*Journal of Exposure Science and Environmental Epidemiology* (2011) 21, 20–30


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# Personal exposure to ultrafine particles

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Personal exposure to ultrafine particles (UFP) can occur while people are cooking, driving, smoking, operating small appliances such as hair dryers, or eating out in restaurants. These exposures can often be higher than outdoor concentrations. For 3 years, portable monitors were employed in homes, cars, and restaurants. More than 300 measurement periods in several homes were documented, along with 25 h of driving two cars, and 22 visits to restaurants. Cooking on gas or electric stoves and electric toaster ovens was a major source of UFP, with peak personal exposures often exceeding 100,000 particles/cm$^3$ and estimated emission rates in the neighborhood of $10^{12}$ particles/min. Other common sources of high UFP exposures were cigarettes, a vented gas clothes dryer, an air popcorn popper, candles, an electric mixer, a toaster, a hair dryer, a curling iron, and a steam iron. Relatively low indoor UFP emissions were noted for a fireplace, several space heaters, and a laser printer. Driving resulted in moderate exposures averaging about 30,000 particles/cm$^3$ in each of two cars driven on 17 trips on major highways on the East and West Coasts. Most of the restaurants visited maintained consistently high levels of 50,000–200,000 particles/cm$^3$ for the entire length of the meal. The indoor/outdoor ratios of size-resolved UFP were much lower than for $PM_{2.5}$ or $PM_{10}$, suggesting that outdoor UFP have difficulty in penetrating a home. This in turn implies that outdoor concentrations of UFP have only a moderate effect on personal exposures if indoor sources are present. A time-weighted scenario suggests that for typical suburban nonsmoker lifestyles, indoor sources provide about 47% and outdoor sources about 36% of total daily UFP exposure and in-vehicle exposures add the remainder (17%). However, the effect of one smoker in the home results in an overwhelming increase in the importance of indoor sources (77% of the total).
*Journal of Exposure Science and Environmental Epidemiology* (2011) **21,** 20–30; doi:10.1038/jes.2009.59; published online 20 January 2010

**Keywords:** *cooking, restaurants, gas stoves, electric stoves, vehicles, tobacco smoke.*

## Introduction

Ultrafine particles (UFP) are increasingly studied because of considerations of their toxicology and possible human health effects (Oberdörster et al., 2005; Bräuner et al., 2007a, b; Stölzel et al., 2007). Outdoor UFP concentrations were measured for 1–2 years in several US cities (U.S. EPA Supersites program; Solomon et al., 2008), and near roadways in California (Zhu et al., 2002) and also in Erfurt and Augsberg, Germany (Wichmann et al., 2000). Residential indoor concentrations have also been reported (Abt et al., 2000; Dennekamp et al., 2001; Long et al., 2001; Wallace and Howard-Reed, 2002; Klepeis et al., 2003; He et al., 2004; Wallace, 2000, 2005, 2006; Wallace et al., 2004, 2008; Hoek et al., 2008). A number of studies of exposures while driving have been done (Westerdahl et al., 2005; Fruin et al., 2008; Zhu et al., 2008). However, exposures in locations such as restaurants or while operating everyday sources of UFP close to one's person (stoves, toasters, ovens, hair dryers, blenders, steam irons) have seldom been previously reported (Afshari et al., 2005). Documenting personal exposures from all major sources is a necessary first step in determining cost-effective health risk reduction measures. Therefore this study has concentrated on personal exposures (using a portable monitor kept close to the person) to UFP from common sources in various locations (homes, cars, restaurants).

## Methods

A portable condensation particle counter (CPC), the Model 3007 (TSI, Shoreview, MN, USA) was employed for the major portion of the study, which took place during 2006–2009. The Model 3007 uses isopropanol to form a supersaturated vapor that condenses around ultrafine particles and causes them to grow to a size detectable by a laser. It can detect particles between about 10 nm and 1 $\mu$m in diameter. The time constant is 9 s to achieve 95% response. The accuracy for concentrations up to 100,000 particles/cm$^3$ is stated by the manufacturer to be within $\pm$20%; above 100,000 particles/cm$^3$ reported values are underestimates due to particle coincidence in the sensing chamber. It is battery-powered; the batteries last for 5–10 h. Each charge of isopropanol also lasts for comparable times. The flow rate is 0.7 l s$^{-1}$, and the data storage capacity is about 50,000 data points.

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Received 21 February 2009; accepted 29 September 2009; published online 20 January 2010

Although the Model 3007 counts all particles up to 1 $\mu$m, evidence from observed size distributions suggests that UFP make up a majority of the particle numbers in most situations. For aged outdoor air, UFP has accounted for 76% (in Beijing, China) to about 90% (in three U.S. cities in the Supersites program) of the total particle number (Wu et al., 2008). For freshly created particles from indoor sources, UFP typically accounted for 87–98% of the total (Wallace and Howard-Reed, 2002). As almost all of our measurements (except those for background levels before beginning experiments) deal with periods during and immediately after a nearby source is creating UFP, we believe the Model 3007 is measuring UFP almost exclusively. The Model 3007 was used to study exposures from a variety of sources in homes; exposures while driving; and exposures while dining in restaurants. In all cases, the sampling time was 1 s.

For one experiment on cigarette smoke, a Scanning Mobility Particle Sizer (SMPS) (TSI, Shoreview MN, USA) was employed. This instrument is capable of determining the number concentrations of about 100 size categories ranging from 10 to 400 nm. The SMPS is considered a reference instrument for determining sizes, and is said by the manufacturer to have an uncertainty of about 8% in estimating number concentrations.

*Exposures in Homes*

Two homes, one in Virginia and one in California, were the main locations for our study. For the home exposures, the instrument was generally placed in one room on the ground floor and operated by line current so that it could run continuously, up to the point where the alcohol needed to be replenished.

The Virginia home is a three-story 385 $m^3$ town house equipped with central air, a gas stove with electronic ignition, and an electric toaster oven. The forced-air ventilation system is kept on at nearly all times to promote mixing. The furnace is gas-fired and an external electric-powered compressor operates a central air-conditioner. Multiple measurements of air change rates were made in the Virginia home over a period of 4 years before this study, and the average air change rate was 0.65 $h^{-1}$ (Wallace and Howard-Reed, 2002). This rate showed strong seasonal variations, including variations because of keeping windows open during much of the summer. For studies of cooking as a source of UFP, the Model 3007 was generally placed on a coffee table in the living room. The living room is on the same floor as the kitchen; the rooms are connected by an open doorway (without a door). For studies of hair dryers and similar devices, the measurements were made in a bathroom with the door open. Ironing was conducted in the basement and the monitor was placed on a coffee table in the same room as the iron. A laser printer was located in an upstairs office, and the monitor was placed in the office for those measurements. All measurements were carried out with all windows closed. Air change measurements were not made; however, based on previous data from this home, air change rates with windows closed varied over a relatively small range from 0.25 to 0.5 $h^{-1}$. A 9-point traverse of the duct system with an anemometer established that with the central fan on, approximately six house volumes (2300 $m^3$) circulated through the system. As the duct system at no point was outside the conditioned volume, operating the fan caused no measurable effect on the outdoor air change rate.

To determine emission rates, the maximum concentration observed was multiplied by an appropriate mixing volume and divided by the time the source was on:

$$S = \frac{C_{\max} V}{t} \quad (1)$$

$S$ is source strength or emission rate (particles/min); $C_{\max}$ is the maximum concentration ($cm^{-3}$), $V$ is mixing volume ($cm^3$), $t$ is time (min).

For cooking, the mixing volume selected was the volume of the first floor (130 $m^3$), since the particles generally did not have time to become well-mixed throughout the house. For the hair dryer tests, the volume was that of the bathroom (12 $m^3$). For the steam iron, the volume was that of the basement den (65 $m^3$). For the laser printer, the volume was that of the upstairs office (30 $m^3$). For the cigarette, the volume in the test with the SMPS was that of the entire Virginia house (385 $m^3$).

The California home is a two-story detached home with an area of 200 $m^2$ and a volume of 460 $m^3$. Previous studies have shown it behaves approximately as a single compartment when its interior doors are open and exterior doors are closed (Howard-Reed et al., 2002). Most measurement experiments were conducted in the kitchen with the monitor placed on a nearby table in the adjacent breakfast nook, approximately 2.8 m from the stove and 4.6 m from the toaster. The area is open between the kitchen and the breakfast nook. For the electric heater and cigarette experiments, the monitor and source were placed in a 44 $m^3$ bedroom in the house with the bedroom door closed. Multiple measurements of air change rates with windows closed averaged 0.3 $h^{-1}$.

Some studies took place in a 340 $m^3$ 1-story uninhabited manufactured house on the campus of the National Institute for Standards and Technology (NIST). This house had a gas stove, an electric stove, and an electric toaster oven. Only a subset of the electric stove experiments using the Model 3007 are reported here. More than 250 experiments employing a nano-SMPS to measure all three sources are reported in another article (Wallace et al., 2008). The Model 3007 monitor was placed in the master bedroom, which was connected to the foyer by an open door; the foyer was part of a larger space including the kitchen, where the stoves and toaster oven were located. The central fan in the heating, ventilating, and air conditioning (HVAC) system was on at all times to promote mixing.

*Indoor–Outdoor Relationships*

Personal exposure indoors includes exposure to particles penetrating from outdoors. To determine the magnitude of this effect, measurements were made indoors and outdoors at the NIST test house. The measurements employed a SMPS (TSI, Shoreham, MN, USA) capable of measuring particles from 2 to 100 nm in diameter. A switching mechanism allowed automatic switching from outdoors to indoors for several days at a time. About 24 days of sampling were conducted with windows closed, and another 21 days with one window open 8 cm (3 inches). Four consecutive 2.5-min samples were collected indoors and then outdoors. This approach allowed for dropping the first one or two samples of each cycle, if an effect because of adjusting to different temperature–humidity conditions was encountered.

*Exposures While Driving*

The driving studies involved two cars. The Virginia car was a Volkswagen Golf gasoline-powered hatchback. The California car was a Lexus RX-300 sports utility vehicle.

For both cars, the Model 3007 was placed on the front passenger seat while driving. For the Virginia car, windows were generally open during periods when vehicles such as school buses were being purposefully tracked, to have a better chance of detecting particular sources. For longer trips, the windows were normally closed with the outdoor intake fan on a low setting. For the California car, the driver's window was fully open, and the passenger window was opened by 8 cm (3 inches). Air exchange rates were measured for this car under different conditions of speed and fan and window settings (Ott et al., 2008).

*Exposures in Restaurants*

For these studies, the Model 3007 was turned on while leaving home and operated while walking to the restaurant from the parking lot to provide a background (outdoor) concentration. In the restaurant, the instrument was placed either on the seat in a booth, a nearby chair, or the table. The volume of the restaurant was calculated by walking the length and width and estimating or measuring ceiling height. Occasionally a DM S50 Sonic Measure (Zircon, Campbell, CA, USA) distance-scaling instrument that measures the time of a reflected ultrasonic signal was used to measure the distances to walls or ceilings. Records were kept of the number of people or occupied tables. All restaurants were nonsmoking.

## Results

*Quality Control*

The Model 3007 sampling rate was checked periodically and found to be within 5% of $0.71\,s^{-1}$. A second instrument was run side-by-side with one of the main instruments for approximately 100 h to determine precision and relative bias. In a typical experiment, the relative bias of the instruments was 1–3% and the mean precision, calculated as the sum of the absolute values of the difference divided by the sum of each measurement pair, was 2.3%. In one typical experiment, for 22,973 1-s measurements, the $R^2$ value was 0.998 (Figure 1). The Model 3007 occasionally shows a drop of 5–15% in a 1-s reading. The frequency of this occurrence is less than 1% of measurements, so it does not affect the average concentration noticeably. After our data collection was completed, TSI reported that the problem had been identified as a momentary leakage of the isopropanol into the sensing area; this can be corrected by elevating the front end of the monitor by 5° (K. Erickson, TSI, personal communication).

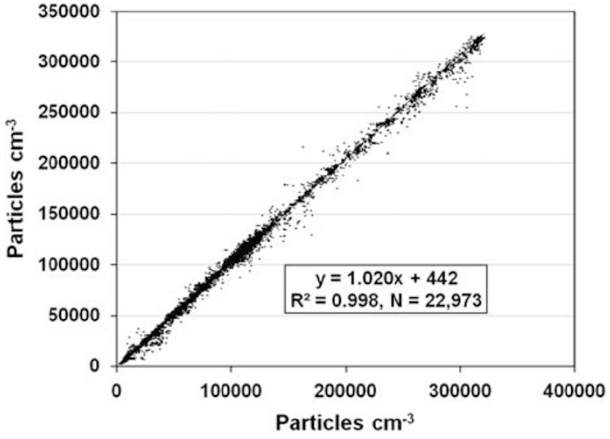


**Figure 1.** Relative bias and precision of collocated Model 3007 monitors. Note the good agreement beyond the nominal 100,000 particles/cm$^3$ limit.

*Exposures in Homes*

In the Virginia home, more than 250 activities relating to about a dozen different sources were recorded. The sources, activities, room volumes for calculating emission rates, and ranges of concentrations are provided in Table 1. The median background (indoor) number concentration was 2,700 particles/cm$^3$ (5th percentile 900 particles/cm$^3$, 95th percentile 9000 particles/cm$^3$). The concentrations in the table have been corrected for the associated background concentrations.

*Cooking* Ninety-five experiments employed a gas stove, 54 an electric toaster oven, and 23 both simultaneously. Deep-frying tortillas on the gas stovetop burner followed by baking them in the oven produced number concentrations in the living room that exceeded 400,000 particles/cm$^3$. Baking muffins at 350°F or potatoes at 450°F, the latter followed by broiling salmon in the gas oven, resulted in number concentrations between 100,000 and 200,000 particles/cm$^3$.

**Table 1.** Tests of cooking and other activities in the Virginia home and NIST research house.

| Appliance | Activity, model, food type | *N* | Monitor site | Volume ($m^3$) | Range of peak concentrations[a] ($\times 10^3$ particles $cm^{-3}$) | Range of 1-h averages[a] ($\times 10^3$ particles $cm^{-3}$) |
|---|---|---|---|---|---|---|
| Air popper | Popcorn | 1 | LR | 130 | 165 | 57 |
| Candles | Candles | 5 | LR | 130 | 117–186 | 53–78 |
| Cleansers | Housecleaning | 4 | LR | 130 | 123–330 | 40–171 |
| Curling iron | Two models | 3 | Bathroom | 12 | 127–324 | 40–214 |
| Electric coil, toaster oven | Bacon, eggs, Eng. muffin | 3 | NIST MBR | 350 | 42–48 | 24–28 |
| Electric toaster | 2-slot model | 1 | LR | 130 | 240 | 104 |
| Electric toaster oven | Toast or empty | 4 | Kitchen | —[b] | 134–396 | 24–146 |
| Electric toaster oven | Toast | 35 | LR | 130 | 15–212 | 8–112 |
| Electric toaster oven | Toast (multiple slices) | 1 | LR | 130 | 250 | 183 |
| Electric toaster oven | English muffin | 2 | NIST MBR | 350 | 38–64 | 28–51 |
| Electric toaster oven | Bagel, toast | 7 | Upstairs | 385 | 14–41 | 8–25 |
| Fireplace | Fire lit | 1 | LR | 130 | 130 | 2 |
| Gas burner | Boiling water | 4 | Kitchen | — | 135–243 | 67–88 |
| Gas burner | Various cooking | 29 | LR | 130 | 24–300 | 7–140 |
| Gas burner | Fried eggs | 1 | NIST MBR | 350 | 60 | 34 |
| Gas burner | Various cooking | 3 | Upstairs | 385 | 35–98 | 20–61 |
| Gas burner and gas oven | Tortillas | 1 | LR | 130 | 439 | 212 |
| Gas burner and toaster oven | Mostly toast and coffee | 26 | LR | 130 | 70–358 | 22–76 |
| Gas burner and toaster oven | Toast and coffee | 2 | Upstairs | 385 | 17–48 | 11–25 |
| Gas burners | Boiling water—2 burners | 2 | Kitchen | — | 346–365 | 123–157 |
| Gas burners | Various cooking | 12 | LR | 130 | 15–306 | 8–161 |
| Gas burners | Grilling tuna | 1 | Upstairs | 385 | 141 | 95 |
| Gas burners and gas oven | Roast beef, boiled potatoes | 1 | LR | 130 | 239 | 111 |
| Gas dryer, vented | Clothes dryer | 1 | Basement | 130 | 18 | 12 |
| Gas oven | Baking potato | 5 | Kitchen | — | 229–410 | 126–230 |
| Gas oven | Baked potato | 4 | LR | 130 | 145–314 | 87–199 |
| Gas oven | Broiled fish | 4 | LR | 130 | 321–432 | 127–386 |
| Gas oven | No food | 10 | LR | 130 | 141–320 | 52–129 |
| Gas oven | Self-cleaning oven | 1 | LR | 130 | 192 | 55 |
| Gas oven | Baked potato | 1 | NIST MBR | 350 | 69 | 45 |
| Gas oven | English muffin | 1 | NIST MBR | 350 | 20 | 11 |
| Gas oven | No food | 1 | Upstairs | 385 | 81 | 54 |
| Hair dryer | Several models | 9 | Bathroom | 12 | 2–312 | 2–158 |
| Laser printer | Printing 10 pages | 3 | Upstairs office | 30 | 6–19 | 4–9 |
| Match | Match | 1 | Kitchen | — | 235 | 11 |
| Match | Match | 1 | LR | 130 | 20 | 7 |
| Space heater | Heat | 2 | Basement | 130 | 8–17 | 7–13 |
| Steam iron | Ironing | 2 | Basement | 130 | 21–148 | 11–86 |

LR, Living Room; MBR, Master Bedroom; NIST, National Institute for Standards and Technology.
[a]Corrected for background.
[b]Open space—no mixing volume applicable.

Sautéing shrimp on the gas stovetop burner in the kitchen resulted in peak concentrations between 200,000 and 300,000 particles/$cm^3$ in the living room, but operating a laser printer in the upstairs office resulted in a small increase of about 1000 particles/$cm^3$ (Figure 2).

Frying bacon at low heat and eggs at high heat on the gas burner in the kitchen resulted in peak number concentrations in the living room that exceeded 200,000 and 250,000 particles/$cm^3$, respectively (Figure 3). Later that day, a new electric toaster oven that had never been used before was tested repeatedly on the 4-min toasting cycle, but without any bread introduced into the oven. The first test produced higher concentrations than the others (Figure 3). This result could be due to a coating on the coils or to an accumulation of dust on the coils.

An electric stove was used for 21 experiments at the NIST test house. Early experiments on the new stove were done without food; later experiments involved boiling different amounts of water and more complex cooking. Peak concentrations ranged from 6000 to 145,000 particles/$cm^3$.

A brand-new electric stove was tested without food at first. Concentrations produced by the oven in both baking and broiling modes were near 200,000 particles/$cm^3$ for the first use, but dropped by 50% for the second baking test and by

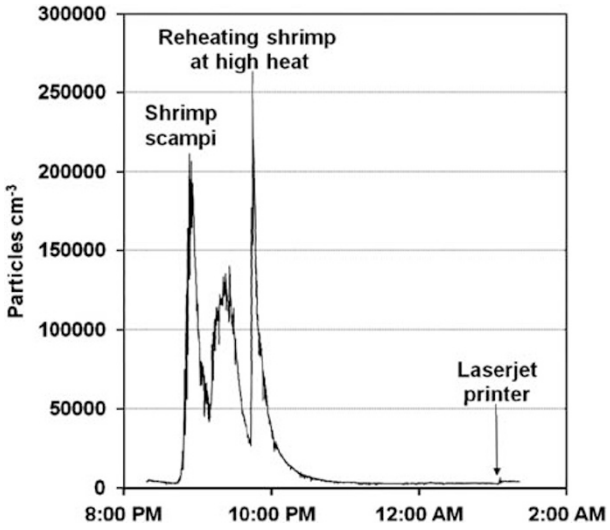


**Figure 2.** Sauteing shrimp on gas stovetop burner; printing on laser printer.

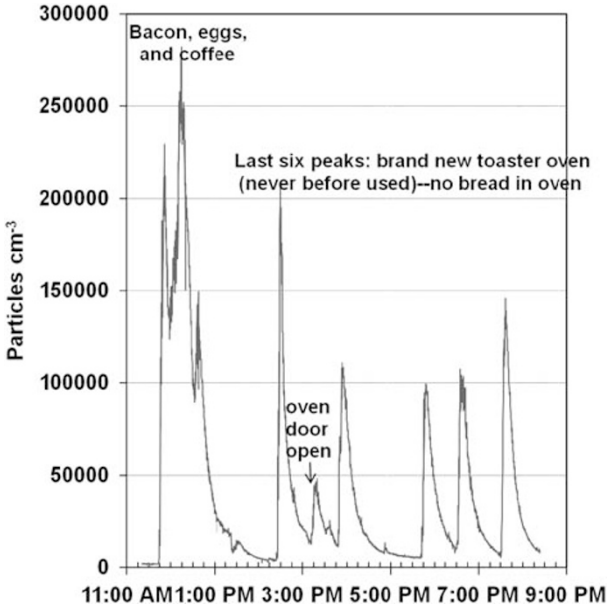


**Figure 3.** Frying bacon (low heat) and eggs (high heat) on stovetop burner; boiling water for coffee; testing new electric toaster oven by 4-min toasting cycle repeated six times.

an additional 25% for the second broiling test a short time later (Figure 4). Earlier that day the effect of turning on an electrostatic precipitator particle filter in the HVAC system was studied. The arrow in Figure 4 marks the time it was turned on and the immediate steepening of the decay curve. A calculation of the effect showed that it increased the natural decay curve from 1.5 to 2.5 $h^{-1}$.

*Electric sources* Ironing with a steam iron in the basement produced levels in the basement that averaged about 100,000

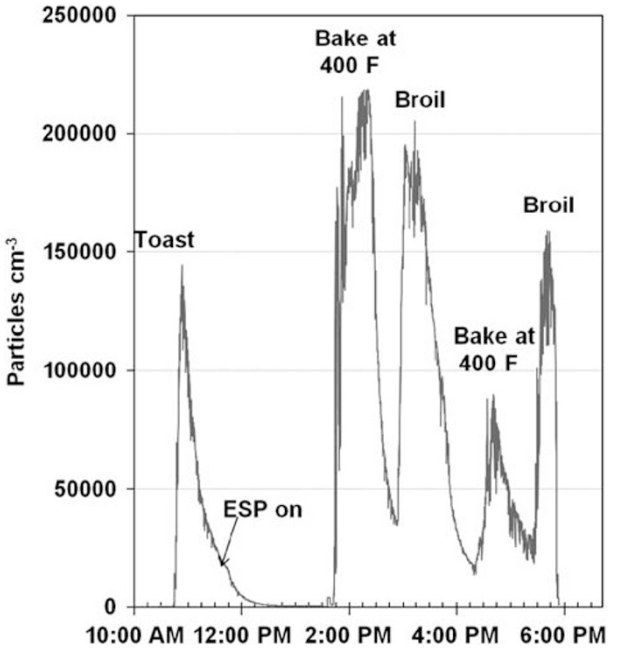


**Figure 4.** Making toast on toaster oven; turning on electrostatic precipitator (ESP) filter in heating, ventilating, and air conditioning system during decay period; testing new electric oven baking and broiling operations (two tests each with no food).

particles/$cm^3$ over 1 h. Popping corn on an air popper in the kitchen produced levels in the living room $>150{,}000$ particles/$cm^3$.

The effects of an electric toaster, an electric space heater, and an electric hair straightener were compared. Both the toaster and hair straightener are capable of increasing personal exposure to peaks ranging between 200,000 and 300,000 particles/$cm^3$, but the electric space heater had almost no effect on UFP numbers.

*Cigarette emissions* Although many studies have calculated particle mass emissions from cigarettes, few have provided number emission rates (Klepeis et al., 2003). In one experiment, a single Marlboro cigarette was smoked in the kitchen of the Virginia town house on 24 December 1997. The central HVAC fan was on to provide mixing. An SMPS located in the basement recorded the number and size of the resulting particles, from 10 to 445 nm. The resulting size distribution peaked at about 100 nm (Figure 5).

The increase in particle number was approximately 5000 per $cm^3$. Assuming no losses because of coagulation, ventilation, and deposition while the particles traveled from the kitchen to the basement, we can multiply this value by the volume of the house (385 $m^3$) to achieve a rough estimate of the emission rate of $2 \times 10^{12}$ particles/cigarette. This is certainly an underestimation of the total particles generated, because it required several minutes for the SMPS in the basement to respond, and as the UFP traveled there partly

through the rooms and stairwells and partly through the ductwork. Both routes, but particularly the ductwork, present surfaces and right angle turns that would increase deposition.

It may also be noted that a substantial portion of the particle output exceeded 100 nm in diameter. The particles greater than 100 nm (100–400 nm) increased from a background of 600–4100 particles/cm$^3$, for a total increase of 3500 particles/cm$^3$.

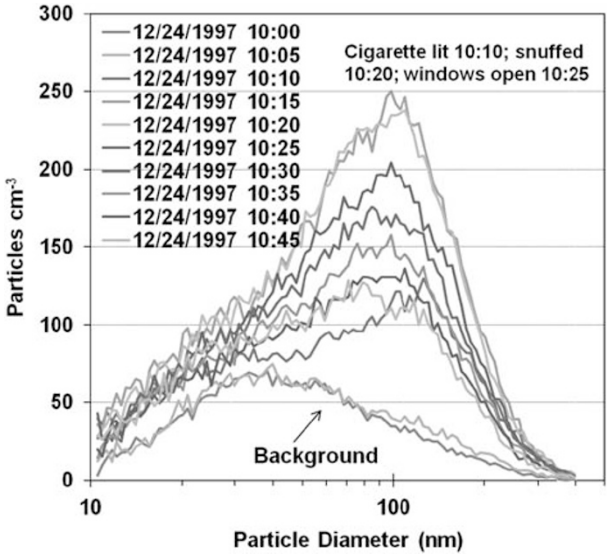


**Figure 5.** Size distribution (10–400 nm) of a cigarette smoked in the Virginia house.

The next 12 experiments took place in the California home with a Model 3007. The first of these took place on 26 July 2005. In this ''whole-house'' experiment, a cigarette was smoked in the kitchen and measured by a Model 3007 in the living room. The resulting increase in particles was 11,000 cm$^3$ over a smoking time of 11 min. Using the same assumptions as above, this corresponds to an emission rate of $5.1 \times 10^{12}$ particles/cigarette. Another ''whole-house'' experiment resulted in an emission rate estimate of approximately $1.0 \times 10^{12}$ particles/cigarette. Nine additional experiments were run with a Marlboro cigarette in a small (44 m$^3$) bedroom using the Model 3007. A typical increase of about 100,000 particles/cm$^3$ over the background was observed, leading to an average emission rate estimate of 4.8 (SD 1.7) $\times 10^{12}$ particles/cigarette.

*Other combustion sources* Two candles burned for 40 min in the living room showed extended periods above 100,000 particles/cm$^3$. Lighting a match close to the monitor produced momentary levels $>200{,}000$ particles/cm$^3$; lighting an identical match in the kitchen could still elevate levels in the living room by about a factor of 3.

*Summary of all sources* Background-corrected concentrations and emission rates with estimated uncertainties for the sources measured in the Virginia home are provided in Table 2 and Figure 6, and for the California home in Table 3.

**Table 2.** Emission rates from indoor sources.

| Appliance | Event | *N* | Range of emission rates ($\times 10^{12}$) (particles/min$^1$) | Mean emission rate ($\times 10^{12}$) (particles/min$^1$) | SD ($\times 10^{12}$) (particles/min$^1$) | SE ($\times 10^{12}$) (particles/min$^1$) | Range of 1-h averages ($\times 10^3$) (particles/cm$^3$) |
|---|---|---|---|---|---|---|---|
| Gas stove and toaster oven | Cooking | 23 | 1.1–11.6 | 5.11 | 3.04 | 0.63 | 6–94 |
| Gas clothes dryer | Drying clothes | 6 | 2.2–5.6 | 4.40 | 1.60 | 0.65 | 16–50 |
| Air popper | Popping corn | 4 | 1.4–6.0 | 4.26 | 2.03 | 1.01 | 19–55 |
| Electric toaster | Toasting | 1 | | 3.8 | | | 99 |
| Match | Lighting candles | 3 | 2.3–5.0 | 3.65 | 1.91 | 1.10 | 4.4–26 |
| Spray cleaner | Housecleaning | 6 | 1.5–3.2 | 2.60 | 0.61 | 0.25 | 33–162 |
| Electric toaster oven | Cooking | 54 | 0.19–6.9 | 2.11 | 1.34 | 0.18 | 7–144 |
| Gas stove | Cooking | 95 | 0.37–6.9 | 1.89 | 1.71 | 0.18 | 8–223 |
| Electric stove | Cooking | 21 | 0.24–4.4 | 1.25 | 1.08 | 0.24 | 6–145 |
| Cigarette | Smoking | 13 | 0.33–1.0 | 0.48 | 0.17 | 0.05 | 4–14 |
| Electric mixer | Preparing food | 5 | 0.02–1.5 | 0.57 | 0.72 | 0.32 | 0.004–61 |
| Candles | Burning candles | 10 | 0.13–1.0 | 0.56 | 0.25 | 0.08 | 9–139 |
| Curling irons | Grooming | 3 | 0.14–0.4 | 0.29 | 0.13 | 0.07 | 0.005–193 |
| Steam iron | Ironing | 6 | 0.04–0.35 | 0.24 | 0.11 | 0.04 | 10–81 |
| Hair dryers | Grooming | 8 | 0.0007–0.7 | 0.23 | 0.22 | 0.08 | −0.01–137 |
| Space heater | Heating | 3 | 0.03–0.29 | 0.13 | 0.13 | 0.08 | 3.6–57 |
| Hair straightener | Grooming | 1 | | 0.11 | | | 16 |
| Laser printer | Printing 10 pages | 3 | 0.03–0.10 | 0.06 | 0.04 | 0.02 | 0.068–4.2 |
| Vacuums | Housecleaning | 2 | 0.02–0.10 | 0.06 | 0.06 | 0.04 | 0.31–3.5 |
| Fireplace | Fire lit | 1 | | 0.003 | | | 0.14 |

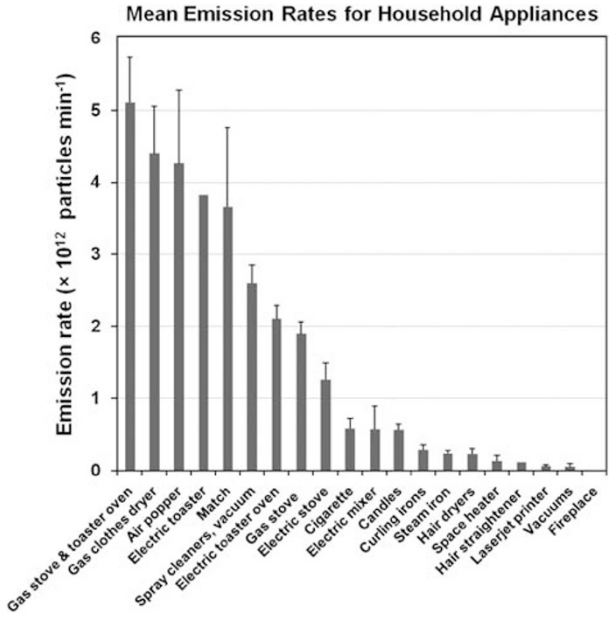


**Figure 6.** Mean emission rates (with SE) of 19 categories of sources.

Emission rates from the gas stove in the California home were generally similar (around $10^{12}$ particles/min) to those in the Virginia home.

*Indoor–Outdoor Relationships*

For the experiments at the NIST test house, comparing the first sample of each 4-sample set indoors and outdoors showed no apparent influence of the change in temperature and relative humidity encountered on changing from indoors to outdoors or vice versa, so all samples were retained for analysis. The ratios with all windows closed or with one window open are shown in Figure 7. For the case with closed windows, only 5% of particles $<10$ nm in diameter are able to remain airborne in the house, and $<20\%$ of all particles $<64$ nm remain airborne in the house. A single open window results in a marked increase in the penetration of these particles, but even these ratios are below those seen for larger particles ($PM_{2.5}$ and $PM_{10}$). Although these measurements cut off at 64 nm, and the penetration will probably continue to increase at larger particle diameters, because the particles $<64$ nm probably account for the majority of particles, it seems reasonable to estimate the total infiltration ratio for ultrafine particles at about 30%.

*Exposures While Driving*

Results for 17 drives in California and the East Coast are provided in Table 4. The travel times of the 17 drives ranged from 25 to 352 min, and the in-traffic mean concentrations ranged from 17,600 to 48,100 particles/cm$^3$. Both background outdoor concentrations (9000–10,000 particles/cm$^3$) and vehicle interior concentrations (29,000–34,000 particles/cm$^3$) were very similar on the East and West Coast highways. These readings are quite similar to those noted in an 8-month study in Montreal: 38,000 particles/cm$^3$ in the evening, 31,000 particles/cm$^3$ in the morning (Weichenthal et al., 2008). They are lower than those reported on Los Angeles freeways by Westerdahl et al. (2005) and Zhu et al. (2008), which probably is because of the exceptionally high volume of heavy duty diesel-powered trucks on the Los Angeles freeways selected for their studies (Zhu, 2008).

*Exposures in Restaurants*

Trips to 22 restaurants are documented in Table 5. Restaurants in which the kitchens were separated from the dining

**Table 3.** Concentrations and emission rates for California home.

| Action | Time of action (min) | Peak concentration ($\times 10^3$) (particles/cm$^3$) | Emission rate ($\times 10^{12}$) (particles/min$^1$) |
|---|---|---|---|
| *Electric appliances* | | | |
| Toasting English muffin in toaster | 4 | 174 | 17 |
| Empty toaster | 4 | 118 | 12 |
| Infrared electric heater | 10 | 21 | 0.8 |
| *Testing gas burners—no food* | | | |
| Gas—no grill | 20 | 7 | 0.1 |
| Gas—no grill | 32 | 16 | 0.2 |
| Gas—with grill | 20 | 17 | 0.3 |
| Gas burner | 12 | 98 | 3.3 |
| Gas burner | 12 | 132 | 4.4 |
| Empty stainless steel frying pan | 4 | 43 | 4.3 |
| Empty pan | 10 | 156 | 6.2 |
| Gas—sauce pan no water | 20 | 51 | 1.0 |
| *Boiling water* | | | |
| *In stainless steel pan* | | | |
| 1 min | 1 | 1 | 0.4 |
| 2 min | 2 | 11 | 2.1 |
| 3 min | 3 | 18 | 2.3 |
| 4 min | 4 | 22 | 2.2 |
| 5 min | 5 | 13 | 1.0 |
| 6 min | 6 | 9 | 0.6 |
| 10 min | 10 | 16 | 0.6 |
| 10 min | 10 | 112 | 4.5 |
| 20 min | 20 | 3 | 0.05 |
| In Pyrex glass beaker | 10 | 60 | 2.4 |
| *Cooking on gas stove* | | | |
| Boiling eggs | 4 | 86 | 8.6 |
| Boiling one egg | 5 | 155 | 12 |
| Scrambled eggs ($N=7$) | 4 | 0–192 | 0–19 |
| Scrambled eggs, toast | 4 | 50 | 5.0 |
| Fried eggs | 5 | 74 | 5.9 |
| Soup | 1.5 | 0 | 0.0 |
| Soup | 10 | 40 | 1.6 |
| Soup | 13 | 102 | 3.1 |

areas, such as the large steak house in San Mateo, had relatively low indoor concentrations ($16 \times 10^3$ particles/cm$^3$). In contrast, restaurants with open flame cooking or grills located in the dining areas near the patrons had relatively high ultrafine particle concentrations, such as the Thai restaurant in San Francisco ($166 \times 10^3$ particles/cm$^3$) and the Indian restaurant in San Carlos ($222 \times 10^3$ particles/cm$^3$; Figure 8). The relatively high ultrafine particle counts measured during three dates at a buffet breakfast at a Sacramento hotel restaurant was attributed to a small 2-grill gas burner used by a chef to cook omelettes for the breakfast guests. A French restaurant in Fairfax, VA, USA, maintained a concentration $>200{,}000$ particles/cm$^3$ for 2 h (Figure 9). Mean concentrations for all 22 restaurants of $94{,}000 \pm 69{,}000$ particles/cm$^3$ were more than an order of magnitude higher than background outdoor levels of $7900 \pm 4000$ particles/cm$^3$.

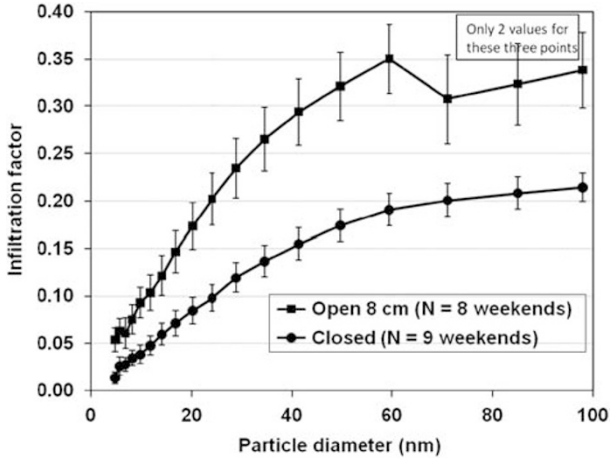


**Figure 7.** Size-resolved indoor/outdoor number concentration ratios (infiltration factors) for two cases: all windows closed or one window open 3 inches (8 cm). Values measured in uninhabited NIST test house on weekends. Error bars are SE.

## Discussion

Although the upper limit for the Model 3007 is said to be 100,000 particles/cm$^3$, correction factors are available for higher concentrations (Hämeri et al., 2002). These correction factors suggest that Model 3007 readings up to 200,000 particles/cm$^3$ are underestimated by about 10%, with increasing underestimates at higher concentrations. We have chosen not to make these corrections, as the number of values $>200{,}000$ particles/cm$^3$ was quite small. It should be noted, however, that when we report values $>100{,}000$ particles/cm$^3$, we believe these are dependable underestimates in that the actual values are at least this high.

For many of these sources, we have only a few measurements, and the measurements themselves are uncertain by about 20%. For other sources, however, such as home cooking and penetration of outdoor particles into homes we are building on a base of literally thousands of measurements in particular homes over periods exceeding 1 year.

Exposures for the cook (monitor in kitchen) were usually several times higher than exposures in the remainder of the house, reaching levels up to 400,000 particles/cm$^3$. Exposures were also very high (up to 300,000 particles/cm$^3$) for use of hair dryers and curling irons. Exposures were high (in the 100,000–200,000 particles/cm$^3$ range) for the use of the air corn popper, candles, and the steam iron. Exposures were relatively low ($<20{,}000$ particles/cm$^3$) for the use of

**Table 4.** Particle counts measured by Model 3007 in vehicles (thousands of particles/cm$^3$).

| Trip | Date | Highway travel route | Time (min) | On-road mean | Background mean | On-road SD | Background SD |
|---|---|---|---|---|---|---|---|
| 1 | 5/24/2007 | Reston, VA, to Gaithersburg, MD | 50 | 33 | 12 | 20 | 0.2 |
| 2 | 5/24/2007 | Gaithersburg, MD, to Reston, VA | 50 | 48 | 9 | 26 | 0.7 |
| 3 | 5/31/2007 | Reston, VA, to Gaithersburg, MD | 35 | 23 | 8 | 16 | 0.4 |
| 4 | 5/31/2007 | Gaithersburg, MD, to Reston, VA | 47 | 43 | 14 | 24 | 1.7 |
| 5 | 8/10/2007 | Reston, VA, to New York City, NY | 316 | 20 | 8 | 9 | 0.4 |
| 6 | 8/14/2007 | Boston, MA, to New Hampshire | 51 | 18 | 3 | 12 | 2.8 |
| 7 | 8/20/2007 | Berkshires, MA to Reston, VA | 352 | 34 | — | 21 | — |
| 8 | 8/23/2007 | Reston, VA, to Gaithersburg, MD | 25 | 46 | 12 | 19 | 0.5 |
| 9 | 8/27/2007 | Reston, VA, to Washington, DC | 68 | 23 | 8 | 15 | 1.6 |
| 10 | 9/19/2007 | Reston, VA, to Gaithersburg, MD | 50 | 38 | 5 | 17 | 1.2 |
| 11 | 10/1/2007 | Reston, VA, to Gaithersburg, MD | 37 | 42 | 8 | 23 | 1.0 |
| 12 | 10/1/2007 | Gaithersburg, MD, to Reston, VA | 27 | 39 | — | 24 | |
| 13 | 7/5/2006 | Menlo Park-Los Altos & Return, CA | 35 | 44 | 9 | 53 | 1.4 |
| 14 | 8/9/2006 | Redwood City to San Francisco, CA | 39 | 18 | 11 | 9 | 0.3 |
| 15 | 8/9/2006 | San Francisco to Redwood City, CA | 41 | 30 | 7 | 26 | — |
| 16 | 5/25/2007 | Redwood City to Sacramento, CA | 153 | 22 | 8 | 13 | — |
| 17 | 5/28/2007 | Sacramento to Redwood City, CA | 183 | 30 | 10 | 17 | — |
| East Coast (VA, MD, NY, NH, MA, DC) Roads | | | 1108 | 33.9 | 8.8 | 18.7 | 1.0 |
| West Coast (California) Roads | | | 451 | 29.0 | 8.9 | 23.3 | 0.8 |

**Table 5.** Particle counts measured by Model 3007 in 22 restaurants while dining.

| No. | Description, town[a] | Patron count | Duration (min) | Volume ($m^3$) | Mean ($\times 10^3\,cm^{-3}$) | SD ($\times 10^3\,cm^{-3}$) | Outdoor background ($\times 10^3\,cm^{-3}$) |
|---|---|---|---|---|---|---|---|
| 1 | Mexican Restaurant, MP | 16 | 27 | 174 | 28 | 16 | 7.2 |
| 2 | Sports Tavern, RC | 14 | 37 | 639 | 126 | 46 | 10 |
| 3 | Family Mexican Restaurant, RC | 3 | 30 | 211 | 10 | 3.2 | 5.4 |
| 4 | American Restaurant, PV | 75 | 75 | 611 | 22 | 11 | 2.1 |
| 5 | Sports Tavern, MP | 15 | 29 | 548 | 88 | 13 | 2.2 |
| 6 | Thai Restaurant, SF | 6 | 67 | 302 | 166 | 43 | 12 |
| 7 | Italian Restaurant, SM | 25 | 88 | 385 | 25 | 6.3 | 10 |
| 8 | Hotel Restaurant 1, Sac | 77 | 69 | 810 | 98 | 23 | 10 |
| 9 | Hotel Restaurant 2, Sac | 68 | 35 | 810 | 97 | 33 | 8.5 |
| 10 | Hotel Restaurant 3, Sac | 23 | 44 | 810 | 58 | 18 | 14 |
| 11 | Seafood Dinner Restaurant, SM | 53 | 103 | 652 | 110 | 44 | 4.3 |
| 12 | Sandwich-Pizza-Bakery, WS | 6 | 77 | — | 61 | 35 | 9.5 |
| 13 | Elegant Indian Restaurant, BG | 58 | 69 | 790 | 192 | 39 | 9.3 |
| 14 | Seaside Beach Restaurant, HMB | 112 | 90 | 1422 | 77 | 30 | 7.3 |
| 15 | Family Tavern, SF | 34 | 128 | 448 | 109 | 51 | 18 |
| 16 | Italian Restaurant 2, SM | 41 | 76 | 284 | 37 | 6.8 | 3.7 |
| 17 | Steak House, HD | 124 | 73 | 610 | 16 | 6.3 | 3.6 |
| 18 | Sicilian Restaurant, RC | 24 | 60 | 490 | 69 | 4.0 | 7.6 |
| 19 | Indian Restaurant, SC | 16 | 75 | 360 | 222 | 27 | 3.9 |
| 20 | Seafood Grill, RS | 80 | 104 | 960 | 37 | 7.2 | 6 |
| 21 | Chinese restaurant, GA | 60 | 75 | 1200 | 203 | 43 | 12 |
| 22 | French restaurant, FA | 30 | 60 | 132 | 228 | 11 | 6.3 |
| Mean | | 43.6 | 67.8 | 602.3 | 94.5 | 23.5 | 7.9 |
| SD | | 34.4 | 26.5 | 333.2 | 68.9 | 16.0 | 4.0 |

[a]Towns: MP = Menlo Park, CA; PV = Portola Valley, CA; WS = Woodside, CA; RC = Redwood City, CA; SF = San Francisco, CA; SM = San Mateo, CA; BG = Burlingame, CA; HD = Hillsdale, CA; HMB = Half Moon Bay, CA; Sac = Sacramento, CA; SC = San Carlos, CA; RS = Reston, VA, GA = Gaithersburg, MD , FA = Fairfax City, VA.

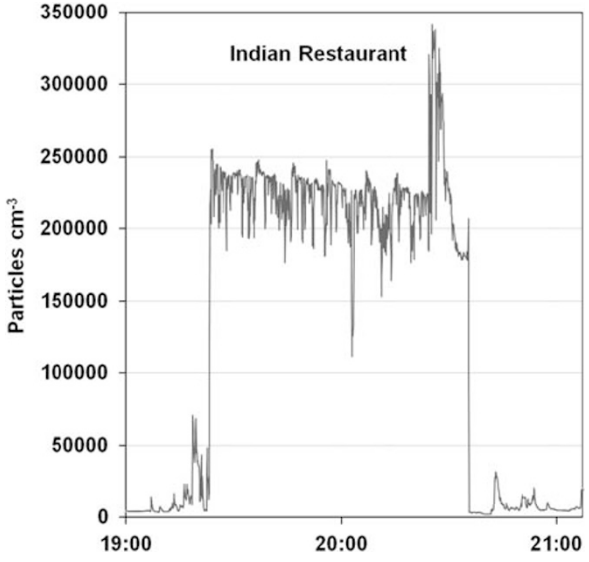


**Figure 8.** Indian restaurant in San Carlos, CA.

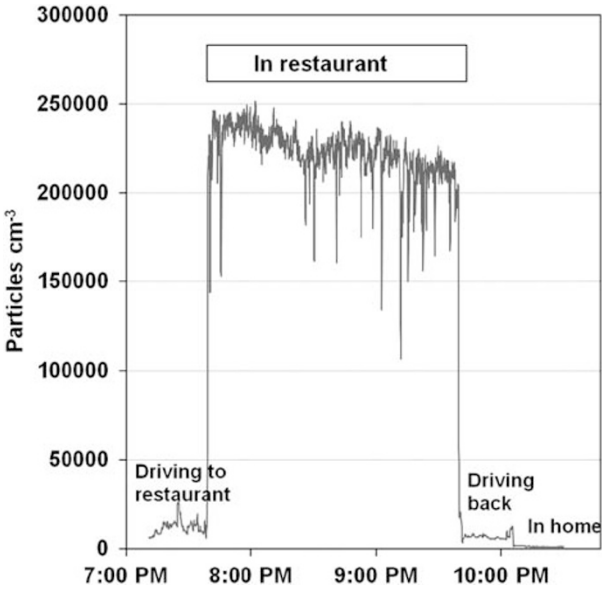


**Figure 9.** French restaurant in Fairfax, VA.

the laser printer, fireplace, and space heater. Exposures in restaurants were relatively high (94,000 particles/$cm^3$) whereas exposures in vehicles were lower (29,000–33,000 particles/$cm^3$). Exposures in these suburban homes without sources were quite low (3000 particles/$cm^3$).

The simple Eq. (1) used to determine emission rates ignores losses because of ventilation, deposition, and coagulation. However, most cooking episodes (e.g., boiling

water for tea or coffee, making toast) were completed in 4 min, allowing little time for losses because of ventilation and deposition. Assuming a rather high air change rate of $0.5\,h^{-1}$ and a high deposition rate of $1\,h^{-1}$, the losses due to these processes over 4 min would be about 10%. On the other hand, more extensive losses due to coagulation may have occurred in cases with high concentrations (>100,000 particles/$cm^3$). Therefore our results may be underestimates of actual emission rates, although as they incorporate coagulation losses they may result in better estimates of exposure. Our finding of cooking emission rates in the neighborhood of $10^{12}$ particles/min may be compared with the finding in Wallace et al. (2008) of emission rates around $10^{13}$ particles/min. These values are not in disagreement, as this study uses the Model 3007 with a lower cutoff at 10 nm, whereas the Wallace et al. (2008) study employed a nano-DMA with a lower cutoff at 2 nm; about 10 times as many particles were noted in the 2–10 nm region as in the region >10 nm, which accounts for the order of magnitude difference in the estimated emission rate.

The exposures documented here are in general considerably higher than those measured outdoors. For example, exposures while driving in both the East Coast and California were about 30,000 particles/$cm^3$, compared with an average outdoor background of about 8000 particles/$cm^3$. Restaurants contributed 1-h averages of more than 90,000 particles/$cm^3$ to exposure, compared with an outdoor background of less than 8000 particles/$cm^3$. And cooking with either a gas or electric stove or toaster oven, using a gas clothes dryer, and use of several small kitchen and personal grooming appliances could also contribute 1-h exposures on the order of several times outdoor concentrations.

A consideration for personal exposure is the effect of outdoor particles penetrating into the home. Because of the high Brownian motion of UFP, they are less likely to penetrate the building envelope, and also deposit more rapidly on surfaces. Whereas typical infiltration factors for $PM_{10}$ and $PM_{2.5}$ are in the neighborhood of 50% (Ott et al., 2000), the infiltration factor for total UFP based on measurements over 18 months in the Reston house was about 30% (Wallace and Howard-Reed, 2002). This means that the effect of outdoor UFP on personal exposures is substantially lower than for $PM_{2.5}$ or $PM_{10}$. This in turn means that indoor sources will provide a higher fraction of total exposure to UFP than is provided by the larger particles.

We can estimate the relative importance of outdoor, indoor, and in-vehicle sources to the exposures of persons not exposed to cigarette smoke. Outdoor concentrations in the two suburban locations ranged between 8000 and 10,000 particles/$cm^3$. People spend about an hour outdoors daily, so their integrated daily exposure (concentration × time) would be about 9000 particles-h/$cm^3$. Indoor concentrations in a suburban town house when no sources were present averaged 2373 particles/$cm^3$ over an 18-month period (Wallace and Howard-Reed, 2002); presumably this is because of penetration of outdoor particles. People spend about 22 h/day indoors, so their baseline exposure because of outdoor particle penetration would be about $2400 \times 22 = 53{,}000$ particle-h/$cm^3$. People spend about 1 h in cars exposed to about 30,000 particles/$cm^3$ so their daily exposure due to auto travel would be about 30,000 particle-h/$cm^3$. Americans spend about 0.2 h/day in restaurants (ATUS, 2009) for an exposure of $90{,}000 \times 0.2 = 18{,}000$ particle-h/$cm^3$. Cooking was associated with a mean concentration of 26,000 particles/$cm^3$ for an average time period of 2.4 h/day (Wallace and Howard-Reed, 2002), corresponding to an integrated daily exposure from cooking of about 62,000 particle-h $cm^3$.

Thus the two largest sources in this scenario are outdoors (penetration into homes and being outdoors) at $53{,}000 + 9000 = 62{,}000$ particle-h/$cm^3$ daily and cooking or eating out at $62{,}000 + 18{,}000 = 80{,}000$ particle-h/$cm^3$ daily. Driving accounted for an additional 30,000 particles-h/$cm^3$. The grand total is 172,000 particle-h/$cm^3$ daily. Dividing this result by 24 h gives an average daily UFP exposure of 7200 particles/$cm^3$, of which about 47% is due to indoor sources, 36% to outdoor sources, and 17% to in-vehicle exposure. We again emphasize that these estimates are based on our measurements in suburban locations. In rural areas, the influence of outdoor UFP would be smaller, and in urban areas, larger.

A similar scenario was developed by Fruin et al. (2008) for Los Angeles residents. Their point estimate for the outdoor contribution to exposure (46%) was somewhat higher than ours (36%), as expected for an urban *vs* a suburban environment. However, because their scenario included 30 m per day on very high traffic density freeways, they find a much higher contribution of in-vehicle exposure (36% compared with our value of 17%).

Using our figure of $4.8 \times 10^{12}$ particles per cigarette and assuming 16 cigarettes are smoked per day in an average-sized house of 400 $m^3$ volume with a typical air exchange rate of $0.75\,h^{-1}$, we find a typical particle exposure because of secondhand smoke of 10,600 particles/$cm^3$, more than doubling the typical exposure because of all other sources. The percentage of particle exposures due to indoor, outdoor, and in-vehicle sources in homes with one smoker would then be 77, 17, and 6%, respectively.

## Conclusions

Our measurements of personal exposure to UFP using a hand-held monitor have identified a number of important indoor sources, ranging from cooking on stoves (both gas and electric) and toaster ovens to hair dryers. These indoor sources are of comparable magnitude (in terms of 24-h personal exposure) to outdoor sources. At least two different physical processes appear to be responsible for the sources of personal exposure to ultrafine particles in everyday life:

combustion (gas stoves, diesel and gasoline engines, candles, cigarettes); and heating elements (hair dryers, electric irons, toasters, space heaters). Because of the large number of these sources and their diversity, future research on exposure and health effects should follow a broad approach. Dealing only with emissions caused by traffic, for example, would leave a multitude of other important sources unaddressed. Thus, future research should include studies on the exposures and effects caused by these many sources of UFP. One of the most important areas that has received little attention is the diverse activities associated with cooking, both for persons at home and for employees and patrons at restaurants.

## Conflict of interest

The authors declare no conflict of interest.

## Acknowledgements

One of us (LW) acknowledges Andrew Persily and members of his group at the National Institute of Standards and Technology for supplying the Model 3007 and the test house on the NIST campus where the indoor–outdoor measurements were made. WO gratefully acknowledges the assistance of Jane McAteer in monitoring several California restaurants, and the Flight Attendants' Medical Research Institute for support. We also acknowledge TSI for loaning a second Model 3007 to determine the precision of the instrument.

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