correlations, particularly in the winter and fall. The PN correlations with CO and NO were almost always lower for the 24-hr average data than for the hourly averages. The high hourly correlations for vehicular pollutants suggest a local traffic source at Upland. Furthermore, the association is higher in the winter, when stagnant conditions and lower inversion layers trap

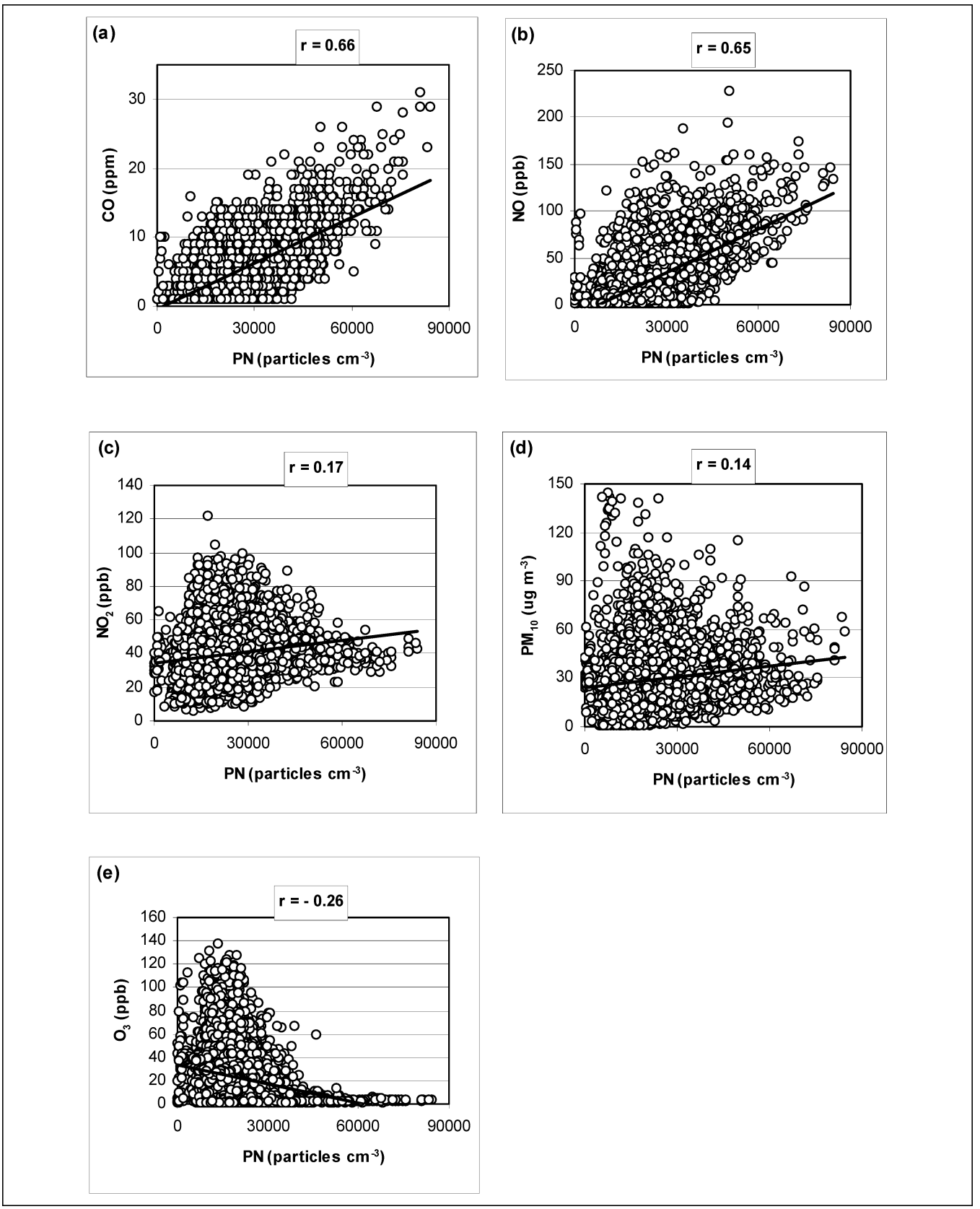


Figure 2. Hourly PN vs. co-pollutant concentrations at Upland for the entire year 2002. (a) CO, (b) NO, (c) NO_2, (d) PM_{10}, and (e) O_3.

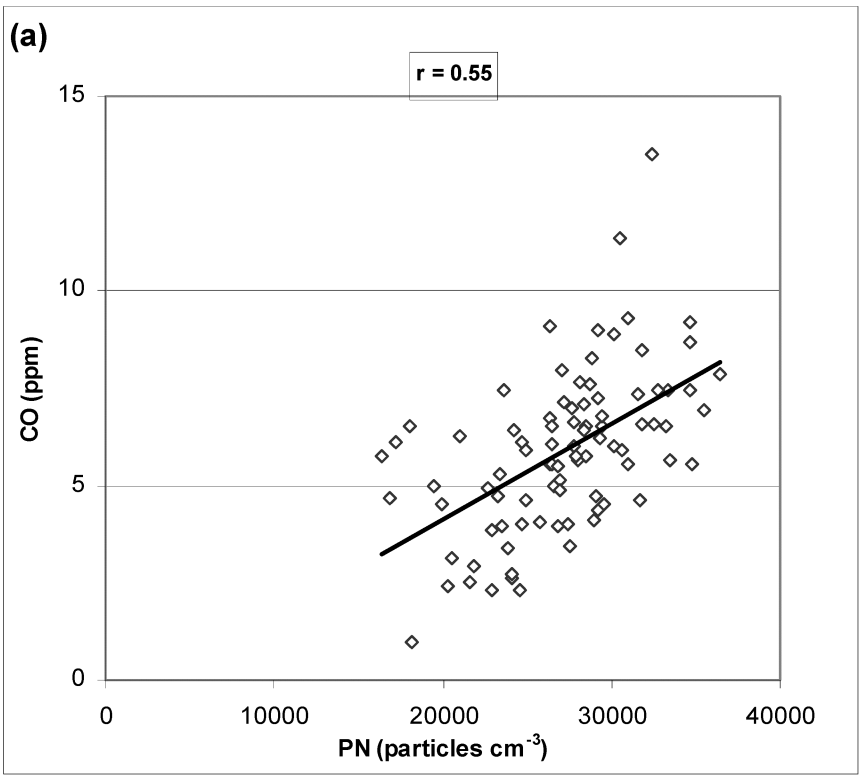
Table 3. Hourly and 24-hr average Pearson correlation coefficient (r) of PN vs. co-pollutant concentrations on a seasonal basis.

	Glendora		Long Beach		Mira Loma		Riverside		Upland	
Winter	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**
CO	0.20	0.19	0.33	0.47	0.17	0.43	0.33	0.43	0.55	0.74
NO	0.35	0.42	0.58	0.48	0.55	0.67	0.41	0.53	0.54	0.72
NO_2	0.37	0.33	0.64	0.58	0.34	0.29	0.38	0.33	0.20	0.35
PM_{10}	0.03	0.30	0.19	0.54	0.51	0.53	0.06	0.01	0.17	0.41
O_3	−0.64	−0.37	−0.42	−0.28	−0.48	−0.65	−0.02	−0.14	−0.38	−0.25
	Glendora		**Long Beach**		**Mira Loma**		**Riverside**		**Upland**	
Spring	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**
CO	0.04	0.14	0.27	0.28	0.46	0.59	0.38	0.61	0.50	0.57
NO	0.49	0.00	0.39	0.41	0.36	0.70	0.38	0.66	0.63	0.59
NO_2	0.16	0.29	0.52	0.34	0.46	0.46	0.30	0.48	0.34	0.30
PM_{10}	0.13	0.37	0.10	0.18	0.14	0.25	−0.11	−0.05	0.06	0.26
O_3	−0.24	−0.40	−0.54	−0.15	−0.23	−0.03	−0.18	−0.04	−0.21	−0.16
	Glendora		**Long Beach**		**Mira Loma**		**Riverside**		**Upland**	
Summer	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**
CO	0.27	0.14	0.46	0.40	0.56	0.49	0.71	0.54	0.52	0.48
NO	0.36	0.10	0.61	0.49	0.67	0.65	0.87	0.65	0.49	0.38
NO_2	0.37	0.28	0.68	0.53	0.78	0.50	0.79	0.70	0.55	0.33
PM_{10}	0.45	0.46	0.31	0.44	0.57	0.26	0.52	0.30	0.16	0.21
O_3	0.07	0.59	−0.17	0.23	0.30	0.11	0.56	0.17	0.05	0.19
	Glendora		**Long Beach**		**Mira Loma**		**Riverside**		**Upland**	
Fall	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**	**24-hr Avg.**	**Hourly**
CO	0.25	0.31	0.88	0.56	0.32	0.47	0.41	0.41	0.53	0.64
NO	0.53	0.19	0.79	0.60	0.11	0.69	0.33	0.25	0.60	0.64
NO_2	0.38	0.35	0.94	0.66	0.20	0.45	0.49	0.33	0.29	0.32
PM_{10}	0.06	0.23	0.71	0.49	0.05	0.33	0.41	0.25	0.27	0.34
O_3	0.04	0.28	−0.10	−0.05	0.13	−0.23	0.52	0.10	−0.13	−0.25

pollutants near the ground and enhance the influence of local sources. Figures 3a and b display the scatter plots of the PN and CO data collected in Upland in the winter season. As mentioned before, the entire data set of 24-hr average values in Figure 3a shows a lower r, while the hourly data in Figure 3b are more correlated. The suggestion that traffic sources from the adjacent freeway heavily influence Upland is supported by the higher correlation for the morning rush-hour period between 5:00 and 8:00 a.m. Most of the higher levels of both pollutants were observed during this morning rush-hour period. As discussed earlier, Upland is situated close to a freeway and, even though it is a receptor site, pollutants generated on the adjacent freeway influence pollutant levels there. The strong diurnal patterns of traffic result in higher levels of hourly correlations compared with 24-hr average correlations.

The lower correlations between PN and both CO and NO in the spring and summer is most likely caused by the different meteorological conditions, which favor increased advection of pollutants from urban Los Angeles areas located upwind of that site. The influence of an aged, transported air mass will act to reduce the modest associations among vehicular co-pollutants that one might expect from local emissions alone. Another factor may be photochemical secondary formation of particles in the atmosphere,[24,28] which would increase particle numbers while not affecting CO or NO levels. Other sites that showed moderate to relatively high levels of correlation among hourly measurements of vehicular pollutants and, thus, are thought to be influenced by local traffic, were Mira Loma and Riverside in the spring and summer.

Several previous studies have explored the relationship between PN and co-pollutants at locations that are heavily influenced by traffic.[8,29,40] Noble et al.[41] found high correlations between CO, NO, and hourly ultrafine particle number (UF) in winter in El Paso, TX, with Pearson correlation coefficients (r) ranging from 0.81 to 0.74 for UF versus CO and UF versus NO, respectively. Both sampling sites in this study were heavily influenced by traffic



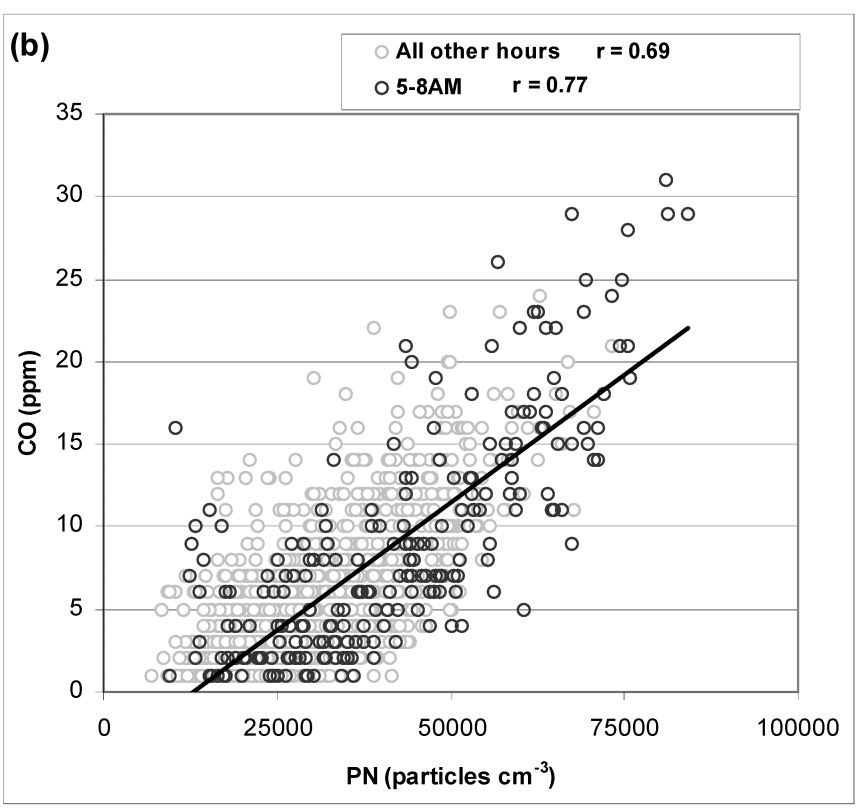


Figure 3. PN vs. CO in Upland during winter, 2002. (a) 24-hr average and (b) hourly.

sources. Cyrys et al.[11] found moderate Spearman correlation coefficients (r) between 0.57 and 0.55 for PN (0.01–2.5 μm) versus CO and NO_2, respectively, on an hourly basis in Erfurt, Germany, for the entire calendar year of 1997. By comparison, the observed winter r in Upland for PN versus CO and NO of 0.74 and 0.72, respectively, are

similar to the El Paso study, and the CO versus PN correlation is actually higher than the Erfurt study.

Morawska et al.[42] also have found lower but significant correlations of PN with CO ($r = 0.45$) and NO_x ($r = 0.40$) over 10 months in Brisbane, Australia, at a site very near a freeway. While high correlations between hourly measurements of vehicular co-pollutants indicate a local traffic source, high correlations of 24-hr average co-pollutant concentrations may signify a different influence. Table 3 shows that the 24-hr correlation coefficients are often higher than their hourly counterparts, especially in the receptor sites, with the exception of Glendora. This trend is more pronounced in the summer season and applies to non-vehicular and secondary co-pollutants as well. The summer season showed high ($r > 0.70$) degrees of correlation of 24-hr PN with CO, NO, and NO_2 at Riverside, and moderately high correlations between PN and CO, NO, and NO_2 at Upland ($r = 0.49–0.55$) and Mira Loma ($r = 0.56–0.78$). In all these cases, the daily average data were more correlated than the hourly data. Thus, while the hourly levels of certain co-pollutants may not peak, dip, or vary in similar diurnal patterns, the 24-hr average levels of all pollutants might show higher correlations during a day of generally poor air quality.

The summer period in Los Angeles is characterized by increased on-shore flow and, thus, enhanced advection of air masses inland.[28,37] Pollutant concentrations, especially at inland sites, are, therefore, influenced by upwind sources and aged air parcels, as opposed to the influence of more local sources during the stagnant conditions in the winter. This may provide an explanation for the higher 24-hr correlations between PN and the more secondary (formed during transport and aging) co-pollutants during the warmer seasons. Interestingly, the highest associations between PN and PM_{10}, CO, NO, and NO_2 in the site of Long Beach, whether based on hourly or 24-hr data, were observed in the fall season. Also, 24-hr concentrations between PN versus CO, NO, and NO_2 were found to be in higher correlation than their corresponding hourly values in the summer period, similar to the rest of the sites.

The scatter plots for PN versus CO, NO, and NO_2 during the summer at Riverside are shown in Figures 4a-f. As already discussed, the correlation is higher in all cases for the 24-hr data than for the hourly data, reaching as high as $r = 0.87$ for NO. Although advection on generally polluted days is causing these high associations, the influence of local sources is not absent. The hourly data (Figures 4b, d, and f) demonstrate that the relatively high hourly correlations given in Table 3 are driven by the high levels during the morning commute hours.

In addition to traffic emissions, another source of ultrafine particles, and, thus, increased PN concentrations, is photochemical reactions in the atmosphere that form new particles by secondary processes.[24,28,43,44] The mechanisms of particle formation are still poorly understood, but it is expected that it is more likely to occur on days of high photochemical activity (an atmosphere with high concentrations of gaseous pollutants and a clear day with a high solar zenith angle). These conditions also will generally favor the production of O_3. Thus, one might expect increases in particle numbers from photochemical processes to accompany increases in O_3 concentrations.

To investigate this relationship further, the Glendora site was selected to minimize the effects of local traffic. As described, the low PN versus CO and NO correlations (Tables 1 and 2) at Glendora indicate that this site is influenced very little by local traffic sources. The summer season at Glendora was examined because this is when photochemistry is expected to be at its highest and it is when the highest hourly PN versus O_3 correlation was found ($r = 0.59$, Table 3). Studies by Shi and Qian, Stanier et al., and Kulmala et al. have observed that production of ultrafine particles by secondary processes are more pronounced in the summer period.[20–22] As shown in Figure 5, the r value of 0.59 increases to a maximum of 0.74 if PN is compared with O_3 levels ~2 hr before (i.e., a 2-hr lag time). Figure 6 shows how the r between PN and O_3 varies as the lag time is changed. The correlation starts increasing with a lag of 1 hr, and the peak correlation was found at 2 hr, as shown in Figure 5. This is followed by a decrease in the association as the lag time is increased to 3 hr or more. Thus, it seems that, while photochemistry is responsible for the increases in both pollutants, an increased association is found when PN levels are compared with O_3 concentrations 1–3 hr earlier. Previous investigations on secondary ultrafine particle formation showed that these particles are initially formed in the size range of 1–2 nm[45,46] and grow to larger sizes (such as those that can be detected by a CPC) either by condensation of low-volatility organics[44,47] on these nuclei or by heterogeneous organic reactions possibly catalyzed by the presence of acids.[27] Low-volatility organics are formed by the oxidation of organic vapors by hydroxyl radicals, and the photolysis of O_3 is one of the mechanisms by which hydroxyl radicals may be produced in the atmosphere.[48] It is hypothesized that the time period of ~1–3 hr for the strengthening of association between O_3 and PN is the time required for all of these processes to occur, starting from the production of hydroxyl radicals to the eventual growth of particles to a detectable size by the CPC, which in our case was larger than ~15 nm. The explanation also can be supported by the findings of Kulmala et al.,[20] where they observed a particle growth rate of 5 nm/hr during the summer. This is consistent with the observed lag time in that it would take ~1–3 hrs for the small particles to grow, by the condensation of

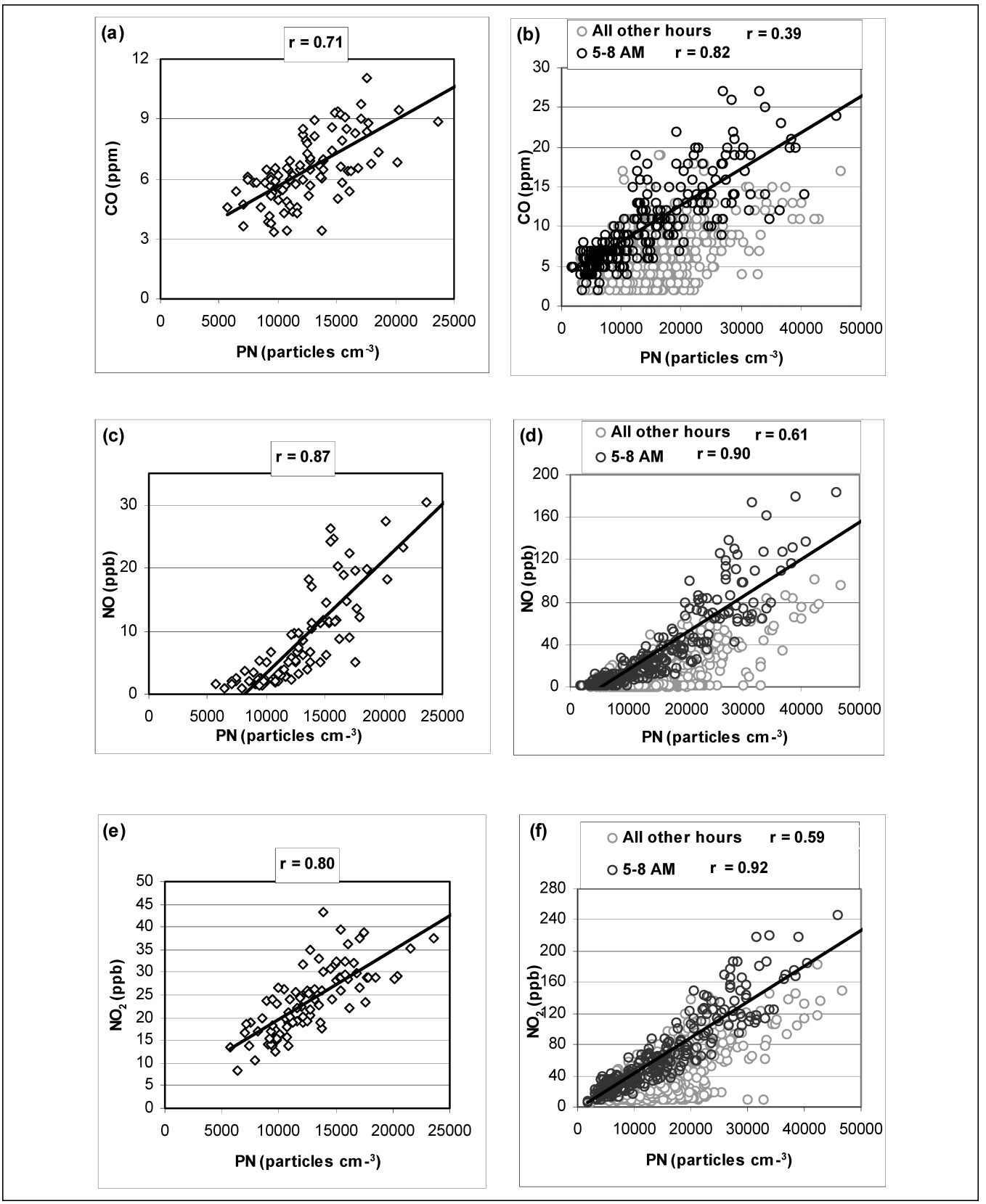


Figure 4. PN vs. CO, NO, and NO_2 in Riverside during summer, 2002. (a) 24-hr average PN vs. CO and (b) hourly PN vs. CO. (c) 24-hr average PN vs. NO. (d) Hourly PN vs. NO. (e) 24-hr average PN vs. NO_2. (f) Hourly PN vs. NO_2.

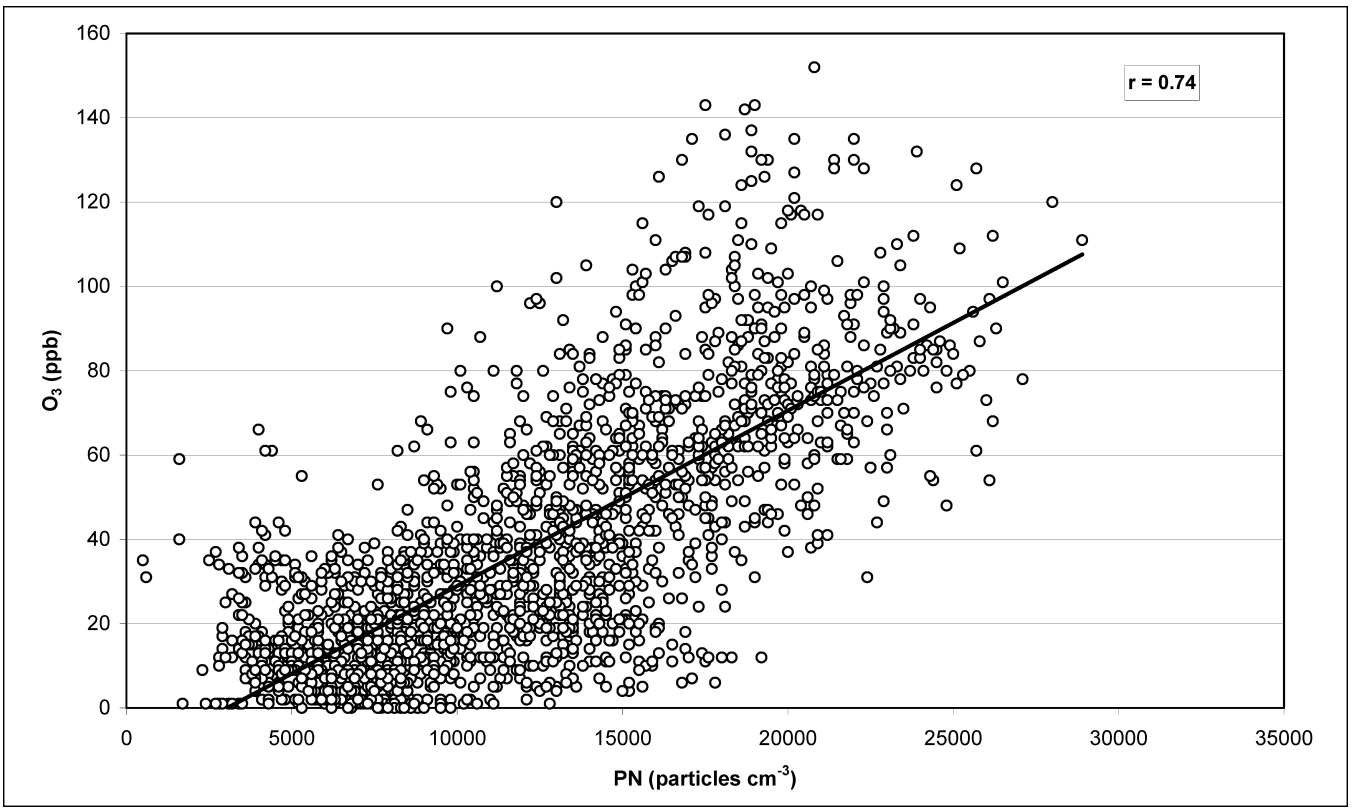


Figure 5. Hourly PN vs. O_3 concentrations in Glendora during summer 2002 with a lag time of 2 hr (PN vs. O_3 2 hr earlier).

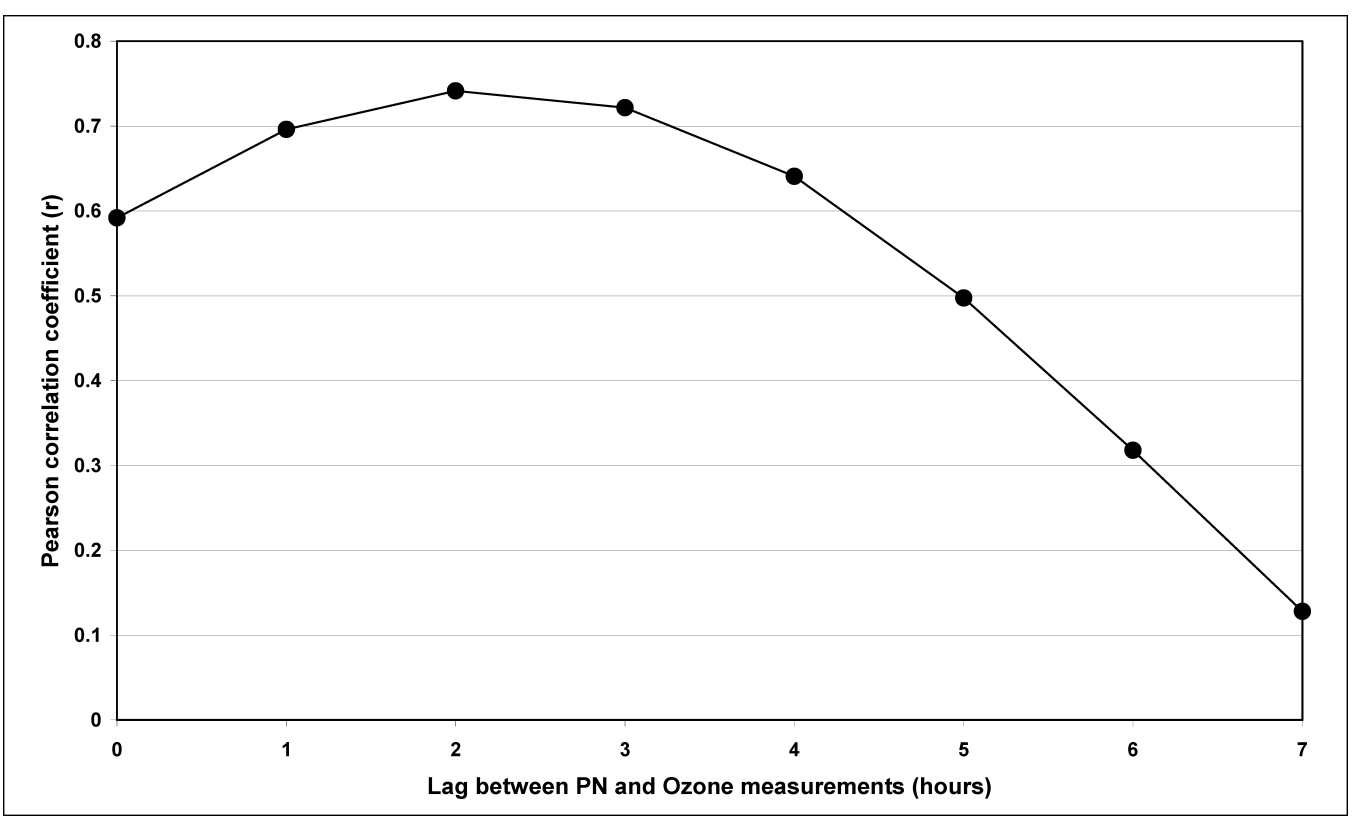


Figure 6. Pearson correlation coefficient, *r*, vs. lag time between measurements of O_3 and PN.

photochemically formed organics, to the size range of 15 nm, which then can be detected by the CPC.

Concurrent measurements at different sites allow the investigation of the geographical homogeneity of PN concentrations across the Los Angeles Basin. This is important for epidemiologic studies that require predictions of human exposure based on a limited number of sampling locations. Table 4 shows the Pearson correlation coefficient, *r*, from intersite correlations of concurrently measured PN concentrations. For all four seasons, correlations of hourly data across sites results in relatively low *r* values, mostly below 0.3. In almost all cases, the 24-hr average PN concentrations were more correlated across sites than were the hourly data. The spatial correlations between the 24-hr PN concentrations increase in the spring and summer periods, especially among the receptor sites of Upland, Mira Loma, Riverside, and Glendora. As was previously mentioned, meteorological conditions that favor a poor air quality day will lead to high particle number and other pollutant levels across the basin, even if the timing of the daily peaks varies enough to cause minimal hourly associations among sites. Examined on a yearly basis, however, the *r* values are low enough to indicate that particle number concentrations are spatially inhomogeneous and that using central site monitoring to predict population exposure to PN on an hourly or even daily basis would not be accurate. Although this finding was somewhat anticipated, the analysis presented here is the first of its kind in the Los Angeles Basin and has serious implications in regulatory and compliance monitoring. For instance, Glendora and Upland, which are only 6 km apart, show moderate to relatively high levels of correlation in the range of 0.63–0.71 of 24-hr average PN for three seasons (spring, summer, and fall), but have an *r* of only 0.23 in the winter, when stagnate weather conditions limit transport. Similarly, the correlation coefficients based on hourly concentrations range from 0.33 in the winter to 0.55 in the summer.

Table 4. 24-hr average and hourly (*italic*) Pearson correlation coefficient (*r*) of PN vs. PN across sites on a seasonal basis.

Winter	**Glendora**	**Long Beach**	**Mira Loma**	**Riverside**	**Upland**
Glendora	—	0.17	0.23	−0.63	0.23
Long Beach	*0.25*	—	−0.20	0.58	0.38
Mira Loma	*0.11*	*0.13*	—	0.32	0.33
Riverside	*−0.41*	*0.13*	*0.22*	—	0.62
Upland	*0.33*	*0.44*	*0.42*	*0.12*	—
Spring	**Glendora**	**Long Beach**	**Mira Loma**	**Riverside**	**Upland**
Glendora	—	0.50	0.13	0.33	0.71
Long Beach	*0.27*	—	0.54	0.21	0.70
Mira Loma	*0.12*	*0.13*	—	0.75	0.63
Riverside	*−0.15*	*0.07*	*0.27*	—	0.60
Upland	*0.37*	*0.29*	*0.49*	*0.45*	—
Summer	**Glendora**	**Long Beach**	**Mira Loma**	**Riverside**	**Upland**
Glendora	—	0.32	0.46	0.52	0.63
Long Beach	*0.40*	—	0.41	0.54	0.31
Mira Loma	*0.22*	*0.01*	—	0.70	0.51
Riverside	*0.13*	*0.01*	*0.13*	—	0.78
Upland	*0.55*	*0.04*	*0.08*	*0.25*	—
Fall	**Glendora**	**Long Beach**	**Mira Loma**	**Riverside**	**Upland**
Glendora	—	−0.18	0.27	0.43	0.66
Long Beach	*0.25*	—	0.30	0.19	0.09
Mira Loma	*0.09*	*0.12*	—	0.08	0.47
Riverside	*0.03*	*0.05*	*0.22*	—	0.21
Upland	*0.44*	*0.23*	*0.45*	*0.14*	—

Figure 7a demonstrates why the Glendora/Upland association is moderate (r = 0. 55) on an hourly basis in the summer, despite the proximity of the two sites. The scatter plot shows that, during the morning traffic hours from 5:00 to 8:00 a.m., Upland experiences an excess of particle numbers relative to Glendora because of the local vehicular sources. When this time period is excluded from the analysis, the *r* increases to 0.70. The clustering of the non-morning data about the 1:1 line shows that the general PN levels at these sites are comparable. Because these two sites are separated by ~6 km east to west, it will take time for an air mass at Glendora to be transported to Upland downwind. Figure 7b shows the same data as Figure 7a, but the Upland data are now compared with Glendora data measured 1 hr previously. The *r* of the non-morning traffic data increases to 0.74, and it is clear that the amount of scatter around the 1:1 line decreases. Figures 7c and d show lag times of 2 and 3 hr between the sites, and the *r* are observed to decrease while the scatter visibly increases. A 1-hr lag suggests an approximate wind speed of 6 km/hr, which is typical at those sites at that time of year.[24] Thus, the Upland site is influenced by both local traffic sources (especially in the morning) and the advection of air masses from upwind. Similar to Upland, the measured PN concentrations in any other location, for any season and time of day, are a combination of the contributions of three major sources, that is, traffic, long-range transport via advection, and photochemistry (in the summer). The regional nature of the latter two source/formation mechanisms (i.e., long-range transport and photochemistry), would tend to create a spatially homogeneous ultrafine aerosol, but the varying degrees to which vehicular sources influence a specific sampling site disrupt this homogeneity.

Again, an attempt was made to find correlations among individual co-pollutants at proximal sites, for example, Glendora versus Upland and Mira Loma versus Riverside, by taking the hourly concentrations for the entire year 2002. At Glendora and Upland, the intra-species

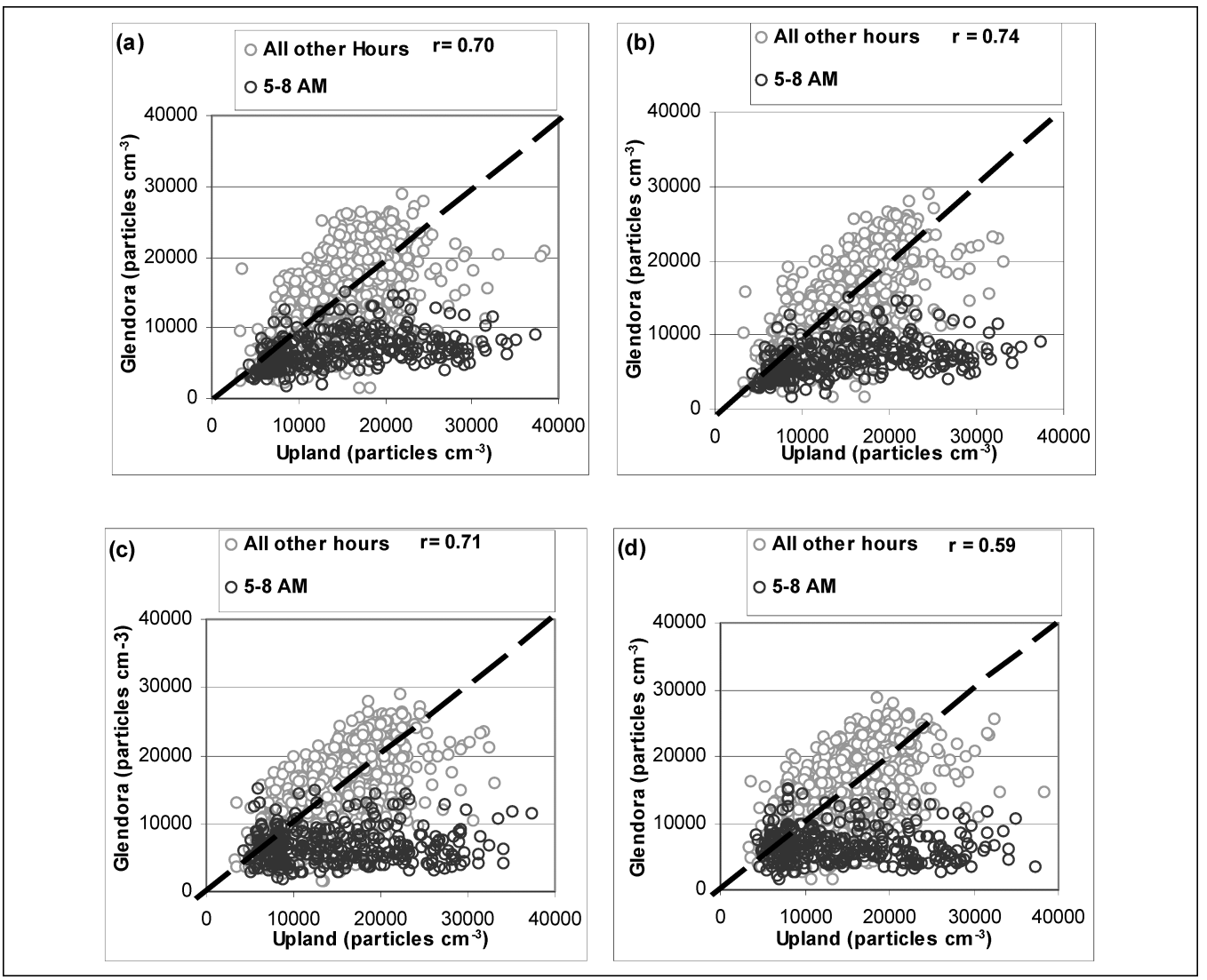


Figure 7. Comparison of PN concentrations in Upland and Glendora during summer 2002. (a) No lag time, (b) 1-hr lag time, (c) 2-hr lag time, and (d) 3-hr lag time. Dashed lines indicate the ideal 1:1 relationship and therefore, does not fit the data.

associations for CO, NO, NO_2, and O_3 are found to be 0.12, 0.48, 0.73, and 0.92, respectively. For Riverside and Mira Loma, the correlations for CO, NO, NO_2, and O_3 were found to be 0.65, 0.79, 0.83, and 0.95, respectively. It is important to note that even pollutants like CO and NO with typically local character are found to be correlated. The high association between O_3 obviously signifies regional nature of the pollutant. The most important conclusion from this analysis is that the spatial inhomogeneity of PN at Riverside and Mira Loma is higher than that for CO or NO, which makes the analysis of PN associations at proximal sites extremely informative. The 24-hr average yearly data also were analyzed to determine the inter-correlations between PN, PM_{10}, and gaseous co-pollutants at all five sites. The 24-hr average Pearson correlation coefficients for all species are shown in Table 5. The associations between PN and the other pollutants already were discussed and presented in Table 2. The varying degree of correlations among CO, NO, and NO_2 in each site (*r* values ranging from 0.29 to 0.84), probably reflects differences in the mix of traffic sources (i.e., gasoline vs. diesel engines) impacting each location. It is interesting to point out that in almost every site, the 24-hr concentrations of O_3 are negatively correlated with those of CO and NO when examined over an entire year. O_3 reaches its highest concentrations in the summer time, during the peak of the photochemical period, whereas the concentrations of CO and NO are maximum in the winter, when inversions and the depressed mixing depth of the atmosphere tend to increase the contribution of local vehicular emissions to air pollution. Riverside and Mira Loma show similar *r* values for PN versus the co-pollutants as well as among CO, NO, and NO_2. This similarity may be explained by the close proximity of the two sites.

Table 5. 24-hr average Pearson correlation coefficient, *r*, between all species sampled for the entire calendar year 2002, all sites.

Long Beach	**PN**	**CO**	**NO**	**NO_2**	**PM_{10}**	**O_3**
PN	1.00	—	—	—	—	—
CO	0.50	1.00	—	—	—	—
NO	0.48	0.58	1.00	—	—	—
NO_2	0.68	0.66	0.68	1.00	—	—
PM_{10}	0.10	0.08	0.10	0.34	1.00	—
O_3	−0.63	−0.59	−0.63	−0.77	−0.02	1.00
Riverside	**PN**	**CO**	**NO**	**NO_2**	**PM_{10}**	**O_3**
PN	1.00	—	—	—	—	—
CO	0.39	1.00	—	—	—	—
NO	0.32	0.76	1.00	—	—	—
NO_2	0.23	0.68	0.63	1.00	—	—
PM_{10}	−0.32	0.32	−0.55	−0.14	1.00	—
O_3	−0.26	0.07	−0.05	0.37	0.50	1.00
Mira Loma	**PN**	**CO**	**NO**	**NO_2**	**PM_{10}**	**O_3**
PN	1.00	—	—	—	—	—
CO	0.44	1.00	—	—	—	—
NO	0.34	0.69	1.00	—	—	—
NO_2	0.11	0.53	0.55	1.00	—	—
PM_{10}	−0.17	0.21	0.14	0.59	1.00	—
O_3	−0.33	−0.48	−0.56	−0.31	0.29	1.00
Upland	**PN**	**CO**	**NO**	**NO_2**	**PM_{10}**	**O_3**
PN	1.00	—	—	—	—	—
CO	0.63	1.00	—	—	—	—
NO	0.66	0.84	1.00	—	—	—
NO_2	0.08	0.52	0.51	1.00	—	—
PM_{10}	−0.19	0.19	0.19	0.68	1.00	—
O_3	−0.54	−0.26	−0.35	0.26	0.41	1.00
Glendora	**PN**	**CO**	**NO**	**NO_2**	**PM_{10}**	**O_3**
PN	1.00	—	—	—	—	—
CO	0.00	1.00	—	—	—	—
NO	0.30	0.29	1.00	—	—	—
NO_2	0.07	0.50	0.47	1.00	—	—
PM_{10}	−0.18	0.33	0.04	0.56	1.00	—
O_3	−0.31	0.17	−0.24	0.18	0.56	1.00

The degree of correlation between 24-hr concentrations of PM_{10} and gaseous co-pollutants also is variable between sites. While PM_{10} is moderately ($r = 0.34$) associated only with NO_2 at the source site of Long Beach, its degree of correlation with O_3 increases substantially at the receptor sites, with *r* varying from 0.29 in Mira Loma to 0.56 in Glendora, thus reflecting the impact of secondary processes in PM formation. The impact of traffic sources on PM_{10} concentrations in every site also is indicated by the moderate to high correlations between PM_{10} and either CO or NO_2, with *r* values ranging from 0.32 between PM_{10} and CO in Riverside to 0.68 between PM_{10} and NO_2 in Upland.

SUMMARY AND CONCLUSIONS

Ultrafine particles, which dominate the particle number concentration of ambient aerosols, recently have been the focus of several health studies. It has been hypothesized traditionally that these particles originate from vehicular emissions; thus, the concentrations of gases such as CO, NO, or NO_2 that also originate from traffic sources can be used as a surrogate measures of ultrafine PM. The advantage of this approach is that concentrations of these gases are monitored routinely in compliance networks and on personal levels by means of relatively simple and easy-to-use monitors. The validity of the assumption using gases as surrogates of ultrafine PM was tested in this study in five sites of the Los Angeles Basin over the course of one calendar year.

The results presented in this paper indicate that there are overall weak to moderate associations for both hourly and 24-hr average concentrations between PN and other co-pollutants when considered for the whole calendar year of 2002. Results suggest that, at least in Los Angeles, gaseous co-pollutants cannot be used as surrogates of PN to assess human exposure. Comparing particle number concentrations between sites also showed low to modest spatial correlations, a result that further complicates monitoring for ultrafine particles either for regulatory purposes or in support of epidemiologic studies.

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