INVESTIGATING MEASUREMENTS OF FINE PARTICLE ($PM_{2.5}$) EMISSIONS FROM THE COOKING OF MEALS AND MITIGATING EXPOSURE USING A COOKER HOOD

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ABSTRACT

There is growing awareness that indoor exposure to particulate matter with diameter ≤ 2.5μm ($PM_{2.5}$) is associated with an increased risk of adverse health effects. Cooking is a key indoor source of $PM_{2.5}$ and an activity conducted daily in most homes. Population scale models can predict occupant exposures to $PM_{2.5}$, but these predictions are sensitive to the emission rates used. Reported emission rates are highly variable, and are typically for the cooking of single ingredients and not full meals. Accordingly, there is a need to assess $PM_{2.5}$ emissions from the cooking of complete meals.

Mean $PM_{2.5}$ emission rates and source strengths were measured for four complete meals. Temporal $PM_{2.5}$ concentrations and particle size distributions were recorded using an optical particle counter (OPC), and gravimetric sampling was used to determine calibration factors.

Mean emission rates and source strengths varied between 0.54—3.7 mg/min and 15—68 mg, respectively, with 95% confidence. Using a cooker hood (*apparent* capture efficiency >90%) and frying in non-stick pans were found to significantly reduce emissions. OPC calibration factors varied between 1.5—5.0 showing that a single value cannot be used for all meals and that gravimetric sampling is necessary when measuring $PM_{2.5}$ concentrations in kitchens.

Key Words: cooker hood, gas burner, source strength, size distribution, calibration factor

PRACTICAL IMPLICATIONS

Cooking is a key indoor source of $PM_{2.5}$ in most houses and may contribute significantly to personal exposure and adversely affect health if $PM_{2.5}$ concentrations are not maintained below known health-based thresholds.

When determining $PM_{2.5}$ exposure indoors using an optical particle counter (OPC), its measurements should be accompanied by those from a gravimetric sampler to provide a calibration factor for the OPC with which to scale its measurements. OPC calibration factors vary by meal and so it is only possible to use a single factor for all meals by introducing significant uncertainty.

Good exposure mitigation measures in domestic kitchens include the use of a cooker hood that covers the front burners, the use of non-stick frying pans, and cooking methods that avoid the browning or charring of food. This is especially important in airtight dwellings where ventilation may be inadequate or in other houses during the heating season when occupants seek to reduce ventilation rates to obtain thermal comfort or to minimize heating fuel costs.

1 INTRODUCTION

Airborne fine particulate matter with a diameter ≤2.5 μm ($PM_{2.5}$) has been identified as a priority indoor air pollutant in US homes[1]. This is because $PM_{2.5}$ is prevalent and both acute and chronic exposure is linked to an increased risk of adverse health effects, including cardiovascular and respiratory morbidity, and mortality[2]. Existing research has primarily focussed on exposure to $PM_{2.5}$ from ambient sources. But, there is a growing interest in the risks posed by $PM_{2.5}$ from indoor sources because people typically spend most of their time inside; for example, people in the UK spend 70% of their time in their houses[3]. In addition, improvements in dwelling airtightness without compensatory purpose provided ventilation has led to lower ventilation rates[4] and a reduction in contaminant dilution. In many homes, indoor sources have been found to have a greater effect on indoor $PM_{2.5}$ concentrations than those from ambient sources[5], and so emissions from indoor sources might be an increasingly important source of personal $PM_{2.5}$ exposure.

Cooking has frequently been identified as an indoor $PM_{2.5}$ source by *in situ* monitoring in dwellings[6-10]. Frying and grilling were found to increase indoor $PM_{2.5}$ concentrations by up to 30 and 90 times the ambient concentration, respectively[8]. Furthermore, Chan *et al.*[11] found emission events exceeding 30 minutes were more frequent around meal times, with the highest occurrence between 17:00 and 21:00, suggesting that the cooking of an evening meal is an important $PM_{2.5}$ emission source. Elevated risks of lung cancer, particularly in women, are associated with emissions from cooking and with poor kitchen ventilation[12]. Cooking using traditional Woks in kitchens without a cooker hood is associated with an increased lung cancer risk for non-smoking Taiwanese women[13], and several known carcinogens have been identified as constituents of cooking emissions[14]. The cooking method and conditions are not exclusive to Taiwan or Asia. Additionally, in a risk assessment of inhalation exposure to trace elements when cooking in under-ventilated spaces, estimated carcinogenic and non-carcinogenic risks were higher than safe levels for most elements considered[15]. However, concern has been raised about relating health effects directly with the diameter of particles alone, since $PM_{2.5}$ is a complex mixture of particles with a range of characteristics including size, water solubility, chemical composition, and metal content[16].

Large scale *in situ* monitoring is often invasive, and cost and time prohibitive. An alternative is to model stocks of dwellings to estimate exposures at the population scale and to predict the impacts of interventions. Model predictions are sensitive to the emission rates used[17], and reported emissions from cooking are highly variable, even for the same cooking methods and ingredients; for example, reported emission rates for toasting vary between 0.11 mg/min[8] and 9.5 mg/min[18]. Accordingly, stock models must account for the uncertainty in emission rates in their predicted concentrations, and modelling frameworks have been developed to do this[17].

There are five main factors that affect cooking emissions. Firstly, the cooking method has a clear effect on the emission rate. Dry, water based, and oil based cooking processes all have very different emission rates, and oil based methods, such as frying, have the highest[19]. Similarly, burned food, grilling/broiling, and frying are found to have the highest mean emission rates[8,20,21]. Higher emission rates are found from stir frying than pan frying, attributable to higher temperatures[22]. Higher particle numbers and mass concentrations at higher cooking temperatures are found by some[23] but not all; [24] maybe because the oil *smoke point*, the temperature at which the oil visibly smokes, was not reached.

Secondly, there is evidence that ingredients influence $PM_{2.5}$ emissions, and oil type is perhaps the most significant[23,25]. The oil smoke point is important, but so are the composition and water content[19,26,27]. Emission rates from the heating of different cooking oils have been found to vary[26,28]. Corn, coconut, and olive oils are found to have higher emission rates than from soybean, safflower, canola, and peanut oils[26]. This difference was mostly related to the smoke point of the oils, except that olive oil generated $PM_{2.5}$ at the same temperature as corn and coconut oils despite having a higher smoke point. In contrast, corn and soybean oils are found to have a lower emission rate than rapeseed and sunflower oils[28].

Thirdly, the effects of non-essential additives, such as seasonings, on emission rates when heating oil have been investigated[29]. In controlled laboratory tests, the addition of sea and table salt to canola oil reduced the $PM_{2.5}$ emission rate. A similar test reduced emission rates by 56% when salt was added to corn oil[30].

Fourthly, the food type is important. A positive correlation is found between the fat content of foods and their emission rate[23,25,31]. Additionally, the water content of foods could impact particle size distribution when grilling ground beef[32].

Finally, there is some evidence that the cooking equipment used is influential. For example, adsorbed organic matter on the surface of pans may generate particulate matter when heated[19]. Heating an empty pan is found to emit ultra-fine particles (UFP) but not $PM_{2.5}$[33]. Additionally, higher emission rates are reported when using gas burners rather than electric hobs (also known as a cooktop)[19,34], but not by others[23], possibly due to the confounding effect of hob temperature.

There are numerous studies investigating cooking emissions and influencing factors by the cooking of single ingredients. Examples include toasted bread, 9.5±10.8 mg/min, fried chicken breast, 15.2 mg/min, and deep fried French fries, 0.34±0.03 mg/min[18]. However, the cooking of individual components, rather than full meals, may not be representative of typical home meal preparation. The addition of ingredients at various stages of the cooking process may affect $PM_{2.5}$ emission rates; for example, by amending cooking temperatures. However, the need to protect public health is arguably more important than proving this hypothesis, and so there is a need to investigate the effects of repetitively cooking multiple food types concurrently in a controlled and systematic manner, because it has not been done before and the data can be used to inform policy.

It is also important to consider the impact of mitigation strategies on emission rates. It is unreasonable to expect cooking to be removed from dwellings *en masse*. Instead, emitted pollutants need to be extracted at their source using a cooker hood (also known as a *range*, *exhaust*, or *extractor* hood). This device reduces exposure risks by capturing emitted pollutants before they mix with kitchen air. Ventilation requirements for kitchens vary around the world. In the UK, kitchens in new dwellings are required to have an intermittent extract rate of 60 l/s, or 30 l/s through a cooker hood, but there is no requirement to modify the ventilation strategy in existing dwellings[35]. Whereas in the Netherlands, a kitchen ventilation rate of 21 l/s is required in new dwellings[36]. In comparison, ASHRAE Standard 62.2 recommends 50 l/s and 150 l/s with and without a cooker hood[37], respectively. Mullen *et al.*[38] found lower concentrations of CO, NO and NO_x in households that reported using kitchen ventilation than those that did not, and a simulation of Southern Californian dwellings predicted that using cooker hoods in all dwellings increased the percentage of homes meeting air quality standards[39].

The ability of a cooker hood to capture particles is indicated by its capture efficiency (CE). Singer *et al.*[40] defined CE as the percentage of emitted particles that are extracted before they mix with room air,

whereas Lunden *et al.*[22] defined CE as the percentage of emitted particles that are extracted either directly or during operation. A cooker hood's CE is a function of the airflow rate through the hood, physical features that entrain pollutants towards the device, the installation height and capture volume, and the burner location and coverage by the hood[40]. Lunden *et al*.[22] found particle CEs of 4-39% for stir frying on the front burner, and 70-99% for the back burner. Singer *et al*.[40] reported similar findings. Lunden *et al.*[22] also found that CEs for particulates were different to those for gases, and that this difference varied between hoods from different manufacturers.

This paper aims to advance the understanding of $PM_{2.5}$ emissions that occur when cooking full main meals. It does this by cooking four hot meals commonly prepared in Northern European countries under controlled conditions using a kitchen laboratory. The cooking methods are varied to help identify parameters that affect emissions, and the cooker hood is explored as an exposure mitigation measure.

2 METHODS AND MATERIALS

2.1 Laboratory Facilities

All of the experiments were performed under controlled ventilation conditions in a test chamber with a depth of 3.65 m, width of 2.66 m, height of 2.68 m, and volume of V = 26.02±0.08 m^3. The chamber layout, including its dimensions, the placement of the cooker hood and the size of cabinets, is comparable to the EN 61591[341] standard for kitchen test facilities. The only addition was the use of a ceiling diffuser to supply air, intended to simultaneously enhance the mixing of the space and minimize stratification and disturbances of the airflow under the cooker hood[42]. Supply air was delivered by an HVAC unit equipped with an AFPRO F7 filter. To minimize uncontrolled infiltration and exfiltration, the supply air flowrate was adjusted so that the pressure difference between the test chamber and its surroundings was less than 0.5 Pa when measured by a Halstrup-Walcher EMA 84 digital pressure gauge.

Tests were performed for two ventilation scenarios that varied the flow rate and location of the ventilation extract. The first is a *low* ventilation scenario with an airflow rate of 21 l/s (75 m^3/h), the minimum required in domestic kitchens by the Netherlands Building Regulations[36]. It was achieved using a single extract grille located in the middle of the chamber 94 cm below the ceiling; see Position

A in Figure 1. The second is a *high* ventilation scenario achieved using a cooker hood with a flow rate of 83 l/s (300 m^3/h). The hood's airflow rate was controlled by a centrifugal fan located outside the chamber and set by measuring the pressure drop over an orifice plate following EN ISO 5167-2[43]. The main tests were conducted using the low ventilation scenario (see Sections 2.3.1-2.3.3) but the high ventilation scenario was used to support additional analyses (see Section 2.3.4).

Full mixing conditions were assumed. An SMC TR16 fan was used during the low ventilation scenario to enhance air mixing, but not for the *high* scenario to avoid reducing the capture efficiency of the cooker hood by disturbing the airflow under it; see Section 3.5.

A Pelgrim GK 564 gas stove with four burners was used as a heat source; see Figure 1. The maximum flowrates, $\dot{Q}$ (l/min), and power, H (kW), per burner were $\dot{Q}$ = 3.2 l/min and H = 1.7 kW for the back left burner, $\dot{Q}$ = 4.5 l/min and H = 2.4 kW for the back-right burner, $\dot{Q}$ = 2.5 l/min and H = 1.3 kW for the front-left burner, and $\dot{Q}$ = 1.9 l/min and H = 1.0 kW for the front-right burner.

2.1.1 Cooker Hood

A ducted cooker hood is a ventilation device located immediately above a stove that aims to capture and remove contaminants emitted by combustion and cooking before they mix in a space. It should cover the front-burners and contain a *damp-buffer* (where the sides that frame the hood protrude below its central horizontal plate) to ensure high CEs[40,44]. A consumer standard ATAG WS9011QAM ducted cooker hood was selected to meet the criteria of the high ventilation scenario and installed 70 cm above the kitchen counter (see Figure 1), which approximately agrees with Singer *et al.*[40] and Lunden *et al*[22]. The cooker hood is 90 cm wide and 53.5 cm deep, and is equipped with a small damp-buffer of height 3 cm (see Figure S1). The exhaust hood extended over the front burners whose centres were 40 cm from the wall. When frying pans were used on the front burners, the smaller pan (Ø=24 cm) was completely covered by the hood, whereas the larger pan (Ø=28 cm) protruded by 0.5 cm.

2.2 $PM_{2.5}$ Measurement Equipment

The $PM_{2.5}$ concentration and particle size distribution were measured in the test chamber using a Grimm 11-R Mini Laser Aerosol Spectrometer optical particle counter (OPC), factory calibrated using dolomite dust. It measures particles with diameters between 0.25-32 µm and classifies them into 31 size bins. It

detects concentrations between 0.1 µg/m^3 and 100 mg/m^3, and identifies particle counts of up to 2 million particles per litre, at a sampling frequency of 6 seconds. The OPC was placed below the extract grille at 1.25 m above the floor, at point A in Figure 1. There are number of known issues with using OPCs to measure $PM_{2.5}$[60] and the consequences are discussed in Section 3.

OPCs pass an aerosol through a laser beam to measure the degree of light scattering, which varies according to the mass concentration, size, shape, and composition of its particles[45]. They are calibrated using test dust with known properties, commonly solid, spherical, and non-absorptive polystyrene latex spheres with a defined distribution of diameters[46]. If the physical or optical properties of the measured particles differ from those of the test dust, the mass concentrations reported by the OPC must be corrected[18]. The correction is made by multiplying a measured concentration by a calibration factor[45]. Additionally, the OPC may also underestimate the concentrations if a significant proportion of the emitted particles are smaller than the lower detection limit of the device[61]. Accordingly, concurrent gravimetric sampling (GS) was used to determine the true mass concentrations using filter-based GS devices for each test meal. Air was drawn through a TECORA fine air inlet, a low volume sampler head, and a glass-fibre 47 mm filter at 2.3 m^3/h using Gilian Aircon2 electric pumps for a defined period of time, following EN12341[47]. The volumetric flow rates were checked with a calibrated Yokogawa RAGL rotameter before and after each collection period. Each filter was weighed before and after a test in accordance with MDHS 14/4[48], and so the mass increase, flow rate, and measurement time are used to calculate the mean concentration. The calibration factor is the mean average of the ratios of the GS and OPC mean concentrations. Three GS devices were placed at the same height as the OPC and 0.5 m to the left, right, and rear of it; see Positions B-D in Figure 1.

2.3 Test Meals and Cooking Methods

To derive a typical portion size, we used data reported for the Dutch National Food Consumption Survey (DNFCS)[49], a periodic survey of food consumed by the Dutch population. In the DNFCS, data is weighted and aggregated by age and sex, where each group is designed to be representative by age, region of the country, degree of urbanisation, and education level. We used a portion size in line with those reported by the DNFCS for the 31-50 age group for men (n=348) and women (n=351) because it

covers the majority of the adult Dutch population and thus indicates typical adult portion sizes. Four meals were chosen comprising carbohydrates, vegetables, and meat based protein sources, because the DNFC indicates that <7% of the population has special eating habits that include vegetarianism. The proportions of meat, potatoes and vegetables were guided by the median mass eaten per consumption day by men and women aged 31-50 years. This data was used to formulate meal types that could be cooked using a stove with a high degree of repeatability.

2.3.1 Test Meal Descriptions

The ingredients of the four meals are given in Table 1, and were selected because they are typically Dutch and broadly European. Meal 1 is a reference meal whose emissions may be compared to the others. The ingredients for Meals 1 and 2 were informed by the DNFCS[49] and the types of meat are consistent with common Western cooking ingredients[14]. The mass of each solid ingredient and volume of each liquid were constant and estimated for a median man and woman. All ingredients were supermarket brand basic ingredients except for the *straight to wok* noodles, which were branded. Fresh ingredients were refrigerated at 4ºC before cooking whereas the canned and dry ingredients, oil and additives were all stored at room temperature. Solid ingredients were weighed using a Zhongshan Camry EK9210K electronic kitchen scale and the salt was weighed with a Mettler AM50 microbalance. Liquid ingredients were measured using a 25 ml measuring cylinder, and 250 ml and 1000 ml beakers.

2.3.2 Cooking Equipment

A cooking protocol was developed based on gas flow rates and cooking time, following Lunden *et al*.[22]. The gas flow rate was controlled with the cooker controls and monitored using 2 parallel Bronkhorst F-201EV mass flow controllers. It was displayed in real-time, summed, and adjusted to meet required flow rates. Pan temperatures were identified during preliminary tests using a ThermaCAM™ SC640 FLIR thermal camera following Buonanno *et al.*[23]. The time taken to reach a required temperature and the gas flow rate were noted. Thereafter, only the gas flow rate and the time were used to control pan temperature. Four pans were used: (i) a TEFAL Titanium Pro 28 cm non-stick stir-fry pan; (ii) a TEFAL Talent Pro 24 cm non-stick frying; (iii) a BK Conical Glass stainless steel 2 litre saucepan; and (iv) a

BK Conical Glass stainless steel 1.5 litre saucepan. Hereon, the frying pans are denoted by their diameter and the saucepans by their volume. All saucepans were covered by a lid during cooking.

Before each test, the pans and cooking materials were cleaned in warm water with standard dishwashing soap, rinsed with tap water, and dried. At the end of each cooking period all burners were turned off and a lid was placed on any frying pan to prevent continued emissions, and to give a clear end to the test. The $PM_{2.5}$ concentrations in the test chamber were monitored for a further 30 minutes. Between tests, the chamber was purged of $PM_{2.5}$ by increasing the exhaust ventilation rate and opening the door to the laboratory until concentrations returned to background levels of <1 $\mu g/m^3$.

To investigate the repeatability of emission rates during the cooking period, each meal was cooked 6 times using the low ventilation scenario; see Table 2. Gravimetric measurements were made during the final repetition to determine calibration factors; see Section 2.2.

2.3.3 Cooking Instructions

The steps required to cook each meal are described here but the ingredient measures are only given in Table 1 for brevity. Meal 1 begins at minute 0 by heating olive oil in a 28 cm frying pan located on the front-left burner with a gas flow rate of 2.5±0.1 l/min. At minute 3, when the pan reaches approximately 160°C, the chicken is added and at minute 4 the gas flow is reduced to 1.3±0.1 l/min. At minute 8 the back left burner is ignited and a 2 litre saucepan containing the water and beans is placed over it, giving a total gas flow rate of 4.2±0.1 l/min. At minute 10 the front-right burner is ignited and a 24 cm frying pan containing olive oil is placed over it, giving a total gas flow rate of 5.3±0.2 l/min. At minute 13 the sliced potatoes are added to the 24 cm frying pan and all ingredients are cooked for a further 15 minutes until the test ends at minute 28. Throughout the cooking period the chicken is turned every 5 minutes and the potatoes stirred for 30 seconds at 3 minute intervals.

Meal 2 follows the method of Meal 1 with one main exception: the potatoes are boiled in water instead of fried. At minute 8 the back-left burner is ignited, giving a total gas flow rate of 4.2±0.1 l/min, and a 1.5 litre saucepan containing the potatoes and water is placed over it. At minute 13 the front-right burner is ignited to boil the French beans, giving a total gas flow 5.3±0.2 l/min. In this test, the potatoes are not

stirred during cooking. The test ended at minute 28, therefore the beans were cooked for less time than in Meal 1, but were cooked by the end of the test.

Meal 3 begins at minute 0, by heating olive oil in the 28 cm frying pan over the rear-right burner with a gas flow rate of at 4.5±0.1 l/min. At minute 3 the bacon is added and stirred constantly. At minute 7 the gas flow rate is reduced and the onion and garlic added to the bacon. Simultaneously, the back-left burner is ignited under a 2 litre saucepan containing water for the pasta, giving a total gas flow rate of 4.1±0.1 l/min. At minute 9 the minced beef is added and the gas flow rate increased to 4.6±0.1 l/min. At minute 13 the tinned tomatoes are added to the mince, mixed thoroughly, and stirred thereafter at 3 minute intervals until the sauce has simmered for 15 minutes in total. At minute 18 the rear-right burner is reduced giving a total gas flow rate of 3.9±0.1 l/min, and the pasta is added to the boiling water and cooked for 10 minutes. At minute 21 the back-left burner is reduced giving a total gas flow rate of 1.9±0.1 l/min. The test ends at minute 28.

Meal 4 only uses the 28 cm frying pan located over the rear-right burner with an initial gas flow rate of 4.5±0.1 l/min. After heating the olive oil, at minute 3 the diced chicken is added, turned at minute 4, and the burner reduced to 1.1±0.1 l/min at minute 5. At minute 8 the chicken is removed from the pan, the total gas flow rate is increased to 4.4±0.1 l/min, and the olive oil is added. At minute 10 the vegetables are added, spread thinly, and stirred continuously. At minute 15 the gas flow rate is reduced to 1.1±0.1 l/min, and the chicken and noodles added to the pan and cooked until the test ends at minute 17.

2.3.4 Additional Tests

Four additional sets of tests were conducted to investigate specific areas of interest, which are reported in Section 3.4. Firstly, to investigate $PM_{2.5}$ emission rates from the gas stove, two *blank* tests were conducted; see Table 2. These followed the gas flows and timings from the reference meal (see Section 2.3.3) for the low ventilation scenario (see Section 2.1), but neither was food cooked nor pans heated.

Secondly, three tests investigated factors that the literature indicates may affect $PM_{2.5}$ emissions during cooking; see Table 2. The reference meal was used with three separate substitutions: (i) any olive oil used for frying was substituted with *Croma* brand "Bakken en Braden" liquid margarine to investigate the findings of Torkmahalleh *et al.*[26] who found that particle emissions varied with oil type; (ii) the non-

stick frying pans were replaced by stainless steel pans; and (iii) the chicken was seasoned with 1 g of salt before frying, to investigate whether the findings from Torkmahalleh *et al.*[29] could be applied to realistic cooking methods. Here, Torkmahalleh *et al.* found that adding salt to oil before heating reduced particle emissions under controlled laboratory conditions.

Thirdly, to investigate the reduction potential of extracting the $PM_{2.5}$ at source, the 4 test meals were prepared using the high ventilation scenario with air solely extracted through the cooker hood.

Finally, Lunden *et al.*[22] found higher particle CEs when frying on the back burners of a stove and so this was investigated using the reference meal with the frying pans relocated to the back burners; see Table 2.

2.4 Data Processing and Statistical Analysis

The measurements of $PM_{2.5}$ concentration over time for each test are used to compute a source strength and an emission rate. Several methods of calculating an emission rate are described in the literature that take the *mass balance* model of Ott *et al*.[50] as their basis, but vary by their assumptions about the test conditions or the emission characteristics[50]. The most common method assumes a constant emission rate and either uses the measured concentration at the end of the emission period[20], or calculates the theoretical peak concentration that should occur at the end of that period in a perfectly mixed space[18], to determine the emission rate. Variations of the *peak estimation* method have been used by Dacunto *et al*.[18], He *et al.*[8], Jiang *et al.*[51], Lee *et al.*[52], and Olson and Burke[20].

Trials indicated that the emission rate is not constant during the cooking period. Therefore, an alternative method that assumes a variable emission rate is used to determine an average emission rate for the cooking period. It uses the principle that the *area-under-the-curve* of a plot of concentration over time, t (s), is equivalent to the total mass emitted, the source strength, G (µg). The source strength is then divided by the emission period to give an average emission rate[50]. Here, the cooking time and emission period, T (s), are considered identical. The average emission rate, $\overline{g(T)}$ (µg/s), is given by

$$\overline{g(T)} = \Phi V \overline{C(T)} + \frac{VC(T)}{T} \quad (1)$$

where Φ (s^{-1}) is the total decay rate (the sum of ventilation, deposition, agglomeration, and evaporation rates), V (m^3) is the test chamber volume, $\overline{C(T)}$ (µg/m^3) is the average concentration over the emission period, and $C(T)$ (µg/m^3) is the concentration at the end of the emission period. Then, $G = \overline{g(T)}\,T$. Generally, $\overline{g(T)}$ is reported in mg/min and G in mg. The model is based on assumptions of air homogeneity, zone isolation, and perfect mixing[53].

All parameters are known or obtained from measurements, except for Φ, which is determined from the log-linear regression of concentrations measured during a 30 minute decay period immediately after the emission period; see Section 2.3.2 and Dacunto *et al.*[18]. The precision of estimates in Φ and the assumptions of the model are determined from the regression where a coefficient of determination (R^2) indicates the proportion of the variance in Φ that is predictable from the measurements of concentration over time. The standard error (α) in Φ describes uncertainty in its value. The propagated error in $\overline{g(T)}$ is the root of the sum of the squares of uncertainty in each parameter, obtained by perturbing each one by its standard error, following Hughes and Hase[54]. The calculation of Φ, $\overline{g(T)}$, and G, and the uncertainty in them, was made using bespoke MATLAB code[55]. The resulting emission rates were compared using a single factor ANOVA with a 5% significance threshold, and two sample t-tests with Bonferroni correction, which was used to reduce the probability of a Type 1 error. Here, the 5% significance level is divided by the number of tests to give a revised significance of 0.83%. This analysis was conducted using Excel.

By removing contaminants before they are allowed to mix in a space, a cooker hood has the effect of reducing $\overline{g(T)}$ to give a *net* emission rate. To estimate the potential of the cooker hood to do this, $\overline{g(T)}$ was calculated for each meal (see Section 2.3.1) under low and high ventilation conditions and compared to give a percentage reduction in $\overline{g(T)}$ for each meal.

3 RESULTS AND DISCUSSION

$PM_{2.5}$ concentrations were measured over time following the methods given in Section 2 for each of the n=4 meals described in Table 1. This section presents results and discusses the scaling of the data, the emission characteristics for each meal, meal emission rates, confounding factors, the effectiveness of the cooker hood, and further emissions from the gas burners.

3.1 Calibration Factors

In order to interpret the measurements of the OPC, a mean calibration factor, $\bar{C}_n$, for each meal was calculated following the method described in Section 2.1, and these have been used to scale the mass measurements of the OPC described hereon. There is a marked variation by meal: $\bar{C}_1$=3.9±0.15; $\bar{C}_2$=5.0±0.096; $\bar{C}_3$=2.7±0.039; and $\bar{C}_4$=1.5±0.045. Full results from the gravimetric sampling tests can be found in Table S1. Gravimetric samples were only collected during a single repetition of each meal, with samples collected on filters in 3 locations within the test chamber. Therefore, the uncertainty in the calibration factors does not account for the variation between repetitions, which may be larger. The variation between meals indicates the composition and optical properties of the emitted $PM_{2.5}$, and proportion of particles below the detection limit of the OPC, varied between meals. They also show that the OPC consistently underestimates particle mass when cooking meals, which agrees with Wang *et al.*[45] whose OPCs were used to measure $PM_{2.5}$ in houses. Wang suggests that this is caused by coincidence losses, deviations in the refractive coefficient, or the presence of high concentrations of particles that are smaller than the OPC's detection limit. Each filter was removed from its transport cassette and equalized in a climate chamber for over 4 hours prior to its second weighing. Trapped aqueous aerosols are likely to have evaporated and so are a source of calibration factor bias because the OPC is known to detect them. These calibration factors are higher than others found in literature[18,51]. However, the calibration factors given by Dacunto *et al.*[18] and Jiang *et al.*[51] are not determined using a Grimm calibrated with Dolomite dust and so they are not directly comparable to those given here.

The highest calibration factor was found for Meal 2 whose particle size distribution (see Figure 4a) indicates that it emitted the highest proportion of small particles, and so this supports the theory that the deviation might be partially caused by small particles whose diameters are less than those detected by the Grimm. The differences in $\bar{C}_n$ could also be attributable to changes in the particle composition. Some other affecting factors are discussed in Section 3.4.

3.2 Emission Characteristics

Figure 2 shows $PM_{2.5}$ concentrations measured in the test chamber over time during, and after, the cooking period. Consistent gradients show steady emission rates and steeper gradients correspond to

higher emission rates. Although the concentrations were logged at 6 second intervals (see Section 2.2), they are smoothed here over 1 minute intervals for illustrative purposes. An additional figure in the supplementary information indicates key moments in the cooking process. All meals show repeatable temporal trends in $PM_{2.5}$ concentration, although there is considerable variance in the magnitude of the concentrations between tests. Cooking is a complex process, and although the process and ingredients were standardized between repetitions, some level of variation would be expected. In particular, although ingredients were purchased from the same location, their exact composition was not tested.

When cooking Meals 1 and 2 (chicken, beans and fried or boiled potatoes, respectively), the $PM_{2.5}$ concentrations initially increase steadily for about 20-25 minutes, and then increase more rapidly for the remainder of the cooking period. It is not immediately clear what caused this change, but it is possible that over time, the frying pan temperature increased and moisture was removed from the fried ingredients leading to Maillard browning. Additionally, it may be caused by particles below the Grimm's detection limit coagulating over time until they reach a detectable size. The consistent changes in gradient exhibited by Figures 2a and 2b show that Meals 1 and 2 share similar emission characteristics, and this is to be expected given their shared ingredients and similar cooking processes; see Table 1. However, Figures 2c and 2d show that Meals 3 and 4 are distinctly different.

During the cooking of Meal 3 (*pasta bolognaise*), the $PM_{2.5}$ concentration initially increases rapidly, correlating with high temperature frying. The changes between minutes 7 and 13 correspond to the adding of ingredients and changes in gas flowrates. The $PM_{2.5}$ emission rate appears to reduce substantially after the early peak, after the onions and garlic are added and the gas flow turned down. During the final repetition of Meal 3, concentrations were notably higher, as they appeared to increase for longer. The gas supply rate was identical the other repetitions and so the reason for this variation is not clear, but may be related to a variations in the ingredients.

Meal 4 (*stir-fry*) also exhibits a high-low-high emission pattern and two distinct peaks. The first peak occurs when frying the chicken, the second when frying the vegetables, and the reduction in concentration occurs immediately after the chicken is removed from the 28 cm frying pan, and prior to the addition of vegetables; see Section 2.3.3.

3.3 Emission Rates

Figure 2f and Table 3 show that the mean $PM_{2.5}$ emission rates ($\overline{g(T)}$) and source strengths (G) measured for the four meals described in Section 2.3.1 vary between 0.54-3.7 mg/min and 15-68 mg with 95% confidence, respectively. Estimated decay rates (see Table S2) ranged from 4.7±0.041 h^{-1} to 6.1±0.042 h^{-1}, although it should be noted that they have little physical meaning in this context because they are a function of particle deposition, agglomeration, evaporation, and other processes, and because the mixed volume of air may not equal the room volume. The volume term in Equation 1 is an important source of bias because the room volume we apply (see Section 2.1) includes cupboards (10% of the room volume), people, and equipment, and assumes that the $PM_{2.5}$ is equally mixed within all of these entities. However, is impossible to determine the validity of this assumption with the measurements made and so it is possible that the bias in $\overline{g(T)}$ and G could be up to 10% of their values. Volume bias is rarely considered; for example, the emission rates reported in Section 1[8,11,18,21] all give a volume but do not describe its calculation. Accordingly, future measurements of emission rates and G should seek to minimize the difference between mixed and space volumes. The coefficients of determination for the decay rate were $R^2 >$0.97 for Meals 1-4 indicating excellent mixing; see Section 2.4 and Sherman[53].

Similar variance in emission rate has been found by Fortmann *et al.*[21] who measured 2.92 mg/min when stir-frying using an electrically heated hob and 3.36 mg/min and 1.54 mg/min when stir-frying (2 tests) on a gas burner, which are comparable to the mean $\overline{g(T)} = 3.2\pm0.24$ mg/min for Meal 4. Fortmann *et al.* also cooked a full meal using an oven and cooktop and measured the emission rate to be 2.45 mg/min, which lies between the mean emission rates of Meals 3 and 4. Dacunto *et al.*[18] cooked three single dishes on an electric hot plate and in an aluminium frying pan. First, they measured an emission rate of 0.4 mg/min and $G = 5.7$ mg for chicken, vegetables, and soy sauce stir fried in olive oil (2 tests), which are less than those for all meals cooked here. However, when they pan fried chicken drumsticks or thighs (on the bone with skin on) for 16-28 minutes (6 repeats) a significant increase was found where the emission rate was 2.5±0.9 mg/min and G was 62.2±16.9 mg. Their emission rate lies between the means for Meals 3 and 4 whereas their G is greater than those for our meals and indicates that they cooked for longer. When Dacunto *el al.* pan fried chicken breast in olive oil so that it was 25-50% charred, $\overline{g(T)}$ =15.2 mg/min and $G = 289$ mg. These high values are most likely caused by the charring, whereas

our meals only experienced Maillard browning. He *et al.*[8] derived emission rates for a range of cooking events, including complex and simple meals, from measured $PM_{2.5}$ mass concentrations over a 48 hour period in 15 domestic kitchens using an OPC without a calibration factor (see Section 2.2). General cooking (37 events) gave a median $\overline{g(T)}$ of 0.11 mg/min ($\sigma = 0.99$ mg/min), frying (4 events) gave a median 2.68 mg/min ($\sigma = 2.18$ mg/min), and stove cooking gave a median of 0.24 mg/min ($\sigma = 1.29$ mg/min). The lack of control over ventilation rates and emission periods, and the lack of a calibration factor, mean there are large uncertainties in these values, but the frying broadly agrees with the emission rates from the meals cooked here whereas the *cooking* and *stove* events are much lower. Nevertheless, it is reassuring that the values of $\overline{g(T)}$ and G given in Table 3 appear plausible given the context provided here.

Our values of G and $\overline{g(T)}$ can also be compared to those derived from *in situ* measurements of multiple household activities that include cooking to give a broader understanding of their significance. Chan *et al.*[11] calculated emission rates for 836 cooking and non-cooking events in 18 dwellings in California, finding a mean source strength and emission rate of 30 mg and 1.72 mg/min, respectively, which are broadly similar to ours. Dacunto *et al.*[18] identified source strengths and emission rates of 1.4 mg and 0.1 mg/min for oven cooked frozen pizza, 72.5 mg and 9.5 mg/min for toasting of bread (90-95% charred), 18.3 mg and 1.6 mg/min for fried salmon, 24.3 mg and 2.1 mg/min for fried pork chop, 19.9 mg and 3.8 mg/min for cigarette smoking, 16.9 mg and 1.3 mg/min for the burning of stick incense, and 215.4 mg and 16.4 mg/min for an open fire. For an equivalent release period, the $\overline{g(T)}$ for Meals 1-4 are greater than those Dacunto *et al.* found for cooking pizza, less than cigarette smoking, and substantially less than an open fire.

Table 3 shows that the particle counts exceeded 2×10^6 particles/litre in 10 of the 24 tests. Here, the Grimm may experience *coincidence* errors where multiple particle may be seen as one larger particle. One might expect coincidence errors to affect the regression analysis, yet the values of R^2 are close to unity and $\alpha < 1\%$ of Φ. For a further discussion of coincidence errors see Section 3.6.

The emission rates in Table 3 have been calculated using an *area-under-the-curve* method that assumes a variable emission rate; see Section 2.4 for a justification. However, several other studies[8,11,18] apply

the *theoretical peak estimation* (also known as the *phantom curve*) method that assumes a constant emission rate, which Figure 2 shows to be untrue when cooking meals. For a direct comparison this method was applied to our data and the constant emission rate was estimated to be 20–56% higher than $\overline{g(T)}$ for Meal 1, up to 70% lower for Meal 3, and between 0–12% lower for Meal 4. Clearly, there are significant and non-uniform differences between the two methods. However, the *area-under-the-curve* method is appropriate in this context because it is exact, and also general because it makes no assumptions about the change in the emission rate over time[50]. Accordingly, we argue that it is appropriate to apply it to the cooking of meals and that it should be used in future studies of this type for accuracy and to ensure a fair comparison between tests.

3.4 Factors Affecting Emission Rates

The emission rates given in Table 3 for each meal further highlight the general repeatability of the tests, which is encouraging given the number of unknown or uncontrollable factors involved in the cooking of foods. Table 3 also shows there are differences between meals, even when they have similar ingredients. For example, Meals 1 and 2 differ only by the cooking method applied to their potatoes; those in Meal 1 are fried, whereas those in Meal 2 are boiled. Here, Meal 1 has the lowest emission rates even though frying is known to be a strong source of $PM_{2.5}$ emissions[19]. Boiling the potatoes emits water vapour, and increased humidity is known to affect the performance of light scattering measurement devices[56]. Additionally, high relative humidity (RH) has been linked to the hygroscopic growth of particles[56,57], and it is possible that the boiling of potatoes created aqueous aerosols that were counted by the Grimm. Measurements of RH may have indicated any measurement errors and this remains a confounding factor. Furthermore, frying is responsible for the emission of ultra-fine particles, whose diameters are below the detection capability of the Grimm[14,23]. A single factor ANOVA indicates the mean emission rate for all meals is not the same ($p < 0.05$). Multiple two sample t-tests with Bonferroni correction (see Section 2.4) indicate that the emission rates of Meals 1 and 2 are not significantly different ($p > 0.0083$). This suggests that the emissions from frying could have less influence on the overall emission rate when cooking meals or there are other experimental explanations that were not measured, such as pan temperature. These tests also suggest that the emission rates from Meals 1 and 3

476 are significantly different, and that Meal 4 emissions differ from all others ($p < 0.0083$), but those for
477 Meal 2 and 3 are not significantly different ($p > 0.0083$).

478 Meal 1 was varied from the base case (see Section 2.3.4) in three ways: (i) using liquid margarine instead
479 of olive oil; (ii) using a stainless-steel pan instead of a non-stick pan, and (iii) by adding salt. Figure 3
480 and Table 4 show that frying in a stainless steel pan had an immediately obvious effect on the emission
481 rates, with the mean emission rate increasing by 940%. Given that the same volume of oil and mass of
482 the ingredients were used for all tests, the higher emission rates may be a function of the thermal
483 conductivity of the pans, their surface temperatures, and the adhesion between the food and the pan.
484 Here, both the chicken and the potatoes were observed sticking to the stainless steel pan in some tests
485 and the surface of the pan charred, which could have been reduced by adding more oil, itself a known
486 source of $PM_{2.5}$. This suggests that using a non-stick pan can minimize $PM_{2.5}$ emission during frying.
487 The new stainless steel pan produced the highest emission rate, which then decreased with each
488 subsequent repetition. This indicates there may be an aging effect that is a function of the changing
489 properties of the pan's surface, which may have continued with further repetitions.

490 The tests with the liquid margarine and with salt show an increase in the mean emission rate of 11% and
491 47%, respectively, when compared to the reference meal. A *t*-test suggests these changes are non-
492 significant ($p > 0.05$). However, these small differences cannot be ruled out and may be detected by
493 further tests. These results disagree with the findings of Torkmahalleh *et al.*[27,29] for salt. Torkmahalleh
494 *et al.*[27,29] added salt to oil before heating, whereas in these tests the salt was added to the chicken before
495 frying. A better comparison may be found with Torkmahalleh *et al.*[33] who found higher particle
496 emissions for grilled salted meat than unsalted meat. Our tests found a small increase in emissions,
497 similar to Torkmahalleh *et al.*[33], however it is not statistically significant. Additionally, the change in
498 emission rate when margarine is used is also not statistically significant, despite previous suggestions
499 that the oil type is a significant factor[17].

500 Custom calibration factors were not obtained for each of these variations. In the absence of data, $\bar{C}_1$=3.9
501 was used for all three variations, which is a limitation and source of uncertainty.

3.5 Cooker Hood Capture Efficiency

The 4 meals were prepared whilst using an extracting cooker hood located immediately over the burners (see Sections 2.3.4 and 2.4). Table 5 gives their emission rates and reductions in their means when compared to those given for the main tests in Table 3. The percentage reductions in Table 5 are equivalent to CEs defined by Lunden *et al.*[22]; see Section 1. The reductions are >90% for all four meals. Additionally, Meal 1 was prepared with the fried components cooked on the back burners, closest to the wall. This resulted in slightly higher reductions that, when tested using a two-sample *t*-test ($p > 0.05$), are statistically non-significant. This is surprising because Lunden *et al.*[22] found particle CEs of 4-39% and 70-99% for stir-frying on front and back burners, respectively. Singer *et al.*[40] tested 15 different hoods and reached the same conclusion. In these studies, the coverage of the front burner by the hoods was variable. For the two hoods with better burner coverage, Lunden *et al.*[22] measured particle CEs 60-80% and <60%, for stir frying on the front burner, compared to CEs close to 100% for frying on the rear burner. Of the 15 hoods studied by Singer *et al.*[40], two had coverage >75% during all tests, suggesting good coverage of the front burners. CEs for these hoods measured between 75-100%. The exact coverage provided by the hoods was not reported in either study, and both found CEs lower than those measured here, even at higher exhaust flow rates. It is possible that the high emission reduction found here is explained by the good coverage of the front burners by the hood (see Section 2.1.1) and by the presence of a damp-buffer[44]. The small volume of the test chamber encouraged a closed-loop airflow pattern that allowed particulates to be captured by the hood long after they were emitted having circulated around the chamber, and so a CE determined in this way is biased and under-estimates occupant exposure risk. These inherent biases in the technique can be overcome by cancelling out the impact of room concentrations. In theory, one way to do this is to increase the chamber volume and ventilate the chamber far from the hood, or to conduct tests in a full-scale residence[62]; unfortunately this is impractical to do without inducing new systematic errors, such as poor mixing. A better way to do it is to use a steady-state capture efficiency test method[58] that ignores uncaptured pollutants. The method used here may indicate the actual performance of the cooker hood in homes with a kitchen with a similar volume to the test chamber, which are common in the English housing stock[63], but it may be less

indicative of performance in houses with larger kitchens or in open plan living spaces where air circulation does not occur.

Meals 3 and 4 both resulted in higher reductions than Meals 1 and 2. However, it was only possible to calculate emission rates for two of the five repetitions of Meal 3, because the chamber $PM_{2.5}$ concentrations were too low in other tests. In these tests, the log-linear regression indicated the concentration increased during the decay period, which may be due to incomplete mixing, and so these tests were discounted. The total decay rates (Φ) ranged from 0.076±0.04 $3h^{-1}$ to 5.6±0.15 h^{-1} (see Table S4). Here, R^2 values decreased substantially (R^2 <0.83 for accepted tests) and the α of the decay rate increased, indicating a decrease in the mixing quality when compared to the initial tests (see Section 3.2). These are limitations of the method, as the chamber concentrations are likely to be low when using a functioning cooker hood with a high capture-efficiency. Particles will have deposited on surfaces during all tests, but the cooker hood may have changed the velocity profile around the cooker and the deposition rates. The method used here accounts for this change by identifying how much the emission rate has effectively reduced, and so is measuring an *apparent* capture efficiency, rather than a true capture efficiency. This metric is useful because it can be used to estimate indoor $PM_{2.5}$ concentrations from a known source and used to inform regulations, but there is significant uncertainty in the measurements.

A standard method to derive capture efficiencies using ideal gases has been proposed[58]. And, although it does not fully account for particle behaviour, there is lower measurement uncertainty. Singer *et al.*[40] calculated their hood CEs by using CO_2 as a tracer and measuring its concentration in the exhaust duct. However, Lunden *et al.*[22] followed the method given in Section 2.3.4 arguing that particle losses in the hood and ductwork bias both the measured concentrations and the particle CE. This highlights issues with all methods of measuring cooker hood CE for different pollutants and is an area of ongoing work. The airflow rate through the cooker hood of 83 l/s is high when compared to the 50 l/s, 30 l/s, and 21 l/s required by ASHRAE, in the UK, and in the Netherlands, respectively. This flowrate is clearly effective but it had noise and energy penalties. The airflow rate was not varied but the relationship between it and the apparent capture efficiency may be non-linear, and so it could be possible to reduce it without affecting performance significantly. Further work is required.

3.6 Particle Size Distributions

The distribution of the optical diameter of particles varies over the cooking and decay periods. Figure 2 shows that the peak concentration occurs towards the end of the cooking and emission period for Meals 1 and 2. Therefore, the vast majority of particles discharged during the cooking period have already been emitted and so the peak concentration is chosen as a suitable moment to analyse the variance in particle diameter for all meals. Figure 4a shows the distribution of particle diameters between $0.25 - 10\ \mu m$ at the peak concentration time, averaged over the 6 tests for each meal type. It shows that they are similar for all four meals, with more particles emitted in the smaller size fractions. Figure 4a shows that when compared to Meal 1, Meal 2 emitted more particles in the smallest size fractions. Figure 2 shows that Meals 3 and 4 had higher peak concentrations than Meal 1 and 2, and Figure 4a confirms that they also had higher particle counts. The distribution for Meal 4 is weighted more towards the larger particle sizes. This agrees with previous findings that particle diameter increased at higher frying temperatures[14]. Table 3 indicate that the Grimm may have experienced coincidence errors in some repetitions of Meals 3 and 4. These increase uncertainty in Figure 4 where large size bins may be over populated and smaller bins under populated in affected meals.

The three variations in the preparation of the Meal 1 base case (see Sections 2.3.4 and 3.3) altered the emission rates and the particle size distributions; see Figure 4a and 4b, respectively. Using liquid margarine resulted in lower particle counts in the larger size fractions ($> 2.5\ \mu m$ in diameter). Frying in stainless steel resulted in higher emissions overall, but the particle size distribution is similar to the base case meal. With the addition of salt, the distribution is weighted towards the smaller size fractions ($< 2.5\ \mu m$).

Figure 4c illustrates the particle size distributions when the cooker hood was used. Cooking Meal 1 on the back burners rather than the front burners also changed the particle size distributions within the chamber. Table 5 shows the cooker hood captured a slightly greater mass of particles when cooking on the back burners. However, the differences between Figures 4a and 4c suggest that the cooker hood captures a greater number of smaller particles ($< 0.4\ \mu m$) when frying on the front burners. It is not clear why this occurs, but the particle size distributions are compared at a single moment in time and so

may change at other times. Also, more of the larger particles ($> 0.65\ \mu m$) are captured in Meal 3 than Meal 4, although it is not clear why this has occurred.

3.7 Gas Burners

$PM_{2.5}$ concentrations were measured during two *blank* tests (see Section 2.3.4) where the combustion elements of the Meal 1 preparation were followed (outlined in Section 2.3.3) for the low ventilation scenario, but no food was cooked. The concentrations are generally <1 $\mu g/m^3$, and so it was impossible to identify any decay at the end of the test once the stove burners were switched off. This shows that the emissions of $PM_{2.5}$ from the gas burners can be considered negligible, and so have not contributed to the temporal variation in $PM_{2.5}$ concentration shown in Figure 2 or the emission rates given in Table 3. However, gas burners are a known emitter of ultrafine particles and nitrogen oxides[21,23], and both pollutants are associated with negative health effects[39].

3.8 Impacts

The $PM_{2.5}$ source strengths and emission rates of the 4 meals suggest that cooking for a prolonged period in a house without adequate ventilation could lead to indoor $PM_{2.5}$ concentrations that exceed those found outside and could negatively affect the health of occupants; see the health risks discussed in Section 1. This is especially likely in airtight dwellings where ventilation may be inadequate and during the heating season when occupants may seek to reduce ventilation rates to minimize heating fuel costs. Section 3.5 shows that a cooker hood can be used to reduce the $PM_{2.5}$ emission rate during cooking, although it is not yet clear what combination of airflow rates and capture efficiencies should be prescribed by standards or norms. Here, the emission rates in Table 3 can be used to derive appropriate ventilation rates and capture efficiencies for cooker hoods following the statistical method described by Salthammer[59]. The emission rates and their standard errors can also be used as stochastic inputs to stock-scale models of housing used to estimate exposure and predict the chronic health impacts from exposure to $PM_{2.5}$ from cooking and the positive changes that may arise from mitigation measures, such as the installation and use of a cooker hood.

4 CONCLUSIONS

This work shows that the cooking of meals emits $PM_{2.5}$. The emission rate varies over time as a particular meal is cooked and is caused by a range of factors, many of which are unquantifiable. However, frying, the browning of food, the presence of oil or fat, the pan temperature, and the pan type all contribute.

It is possible to reduce $PM_{2.5}$ emissions by using a cooking method that does not brown or char the food and by using a non-stick pan when frying. Other methods were tested that have been shown elsewhere to affect $PM_{2.5}$ emission rates, such as replacing oil with liquid margarine and adding salt, but were found to have a minimal effect only.

The apparent capture efficiency of a cooker hood at a particular airflow rate is an indication of the proportion by which it reduces an emission rate. This *net* emission rate of $PM_{2.5}$ from the cooking of meals was reduced substantially by using a cooker hood with good coverage of all burners at a high airflow rate. Although the apparent capture efficiency has the advantage of being derived from measurements of $PM_{2.5}$ concentrations, there is significant uncertainty in its measurement caused by systematic biases.

Measuring capture efficiencies using ideal gases under steady-state conditions is an ideal test method because it is independent of room dynamics and contaminant interactions. However, in real-world environments cooking is rarely done under steady-state conditions and its pollutants infrequently act as ideal gases. In particular these methods do not account for particle behaviour, but they are less uncertain and only measure the ability of a hood to capture pollutants at their source. This first-order approximation is acceptable for rating cooker hoods, but a detailed estimation of occupant exposure inside a dwelling may need to consider these extra factors.

The calibration factors obtained for each meal varied away from unity and so the optical properties of the $PM_{2.5}$ emitted by cooking differ from those of the calibration source, here dolomite dust. Therefore, when measuring $PM_{2.5}$ concentrations in domestic kitchens using an optical particle counter calibrated using dolomite dust they must be corrected using an appropriate calibration factor before negative health effects can be estimated from them with any accuracy. Furthermore, the calibration factors are shown to vary by meal and so it is not possible to use a single factor for all meals without introducing significant uncertainty. Calibration factors can be obtained either from concurrent gravimetric sampling, or values

from the literature can be used with significant uncertainty. It is likely that devices calibrated by sources other than dolomite dust will also require calibration factors.

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FIGURES

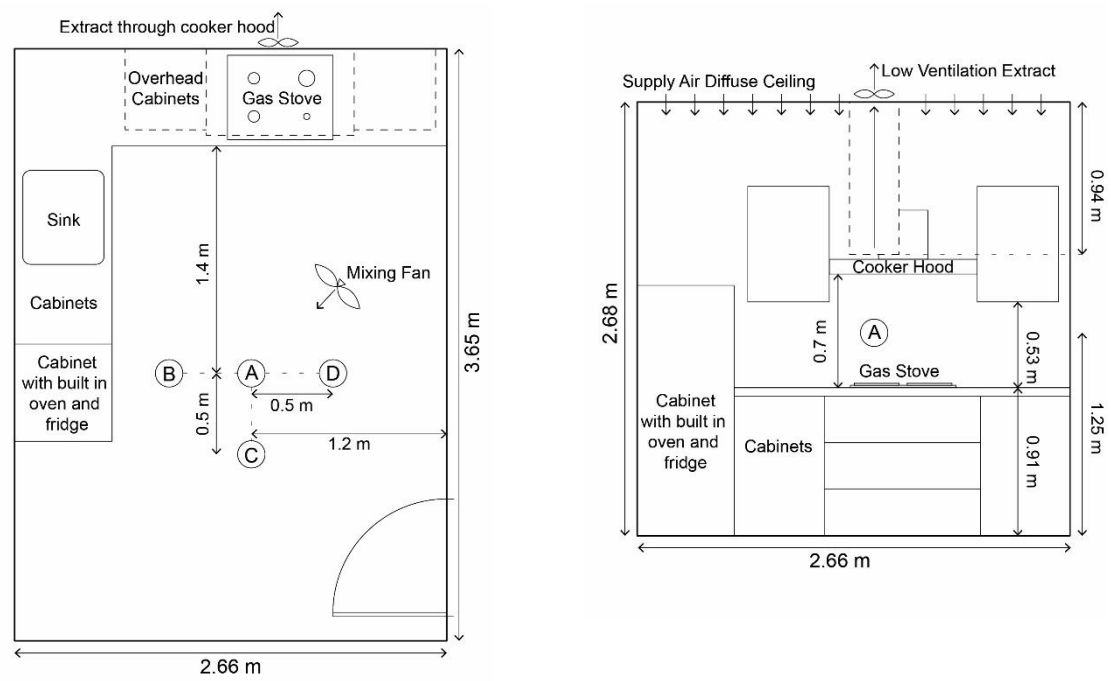
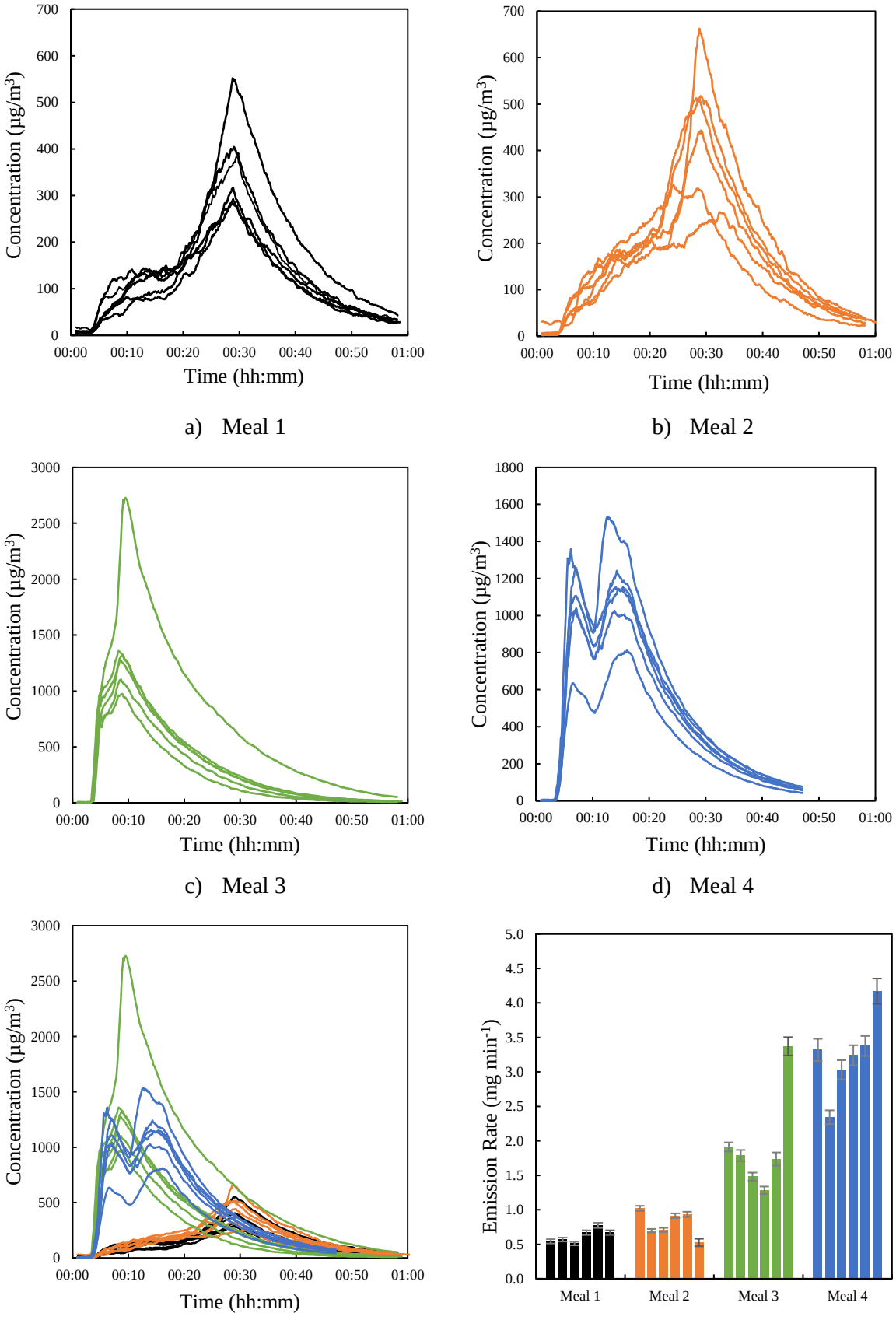


Figure 1

A = location of Grimm 11-R Mini-LAS; B, C & D = gravimetric sampling locations



a) Meal 1

b) Meal 2

c) Meal 3

d) Meal 4

e) All Meals

f) $PM_{2.5}$ Emission rates

Figure 2

9

10

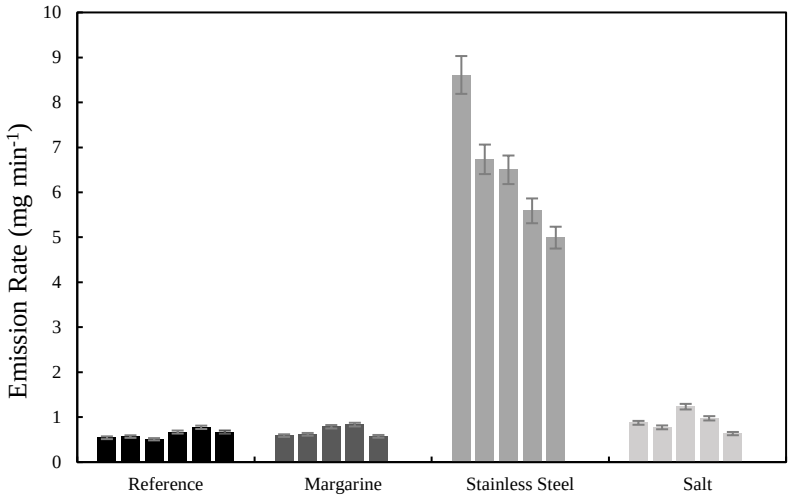


Figure 3

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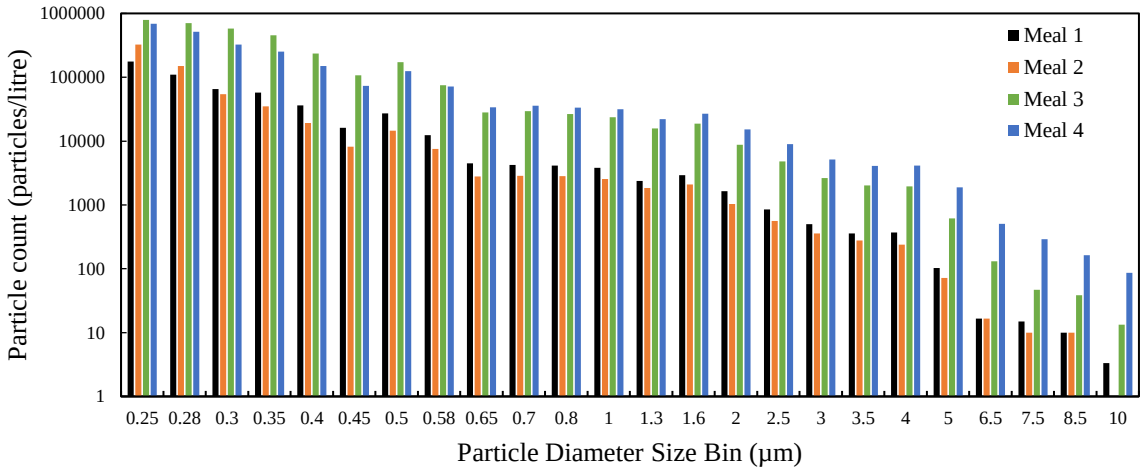
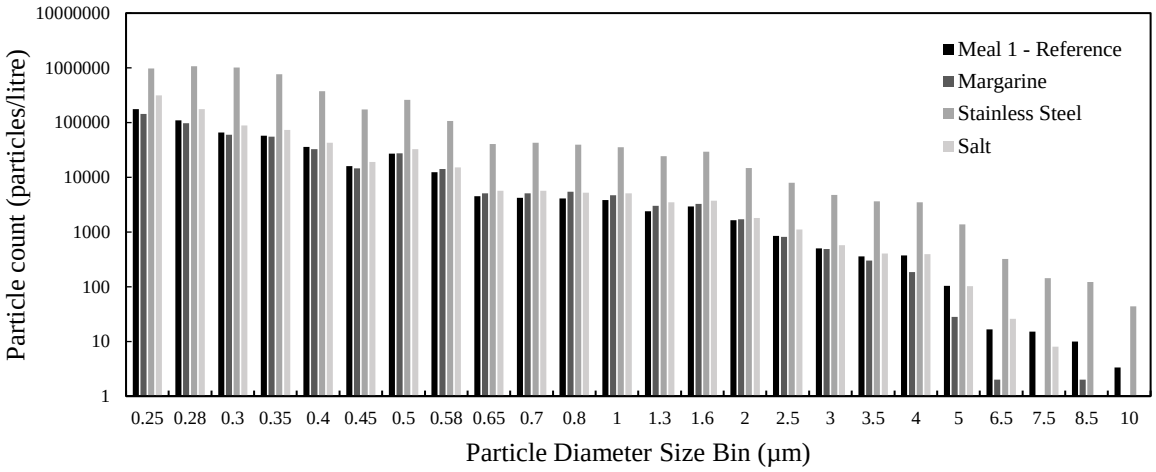
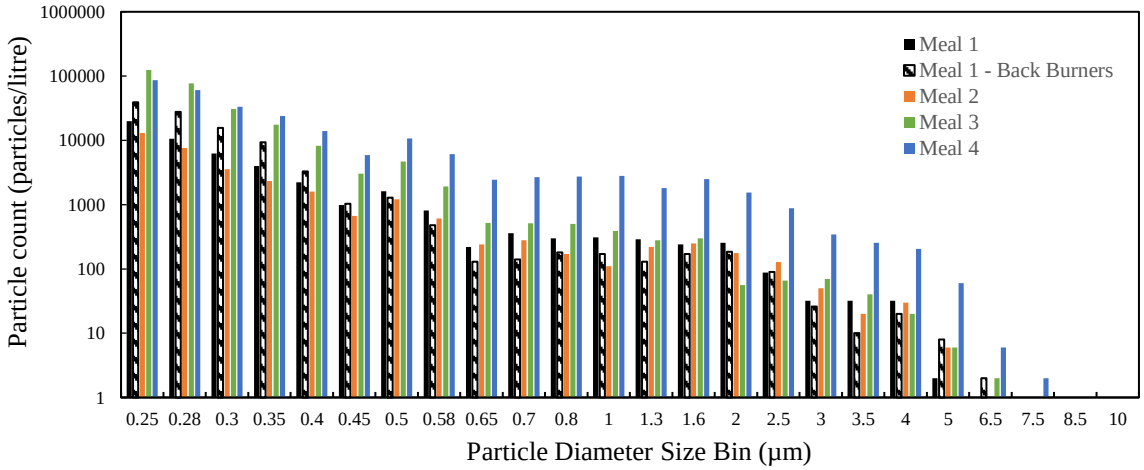


a) Base test meals, mean of 6 tests.



b) Factors potentially affecting emissions, mean of 5 tests



c) Reduction potential, mean of 5 tests

Figure 4

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TABLES

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Table 1

Meal	Ingredient	Measure	Cooking Instructions
1 *Reference* meal 28 minutes 6 repetitions	Chicken breast fillet	200g	Shallow fry in olive oil
	Olive oil	10ml	For the chicken
	Pre-sliced pre-cooked potatoes (5-10mm thickness)	330g	Fry in olive oil
	Olive oil	50ml	For the potatoes
	French/green beans	280g	Boil in water
	Water	750ml	For beans
2 28 minutes 6 repetitions	Chicken fillet	200g	Shallow fry in olive oil
	Olive oil	10ml	For the fillet
	Potatoes sliced in half	330g	Boil in water
	Water	600ml	For the potatoes
	French/green beans	280g	Boil in water
	Water	750ml	For the beans
3 Pasta Bolognese 28 minutes 6 repetitions	Dried farfalle durum wheat pasta	150g	Boil in water
	Water	1500ml	For the pasta
	Smoked lean bacon lardons (24% fat*)	125g	Fry in olive oil
	Chopped onion	115g	Fry in olive oil
	Finely chopped garlic	20g	Fry in olive oil
	Olive oil	10ml	For the fried ingredients
	Minced/ground beef (≤12% fat*)	200g	Fry in own fat
	Tinned/canned chopped tomatoes	400g*	Add to fried ingredients
4 Stir Fry 17 minutes 6 repetitions	Pre-sliced chicken breast	200g	Stir-fry in olive oil
	Olive oil	10ml	For the chicken pieces
	Pre-chopped fresh vegetables:	330g	Stir-fry in olive oil
	White cabbage	27%*	
	Red pepper /capsicum	20%*	
	Leek	20%*	
	French/green beans	20%*	
	Bean sprouts	13%*	
	Straight to wok Noodles	150g	Stir-fry in olive oil
	Olive oil	20ml	For the vegetables

Notes: All ingredients are fresh unless indicated and have not been frozen and defrosted. No seasoning was used. The olive oil was 95% refined and 5% extra virgin*. The prefix "pre" shows ingredient purchased in the described form. Symbol * denotes data taken from packaging.

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Table 2

Experiment name	N	Cooking Duration (min)	Ventilation Rate (m^3/h)
Meal 1 (*reference*)*	6	28	75
Meal 2	6	28	75
Meal 3	6	28	75
Meal 4[2]	6	17	75
"Blanks"	2	28	75
Meal 1 – Margarine	5	28	75
Meal 1 – Stainless steel pan	5	28	75
Meal 1 – Season meat with salt	5	28	75
Meal 1 (*reference*)*	5	28	300
Meal 2	5	28	300
Meal 3	5	28	300
Meal 4	5	17	300
Meal 1 - frying at backburners	5	28	300

23 *, Standard conditions: coated pan, frying on front-burner, in olive oil, gas stove

Table 3

	Emission Rate, $\overline{g(T)}$ (mg min^{-1})				Source Strength, g_{source} (mg)			
Test	Meal 1	Meal 2	Meal 3	Meal 4	Meal 1	Meal 2	Meal 3	Meal 4
1	0.54 ± 0.031	1.0 ± 0.038	1.9 ± 0.066 *	3.3 ± 0.16 *	15 ± 0.88	29 ± 1.1	55 ± 1.9 *	56 ± 2.8 *
2	0.57 ± 0.029	0.70 ± 0.025	1.8 ± 0.082 *	2.3 ± 0.10 *	16 ± 0.80	20 ± 0.71	50 ± 2.3 *	40 ± 1.7 *
3	0.51 ± 0.025	0.71 ± 0.030	1.5 ± 0.059 *	3.0 ± 0.14 *	14 ± 0.71	20 ± 0.83	41 ± 1.6 *	52 ± 2.4 *
4	0.67 ± 0.033	0.91 ± 0.034	1.3 ± 0.055	3.2 ± 0.15 *	19 ± 0.94	26 ± 0.96	36 ± 1.5	55 ± 2.5 *
5	0.77 ± 0.038	0.93 ± 0.038	1.7 ± 0.10 *	3.4 ± 0.14 *	22 ± 1.1	26 ± 1.1	49 ± 2.7 *	58 ± 2.5 *
6	0.67 ± 0.035	0.52 ± 0.054	3.4 ± 0.13 *	4.2 ± 0.18	19 ± 0.98	16 ± 1.7	95 ± 3.7 *	70 ± 3.1
Mean ± α_g†	0.62 ± 0.041	0.80 ± 0.076	1.9 ± 0.30	3.2 ± 0.24	17± 1.1	23 ± 2.0	54 ± 8.6	55 ± 3.9
Standard Deviation	0.10	0.19	0.74	0.59	2.8	4.9	21	9.6

* particle count exceeded 2,000,000 particles/litre

†standard error

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Table 4

Test	Meal 1 Emission Rate, $\overline{g(T)}$ (mg min^{-1})				Meal 1 Source Strength, g_{source} (mg)			
	Reference	Margarine	Stainless Steel	Salt	Reference	Margarine	Stainless Steel	Salt
1	0.54 ± 0.031	0.59 ± 0.029	8.6 ± 0.42 *	0.87 ± 0.043	15 ± 0.88	17 ± 0.82	240 ± 12 *	24 ± 1.2
2	0.57 ± 0.029	0.62 ± 0.030	6.7 ± 0.33 *	0.77 ± 0.043	16 ± 0.80	17 ± 0.85	190 ± 9.2 *	22 ± 1.2
3	0.51 ± 0.025	0.79 ± 0.038	6.5 ± 0.32 *	1.2 ± 0.062	14 ± 0.71	22 ± 1.1	180 ± 8.9 *	35 ± 1.7
4	0.67 ± 0.033	0.83 ± 0.041	5.6 ± 0.28 *	0.97 ± 0.048	19 ± 0.94	23 ± 1.2	160 ± 7.8 *	27 ± 1.3
5	0.77 ± 0.038	0.57 ± 0.031	5.0 ± 0.24 *	0.63 ± 0.036	22 ± 1.1	16 ±0.86	140 ± 6.8 *	18 ± 1.0
6	0.67 ± 0.035				19 ± 0.98			
Mean ± α_g†	0.62 ± 0.041	0.68 ± 0.054	6.5 ± 0.62	0.90 ± 0.10	17 ± 1.1	19 ± 1.5	180 ± 17	25 ± 2.8
Standard Deviation	0.10	0.12	1.4	0.23	2.8	3.4	39	6.3
Increase (%)		9	940	44		9	940	44

* particle count exceeded 2,000,000 particles/litre

†standard error

Table 5

Test	Emission Rate, $\overline{g(T)}$ (mg min^{-1})					Meal 1 Source Strength, g_{source} (mg)				
	Meal 1	Meal 1 – Back Burners	Meal 2	Meal 3	Meal 4	Meal 1	Meal 1 – Back Burners	Meal 2	Meal 3	Meal 4
1	0.027 ± 0.0043	0.040 ± 0.0020	0.023 ± 0.0036	- *	0.093 ± 0.038	0.75 ± 0.12	1.1 ± 0.056	0.64 ± 0.10	- *	1.6 ± 0.65
2	0.064 ± 0.017	0.029 ± 0.0022	0.053 ± 0.0056	0.0068 ± 0.0010	0.072 ± 0.022	1.8 ± 0.48	0.80 ± 0.062	1.5 ± 0.16	0.19 ± 0.029	1.2 ± 0.38
3	0.031 ± 0.0039	0.053 ± 0.011	0.071 ± 0.017	0.0080 ± 0.00094	0.038 ± 0.017	0.87 ± 0.11	1.5 ± 0.31	2.0 ± 0.47	0.22 ± 0.026	0.65 ± 0.30
4	0.024 ± 0.0045	0.024 ± 0.0056	0.015 ± 0.0021	- *	0.027 ± 0.006	0.68 ± 0.13	0.68 ± 0.16	0.42 ± 0.060	- *	0.45 ± 0.10
5	0.062 ± 0.012	0.026 ± 0.0039	0.037 ± 0.0039	- *	0.23 ± 0.075	1.8 ± 0.25	0.73 ± 0.11	1.0 ± 0.11	- *	3.9 ± 1.3
Mean ± α_g†	0.042 ± 0.0089	0.034 ± 0.0054	0.040 ± 0.010	0.0074 ± 0.00060	0.091 ± 0.036	1.2 ± 0.25	0.96 ± 0.15	1.1 ± 0.29	0.15 ± 0.04	1.6 ± 0.61
Standard Deviation	0.020	0.012	0.023	0.00084	0.081	0.56	0.34	0.64	0.024	1.4
Reduction (%)	93	94	95	99.6	97	93	94	95	99.7	97

* concentrations too low to estimate decay rate

†standard error

Supplementary Materials

Investigating Measurements of Fine Particle ($PM_{2.5}$) Emissions from the Cooking of Meals and mitigating exposure using a cooker hood

Catherine O'Leary[1, 2], Yvonne Kluizenaar[2], Piet Jacobs[2], Wouter Borsboom[2], Ian Hall[3], Benjamin Jones[*1]

1. Department of Architecture and Built Environment, University of Nottingham, Nottingham, NG7 2RD, United Kingdom
2. Netherlands Organisation for Applied Scientific Research (TNO), Delft, The Netherlands
3. Division of Respiratory Medicine, School of Medicine, University of Nottingham, Nottingham, NG7 2UH, UK.

***Corresponding author: benjamin.jones@nottingham.ac.uk**

FIGURES

16

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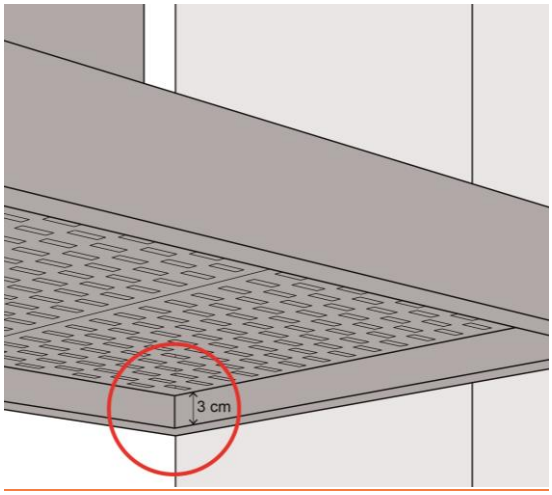
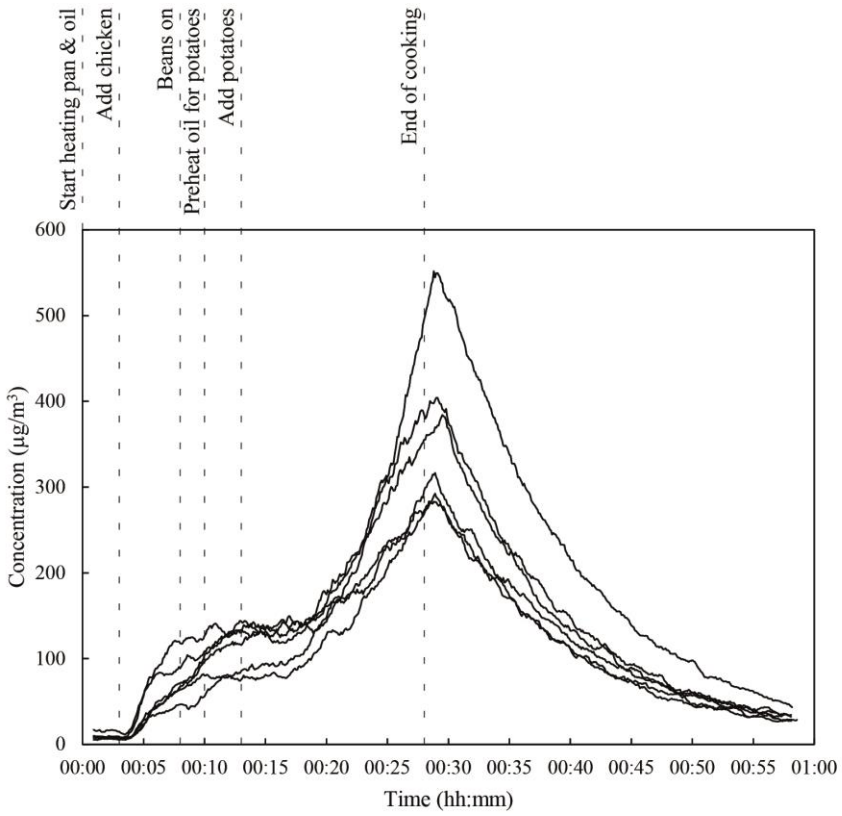


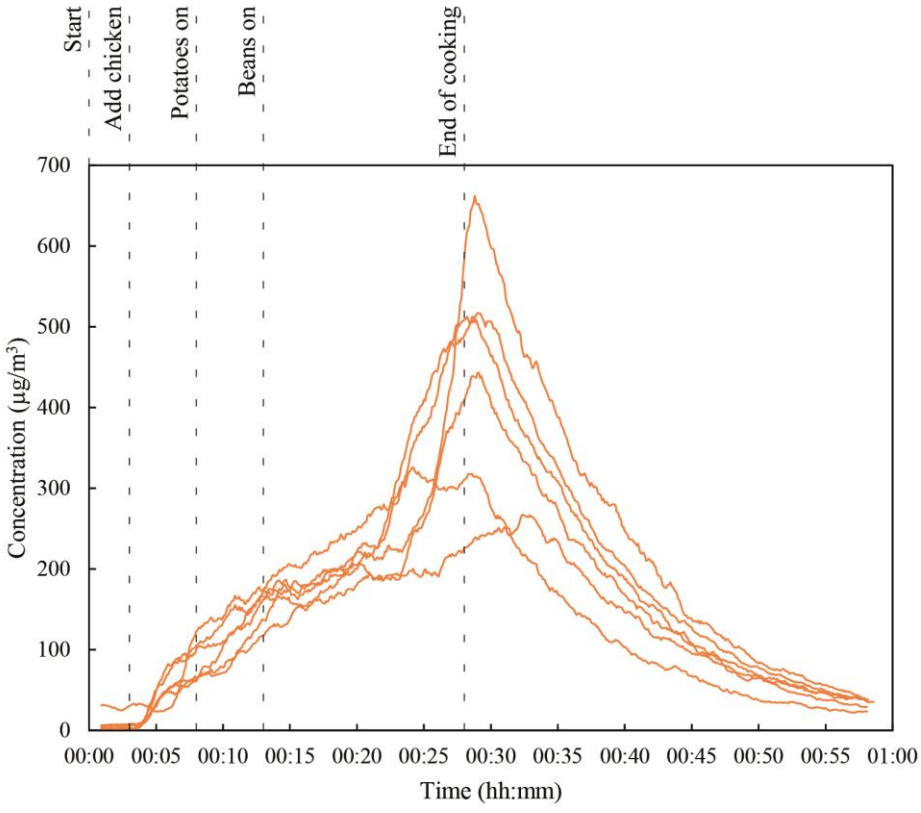
a) Photo of the test rig, with damp-buffer circled

b) 3D diagram of cooker hood showing the damp-buffer

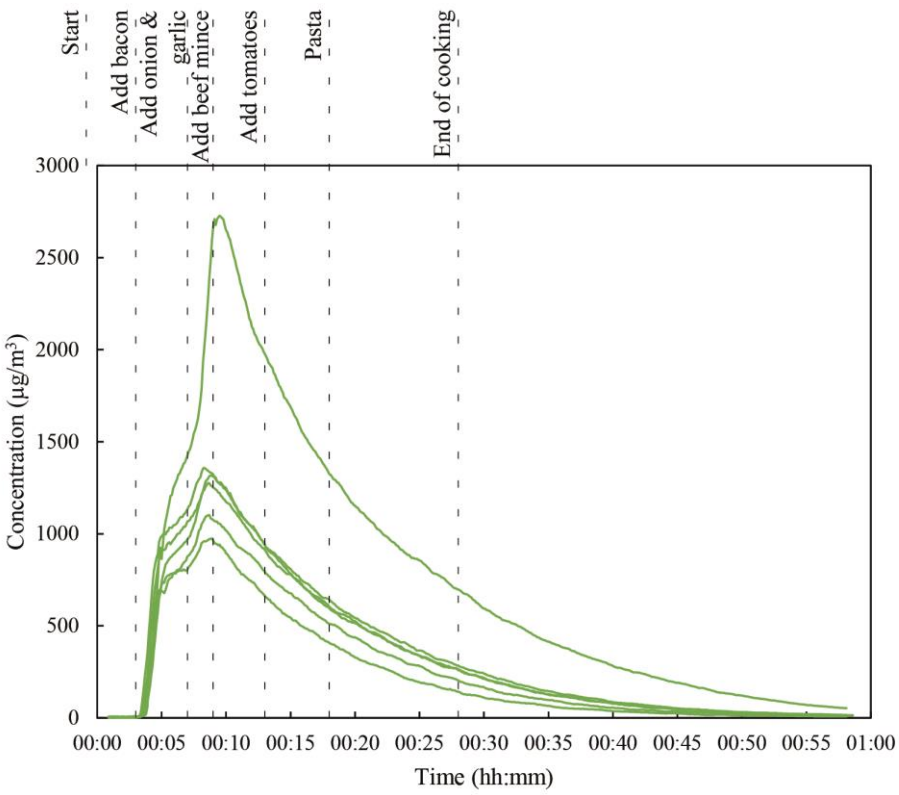
Figure S1 – The test rig



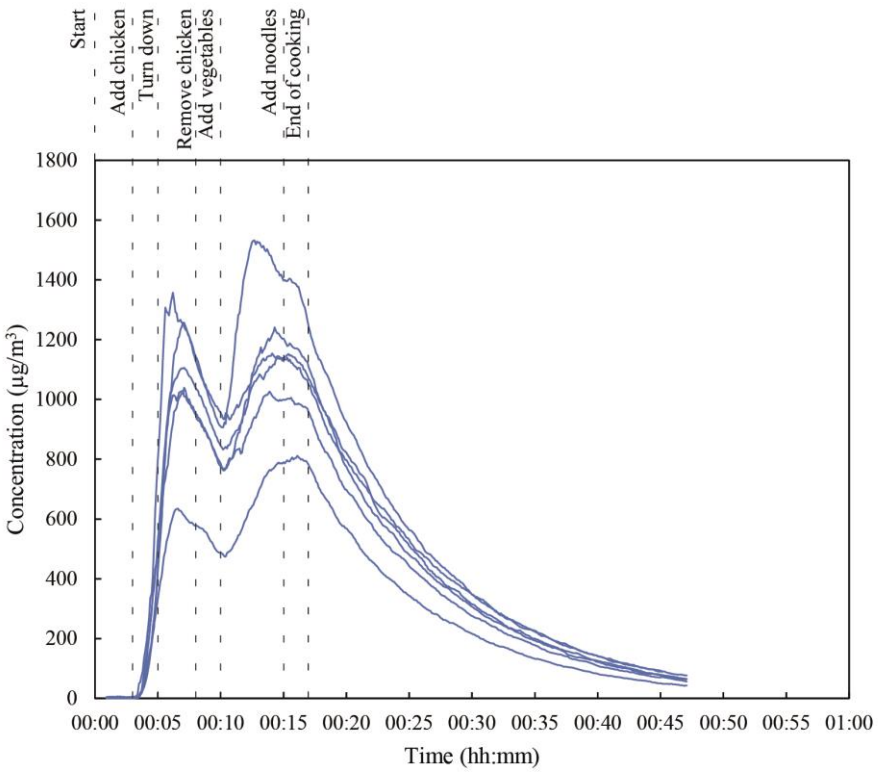
a) Meal 1



b) Meal 2



c) Meal 3



d) Meal 4

Figure S2

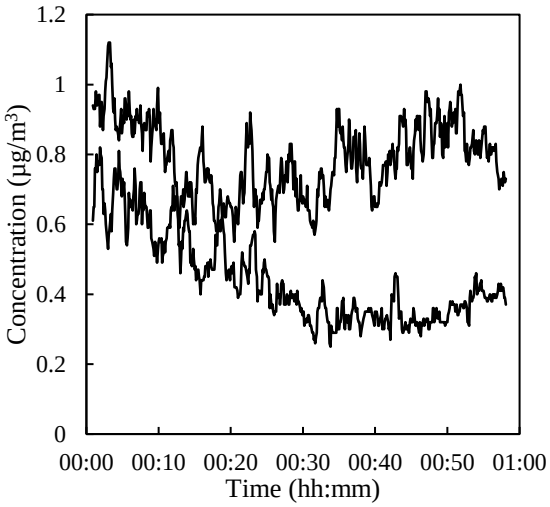

1.2
1
0.8
0.6
0.4
0.2
0
Concentration (μg/m3)
00:00
00:10
00:20
00:30
00:40
00:50
01:00
Time (hh:mm)

Figure S3

TABLES

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Table S1

	Sampling Time (s)	Total Mass (µg)			Average Concentration (µg/m^3)				Calibration Factor ± Standard Error
		Filter 1	Filter 2	Filter 3	Filter 1	Filter 2	Filter 3	Grimm	
Meal 1	3480	310	340	300	139	153	135	36.3	3.92 ± 0.15
Meal 2	1680	180	170	170	168	158	158	32.3	4.99 ± 0.096
Meal 3	3240	1620	1570	1650	783	758	797	290*	2.69 ± 0.039
Meal 4	2760	1020	1030	1120	578	584	635	401	1.50 ± 0.045
Pump flow rate was 2.3 m^3/h (0.64 l/s) during all tests									

* peak particle count exceeded maximum 2,000,000 particles/litre

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Table S2

Test	Decay Rate (h^{-1})							
	Meal 1		Meal 2		Meal 3		Meal 4	
	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2
1	4.8 ± 0.044	0.98	5.9 ± 0.039	0.99	5.9 ± 0.035 *	0.99	5.3 ± 0.016 *	1.00
2	4.7 ± 0.039	0.98	5.3 ± 0.037	0.99	5.8 ± 0.038 *	0.99	5.7 ± 0.019 *	1.00
3	5.0 ± 0.046	0.98	5.6 ± 0.053	0.97	5.8 ± 0.055 *	0.97	5.5 ± 0.016 *	1.00
4	5.3 ± 0.040	0.98	5.6 ± 0.027	0.99	6.0 ± 0.062	0.97	5.5 ± 0.017 *	1.00
5	5.2 ± 0.028	0.99	6.1 ± 0.042	0.99	5.9 ± 0.034 *	0.99	5.6 ± 0.017 *	1.00
6	5.1 ± 0.049	0.97	4.7 ± 0.041	0.98	5.2 ± 0.025 *	0.99	5.6 ± 0.016	1.00
Mean ± α_Φ†	5.0 ± 0.10		5.5 ± 0.21		5.7 ± 0.12		5.5 ± 0.06	

* particle count exceeded 2,000,000 particles/litre

†standard error

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Table S3

	Decay Rate (h^{-1})					
Test	Margarine		Stainless Steel		Salt	
	Decay	R^2	Decay	R^2	Decay	R^2
1	5.4 ± 0.045	0.98	4.5 ± 0.014 *	1.00	5.7 ± 0.035	0.99
2	5.5 ± 0.041	0.98	3.9 ± 0.012 *	1.00	5.5 ± 0.037	0.99
3	5.6 ± 0.034	0.99	4.5 ± 0.013 *	1.00	5.8 ± 0.032	0.99
4	5.4 ± 0.031	0.99	4.4 ± 0.012 *	1.00	5.7 ± 0.031	0.99
5	5.0 ± 0.044	0.98	3.7 ± 0.010 *	1.00	5.3 ± 0.039	0.99
Mean ± α_Φ†	5.4 ± 0.11		4.2 ± 0.16		5.9 ± 0.09	

* particle count exceeded 2,000,000 particles/litre

†standard error

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Table S4

Test	Decay Rate (h^{-1})									
	Meal 1		Meal 1 - Back Burners		Meal 2		Meal 3		Meal 4	
	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2
1	2.7 ± 0.11	0.66	1.2 ± 0.062	0.55	1.8 ± 0.13	0.39	- *	-	3.2 ± 0.12	0.71
2	3.9 ± 0.12	0.77	0.70 ± 0.029	0.65	4.7 ± 0.17	0.72	0.076 ± 0.043	0.010	3.0 ± 0.11	0.73
3	2.3 ± 0.11	0.60	2.0 ± 0.059	0.79	4.5 ± 0.17	0.71	0.36 ± 0.047	0.16	2.3 ± 0.073	0.76
4	1.9 ± 0.12	0.45	1.1 ± 0.059	0.54	1.3 ± 0.14	0.24	- *	-	2.1 ± 0.088	0.66
5	3.8 ± 0.13	0.75	1.7 ± 0.067	0.67	3.6 ± 0.14	0.70	- *	-	5.6 ± 0.15	0.83
Mean ± α_Φ †	2.9 ± 0.40		1.3 ± 0.22		3.2 ± 0.68		0.22 ± 0.14		3.3 ± 0.68	

* concentrations too low to estimate decay rate

†standard error

30

Supplementary Materials

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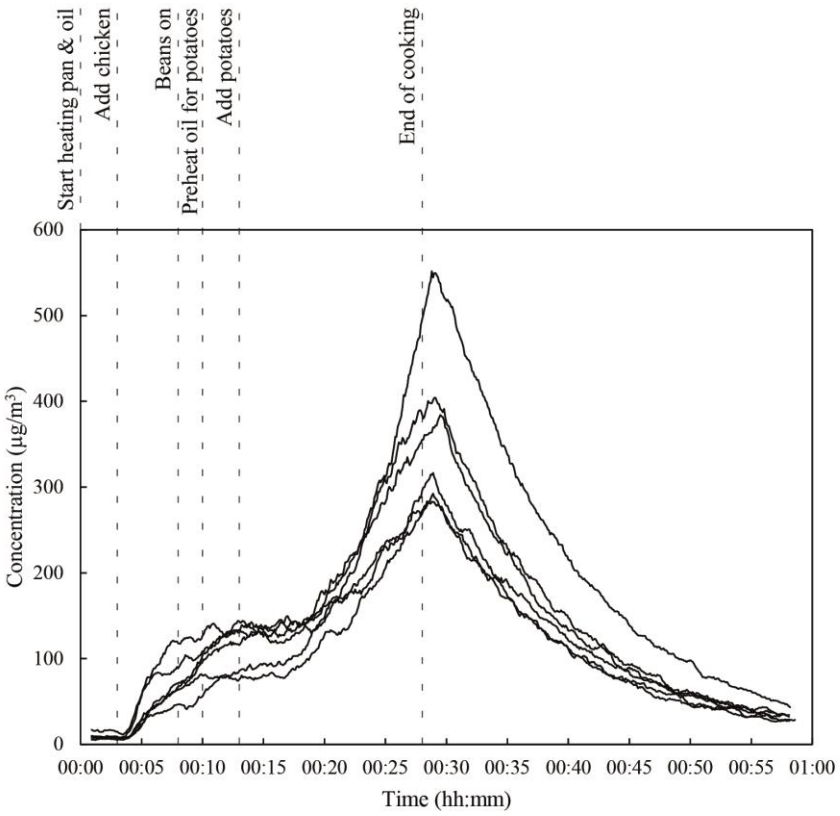
FIGURES

16

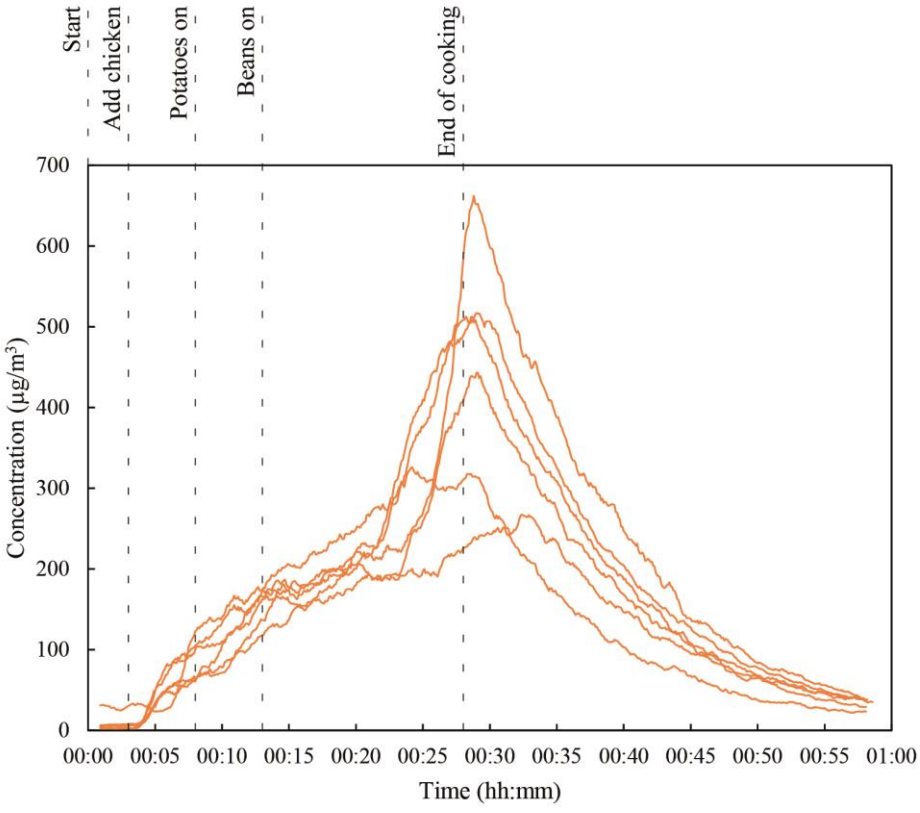
a) Photo of the test rig, with damp-buffer circled

b) 3D diagram of cooker hood showing the damp-buffer

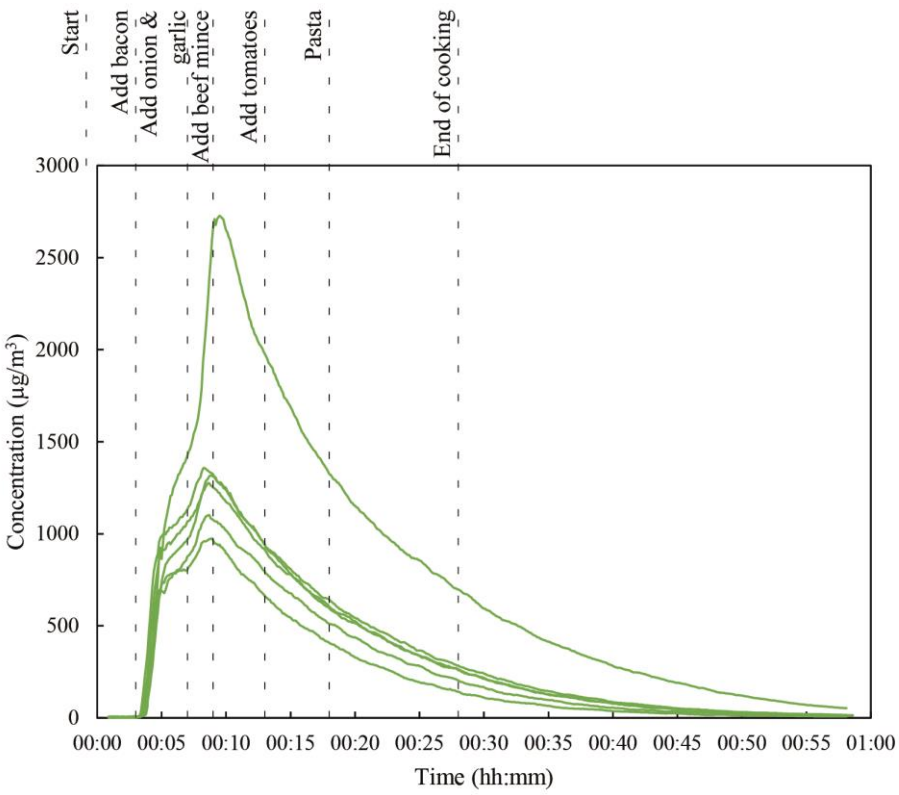
17 Figure S1 – The test rig



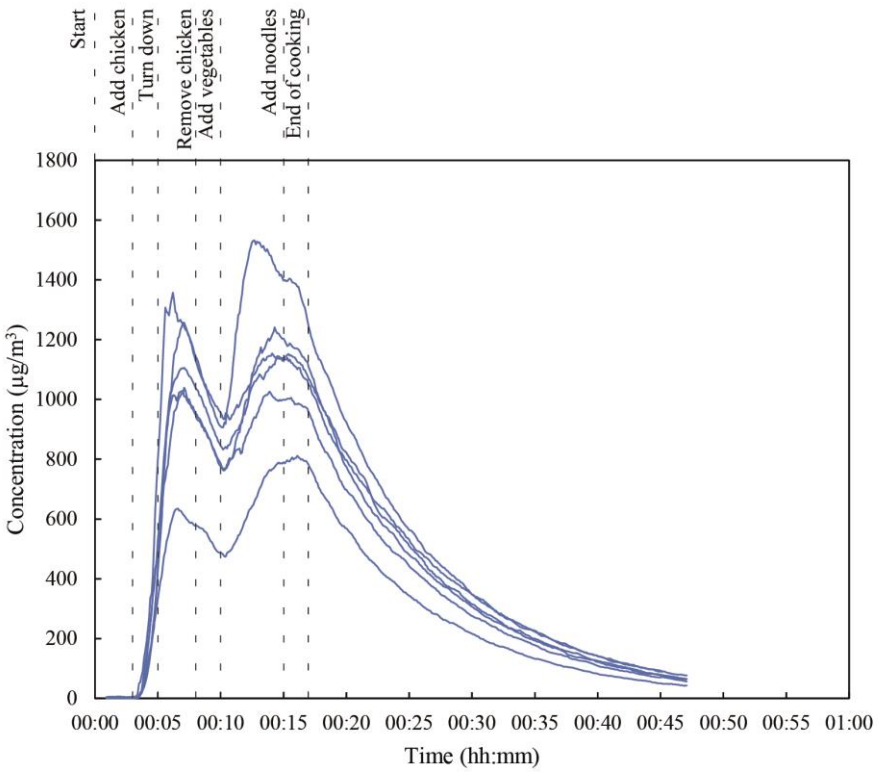
a) Meal 1



b) Meal 2

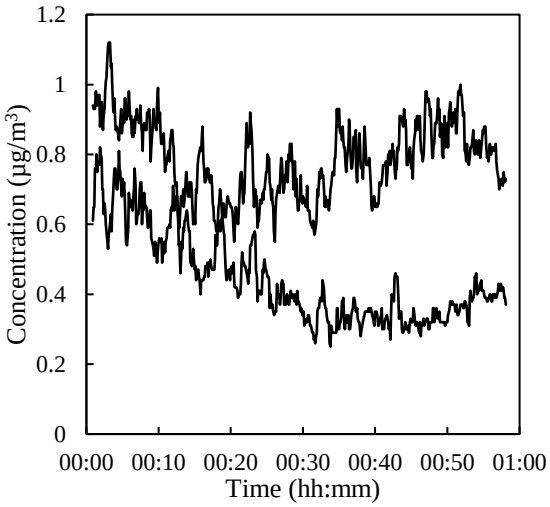


c) Meal 3



d) Meal 4

Figure S2



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TABLES

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Table S1

	Sampling Time (s)	Total Mass (µg)			Average Concentration (µg/m^3)				Calibration Factor ± Standard Error
		Filter 1	Filter 2	Filter 3	Filter 1	Filter 2	Filter 3	Grimm	
Meal 1	3480	310	340	300	139	153	135	36.3	3.92 ± 0.15
Meal 2	1680	180	170	170	168	158	158	32.3	4.99 ± 0.096
Meal 3	3240	1620	1570	1650	783	758	797	290*	2.69 ± 0.039
Meal 4	2760	1020	1030	1120	578	584	635	401	1.50 ± 0.045
Pump flow rate was 2.3 m^3/h (0.64 l/s) during all tests									

* peak particle count exceeded maximum 2,000,000 particles/litre

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Table S2

Test	Decay Rate (h^{-1})							
	Meal 1		Meal 2		Meal 3		Meal 4	
	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2
1	4.8 ± 0.044	0.98	5.9 ± 0.039	0.99	5.9 ± 0.035 *	0.99	5.3 ± 0.016 *	1.00
2	4.7 ± 0.039	0.98	5.3 ± 0.037	0.99	5.8 ± 0.038 *	0.99	5.7 ± 0.019 *	1.00
3	5.0 ± 0.046	0.98	5.6 ± 0.053	0.97	5.8 ± 0.055 *	0.97	5.5 ± 0.016 *	1.00
4	5.3 ± 0.040	0.98	5.6 ± 0.027	0.99	6.0 ± 0.062	0.97	5.5 ± 0.017 *	1.00
5	5.2 ± 0.028	0.99	6.1 ± 0.042	0.99	5.9 ± 0.034 *	0.99	5.6 ± 0.017 *	1.00
6	5.1 ± 0.049	0.97	4.7 ± 0.041	0.98	5.2 ± 0.025 *	0.99	5.6 ± 0.016	1.00
Mean ± α_Φ†	5.0 ± 0.10		5.5 ± 0.21		5.7 ± 0.12		5.5 ± 0.06	

* particle count exceeded 2,000,000 particles/litre

†standard error

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Table S3

Test	Decay Rate (h^{-1})					
	Margarine		Stainless Steel		Salt	
	Decay	R^2	Decay	R^2	Decay	R^2
1	5.4 ± 0.045	0.98	4.5 ± 0.014 *	1.00	5.7 ± 0.035	0.99
2	5.5 ± 0.041	0.98	3.9 ± 0.012 *	1.00	5.5 ± 0.037	0.99
3	5.6 ± 0.034	0.99	4.5 ± 0.013 *	1.00	5.8 ± 0.032	0.99
4	5.4 ± 0.031	0.99	4.4 ± 0.012 *	1.00	5.7 ± 0.031	0.99
5	5.0 ± 0.044	0.98	3.7 ± 0.010 *	1.00	5.3 ± 0.039	0.99
Mean ± α_{Φ}†	5.4 ± 0.11		4.2 ± 0.16		5.9 ± 0.09	

* particle count exceeded 2,000,000 particles/litre

†standard error

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Table S4

Test	Decay Rate (h^{-1})									
	Meal 1		Meal 1 - Back Burners		Meal 2		Meal 3		Meal 4	
	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2	Decay	R^2
1	2.7 ± 0.11	0.66	1.2 ± 0.062	0.55	1.8 ± 0.13	0.39	- *	-	3.2 ± 0.12	0.71
2	3.9 ± 0.12	0.77	0.70 ± 0.029	0.65	4.7 ± 0.17	0.72	0.076 ± 0.043	0.010	3.0 ± 0.11	0.73
3	2.3 ± 0.11	0.60	2.0 ± 0.059	0.79	4.5 ± 0.17	0.71	0.36 ± 0.047	0.16	2.3 ± 0.073	0.76
4	1.9 ± 0.12	0.45	1.1 ± 0.059	0.54	1.3 ± 0.14	0.24	- *	-	2.1 ± 0.088	0.66
5	3.8 ± 0.13	0.75	1.7 ± 0.067	0.67	3.6 ± 0.14	0.70	- *	-	5.6 ± 0.15	0.83
Mean ± α_Φ †	2.9 ± 0.40		1.3 ± 0.22		3.2 ± 0.68		0.22 ± 0.14		3.3 ± 0.68	

* concentrations too low to estimate decay rate

†standard error

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