Chapter 18

Ultrafine Particles on and Near Roadways

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18.1 Introduction

Ultrafine particle (UFP) refers to discrete particles with diameters less than 100 nm in ambient air that exist either as solid particles or liquid droplets. These particles originate from many different stationary and mobile sources [1] as well as from photochemical production as a nucleation event, whereby new particles are formed in the atmosphere from gaseous precursors [2]. The size, chemical compositions, and other physical and biological properties of UFPs are highly variable from place to place owing to the differences in the weather, seasonal patterns, and sources. UFP properties are also influenced by changes they undergo in the atmosphere. A complete description of the atmospheric UFPs would include an accounting of the chemical composition, morphology, and size of each particle and the relative abundance of each particle type as a function of particle size. However, the physical and chemical characteristics of particles are usually measured separately. Size distributions by particle

number, used to calculate surface area and volume distributions, often are determined by physical means, such as electrical mobility of suspended UFPs. Chemical composition usually is determined by the analysis of collected samples although some species can be measured in situ. Although UFPs dominate the atmospheric particle number concentrations, they amount to a small contribution of atmospheric particle mass concentrations. A given mass of UFPs, however, has 100-1000 times more surface area than an equal mass of fine particles (0.1 μ m < diameter < 2.5 μ m) and approximately 10⁵ times more surface area than an equal mass of coarse particles $(2.5 \, \mu \text{m} < \text{diameter} < 10 \, \mu \text{m})$ [3], which may significantly affect their relative toxicity and reactivity. Because of their increased number and surface area, UFPs are particularly important in atmospheric chemistry and environmental health. This chapter focuses on UFPs from vehicle emissions, which usually constitute the largest source of UFPs in an urban environment.

18.2 **Health Effects of Ultrafine Particles**

Epidemiological studies all over the world have consistently linked increases in particulate matter (PM) exposure to increases in mortality and morbidity [4, 5]. More complex is the question of which PM components or attributes are most important in causing these health effects, among which UFP is a likely candidate. Certain toxicological investigations suggest that atmospheric UFPs may be responsible for some of these adverse effects. The thinking is that UFPs can easily find their way from the lungs into the bloodstream and then lead to systemic inflammatory changes [6]. Although epidemiological studies to date have not established whether it is particle mass, surface area, or number concentrations that may be responsible for these observed health outcomes attributable to PM [7–10], some suggestive evidence from epidemiological studies points to UFPs being related to respiratory health endpoints [9].

18.2.1 **Toxicological Studies**

Depending on particle size and shape, UFPs deposit in various regions of human respiratory system by the complex action of several aerosol deposition mechanisms [11]. UFPs' deposition efficiency in the respiratory system increases significantly as particle size decreases [12, 13]. The larger surface area per unit mass of UFPs means they can transport relatively more toxic surface adsorbed materials than larger particles of the same mass. The recent recognition that UFPs are more toxic when inhaled than larger particles suggests that their ability to be absorbed into tissues and the circulation, and their greatly increased surface area, might be important factors in determining the cardiopulmonary toxicity [14]. The inflammatory impact correlates closely with the particle surface area. Particles of diameter 20-50 nm have almost twice the inflammatory potency compared to particles of 200-500 nm diameter [6, 14-18]. As a result of their small size and large surface area, UFPs have greater pulmonary deposition efficiency among asthmatics [19] and crosscellular walls and localize in the mitochondria [20]. Vehicularemitted UFPs near roadways are particularly rich in redox active chemicals that lead to systemic inflammation [21].

Epidemiological Studies 18.2.2

It should be noted that relatively few population studies have been published on the health effects of UFPs. The first epidemiology study was published only a few years ago, which is in contrast to the hundreds of epidemiology studies on PM₁₀ and PM_{2.5} published over the last two decades. With improvement of particle measurement technologies, the adverse health effects of PM become stronger when smaller particle sizes are considered [22]. A daily mortality study in Ehrfurt, Germany, was the first epidemiology study that examined and found significant associations between exposure to UFPs and mortality from respiratory and cardiovascular disease. Recently epidemiological studies, dealing with short-term effects in adults and children with asthma and daily mortality, have addressed the role of UFPs [8, 9, 23, 24]. These studies concluded that health effects were more closely associated with the number of UFPs than the mass of the fine particles.

Vehicle-Emitted Ultrafine Particles 18.3

Emission inventories suggest that the majority of fine and UFPs found in the urban atmosphere come from engine combustions [25-27]. Emissions from motor vehicles have substantially changed

over the last decade because of new fuels, changed engine designs. and improved emission-control technology. Studies of the health effects associated with exposure to motor vehicle exhaust are further complicated by the changing nature of emissions over time. There is a growing concern over the impact of UFPs emitted from diesel vehicles on human health. Diesel exhaust has been declared a probable human carcinogen by the US Environmental Protection Agency, the International Agency for Research on Cancer, the World Health Organization, and the US National Institute of Occupational Safety and Health. The state of California designated it as a toxic air contaminant. Most health effects research on diesel emissions has focused on their possible contribution to lung cancer. Recently, concerns have also been raised about the potential effect of diesel exhaust on enhancing human allergic responses and exacerbating asthma. It is less well recognized that gasoline-powered vehicles also emit relatively high concentrations of UFPs. This is particularly evident with the direct injection gasoline engine, which shares some important design features with the diesel engine. The mass of particles emitted from diesel vehicles is regulated by legislation. The number of particles emitted is not regulated for any type of vehicle in the US, but may be an important indicator of the health impacts. UFPs are believed to penetrate deeper into the lungs and to stay there longer than larger particles. The mass of one particle with a diameter of 3 µm is equal to the mass of 1 million particles with a diameter of 0.03 μm. Most particles from gasoline engines fall below 0.1 µm. This is why they contribute little to the mass of particles collected during a conventional emissions test.

UFPs from vehicular exhaust typically exhibit a bi-modal size distribution with a primary mode between 10 and 30 nm, and a secondary mode between 50 and 70 nm (Fig. 18.1) [28–30]. The primary mode has been found to exist in significant amounts only in-cabin and on or near freeways [31, 32]. Also shown in Fig. 18.1 are the modal diameters (μ_g), geometric standard deviations (σ_g), and typical particle morphologies for each mode [33]. The size distribution of UFPs evolves rapidly when moving away from the source or penetrating into the in-cabin and indoor environments through atmospheric diffusion, coagulation, filtration, deposition, condensation/evaporation, and other particle dynamic processes [32, 34–36].

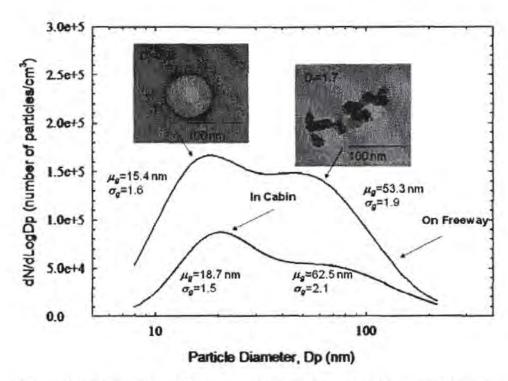


Figure 18.1 Typical on-freeway and in-cabin ultrafine particle size distributions and morphology (data source: Zhu et al. (2007) [31] and Barone and Zhu (2008) [33]).

18.4 Spatial Profiles of Ultrafine Particles Near Roadways

Time spent in traffic and living close to roadways have been linked to adverse respiratory and cardiovascular health [37–40]. Children living near high traffic flows have six times the odds of developing all types of cancer [41]; have increased risk of asthma [39, 42], and have a higher prevalence of most respiratory symptoms [43, 44] than those living farther away or near lower traffic flows. The observed risk was greatest for children living within 100 m of a road [40, 45]. Since the majority of particle number generated by vehicle exhaust have been reported in the size range of 20–130 nm for diesel engines [29] and 20–60 nm for gasoline engines [46], it is important and necessary to quantify UFP emission levels, and to determine UFP behavior after emission as they are transported away from the emission source — busy roads and freeways, into in-cabin and indoor environments where actual human exposure occurs.

Very high concentrations of UFPs and sharp concentration gradients were reported within 100 m downwind of freeways [32, 47]. Meteorological conditions seem to be the most important factor influencing such decay [32, 35, 47–49]. UFP concentration decay profiles observed during the day (red lines with open circles) and at nighttime (blue lines with solid circles) were on different side of the freeway due to reversed wind directions (Fig. 18.2). The lower wind speed and less atmospheric dispersion at night make UFPs decay at a slower rate. In contrast to daytime, where UFPs at 300 m downwind of the freeway were comparable to those measured 300 m upwind of the freeway, nighttime downwind particle number concentrations at 500 m were still significantly greater than those measured at the upwind site. This implies that freeway emissions have a much broader effect on local air quality at night.

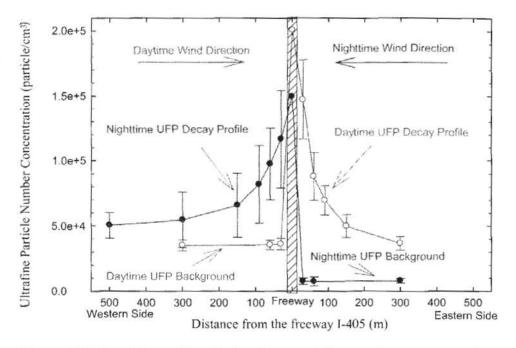


Figure 18.2 Spatial profile of ultrafine particle number concentrations near the I-405 freeway during day and night (data source: Zhu et al. (2002) [32] and Zhu et al. (2006) [48]). See also Color Insert.

Besides a sharp decay of number concentrations, UFP size distributions also change when one moves away from freeways. For example, number concentrations for smaller particles, dp < 50 nm, dropped significantly with increasing distances from the freeway, but for larger ones, dp > 100 nm, number concentrations decreased

only slightly [32]. This suggests besides atmospheric dispersion, particle dynamics such as coagulation, deposition, evaporation, and condensation are also important for UFP transformation near roadways.

It should be noted, nighttime traffic volume was 25% of daytime volume but generated about 80% of the daytime particle number concentration (Fig. 18.2). Lower wind speed and weaker atmospheric dilution alone could not explain this observed discrepancy because carbon monoxide (CO) concentrations at 30 m downwind of the freeway at night (0.5 ppm) were about 25% of what observed during the daytime (2.0 ppm) [48]. Besides the dilution effect, the lower temperature at night may also contribute to this observed discrepancy. Colder ambient temperatures contribute to significantly increased nuclei mode particle formation in vehicle exhaust sampled on the freeway and in dilution tunnel experiments [50]. This observation suggests that there could be significant differences in the formation of UFPs between day and night. Higher particle number concentrations were also observed in winter than in summer near the I-405 freeway [49].

UFP morphologies also change downwind from roadways. The fraction of aggregates was found to be significantly greater on-freeway than 90 m downwind [33]. Since aggregates are a primary aerosol (directly emitted), this may indicate that secondary aerosol (formed in the atmosphere) becomes more prominent with increasing distance from the freeway. The result is consistent with a previous study in Plymouth, UK, showing larger fractions of aggregates near the roadway than in urban background [51]. The fraction of total aerosol present as particles with multiple inclusions was significantly less on-freeway than 90 m downwind [33]. The increase in particles with multiple inclusions with increasing distance from the freeway suggests that dilution does not prevent particles from colliding and merging. Thus, coagulation may play a role in altering the number size distribution and deserve further investigation near roadways.

18.5 **Ultrafine Particles on Freeways**

Typical time-resolved UFP size distributions measured on two freeways are shown as contour plots in Fig. 18.3, where x-axis

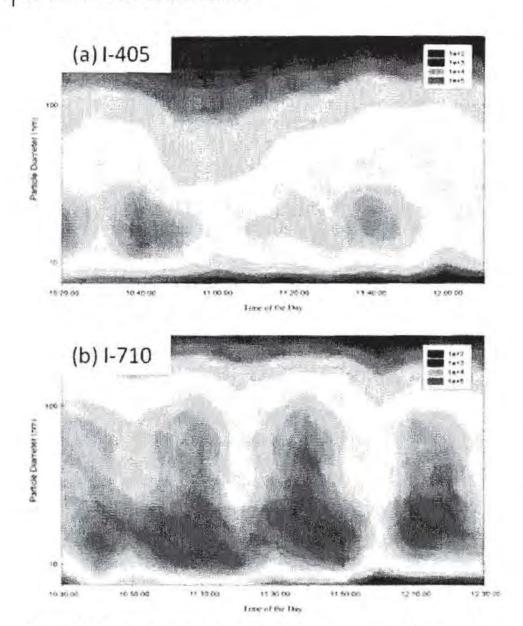


Figure 18.3 Typical contour plot of ultrafine particle number based size distribution on (a) a gasoline-vehicle-dominated freeway (I-405) and (b) a heavy-duty diesel-truck-dominated freeway (I-710) (data source: Zhu et al. (2008) [52]). See also Color Insert.

presents the time at which data were collected, y-axis represents the particle size in log scale, and the color intensity indicates normalized particle number concentration (dN/dLogDp) for a given size at a given time [52]. Same concentration scale was used for both freeways. Traffic on the I-405 freeway was dominated by gasoline-powered cars and light trucks, with less than 5% of vehicles being heavy-duty diesel trucks (HDDT), whereas there were more than

25% of HDDT on the I-710 freeway. Data were collected when the vehicle ventilation system was set to air conditioning on and recirculation off. Previously we found vehicle ventilation settings affect the penetration characteristics of UFPs into vehicles [53]. The current ventilation settings were chosen to ensure passengers' comfort inside the vehicle. The same ventilation settings were used for both freeways. In general, particle number concentrations were lower on the I-405 freeway than on the I-710 freeway as indicated by more green color. For both freeways, hot spots usually occurs around 10-30 nm corresponding to a primary nuclei mode. There are periods between hot spots when particle number concentrations are lower on both freeways. This happens due to the fact that each 2 h time series consisted of two round trip loops on the freeway. The lower concentration periods correspond to the turning points at which the vehicle was driven off the freeways to use local streets to get back on the freeway in the opposite direction. In Fig. 18.3b, a second mode around 60-100 nm usually occurs suggesting bi-modal size distributions on the I-710 freeway. This is in agreement with what we observed inside passenger vehicles on the I-710 freeway previously [53]. On the I-405 freeway, the primary mode was also around 10-30 nm, but the second mode seems broader and less obvious.

18.6 **Ultrafine Particle Penetrating Into Indoor Environments**

To understand the impact of freeway-generated UFPs on indoor levels, we measured UFPs inside four apartments located within 300 m of the I-405 in southern California [36]. Data were collected simultaneously in and out of the apartments without indoor UFP sources (Fig. 18.4). Outdoor UFPs show a mode around 20 nm. No such mode was observed indoors where the UFP concentrations were much lower and more stable than outdoors. This indicates that UFP dynamics during the process of penetrating from outdoors to indoors are important in affecting indoor UFP levels. Lower indoor UFP levels were observed inside the apartment located 200 m away from the I-405 than the one located 50 m away. This indicates that the sharp UFP concentration spatial profiles near I-405 (Fig. 18.2) influence indoor UFP levels.

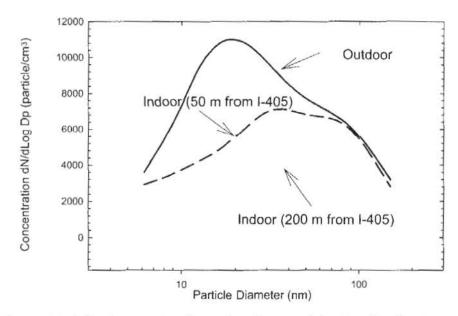


Figure 18.4 Outdoor and indoor ultrafine particle size distributions near the I-405 freeway during daytime (data source: Zhu et al. (2005) [35]).

Indoor-to-outdoor (I/O) ratios for UFP number concentrations depended strongly on particle size and on the nature of indoor ventilation mechanisms. Under infiltration conditions with air exchange rates ranging 0.31–1.11 h⁻¹, the highest I/O ratios (0.6–0.9) were usually found for larger UFPs (70–100 nm), while the lowest I/O ratios (0.1–0.4) were observed for UFPs of 10–20 nm.

18.7 Ultrafine Particle Penetrating Into In-Cabin

UFP concentrations observed on freeways are typically in the range of 100,000-500,000 particles cm⁻³, much higher than ambient background, which is usually on the order of 5000-50,000 particles cm⁻³ [54]. This means the highest human exposure to UFPs occurs while driving on roadways. In contrast to previously studied PM₁₀, PM_{2.5}, and gas-phase pollutants [55–57], UFP in-cabin levels were found to change greatly with respect to vehicle type and ventilation settings. The in-cabin microenvironment (\sim 6% of people's daily time budget) was estimated to contribute approximately 10-50% of people's daily exposure to UFPs from traffic [31].

UFPs measured inside passenger vehicles [31] and school buses [58] showed tremendous variability within and across vehicles

driven on various roadways (Table 18.1). Data were collected inside passenger vehicles and school buses of different ages on major California and Texas freeways with different traffic volumes and mixes. For the same vehicle, the highest UFP levels were observed on I-710 with the highest percentage of HDDT. Across vehicles, UFP levels were much lower inside newer vehicles driven on the same freeway.

Car age and ventilation conditions play major roles in the in-cabin commuter exposure to freshly emitted UFP. The average in-cabin to on-roadway (I/O) ratio was found around 50% under fan off and recirculation (RC) off. Switching to fan on RC on leads to a significant 1/0 ratio decrease to 20% due to less UFP exchange between incabin and outside. In addition, significant I/O ratio changes were associated with the ventilation airflow rate change.

Table 18.1 Average (change to ±STDEV) UFP concentrations inside different vehicles on various roadways with different traffic volume and mix (data source: Zhu et al. (2007) [31] and Zhang and Zhu (2010) [58]).

Vehicle type	Age	Freeway	Traffic volume (vehicles min ⁻¹)	HHDV (%)	UFP concentration (10 ³ particles cm ⁻³)
VW Jetta	5	I-110	100	0	25±7
VW Jetta	5	I-405	230	5	200 <u>+</u> 48
VW Jetta	5	I-710	200	25	290 <u>+</u> 67
PT Cruiser	1	I-110	100	0	22 <u>+</u> 5
PT Cruiser	1	I-405	230	5	85 <u>+</u> 19
PT Cruiser	1	I-710	200	25	150 <u>+</u> 34
School Bus	18	I-37	40	3	62 <u>+</u> 23
School Bus	2	I-37	40	3	18 <u>+</u> 8

18.8 Summary

Although UFPs represent 80% of atmospheric particles in terms of number concentrations, due to their small size, they are not captured by currently regulated PM mass concentrations (i.e., PM₁₀ and PM_{2.5}). In an urban environment, motor vehicle emissions usually

constitute the most significant source of primary UFPs. Very high levels of UFPs and sharp concentration decay profiles have been reported near roadways. It suggests that people, who live and work, near major traffic sources, will have much higher UFP exposure than those who live farther away from such sources. Significant amount of traffic-related UFPs can penetrate into indoor and in-cabin microenvironments where actual human exposure occurs. Particle size and ventilation settings are important factors determining the I/O ratios.

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Reviews

"At a time when we are concerned about the potential hazards of engineered nanomaterials, this book gives a timely and delightful overview of the variety of nanoparticles that exist in our environment — from noble metal nanoparticles in ore systems and nanodust in the solar system to magnetic nanoparticles in homing pigeons and photonic crystals in beetles. It is an enjoyable and a useful companion for anyone interested in knowing about the potential hazards of nanomaterials and those seeking inspiration from nature to create functional materials."

Dr Ai Lin Chu - Senior Editor, Nature Nanotechnology

Description

While man strives to synthesize and utilize functional nanomaterials, Nature's own laboratory has perfected the production of a range of highly selective nanomaterials. Natural nanomaterials (and even natural nanotechnologies) are all around us, which inevitably leads to these questions: What are these natural nanomaterials made of? Where can we find them? What can they do? Answering these questions will lead to a better understanding of the world around us and facilitate new and environmentally friendly ways of creating and manipulating nanoscale materials for the next generation of new technologies.

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