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Ultrafine Particulate Exposures in Indoor, Outdoor, Personal and Mobile Environments: Effects of Diesel, Traffic, Pottery Kiln, Cooking and HEPA Filtration on Micro-environmental Particle Number Concentration

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We measured ultrafine particulate material (UFPM) number concentrations in a variety of indoor, personal and mobile environments, using a P-Trak condensations particle counter (TSI model 8525). Indoor UFPM levels indicated influx of outdoor air plus indoor combustion sources (e.g. toasting and cooking, yielding UFPM concentrations >500000 particles/cm^3). Influx of UFPM into the indoor environment from unexpected sources (e.g. a home pottery kiln) was readily detected. In a moving passenger car environment, prolonged exposures to UFPM concentrations >100000 particles/cm^3 were observed related to heavy traffic, especially diesel vehicles. Data logging at 1 s revealed transient UFPM peaks not demonstrable with longer interval averaging. Indices of continuous, average and cumulative exposures are presented. This type of sensitive and portable instrumentation should be highly useful in studies of UFPM in mobile and personal micro-environments.

Keywords: cooking; diesel; filtration; pottery kiln; ultrafines

INTRODUCTION

Ultrafine particles, which are defined as particles <0.1 μm in diameter, are on a mass basis only a minor component of the atmospheric aerosol (~1% by weight), but on a number basis they are a major constituent (~86% by count). Ultrafine particles can be generated by a variety of urban/industrial processes, although some specific emitters are recognized as major sources of ultrafines (e.g. diesel vehicles). Toxicological studies suggest that ultrafines have the potential to reach both the upper and lower respiratory tract and to produce adverse health effects. It has been shown that ultrafine particles produce inflammatory effects in the lung that may be the result of oxidative stress. In this study we have utilized a new portable ultrafine particle counter to measure the concentrations of ultrafine particles in numerous environments.

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MATERIALS AND METHODS

A P-Trak condensation particle counter (TSI model no. 8525) was used to assess the number concentration of ultrafine particulate material (UFPM) in various environmental situations. This portable instrument grows UFPM in a chamber containing supersaturated alcohol vapor and subsequently counts them using a laser. Concentrations are measured from 0 to 500000 particles/cm^3 for particles in the size range 0.02–>1.0 μm. Data logging at intervals as short as 1 s facilitates observation of rapid fluxes in UFPM concentrations. This instrument does not measure particle size or chemical composition. Cumulative sampling duration was >200 h, and peak, average and cumulative exposures were examined.

RESULTS

We operated the P-Trak as a mobile and personal sampler under a variety of exposure conditions. Indoor counts ranged from zero at the outlet of a HEPA filter unit to several hundred particles/cm^3 in the home at baseline (Figs 1 and 2). Counts in the

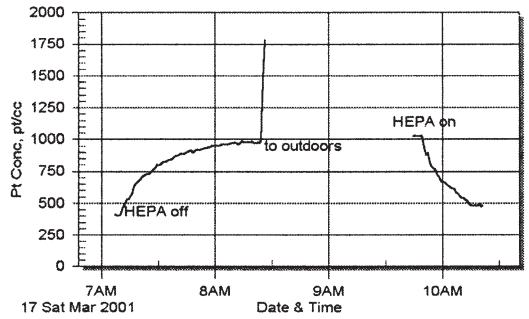

Fig. 1. Effect of a HEPA filtration unit in a residential home bedroom.

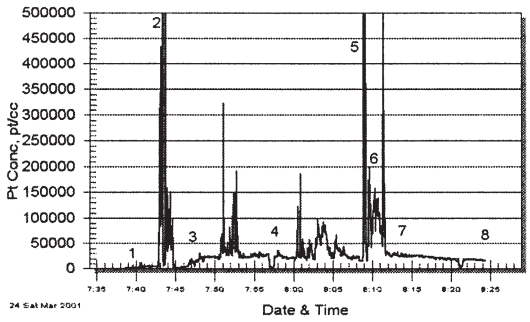

Fig. 2. Kitchen cooking effects on UFPM concentration. A typical sampling run. (1) 7:38 a.m., 1300 early level; move sampler by stove vent, level to 5000 (= outdoor); turn on stove burner (electric ring). (2) Note variation up to max 500000 when sampler moved up and down to breathing zone level before exhaust fan turned on. (3) Level to 6000–2000 on counter top; 7:47 a.m., counter top 22 000. (4) 7:55 a.m., out of kitchen to hallway to upstairs, 1700; back to kitchen, ~25000 by counter top. (5) Up to 35000 and more above stove with exhaust fan on; 8:09 a.m., turned off exhaust fan, meter at breathing level up to 500000+. (6) Levels on counter ~100000+, exhaust fan on, fumes spread; levels by exhaust fan <100000. (7) To counter top, 8:12 a.m., ~30000; 8:20 a.m., counter; back to stove, vent off; hallway, living room, kitchen, 18000. (8) Outside 8:28 a.m.; into car 8:30 a.m.; ~3400 inside car, then up to ~5000 as equilibrates with outside air.

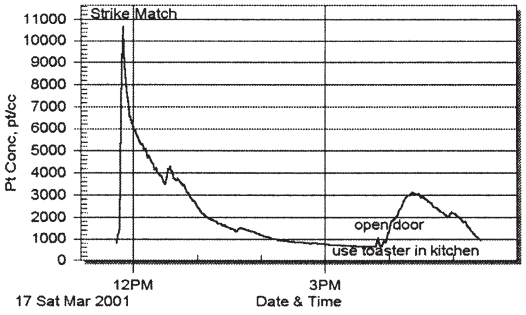

Fig. 3. Point source UFPM, decay and toaster UFPM dispersion.

home increased as doors were opened and were generally related to influx of outdoor air, where baselines of from 2000 to >10000 particles/cm^3 were observed. Cooking activities such as toasting caused concentrations to exceed the maximum detectable (500000 particles/cm^3); the results clearly demonstrate dispersion of the particles throughout the home (having forced air continuous ventilation) and the time course of clearance (Figs 2 and 3).

Re-entrainment of UFPM from the exhaust of a well-ventilated ceramics kiln (electric) was detected and corrected (Fig. 4a). In addition, polycarbonate filter samples from the kiln exhaust were examined by scanning electron microscopy/energy dispersive X-ray spectroscopy (SEM/EDS) (Fig. 4b), which revealed few ultrafine particles within the resolution limits of the SEM (>100 nm). However, aggregates of UFPM consistent with origin from the ceramics and glaze materials were identified, as well as fine sulfates, as seen in the ambient air.

In a mobile environment inside a passenger vehicle (usually with the windows closed), there was rapid penetration of UFPM from outdoors and passing diesel vehicles (school buses and trucks) resulted in a rise from baseline up to >500000 particles/cm^3 (Fig. 5a and b). Note a sharp peak at the end of the sampling shown in Fig. 5b, as the P-Trak was carried past a small area set aside for smokers, near the entrance to the building.

A summary of UFPM data from the first 67 sample collections (indoors and outdoors) is presented in Fig. 6a and b. In addition to the average data (Fig. 6a), an index of cumulative exposure is presented, calculated by multiplying the average UFPM number concentration by the duration (min) of each sample (Fig. 6b).

Nearly continuous sampling (except for the time required to change batteries and recharge the alcohol chamber) during two round trips from Syracuse to New York and back using the same route (~900 km round trip) revealed large variations in UFPM counts, mostly associated with diesel truck traffic immediately ahead of the passenger car and with generally heavy stop-and-go urban traffic (see Fig. 7a–c). The average particle concentration over the 12–13 h for the second trip (not shown) was remarkably similar to that for the first trip (Table 1).

Prolonged exposures to concentrations >100000 particles/cm^3 commonly occur in traffic behind or near diesel vehicles and persistent levels on highways increase with general traffic volume. Acute upper respiratory symptoms (headache, nasal/sinus congestion and rhinitis) were noted by both driver and passenger in the car at very high exposure levels (>150000 particles/cm^3). The sensory recognition of these symptoms occurred within <1 min of the sharp increases associated with diesel traffic.

Table 1. Summary UFPM data from three segments of a round trip from Syracuse to New York and back

Trip		A	B	C
Sampling duration	Hours	3:42	2:59	6:52
	Minutes	222	179	412
Start time		7:39	11:22	14:40
Stop time		11:22	14:21	21:32
Average	particles/cm^3	28145	31779	29891
Cumulative	particles/cm^3 min	6248190	5688441	12315092
Maximum	particles/cm^3	158000	128600	190200
Min	particles/cm^3	23870	4320	6204

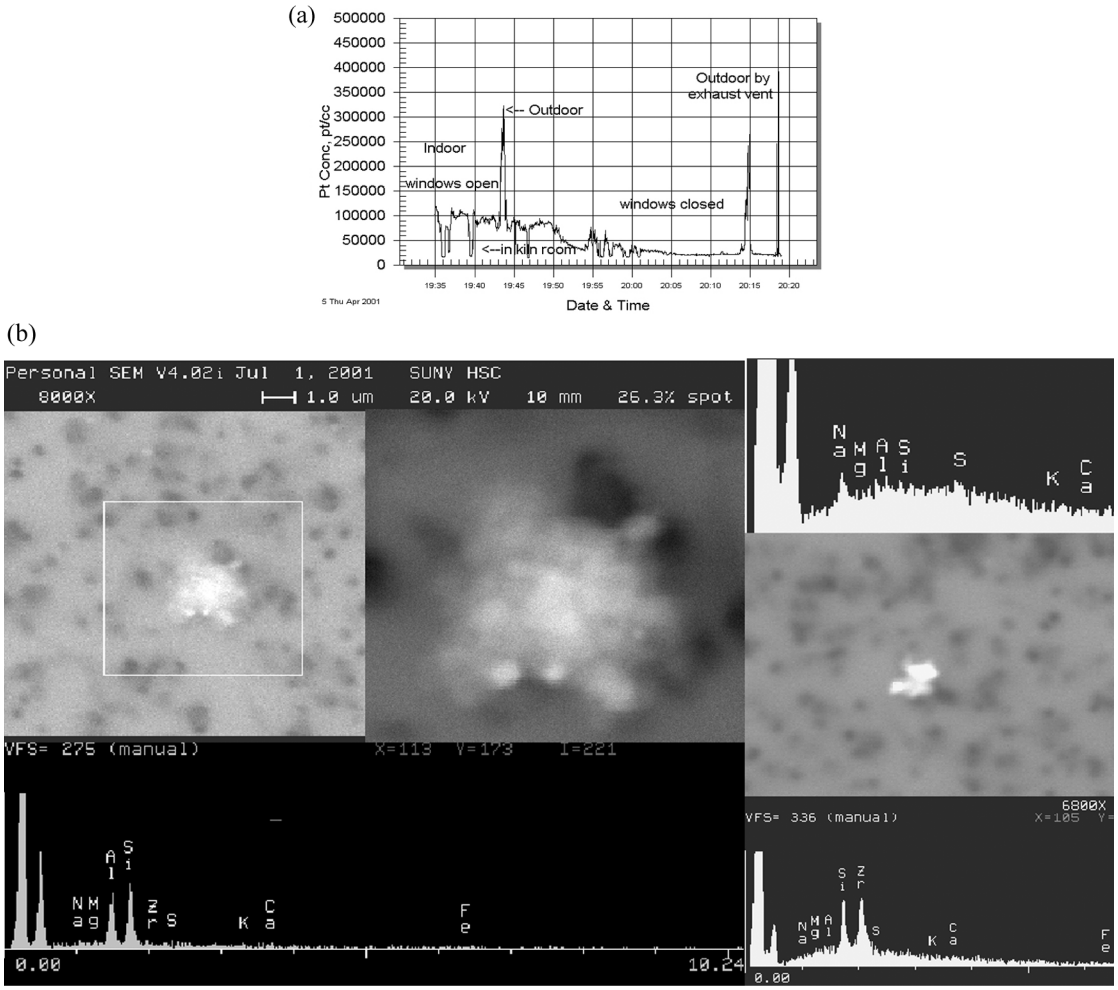

Fig. 4. (a) Re-entrainment of UFPM from kiln exhaust. (b) Examples of particles in kiln exhaust; 30 min sample. Aluminum silicate aggregate, sulfates (ambient) and zirconium silicate.

DISCUSSION

There is scant data on particle numbers in indoor environments. Abraham (1998a) reported direct SEM counting of fine PM on membrane filters from indoor environments and the effects of use of a HEPA filter system. He counted particles limited by the resolution of SEM to ~>100 nm and sampled over 24–48 h. Our tests of UFPMs used the same rooms and HEPA system as he did, but counted particles >20 nm with the TSI P-Trak system, sampled at 1 s intervals. Most (~80%) of the particles counted in his SEM study were carbonaceous, with sulfates the next most prevalent particle (~17%). Abraham demonstrated a signifi-

cant 62% reduction (from 85.5 to 32.5 particles/cm^3) with use of the HEPA filter system. Our studies of UFPMs, with a much finer time resolution than is possible with SEM sampling by membrane filters, confirmed the reduction in particle concentration with HEPA filtration, operating in the same rooms studied by Abraham. Furthermore, by instantaneous monitoring of sources of air from ventilation ductwork and from the outflow of the HEPA filter unit (0 particles/cm^3) it was clear that in a real world situation the measured PM concentration was an equilibrium between incoming air and filtration activity.

In a second study Abraham (1998b) used membrane filter sampling and SEM/EDS analysis to count and analyze sub-micrometer PM indoors and outdoors and with personal sampling. Particles >100 nm were analyzed. Total PM concentrations ranged up to 500/cm^3 and indoor concentrations clearly paralleled those outdoors. Our results with UFPMs, as expected, show higher number concentrations than the SEM filter counting methodology. Similar trends for indoor/outdoor particle concentrations are shown. Other than indoor combustion sources, the major determinant of indoor UFP concentration is clearly the infiltration of outdoor air.

Abraham (1998b) also demonstrated compositional differences between indoor and outdoor sulfate fine particles, with more sulfur-only (most likely ammonium sulfates) or sodium sulfate particles outdoors and more potassium-rich sulfates indoors. Based on our observations on the high concentrations of UFPMs generated by indoor combustion sources, it

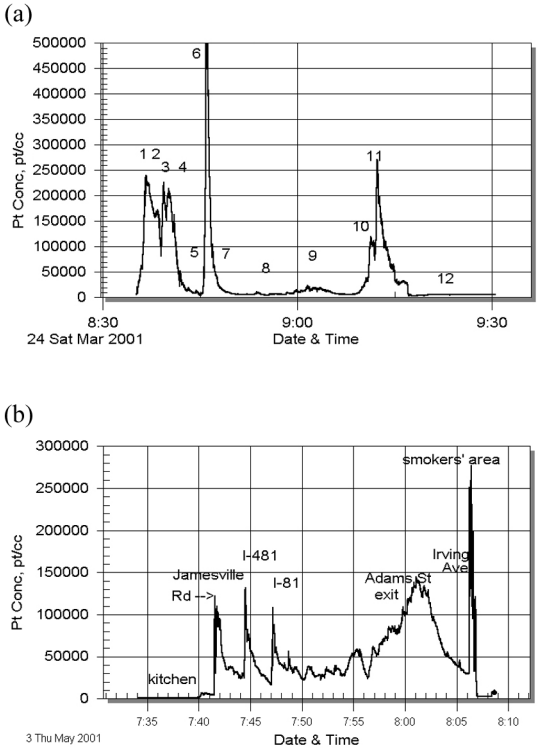

Fig. 5. (a) Sampling UFPM in a mobile environment. (1) Following ~50 ft behind school bus; (2) bus coasting down hill; (3) accelerating up hill; (4) pass bus; (5) at school parking lot; (6) as same bus approaches school parking lot; (7) into village; (8) onto street with light traffic; (9) onto I-481 south (light traffic); (10) onto I-81 north (no trucks in sight); (11) downtown exit (smell of diesel exhaust on Adams St.); (12) in laboratory at university. (b) Morning commute and environmental tobacco smoke.

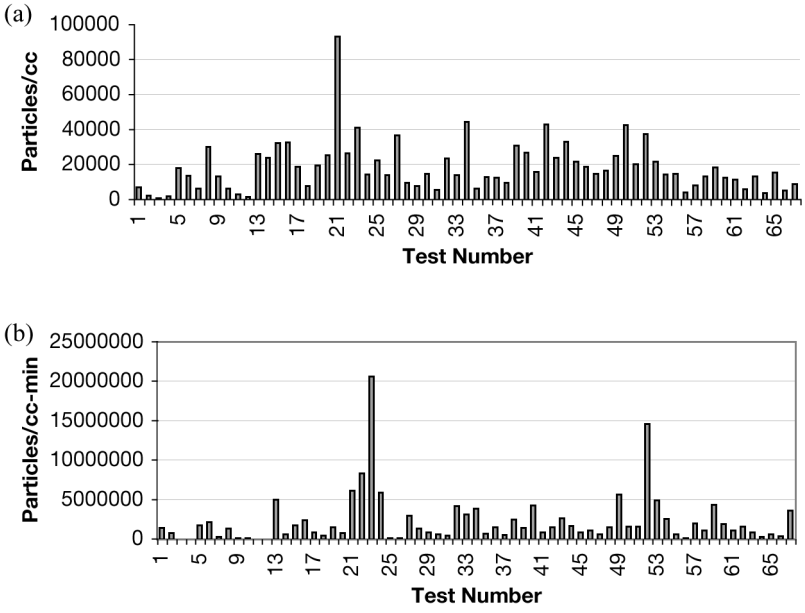

Fig. 6. (a) Average ultrafine particle concentration in 67 tests (>128 h). (b) Cumulative ultrafine particle exposures in 67 tests.

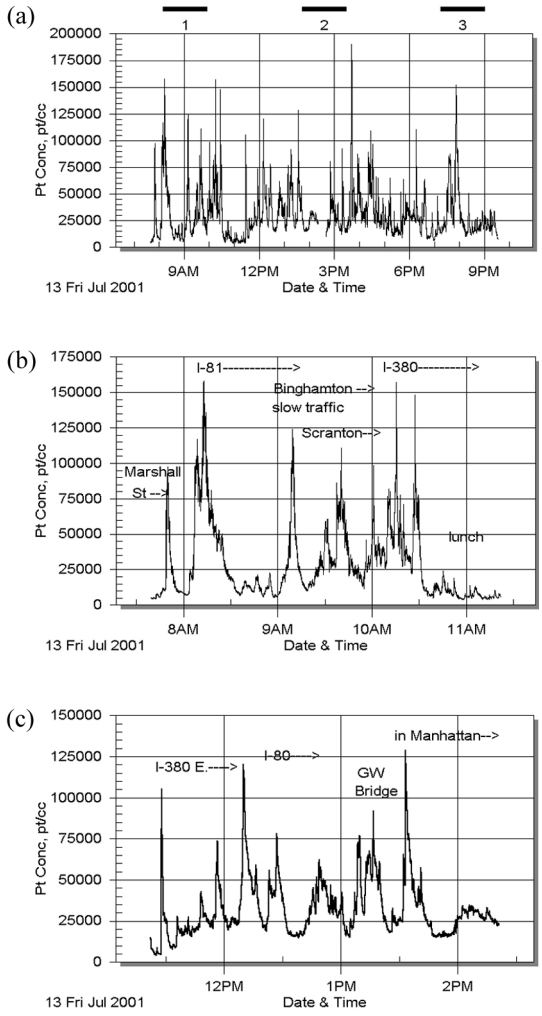

Fig. 7. (a) Long distance UFPM exposure inside car: 13 h+ round trip, Syracuse to New York and back. (b) Syracuse to New York: UFPM exposure inside car. Note the prolonged very high UFPM levels related to variable traffic conditions in the city and on the highway. Longer time averaging does not reveal these. (c) UFPM inside car in the vicinity of New York.

appears likely that the source of the K-rich sulfates is from combustion of K-rich materials such as foods, candles, etc. Whether K-containing UFPMs aggregate with sulfates from outdoor sources or are generated as a single entity remains to be determined.

Dennekamp *et al.* (2001) measured UFPM and NO_x in simulated kitchens, using a TSI 3934 scanning mobility particle sizer and a 3022A condensation particle counter. Their measurements were more detailed than those we report and limited to a single non-mobile controlled laboratory room, but comparisons on reported UFPM number concentrations are worth noting. The time resolution of their measurements was 2.5 min, with averaging over 5 min, whereas ours was 1 s. They compared gas versus electric ring cookers, a grill and an oven in a 70 m^3 room with the windows closed and no mechanical ventilation. Our measurements were in a real home environment with a forced air ventilation system and exhaust ventilation in operation at times. Their table 2 presents the maximum 5 min concentrations above background (their number concentrations are presented for the size range 10–500 nm, our results are not size resolved but are for UFPM >20 nm). As we tested only electric rings, only those results will be compared. They observed peak concentrations (averages, UFPM/cm^3) of: one ring, 94000; four rings, 111000 (no increase when pan heated to boil water); grill only, 77000; toast, 134000. Our results show much higher short-term peaks (>500000 UFPM/cm^3) for similar cooking activities.

Observed decay rates for UFPMs in the closed unventilated laboratory was 30 min for a 50% loss. Coagulation of the particles was the most likely reason. Dennekamp *et al*. have discussed the importance of outside concentrations and of measuring and modeling individual (personal) exposures. Our studies provide a measurement methodology and some data enabling direct measures of personal exposures to UFPMs. In our real world ventilated room (~5 × 5 × 3 m) the time for a 50% loss of UFPM from a point source (lighted match) was ~20–30 min.

Wallace (2000) measured PM in an occupied town house and Abt *et al.* (2000) characterized indoor particle sources: both found toasting (among other activities) contributed to indoor PM. Neither of these studies however measured UFPMs. Dennekamp *et al.* (2001) stated: 'Although no experimental studies of human exposure to indoor UFPs have been undertaken, the concentrations found indoors are as high as those found outdoors during episodes of high air pollution, when people have been shown to demonstrate health effects (Peters *et al.*, 1997)'.

Both our studies and those of Dennekamp *et al.* (2002) present initial data expanding the available database of information on UFPM exposures. This type of sensitive and portable instrumentation should be highly useful in studies of UFPM in mobile and personal micro-environments.

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