Ultrafine Particles: 3 Years of Measurements in the NIST Test House

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SUMMARY
NIST has supported research characterizing ultrafine particle sources and dynamics for more than a decade. Over 90 % of ultrafine particles (UFP) produced by stovetop cooking on both gas and electric stoves were <10 nm in diameter. Emission rates of up to 10^{14} min^{-1} were noted. Coagulation was a dominant removal mechanism and was modeled with some success. Kitchen exhaust fan efficiencies varied from <10 % for particles <5 nm to nearly 100 % for particles >10 nm. Using the NIST test house, penetration coefficients and deposition rates were estimated under real-world conditions for particles in closed-window and open-window configurations. Infiltration factors using dedicated automated air change rate measurements varied from 0.02 for the smallest (<5 nm) particles to >0.5 for larger (30 nm to 100 nm) sizes.

IMPLICATIONS
Ultrafine particles (UFP) have been linked to adverse human health effects such as oxidative damage to DNA and mortality. Characterizing concentrations and identifying important sources and removal mechanisms can lead to more effective mitigation of UFPs.

KEYWORDS
Indoor air, particles, cooking, deposition, air change rates, filters, exhaust fans

INTRODUCTION
UFPs have been associated with morbidity and mortality (Stölzel, 2008). However, they are not monitored regularly or regulated, and their major indoor sources and resulting size distributions have been examined in few studies. Indoor dynamics such as coagulation and deposition are seldom considered with sufficient rigor. Indoor-outdoor relations such as infiltration factors and penetration coefficients are also seldom studied. Also, equipment capable of measuring the entire range from 2-100 nm has only recently been made available. Finally, research on mitigation techniques such as kitchen exhaust fans and air filters is desirable.

Recognizing these research needs, NIST has used a test house (340 m^3) on the Gaithersburg, MD campus to characterize sources, indoor concentrations and processes, outdoor particle infiltration, and mitigation techniques under real-world conditions. Size-resolved (2 nm to100 nm) ultrafine particles (UFP) from indoor sources such as gas and electric stoves and power tools were measured to determine emission rates and coagulation rates. Indoor-outdoor relationships were also studied to determine penetration, deposition, and infiltration rates. The ability of exhaust fans to reduce UFP exposure from cooking was also characterized

METHODS

A Scanning Mobility Particle Sizer (SMPS) employing a nano-differential mobility analyzer (nano-DMA) and a water-based condensation particle counter (CPC) was employed for all UFP measurements. Two sheath flow rates (15 L/min and 6 L/min) were employed to measure two UFP size ranges (2 nm to 64 nm and 3 nm to100 nm). Corresponding aerosol flow rates were 1.5 L/min and 0.6 L/min. A strong radioactive source (Kr-85) was employed to adequately charge the smallest particles. A dedicated automated tracer gas system measured sulphur hexafluoride (SF_6) every minute in 10 indoor locations. The SF_6 was injected every 4 h.

Cooking studies

Two stoves were employed—one with natural gas, the other with electric heating elements. An electric toaster oven was also used. Cooking experiments included testing the gas burner flame or the electric stovetop coil with no pots or food to determine the impact of the fuel itself on UFP generation. Other cooking tests included boiling water on the stovetops, frying with cooking oil, and baking or broiling using the stove ovens. The toaster oven was also used for toasting and baking. Approximately 150 experiments were carried out over a 3-year period from 2007-2009.

Power tools

Power tools were tested using two 2-min operations (separated by 1 min) with the tool turned on but no wood or other materials being drilled or sawed. A few other products with electric motors (vacuum cleaners, shavers, etc.) were also tested.

Kitchen exhaust fans

The effect of kitchen exhaust fans on UFP exposure was studied using an inexpensive fan with low capacity (25 L/s to 50 L/s) and a higher quality fan with settings ranging from 100 L/s to 200 L/s. The gas stove was employed and operated for 15 min with the exhaust fan either on or off. The concentrations were measured over the next hour and the efficiency calculated by comparing the results with the fan off and on.

In all three types of experiments above, the SMPS was located either in the room with the source or in a distant bedroom to determine the range of exposures from persons operating the source to those in other parts of the house.

Indoor-outdoor relations

The SMPS was attached to a 20-cm probe extending outdoors from the MBR. A "Y"-tube equipped with a solenoid valve switched the sampling from inside to outside every 10 min. The indoor tube was equipped with a tube of equal length to the outdoor probe to equalize the losses from deposition in the tubes. The 10-min sampling time in each microenvironment allowed for four consecutive samples in each. If the first of the four samples showed differences with the following three, it was considered to be affected by the change in environmental conditions and the sample was discarded. Most of the time, it was not necessary to discard it. Sampling was conducted only on weekends to prevent any indoor sources from affecting the results. Three conditions were employed: closed windows, one window open 7.5 cm, two windows open 15 cm each.

Data analysis

Emission rates from indoor sources were estimated for the cooking experiments. A dynamic model included coagulation terms with the standard Fuchs corrections, van der Waals/viscosity corrections, and fractal corrections together with estimated deposition rates and measured air change rates to estimate size distribution evolution during decay following cessation of the source. Best least-square fits to the observed size distributions were sought by adjusting parameters including Hamaker constants. Details of the model may be found in Wallace et al., (2008).

For the kitchen exhaust fan experiments, size-resolved particle concentrations were integrated over one hour for both conditions (fan off, fan on) and the efficiency calculated as 1-ratio (fan on/fan off).

For the indoor-outdoor experiments, a recursive model employed the following difference equation based on the mass-balance model with no indoor sources:

$$C_{in}(\mathrm{t}+\Delta \mathrm{t}) = PaC_{out}(\mathrm{t})\Delta \mathrm{t} + C_{in}(\mathrm{t})(1-(a+k)\Delta \mathrm{t}))$$

where C_{in} = indoor number concentration; C_{out} = outdoor number concentration; P = penetration coefficient; a = air change rate; and k = deposition rate. Since the air change rate a was measured continuously, the only unknown parameters are P and k. These were varied iteratively by minimizing the sum of the absolute differences between predicted and observed values of C_{in}. Since this approach requires a complete data series for C_{out}, the missing outdoor values due to indoor sampling were interpolated linearly. Missing indoor values were not filled in. The average infiltration factor was then calculated from the equation $F_{inf} = P\langle a\rangle/(\langle a\rangle+\mathrm{k})$, where $\langle a\rangle$ is the weekend-averaged air change rate. The result was compared to the ratio $\langle C_{in}\rangle/\langle C_{out}\rangle$ as a check on the estimates of P and k.

Quality assurance

The SMPS was calibrated by the manufacturer immediately before experiments began. The instrument is considered by NIST to be a reference instrument for particle diameters. Due to the absence of any national or international standard for particle number concentrations, it is not possible to estimate the uncertainty in the number concentrations. The manufacturer suggests a value of 12 %. Flow rates were measured in triplicate and required to agree within 2 % before beginning each experiment. The tracer gas system was calibrated over the range of 20 ppb to 200 ppb SF_6 approximately every two weeks. Uncertainties are estimated to be 15 % to 20 %.

RESULTS

Cooking experiments

About 150 cooking tests with and without food were completed. Number concentration ranges, geometric mean diameters, and emission rates for the size range from 2 nm to 64 nm are provided (Table 1). Typically 90 % of the particles emitted by the stovetop cooking were <10 nm, whereas for the gas and electric ovens and the electric toaster oven more than half of the particles were usually >10 nm. Normal indoor number concentrations were near 1000 cm^{-3}, so the peak concentrations of 10000 cm^{-3} to 10^6 cm^{-3} were 1-3 orders of magnitude greater.

Table 1. UFP number concentrations and emission rates from cooking on gas and electric stoves and an electric toaster oven.

Mode	Number of tests	Geometric mean diameter (nm)[a]	Peak concentration (2 nm to 64 nm) ($\times 10^3$ cm^{-3})[b]	Emission rate ($\times 10^{12}$ min^{-1})[b]
Gas stove: no food cooked		Range of values		
Burner (SMPS in Kitchen)	9	4.4-7.0	290-2200	N/A
Burner (SMPS in MBR)	19	4.0-7.0	90-740	4.6-13
Oven (Bake/Broil)	14	4.3-24	48-450	0.3-5.1
Gas stove: food cooked or water boiled				
Burner (SMPS in Kitchen)	1	8.7	1000	N/A
Burner (SMPS in MBR)	11	5.5-20	24-190	0.4-7.0
Oven (Bake/Broil)	5	4.6-18	22-140	0.4-1.1
Electric stove				
Stovetop Coil: no food cooked	31	3.2-22	7.1-350	0.6-11
Stovetop Coil: food cooked	31	6.1-31	9.0-310	0.14-14
Oven (Bake/Broil): no food cooked	12	5.2-30	3.3-47	0.06-0.8
Electric Toaster Oven				
Oven in MBR: no food	5	28-32	740-1500	3.1-6.4
Oven in Kitchen: no food	7	16-18	230-340	4.1-6.0
Oven in MBR: food	6	22-49	31-210	1.8-3.7

[a] Uncertainties estimated to be <5 %

[b] Uncertainties estimated to be 12 %

Power tools

The highest concentration observed was with the circular saw, which exceeded 800,000 cm^{-3} in the test in the master bedroom with the door closed. Although only operated for 4 minutes, the concentration was still elevated by about a factor of 8 (8000 cm^{-3} vs 1000 cm^{-3}) an hour later. The initial mode for most of the power tools was less than 10 nm, evolving over time to somewhat larger values due mostly to coagulation and partly to faster deposition on surfaces by the smallest UFP (Table 2).

Table 2. Summary of Power Tools Tested and Peak Number Concentrations[a]

Power Tool	Time on (min)	Location	N ≤10 nm (10^3 cm^{-3})	N>10 nm (10^3 cm^{-3})	Mode[b] (nm)
circular saw	4	Same room	701	553	8.3
		Different room	59	62	8.8
jig saw	4	Same room	430	200	7.0
		Different room	16	12	7.8
belt sander	4	Same room	235	87	6.0
		Different room	13	7	6.6
reciprocating saw	4	Same room	88	50	7.1
		Different room	3	2	6.4
drill	10	Same room	56	21	7.2
		Different room	7	2	6.9
vacuum cleaner	20	Same room, door closed	28	22	8.4
		Same room, door open	9	8	7.6
hedge clippers	4	Same room	0	0	N/A
compressor	5	Same room	0	0	N/A
pump	20	Same room	0	0	N/A
shaver	10	Same room	0	0	N/A

[a] Number concentration errors estimated by manufacturer to be about 12 %.
[b] Uncertainties estimated to be <5 %

Kitchen exhaust fan

Efficiency was low for particles < 5 nm, indicating the difficulty in entraining these particles into the plume. Efficiency was also reduced if the front burner was used, compared to the back burner. The 100 L/s fan efficiency was always larger than that for the 50 L/s fan and reached values > 90 % for the larger particles (10 nm to 14 nm). For further details, see the companion article by Rim et al (2011) in this volume.

Indoor-outdoor studies

With all windows closed, the penetration coefficient *P* ranged from 0.2 for the smallest (8 nm to 10 nm) particles) to about 0.55 for the 30 nm to100 nm particles. With one window open 7.5 cm, *P* ranged from 0.6-0.7 for the smallest (5 nm to 10 nm) particles to about 0.75 for the larger particles. With both windows open to 15 cm each, the penetration coefficient was close to unity for all particle sizes from 10 nm to100 nm. For all three window settings, the deposition rate *k* decreased monotonically with increasing particle diameter from values >1 h^{-1} for the 5 nm particles to about 0.3 h^{-1} for the 100 nm particles. Uncertainty estimates for *P* and *k* ranged from 1 % to 4 %. The infiltration factor increased monotonically with both particle diameter and air change rate, from a range of 0.02 to 0.09 for the smallest (5 nm) particles to a range for the largest (100 nm) particles between 0.15 to 0.27 for the windows closed case; 0.3 to 0.46 for the one window open case; and 0.15 to 0.82 for the case with two windows open. F_{inf} uncertainty estimates propagated from uncertainties in P, k, and a ranged from 20% to 30%.

DISCUSSION

Total number concentrations due to cooking were as much as 10 times greater than reported in previous studies that were limited to particle sizes above 10 nm. Emission rates ranged up to 10^{14} min^{-1}. Because of the high concentrations of up to 10^6 cm^{-3}, coagulation was the dominant process affecting the evolution of the size distribution after the source was turned off. Models were incapable of matching this evolution without corrections for van der Waals and viscosity forces. The low infiltration factors (2 % to 30 %) suggest that for homes of comparable air tightness to the NIST test house, outdoor sources of these ultrafine particles will not contribute substantially to human exposure if indoor sources are present and windows are closed. Deposition rates of 0.3 h^{-1} to >1 h^{-1} are greater than model predictions by about an order of magnitude. However, these rates include deposition in ductwork due to having the central fan on at all times, and are therefore expected to be higher than natural deposition rates.

The combination of low infiltration factors and powerful indoor sources suggests that UFP exposures will be more highly influenced by indoor sources than will $PM_{2.5}$ exposures. Since in many cases, the major indoor source is cooking, this suggests that a focus on reducing exposures while cooking will often be the most efficient mitigation strategy. Kitchen exhaust fans have the capability of reducing exposures of the larger UFP (10 nm to100 nm) by up to 90 % during and shortly after cooking. Exhaust fan efficiencies are lower for the 2 nm to10 nm sizes, for which reduction of exposure may require air filters.

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