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Ambient Fine and Ultrafine Particle Measurements and Their Correlations with Particulate PAHs at an Elementary School Near a Highway

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ABSTRACT

Ambient particulate matter (PM) and particle-bound polycyclic aromatic hydrocarbon (PAH) concentrations were measured continuously for 70 days at a Korean elementary school located near a highway. The PM_{10}, $PM_{2.5}$, and PM_1 values were measured with a light-scattering, multi-channel, aerosol spectrometer (Grimm, Model 1.107). The number concentrations of the particles were measured using a scanning mobility particle sizer and counter (SMPS+C) which counted particles from 11.1 to 1083.3 nm classified in 44 channels. Particle-bound PAHs were measured with a direct reading, photoelectric aerosol sensor. The daily NO_2, SO_2, and CO concentrations were obtained from a national air-monitoring station located near the school. The average concentrations of PM_{10}, $PM_{2.5}$, and PM_1 were 75.3, 59.3, and 52.1 $\mu g/m^3$, respectively. The average number concentration of the ultrafine particles (UFPs) was 46,307/cm^3, and the averaged particle-bound PAHs concentration was 17.9 ng/m^3 during the study period. The ambient UFP variation was strongly associated with traffic intensity, particularly peak concentrations during the traffic rush hours. Particles <100 nm corresponded to traffic-related pollutants, including PAHs. Additional long-term monitoring of ambient UFPs and high-resolution traffic measurements should be carried out in future studies. In addition, transient variations in the ambient particle concentration should be taken into consideration in epidemiology studies in order to examine the short-term health effects of urban UFPs.

Key words: Ultrafine particles, Polycyclic aromatic hydrocarbons, Particulate matter, Traffic-related pollutants, Highway

1. INTRODUCTION

Epidemiological studies have demonstrated that airborne particulate matter (PM) is associated with adverse health outcomes, including increased respiratory and cardiovascular illnesses and hospitalization rates (Pope and Dockery, 2006; Brunekreef and Forsberg, 2005; Pope *et al.*, 2002; Dockery, 2001). However, it is unclear which PM properties are responsible for the health effects. Ambient PM is composed of heterogeneous compounds varying in size, number, chemical composition, surface area, concentration, and source (Brook *et al.*, 2004). It has been suggested that ambient PM size is an important factor in toxicity, with biological activity increasing as the particle size decreases (Oberdorster, 1996).

Ambient particles are measured as PM mass and classified according to aerodynamic diameter into the following size fractions: particles <10 μm (PM_{10}), coarse thoracic particles of 2.5 to 10 μm ($PM_{2.5\text{-}10}$), and fine particles <2.5 μm ($PM_{2.5}$) or <1 μm (PM_1). Ultrafine particles (UFPs) are receiving increasing interest in the scientific community due to their small size, <0.1 μm and potential health effects. Traffic-related emissions are the primary source of ambient urban fine and ultrafine particles (Fine *et al.*, 2004; Zhang *et al.*, 2004). Particles emitted from vehicles are <0.1 μm in size and are the major contributor to urban particulate pollution (Kittelson, 1998). In addition, they can increase allergic sensitization (Diaz-Sanchez *et al.*, 2000; Diaz-Sanchez *et al.*, 1999). Several studies have shown that a large proportion of urban UFPs consist of primary combustion products from traffic emissions such as diesel exhaust particles which include organic compounds, elemental carbon, soot, and metals (Kim *et al.*, 2002; Bockhorn, 2000; Cass *et al.*, 2000).

UFPs correspond to nucleation and Aitken modes and are typically fresh combustion emissions, of which

vehicle engines are the primary source in urban areas (Sioutas *et al.*, 2005). Particles emitted from vehicles are the result of gas phase species nucleation, forming condensed phase species in newly formed particles that have had little chance to grow (*i.e.*, nucleation mode) or in recently formed particles that are actively undergoing coagulation (*i.e.*, Aitken mode) (Araujo and Nel, 2009). UFPs are likely more toxic than larger particles (Li *et al.*, 2003; Donaldson *et al.*, 2002) because of their size. The larger numbers and surface area per unit mass of UFPs are important toxicologically due to the enhanced tissue penetration and oxidative stress that induces cellular damage (Oberdorster *et al.*, 2005; Li *et al.*, 2003). In addition, urban UFPs have high concentrations of toxic substances, including soluble metals, polycyclic aromatic hydrocarbons (PAHs), and quinines, all of which contribute to inflammation and oxidative stress (Li *et al.*, 2008; Nel *et al.*, 2006).

High ambient PAH concentrations are associated with increased urban vehicle traffic volumes because most PAHs are generated from incomplete combustion (Sapkota and Buckley, 2003; Levy *et al.*, 2001). Ambient PAHs are partitioned between the particulate and gas phase, primarily as fine and sub-micrometer particles $<$1-3 μm in size (Kaupp and McLachlan, 2000), and, therefore, they can penetrate deep into the bronchial system.

Epidemiologic panel studies have evaluated the short-term effects of temporal environmental concentrations on health outcomes. Ambient particles may peak during rush hour when rapid changes occur in the concentration of vehicle exhaust particles in ambient urban air (Zhu *et al.*, 2002). Continuous, real-time PM monitoring is necessary in order to characterize transient variations in atmospheric PM and to understand the effects of these variations on human health. Studies based on continuous measurements of particle size distribution have indicated that UFP concentrations are positively correlated with traffic volume (Buonocore *et al.*, 2009; Fruin *et al.*, 2008), nitrogen oxide (Kwasny *et al.*, 2010; Noble *et al.*, 2003), and black carbon (Gonzalez *et al.*, 2011), with different size distributions and temporal trends (Kim *et al.*, 2002; Zhu *et al.*, 2002) in different urban areas. However, few studies have evaluated airborne UFP concentrations in Korean urban centers (Park *et al.*, 2008).

This study presents the results of continuous particle and gaseous air pollutant measurements in an urban area in order to determine the temporal variation in ambient fine PM, UFP, and particle-bound PAH (p-PAH) concentrations at an urban elementary school in Korea. We examined the diurnal variation in particle concentrations in relation to traffic patterns and assessed their correlations with other pollutants, especially p-PAHs that originate from vehicle emissions. The results of this study provide information on the potential exposure of schoolchildren to particulate pollutants.

2. MATERIALS AND METHODS

2.1 Monitoring Site

Monitoring was conducted on the roof of a four-story elementary school (37° 28′05″N, 126° 40′41″E) in Namgu, a mixed commercial and residential area of the city of Incheon, South Korea. The school was located near a main road and approximately 200 m from a six-lane highway with an average traffic volume of 98,000 vehicles/day. The site was also located within 5 km of the Incheon industrial complex. Therefore, traffic and plant operations were considered the main ambient air pollution sources.

2.2 Measurements

The real-time concentration of particles was measured continuously for 67 days from March 31 to June 6, 2009 at the fixed monitoring site. Particulate matter mass was measured with a multi-channel (31 sizes, 0.25-32 μm) aerosol spectrometer (Model 1.107, Grimm Aerosol Technik, Germany), which sampled the particles at a rate of 1.2 L/min using a laser light scattering optical particle counter. The particle counts from each of the channels were converted to size categories of PM_{10}, $PM_{2.5}$, and PM_1 every 5 min. The daily gravimetric PM measurements were collected in parallel (45 to 50 days) by low-volume (16.7 L/min for 23-hr) particulate samplers (FH95, Thermo, Germany) for comparison to the continuous light-scattering measurements. The PM mass was collected using Teflon filters (47 mm PTEF, Zefluor, 2 μm pore size) with a flow rate of 16.7 L/min for 23 hr (09:00-08:00) and weighed using a micro-balance (Satorious, 0.01 mg reading precision). We found that the measurement from the aerosol spectrometer was in excellent agreement with the PM filter-based measurements ($r^2 >$ 0.98).

The number concentrations of the particles were measured using a scanning mobility particle sizer and counter (SMPS+C) composed of a differential mobility analyzer (DMA; Grimm model 5.5-900) and a condensation particle counter (CPC; Grimm Model 5.400), which counted particles between 11.1 to 1083.3 nm in size and classified them into 44 channels by scanning every 7 min. The instruments can count up to 10^7 particles/cm^3 with 10% accuracy when operated at a 0.3

L/min aerosol and a 3 L/min sheath flow rate. Data were collected with software provided by the manufacturer, which also allows for correction of losses in the system. The UFP number concentration was the sum of 25 channels from between 11.1 and 101 nm in size ($NC_{0.01\text{-}0.1}$). These sizes represented the characteristics of the particles most likely to be produced by vehicle traffic. The accumulation mode particle number $NC_{0.1\text{-}1}$ was calculated by adding the 18 channels between 111 and 930 nm in size, representing a large portion of the sub-micrometer particle mass. Particle number concentrations from a series of six days with <100 measurements and/or those collected for less than 12 hours each day were excluded. Particle-bound PAHs were measured with a photoelectric aerosol sensor direct reading instrument (PAS 2000, EcoChem Technologies) which detected particulate PAHs by measuring the electrons emitted by organic molecules on particles irradiated by ultraviolet light. The instrument is useful for measuring four or more ring PAHs adsorbed to a particle having an aerodynamic size greater than 1 μm with a nominal detection limit of 3 ng/m^3 (Cowen *et al.*, 2001; Burtscher, 1992). However, previous studies have shown that gas-phase PAHs with two to four rings are not detected by the PAS (Ramamurthi and Chuang, 1997).

Daily NO_2, SO_2, and CO concentrations were obtained from a national air-monitoring station located 2 km east of the school. We used 1-hr averages for the continuous measurements. Diurnal variation was assessed for both weekdays and weekends in order to examine the effects of traffic on the particulate concentrations. The Pearson correlation coefficients were used to analyze the relationship between the ambient PM and p-PAHs and the gaseous pollutants.

3. RESULTS AND DISCUSSION

3.1 Mass and Number Concentrations of Particles

The air pollutants data are summarized in Table 1. During the study period, the 1-hour average concentrations (mean $\pm$ SD) of PM_{10}, $PM_{2.5}$, and PM_1 were 75.3 $\pm$ 43.5, 59.3 $\pm$ 34.4, and 52.1 $\pm$ 31.6 μg/m^3, respectively. Their variations were relatively high because the hourly peak and lowest values were included. Daily PM_{10} levels greater than 100 μg/m^3 were observed for ten of the 70 days of the study period. $PM_{2.5}$ and PM_1 accounted for 79% and 68% of the PM_{10} mass concentration, respectively. The 1-hour average $NC_{0.01\text{-}0.1}$, and $NC_{0.01\text{-}0.05}$ were 46,307 $\pm$ 34,644 and 37,915 $\pm$ 30,303 cm^3, respectively. The average P-PAHs concentration was 17.9 $\pm$ 13.8 ng/m^3 during the study period. The highest particle number was observed in the UFP fraction range of 0.01 to 0.1 μm (Fig. 1). The UFPs accounted for 83% of the total particle number concentration.

The results of our study showed relatively high air pollutant levels. The particle number concentrations and PM_{10} and $PM_{2.5}$ values were somewhat higher than levels reported in other areas of Korea (Park *et al.*, 2008) and much higher than the levels reported in European cities (Timonen *et al.*, 2004; Pekkanen *et al.*, 2002). High ambient particle concentrations might have been emitted from the highway near the monitoring site. The p-PAH concentration was not much greater than the PAS measurements in other urban centers, *e.g.*, 29 ng/m^3 in Tokyo, Japan; 52 ng/m^3 in Bangkok, Thailand (Chetwittayachan *et al.*, 2002); and 19 to 92 ng/m^3 in Basel, Switzerland (Junker *et al.*, 2000). Although PAS measurements are limited to the PAH concentration in particles, Chetwittayachan *et al.* found that PAS

Table 1. Means of the 1-hour average concentrations of air pollutants.

	No. of hour	Mean	SD[a]	Percentile			Minimum	Maximum
				25th	50th	75th		
PM^b_{10} (μg/m^3)	1597	75.3	43.5	44.2	63.9	95.2	5.4	271.0
$PM_{2.5}$ (μg/m^3)	1597	59.3	34.4	34.5	50.5	77.2	4.4	212.4
PM_1 (μg/m^3)	1597	52.1	31.6	29.6	43.8	69.1	3.4	191.1
$NC^c_{0.1\text{-}0.5}$ (1,000/cm^3)	1465	7.349	4.569	3.662	6.076	10.218	0.481	25.752
$NC_{0.01\text{-}0.1}$ (1,000/cm^3)	1465	46.307	34.644	21.667	35.979	61.889	2.222	263.426
$NC_{0.01\text{-}0.05}$ (1,000/cm^3)	1465	37.915	30.303	16.584	28.982	50.158	1.377	244.224
P-PAHs[d] (ng/m^3)	1597	17.9	13.8	8.4	14.4	23.1	<3	99.5
NO_2 (ppm)	1478	0.033	0.018	0.02	0.029	0.041	0.001	0.113
SO_2 (ppm)	1478	0.01	0.006	0.006	0.009	0.012	0.001	0.059
CO (ppm)	1478	0.7	0.3	0.5	0.6	0.8	0.1	2.6

[a]SD, standard deviation
[b]PM, particulate matter (numbers represent diameter of particles in μm)
[c]NC, number concentration of particles (numbers represent diameter of particles in μm)
[d]P-PAHs, particle-bound PAHs concentration

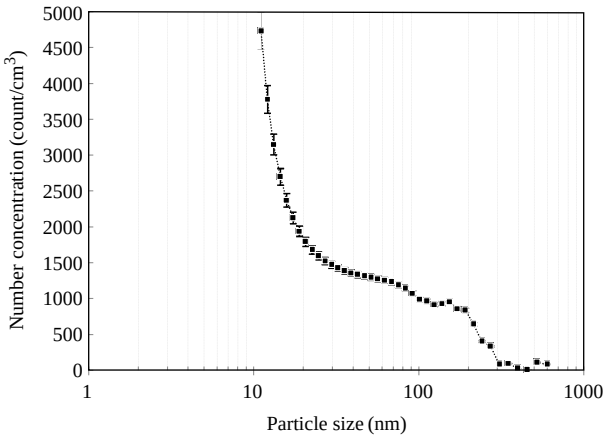

Fig. 1. Average number concentrations (±standard error) of particles from 10.1-1,000 nm in size measured over 64 days.

level exhibited a strong correlation (r^2=0.79) with the total concentration of 11 carcinogenic PAHs as determined by gas chromatography/mass spectrometry (Chetwittayachan *et al.*, 2002).

3.2 Diurnal Trends

Fig. 2 shows the diurnal variations in the gaseous and particulate pollutants. The plot shows the typical traffic-related time-series patterns for the particle measurements. Two peaks, associated with the morning and evening rush hours, were observed in the UFP and p-PAH measurements. The UFP number concentration peaked between 06:00 and 09:00 and then again between 18:00 and 22:00. The diurnal pattern of the p-PAHs was similar to that of the UFP. The concentration of the PAHs remained elevated throughout the morning rush hour, decreased in the afternoon, and increased in the evening. The evening UFP and PAH concentrations continued to rise until the late evening. This might be due to the late evening rush hour (22:00) that commonly occurred on the highway near the monitoring site (KEC, 2004). From 22:00 until midnight, the concentrations of UFP and p-PAHs started to decrease due to decreased traffic activity. The PM mass fractions of PM_{10}, $PM_{2.5}$, and PM_1 had the highest concentrations during the morning rush hour but did not increase as much as the UFPs in the evening.

The pollutants, especially the UFPs and PAHs, decreased on the weekends. The PM mass fraction diurnal trends did not differ between the weekdays and weekends, whereas the UFPs and p-PAHs had different patterns on the weekdays than on the weekends. On the weekends, the variations in the UFP and p-PAH did not show evening rush hour peaks. Fig. 3 shows the diurnal trends of the ambient particles and p-PAHs

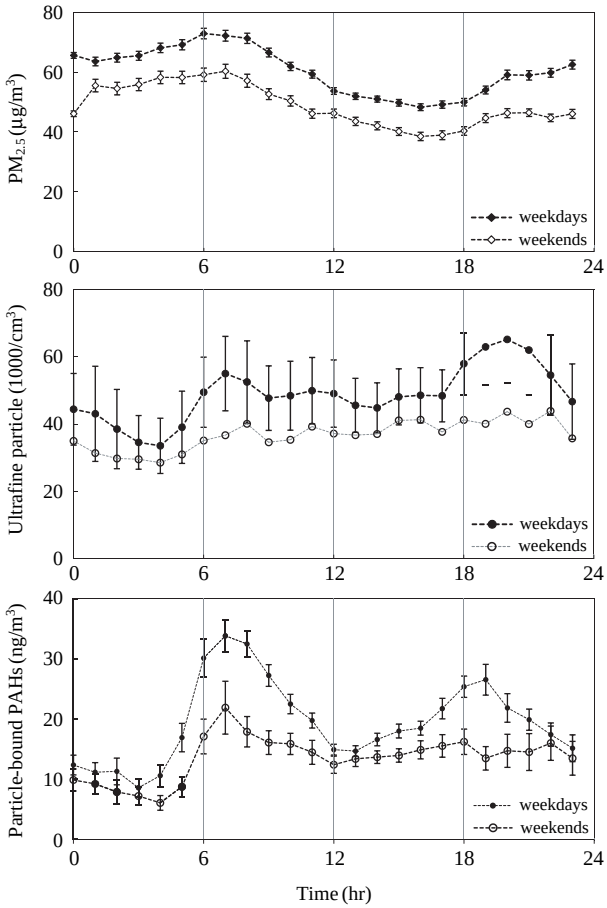

Fig. 2. Hourly mean concentrations (±standard error) of ambient particles and p-PAHs on weekdays and weekends.

concentration on the highest and lowest PAH pollutions day. On the highest PAHs pollution day (Friday), the diurnal pattern of the p-PAHs was similar to that of the UFP and PM_1, showing the typical two peaks during the morning and evening rush hours. However, these similar patterns were not observed on the lowest PAHs pollution day (Sunday).

Normally, traffic emissions are the primary source of urban ambient fine PM pollution (Fine *et al.*, 2004). The fresh particles emitted from vehicles have short life spans as they coalesce with larger particles. In addition, the number size distribution was altered by the aerosol dynamical processes, including dilution with clean air, polluted air entrainment, vapor condensation, and particle coagulation (Wehner *et al.*, 2002). The results of previous epidemiological studies have indicated that short term (1 hr) exposure to DEPs could produce a pulmonary inflammation response in healthy subjects (Salvi *et al.*, 2000; Salvi *et al.*, 1999). Therefore, 24-hr average data might underestimate the exposure-related adverse health effects. However, more

Table 2. Pearson correlation coefficients among the ambient pollutant concentrations.

	PM_{10}	$PM_{2.5}$	PM_1	$NC_{0.1\text{-}0.5}$	$NC_{0.01\text{-}0.1}$	$NC_{0.01\text{-}0.05}$	P-PAHs	NO_2	O_3	SO_2	CO
PM_{10}	1										
$PM_{2.5}$	0.97	1									
PM_1	0.96	1	1								
$NC_{0.1\text{-}0.5}$	0.77	0.78	0.77	1							
$NC_{0.01\text{-}0.1}$	0.47	0.45	0.45	0.86	1						
$NC_{0.01\text{-}0.05}$	0.43	0.42	0.41	0.83	1	1					
P-PAHs	0.38	0.4	0.4	0.55	0.47	0.44	1				
NO_2	0.78	0.77	0.76	0.69	0.45	0.41	0.6	1			
SO_2	0.86	0.82	0.81	0.78	0.63	0.59	0.46	0.77	0.08	1	
CO	0.83	0.81	0.8	0.7	0.44	0.39	0.56	0.85	−0.17	0.82	1

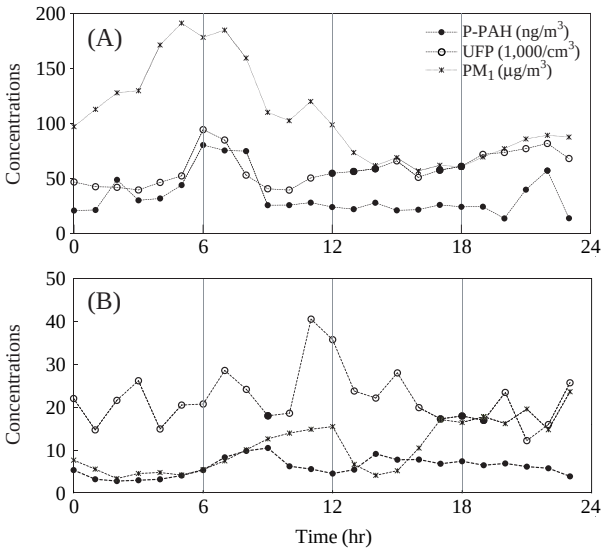

Fig. 3. Hourly mean concentrations of ambient particles and p-PAHs on a highest (A) and lowest (B) PAH pollution day.

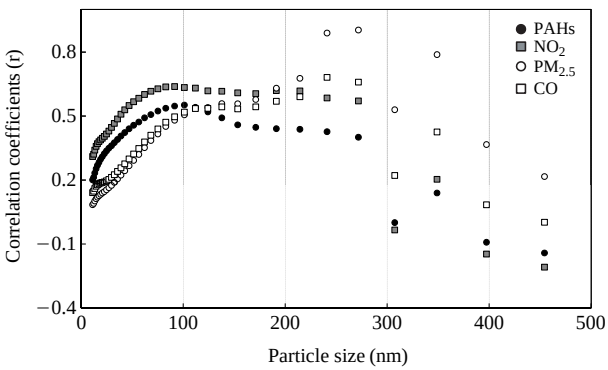

Fig. 4. Pearson correlation coefficients among the 1-hr mean concentrations of $PM_{2.5}$, p-PAHs, NO_2, CO and particle number in 38 size classes (10-454 nm).

research is needed to explore the relationships between various sizes of UFPs and traffic volume or other traffic-related pollutant proxies.

3. 3 Pollutants Concentration Correlations

Ambient particles and gaseous pollutant concentrations were intercorrelated. The Pearson correlation analysis showed a high correlation ($r > 0.95$) among PM_{10}, $PM_{2.5}$, and PM_1. In addition, $PM_{2.5}$ was significantly ($p < 0.001$) correlated with NO_2 ($r=0.77$), SO_2 ($r=0.82$), and CO ($r=0.81$). The UFPs were moderately ($p < 0.05$) correlated with PM_{10} ($r=0.47$), $PM_{2.5}$ ($r=0.45$), NO_2 ($r=0.46$), SO_2 ($r=0.63$), and CO ($r=0.44$).

Fig. 4 illustrates the correlation coefficients of the hourly mean concentrations of 38 particle size classes from 10 to 454 nm and p-PAHs. The correlation coefficients for UFPs with p-PAHs and NO_2 were higher than the correlation of the UFPs with $PM_{2.5}$ and CO. The particle number correlation coefficients with $PM_{2.5}$ and CO increased in relation to particle sizes up to 300 nm. The correlation coefficients between particle number concentration and the p-PAHs and NO_2 remained elevated in relation to particle size up to 100 nm and decreased for larger particle sizes. This result suggests that the particle number concentration of 100 nm particles originated predominantly from traffic emission particles, DEPs in particular.

With respect to human health, UFPs from diesel exhaust have mucosal adjuvant properties and can exacerbate allergic inflammation, even turning a harmless neoantigen into an allergen that induces allergen-specific IgE (Diaz-Sanchez *et al.*, 2000; Diaz-Sanchez *et al.*, 1999; Nel *et al.*, 1998). Studies using a rat model suggest that ambient UFPs act as adjuvant substances for allergic sensitization, probably due to their high oxidant potential (Li *et al.*, 2009; de Haar *et al.*, 2006).

4. CONCLUSIONS

The results of our study showed relatively high air pollutant levels at an elementary school located near a highway. In the daily change profiles of the measured

particulate pollutants, UFPs and p-PAHs showed clearer, higher peak concentrations in the morning and evening compared to the PM mass fraction, indicating that they were strongly related to the high traffic rates during the morning and evening rush hours. Throughout the day, the ambient particle variation showed marked peaks that could lead to high acute exposure, especially during the morning and evening rush hours. These fresh particles emitted from vehicles have short life spans as they coalesce with larger particles. Therefore, 24-hr average data might underestimate the exposure-related adverse health effects.

In the correlation analysis, the correlations of size-resolved particles with particle-bound PAHs and other gaseous pollutants depended on particle size. The correlation coefficients between size-resolved particle concentrations and traffic-related pollutants, such as p-PAHs and NO_2, increased up to 100-nm particle sizes and declined with larger particles. This result suggests that the particle number concentration of 100 nm particles originated predominantly from traffic emission particles, DEPs in particular.

In conclusion, ambient UFP variation is strongly associated with traffic intensity, particularly peak concentrations occurring during the traffic rush hours. Particles < 100 nm corresponded to traffic-related pollutants, including PAHs. Additional long-term monitoring of ambient UFPs and high-resolution traffic measurements should be explored in future studies. In addition, transient variations in the ambient particles concentrations should be taken into consideration in future epidemiology studies in order to examine the short-term health effects of urban UFPs.

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