

Review of Measurement Methods and Compositions for Ultrafine Particles

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Abstract

Impactor, virtual impactor/aerosol concentrator, and aerodynamic lenses are used to separate the ultrafine particle (UP) fraction from other particle sizes for chemical analysis. Cascade impactors, such as the Micro-Orifice Uniform Deposit Impactor (MOUDI), are most commonly used in field studies, with sampling onto substrates amenable to different chemical analyses. Impactors need sufficient sampling flow rates and homogeneous deposits on the impaction surfaces for multiple chemical analyses. Mass, elements, ions, and carbon fractions can be measured on these substrates by several analytical methods. Specific organic compounds measured by solvent extraction require substantial mass loadings that can only be obtained by compositing samples from several measurement periods unless aerosol concentrators or high-volume sampling devices are used. Thermal desorption-gas chromatographic/mass spectrometry has potential to obtain organic speciation with small sample sizes. Studies of UP composition began in the late 1990s, with 25 ambient studies surveyed here. These are mostly from urban areas. Organic material, including polycyclic aromatic hydrocarbons (PAHs), usually constituted the most abundant portion of UP, with high elemental concentrations found near industrial sites. Much of the UP < 50 nm appears to be semi-volatile, consistent with it being composed by organic materials such as hopanes from engine oils or condensed secondary organic aerosol such as organic acids.

Keywords: Ultrafine particles; Chemical composition; Particle size distribution; Carbonaceous aerosol.

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INTRODUCTION

Ultrafine particles (UP) are loosely defined as those with diameters in the range of ~1 nm to 100 nm (nanometers). UP are bigger than air molecules (~0.3 nm), but smaller than the upper limits of PM_{2.5} or PM₁₀ (particles with aerodynamic diameters less than 2.5 and 10 micrometers [μm], respectively) regulated by the U.S. National Ambient Air Quality Standards (NAAQS; U.S.EPA, 2006). Although UP do not contribute large quantities to PM_{2.5} or PM₁₀ mass, they dominate the number concentration and most of the surface area. UP are produced by condensation of hot vapors in fresh combustion emissions, or form naturally when gases oxidize to compounds with lower vapor pressures and spontaneously nucleate or condense on other small particles (Kulmala *et al.*, 2004). Owing to their small sizes and high mobilities, UP diffuse rapidly and may combine with each other, with larger particles, and with nearby deposition surfaces within minutes to hours. UP may contain organic material, transition metals, sulfuric acid (H₂SO₄), and free radicals. UP often consist of volatile components at atmospheric temperatures, and the high curvature of the smallest particles (< 10 nm) favors evaporation over larger particles of the same composition (the Kelvin effect). Gases evaporated from small particles may re-condense on larger ones, thereby shifting the distribution toward larger particle diameters (Zhang and Wexler, 2002).

UP are often considered deleterious when they are inhaled or ingested into the human body (Oberdörster *et al.*, 1995). They penetrate to the lower parts of the lung (Daigle *et al.*, 2003) where their large numbers can defeat defensive mechanisms. Thereafter, they can transport through the bloodstream or lymphatic system to vital organs (Oberdörster *et al.*, 2004). However, UP can also be lifesavers. The same inhalation and transport properties can rapidly deliver medicines to specific locations. Iron (Fe) nanoparticles can be magnetically directed to specific locations without invasive procedures.

UP are produced by either pollution sources or for commercial applications and can result in adverse health effects in the ambient and workplace environments if they are not appropriately controlled (Biswas and Wu, 2005; Chow *et al.*, 2005c; Chow and Watson, 2006). Control methods are more complex and cost more than those for larger particles.

This review examines the state of knowledge concerning UP chemical composition. Specific objectives are to explain and evaluate measurement methods for UP chemical composition, summarize UP chemical composition, compare them among different times and places, and specify gaps and uncertainties in the current knowledge base regarding UP composition and potential methods of filling those gaps.

MEASUREMENT METHODS FOR ULTRAFINE PARTICLE CHEMICAL COMPOSITION

PM composition measurements require collection of materials on substrates that can then be analyzed by precise laboratory methods (Chow, 1995). Since UP mass concentrations are low compared to other size fractions (e.g., PM_{2.5} and PM₁₀), large sample volumes or sensitive analytical methods are needed. Inertial size-selective inlets for UP present design and application challenges. *In situ* single particle mass spectrometers are emerging technologies for UP chemical characterization (Middlebrook *et al.*, 2003).

Particle measurement systems

A generic particulate sampling system includes size-selective inlet, sampling surface, denuders, filter holders, flow controllers, and pumps (Watson and Chow, 2001). Although simple in concept, practical implementation requires a careful integration of the components specific to the sampling objectives. The nature of the aerosol being sampled, environmental sampling conditions (e.g., temperature and relative humidity), and the types of chemical analyses applied to the filter deposit must be evaluated before the sample is taken.

UP size-selective inlets

Impactors, virtual impactor/aerosol concentrators, and aerodynamic lenses have been devised to separate the UP fraction from larger particles.

Impactors

Stacked or cascade impactors obtain particle size ranges in series. At near-ambient pressures, the lower size ranges of cascade impactors were once limited to ~100 nm because it was impractical to make jet widths small enough and flow rates high enough to permit impaction of smaller particles (Marple, 2004). A filter located after the final stage would collect all of the smaller particles, but this was insufficient to characterize the UP fraction.

UP sizes are similar to the mean free path of air molecules (66.4 nm at 293°K and atmospheric pressure), the distance a typical air molecule travels before encountering another molecule. The mean free path decreases at lower pressures. The Cunningham slip correction factor is used in impactor design equations to account for differences in particle movements as they become less equally bombarded by air molecules. By operating impactor stages at lower pressures, size cuts can be reduced. Smaller nozzle widths decrease downstream pressures as well as increasing the velocity through the nozzle, which also lowers the cut-point. UP sampling on substrates became practical in the late 1970s and early 1980s with the perfection of low pressure impactors (Berner, 1972; 1976a; 1976b; 1984; Hering *et al.*, 1979a; 1979b; Marple *et al.*, 1981; 1991; Wang and

John, 1988; Hillamo and Kauppinen, 1991). Table 1 summarizes the cut points for different cascade impactors.

The 13-stage Low Pressure Impactor (LPI; Hering *et al.*, 1979a; 1979b) uses a single nozzle for each stage to concentrate collected particles into a spot on the impaction surface. It was designed to use metal strips as impaction substrates that could be analyzed for sulfur by flash volatilization (Roberts and Friedlander, 1976). The small amount of deposit collected by the UP stages, the non-uniformity of the deposit, and the collection substrate are not amenable to a broad range of chemical composition measurements.

The eight-stage Berner Low Pressure Impactor (BLPI; Berner, 1972; 1976a; 1976b; 1984; Hillamo and Kauppinen, 1991) contains multiple nozzles (1 to 250) in each impaction plate. Variation in the number of nozzles per stage creates uncertainties for chemical analysis. Halder *et al.* (1999) modified their elemental detection system to rotate the samples in front of the excitation beam. This is not satisfactory for other methods where a portion of the deposit is used to extrapolate results for the entire sample.

The 11- to 13-stage Andersen Low Pressure Impactor (ALPI; Yamasaki *et al.*, 2000; Pagels *et al.*, 2005; Vaaraslahti *et al.*, 2005; Zervas and Dorlhene, 2006) has multiple jet impaction plates, similar to the BLPI. These are individually manufactured and calibrated. Table 1 shows several versions of the ALPI with flow rate variations from 20-24.5 liters per minute (L/min).

The 13-stage Electrical Low Pressure Impactor (ELPI, Dekati Instruments, Finland) uses a unipolar corona discharge to impact an unit charge on each particle (Marjamäki *et al.*, 2000). The charged particles then travel through a series of impactors and are deposited onto stages based on their aerodynamic diameters. Each impactor stage is electronically isolated and the accumulated charge on each substrate is proportional to the number of particles deposited on that stage. ELPIs have been used in a variety of source characterization studies for diesel (e.g., Shi and Harrison, 1999; Arnold *et al.*, 2006; Mamakos *et al.*, 2006), gasoline (e.g., Maricq *et al.*, 1999), wood combustion (e.g., Hays *et al.*, 2003), and power plant (e.g., Yi *et al.*, 2006) emissions, as well as for indoor (e.g., Mosley *et al.*, 2001) and outdoor (e.g., Gouriou *et al.*, 2004) studies and characterization of pharmaceuticals (Glover and Chan, 2004).

The 12-stage Small Deposit Low Pressure Impactor (SDI; Maenhaut *et al.*, 1996) was developed for compatibility with Proton Induced X-ray Emission (PIXE) spectroscopy to measure elemental composition. It is called “small deposit” because the sample is focused within an 8 mm diameter spot.

The Micro-Orifice Impactor (MOI; Kuhlmeier *et al.*, 1981) was similar to the BLPI, but it evolved into the 11-stage Micro-Orifice Uniform Deposit Impactor (MOUDI; Marple *et al.*, 1991; MSP, 1999) by slowly rotating the impaction plates under each nozzle, thereby meeting the need for homogeneous deposits. The Nano-MOUDI (Fujitani *et al.*, 2006) provides greater detail in the UP size fraction. MOUDI is most commonly used for UP composition studies.

Table 1. Different cut points for different cascade impactors.

Impactor	Flow Rate (L/min)	Total Stages	Cut Point Ranges (μm)	UP Stages	UP Cut Points (nm) [*]
Low Pressure Impactor (LPI) ^a	10 or 30	13	< 0.03–10	5	30, 60, 108, 170
Berner Low Pressure Impactor (BLPI) ^b	30	8	0.034–11.3	4	34, 65, 94, 160
Electrical Low Pressure Impactor (ELPI) ^c	10 or 30	13	0.007–10	4	30, 60, 108, 170
Small Deposit Area Impactor (SDI) ^d	11	12	0.045–8.5	3	45, 90, 150
Andersen Low Pressure Impactor (ALPI) ^e	22.2	12	0.056–9.5	2	56, 130 ^f
(Dylec LP-20)	20	12	0.06–11	2	60, 120 ^g
	24.2	11	0.11–11	1	110 ^h
	24.5	13	0.01–10.85	3	10, 50, 120 ⁱ
Micro Orifice Uniform Deposit Impactor (MOUDI) ^j	10 or 30	10	0.056–18	3	56, 100, 180
Nano-MOUDI ^k	10 or 30	13	0.010–18	4	10, 18, 32, 56

^a Dekati, Ltd., Tampere, Finland, (Hering *et al.*, 1979a; 1979b).

^b (Berner, 1972, 1976a, 1976b; Hillamo and Kauppinen, 1991).

^c Dekati, Ltd., Tampere, Finland, (Shi *et al.*, 1999; Keskinen *et al.*, 2003).

^d Dekati, Ltd., Tampere, Finland, (Maenhaut *et al.*, 1996).

^e Andersen Instruments, now Thermo Fisher Scientific, Waltham, M.A.; Model LP-20, Tokyo Dylec, Tokyo, Japan (Yamasaki *et al.*, 2000; Pagels *et al.*, 2005; Vaaraslahti *et al.*, 2005; Zervas and Dorlhene, 2006).

^f (Yamasaki *et al.*, 2000)

^g (Hasegawa *et al.*, 2004)

^h (Kawanaka *et al.*, 2004)

ⁱ (Ma *et al.*, 2004)

^j MSP Corporation, Shoreview, MN, (Marple *et al.*, 1990; 1991; MSP, 1999).

^k MSP Corporation, Shoreview, MN, (MSP, 2004).

^{*} Nominal cut points. Actual cut points vary from instrument to instrument depending on calibration.

Although cascade impactor substrates are sometimes greased or oiled to minimize re-entrainment and bounce from one stage to the next, these coatings interfere with chemical analyses (Fujitani *et al.*, 2006; Wang *et al.*, 2005). This is more of an issue for larger, and drier, soil particles than for PM_{2.5} which are often inherently oily. Serious particle bounce is indicated by soil-related elements in the lower impaction stages.

Aerosol concentrators

Virtual impactors are often used as aerosol concentrators (e.g., Sioutas *et al.*, 1995) to obtain aerosol concentrations higher than those found in ambient air. Several UP concentrators have been developed (Gordon *et al.*, 1999; Gupta *et al.*, 2004a; 2004b; Kim *et al.*, 2000a; 2000b; 2001b; 2001a; Misra *et al.*, 2004; Sioutas *et al.*, 1999) where UP are drawn through an inlet that removes the large particles (e.g., > 0.15 µm), then into a chamber saturated with water vapor over a warm water reservoir. UP subsequently pass through a condensing area at lower temperature where the particles grow as water vapor condenses on them. These grown particles then pass through a 1.5 µm cut-point virtual impactor where they are separated from most of the airstream. They then pass through a drier where the water is evaporated and they return to their UP size ranges. The implementation of Kim *et al.* (2000b) operates at 120 L/min and the minor flow can vary from 3 to 12 L/min. This provides for an enrichment of 10 to 40 times the ambient UP concentration. Although constructed primarily for animal exposure studies, this type of concentrator inlet can be used to obtain large quantities of UP on filters amenable to different chemical analysis methods. Owing to the different hygroscopic properties of UP, the sampled composition and size distribution may differ from that of the ambient air.

UP aerodynamic lenses

The aerodynamic lens (Liu *et al.*, 1995a; 1995b; Middha and Wexler, 2005; Petrucci *et al.*, 2000) consists of apertures of varying sizes in a series. A particle beam is produced when a particle-laden gas expands through a nozzle into a vacuum. Particles move closer to the axis when their aerodynamic diameters are less than a critical value and experience small radial drag forces. They stay close to the axis during nozzle expansion and therefore form a narrow particle beam downstream. The major effects that limit the minimum beam width are Brownian motion and lift forces on particles during the nozzle expansion. Aerodynamic lenses are used almost exclusively on single particle spectrometers for particle sizes < 100 nm. Because the lower size range of particle mass spectrometers is limited, and quantification is less than 100% efficient, these instruments underestimate particle counts.

Sampling substrates

Sampling substrates must be matched to the analysis purpose (Chow, 1995). For impactors such as the MOUDI, the substrates must be thin enough that they do not interfere with the narrow gap between the nozzle exit and the impaction plate. The substrates cannot contain the substances being measured, so it is often necessary to operate several instruments in parallel to accommodate a number of filter media. MOUDIs can be obtained for either 37 mm or 47 mm diameter substrates.

Ringed Teflon-membrane filters have been found to pop up the retainer ring on the MOUDI filter holder and have been used after the last impactor stage that allows the flow for the impactors to be drawn through. Because filter porosity is not needed in an impactor such as the MOUDI, 37 or 47 mm disks of non-porous Teflon fluorinated ethylene propylene copolymer (FEP) films of 0.002 thickness (DuPont) are used. These disks are soaked in methanol overnight, rinsed with distilled-deionized water (DDW) and dried in a vacuum chamber to remove contaminants. These substrates are amenable to mass, elemental, and ion determinations.

Aluminum (Al) foil in 37 or 47 mm disks (Reynolds Aluminum; Gresham, OR) are often used as substrates for carbon analysis. The disadvantage of foil is that it is highly reflective and melts at ~600°C when heated in an inert atmosphere. This makes the separation of elemental from organic carbon (EC, OC) uncertain when measured by thermal/optical methods (Chow *et al.*, 1993; 2007). One solution is to apply the pyrolysis correction on the quartz backup filter to the total carbon (TC) measured on the Al substrates.

Other substrates (e.g., Pallflex TX40HI20 and T60A20 Teflon-coated glass-fiber filters) can be used for ion analyses and for specific organic compounds, but not for TC analyses owing to their Teflon coating. Glass-fiber filters contain borosilicate glass filaments and should not be considered for particle sampling (Coutant, 1977; Spicer and Schumacher, 1979; Witz *et al.*, 1983; Lin and Friedlander, 1988). Fujitani *et al.* (2006) showed that mode diameter varies from 56 nm for ELPI with Al foil, to 100 nm for a Nano-MOUDI with Al foil, to 260 nm for ELPI with quartz-fiber filters for diesel exhaust particles. Heavy loadings on ELPI-Al (similar to those of polycarbonate substrates) experienced particle blow-off, resulting in a smaller mode diameter.

Etched polycarbonate-membrane filters have low elemental blank levels and are appropriate for elemental and ion analyses but not for thermal evolution carbon analysis (Watson *et al.*, 2005). They are the best filter media for single particle analysis by electron microscopy because particles are easily distinguished from the flat filter surface. The filters hold an electrostatic charge that influences mass measurements unless effort is invested in discharging them with a small radioactive source (Engelbrecht *et al.*, 1980). Electrostatic discharging is good practice for all filter media, even though others do not retain as much charge as the polycarbonate membranes.

UP laboratory analysis methods

Chow (1995), Solomon *et al.* (2001), Wilson *et al.* (2002), and Fehsenfeld *et al.* (2004) review laboratory chemical analysis methods applicable to UP and other PM samples. Filter substrates are analyzed for mass by gravimetry (Engelbrecht *et al.*, 1980; Feeney *et al.*, 1984; Witz *et al.*, 1988; 1990), elements by X-ray Fluorescence (XRF; Watson *et al.*, 1999), PIXE (Kasahara, 1999), Instrumental Neutron Activation Analysis (INAA; Dams *et al.*, 1970; Zoller and Gordon, 1970; Olmez, 1989), Atomic Absorption Spectrometry (AAS; Ranweiler and Moyers, 1974; Fernandez, 1989), Inductively-coupled Plasma (ICP) with Atomic Emission Spectroscopy and ICP with Mass Spectrometry (ICP/AES and ICP/MS; Fassel and Kniseley, 1974; McQuaker *et al.*, 1979; Lynch *et al.*, 1980; Harman, 1989; Wang *et al.*, 1998; Tan *et al.*, 2002; Lough *et al.*, 2005), ions by ion chromatography (IC; Chow and Watson, 1999), Automated Colorimetry (AC; Butler *et al.*, 1978; Mueller *et al.*, 1978; Fung *et al.*, 1979; Pyen and Fishman, 1979), and carbon (OC, EC, and carbon fractions) by thermal or thermal/optical methods (Chow *et al.*, 1993; 2001; 2004; 2005a; 2007; Watson *et al.*, 2005).

Organic speciation

The most common method used for speciated organic compounds (such as PAHs, alkanes, alkenes, and polar organics) in PM samples is solvent extraction (SE), followed by gas chromatography (GC)/MS, time-of-flight (TOF)/MS, or flame ionization detection (FID). Combined Fourier transform infrared (FTIR)/MS techniques or high performance liquid chromatography (HPLC)/MS are also used. HPLC is a form of IC that uses columns and eluents specific to water soluble carbon, especially organic acids. Large sample deposits are required for these analyses, much more than is available in the UP fraction unless acquired from high-volume sampling, impactors, or aerosol concentrators.

Thermal desorption (TD)-GC/MS (e.g., Ho and Yu, 2004; Hays and Lavrich, 2007) is an emerging technology in which a small section of the substrate can be placed directly into the GC injector and heated. Organic materials are volatilized and detected by GC/MS or GC/MS-FID. Very small samples, such as UP samples from impactors, can be analyzed by this technique.

Thermal denuders can also be used with other UP detection devices, such as the scanning mobility particle sizer (SMPS), to infer aerosol composition (Hasegawa *et al.*, 2004). Particles are drawn through an inlet that is cycled between ambient and a selected higher temperature and the SMPS size distributions are compared. Temperatures in the range of 300°C are often used to include sulfates, nitrates, and many OC compounds. The difference between the heated and unheated measurements separates the stable from the “semi-volatile” fraction of the aerosol.

Another approach is to use a humidifier between two SMPSs to determine the hygroscopicity of the sampled aerosol. This is termed the Tandem Differential Mobility Analyzer (TDMA; Rader and McMurry, 1986). Sodium chloride, ammonium sulfate, ammonium nitrate, and some

organic compounds grow to larger sizes when humidified. Other materials, such as EC, often do not grow as much. This gives a qualitative indication of the potential chemical composition.

Individual particle analysis

Single particles are characterized by optical or electron microscopy. Optical microscopy (Lee and Kelly, 1980; Janocko *et al.*, 1982; Casuccio *et al.*, 1983; 1989; Dattner *et al.*, 1983; Lucas *et al.*, 1988) is useful for coarse particles with sizes much larger than the wavelength of light (0.3 to 0.7 μm). Electron microscopy (EM; such as scanning EM [SEM] or transmission EM [TEM]) is needed to characterize smaller particles and their size distribution. These methods provide information on particle color, shape, size, and composition.

ULTRAFINE PARTICLE COMPOSITION

Table 2 summarizes measurements from 25 studies of UP chemical composition. It lists the sampling location, types of measurements taken, the number of samples, and the major findings with respect to UP composition. Most of the studies acquired other UP measurements, such as particle number and size distribution. These are included in the table for future reference. Several general observations can be made with respect to data reporting, measurement locations, measurement methods, and aerosol composition.

Table 2. Summary of studies on ultrafine particle (UP) composition.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																																																
Hughes <i>et al.</i> (1998)	<ul style="list-style-type: none"> One site: Pasadena, CA. Five 24-hr average samples every sixth day from 1/1996 to 2/1996. 	<ul style="list-style-type: none"> A DMA/CPC (Models 3071 and 3760) every 90s in 175 channels for $0.017 < d_p < 0.25 \mu\text{m}$ (including 120 channels of UP). A laser OPC (Model ASASP-X) every 4-10s in 31 channels for $0.09 < d_p < 3.0 \mu\text{m}$. A ten-stage MOUDI (Model 100) and a non-rotating MOI (Model 110) (reports bottom six stages $[0.056 < d_p < 1.8 \mu\text{m}]$ and after filter). MOUDI and MOI were preceded with a Teflon-coated cyclones to remove coarse particles ($d_p > 1.8 \mu\text{m}$). The two UP stages were 56 and 97 nm. A low-volume fine PM sampler. 	<ul style="list-style-type: none"> Acquired mass elements (by INAA), carbon (OC and EC) by TOT, anions (Cl⁻, NO₃⁻, SO₄²⁻) by IC, and cations (NH₄⁺) by ICM using RFA. Data presented in tables for daily UP concentrations, stacked bar charts for MOUDI concentrations, and pie charts without percentages for each component. 	<ul style="list-style-type: none"> Particle number concentration $1.3 \times 10^4 \pm 8.9 \times 10^3 \text{ \#}/\text{cm}^3$. Averaged 24-hr UP mass was $0.6\text{-}1.2 \mu\text{g}/\text{m}^3$. <table border="1"> <thead> <tr> <th rowspan="2">Date</th> <th colspan="2">UP mass ($\mu\text{g}/\text{m}^3$)</th> </tr> <tr> <th>fine mass $d_p < 1.8 \mu\text{m}$</th> <th>56-97 nm</th> </tr> </thead> <tbody> <tr> <td>1/23/96</td> <td>21.5</td> <td>0.36</td> </tr> <tr> <td>1/29/96</td> <td>25.5</td> <td>0.20</td> </tr> <tr> <td>2/4/96</td> <td>14.9</td> <td>0.24</td> </tr> <tr> <td>2/10/96</td> <td>24.9</td> <td>0.91</td> </tr> <tr> <td>2/17/96</td> <td>14.0</td> <td>0.20</td> </tr> <tr> <td>2/17/96</td> <td>14.0</td> <td>0.76</td> </tr> </tbody> </table>	Date	UP mass ($\mu\text{g}/\text{m}^3$)		fine mass $d_p < 1.8 \mu\text{m}$	56-97 nm	1/23/96	21.5	0.36	1/29/96	25.5	0.20	2/4/96	14.9	0.24	2/10/96	24.9	0.91	2/17/96	14.0	0.20	2/17/96	14.0	0.76	<ul style="list-style-type: none"> Used particle density of $1.7 \text{ g}/\text{cm}^3$ to convert equivalent number concentration to mass distribution. OC ($0.26 \mu\text{g}/\text{m}^3$) and EC ($0.06 \mu\text{g}/\text{m}^3$) contributed 46 to 62% of UP mass. Trace metals accounted for a few % to ~25% of UP mass with Fe (average/maximum $67.5/148.3 \text{ ng}/\text{m}^3$) being most abundant followed by Ti ($7.65/110.2 \text{ ng}/\text{m}^3$), Cr ($7.32/126.2 \text{ ng}/\text{m}^3$), Zn ($3.68/6.56 \text{ ng}/\text{m}^3$) and Ba ($1.04/2.8 \text{ ng}/\text{m}^3$). Lanthanide series elements (used as catalysts) were found for La and Ce with an average/maximum of $0.11/0.51 \text{ ng}/\text{m}^3$ and $0.19/0.82 \text{ ng}/\text{m}^3$, respectively. 																									
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Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Hughes <i>et al.</i> (1999)	<ul style="list-style-type: none"> Four sites in CA, three urban and one background: <ul style="list-style-type: none"> Long Beach Fullerton Riverside Santa Catalina Islands (background site). From 9/21/1999 to 10/2/1999 for: <ul style="list-style-type: none"> 0700-1100 PDT (Long Beach) 1100-1500 PDT (Fullerton) 1500-1900 PDT (Riverside) Starting at 1300 PDT for 24-hr on 9/21/1999 (Santa Catalina Island). Six four-hr and one 24-hr average samples for UP (four samples each at three sites plus two samples at Riverside and one sample at Santa Catalina Island). 	<ul style="list-style-type: none"> EAA ($17 < d_p < 250$ nm; Model 3030) every seven min at three urban sites A laser OPC ($0.09 < d_p < 2.6$ μm; Model ASAP-X) every five sec in 21 channels at the Long Beach and Riverside sites. Two ATOFMS at the three urban sites. One ten-stage MOUDI (Model 100) and non-rotating MOI (Model 110) report the bottom six stages ($0.056 < d_p < 1.8$ μm) and after filter. Low-volume total and fine PM sampler. The two UP stages are 56 and 97 nm 	<ul style="list-style-type: none"> Acquired mass elements by INAA, carbon (OC and EC) by TOT, anions (Cl^-, NO_3^-; and SO_4^{2-}) by IC, and cations (NH_4^+) by ICM using RFA. VOCs and total non-methane organic particle compounds by EPA method TO-12 (Data not reported). Data presented in percent average mass distribution with a stacked bar chart for MOUDI concentrations, and mass spectra from ATOFMS. 	<ul style="list-style-type: none"> Particle number concentrations $\sim 10^4$ #/cm³. UP composition for the MOUDI stacked bar charts were too small to quantify UP composition. 	<ul style="list-style-type: none"> Particle density of 1.3 g/cm³ used to convert equivalent particle number concentration to mass distribution. Nominal cut points for MOUDI and MOI were not reported. Different numbers of impactor samples were taken at the different sites on different days, precluding site-to-site comparison. Unimodal particle mass distribution found at Riverside, with the peak around 0.5 μm, larger than the mode at Long Beach. UP mass was ~ 1-1.5 μg/m³, with organic compounds being the largest contributor at urban sites. Santa Catalina Island UP was mostly inorganic, but not well identified. Example mass spectra for ATOFMS showed geometric mass d_p of 0.37 μm for NH_4NO_3 containing particles with OC inclusion, 0.42 μm for OC, and 0.98 μm for predominantly EC with some NH_4NO_3. No information given for UP.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																																	
Cass et al. (2000)	<ul style="list-style-type: none"> Seven sites in CA: <ul style="list-style-type: none"> L.A. Pasadena Fullerton Azusa Mira Loma Diamond Bar Riverside. Several 24-hr average samples from 1/1996 to 11/1997. 	<ul style="list-style-type: none"> DMA/CPC (TSI Models 3071 and 3760). EAA (TSI Model 3030). OPC (Model ASASP-X). Two MOUDIs (Model 100) (0.056 < d_p < 1.8 μm in six stages). 	<ul style="list-style-type: none"> Acquired mass, elements by INAA, carbon (OC and EC) by TOT, anions (Cl⁻, NO₃⁻, and SO₄²⁻) by IC, and cations (NH₄⁺) by ICM using RFA. Data presented in stacked bar charts and pie charts. 	<ul style="list-style-type: none"> UP mass averaged 0.8 μg/m³ ranging from 0.55 to 1.16 μg/m³. UP chemical composition: <table border="1" style="margin-left: 20px;"> <thead> <tr> <th>Observables</th> <th>Average</th> <th>Range</th> </tr> </thead> <tbody> <tr> <td>Mass (μg/m³)</td> <td>0.8</td> <td>0.55-1.16</td> </tr> <tr> <td>OC x 1.4</td> <td>50%</td> <td>32-67%</td> </tr> <tr> <td>EC</td> <td>8.7%</td> <td>3.5-17.5%</td> </tr> <tr> <td>Cl⁻</td> <td>0.5%</td> <td>0-2.5%</td> </tr> <tr> <td>NO₃⁻</td> <td>6.8%</td> <td>0-19%</td> </tr> <tr> <td>SO₄²⁻</td> <td>8.2%</td> <td>0.8-18%</td> </tr> <tr> <td>NH₄⁺</td> <td>3.7%</td> <td>0-33%</td> </tr> <tr> <td>Na⁺</td> <td>0.6%</td> <td>0-2.5%</td> </tr> <tr> <td>Metal oxides</td> <td>14%</td> <td>0.6-26%</td> </tr> <tr> <td>Unknown</td> <td>7.5%</td> <td>0-29%</td> </tr> </tbody> </table> 	Observables	Average	Range	Mass (μg/m ³)	0.8	0.55-1.16	OC x 1.4	50%	32-67%	EC	8.7%	3.5-17.5%	Cl ⁻	0.5%	0-2.5%	NO ₃ ⁻	6.8%	0-19%	SO ₄ ²⁻	8.2%	0.8-18%	NH ₄ ⁺	3.7%	0-33%	Na ⁺	0.6%	0-2.5%	Metal oxides	14%	0.6-26%	Unknown	7.5%	0-29%	<ul style="list-style-type: none"> Estimated UP (i.e., PM_{0.1}) emissions were 13.3 tonnes per day in Southern California with 43% from on-road vehicles, 32% from stationary fuel combustion, and 10% from other mobile sources (e.g., diesel engines, off-road vehicles). Primary UP emissions were estimated to consist of 65% organic compounds, 7% EC, 7% sulfate and 4% trace elements. UP composition varied by location and season. Mass particle size distribution peaked at 0.5 μm consisting largely of carbonaceous aerosol, followed by (NH₄)₂SO₄ and NH₄NO₃. Abundant trace metals in UP were K (88 ng/m³), Na (85 ng/m³), and Ba (19 ng/m³). Abundant transition metals in UP were Fe (186 ng/m³), Ti (43 ng/m³), Cr (6.7 ng/m³) and Zn (3.8 ng/m³).
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Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																																																
Chung <i>et al.</i> (2001)	<ul style="list-style-type: none"> • One site: Bakersfield, CA • 8-hr average samples acquired from 1000-1800 PST on seven days from 1/14/1999 to 1/23/1999. • 1/14/1999 and 1/21/1999 were chemically analyzed. 	<ul style="list-style-type: none"> • Two MOUDIs (Model 110) (0.056 < d_p < 1.8 μm in six stages) equipped with AIHL-designed cyclones. • Collocated MOUDIs with filter-based sampling. 	<ul style="list-style-type: none"> • Acquired mass elements by PIXE and XRF, carbon (OC and EC) by TOT, and ions (Na^+, K^+, Ca^+, NH_4^+, Cl^-, NO_3^-, PO_4^{3-}, and SO_4^{2-}) by IC. • Data presented in stacked bar charts; percent chemical composition by pie charts (without mass concentration), and time series. 	<p>Particle number concentrations from $\sim 10^{10}$ to 10^{11} #/cm³</p> <p>Ultrafine percentage composition:</p> <table border="1"> <thead> <tr> <th>Observables</th> <th>Polluted day (1/14/1999)</th> <th>Clean day (1/21/1999)</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>NA</td> <td>NA</td> </tr> <tr> <td>OC</td> <td>24%</td> <td>14%</td> </tr> <tr> <td>EC</td> <td>5%</td> <td>5%</td> </tr> <tr> <td>NO_3^-</td> <td>11%</td> <td>8%</td> </tr> <tr> <td>SO_4^{2-}</td> <td>5%</td> <td>5%</td> </tr> <tr> <td>Other S</td> <td>5%</td> <td>5%</td> </tr> <tr> <td>Na^+</td> <td>7%</td> <td>8%</td> </tr> <tr> <td>Cl^-</td> <td>0%</td> <td>2%</td> </tr> <tr> <td>Al</td> <td>6%</td> <td>14%</td> </tr> <tr> <td>Si</td> <td>4%</td> <td>0%</td> </tr> <tr> <td>K</td> <td>3%</td> <td>2%</td> </tr> <tr> <td>Ti</td> <td>2%</td> <td>3%</td> </tr> <tr> <td>Fe</td> <td>1%</td> <td>2%</td> </tr> <tr> <td>Ca</td> <td>20%</td> <td>18%</td> </tr> <tr> <td>Other metals</td> <td>8%</td> <td>13%</td> </tr> </tbody> </table>	Observables	Polluted day (1/14/1999)	Clean day (1/21/1999)	Mass	NA	NA	OC	24%	14%	EC	5%	5%	NO_3^-	11%	8%	SO_4^{2-}	5%	5%	Other S	5%	5%	Na^+	7%	8%	Cl^-	0%	2%	Al	6%	14%	Si	4%	0%	K	3%	2%	Ti	2%	3%	Fe	1%	2%	Ca	20%	18%	Other metals	8%	13%	<ul style="list-style-type: none"> • No specific cut points were given for MOUDI stages. • Ca, (the geological elements), and OC (54 ± 62 ng/m³) were major UP components. • UP at Bakersfield were alkaline while fine particles were neutral based on anion/cation balance (UP Ca^{++} was 64 ± 57 ng/m³ and estimated 25% more cations than anions).
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Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances		Comments
				Urban Site	Rural Site	
Pakkanen <i>et al.</i> (2001a)	<ul style="list-style-type: none"> Two sites in Finland: – Vallila, Helsinki (urban site 2 km NE of city center) – Luukki, Espoo (rural site). 24- or 48-hr average samples starting at 0900-1000 LST were acquired once per month from 6/12/1996 to 6/5/1997 (10 valid pairs). 	<ul style="list-style-type: none"> Two ten-stage BLPs sampling for $0.035 < d_p < 7.5 \mu\text{m}$ with pre-impactor cut-size of $\sim 15.7 \mu\text{m}$ (Liu and Pui, 1981). The three UP stages are 35, 67, and 93 nm. Collocated BLPs and VIs for validation (Pakkanen and Hillamo, 2002). 	<ul style="list-style-type: none"> Acquired mass, elements by ICP/MS, (an additional six BLP samples [60 individual stages] by PIXE and INAA for comparison), ions (Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Mg^{++}, Ca^{++}, MSA, O_x, malonate, succinate, glutarate) by IC. Data presented in tables, species size distribution, and ion balance as a fraction of particle sizes and size distribution. 	Average mass and chemical composition (ng/m^3) are:		<ul style="list-style-type: none"> Elemental and ion analyses explained 15-20% of UP mass. The remaining UP mass was attributed 70% to carbonaceous aerosol and 10% to water, which were not measured. Contribution of UP to $\text{PM}_{2.5}$ mass was 7% at urban and 8.5% at rural sites. Most abundant metals were Na, K, Ca, Fe, and Zn, ranging from 0.7 to $5 \text{ ng}/\text{m}^3$. Most abundant organic ions were O_x and MSA, contributing to UP at 0.5% and 0.37%, respectively. Contribution of UP to $\text{PM}_{2.5}$ for B, Ni, Se, and Ag was 10-20%. Aiken mode ($d_p < 0.15 \mu\text{m}$) often observed with mean mode diameter of 0.06 and $0.12 \mu\text{m}$. Abundance of Mg, Ca, Sr, and Ba at Aitken mode was attributed to vehicle exhaust. Abundance of Fe, Co, Ni, and Mo was attributed to heavy fuel oil combustion.
				Observables	Urban Site	
Mass	490	520				
NO_3^-	4	11				
SO_4^{2-}	32	40				
NH_4^+	22	25				
B	0.27	0.13				
Na ⁺	2.1*	3.2*				
Na	2.9*	1.3*				
Mg^{++}	0.69*	0.92*				
Mg	0.26*	0.37*				
Cl	1.7	1.2				
K ⁺	2.0*	1.5*				
K	0.86*	1.3*				
Ca^{++}	4.8*	6.5*				
Ca	2.2*	1.9*				
V	0.16	0.092				
Fe	1.8	0.73				
Co	0.024	0.023				
Ni	0.24	0.24				
Zn	0.81	0.70				
Sr	0.01	0.009				
Mo	0.015	0.013				
Ba	0.058	0.03				
Ox	2.1	1.9				
MSA	1.3	1.7				

* Na^+/Na , Mg^{++}/Mg , K^+/K , and Ca^{++}/Ca ratios often exceeded unity, indicating an overestimation of cations or underestimation of anions.

- Anion deficit was found for $d_p < 0.2 \mu\text{m}$ and $d_p > 1 \mu\text{m}$
- SO_4^{2-} size distribution showed MMD $\sim 0.5 \mu\text{m}$

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																		
Geller <i>et al.</i> (2002)	<ul style="list-style-type: none"> Two sites in the L.A. Basin, CA: <ul style="list-style-type: none"> Downey (Diesel emission-dominant site) Riverside (Downwind receptor site). Three-hour samples taken from 3/12/2001 to 3/29/2001 at: <ul style="list-style-type: none"> 0700-1000 PST (morning) 1100-1400 PST (midday) 1500-1800 PST (evening). Total of 36 UP samples (= 3 samples/day x 6 days x 2 sites). 	<ul style="list-style-type: none"> USC UPC (concentrator) operated at 110 L/min intake and 5 L/min output flow rate (enrichment factor of 22) followed by nano-MOUDI (Model 115-1, nominal cut points: 10, 18, 32, and 56 nm). A modified MOUDI (Model 100) (100, 180, and 320 nm) operated at 10 L/min. Concentrations compared with collocated ELPI (Model 393501) and CPC (Model 3022A) measurements. 	<ul style="list-style-type: none"> Acquired mass, carbon (OC and EC) by TMO, and ions (NO_3^- and SO_4^{2-}) by IC. Data presented in an average histogram of five or six stages and scatterplots. No elemental concentrations or Nano MOUDI composition data were reported. 	<ul style="list-style-type: none"> Particle number concentrations $2.3\text{-}6.9 \times 10^3$ #/cm³ at Downey and $9.3 \times 10^3 - 1.1 \times 10^4$ #/cm³ at Riverside. UP average mass was 0.81-1.92 µg/m³ at Downey and 0.65-0.80 µg/m³ at Riverside. <table border="1" style="margin-left: auto; margin-right: auto;"> <thead> <tr> <th>UP (<100 nm) composition *</th> <th>Downey ng/m³</th> <th>Riverside ng/m³</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>500-4,200</td> <td>300-1,800</td> </tr> <tr> <td>OC</td> <td>200-1,000</td> <td>200-400</td> </tr> <tr> <td>EC</td> <td>10-310</td> <td>20-70</td> </tr> <tr> <td>NO₃⁻</td> <td>ND - 400</td> <td>ND - 80</td> </tr> <tr> <td>SO₄²⁻</td> <td>ND - 120</td> <td>ND - 40</td> </tr> </tbody> </table> <p>* Estimated value from histogram (in Figures 4 and 6 of Geller <i>et al.</i> (2002).</p>	UP (<100 nm) composition *	Downey ng/m ³	Riverside ng/m ³	Mass	500-4,200	300-1,800	OC	200-1,000	200-400	EC	10-310	20-70	NO ₃ ⁻	ND - 400	ND - 80	SO ₄ ²⁻	ND - 120	ND - 40	<ul style="list-style-type: none"> Define UP as < 180 nm and also as 10-100 nm. OC was predominant in UP at both sites, accounting for 50-80% of the total mass. Bimodal mass distribution with nuclei (32-56 nm) and accumulation (100-180 nm) modes was found at Downey. Distinct mode of UP mass and OC in the 32-56 nm range was independent of time of day at Downey. OC drove mass size distribution at the diesel-dominated Downey site. Morning peaks (32-56 µm) resulted from condensation of vehicle-emitted organic vapors. Average UP OC/EC ratios were 2.4, 2.7, and 3.4 at Downey, and 7.5, 9.9 and 7.5 at Riverside for morning, midday, and evening periods, respectively. Higher OC/EC ratios at Riverside were attributed to gasoline-fueled vehicle emissions.
UP (<100 nm) composition *	Downey ng/m ³	Riverside ng/m ³																					
Mass	500-4,200	300-1,800																					
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NO ₃ ⁻	ND - 400	ND - 80																					
SO ₄ ²⁻	ND - 120	ND - 40																					

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																																										
Kim <i>et al.</i> (2002)	<ul style="list-style-type: none"> Two sites in CA <ul style="list-style-type: none"> Downey (Diesel-emission dominated site) Riverside (Downwind receptor site) Approximately five months, from 9/19/2000 to 1/31/2001 at Downey, and 2/5/2001 to 6/30/2001 at Riverside. 24-hr average MOUDI measured once per week during a weekday (starting at 0600 PST). Acquired approximately 20 samples. 	<ul style="list-style-type: none"> SMPS (Model 3936) measured 15-min average particle number concentration ($14 < d_p < 500$ nm). A dual beam aethalometer (Model AE-20) measured 5-min $PM_{2.5}$ BC. Three MOUDIs (Model 110) (using 0.1, 0.35, 1.0, 2.5 and 10 μm stages) Five-min average wind speed, wind direction, temperature and relative humidity. 	<ul style="list-style-type: none"> Acquired mass, elements by XRF, carbon (OC and EC) by TMO, ions (NO_3^- and SO_4^{2-}) by IC. Data presented in tables and pie charts. Correlations and scatterplots were given over a diurnal time series. 	<ul style="list-style-type: none"> Geometric mean (concentration range): <table border="1" style="margin-left: 20px;"> <thead> <tr> <th></th> <th>Downey</th> <th>Riverside</th> </tr> </thead> <tbody> <tr> <td>Mass (μg/m³)</td> <td>4.11 (1.12-8.89)</td> <td>1.34 (0.42-3.65)</td> </tr> <tr> <td>OC (μg/m³)</td> <td>2.75 (2.62-3.01)</td> <td>1.35 (0.72-3.90)</td> </tr> <tr> <td>EC (μg/m³)</td> <td>0.67 (0.58-0.75)</td> <td>0.13 (0.04-0.19)</td> </tr> <tr> <td>NH_4NO_3 (μg/m³)</td> <td>0.08 (0.02-0.14)</td> <td>0.10 (ND - 0.33)</td> </tr> <tr> <td>$(NH_4)_2SO_4$ (μg/m³)</td> <td>0.15 (0.078-0.11)</td> <td>0.09 (0.05-0.2)</td> </tr> <tr> <td>Ti (ng/m³)</td> <td>3.2 (0.2-9.1)</td> <td>0.87 (0.22-2.9)</td> </tr> <tr> <td>V (ng/m³)</td> <td>1.8 (0.2-15.9)</td> <td>0.17 (0.16-0.33)</td> </tr> <tr> <td>Cr (ng/m³)</td> <td>5.1 (0.2-37)</td> <td>13 (0.17-6.7)</td> </tr> <tr> <td>Mn (ng/m³)</td> <td>2.5 (0.3-5.7)</td> <td>0.58 (0.3-2.03)</td> </tr> <tr> <td>Fe (ng/m³)</td> <td>40.5 (10.4-133)</td> <td>10 (1.23-33.8)</td> </tr> <tr> <td>Ni (ng/m³)</td> <td>3.6 (0.1-12.3)</td> <td>1.83 (0.13-13.6)</td> </tr> <tr> <td>Cu (ng/m³)</td> <td>6.4 (0.6-24.6)</td> <td>1.46 (0.18-9.54)</td> </tr> <tr> <td>Zn (ng/m³)</td> <td>7.5 (1.4-38.6)</td> <td>1.45 (0.11-6.11)</td> </tr> </tbody> </table> 		Downey	Riverside	Mass (μ g/m ³)	4.11 (1.12-8.89)	1.34 (0.42-3.65)	OC (μ g/m ³)	2.75 (2.62-3.01)	1.35 (0.72-3.90)	EC (μ g/m ³)	0.67 (0.58-0.75)	0.13 (0.04-0.19)	NH_4NO_3 (μ g/m ³)	0.08 (0.02-0.14)	0.10 (ND - 0.33)	$(NH_4)_2SO_4$ (μ g/m ³)	0.15 (0.078-0.11)	0.09 (0.05-0.2)	Ti (ng/m ³)	3.2 (0.2-9.1)	0.87 (0.22-2.9)	V (ng/m ³)	1.8 (0.2-15.9)	0.17 (0.16-0.33)	Cr (ng/m ³)	5.1 (0.2-37)	13 (0.17-6.7)	Mn (ng/m ³)	2.5 (0.3-5.7)	0.58 (0.3-2.03)	Fe (ng/m ³)	40.5 (10.4-133)	10 (1.23-33.8)	Ni (ng/m ³)	3.6 (0.1-12.3)	1.83 (0.13-13.6)	Cu (ng/m ³)	6.4 (0.6-24.6)	1.46 (0.18-9.54)	Zn (ng/m ³)	7.5 (1.4-38.6)	1.45 (0.11-6.11)	<ul style="list-style-type: none"> Unimodal size distribution with mode 30-40 nm at Downey and bimodal at Riverside with an increase in accumulation mode (0.1-2.5 μm). An increase in sub-100 nm during 1400-1600 PST was found at Riverside. MOUDI quartz-fiber after filter OC may have positive or negative artifacts. Fe was most abundant of the transition metals in UP, followed by Cu and Zn (relatively high V, Cr, and Ni were also found). UP V was 11 times higher at Downey than at Riverside, reflecting the impact from refineries at Downey.
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				<ul style="list-style-type: none"> Average Percentage: <table border="1" style="margin-left: 20px;"> <thead> <tr> <th></th> <th>Downey (12/19/2000-1/25/2001)</th> <th>Riverside (3/1/2001-4/19/2001)</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>NA</td> <td>NA</td> </tr> <tr> <td>OC</td> <td>64%</td> <td>73%</td> </tr> <tr> <td>EC</td> <td>18%</td> <td>8%</td> </tr> <tr> <td>NO_3^-</td> <td>2%</td> <td>1%</td> </tr> <tr> <td>SO_4^{2-}</td> <td>4%</td> <td>5%</td> </tr> <tr> <td>Others*</td> <td>2%</td> <td>0%</td> </tr> <tr> <td>Metals</td> <td>3%</td> <td>13%</td> </tr> </tbody> </table> 		Downey (12/19/2000-1/25/2001)	Riverside (3/1/2001-4/19/2001)	Mass	NA	NA	OC	64%	73%	EC	18%	8%	NO_3^-	2%	1%	SO_4^{2-}	4%	5%	Others*	2%	0%	Metals	3%	13%																			
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Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																												
Viidanoja <i>et al.</i> (2002a)	<ul style="list-style-type: none"> One site in Finland: Vallila, Helsinki (Urban site, 2 km NE of city center). 24-hr average samples were acquired starting at noon LT from 5/1/2000 to 7/15/2000. 	<ul style="list-style-type: none"> Two 12-stage SDIs sampling for 0.045 < d_p < 8.4 μm. The second SDI was equipped with a Teflon front filter to remove particles and estimate organic artifact. The two UP stages are 45 and 88 nm. Collocated SDIs and VI for comparison. An aethalometer (based on an absorption coefficient of 19 m^2/g). 	<ul style="list-style-type: none"> Acquired carbon (OC and EC) by TOT (OC at 310, 480, 615, and 900°C in helium; EC at 675, 750, 825, and 920°C in 10% oxygen in helium). OC and EC were determined both with (by laser) and without (by helium and oxygen phases) optical pyrolysis (OP) corrections. A pyrolysis correction factor determined by TOT with VI samples is used to estimate pyrolytic OC in SDI samples without OP correction by laser. Data presented in tables for SDI stages and for OC and EC size distribution. 	<ul style="list-style-type: none"> Nearly 100% of OC found at 45 and 88 nm stages are gaseous OC. The percentage of gaseous OC decreases to 20-40% of total OC (gas and particles) for the larger particles. UP Composition ($\mu\text{g C}/\text{cm}^3$): <table border="1" style="margin-left: 20px;"> <thead> <tr> <th>Carbon</th> <th>d_p (45-88 nm)</th> <th>d_p (< 45 nm)</th> <th>after filter</th> </tr> </thead> <tbody> <tr> <td>OC (Particle)</td> <td>4.5</td> <td>2.6</td> <td>5.3</td> </tr> <tr> <td>OC (Gaseous)</td> <td>2.8</td> <td>2.8</td> <td>5.2</td> </tr> <tr> <td>OC (6/20-21)</td> <td>3.8</td> <td>2.6</td> <td>5.2</td> </tr> <tr> <td>OC (6/27-28)</td> <td>3.8</td> <td>2.6</td> <td>5.4</td> </tr> <tr> <td>EC (6/20-21)</td> <td>1.9</td> <td>0.5</td> <td>0</td> </tr> <tr> <td>EC (6/27-28)</td> <td>0.8</td> <td>0.4</td> <td>0</td> </tr> </tbody> </table>	Carbon	d_p (45-88 nm)	d_p (< 45 nm)	after filter	OC (Particle)	4.5	2.6	5.3	OC (Gaseous)	2.8	2.8	5.2	OC (6/20-21)	3.8	2.6	5.2	OC (6/27-28)	3.8	2.6	5.4	EC (6/20-21)	1.9	0.5	0	EC (6/27-28)	0.8	0.4	0	<ul style="list-style-type: none"> Reported 11-23% (median of 20%) positive organic artifact by fine fractions (d_p < 2.3 μm) of VI as compared to SDI. OC pyrolysis in VI samples averaged 10% (1-22%) of total OC. SDI, like other impactors, was susceptible to sampling artifact and OC pyrolysis during thermal analysis. Carbonaceous aerosol in UP was overestimated without removing gaseous organic. OC pyrolysis was most prevalent on stages 4 (0.142-0.235 μm) and 5 (0.235-0.38 μm), followed by stages 6-8 (0.038-0.58, 0.058-0.8, and 0.8-1.06 μm, respectively). OC exhibited bimodal peaks at 0.2-0.8 μm and 2-5 μm. EC exhibited unimodal peaks at 0.2-0.5 μm. EC was low (0.4-0.5 $\mu\text{g C}/\text{cm}^3$) for d_p < 45 nm, EC was 40-50% of OC at 88 nm.
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Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Ishizaka and Adhikari (2003)	<ul style="list-style-type: none"> Two sites in Japan: <ul style="list-style-type: none"> Nagoya (urban site with industrial pollution) Mikuni (coastal atmosphere). Sampling continuously for ~two to five days on five days: <ul style="list-style-type: none"> 12/17/1998 12/18/1998 12/22/1998 1/20/1999 1/28/1999. 	<ul style="list-style-type: none"> A CPC (TSI Model 2025A) and a diffusion battery (TSI Model 3041) with switching value (TSI Model 3042A) in 15 channels for $0.003 < d_p < 0.2 \mu\text{m}$. A CCN counter. (Mee, Model 130) is activated at 0.5% super saturation. An LPC (Kanomax Model TF-500) for $d_p > 0.3 \mu\text{m}$. A thermal analysis system consisting of a heater (100–500°C), an aerosol sensor, and a data recorder. Sampling flow rate was 28.3 L/min for two to five days at Nagoya and 50 L/min for two to four days at Mikuni. 	<ul style="list-style-type: none"> Compared continuous thermal analytical curves with CCN concentrations based on rate of evaporation, and then inferred to its chemical compositions. Data presented in tables, histogram, and time-series plots. 	<ul style="list-style-type: none"> No UP composition was reported. Divided chemical compounds into three temperature ranges: <ul style="list-style-type: none"> Group I (0–150°C) highly volatile compounds: <ul style="list-style-type: none"> H_2SO_4, water vapor, formic acid, acetic acid, NH_4Cl, $(\text{NH}_4)_2\text{SO}_4$, OC, organic nitrogen, terpenes, and dioctyl phthalate Group II (150–300°C) intermediately volatile compounds: <ul style="list-style-type: none"> $(\text{NH}_4)_2\text{SO}_4$, NH_4HSO_4, NH_4NO_3, SOA, and diesel exhaust Group III (> 350°C) refractory compounds: <ul style="list-style-type: none"> Soot/BC, polymerized organic compounds, CaCO_3, and NaCl CCN decreased abruptly at 100°C and then decreased linearly to 12% of the initial concentration at 200°C for Nagoya samples and at 250°C for Mikuni samples. 	<ul style="list-style-type: none"> Used thermal analysis (i.e., volatility) to infer CCN composition. ~80% of CCN evaporated below 300°C with the majority of the volatile species, and the concentration of CCN remained constant between 300 and 500°C. Anthropogenic organic material, such as that from automotive exhaust, contributed to CCN in the polluted atmosphere. Thermal curve for inorganic ions were similar to CCN. This suggested that $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 contributed greatly to CCN at urban Nagoya site and $(\text{NH}_4)_2\text{SO}_4$ contributed greatly to CCN at coastal Mikuni site. Thermal curve of carbon and sea salt were different from CCN with organic material contributing to CCN. Soot particles did not contribute to CCN.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																				
Phares <i>et al.</i> (2003)	<ul style="list-style-type: none"> One site at East Houston, TX (near channel view, less than 1 km from ship channel) as a part of the U.S. EPA Supersite program. Continuous sampling from 8/23/2000 to 9/18/2000. 	<ul style="list-style-type: none"> An RSMS II (0.014 <math>d_p < 1.3 \mu m</math>), with a laser ablation time-of-flight MS characterizing single particle with positive/negative ion detection for ~110 min scan. 	<ul style="list-style-type: none"> Particles in 60 composition classes of nine sizes were selected (35, 50, 70, 100, 140, 170, 320, 590, and 1,140 nm). Data presented in number of spectra obtained in each compound class and wind roses (to infer pollution sources). 	<p>Major UP Composition includes:</p> <table border="1"> <thead> <tr> <th>Spectrum</th> <th>% of UP</th> </tr> </thead> <tbody> <tr> <td>Carbon</td> <td>16</td> </tr> <tr> <td>Si/Silicon Oxide</td> <td>30</td> </tr> <tr> <td>K</td> <td>31</td> </tr> <tr> <td>V/Vanadium Oxide</td> <td>0.64</td> </tr> <tr> <td>Fe</td> <td>6.1</td> </tr> <tr> <td>Zn</td> <td>1.7</td> </tr> <tr> <td>NaCl</td> <td>7.5</td> </tr> <tr> <td>Lime</td> <td>1.0</td> </tr> <tr> <td>Amines</td> <td>2.4</td> </tr> </tbody> </table>	Spectrum	% of UP	Carbon	16	Si/Silicon Oxide	30	K	31	V/Vanadium Oxide	0.64	Fe	6.1	Zn	1.7	NaCl	7.5	Lime	1.0	Amines	2.4	<ul style="list-style-type: none"> Obtained approximately 27,000 single particle spectra for individual particle composition, but no quantitative chemical composition was given. Houston ship channel aerosol is dominated by refineries, incinerators, and chemical plants as evidenced by emitted Si, V, amines, metals, and lime in UP.
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Rhoads <i>et al.</i> (2003)	<ul style="list-style-type: none"> One site at Atlanta, GA, as part of the August 1999 "Southern Oxidants Study Supersite Experiment." Sampled 120 hours over 20 days (from 8/10/1999 to 8/31/1999) and obtained spectra from 15,989 particles. 	<ul style="list-style-type: none"> An RSMS-II (0.014 <math>d_p < 1.3 \mu m</math>), with a laser ablation time-of-flight MS, characterizing single particle with positive/negative ion detection by LDI/MS. 	<ul style="list-style-type: none"> Particles in 70 composition classes of 13 size fractions, including 14, 30, 47, 59, 105, 153, 204, 254, 354, 491, 823, 995, and 1,285 nm. Data presented in number of spectra obtained in each compound class, by wind roses in eight directions, and by fractions of total as overall distributions of spectrum during study. 	<p>Major UP composition includes:</p> <table border="1"> <thead> <tr> <th>Spectrum</th> <th>% of UP*</th> </tr> </thead> <tbody> <tr> <td>OC</td> <td>74</td> </tr> <tr> <td>EC</td> <td>1.5</td> </tr> <tr> <td>NO₃⁻</td> <td>2</td> </tr> <tr> <td>Na</td> <td>1</td> </tr> <tr> <td>K</td> <td>8</td> </tr> <tr> <td>Ca</td> <td>2</td> </tr> <tr> <td>Fe</td> <td>3</td> </tr> <tr> <td>Others</td> <td>8.5</td> </tr> </tbody> </table> <p>*Based on ~15,000 single particle spectra.</p> <ul style="list-style-type: none"> Pie charts of UP composition are grouped by carbonaceous species, NO₃⁻, minerals*, SiO₂, Fe, other metals (specific elements were not given), and unknown categories. 	Spectrum	% of UP*	OC	74	EC	1.5	NO ₃ ⁻	2	Na	1	K	8	Ca	2	Fe	3	Others	8.5	<ul style="list-style-type: none"> Obtained approximately 15,000 spectra for individual particle composition but no quantitative chemical compositions were given. No distinguishable differences were found for minerals, SiO₂, Fe, and other metals. No S or SO₄⁻ were found in compound classes. Many of the major compound classes appeared in same size and/or wind direction ranges, indicating emissions from specific sources. 		
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Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Cohen <i>et al.</i> (2004)	<ul style="list-style-type: none"> One site at New York, NY, ~400 meters east of the former World Trade Center (WTC). Ten sampling periods of 3-4 days each during 9/20/2001 to 12/14/2001. 	<ul style="list-style-type: none"> Four UDM. One EAS (TSI Model 3100) preceded by five-stage MI (i.e., MI-EAS). One CNC (TSI Model 3020). One MOUDI (Model 100) with eight stages (0.1, 0.17, 0.29, 0.56, 1.0, 1.8, 3.8 and 15 μm). 	<ul style="list-style-type: none"> Acquired mass, carbon (OC and EC by TOT), and PAH (Offenberg <i>et al.</i>, 2003) in MI-EAS backup filters ($d_p < 0.5 \mu\text{m}$). Data presented in atomic force microscopy (AFM) images, histogram, and time series. 	<ul style="list-style-type: none"> Particle number concentrations ranged from $1-5 \times 10^4 \text{ \#}/\text{cm}^3$, peaking at $7 \times 10^4 \text{ \#}/\text{cm}^3$. No quantitative UP mass or chemical composition were reported. PAH ranged from 10 to $1,500 \text{ ng}/\text{m}^3$ for $d_p < 0.5 \mu\text{m}$. PAHs (BAA, PHE, FLT, pyrene, and chrysene/triphenylene) comprised 55% of PAHs for $d_p < 0.5 \mu\text{m}$. About 7% of TC was organic for $d_p < 0.5 \mu\text{m}$. 	<ul style="list-style-type: none"> UP and fine PM were not elevated during the study. No acidic or fibrous particles were found in UP fractions. Morphology of fine PM collected soon after the WTC collapse with major fires showed agglomerates of small particles under incomplete combustion.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Fine <i>et al.</i> (2004)	<ul style="list-style-type: none"> Two sites in CA: <ul style="list-style-type: none"> Downtown L.A. at USC Riverside. Four times daily (i.e., 0700-1030 PDT; 1100-1430 PDT; 1500-1830 PDT; 1900-0630 [the following day] PDT) 3.5- and 11.5-hr averaged samples during summer (8/12/2002-8/16/2002 at USC; and 8/26/2002-8/30/2002 at Riverside) and Winter (1/13/2003-1/17/2003 at USC; and 1/27/2003-1/31/2003 at Riverside). 	<ul style="list-style-type: none"> A High-Vol sampler with slit impactor operated at 500 L/min to separate accumulation (0.18-2.5 μm) and UP (0-0.18 μm) size ranges. A MOUDI (Model 110). MOUDI stages were chosen to correspond to the size cut points of the High-Vol sampler. 	<ul style="list-style-type: none"> Acquired mass and carbon (OC and EC) by TOT. Derivatized and non-derivatized PAHs (including 17α(H), 21β(H), and 29-norhopane, 17α(H), 21β(H)-hopane, 22,29,30-trisnorhopane and COR) and organic acids measured after derivatization as methylated acids by GC/MS. Solvent extraction in 50 mL of 19:1 mixture of DCM and methanol, followed by nitrogen evaporation. Data presented in histograms and correlations of EC and PAHs. 	<ul style="list-style-type: none"> UP mass up to 5 and 10 $\mu\text{g}/\text{m}^3$ were reported at USC and Riverside, respectively, with high UP concentrations during midday and evening. Levogluconan (a marker for vegetative burning) was often not detectable, with highest average of 50 ng/m^3. Cholesterol (a marker for cooking) ranged from 0.01-0.2 ng/m^3. 1,2-benzenedicarboxylic acid (a SOA indicator) ranged from 0.01-2 ng/m^3. Use of specific ng compounds at specific time intervals and location to imply source origins. <p>* Average concentrations were approximated from histograms.</p>	<ul style="list-style-type: none"> Defined UP as <180 nm. No quantitative data was presented. Particle mass distributions suggested photochemical origins during summer at both sites and during winter at the Riverside site. Particle growth was not significant during dry and hot summer, owing to the hydrophobic nature of EC and nonpolar organics in UP. Abundant hopanes (0.2-1.1 ng/m^3) in UP during the morning at the USC site suggested fresh vehicle emissions (i.e., from motor oil). Abundant BaP (0.05-1.5 ng/m^3) and COR found in UP suggested poor spark-ignition combustion in gasoline-fueled vehicles.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Gasparini <i>et al.</i> (2004)	<ul style="list-style-type: none"> One site at Aldine, TX (north and downwind of Houston metropolitan area) as part of the U.S. EPA Supersite program. From 6/2001 to 10/2001. 	<ul style="list-style-type: none"> Two HF-DMAs alternating operation between single DMA mode for size distribution and TDMA mode for measurements of size-resolved aerosol properties such as hygroscopic growth and volatility for particle size range of $25 < d_p < 700$ nm. Hygroscopic measurements made with TDMA were analyzed in eight sizes: 25, 36, 53, 77, 122, 163, 236, and 344 nm. 	<ul style="list-style-type: none"> Based on hygroscopicity, separate particles into four categories: pure and mixed insolubles and pure and mixed solubles. Data presented in mass and 3-D volume distribution using 11-point second order Savitzky-Golay smoothing function, TDMA growth factor distributions, and size-resolved hygroscopicity. 	<ul style="list-style-type: none"> Divided UP composition into four categories based on their hygroscopicity: <ul style="list-style-type: none"> pure and mixed water solubles: <ul style="list-style-type: none"> inorganic salts, acids, and oxidized organics pure and mixed water insolubles: <ul style="list-style-type: none"> soot, organic compounds, metals, and dust. 	<ul style="list-style-type: none"> By combining size distribution with aerosol hygroscopicity, the size-resolved aerosol composition was inferred. Vehicle related OC and EC emissions during morning traffic hours resulted in simultaneous increase in pure insoluble material throughout the size ranges up to 344 nm. Particles > 0.1 μm exhibited bimodal growth, with increasing importance of the hygroscopic mode with increasing dry particle size. During two episodes, pronounced increase in small particle concentrations were followed by particle growth. It appeared that condensation of organic compounds was responsible for the initial growth, while condensation of inorganic compounds was responsible for the continued growth. Morning traffic resulted in an increase in pure insoluble material throughout the 700 nm size range.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Hasegawa <i>et al.</i> (2004)	<ul style="list-style-type: none"> Two sites in Japan: <ul style="list-style-type: none"> A roadside site at the Ikegami-Shincho crossing in Kawasaki City, Kanagawa Prefecture An ambient suburban site at the National Institute for Environmental Studies in Tsukuba City, Ibaraki Prefecture. From 2/18-27/2002 (Period I) and from 9/20/2002 to 10/1/2002 (Period II) at Kawasaki, and from 3/1/2002 to 3/14/2002 at Tsukuba. 	<ul style="list-style-type: none"> An SMPS consisting of DMA (Model 3081) and CPC (model 3025A) for 10 min scan of 18.4 <math>d_p</math> < 865 nm in 107 channels. A thermal denuder (Dekati), heated at 250°C preceded DMA (Grimm Model 5.5-900) and CPC (Grimm Model 5.403) for 14.9 <math>d_p</math> < 742 nm during Period II. 13 stage ALPI (Tokyo Dylec Corp. Model LP-20).* Hourly NO, NO₂, O₃, SPM (PM₁₀) (period I) and 3-hr average OC and EC by R&P Model 5400 (Period II). 	<ul style="list-style-type: none"> Acquired OC and EC for 0.06 <math>d_p</math> < 11 μm at 11 stages. Data presented in carbon size distribution, number distribution, wind pollution roses, and temporal variations. 	<ul style="list-style-type: none"> At 250°C the peak at 30 nm (containing volatile species such as NO₃⁻, SO₄⁻ and organics) disappeared. The second peak at 90 nm (consisting of solid material, mainly soot and EC) remained. EC/TC ratio was larger for <math>d_p > 60</math> nm as compared to those in the 60-120 nm ranges, suggesting abundance of volatile compounds for <math>d_p > 60</math> nm. 	<ul style="list-style-type: none"> Increase in UP corresponded to total traffic and high concentration of diesel engine vehicles. Particle concentration for 18.4 <math>d_p < 50</math> nm increased in the morning under calm winds, which corresponded to the increased EC and NO. The 30 nm peaks at the suburban Tsukuba site in the afternoon implies continued impacts from vehicle exhaust as well as photochemical reactions. Thermal denuder suggested that most UP evaporated at 250°C.

* See Table 1 for cut points.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																																														
Kawanaka <i>et al.</i> (2004)	<ul style="list-style-type: none"> One site at Saitama University in Saitama City, north of Tokyo, Japan. 42 days during 1/16/2003 to 2/27/2003 with filter change occurring every week. 	<ul style="list-style-type: none"> 12-stage ALPI (Tokyo Dylec Model LP-20)* for $0.11 < d_p < 11 \mu\text{m}$. * See Table 1 for cut points. 	<ul style="list-style-type: none"> Acquired 9 PAHs after DCM extraction (i.e., BbF, BkF, BaP, BeP, PER, IND, DBahA, BghiP, COR) by GC/EI-MS, and 2-NF by GC/NCI-MS. Mutagenicity determined by Ames test using <i>Salmonella typhimurium</i> strains TA98 and YG1024 under conditions with and without S9 mix. Data presented in tables as percent cumulative frequency and in histogram for particle size distribution. 	<table border="1"> <thead> <tr> <th>Type of PAH</th> <th>PAH** (ng/g PM)</th> <th>UP Cumulative frequency of PAH and 2-NF</th> </tr> </thead> <tbody> <tr> <td>Total PAH</td> <td>170</td> <td>NA</td> </tr> <tr> <td>BbF</td> <td>32</td> <td>3.6</td> </tr> <tr> <td>BkF</td> <td>26</td> <td>3.4</td> </tr> <tr> <td>BeP</td> <td>23</td> <td>3.6</td> </tr> <tr> <td>BaP</td> <td>21</td> <td>3.1</td> </tr> <tr> <td>PER</td> <td>5</td> <td>4.1</td> </tr> <tr> <td>IND</td> <td>22</td> <td>3.2</td> </tr> <tr> <td>DBahA</td> <td>3.2</td> <td>2.9</td> </tr> <tr> <td>BghiP</td> <td>27</td> <td>4.2</td> </tr> <tr> <td>COR</td> <td>16</td> <td>4.8</td> </tr> <tr> <td>2-NF</td> <td>9</td> <td>5.8</td> </tr> </tbody> </table> <table border="1"> <thead> <tr> <th>Type of Mutagenicity</th> <th>Mutagenicity/PM (Revertants/mg)**</th> </tr> </thead> <tbody> <tr> <td>TA98-S9</td> <td>1,050</td> </tr> <tr> <td>TA98+S9</td> <td>800</td> </tr> <tr> <td>YG1024-S9</td> <td>9,000</td> </tr> <tr> <td>YG1024+S9</td> <td>2,500</td> </tr> </tbody> </table>	Type of PAH	PAH** (ng/g PM)	UP Cumulative frequency of PAH and 2-NF	Total PAH	170	NA	BbF	32	3.6	BkF	26	3.4	BeP	23	3.6	BaP	21	3.1	PER	5	4.1	IND	22	3.2	DBahA	3.2	2.9	BghiP	27	4.2	COR	16	4.8	2-NF	9	5.8	Type of Mutagenicity	Mutagenicity/PM (Revertants/mg)**	TA98-S9	1,050	TA98+S9	800	YG1024-S9	9,000	YG1024+S9	2,500	<ul style="list-style-type: none"> Define UP as $d_p < 0.11 \mu\text{m}$, fine particles as $d_p < 2.1 \mu\text{m}$, and PM_{course} as $2.1 < d_p < 11 \mu\text{m}$. UP was of 1.5% of PM₁₁. 9 PAHs, total PAH, 2-NF, and mutagenicity were unimodal with a peak at 0.48-0.68 μm. Highest PAHs, 2-NF and mutagenicity per unit PM mass were found in UP fractions, suggesting UP were efficient at carrying mutagenic compounds which cause adverse health effects. Based on size distribution of PM between 0.11 and 11 μm, 2.9-5.8% of PAHs and 2-NF, and 5.1-5.9% of mutagenicity were found in UP. Attributed PAH to emissions from combustion sources.
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BbF	32	3.6																																																	
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BeP	23	3.6																																																	
BaP	21	3.1																																																	
PER	5	4.1																																																	
IND	22	3.2																																																	
DBahA	3.2	2.9																																																	
BghiP	27	4.2																																																	
COR	16	4.8																																																	
2-NF	9	5.8																																																	
Type of Mutagenicity	Mutagenicity/PM (Revertants/mg)**																																																		
TA98-S9	1,050																																																		
TA98+S9	800																																																		
YG1024-S9	9,000																																																		
YG1024+S9	2,500																																																		

** approximated from histogram in Figures 2 and 4 of Kawanaka *et al.* (2004).

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Ma <i>et al.</i> (2004)	<ul style="list-style-type: none"> One site at Kansai, Japan, with industrial parks such as textiles, general machinery, and chemical/metal products with a 45 km radius. Two times per day (0900-1800 and 1800-0900 LT) from 12/10/2002 to 12/25/2002. 	<ul style="list-style-type: none"> Two 13-stage LPAIs* for $0.01 < d_p < 10.85 \mu\text{m}$. OPC (Model KC-01D, RION) for 15-min average with cut point at 0.3, 0.5, 1.0, 2.0, 5.0, and 7.5 μm. Hourly SO_2 (GFS-32, DKK) NO_x (GLN-32, DKK), and $\text{PM}_{2.5}$ TEOM. 	<ul style="list-style-type: none"> Acquired mass, elements by PIXE, and carbon (OC and EC) by TOR. Data presented in percent of UP particle size distribution by elements in three-dimensional (3-D) size distribution of OC and EC, and used factor analysis to identify source contribution. 	<ul style="list-style-type: none"> 3-D graphs show low UF metal concentration with abundant S ($\sim 2.00 \text{ ng/m}^3$), detectable K ($< 100 \text{ ng/m}^3$), and trace levels ($< 0.2 \text{ ng/m}^3$) of Mn and Cr, indicative of gas-to-particle conversion in the atmosphere and by the condensation of gas vaporized in the combustion process. 	<ul style="list-style-type: none"> Only one LPAI stage contained UP. Peak EC concentration was in the range of 0.12-0.29 μm, while peak OC was found at 0.29-0.67 μm. High OC/EC ratio suggested that organic compounds were emitted as primary particles from combustion sources in Kansai. No specific source apportionment results were given for UP. Factor analysis indicated automobile exhaust, fossil fuel contribution, refuse incinerations, iron industry, and geological material contributed to $\text{PM}_{2.5}$.

* See Table 1 for cut points.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Miguel <i>et al.</i> (2004)	<ul style="list-style-type: none"> One site at Claremont, CA. 24-hr average samples, (midnight to midnight), sampled once a week from 10/2001 to 7/2002. Samples were composited monthly for analysis (~10 samples) as UP (0-0.18 μm), accumulation (0.18-2.5 μm), and coarse (2.5-10 μm) modes. 	<ul style="list-style-type: none"> A MOUDI (Model 110, used 4 or 5 stages). Meteorological data for wind speed, wind direction, temperature, and relative humidity were acquired. 	<ul style="list-style-type: none"> Acquired mass, carbon (OC and EC), by unspecified method, 12 PAHs extracted with DCM and analyzed by HPLC-fluorescence, and ions (NO_3^- and SO_4^{2-}) by IC (Dionex Model DX-100). 12 PAHs were grouped by sub-cooled vapor pressure: 1) a PHE-FLT semi-volatile PAH group ($\log[P_{\text{L}}^{\circ}]$ from -0.95 to -2.06) including PHE, ANT, PYR, and FLT; and 2) a BAD-IND less volatile and particle-phase PAHs ($\log[P_{\text{L}}^{\circ}]$ -3.22 to -7.04) including BAA, CRY, BgP, BaP, and BbF. Data presented in monthly particle size distribution of PAHs and EC including one UP mode and examined correlations of chemical components with temperature. 	<ul style="list-style-type: none"> EC, OC, and SO_4^{2-} in UP mode accounted for 43%, 24%, and 4% of total EC, OC, and SO_4^{2-} mass, respectively. PHE-FLT and BAA-IND in UP mode accounted for 30% and 40% of the PAH mass. 	<ul style="list-style-type: none"> UP defined as $d_p < 0.18 \mu\text{m}$. EC size distribution did not vary over the study period with abundant UP and accumulation modes. Correlations with temperature were high for SO_4^{2-} and EC in UP mode and OC and NO_3^- in coarse mode. As temperature decreased, particle-phase PAH concentration (i.e., the BAA-IND group) increased with decreasing volatile species, suggesting the increased partitioning from vapor phase and increased photostability during the winter. Similar size distributions were found for monthly composite target PAHs from October to February. Coarse fraction PAHs were found from March to July.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Tolocka <i>et al.</i> (2004b)	<ul style="list-style-type: none"> One site at southeast Baltimore, MD, as part of the U.S. EPA Supersite program. Continuous sampling from 4/2002 to 5/2002 to obtain two-hr average spectra with ~2,000 particles/day. 	<ul style="list-style-type: none"> An RMS III (45 < d_p < 1,250 nm), with a laser ablation time-of-flight MS, characterizing single particles with positive/negative ion detection for ~110 min scan by LDI/MS. A continuous NO_3^- monitor (by flash vaporization). 	<ul style="list-style-type: none"> Particles in nine sizes were selected (45, 50, 90, 115, 140, 220, 440, 770, and 1,250 nm). Metals are categorized into: 1) pure, 2) mixed, and 3) unconfirmed classes. Evaluated positive ion spectra using m/z 30 (NO^+) as indicator for NO_3^-, and calculated NO_3^- from the hit rate and number of particles collected in a particular time span. Data presented in time series spectrum, number concentrations, and number by wind roses in eight directions. 	<ul style="list-style-type: none"> No absolute UP concentrations, used ART 2-a neural network algorithm, assigned NO_3^- spectra into "pure" and "internally mixed" classes. Identified events on the basis that concentration increased 10 fold over the baseline concentration levels. Reported pure or fresh (50-90 nm) and aged (110-220 nm) NO_3^- events, coincided with an increase in the number of mixed particles containing NO_3^-, suggesting that they were formed by condensation of NH_4NO_3 onto pre-existing particles. 	<ul style="list-style-type: none"> Obtained approximately 75,000 size particle spectra in the positive ion mode. UP NO_3^- events were found during low temperatures and high RH. Partitioning of NH_4NO_3 to particle phase was influenced by the particle number concentrations and chemical composition during NO_3^- events. V, a marker for residual oil combustion, was observed from all wind directions. Fe and Pb were observed from the east-northeast, whereas As and Pb were observed from the south-southeast.
Fang <i>et al.</i> , (2005; 2006a; 2006b)	<ul style="list-style-type: none"> One traffic sampling site, west of Taichung, Taiwan. Three-day (~4,320 min) average samples acquired from 11/2004 to 1/2005. 	<ul style="list-style-type: none"> One MOUDI (Model 100; $0.056 < d_p < 18 \mu\text{m}$) in 10 stages. One nano-MOUDI (Model 115); $10 < d_p < 56 \text{ nm}$. 	<ul style="list-style-type: none"> Acquired mass and ions (Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Ca^{2+}, and Mg^{2+}) by IC (Dionex-100) and elements (Mg, Cr, Fe, Cu, Zn, and Pb) by AAS from Teflon filters. 	<ul style="list-style-type: none"> UP concentrations were 39, 35, and $37 \mu\text{g}/\text{m}^3$ for the < 18, 32, and 56 nm stages, respectively. UP ($d_p < 0.1 \mu\text{m}$) consisted of 16.9% of total PM mass ($d_p < 18 \mu\text{m}$). Major ionic species of SO_4^{2-}, NH_4^+, and Mg^{2+} have higher concentrations for $d_p < 56 \text{ nm}$. Cumulative fractions for UP are: 21.1% for SO_4^{2-}, 24% for NH_4^+, and 21% for Mg^{2+}. Cumulative fractions for $d_p < 56 \text{ nm}$ was 12.3 for elements (Mg, Ca, Cr, Fe, Zn, and Pb). 	<ul style="list-style-type: none"> Used cumulative fractions to interpret the UP fraction. Ion concentrations were reported as $\mu\text{g}/\text{g}$, and metal concentrations as mg/g or as $\mu\text{g}/\text{m}^2$ for filter density, inconsistent with mass concentrations of $\mu\text{g}/\text{m}^3$.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Herner <i>et al.</i> (2005; 2006a; 2006b)	<ul style="list-style-type: none"> • Six sites in Northern California: <ul style="list-style-type: none"> – Bodega Bay (coastal) – Davis (urban) – Sacramento (urban) – Modesto (urban) – Bakersfield (urban) – Sequoia National Park (regional background) – Western slope of the Sierra Nevada mountains. • From 12/16/2000 to 2/3/2001 for the following intensive operating periods (IOPs): <ul style="list-style-type: none"> – IOP1: 12/15-18/2000 – IOP2: 12/26-28/2000 – IOP3: 1/4-7/2001 – IOP4: 1/31/2001-2/3/2001 	<ul style="list-style-type: none"> • Two collocated MOUDI (Model 110) at each site. • SMPS measurements at Modesto during 1/4-7/2001. • PM₁₀, PM_{2.5} and PM_{1.8} and gaseous NH₃ and HNO₃ were acquired. 	<ul style="list-style-type: none"> • Acquired mass, carbon (OC and EC) by TOT, ions (Na⁺, K⁺, Ca⁺⁺, NH₄⁺, NO₃⁻, PO₄³⁻, and SO₄²⁻), elements (Al, Si, P, S, Cl, K, Ca, Sc, Ge, Se, Br, I, and Ce) by XRF, and elements (Si, S, Ca, Ni, Cu, and Zn) by ICP/MS. 	<ul style="list-style-type: none"> • OC and EC constituted 90% for d_p < 0.18 µm and ~98% for UP. • Maximum UP of ~2.4 µg/m³ was found at Sacramento and Bakersfield. • Daytime UP concentrations were ~50% lower than nighttime concentrations. • UP concentrations did not increase during multi-week stagnation in the SJV (consistent with coagulation as a dominant removal mechanism for UP) though PM_{2.5} increased by 7 fold.. 	<ul style="list-style-type: none"> • Two types of particles were identified: hygroscopic SO₄²⁻, NO₃⁻, and NH₄⁺; and less hygroscopic OC and some EC. • Carbon and hygroscopic particles exist separately in the SJV until coagulation mixes them in the accumulation mode. • UP elemental concentrations (ng/filter) were low, typically < 0.1 or 0.01 µg/m³.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																		
Lin <i>et al.</i> (2005)	<ul style="list-style-type: none"> Roadside in an unspecified city in southern Taiwan. 13 samples collected during seven sampling periods from 2/2004 to 4/2004. 	<ul style="list-style-type: none"> MOUDI (Model 110) and nano-MOUDI (Model 115) with Si grease applied. 	<ul style="list-style-type: none"> Mass and elements (Na, Mg, Al, Si, Ca, V, Cr, Mn, Fe, Ni, Cu, Zn, Sr, Ag, Cd, Sb, Ba, and Pb) by ICP/MS (Agilent 7500 series) and Si by ICP/AES (Perkins Elmer Optima 2000 DV). 	<p>Nanoparticles ($10 < d_p < 56$ nm) contain greater quantities of traffic-related metals (Pb, Cd, Cu, Zn, Ba, and Ni).</p> <ul style="list-style-type: none"> UP by mass were: <table border="1" style="margin-left: 20px;"> <thead> <tr> <th>Element</th> <th>% of UP mass in nanoparticles</th> </tr> </thead> <tbody> <tr><td>V</td><td>38%</td></tr> <tr><td>Cr</td><td>28%</td></tr> <tr><td>Ni</td><td>30%</td></tr> <tr><td>Zn</td><td>22%</td></tr> <tr><td>Ag</td><td>37%</td></tr> <tr><td>Cd</td><td>50%</td></tr> <tr><td>Sb</td><td>64%</td></tr> <tr><td>Pb</td><td>24%</td></tr> </tbody> </table>	Element	% of UP mass in nanoparticles	V	38%	Cr	28%	Ni	30%	Zn	22%	Ag	37%	Cd	50%	Sb	64%	Pb	24%	<ul style="list-style-type: none"> Reported nanoparticles ($d_p < 10$-56 nm) and UP measurements of 23 ± 11 and $31 \pm 19 \mu\text{g}/\text{m}^3$, respectively.
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				<ul style="list-style-type: none"> Ag, Cd, Sb are more abundant in nucleation/condensation mode. V, Zn, Ag, Cd, Sb, Ba, and Pb are abundant in UP diesel exhaust, while Mn, Cu, and Sr are abundant in UP gasoline exhaust. 																			

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																																																																																																				
Sardar <i>et al.</i> (2005)	<ul style="list-style-type: none"> Four sites in CA: <ul style="list-style-type: none"> USC (Source site) Long Beach (LB) (Source site) Upland (UL) (receptor site) Riverside (RS) (receptor site). Sampled 14 days at each site over three seasons: <ul style="list-style-type: none"> Fall (9/2002 to 11/2002) Winter (12/2002 to 3/2003) Summer (6/2003 to 8/2003). ~12 samples (one 14-day integrated sample x 4 sites x 3 seasons). 	<ul style="list-style-type: none"> A modified MOUDI (Model 110), Nano-MOUDI (Model 115-1), and an ELPI (Model 393501). An SMPS/CPC system (Models 3966/3022A) every 15 min scans for 22 <math>d_p < 948\text{ nm}</math> during fall 2002 at Riverside and Long Beach, during winter 2002 at USC and during summer 2003 at Riverside. Hourly gases NO, NO₂, CO and O₂. 	<ul style="list-style-type: none"> Acquired 14-day integrated mass, carbon (OC and EC) by TOT, and ions (NO₃⁻ and SO₄²⁻) by IC. Data presented in tables, percent contribution to total, histogram, and time series plots. 	<p>UP mass ranged 0.8 to 3.5 μg/m³, peaking during fall.</p> <p>UP composition (ng/m³)</p> <table border="1"> <thead> <tr> <th>Observables</th> <th>USC</th> <th>LB</th> <th>UL</th> <th>RS</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>3490</td> <td>3100</td> <td>2870</td> <td>2890</td> </tr> <tr> <td>OC</td> <td>1980</td> <td>2150</td> <td>1880</td> <td>1450</td> </tr> <tr> <td>EC</td> <td>1190</td> <td>181</td> <td>153</td> <td>704</td> </tr> <tr> <td>NO₃⁻</td> <td>20</td> <td>35</td> <td>27</td> <td>70</td> </tr> <tr> <td>SO₄²⁻</td> <td>204</td> <td>154</td> <td>47</td> <td>61</td> </tr> </tbody> </table> <table border="1"> <thead> <tr> <th colspan="5">Winter</th> </tr> <tr> <th>Observables</th> <th>USC</th> <th>LB</th> <th>UL</th> <th>RS</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>1490</td> <td>1240</td> <td>1450</td> <td>1310</td> </tr> <tr> <td>OC</td> <td>902</td> <td>711</td> <td>1050</td> <td>588</td> </tr> <tr> <td>EC</td> <td>278</td> <td>104</td> <td>21</td> <td>38</td> </tr> <tr> <td>NO₃⁻</td> <td>< 2.8</td> <td>< 3.0</td> <td>< 2.6</td> <td>< 2.0</td> </tr> <tr> <td>SO₄²⁻</td> <td>< 2.8</td> <td>130</td> <td>< 2.6</td> <td>< 2.0</td> </tr> </tbody> </table> <table border="1"> <thead> <tr> <th colspan="5">Summer</th> </tr> <tr> <th>Observables</th> <th>USC</th> <th>LB</th> <th>UL</th> <th>RS</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>2050</td> <td>860</td> <td>1420</td> <td>1530</td> </tr> <tr> <td>OC</td> <td>658</td> <td>378</td> <td>476</td> <td>508</td> </tr> <tr> <td>EC</td> <td>28</td> <td>15</td> <td>66</td> <td>124</td> </tr> <tr> <td>NO₃⁻</td> <td>< 1.5</td> <td>< 1.0</td> <td>< 1.5</td> <td>64</td> </tr> <tr> <td>SO₄²⁻</td> <td>495</td> <td>156</td> <td>203</td> <td>209</td> </tr> </tbody> </table>	Observables	USC	LB	UL	RS	Mass	3490	3100	2870	2890	OC	1980	2150	1880	1450	EC	1190	181	153	704	NO ₃ ⁻	20	35	27	70	SO ₄ ²⁻	204	154	47	61	Winter					Observables	USC	LB	UL	RS	Mass	1490	1240	1450	1310	OC	902	711	1050	588	EC	278	104	21	38	NO ₃ ⁻	< 2.8	< 3.0	< 2.6	< 2.0	SO ₄ ²⁻	< 2.8	130	< 2.6	< 2.0	Summer					Observables	USC	LB	UL	RS	Mass	2050	860	1420	1530	OC	658	378	476	508	EC	28	15	66	124	NO ₃ ⁻	< 1.5	< 1.0	< 1.5	64	SO ₄ ²⁻	495	156	203	209	<ul style="list-style-type: none"> UP defined as $d_p < 180\text{ nm}$, assuming particle density 1.6 g/cm³, equivalent mobility diameter is 130 nm. Converted SMPS number to mass concentrations by assuming density of 1.6 g/cm³. Highest UP and EC mass found at USC, probably due to the impact of nearby freeway traffic with diesel vehicle emissions. UP composition varied in the range of 52-69% for OC, 1-34% for EC, 0-4% for NO₃⁻, and 0-24% for SO₄²⁻. Average UP across sites and seasons were 52% OC, 10% EC, 1% NO₃⁻, and 8% SO₄²⁻. Found OC mode at 18-56 nm during summer, suggesting photochemical SOA formations.
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Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments
Huang <i>et al.</i> (2006)	<ul style="list-style-type: none"> • Zhuijiang Tunnel (near Guangzhou, Southern China). • 12 sampling periods from 12/20/04 to 1/20/05 and nine periods from 7/20/04 to 8/20/04. • Tunnel length: 1,238 m (721 m underwater). • Tunnel Bores: Two, with three lanes in one traffic direction. • Average Vehicle Speed: 15-50 km/hr. • 94,173 vehicles tracked over six sampling periods: <ul style="list-style-type: none"> - 18% heavy duty - 57% light duty - 25% motorcycles. 	<ul style="list-style-type: none"> • MOUDI (10 stages). 	<ul style="list-style-type: none"> • Carbon (OC and EC) by NIOSH method • Ions (Cl⁻, NO₃⁻, SO₄⁼, Na⁺, K⁺, Mg⁺⁺, Ca⁺⁺, NH₄⁺) by IC. 	<ul style="list-style-type: none"> • Reconstructed mass showed abundance of OC and EC in UP with few percentage for ions. 	<ul style="list-style-type: none"> • MMD for EC at 0.42 μm for tunnel as compared to 0.1 μm in the literature. • No mass was measured. • Diesel vehicle exhaust included high engine loads and low combustion efficiency. • Residence time and optical properties of EC in South China were difficult to measure. • Internal mixing of EC with SO₄⁼ led to faster atmospheric removal by wet deposition.

Table 2. Continued.

Study	Sampling Location, Period, and Number of Samples	Measurement Methods	Observables and Data Presentation Method	Relative UP Abundances	Comments																																																
Watson <i>et al.</i> (2007)	<ul style="list-style-type: none"> Two sites in California's San Joaquin Valley <ul style="list-style-type: none"> Fresno (urban) Angiola (rural) From 12/26/2000 to 2/3/2001 for the following intensive operating periods (IOPs): <ul style="list-style-type: none"> IOP2: 12/26-28/2000 IOP3: 1/4-7/2001 IOP4: 1/31/2001-2/3/2001 34 samples collected during: <ul style="list-style-type: none"> 0000-0500 (6) 0500-1000 (10) 1000-1600 (10) 1600-2400 (8) 	<ul style="list-style-type: none"> Three collocated MOUDI (Model 110; 8 stages) at each site. PM_{2.5} collocated filter samples were acquired. 	<ul style="list-style-type: none"> Acquired mass, carbon (OC and EC) by IMPROVE TOR/TOT method, ions (Cl⁻, NO₃⁻, SO₄²⁻, Na⁺, K⁺, NH₄⁺) and elements (Na-U) by XRF. 	<ul style="list-style-type: none"> UP: 56 nm < d_p < 100 nm. <table border="1"> <thead> <tr> <th>Observables</th> <th colspan="2">Average ± Standard Deviation (µg/m³)</th> </tr> <tr> <td></td> <th>Angiola</th> <th>Fresno</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>4.4 ± 3.5</td> <td>2.4 ± 1.3</td> </tr> <tr> <td>OC</td> <td>2.9 ± 3.3</td> <td>2.2 ± 1.2</td> </tr> <tr> <td>EC</td> <td>0.55 ± 0.26</td> <td>0.47 ± 0.30</td> </tr> <tr> <td>NO₃⁻</td> <td>ND</td> <td>0.11 ± 0.07</td> </tr> <tr> <td>SO₄²⁻</td> <td>ND</td> <td>ND</td> </tr> <tr> <td>Fe</td> <td>0.038 ± 0.054</td> <td>0.086 ± 0.093</td> </tr> </tbody> </table> <table border="1"> <thead> <tr> <th>Observables</th> <th colspan="2">Average ± Standard Deviation (UP% of Total)</th> </tr> <tr> <td></td> <th>Angiola</th> <th>Fresno</th> </tr> </thead> <tbody> <tr> <td>Mass</td> <td>9.4 ± 9.3</td> <td>3.0 ± 2.1</td> </tr> <tr> <td>OC</td> <td>22 ± 16</td> <td>6.6 ± 3.6</td> </tr> <tr> <td>EC</td> <td>23 ± 9</td> <td>10.5 ± 4.6</td> </tr> <tr> <td>NO₃⁻</td> <td>ND</td> <td>0.34 ± 0.09</td> </tr> <tr> <td>SO₄²⁻</td> <td>ND</td> <td>ND</td> </tr> <tr> <td>Fe</td> <td>17 ± 26</td> <td>14 ± 17</td> </tr> </tbody> </table>	Observables	Average ± Standard Deviation (µg/m ³)			Angiola	Fresno	Mass	4.4 ± 3.5	2.4 ± 1.3	OC	2.9 ± 3.3	2.2 ± 1.2	EC	0.55 ± 0.26	0.47 ± 0.30	NO ₃ ⁻	ND	0.11 ± 0.07	SO ₄ ²⁻	ND	ND	Fe	0.038 ± 0.054	0.086 ± 0.093	Observables	Average ± Standard Deviation (UP% of Total)			Angiola	Fresno	Mass	9.4 ± 9.3	3.0 ± 2.1	OC	22 ± 16	6.6 ± 3.6	EC	23 ± 9	10.5 ± 4.6	NO ₃ ⁻	ND	0.34 ± 0.09	SO ₄ ²⁻	ND	ND	Fe	17 ± 26	14 ± 17	<ul style="list-style-type: none"> Nearly all of the UP was OC and EC at Angiola and Fresno. UP fractions of OC and EC were higher at Angiola (22 and 23%, respectively) than at Fresno (6.0 and 9.9%, respectively).
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NO ₃ ⁻	ND	0.11 ± 0.07																																																			
SO ₄ ²⁻	ND	ND																																																			
Fe	0.038 ± 0.054	0.086 ± 0.093																																																			
Observables	Average ± Standard Deviation (UP% of Total)																																																				
	Angiola	Fresno																																																			
Mass	9.4 ± 9.3	3.0 ± 2.1																																																			
OC	22 ± 16	6.6 ± 3.6																																																			
EC	23 ± 9	10.5 ± 4.6																																																			
NO ₃ ⁻	ND	0.34 ± 0.09																																																			
SO ₄ ²⁻	ND	ND																																																			
Fe	17 ± 26	14 ± 17																																																			

^aSampling and Monitoring Methods.

- Aethalometer..... A paper tape light-absorption measurement of Black Carbon (BC) (Model RTAA-900, Andersen Instruments, Inc. [Thermo Electron, Inc.], Atlanta, GA)
- ALPI..... Andersen Low pressure impactor, operated at 20 L/min, with 12 stages at < 0.06, 0.12, 0.29, 0.48, 0.68, 1.2, 2.1, 3.5, 5.1, 7.6, and 11 μm (Model LP-20, Tokyo Dylec, Tokyo, Japan, (Kasahara *et al.*, 1996; Ma *et al.*, 2001). See Table 1 for different variations of ALPI)
- ATOMF..... Aerosol time-of-flight Mass Spectrometers (Model 3800-030/3800-100, TSI, Inc., St. Paul, MN) to obtain continuous particle number distributions and single particle chemical composition in the size range of $0.2 < dp < 5 \mu\text{m}$
- BLPI..... Berner low pressure impactor with sampling flow rate of 30 L/min. Nominal cut points of eight stages (Berner and Lurzer, 1980)
- CCN Counter..... Cloud Condensation Nuclei Counter (Model 130, Mee Industries, Monrovia, CA)
- CPC..... Condensation Particle Counter, also termed Condensation Nuclei Counter (CNC) including: a) TSI (Models 3022A and 3760, TSI, Inc., Shoreview, MN), and b) Grimm (Model 5.403, Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany)
- DMA..... Differential Mobility Analyzer including: a) TSI (Model 3071, TSI, Inc., Shoreview, MN), and b) Grimm (Model 5.5-900, Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany)
- HF-DMA..... High-flow differential mobility analyzer (Aerosol Dynamics, Inc., Berkeley, CA; Stolzenburg *et al.*, 1998) Nano DMA TSI (Model 3085, TSI, Inc., Shoreview, MN), selects particles < 150 nm
- EAA..... Electrical Aerosol Analyzer (Model 3030, TSI, Inc., Shoreview, MN), measures particle number concentrations in the range of $0.017 < dp < 0.250 \mu\text{m}$
- EAS..... Electrostatic aerosol sampler. A two-stage (one charging and one collecting stage) electrostatic precipitator that deposits particles onto collection surface for chemical analysis and microscopic evaluation. (EAS, Model 3100, TSI, Inc., Shoreview, MN)
- ELPI..... Electrical low-pressure impactor (Model 393501, Dekati Ltd. Osuusmylynkatu 13 TSI, Finland), operated at a flow rate of 10 or 30 L/min
- High-Vol..... High volume sampling system with slot impactor and cut point at 0.18 μm at flow rate of 500 L/min (Misra *et al.*, 2002) LPC Laser Particle Counter (Model TF-500, Kanomax, Andover, NJ, Ishizaka and Adhikari, 2003)
- MI..... Mercer impactors, five impaction stages (0.46, 0.88, 1.68, 3.26 and 6.32 nm) (In-Tox products, Albuquerque, NM)
- MOI..... Micro-orifice impactor (MSP Corporation, Minneapolis, MN)
- MOUDI..... Micro-orifice uniform deposit impactor, with eight stages at 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6, and 18 μm (Model 100, MSP Corporation, Minneapolis, MN). Model 110 (MSP Corporation, Minneapolis, MN) is a 10-stage impactor which adds 0.056 and 0.1 μm to the Model 100 stages. Both models operate at 30 L/min
- Modified MOUDI..... Model 110 (MSP Corporation, Minneapolis, MN) with 2/3 of acceleration nozzle plates masked, operated at 10 L/min
- Nano-MOUDI..... Model 115-1 (MSP Corporation, Minneapolis, MN) operated at 10 L/min, with 4 stages at 10, 18, 32, and 56 nm
- OPC..... Optical Particle Counter: a) Particle Measuring Systems (Model ASASP-X, Particle Measuring Systems, Boulder, CO) in 31 channels for the size range $0.09 dp < 2.6 \mu\text{m}$ every five sec, and b) RION (Model KC-01D1, RION, Tokyo, Japan)
- R&P 5400..... Continuous carbon analyzer for OC and EC (Rupprecht and Patashnick Co, Albany, NY)
- RSMS II..... Rapid Single-particle mass spectrometer, Version 2. (Malina *et al.*, 2000; Phares *et al.*, 2001; 2002)
- RSMS III..... Rapid single-particle mass spectrometer, Version 3. A laser ablation time-of-flight mass spectrometer that simultaneously detects positive and negative ions. (Lake *et al.*, 2003; 2004; Tolocka *et al.*, 2005; 2006)
- SDI..... Small deposit area impactor (a 12-stage [0.045, 0.090, 0.150, 0.235, 0.380, 0.580, 0.800, 1.06, 1.61, 2.60, 4.07, and 8.5 μm], low-pressure, multi-nozzle inertial impactor with a deposit area < 8 mm in diameter, 53 deposit areas, operated at 11 L/min (Maenhaut *et al.*, 1996)
- SMPS..... Scanning Mobility Particle Sizer (Model 3936, TSI, Inc., Shoreview, MN)
- TEOM..... Tapered element oscillating microbalance (Series 1400a, Thermo Fisher Scientific, formerly Rupprecht & Patashnick Co., Inc., Albany, NY)

- TDMA Tandem differential mobility analyzer, operating two DMAs in series (Rader and McMurry, 1986)
 UCPC Ultrafine Condensation Particle Counter (Model 3025A, TSI, Inc., St. Paul, MN)
 UDM Ultrafine diffusion monitor, a low-flow rate (200 cm³/min) diffusion monitor that deposits particles by diffusion onto the walls of the channel (Cohen *et al.*, 2000). UP are collected onto two nanofilm detectors and one non-reactive (silicon) detector (detectors are stored under nitrogen after sampling) to quantify both total and acidic particles by atomic force microscopy (AFM)
 USC UPC..... UP Concentrator that enriches particle concentrations by a factor of 40. Air is first drawn at a flow of 2,201 L/min through a saturation-condensation system that grows particles to 2-3 μm droplets, separated by virtual impactors, passed through diffusion dryer, and then returned to original size (Kim *et al.*, 2000a)
 VI Virtual impactor, operated at 16.7 and 1.7 L/min for 2.3 and 15 μm cut point, respectively (Loo and Cork, 1988)

^b **Analysis Methods**

- AAS Atomic Absorption Spectrometry
 GC/EI-MS Gas chromatograph/electron impact ionization-mass spectrometer for organic compounds
 GC/MS Gas chromatograph/mass spectrometry for organic compounds
 GC/NCI-MS Gas chromatograph with negative ion chemical ionizations mass spectrometer for organic compounds
 HPLC High performance liquid chromatography for organic compounds and organic acids
 IC Ion chromatography for anions (Cl⁻, NO₃⁻, PO₄³⁻, SO₄²⁻) and cations (Na⁺, K⁺, NH₄⁺)
 ICM Indophenol colorimetric method for NH₄⁺ analysis
 ICP/MS Inductively coupled plasma/mass spectrometry for elements
 INAA Instrumental neutron activation analysis for elements
 LDI/MS Laser desorption ionization/mass spectrometry
 PIXE Proton-induced X-ray emissions spectrometry analysis for elements
 RFA Alpkem rapid flow analyzer (Model RFA-300) for NH₄⁺ analysis by indophenol colorimetric method
 SAED Selected area electron detection
 TEM Transmission electron microscopy for particle morphology
 TMO Thermal magnesium dioxide carbon analysis method for OC and EC (Fung, 1990)
 TOR Thermal/optical reflectance method for OC and EC (Chow *et al.*, 1993; 2001; 2004; 2005b; 2007)
 TOT Thermal/optical transmission method for OC and EC (Birch and Cary, 1996)
 XRF X-ray fluorescence analysis for elements

^c **Observables**

- CCN Cloud condensation nuclei
 CN Condensation nuclei
 d_p Particle aerodynamic diameter
 HC Hydrocarbon
 MMD Mass medium diameter
 MSA Methane sulfonate
 O_x Oxalate
 PAH Polycyclic aromatic hydrocarbon
 2-NF 2-nitrofluoranthene

ANT anthracene
BAA Benzo[<i>a</i>]anthracene
BaP benzo[<i>a</i>]pyrene
BbF benzo[<i>b</i>]fluoranthene
BeP benzo[<i>e</i>]pyrene
BgP/BghiP	.. benzo[<i>g,h,i</i>]perylene
BkF benzo[<i>k</i>]fluoranthene
COR coronene
CRY chrysene
DBahA dibenz[<i>a,h</i>]anthracene
FLT fluoranthene
IND Indeno[1,2,3- <i>cd</i>]pyrene
PER perylene
PHE phenanthrene
PYR pyrene
PM Particulate matter
SOA Secondary Organic Aerosol
Chemical Species:	
BC Black Carbon
EC Elemental Carbon
OC Organic Carbon
OM Organic Mass (= OC x 1.4)
TC Total Carbon
Cl ⁻ Chloride
NO Nitrogen oxide
NO ₂ Nitrogen dioxide
NO ₃ ⁻ Nitrate
SO ₄ ⁼ Sulfate
NH ₄ ⁺ Ammonium
Na ⁺ Water-soluble sodium
VOC Volatile Organic Compounds
(NH ₄) ₂ SO ₄ Ammonium sulfate
NH ₄ NO ₃ Ammonium nitrate
NH ₄ Cl Ammonium chloride
NH ₄ HSO ₄ ⁼	.. Ammonium bisulfate
Ca ⁺⁺ Water-soluble calcium
H ₂ SO ₄ Sulfuric Acid
CaCO ₃ Calcium carbonate
NaCl Sodium chloride
O ₃ Ozone

Elements:

B..... Boron
 Al..... Aluminum
 Mg..... Magnesium
 Si..... Silicon
 K..... Potassium
 Ti..... Titanium
 V..... Vanadium
 Cl..... Chlorine
 Cr..... Chromium
 Mn..... Manganese
 Fe..... Iron
 Co..... Cobalt
 Ni..... Nickel
 Cu..... Copper
 Zn..... Zinc
 Se..... Selenium
 Sr..... Strontium
 Mo..... Molybdenum
 Ag..... Silver
 Pb..... Lead
 Ba..... Barium
 La..... Lanthanum
 Ce..... Cerium

SPM..... Suspended particulate matter with a cut point at 100% collection efficiency of ~10 µm (~7 µm by 50% collection efficiency, term used in Japan)

UP..... Ultrafine particles

^d Other Abbreviations

AIHL cyclone..... Air Industrial Hygiene Laboratory cyclone, removes particles greater than 1.8 µm at 30 L/min (John and Reischl, 1980)

DCM..... Dichloromethane (CH₂Cl₂)

L.A..... Los Angeles

LDT..... Local daylight time

LST..... Local standard time

ND..... Not detectable

NA..... Not available

PDT..... Pacific daylight time

PST..... Pacific standard time

USC..... University of Southern California

Data reporting

Inconsistent definitions are used for the terms “ultrafine particle” and “nanoparticle.” Many of these definitions are operational, depending on the measurement method. In Table 2, Kawanaka *et al.* (2004) define UP as particles < 110 nm, while Geller *et al.* (2002), Fine *et al.* (2002), Miguel *et al.* (2004) and Sardar *et al.* (2005) define UP as < 180 nm. Chow *et al.* (2005c) recommended the use of < 10 nm for nanoparticles and < 100 nm for UP in the physical sciences.

Many of the references report their results graphically rather than in tables, thereby making it difficult to obtain quantitative concentrations for the different chemical species. Several reporting methods include stacked bar charts of the composition for the different size fractions in which only the major components could be identified. Some references report pie charts with percentage contributions of each chemical component. These had to be deconvoluted to obtain the absolute concentrations reported in Table 2, and this was only possible when the UP mass was reported with the pie chart. Several of the studies report only a few of the UP size fractions rather than the total for all particles less than 100 nm. The most straightforward references were those that reported the results in a table for each of the species measured and each stage of size-segregation, such as that of Pakkanen *et al.* (2001b).

Data are qualitative or semi-quantitative for the aerosol mass spectrometer studies (Phares *et al.*, 2003; Rhoads *et al.*, 2003; Tolocka *et al.*, 2004a; 2004b). Zhang *et al.* (2004) show that OC is a major component of small particles, demonstrated by the percentage of nucleation events. Erdmann *et al.* (2005) classify single particle spectra, but this is often subjective and it is not possible to compare the results among different investigators, even when they are using the same instrument. Tolocka *et al.* (2004a) shows a clear directionality of UP metals using the short-time resolution available from the single particle mass spectrometer, in which most of the UP Fe is coming from a source to the northeast, whereas most of the UP arsenic (As) derives from sources to the southeast. This type of data can be correlated with source locations to better identify and quantify contributors to excessive concentrations.

Monitoring locations

Of the 25 studies summarized in Table 2, eight (Hughes *et al.*, 1998; 1999; Cass *et al.*, 2000; Geller *et al.*, 2002; Kim *et al.*, 2002; Fine *et al.*, 2004; Miguel *et al.*, 2004; Sardar *et al.*, 2005) are from different locations within the Los Angeles metropolitan area. In central California, Chung *et al.* (2001) sampled at urban Bakersfield and Watson *et al.* (2007) sampled at the urban Fresno and non-urban Angiola sites. Herner *et al.* (2005; 2006a; 2006b) characterized UP at six California locations including the cities of Modesto and Sacramento, a coastal site (Bodega Bay), Sequoia National Park, and the western slope of the Sierra Nevada. All of these used MOUDI impactors for sampling with a combination of XRF, PIXE, or INAA for elements, IC or AC for

water soluble ions, and thermal/optical reflectance (TOR) or transmission (TOT) for OC and EC measurements (Watson *et al.*, 2005). Fine *et al.* (2004) and Miguel *et al.* (2004) quantified PAHs.

Other U.S. cities include Houston, TX (Phares *et al.*, 2003), Atlanta, GA (Rhoads *et al.*, 2003), and Baltimore, MD (Tolocka *et al.*, 2004a; 2004b), where single particle spectrometers were applied. Gasparini *et al.* (2004) reported TDMA measurements from Houston to evaluate hygroscopic properties. MOUDI measurements were also taken by Cohen *et al.* (2004) in New York near the World Trade Center after 9/11/2001 and results are influenced by that event.

In Europe, Pakkanen *et al.* (2001a) measured UP with a BLPI at urban Helsinki and rural Luukii, Espoo sites in Finland. Viidanoja *et al.* (2002a) used SDIs in Helsinki. Four studies on UP were reported in Japan using ALPI with the cut points shown in Table 1. Kawanaka *et al.* (2004) and Ma *et al.* (2004) sampled at Saitama University and in a Kansai industrial area, respectively. Hasegawa *et al.* (2004) coupled ALPI with a thermal denuder near roadways in Kawasaki. Ishizaka and Adhikari (2003) also applied the thermal denuder method in the industrial area of Nagoya and along the Japanese coast at Mikuni to infer cloud condensation nuclei (CCN) composition. In Asia, Lin *et al.* (2005) conducted a roadside study in southern Taiwan using the MOUDI and Nano-MOUDI, while Huang *et al.* (2006) reported on a tunnel experiment in southern China using a MOUDI.

Most of the studies were performed during the late 1990s and early 2000s. The number of samples taken ranges from two to ~50, which is a small data set from which to draw general conclusions. Sampling locations were largely within highly populated urban centers, with a few in non-urban areas for contrast. Sometimes example results from a few interesting samples were reported, while at other times only the all-sample averages were reported.

UP composition

Organic material (OM = OC×1.4 to account for the unmeasured hydrogen, oxygen, nitrogen, and other material that is associated with organic molecules) was the major component of the UP fraction. Hughes *et al.* (1998) found that OM accounted for 40% to 53% of the UP in Los Angeles during 1996. This was consistent with the results of Cass *et al.* (2000), Geller *et al.* (2002), Kim *et al.* (2002), and Sardar *et al.* (2005) for southern California samples. However, Miguel *et al.* (2004) found that 43% of UP was composed of EC while only 24% came from OC. For northern California, Herner *et al.* (2005; 2006a; 2006b) found that OC and EC constitute ~98% of UP mass. OC and EC in UP were also reported by Huang *et al.* (2006) for a southern China tunnel. Different OC and EC levels could be due to the different sampling sites, sampling times, UP cut points, and measurement methods. In any case, OC and EC were major portions of UP in nearly all of the summarized studies.

When the OC fraction was examined for more specific organic compounds, Miguel *et al.* (2004) found that phenanthrene/fluoranthene and benzo[a]anthracene/indeno[1,2,3-cd]pyrene in

UP mode accounted for large fractions of PAH mass in Southern California. These compounds indicate contributions from fossil fuel combustion sources, especially gasoline- and diesel-powered vehicle exhaust. In Saitama, Japan, Kawanaka *et al.* (2004) found PAH in the range of 3–30 ng/g PM and 3–6% of PAH were found in UP. They also found that these samples were highly mutagenic. Mutagenicity was not observed at urban and suburban locations in Guangzhou, South China, especially for heavy PAHs (> 5 rings; Duan *et al.*, 2005). PAH are semi-volatile in nature and can easily condense on other particles. The larger surface-to-volume ratios in the UP fraction probably encourage this condensation. Cohen *et al.* (2004) measured PM_{2.5} PAH as high as 1,500 ng/m³, reflecting the intensity of emissions from fires soon after the 9/11/2001 World Trade Center attack.

Fine *et al.* (2004) sought contributions from cooking and wood burning by using levoglucosan and cholesterol as organic markers for these sources. Their concentrations were low, with levoglucosan undetected in many samples, although it did achieve a value as high as 50 ng/m³ in one UP sample. Cholesterol was also low (< 0.2 ng/m³), which indicated a negligible contribution from meat cooking to UP in Southern California during the study period.

Mass, elements, and ion concentrations were reported by Pakkanen *et al.* (2001a), while only carbon was reported by Viidanoja *et al.* (2002b). Therefore, mass closure was not achieved, although Pakkanen *et al.* (2001a) found organic acids that could indicate biogenic origins. Phares *et al.* (2003) found that amines constituted 2.4% of the UP mass in Houston, TX. Hasegawa *et al.* (2004) found that nearly all of the UP < 30 nm disappeared when heated to 250°C with a thermal denuder for roadway samples. This is consistent with UP being composed of semi-volatile OM that can evaporate from small particles and condense on larger ones, thereby enhancing particle growth (Zhang and Wexler, 2002).

EC was also present in most UP fractions, but not in the lowest size ranges as indicated by the thermal denuder results of Hasegawa *et al.* (2004) and Ishizaka and Adhikari (2003). This is consistent with the laboratory tests reported by Kittelson (1998) that showed most of the UP < 50 nm fraction evaporating after thermal denuding. This might happen with low-sulfur diesel fuels and after-engine soot removal owing to evaporated lubrication oil that condenses after emission and cooling to ambient temperatures (Zielinska *et al.*, 2004; Vaaraslahti *et al.*, 2005). This hypothesis is somewhat supported by the hopane measurements of Fine *et al.* (2004), as hopanes are believed to originate in engine oils.

Non-carbonaceous substances were found to dominate some of the samples, but this may depend on the measurement method. Single particle spectrometers, for example, draw materials into a vacuum, and some of the semi-volatile organic compounds may evaporate prior to detection. These spectrometers collect UP < 50 nm with < 100% efficiency, even with aerodynamic lens inlets, and it appears that much of the OC resides in this fraction. Phares *et al.* (2003) found that silicon oxides constitute 30% of UP, with potassium (K) constituting another

31% of UP. Carbon was only 16% of the UP in Houston, TX. This may be an artifact of the particle spectrometer measurement system, but it might also indicate that some very fine geological material penetrates to the UP fraction. Using an impactor, Chung *et al.* (2001) found large quantities of calcium (Ca) in the UP fraction at Bakersfield, CA. Ca is also an engine oil additive, and this may be the source. Kim *et al.* (2002) found that Fe was the largest metal component in Los Angeles, ranging from 10 to 130 ng/m³. This may be evidence of particle re-entrainment from previous impactor stages, as mentioned above. Ma *et al.* (2004) found K and traces of manganese (Mn) and chromium (Cr) near Japanese industrial sources. At roadside, Lin *et al.* (2005) found vanadium (V), zinc (Zn), silver (Ag), cadmium (Cd), antimony (Sb), barium (Ba), and lead (Pb) are abundant in UP diesel exhaust, while Mn, cerium (Ce), and strontium (Sr) are abundant in UP gasoline exhaust.

Sulfur (S) was an important, but minor, UP component in most of the studies. This is surprising, since sulfuric acid (H₂SO₄) nucleation is considered to be a major formation mechanism of small particles. Once it begins, in fresh combustion exhaust or as the result of photochemical activity, it appears that organic or metal vapors rapidly condense and constitute most of the UP mass.

CONCLUSIONS AND KNOWLEDGE GAPS

UP chemical composition measurements are most commonly acquired with laboratory measurements from the lower stages of cascade impactors. The MOUDI is usually used for these measurements. XRF, PIXE, and INAA are applied for elements, with thermal/optical methods used for carbon. Ions are measured by IC, AC, or AAS. Single particle mass spectrometers are used for semi-quantitative chemical measurements.

Data are reported in different ways by different authors and are not completely comparable across studies. In the U.S., data have been reported for: the Los Angeles area, central (Fresno, Bakersfield), and northern California (Sacramento, Modesto); Houston, TX; Atlanta, GA; Baltimore, MD; and New York, NY. In Finland, data are available from Helsinki and Luukii, Espoo. In Japan, data are available from Nagoya, Mikuni, Kawasaki, Tsukuba, and Saitama. In Asia, data are available for Taiwan and Southern China. Most of these are urban areas, with some results from roadside, industrial, non-urban, coastal, and regional background sites.

Organic material is the most abundant portion of UP in most, but not all samples. Some have high elemental concentrations, especially from industrial sites. K, Ca, and Fe were found to be important elements in some samples. K originates from biomass burning, and Ca is used as an oil additive. Condensed Fe vapors are often found in industrial processes. Much of the UP < 50 nm appears to be semi-volatile, consistent with it being comprised of organic materials such as

hopanes from engine oils or condensed secondary organic aerosol such as organic acids. PAH were abundant in the UP fraction in a few studies.

Sampling inlets and upper and lower size cuts for UP fractions are not consistent or well-defined. Methods are needed to establish UP fractions, then to translate the results from different sampling methods into a common particle size range. Differences in the analytical methods applied for the measurement of OC, EC, and organic compounds need to be better quantified and standardized. More comparison studies are needed among laboratories for particles specific to the UP fraction.

Standardized concentration units, reporting methods, and common data bases are needed to better compare UP composition measurements among sampling sites. These data bases would allow more efficient and consistent comparisons of results from different studies.

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