# Ultrafine particles from electric appliances and cooking pans: experiments suggesting desorption/nucleation of sorbed organics as the primary source

**Abstract** Ultrafine particles are observed when metal surfaces, such as heating elements in electric appliances, or even empty cooking pans, are heated. The source of the particles has not been identified. We present evidence that particles >10 nm are not emitted directly from the heating elements or the metal surfaces. Using repeated heating of an electric burner, several types of cooking pans, and a steam iron, the increase in the number of particles (>10 nm) can be reduced to 0. After the devices are exposed to indoor air for several hours or days, subsequent heating results in renewed particle production, suggesting that organic matter has sorbed on their surfaces. Also, after a pan has been heated to the point that no increase in particles is observed, washing with detergent results in copious production of particles the next time the pan is heated. These observations suggest that detergent residue and organics sorbed from indoor air are the sources of the particles. We hypothesize that organic compounds are thermally desorbed from the hot surface as gaseous molecules; as they diffuse from the hot air near the pan into cooler air, selected compounds exceed their saturation concentration and nucleation occurs.

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**Practical Implications**

These experiments indicate that as a metal surface is exposed to indoor air or washed with a detergent, organics accumulate that can later be a source of ultrafine particles (UFP) (>10 nm) when the surface is heated. This mechanism may apply to other indoor surfaces that are periodically heated [e.g. steam irons (as shown in this manuscript), electric stovetops, toasters and toaster ovens, hot water or steam radiators, electric baseboards, and electric resistance heaters]. Exposure to such UFP is common, with exposure being highest when an individual is close to the heated surfaces, as occurs during cooking or ironing. This constitutes a route of exposure to these semi-volatile organic compounds (SVOCs) that has not been highlighted in previous studies. Knowledge regarding the source and mechanism of UFP formation during the heating of metal surfaces is relevant to understanding and controlling risks from indoor particle exposures. Studies that characterize the chemical constituents of UFP, including the 2–10 nm size range, are needed to further our knowledge of this important mechanism.

## Introduction

Multiple studies have documented particles from cooking. UFP (diameters < 100 nm) have been shown to be produced by both gas and electric stoves (Dennekamp et al., 2001; Glytsos et al., 2010; Schripp et al., 2011). For gas stoves, some of the particles are emitted by the combustion of the fuel (Dobbins, 2007).

For electric stoves, the particles have been shown to be produced in the absence of food (Wallace et al., 2008). However, is the source the heating element itself, or is it material that has deposited or sorbed on the heating element? Particles may also be produced by the pot or pan, but again the question is the source – is it the pan itself, or material on the pan? This study was undertaken to answer these questions.

## Methods

We used identical condensation particle counters (CPC; Model 3007; TSI, Shoreview, MN, USA) to measure different concentrations of particles >10 nm (in electrical mobility diameter) in over 200 controlled experiments in two homes. The lower size cutoff of about 10 nm was confirmed by Hämeri et al. (2002). The great majority of the particles produced are <100 nm in diameter (UFP), so the instruments are considered UFP monitors. A regression of the two co-located monitors resulted in a slope of 1.05 and an $R^2$ of 97% ($n$ = 24 000 1 s measurements). Fine particles ($PM_{2.5}$) were measured using a laser photometer equipped with a 2.5 μm impactor (Model AM510 Side-Pak; TSI). The monitor was calibrated against gravimetric measurements of tobacco smoke, resulting in a calibration factor of 0.3 being applied to the monitor readings (Dacunto et al., 2013; Jiang et al., 2011).

Two identical single-coil, 120-volt alternating current electric burners were purchased (Proctor-Silex '5th burner'; Hamilton Beach, Southern Pines, NC, USA). One burner used 960 watts and drew 8.2 amperes, the other used 870 watts (amperes not measured, but calculated as 960/120 = 8.0 amperes). Most experiments used 3- to 5-min heating periods. Longer periods (6–15 min) were also employed to study behavior at higher temperatures. The burner itself and three types of pans were tested fully: stainless steel, cast iron, and coated with polytetrafluoroethylene (PTFE). Several other items were also tested, including two larger pans, a toaster oven, a 2-slot toaster, and a steam iron (Table 1). However, these items, with the exception of the steam iron, were too large to allow attainment of uniform temperatures and thus did not allow us to achieve 'clean' (zero or near-zero particle production rates). These experiments are discussed in the Supporting Information. (The steam iron results are included in this main text.)

### Stainless steel pan

Two identical new 8-inch (20 cm) diameter triple-ply stainless steel pans were purchased (All-Clad, Canonsburg, PA, USA). No food or cooking oil was heated on the pans for the duration of the experiments. An older identical pan that had been used for cooking was also employed in some experiments, to study the effect of washing. The two new pans were never washed. The older pan was washed multiple times, using different washing materials, including dishwasher detergents, cleansers containing oxalic acid, and steel wool.

### Cast iron pan

A 5-inch (12.5 cm)-diameter pre-seasoned cast iron pan was purchased. Most commercially available cast iron pans are 'pre-seasoned' by manufacturers because of the rapid accumulation of rust by unseasoned cast iron. The seasoning consists of a light vegetable oil, repeatedly rubbed in and then heated. We considered buying an unseasoned pan, but because they are not common, we bought a pre-seasoned pan. We did not add oil or further seasoning to the pan. This pan was not washed, in accordance with the manufacturer's instructions.

### PTFE-coated pan

A newly purchased 5-inch (12.5 cm)-diameter pan (T-Fal; Groupe SEB, Ecully, France) was employed. The small coated pan was washed according to the manufacturer's instructions (gentle detergent, nylon scrubbing brush, or blue sponge).

### Steam iron

A new steam iron was also bought and tested at both the dry (low and high temperature) and steam (high and low steam) settings.

**Table 1** Dimensions, areas, and mass of tested items

| Description | Diameter top (cm) | Diameter bottom (cm) | Height (cm) | Area bottom (cm$^2$) | Area sides (cm$^2$) | Total area (cm$^2$) | Mass (g) |
|---|---|---|---|---|---|---|---|
| Stainless steel pans | 21.6 | 14.0 | 3.8 | 153.5 | 67.7 | 221 | 703 |
| Small cast iron pan | 12.1 | 10.2 | 2.5 | 81.3 | 28.4 | 109 | 498 |
| Small coated pan | 12.1 | 9.5 | 2.5 | 71.0 | 27.7 | 99 | 137 |
| Large coated wok type | 27.9 | 15.2 | 8.9 | 182.6 | 192.3 | 374 | 1178 |
| Large coated griddle | 30.5 | 29.2 | 3.8 | 670.3 | 113.5 | 784 | 1078 |
| Toaster oven interior | 30 (l) | 29 (w) | 15.2 | | | 3600 | 5280 |
| Toaster (stainless steel with two slots) | 25 (l) | 16 (w) | 21.0 | | | 3900 | 1452 |
| Burner area | 12.7 | 12.7 | | | | 127 | 915 |
| Burner heating element | 94 (l) | 0.6 (w) | | | | 60 | NA |
| Steam iron | 0 | 10.1 | 22.9 | 180.6 | 0 | 181 | 1080 |

l, length; w, width.

Two digital thermometers with metal probes suitable for measuring the temperatures reached by the burner coil and the pans were employed (Model 51; Fluke, Everett, WA, USA).

All experiments were carried out in two private homes. In one home (Home 1), a kitchen (68.2 m³) or a smaller office (25.2 m³) was used. The doors were closed in both rooms, and a small fan employed to promote faster mixing. In the kitchen, the burner was placed in the approximate center of the room – at the end of the counter of the 'peninsula' containing the stove. The Model 3007 was placed on the kitchen table about 2 m from the stove and 1 m from one wall. The fan was placed near the opposite wall. In the smaller room, the burner was on the floor about 1 m from one wall, while the Model 3007 was placed about 2 m away in the center of the room. The fan was placed near the opposite wall.

In the other home (Home 2), most experiments took place in a breakfast nook, 3 m from the burner, with all external windows and doors to the outside closed. Home 2 had a vaulted ceiling and its kitchen was open to the larger volume of the house (460 m³). As in this home, the source was close to the measuring instrument, no attempt was made to calculate total particle production. However, the measured peak concentrations were considered to reflect the relative changes in particle production at different times. Also, several experiments were carried out in a closed room (43 m³) with a small fan to compare with Home 1.

In both homes, a typical experiment involved turning on the burner to its maximum setting and then turning it off after a heating period of 5 min (to the nearest second). Before each experiment, the Model 3007 was turned on to record the background particle concentration (typically 1000–3000 particles/cm³). The peak concentration typically ranged from 20 000 to 200 000 cm⁻³. Subsequent experiments on the same day required waiting until the concentration returned to background levels. The temperature probe was placed in contact either with the burner coil or with the bottom (interior) of the pan.

The two homes differed in the frequency and extent of cooking activities. In Home 1 (two residents), extensive baking of bread and muffins took place several times per week. Also, breakfasts involved toasting muffins in a toaster oven, while most dinners involved cooking of meat or fish along with potatoes or rice and green vegetables. In Home 2 (one resident), there was little indoor cooking, and no cooking took place on the dates in which experiments were performed. Also, both residents in Home 1 spend an unusual amount of time indoors at home, so that SVOCs associated with personal activities other than cooking may be elevated, compared with Home 2.

In Home 1, the total number of particles produced during an experiment was determined by multiplying the peak concentration (minus the background concentration) by the volume of the room. As the peak was typically reached 3–5 min after the end of the heating period, there was little time for particles to be lost by air exchange, deposition, or coagulation, so we did not correct for these loss terms. We refer to this calculated total as the 'particle count'.

## Results

### Electric burner alone

Each of the two electric burners was tested multiple times in each house ($n = 76$ and $n = 43$ for homes 1 and 2, respectively) in succession, with intervening times varying from an hour to several days. The number of particles produced declined over time, eventually reaching zero in most cases (Figure 1). After some days, however, a new set of experiments consistently showed that the burner had 'recovered' in the intervening time and was now capable of producing large numbers of particles. The mean peak burner temperature in Home 1 was 417°C (s.d. 57°C) with a range of 338–542°C. The particle counts for Home 1 ranged from zero to $7.6 \times 10^{12}$. All 76 experiments in Home 1 are documented in Table S1.

Figure 1 also shows the results for the fine particle Sidepak monitor, located next to the UFP monitor. Using the published calibration factor of 0.3 (Dacunto et al., 2013; Jiang et al., 2011), we found that the $PM_{2.5}$ mass concentrations (10-s averages) did not exceed 1 $\mu g/m^3$ greater than background in any of our experiments.
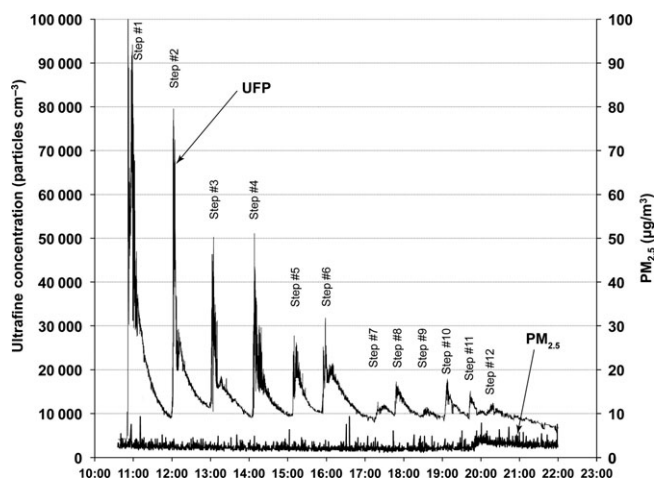


**Fig. 1** Twelve experiments with 5-min heating steps on an electric burner. The peaks typically are reached a few minutes after the end of the heating period. The small increase in the PM2.5 concentration around 20:00 is attributable to infiltration of ambient $PM_{2.5}$ from wood burning activity in the neighborhood, which has been measured previously and often results in a small peak around 8–9 PM. These results are from Home 2; all other Figures are from Home 1

New stainless steel pans

One of the two new stainless steel pans was tested 142 times in Home 1 (Table S2). The first series of 4-min burns required 28 repetitions over a period of 37 days before the pan eventually reached zero particles produced. Although there were several periods when the number counts were low, the pan appeared to accumulate new material between successive heating experiments resulting in a newly replenished reservoir for more particles. The pan was uncovered throughout except for the final week, when it was covered. However, even covered, it accumulated enough material to increase its former output by an order of magnitude. This appears to be evidence that the material is able to deposit on the pan without depending on gravitational settling. Subsequent series of tests required fewer repetitions, varying from 2 to 14, before again reaching zero emissions. As these later tests were completed over a much shorter period than 37 days, it seems likely that the smaller number of repetitions to reach zero emissions reflected less time between repetitions for organic material to accumulate.

In several experiments, a pan was heated for 5 min at a time until it was shown to emit no particles. The pan was then heated for 7 min. The effect was consistently to produce more particles during the 2 min of extra heating time (Figure 2). This result suggests that some material had remained on the pan throughout all the 5-min burns, but the extra two minutes (at higher temperature) released additional material.

When the empty pan sitting on the electric burner was exposed to indoor air for 5 h or less, we found that particle counts following heating for a few minutes ranged from 0 to $9 \times 10^{12}$ (Figure 3). When the empty pan and grill were exposed to indoor air for longer time periods of 100–300 h, the particle counts were relatively high, ranging from $2 \times 10^{12}$ to $1.2 \times 10^{13}$, suggesting that fresh material had been deposited on the metal surface again. Figure 3 suggests that the increase is initially fast but slows over time, possibly approaching an asymptote. This result is consistent with the concept of equilibrium partitioning of organic matter between air and the exposed surfaces of the pan and burner (see Discussion section). Similar results were obtained for the small coated pan (Figures S2–S4).

A repetition of these experiments involving exposing the pan to indoor air for varying periods of time was carried out in the closed room in Home 2. After bringing the burner down to the point of zero emissions, the 'twin' stainless steel pan (which had been open to indoor air for 1776 h) was heated for 5.0 min and resulted in a peak concentration above background of 145 000 $cm^{-3}$ and a total particle production of $6.24 \times 10^{12}$. It required nine heating steps before the pan was reduced to zero particles. Twenty-four hours later, it was heated again and achieved only a small concentration above background of 10 000 $cm^{-3}$ and a total particle production of $0.45 \times 10^{12}$. As can be seen, these total particle counts of $0.45 \times 10^{12}$ following 24 h of exposure and $6.24 \times 10^{12}$ following 1776 h of exposure agree roughly with the Home 1 results pictured in Figure 3 and extend the period when an asymptote may have been reached from a few hundred to 1776 h.

Used stainless steel pan – effect of washing

The used stainless steel pan was employed in another series of 122 tests (Table S3). In these tests, the pan
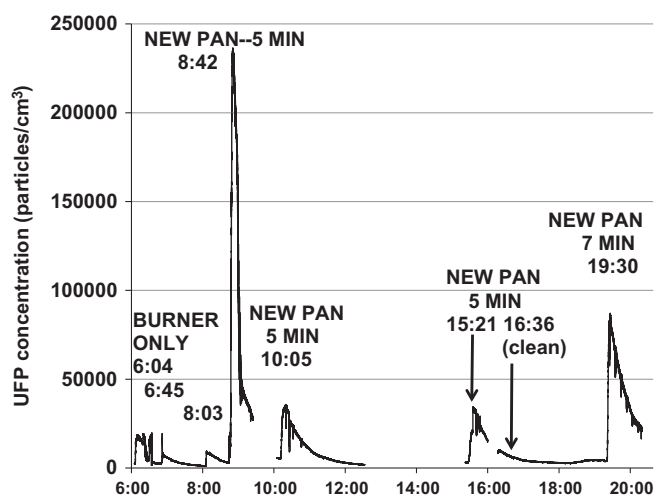


Fig. 2 Effect of increasing heating period from 5 to 7 min on the new empty stainless steel pan. The burner was first heated three times for 5 min each until it produced only a few particles (8:03). The pan was then heated four times for 5-min periods until it produced no particles (16:36). The next heating period (19:30), extended to 7 min, produced a large number of particles (85 000 particles/cm$^3$)
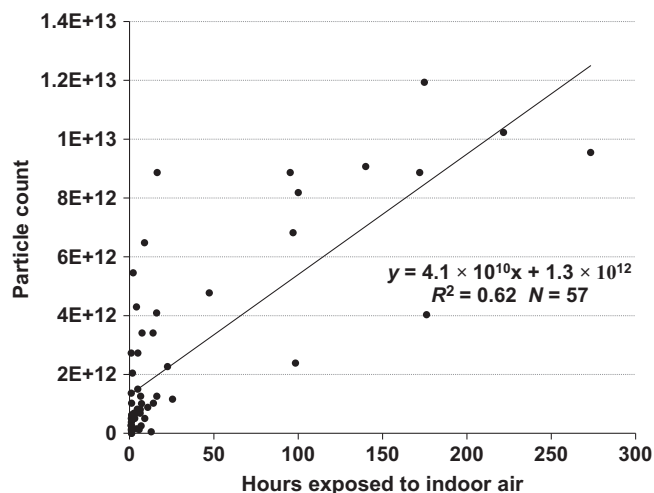


Fig. 3 Particle counts from stainless steel pan heated for 4 min as a function of time exposed to indoor air. Standard errors of the slope and intercept were $0.4 \times 10^{10}$ and $0.3 \times 10^{12}$, respectively

was heated repeatedly as in the experiments on the two new stainless steel pans that had not been used for cooking. For the used pan, we found that its particle counts also could be reduced to zero after repeated heating steps. However, after each series of steps was completed (with the pan showing zero particles produced), the pan was washed, using a variety of washing techniques, most of which involved use of a detergent (Table S4). After each washing using detergent, the freshly washed pan invariably produced a large number of particles, amounting to about 3–4 times as many as the mean of all particles emitted in the subsequent tests. The sequence of particle counts determined from the first test after washing or rinsing to the last in the series (i.e. the point of achieving zero particles produced) is provided in Figure 4. Eight series of tests are pictured. The first test after washing normally has the highest particle count; the subsequent reduction is not always monotonic. This may be due to additional material being added to the pan by airborne sorption between tests (see Discussion section).

Besides the order of experiments after washing (i.e. first heating period after washing, second, and so on), particle counts appeared to depend partially on the duration of the heating (and the consequent increased temperature of the pan). Therefore, a multiple regression was performed on both variables and both were found statistically significant ($P < 0.05$; Table 2).

Coated pan

In all the experiments with the small coated pan, it was eventually found possible to reduce the particle counts to zero or near-zero, although it sometimes required multiple heating steps to achieve the 'clean' (no or few

**Table 2** Multiple regression of particle emission rate on experiment order and pan temperature

|  | $B$ (slope) | Standard error | $t$ (114 d.f.) | $P$-value |
|---|---|---|---|---|
| Intercept | −4.8E + 12 | 4.9E + 12 | −1.0 | 0.3 |
| Order after washing | −2.6E + 11 | 7.4E + 10 | −3.6 | **0.0005** |
| Pan temperature (°C) | 6.8E + 10 | 3.0E + 10 | 2.3 | **0.02** |

d.f., degrees of freedom (i.e. $n-2$).
Bold values are statistically significant.

particles) state. Five series of experiments were carried out. The first two were carried out on the unwashed pan. The next two were after washing in a very small amount of detergent. The last series was begun after leaving the pan exposed to indoor air for about a month. In each series, a short (3-min), medium (4.5–5-min), and long (5–7-min) heating period was employed and the experiments repeated until particle counts reached zero or near-zero for each choice of duration (Table S5).

In general, the first experiment after washing with detergent or after increasing the heating duration (and the resulting pan temperature) produced the highest particle counts. After that, particle counts tended to decrease with each successive experiment in a series (Figure 5). However, at times, the particle counts increased without a corresponding increase in the duration or the temperature. Often these increases were associated with increased time between experiments (e.g. overnight). This may indicate increased deposition of material given the longer time to deposit.

Cast iron pan

Several series of experiments were performed without washing the pan at any time (washing with soap and
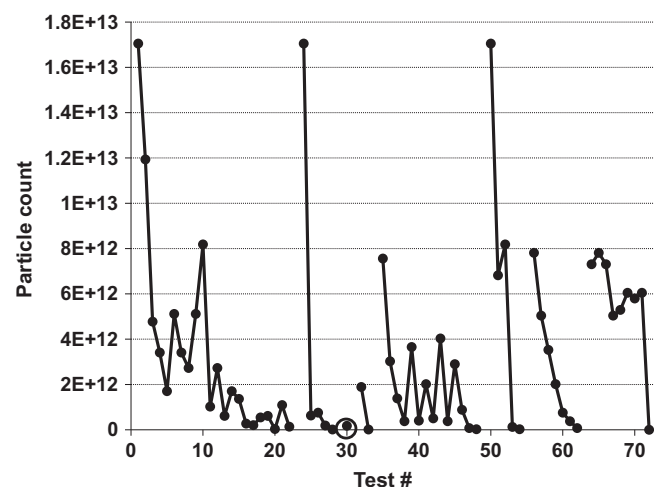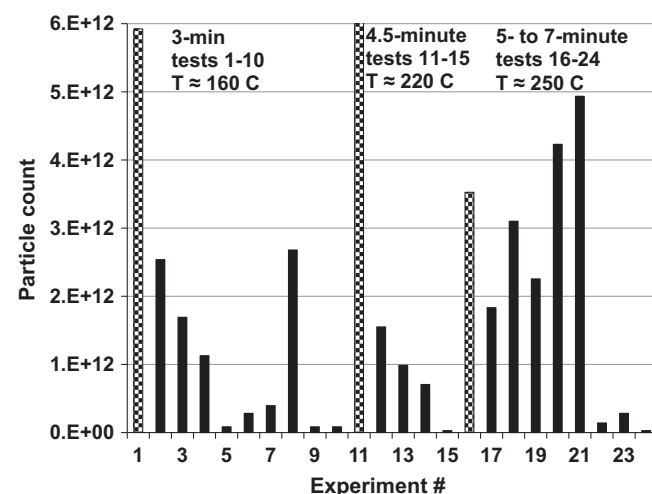


**Fig. 4** Particle counts from repeated heating of a used stainless steel pan. Each of the eight series begins after the pan has been washed and ends when no further particles are observed. Seven series included use of a dishwasher detergent. When the detergent was not used (just rinsing with tap water) (test #30), the particle count was near-zero on the first heating



**Fig. 5** Progression of particle counts per heating period as a function of experiment number and pan temperature for an unwashed coated pan. The figure corresponds to Series 1 in Table S5. The highlighted bars mark the beginning of each heating period–temperature regime. 'Overnight' delays occurred before experiments #8 and #20

water is not recommended by manufacturers, even after cooking, as it can strip the seasoning off). The experiments followed the same protocol employed for the coated pan, with an initial heating period of 3 min, repeated until attainment of 'clean' status, followed by increasing the time in steps of 1 min up to 10 min and then in steps of 2.5 min up to 15 min at which point the temperature of the pan reached an asymptote of about 260°C (Figure 6; Tables S6 and S7). Each time the heating duration was increased (1 min corresponded to about a 15–30°C increase in temperature), a 'new' burst of particles occurred, suggesting mobilization of material previously unaffected by the heating at lower temperatures.

Steam iron

The iron achieved temperatures between about 105 and 125°C within 2 min of being turned on (Figure S5), except for the low temperature dry setting, for which the maximum temperature was about 55°C. Successive tests of the iron at the highest steam temperature resulted in reduction of particle emission rates to 4% of the initial rate after 10 experiments (Figure 7; Table S8).

## Discussion

Although earlier studies have documented increased ultrafine particle concentrations associated with electric stoves and cooking utensils (Schripp et al., 2011; Wallace et al., 2008), the ultimate source of the particles has not been unambiguously identified. Dennekamp et al., (2001) called attention to this problem, commenting that the source could have been from the burner elements themselves or from material deposited on them, or both.
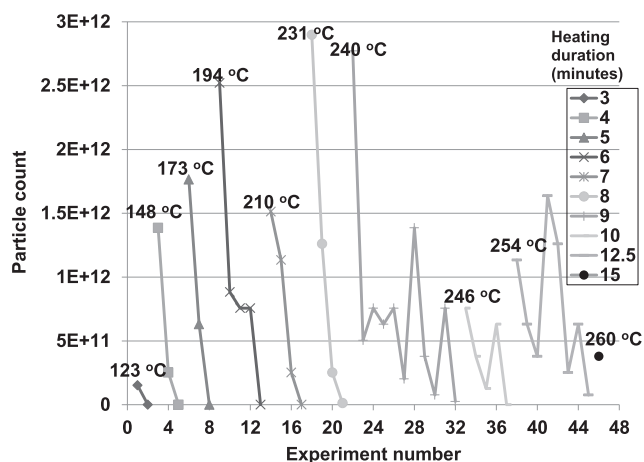
**Fig. 6** Particles produced from small cast iron pan for heating periods of 3–15 min and peak pan temperatures of 120–260°C. In all cases, particle production is forced to zero by repeated heating for the same duration
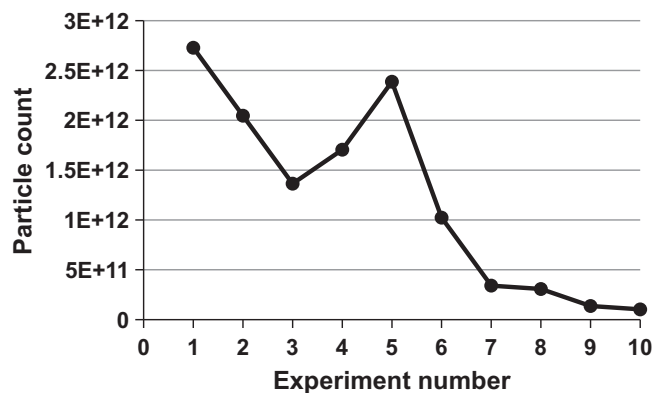
**Fig. 7** Particles produced by steam iron in successive 10-min heating periods

Ultrafine particles produced from heating metallic wires have been observed for many years (Goldsmith et al., 1966; Nolan and Kennen, 1949). However, Scheibel and Porstendörfer (1983) reported that particle production from a silver source required a temperature of 900°C, and Goldsmith et al. (1966) referred to temperatures exceeding 1000°C. More recently, studies of atomic cluster emissions from glowing wires (Fernandez de la Moya et al., 2003; Peineke et al., 2006, 2009) show that emission of atomic clusters is a function of the melting temperature $T_m$ of the wire and begins to increase when temperatures approach $T_m$. For most metals, $T_m$ is in the range of 1000–2000°C (for iron it is 1811°C). As our cast iron pan did not reach temperatures in excess of 290°C, we believe it did not emit atomic clusters that could serve as nuclei for subsequent SVOC condensation and particle growth to >10 nm. The <1000-watt electric burner employed in this study was unable to achieve heating element temperatures >600°C, so we are uncertain whether the burner alone was able to produce particles directly. If the burner or the pans did produce UFP by this mechanism, then the particles could conceivably serve as nuclei for subsequent condensation and growth to become observable by the Model 3007. However, our ability to drive the number of particles >10 nm down to zero suggests that direct production from the heated metallic surfaces was not the source of the observed particles, unless the direct production mechanism also had a limited reservoir of material, which could then be replaced over time, both of which conditions are unlikely. For example, Peineke et al. (2006) show that their heated palladium wire can generate particles at a constant rate for more than 11 h.

Our results suggest that for particles >10 nm, the source is neither the heating element nor the pan *per se*, but organic matter sorbed on the burner and pan. Our experiments show that, in indoor environments, the organic matter can accumulate on the exposed surfaces in a short time (hours or days). Organic matter

can also be deposited on cookware surfaces by washing with detergent, and the material is not then removed even by extensive rinsing.

These conclusions apply to the electric burner itself, to all three types of pans tested – stainless steel, cast iron, and coated – and to the steam iron that was tested. The highest particle counts, for a given heating time and temperature, occur the first time the pan is heated after washing with detergent or after exposure to room air for an extended time. Particle counts then generally decline on subsequent repeated heating steps. However, at later times, the particle counts can increase, particularly if the item has been re-exposed to indoor air for an extended period.

Numerous volatile and semi-volatile organic compounds (VOCs, SVOCs) are commonly found in homes at concentrations ranging from nanograms to micrograms per cubic meter (Polidori et al., 2006; Rudel et al., 2003, 2010; Wallace, 1987; Weschler and Nazaroff, 2008). These compounds are emitted by consumer products, cleaning materials, building materials, combustion, and other processes (Wallace et al., 1987). As demonstrated in studies of window films, including films on the interior surfaces of windows, organics in room air can partition between the gas-phase and indoor surfaces (Butt et al., 2004; Cetin and Odabasi, 2011; Duigu et al., 2009; Gewurtz et al., 2009; Li et al., 2010; Liu et al., 2003; Pan et al., 2012). Indeed, the partitioning of organics between the gas-phase and indoor surfaces has been studied for more than two decades (Zhang et al., 2002 and references therein; Weschler, 2003), beginning with work by Tichenor et al. (1991) that developed equations to describe the sorption and desorption of organics from indoor surfaces. More recently, Weschler and Nazaroff (2012), using measurements from the US EPA's Children's Total Exposure to Persistent Pesticides and Other Persistent Organic Pollutants study, have demonstrated that the concentration of SVOCs in surface films can be estimated from an SVOC's octanol/air partition coefficient ($K_{oa}$) and its gas-phase concentration. In other words, indoor gas-phase organics have a tendency to initially adsorb (physisorb) onto clean indoor surfaces; as organic films develop, indoor organics further absorb into and equilibrate with these surface films. Organic compounds in indoor air appear to be the source of the observed accumulation on an electric iron, electric burners, and pans in the two homes. The amount of organic matter available for such partitioning depends on temperature and relative concentrations in infiltrating outdoor air and on indoor reservoirs such as floors, walls, and dust. However, the amount absorbed is only a small fraction of the total amount available in typical room air. After a pan has achieved near-zero particle counts from repeated 5-min heating periods, extending the 5-min period to a 7-min heating period results in renewed particle production.

The extra time heating increases the temperature of the pan from about 120–150°C to as much as 250–290°C. The higher temperature results in the thermal desorption of less volatile SVOCs that had low desorption rates during shorter heating periods. In this sense, the burner or pans may be viewed as gas chromatographs, releasing ever less volatile compounds as the burner or pan reaches ever higher temperatures. (Note: At temperatures of 250–290°C, some oxidation/decomposition of sorbed organics may also occur.)

The appearance of new particles following washing of the pans is consistent with small residues of organic constituents of the cleaning agents remaining on the pans. Most of the washing experiments employed typical amounts of detergent, but the last two used successively smaller amounts. One used three drops (<0.5 ml) and another used a single drop, each diluted 500 times. Yet the number of particles produced immediately following washing was similar for all three amounts, and for different amounts of flushing with clean water. This result indicates that only a small amount of residual detergent provides sufficient organic matter for particle formation and that the residue is not readily dislodged by tap water flushes (Milenovic et al., 2011; US EPA, 2014). Indeed, rinsing with nearly pure isopropyl alcohol did not eliminate the formation of new particles following washing with detergent.

Simple thermal desorption of organic matter from the surface of a coil or pan does not explain the subsequent observations of UFP. One possible explanation for these observations is based on the fact that organic molecules have vapor pressures that increase with temperature; for some classes of organic compounds (e.g. phthalate esters), the increase with increasing temperature can be relatively large. With this in mind, consider a new stainless steel pan that has various SVOCs sorbed on its surface. When the pan is heated for 5 min, the surface of the pan reaches 155°C, and some of the SVOCs are thermally desorbed. Close to the surface of the pan, the temperature is much higher than room temperature, and the SVOCs in the diffusing 'cloud' have concentrations lower than their saturation vapor pressures. As the 'cloud' of semi-volatile compounds diffuses further into the room, the temperature falls, and at some point, the concentration of selected compounds in the cloud exceeds their saturation vapor pressure – we have a packet of air that is supersaturated in selected SVOCs. Shortly after this point, heterogeneous or homogeneous nucleation occurs and UFP are formed. Additional condensation leads to further particle growth. These newly formed UFP are then 'counted' by the particle counters employed in this study.

Initial calculations suggest that estimated ranges for the flux of organic material toward indoor surfaces are broadly consistent with the relationship between numbers of particles and time of accumulation. The slope

in Figure 3 is $4 \times 10^{10}$ particle counts per hour exposed to room air. For UFP particles with diameters between 5 and 20 nm, estimated mass emission fluxes ranged from 2 to 40 $\mu g/m^2/h$ of exposure to room air for the stainless steel pan. This range was calculated by assuming unit density spherical particles with diameters between 5 and 20 nm and multiplying the mass of a single particle by the particle counts per hour. For total gas-phase SVOC concentrations between 1 and 10 $\mu g/m^3$, a reasonable range for indoors (Rudel et al., 2003; Weschler and Nazaroff, 2008), and a mass transfer coefficient of 3 m/h (Weschler and Nazaroff, 2008), the predicted flux of organic matter to the stainless steel pan is 3–30 $\mu g/m^2/h$ of exposure. This estimate is consistent with the growth rate of 2–32 $\mu g/m^2/h$ measured for the growth of organic films, exclusive of inorganic particles, on exterior window surfaces by Li et al. (2010) and 55–90 $\mu g/m^2/h$ on sheltered 3-mm-diameter glass beads at an outdoor urban site measured by Wu et al. (2008). Hence, this crude exercise suggests that the sorptive flux of gas-phase SVOCs is sufficient to explain the UFP particles produced by the pan (following different intervals of exposure).

In our residential indoor experiments, we found that multiple repeated 5-min heating steps caused a pattern of decreasing particle counts. With as many as 12 or more repeated 5-min heating steps, it was possible to bring the particle counts from our electric burners essentially to zero in both homes. When the burner was first heated above 150°C, a fraction of organic matter on the coil desorbed. Each repeated heating step desorbed additional organic matter until insufficient sorbed organic matter remained to form a detectable number of UFP upon heating. However, over the course of a few days in which the burner was not used, the coil was presumably replenished with SVOCs that sorbed onto its surface from the indoor air; a large burst of particles then occurred when it was first used again.

Schripp et al. (2011) conducted controlled laboratory experiments on 13 electric appliances (including toasters, steam irons, and an electric grill) and reported ultrafine particle counts of 1.1–3.9 $\times 10^{12}$ when these appliances were operated for the first time ('Phase 1' in their terminology). These values are remarkably similar to the values reported in our study for the initial heating regimes. Included in their study were several repeated experiments similar to the multiple repeated heating steps we conducted, and for most items, they saw a decrease in the particle counts similar to the decrease we observed. However, they did not carry out as many repeated heating steps as conducted in the present study, and, in the case of one iron, a full day elapsed between each repeated measurement. Finally, most of the items tested were more complex than the small pans in our study. In agreement with their results, we were unable to reduce the particle counts for the toaster or for the larger pans (10 inch or

greater) to zero, possibly because of not being able to produce sufficiently high and homogeneous temperatures over the entire area of the items. For these reasons, in part, Schripp et al. apparently did not reach a point where the measurable UFP was close to background values. They conclude that their laboratory findings indicate the presence of sorbed semi-volatile compounds 'has an influence on emission strength but is not the main cause of particle release', but they do not identify the major source of the UFP. As noted above, it may be that further testing of the simpler appliances such as the electric grill and the steam irons with more tests and shorter intervals between each test could reduce the particle emissions to near-zero, as was the case with the electric burner and steam iron tested in the present study. We note that the first three tests of our iron also showed little reduction in particle counts, and in fact the fourth test occurred more than 20 h after the third and a renewed increase in particles was noted, possibly due to sorption of additional organics during the time exposed to indoor air. However, subsequent tests over the next 16 h succeeded in driving the particle count toward zero (Figure 7).

Bhangar et al. (2011) studied UFP particle concentrations and exposures in seven homes in northern California, using a water-based CPC capable of measuring particles down to 6 nm diameters. The authors concluded that indoor source activities, 'most notably cooking', caused the highest peak exposures. In one home, heating with a gas furnace caused increases in UFP concentrations. The authors noted, in agreement with our own findings for repeated heating of metal surfaces, that repeated heating of the furnace over a short time resulted in a decline of the source strength.

A serious limitation of this study is the restriction to particles >10 nm. Wallace et al. (2008) showed that particles produced by electric stovetop coils were predominantly (>90%) in the size range from 2.5 to 10 nm, with a mode at about 5 nm. What the present study suggests is that particles >10 nm are not produced by the burner coils or by the stainless steel pans *per se*, but rather by organic matter sorbed on the coils or pans. Whether particles smaller than 10 nm are also produced by organics sorbed on the surface or by the heating elements or the pan *per se* remains unknown. This can be determined using a specialized CPC or a scanning mobility particle sizer capable of counting particles down to approximately 3 nm in size and performing a sufficient number of repeated heating experiments in quick succession to determine whether the <10 nm particle production can be driven to zero.

A further limitation of the present study was our inability to measure SVOC concentrations in either indoor air or coating indoor surfaces. We were also unable to determine the composition of the particles. Further research on the composition of particles associated with heating of metallic surfaces is desirable.

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*Possible health concerns*

These results imply that the process of cooking, which places individuals close to heated cookware surfaces, may bring about high exposures to UFP as a result of the heated cookware, regardless of whether food is present. Various studies have shown that often exposure due to cooking is greater than the exposure that same person will receive in most other everyday activities (Bhangar et al., 2011; Wallace and Howard-Reed, (2002); Wallace and Ott, 2011), although Bekö et al. (2013) document that widespread use of candles in Denmark can rival cooking as a major source of exposure. Other studies have shown that the energy source alone (gas and electric stoves, empty toaster ovens) can also produce very large exposures to UFP (Wallace et al., 2004, 2008). A recent careful study also showed that heated cooking oils could produce UFP (Torkmahalleh et al., 2012). No studies, however, have teased out the separate contributions of the stove or oven, the pan, and the food (including cooking oil). Once the particles are burned off, they do not stay burned off, but instead fresh SVOCs partition to the burner and pan and, if several days elapse, they must be burned off again. Even more importantly, if washing in hot soapy water consistently adds a film of organic material to the pan, the pan will emit large amounts of UFP even without other material being added over the succeeding days. The pan will produce approximately as many particles the next time it is used, whether that is a few hours between meals or a few days. The potential of cooking to cause high personal exposures to UFP has been recognized only recently and has not received extensive study. This study has highlighted a mechanism that facilitates the mobilization of SVOCs from heated metallic surfaces with subsequent inhalation as UFP. Some of the SVOCs found in indoor air, and presumably sorbed to the metal surfaces, are known or suspected to have adverse health effects (Armstrong et al., 2004; Dodson et al., 2012; Rudel et al., 2003). Although the mass of the resulting UFP is small, they have enormous surface area, and their small size allows them easily to enter the blood stream through the alveoli (Oberdörster et al., 2007). The evidence suggests that ordinary cleaning of the cookware with water, soap, and cleansers will not reduce, and in fact exacerbates, the tendency for these ultrafine emissions to occur. These findings also may be relevant to professional chefs and cooks (Yenugadhati et al., 2009) who are exposed to cooking and heated cookware for extended time periods in commercial settings, but this topic is beyond the scope of the present investigation.

## Supporting Information

Additional Supporting Information may be found in the online version of this article:
**Figure S1.** Particle production from old used all-Clad pan as a function of the order in which the experiment was performed following washing of the pan.
**Figure S2.** For the small coated pan heated for 3 min, particles produced are plotted as a function of time exposed to indoor air.
**Figure S3.** Same as Figure S2 for heating durations of 5–6 min.
**Figure S4.** Same as Figure S2 for heating durations of 7 min.
**Figure S5.** Temperatures achieved by steam iron over 11-min tests.
**Figure S6.** Measurements of ultrafine and fine particles using a new toaster without toast in five consecutive 3-min toasting operations followed by toasting a slice of bread.
**Figure S7.** Photo of measuring equipment and items tested in home 1.
**Table S1.** Results from heating the electric burner alone.
**Table S2.** Results from heating the new stainless steel pan.
**Table S3.** Experiments with old used stainless steel pan.
**Table S4.** Washing technique for old stainless steel pan.
**Table S5.** Experiments with small coated pan.
**Table S6.** Temperatures and particle production associated with heating periods of 3–7 min.
**Table S7.** Temperatures and particle production associated with heating periods of 3–15 min: small cast iron pan.
**Table S8.** Particle counts from steam iron.

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