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Particle Emission Factors During Cooking Activities

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Abstract

Exposure to particles emitted by cooking activities may be responsible for a variety of respiratory health effects. However, the relationship between these exposures and their subsequent effects on health cannot be evaluated without understanding the properties of the emitted aerosol or the main parameters that influence particle emissions during cooking. Whilst traffic-related emissions, stack emissions and ultrafine particle concentrations (UFP, diameter < 100 nm) in urban ambient air have been widely investigated for many years, indoor exposure to UFPs is a relatively new field and in order to evaluate indoor UFP emissions accurately, it is vital to improve scientific understanding of the main parameters that influence particle number, surface area and mass emissions. The main purpose of this study was to characterise the particle emissions produced during grilling and frying as a function of the food, source, cooking temperature and type of oil. Emission factors, along with particle number concentrations and size distributions were determined in the size range 0.006-20 µm using a Scanning Mobility Particle Sizer (SMPS) and an Aerodynamic Particle Sizer (APS). An infrared camera was used to measure the temperature field. Overall, increased emission factors were observed to be a function of increased cooking temperatures. Cooking fatty foods also produced higher particle emission factors than vegetables, mainly in terms of mass concentration, and particle emission factors also varied significantly according to the type of oil used.

Keywords: Ultrafine particle concentration; indoor emissions, aggregate, size distribution, cooking activities.

1. Introduction

Numerous studies have found associations between airborne particulate matter and negative effects on human health (Kreyling *et al.*, 2006). A number of epidemiological studies also associated these effects with particle mass concentration, including PM_{2.5} (Pope, 2000) and PM₁₀ (Loomis, 2000), as well as ultrafine particle (UFP) number concentration (Hauser *et al.*, 2001) and overall exposure rate (Siegmann and Siegmann, 1998). Anastasio and Martin (2001) also suggest that health effects from airborne particles are strongly associated with co-exposure to other airborne pollutants.

Although many studies have investigated submicron and UFP in urban ambient air, an important gap in knowledge still exists with respect to indoor environments. In many countries, the majority of people spend most of their time (80–90%) indoors, where cooking represents one of the most

significant particle generating activities (Kamens *et al.*, 1991; Ozkaynak *et al.*, 1996). In addition, UFPs emitted from cooking activities have also been associated with many respiratory ailments, including lung cancer (Dennekamp *et al.*, 2001; Ko *et al.*, 2000; Wallace *et al.*, 2004).

In order to gain a better understanding of the relationship between particulate air pollution and gas cooking, several studies have attempted to measure the particle number concentration and size distribution of particles generated during cooking (Abt et al., 2000a, b; Brauer et al., 2000, Dennekamp et al., 2001; Hussein et al., 2006; Li et al., 1993; See and Balasubramanian, 2006a, b; He et al., 2004; Yeung and To, 2008; Wallace et al., 2004). These studies provided valuable information on the characteristics of particles generated by different cooking methods. For example, Abt et al. (2000a, b) conducted an intensive study in houses in Boston USA, in order to characterise sources of indoor particles and found that cooking activities, cleaning and the movement of people has a significant impact on indoor particle concentrations. He et al. (2004) quantified the emission characteristics of indoor particle sources in 15 houses in Brisbane, Australia, and found that some indoor activities increased indoor particle number concentration by 1.5-27 times the concentrations observed when there was no indoor source in operation. Wallace et al., 2004 performed an 18 month study in a four-bedroom, three level house located near Washington DC, USA and found that selected cooking episodes (mostly frying) produced particle concentrations to the magnitude of 10¹⁴ after only 15 minutes of cooking, more than 90% of which were in the ultrafine range. Li et al. (1993) measured particle size distributions of sub-micrometer aerosols from cooking fumes in an apartment in Taiwan. Aerosols were generated from three types of domestic cooking processes, namely scrambling eggs, frying chicken, and cooking soup, and on average, the corresponding mode diameters of particles concentrations were 40 nm, 50 nm, and 30 nm, respectively. The concentrations of UFP and of nitrogen oxides generated by cooking with both gas and electricity in a laboratory (with no mechanical ventilation and the windows closed) were carried out by Dennekamp et al. (2001). They focused on domestic cooking activities and used a very small amount of vegetable oil (15 mL) to fry 500 g of vegetables or 4 rashers of bacon. They found that gas cooking stoves generated more UFP than electrical ones and that the peak UFP concentration generated on gas by frying fatty foods was significantly higher than that generated by frying vegetables. Their results also showed that cooking processes produced peak numbers of particles in the range 22-72 nm.

More recently, See and Balasubramanian (2006a, b) performed controlled experiments in a domestic kitchen using five different gas-cooking methods. Their results showed that deep-frying

produced the highest concentration of particles, up to 6.0x10⁵ part/cm³, and the mode diameter was approximately 20 nm. Finally, Yeung and To (2008) examined the size distribution of aerosols emitted from commercial cooking activities. Particle number concentration measurements revealed that the particle size distribution followed a lognormal distribution and that the aerosol mode diameter increased as cooking temperature increased. Similarly, at a higher cooking temperature, more aerosols formed in the accumulation mode and bimodal distributions were detected (Siegmann and Sattler, 1996).

Even though a significant amount of number and mass emission factor data exists for cooking activities, a lack of understanding of the parameters that influence those emissions factors still remains. Therefore, the main aim of this study was to evaluate the influence of the temperature, as well as food, oil and stove type on number, surface and mass (in terms of PM_{2.5}) emission factors when grilling (cooking without oil over a hot plate, heated by a gas or electrical stove) and frying (typically deep-frying, when food is immersed in hot oil, heated by gas stove or electrical frying machine). In order to produce useful data which can later be used for exposure assessment, it was also important to analyse other aerosol characteristics such as particle morphology (Melia *et al.*, 1977; Speizer *et al.*, 1980; Ware *et al.*, 1984; Volkmer *et al.*, 1995; Jarvis *et al.*, 1996; Jarvis *et al.*, 1998). For example, UFP distribution is often influenced by the presence of aggregates (Lall and Friedlander, 2006; Lall *et al.*, 2006), whose dynamics differ significantly from those of spherical particles, which can then lead to errors in the calculation of aerosol distributions (Friedlander, 2000). In addition, using data based solely on spherical particles can also lead to an underestimation of surface area and an overestimation of volume (and hence mass) distributions.

2. Experimental analysis

2.1 Sampling site

The experimental campaign was conducted between May and September 2008 in an open-plan laboratory in San Vittore del Lazio (Italy), with a total area of 80 m² and a height of 2.8 m. A domestic kitchen was set-up inside the laboratory, with both gas (mixture of butane and propane) and electrical stoves. Two different ventilation conditions were used during the tests: minimum ventilation (doors and windows closed) and normal ventilation (doors and windows closed with mechanical ventilation in operation). For both sets of conditions, the air exchange rate (AER) was measured on the basis of the CO₂ decay curve (He *et al.*, 2004) using a TSI Model 7515 IAQ-CALCTM, calibrated by the manufacturer at the beginning of the experimental campaign. The AERs

were found to be $0.29 \pm 0.05 \text{ h}^{-1}$ for minimum ventilation and $0.89 \pm 0.11 \text{ h}^{-1}$ for normal ventilation conditions.

2.2 Instrumentation

Particle number concentration and size distribution were measured by a Scanning Mobility Particle Sizer (SMPS 3936, TSI Inc., St. Paul, MN) and an Aerodynamic Particle Sizer (APS 3321, TSI Inc., St. Paul, MN). Mass concentration was also calculated using the SMPS/APS data, on the basis of an algorithm well described by Fine *et al.* (2004) and Sioutas *et al.* (1999), with a particle density value equal to 1.7 g/cm³. To estimate the diffusion losses during sampling, the data obtained by the SMPS/APS were also corrected using the methods reported by Chen *et al.* (1998) and Birmili *et al.* (1997). A Condensation Particle Counter (CPC 3775 TSI Inc., St. Paul, MN) used was calibrated at the TSI laboratory in High Wycombe, UK using monodisperse polystyrene latex spheres several days prior to the beginning of the experimental campaign. Flow rates were continuously checked using a TSI Model 4100 Flow Meter. Data analysis was performed using the Aerosol Instrument Manager[®] and Data Merge[®] (TSI Inc., St. Paul, MN) software.

A FLIR System ThermaCAMTM S45 was used to monitor temperature in real-time, with a thermal sensitivity of 0.08 °C. In order to evaluate the thermal emissivity of the grilling and frying surfaces, the values obtained by the FLIR System were corrected as a function of the temperature measured by several thermocouples, calibrated by the European Accredited Laboratory (LAMI), University of Cassino, Italy.

Morphological analysis was conducted using a Nanometer Aerosol Sampler (Model 3089, TSI Inc.) to collect particles on a mesh TEM copper grid with a carbon/formvar support film, and these samples were analysed through a ZEISS EM10CA TEM. In addition, the authors also made corrections to account for the presence of aggregates using the Idealised Aggregate (IA) theory (Lall and Friedlander, 2006, Lall *et al.*, 2008), which is based on the assumption that aggregates are composed of primary particles, all of which have the same (known) diameter. On the basis of the morphological measurements, primary particle diameter was found to be 30 nm, using the IA theory (see Section 3.1).

2.3 Methodology description

In order to determine indoor particles concentration levels, taking into consideration the contributions from indoor and outdoor sources, the deposition rate of particles on indoor surfaces and the AER, the following equation was used (Koutrakis *et al.*, 1992; Chen *et al.*, 2000, Thatcher and Layton, 1995):

$$\frac{dC_{in}}{dt} = P \cdot AER \cdot C_{out} + \frac{Q_s}{V} + (AER + k) \cdot C_{in}$$
(1)

where C_{in} and C_{out} are the indoor and outdoor particle concentrations, respectively, P is the penetration efficiency, k is the deposition rate, Q_s is the indoor particle generation rate, t is time and V is the efficient volume of the laboratory.

In order to estimate the average emission factor, equation (1) can be simplified by using average values instead of functions, and also by making further assumptions about the experimental conditions reported in (He *et al.*, 2004) as follows:

$$EF = V \cdot \left[\frac{C_{in} - C_{in,0}}{\Delta t} + \left(\overline{AER + k} \right) \cdot \overline{C_{in}} - AER \cdot C_{in,0} \right]$$
(2)

where EF is the average emission factor, C_{in} and $C_{in,0}$ are the peak and initial indoor particle concentrations, respectively, $\overline{AER + k}$ is the average total removal rate, and ΔT is time difference between initial and peak concentration. This equation ignores the effects of particle dynamics such as condensation, evaporation and coagulation, since these are considered to be minor, particularly under the conditions normally encountered in residential environments.

All measurements were made at a distance of 2 meters from the stove and the procedure for the cooking activities (both grilling and frying) was as follows: 5 min of background measurements, followed by measurements throughout the duration of cooking and then a further 30-40 min of measurements after the cooking had ceased. Each emission factor value reported in the results represents the mean value of at least three tests and were calculated according to equation (2). A detailed description of the tests is provided in Table 1.

Table 1. Detailed description of the cooking tests.

Cooking activity	Type of stove	Food description	Cooking time (min)	Notes
Grilling	cheese (70 g) pork meat (130 g) bacon (50 g) eggplants (30 g) 8-10		Two tests were carried out: one with the dial at full power and the other at minimum power. Cooking time includes the time required to heat the grill Grill temperature was controlled and monitored using the thermocamera.	
	electric	cheese (70 g) pork meat (130 g) bacon (50 g)	10	Grill temperature was controlled and monitored using the thermocamera.
Frying	gas	chips (50 g) onion (60 g)	8	Dial at full power. Three different oils were tested: olive oil, peanut oil and sunflower oil.
,	electric	chips (50 g) mozzarella (250 g)	10	Three different oils were tested: olive oil, peanut oil and sunflower oil.

3. Experimental results

3.1 Morphological characterisation

Figure 1 shows an example of the aggregate particles observed during cooking. These aggregates show a branched, chain-like structure with a fractal dimension lower than 2 (Dye *et al.*, 2000; Xiong and Friedlander, 2001). On the basis of these microscopic examinations, the average diameter of the primary particles that made up each aggregate was found to be 32.6 nm, with a standard deviation of 12.4 nm. This value is similar to the values for outdoor aggregates reported by Lee *et al.* (2001), Park *et al.* (2004) and Barone *et al.* (2006). Using the IA theory, as outlined in Section 2.2, a primary particle diameter of 30 nm characterisation was used correct data for the presence of aggregates.

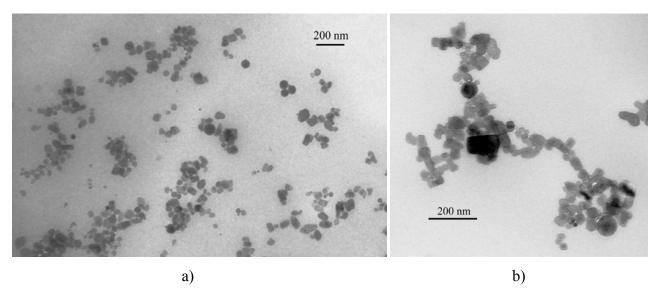


Figure 1. Typical atmospheric particle aggregates sampled by TEM grids in a kitchen during meat grilling using gas stove: a) aggregate particles; and b) close up of a single aggregate particle.

3.2 Characterisation of the grilling source

Table 2 presents the particle number emission factors and the peak concentrations when grilling using the gas and electric stoves. The results showed that the gas stove typically generated more particles than the electric stove. In addition, particle emission rates were found to increase as a function of increasing temperature, and also when the grill was placed on the stove.

Table 2. Summary of the size distribution of the aerosols generated from the stoves: number emission factor (NEF) and number peak value (NPV).

Cooking Activity	NEF particles/min	NPV particles/cm ³	Average temperature and uniformity on the grill at the end of cooking (°C)
Gas stove at full power	$2.1 \pm 0.1 \times 10^{12}$	$9.5 \pm 2.1 \times 10^4$	
Grilling with gas stove at full power	$3.0 \pm 0.2 \times 10^{12}$	$1.5 \pm 0.2 \times 10^5$	325 ± 26
Grilling with gas stove at minimum power	$1.2 \pm 0.1 \times 10^{11}$	$1.7 \pm 0.3 \times 10^4$	37 ± 1.0
Grilling with electric stove at maximum temperature	$1.3 \pm 0.1 \times 10^{12}$	$9.4 \pm 1.9 \times 10^4$	246 ± 7
Grilling with electric stove at minimum temperature	$2.9 \pm 0.2 \times 10^{11}$	$2.6 \pm 0.4 \times 10^4$	178 ± 10

Figure 2 shows the particle number distribution for grilling with the gas at full power. It can be seen that 98% of the emitted particles were in the ultrafine range and the mode diameter of approximately 20 nm is in agreement with the values reported for gas stoves (Dennekamp *et al.*, 2001), stationary gas-fired combustion sources (Chang *et al.*, 2004) and liquefied petroleum gas (LPG) powered vehicles (Ristovski *et al.*, 2005).

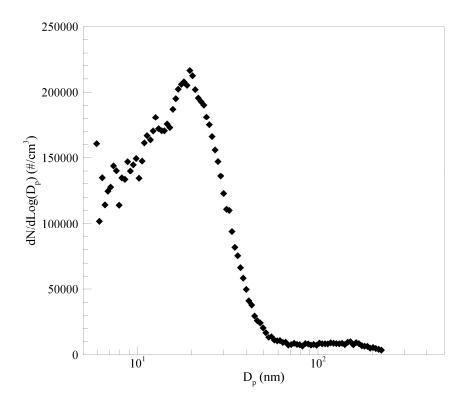


Figure 2. Particle number distribution of the particles emitted during grilling on the gas stove.

3.3 Influence of food type on particle emissions during grilling

Table 3 presents the number, surface area and mass concentration emissions from grilling at maximum power, using a number of different food types. It can be seen that the emission factors for foods containing a high percentage of fat were significantly higher than those for the low fat vegetables. Both surface area and mass emission factors were significantly higher for fatty foods and the ratio of peak mass concentration to background concentration was approximately 40 and 8 for fatty foods and vegetables, respectively. Among the fatty foods analysed, the highest surface area and mass emission factors were observed for bacon, which also contains the highest level of fat compared to the other fatty foods investigated.

Table 3. Summary of the size distribution of aerosols generated using a gas stove at maximum power when grilling cheese, wurstel (pork meat), bacon and eggplants (vegetable). Emission factor (EF), peak value (PV) and mode diameter (MD) for number (N), surface area (S) and mass (M).

	Cheese	Wurstel	Bacon	Eggplants
NEF (part/min)	3.4×10^{12}	3.1×10^{12}	2.8×10^{12}	2.6×10^{12}
SEF (µm²/min)	1.1×10^5	1.3×10^5	2.3×10^5	4.9×10^4
MEF (µg/min)	9.5×10^3	1.0×10^4	4.7×10^3	8.9×10^2
NPV (part/cm ³)	1.1×10^5	1.3×10^5	1.0×10^5	1.2×10^5
SPV (nm ² /cm ³)	4.6×10^9	5.8×10^9	9.8×10^9	2.8×10^9
MPV (μ g/m ³)	383	452	236	78
NMD (nm)	41	43	49	29

During the grilling of fatty foods, the maximum ratio of peak particle number concentration to background concentration was 8.7. The mode was found to be between 40-50 nm for fatty foods and it decreased to a value of around 30 nm for the low fat vegetables (see Figure 3). UFPs were found to account for about 83 % of total submicrometer particles, which is consistent with the findings of Li *et al.* (1991).

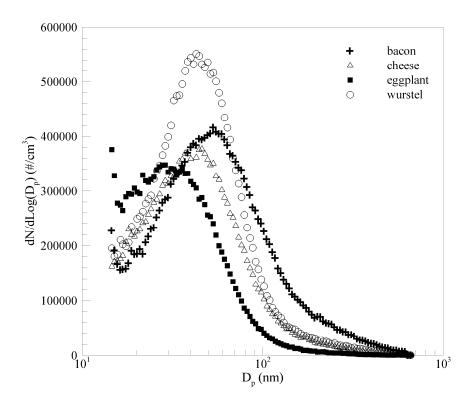


Figure 3. Particle number distribution of aerosols generated using a gas stove at maximum power for the grilling of cheese, wurstel, bacon and eggplants.

3.4 Influence of the temperature on particle emissions during grilling

Temperature has an important impact on the characteristics of particles emitted during cooking activities. Figure 4 illustrates the temperature fields for grilling bacon on a gas stove. The average grill temperature was equal to 242 ± 5.2 °C for the stove operating at maximum power (Figure 4a) and 171 ± 17 °C (Figure 4b) for the stove operating at minimum power. Using the infrared thermal imaging camera, the mean surface temperature of the bacon during the grilling was also determined to be 114 ± 8.1 °C (maximum power) and 82 ± 13 °C (minimum power).

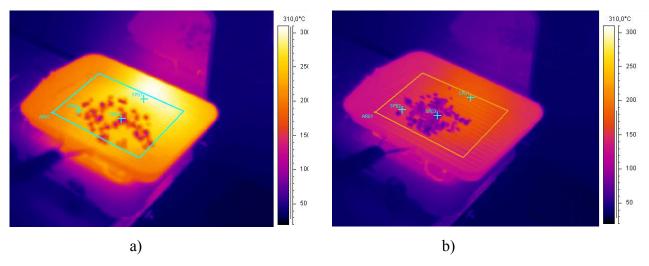


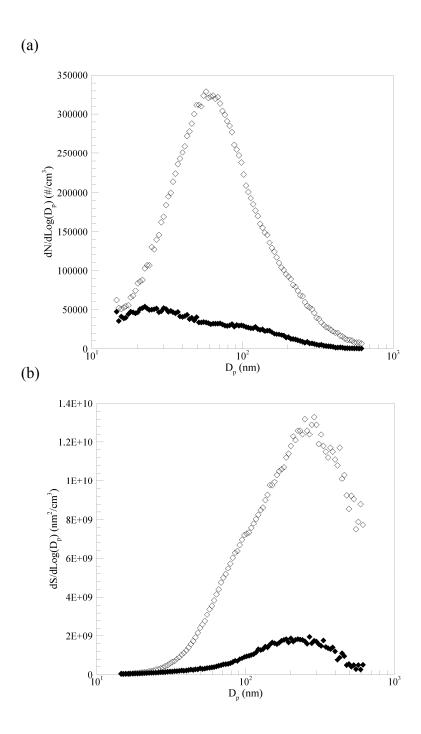
Figure 4. Temperature field for grilling bacon on a gas stove (measured by the FLIR System ThermaCAMTM S45): (a) stove at maximum power, (b) stove at minimum power.

Table 4 presents the emission factors for grilling 50 g of bacon over a gas stove, at both minimum and maximum stove power. It can be seen that grill temperature had a significant impact on emission factors, with an increase of 70% in the case of number concentration, and a 12 fold increase in mass concentration.

Table 4. Summary of emission factors for grilling 50 g of bacon over a gas stove, at both minimum and maximum stove power, including number (N), surface area (S) and mass (M) concentration emission factors (EF).

	Full power	Minimum power	Ratio full power/minimum power
NEF (part/min)	$2.5 \pm 0.2 \times 10^{12}$	$1.5 \pm 0.1 \times 10^{12}$	1.7
SEF (µm²/min)	$2.6 \pm 0.3 \times 10^5$	$3.3 \pm 0.3 \times 10^4$	7.9
MEF (μg/min)	$4.7 \pm 0.5 \times 10^3$	$3.9 \pm 0.2 \times 10^2$	12

Figure 5 shows a graphical representation of the number, surface area and mass distributions obtained for grilling 50 g bacon on a gas stove. In terms of number distribution, the mode diameter is higher when the stove is at maximum power (Figure 5a) ranging from 22-57 nm. This effect was also observed by Dennekamp *et al.* (2001) and Yeung and To (2008). In addition, an increase of the mode diameter can also be observed for the surface area and mass distributions (Figure 5b, c).



(c)

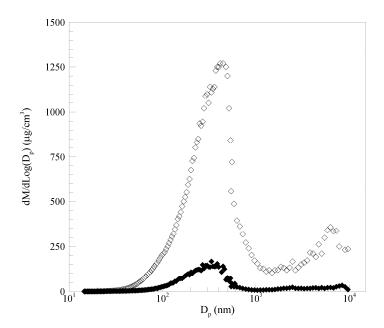


Figure 5. Particle number (a), surface area (b) and mass (c) distributions during the grilling of 50 g bacon on gas stove (◊ - Maximum Power, ♦ Minimum Power).

3.5 Influence of the type of heat source (gas or electric) on emission factors

In order to determine the influence of the type of heat source, emission factors were compared for grilling bacon using both a gas and an electric stove. During the grilling of 50 g of bacon, the mean temperature of the bacon was equal to 71 ± 7.5 °C and to 82 ± 5.8 °C in the case of gas and the electric stove, respectively, with grill temperatures equal to 171 ± 13 °C and 178 ± 6.4 °C. The corresponding emission factors are reported in Table 5.

Table 5. Summary of emission factors for grilling 50 g of bacon over a gas stove and an electric stove, including number (N), surface area (S) and mass (M) concentration emission factors (EF).

	Gas stove	Electric stove	Ratio gas stove/electric stove
NEF (part/min)	$1.5 \pm 0.2 \times 10^{12}$	$1.2 \pm 0.1 \times 10^{12}$	1.3
SEF (µm²/min)	$2.6 \pm 0.3 \times 10^4$	$6.8 \pm 0.5 \times 10^4$	0.38
MEF (µg/min)	$3.0 \pm 0.2 \times 10^2$	$1.7 \pm 0.2 \times 10^3$	0.17

Particle number emission factors were found to be higher when grilling with the gas stove, which can be attributed to the emissions coming directly from the stove. On the other hand, the surface area and mass emissions are only slightly influenced by the stove and mainly depend on the surface temperature of the bacon.

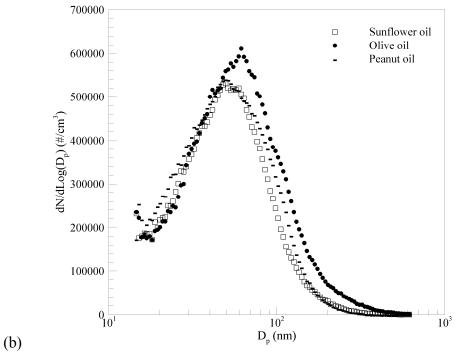
3.6 Influence of oil type on gas stove frying emissions

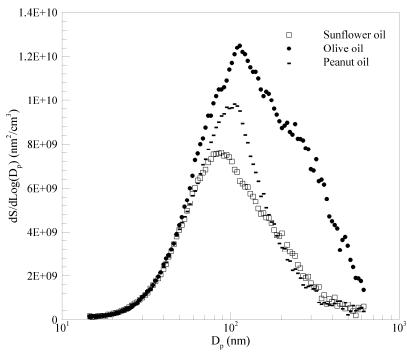
In order to estimate the influence of oil on the emission factors from frying 50 g of chips with the gas stove at maximum power, three different oils were used: i) olive oil, ii) peanut oil and iii) sunflower oil (specifically formulated for frying) (Table 6).

Table 6. Summary of the emission factors for generated using a gas stove at maximum power for the frying of 50 g of chips: emission factor (EF), peak value (PV) and mode diameter (MD) for number (N), surface area (S) and mass (M).

	Olive oil	Peanut oil	Sunflower oil (specific for frying)
NEF (part/min)	1.8×10^{12}	2.3×10^{12}	1.1×10^{12}
SEF (µm ² /min)	2.5×10^5	1.6×10^5	1.2×10^5
MEF (μg/min)	3.3×10^3	1.6×10^3	1.2×10^3
NPV (part/cm ³)	1.2×10^5	1.2×10^5	1.1×10^5
SPV (nm ² /cm ³)	1.1×10^{10}	6.8×10^9	6.0×10^9
MPV (μ g/m ³)	128	68	60
NMD (nm)	61.5	49.6	49.6
Geometric Standard Deviation (nm)	1.91	1.82	1.80

As can be seen from Table 6, the emission factors generated by the olive oil were the highest, indicating that this is not the best type of oil to be used when frying. In contrast, the emissions factors generated by the sunflower oil were the lowest (almost half that of olive oil), indicating that it is the best oil to be used when frying. Figure 6 shows the different number, surface area and mass distributions obtained for each type of oil at cooking temperatures of 226 ± 3 °C, 240 ± 5 °C and 234 ± 3 °C for peanut, olive and sunflower oil, respectively.







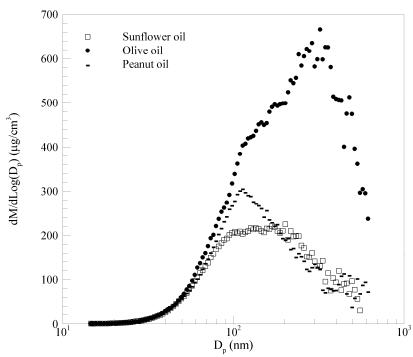


Figure 6. Particle number (a), surface area (b) and mass (c) distributions during the frying 50 g chips with the gas stove at maximum power, using three different kinds of oil: olive oil, peanut oil and sunflower oil.

3.7 Emissions from frying using an electric heat source

In order to compare the emission factors for frying using a gas stove and an electric heat source, an electric frying pan was also used. The frying pan was operated at a constant temperature of 190 °C, which was confirmed by using the infrared thermal imaging camera (Figure 7). The emission factors in frying 50 g of chips and 250 g of cheese (mozzarella) using the three different types of oil (olive oil, peanut oil and sunflower oil) are reported in Table 7.

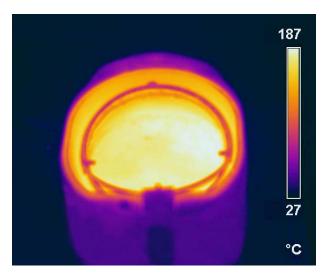


Figure 7. Temperature field on the frying pan during the frying tests.

Table 7. Summary of the emission factors generated using an electric frying pan at 190 °C for frying 50 g of chips and 250 g of cheese (mozzarella) using three different types of oil: emission factor (EF), peak value (PV) and mode diameter (MD) for number (N), surface area (S) and mass (M).

	Chips (olive oil)	Chips (peanut oil)	Chips (sunflower oil)	Cheese (mozzarella) (sunflower oil)
NEF (part/min)	1.2×10^{10}	2.7×10^{10}	1.1×10^{10}	1.5×10^{10}
SEF (µm²/min)	4.0×10^3	7.3×10^3	4.9×10^3	5.0×10^3
MEF (μg/min)	2.0×10^2	6.4×10^{1}	3.2×10^{1}	3.4×10^{1}
NPV (part/cm ³)	2.6×10^4	1.5×10^4	1.4×10^5	1.6×10^5
SPV (nm ² /cm ³)	6.8×10^8	1.3×10^9	5.9×10^8	4.3×10^8
MPV ($\mu g/m^3$)	27	13	12	12

From Table 7 it can be seen that all of the emission factors for the electric frying pan were significantly lower than those generated by frying using a gas stove. The main factor that could reduce the emission factors from frying could be the lower frying temperature (i.e. at or below 190 °C) of the electric frying pan. In addition, the type of food used did not appear to have a significant impact on the emission factors from frying.

4. Conclusions

Overall, it was found that when grilling, the gas stove generated more particles than the electric stove, with negligible differences in emission factors from heating the empty grill at maximum and minimum power on the gas stove. In contrast, the type of food used for grilling did significantly affect emission rates, with foods containing a higher percentage of fat generating higher emission rates. An analysis of the influence of temperature when cooking food on the gas stove also found that particle number, surface area and mass concentration all increased for higher temperatures, for both gas and electric stoves. Experiments to determine the influence of oil type on gas frying found that sunflower oil generated the lowest number, surface area and mass emission factors, whilst olive oil emitted the highest. The emission factors obtained from using an electrical frying pan with the same three oils were found to be well below those observed for frying using a gas stove.

Now that is has been shown that both food and oil type, as well as temperature, have a significant effect on cooking emission, future studies should focus on the morphological and chemical characterisation of the emitted particles, in order to improve knowledge on the characteristics and possible health effects of cooking emissions.

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