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Chemical fingerprints of cooking emissions and their impact on indoor air quality

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Indoor environments host multiple sources of volatile organic compounds (VOCs) that influence the air quality, with cooking being one such significant and complex emission source. VOC emissions from cooking vary with the type of food cooked, ingredients used, cooking methodology, and ventilation, yet their speciation and impact on indoor air remain poorly understood. This study quantifies real-time emission rates of 39 VOCs from three frequently prepared UK meals: stir-fry, curry, and chilli, using a high-sensitivity selected ion flow tube mass spectrometer (SIFT-MS) in a room-scale, semi-realistic kitchen. Across 39 cooking experiments a distinct VOC emission profile for each meal was measured. The emissions were dominated by alcohols (methanol and ethanol, >50% of total emissions), harmful aldehydes (acetaldehyde, 7–23%), and highly reactive monoterpenes (up to 4%). The emissions were found to be influenced strongly by the use of different variants of the same ingredient (freshly chopped and packaged diced onions), spices and cooking behaviours. The secondary chemistry of the resultant VOC emissions was further investigated by simulating the hydroxyl (OH) reactivity and secondary product formation using INCHEM-Py. The model results show that the cooking plumes significantly perturbed the indoor chemistry, with OH reactivity ranging from 50–200 s^{-1} depending on VOC composition. Further simulations of a typical urban London kitchen revealed recipe-dependent impacts on radical (HO_2, RO_2) and secondary pollutant (O_3, PAN, organic nitrates, formaldehyde) formation. Among the meals tested, chillies exhibited the highest potential for secondary pollutant production, followed by curries. These findings highlight the influence of cooking emissions on indoor air quality and secondary chemistry.

Environmental significance

Domestic cooking is a major source of volatile organic compounds (VOCs) in indoor environments and these emissions can vary significantly based on cuisine type and cooking methods. Subsequently, these primary VOC emissions interact with oxidants such as O_3 and OH radicals and produce secondary products with potential health implications. Our study employs real-time chemical characterization of VOCs in scripted cooking of diverse cuisines, revealing distinct emission signatures across the different cooking stages and the influence of different cooking ingredients. The experimentally quantified emission rates are used in an indoor air chemistry model to investigate their impact on indoor air quality. This work reveals relationships between cooking practices and indoor air quality deterioration, providing a quantitative foundation for developing targeted mitigation strategies centred around human behaviour change during cooking activities.

1 Introduction

In high-income countries like the UK, people spend nearly 90% of their time indoors, and about 60% in their homes.[1] While outdoor pollution can have a considerable influence on indoor air pollutant loading, the inability of the pollutants to disperse easily in indoor environments results in their accumulation and significantly elevated concentrations.[2,3] Human activities such as cooking, are an important and perhaps the largest source of gaseous and particulate emissions in indoor environments.[4–8] Amongst the gaseous emissions, volatile organic compounds (VOCs) are particularly important because of the pivotal role they play in atmospheric chemical processes such as chemical reactivity, and secondary pollutant formation.[7,9] Additionally, VOCs such as formaldehyde, BTEX (benzene, toluene, ethylbenzene, and xylene), acrolein, and acetaldehyde are known to have considerable health effects.[10] The VOC emissions from cooking processes are highly influenced by cooking fuels, cooking styles, recipes, and the use of different ingredients like oils, spices, *etc.*[6,11] These changes in cooking ingredients and

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styles are highly localized and depend on several socio-cultural and geographical factors, thereby, making it difficult to generalize conclusions worldwide. For example, cooking vegetables releases high amounts of ethanol and dimethyl sulfide,[11,12] the use of spices releases a considerable amount of monoterpenes,[11,13] and heating oils emits carbonyl (aldehydes and ketones), alkanes, and organic acids.[8,11,14]

While in low and middle-income countries cooking emissions are primarily dominated by the use of biomass-based fuels, in countries such as the UK use of natural gas, and electricity, would suggest that cooking emissions are likely to be influenced by the types of food cooked and the cooking behaviours, rather than the fuel emissions.[3,4] The emissions of particulate matter from cooking processes have been studied extensively before, however, gaseous emissions and speciated VOCs at high-temporal resolution in particular, have rarely been investigated.[15] Furthermore, the focus of such studies has been centred around carbonyls and polycyclic aromatic hydrocarbons (PAHs) because of their potential adverse health effects on humans.[8,15–20] A more comprehensive VOC characterization has been carried out for only large-scale cooking processes such as commercial cooking in restaurants, for a particular cuisine such as Chinese,[9,21–24] and/or for the cooking of individual ingredients in a chamber under highly controlled conditions, that may not resemble real-world household cooking or a full-scale recipe.[12,25]

Variability in cooking styles and ventilation systems produces differences in the VOC emissions between commercial and residential cooking.[15] The recent HOMEChem (House Observations of Microbial and Environmental Chemistry) campaign studied the VOC emissions from cooking and their impact on indoor air quality in detail, particularly vegetable stir-fry and a traditional Thanksgiving meal.[26,27] The study focused on sequential cooking and cleaning activities, and considering the limitations due to the mixing time of air in their test house, the emissions between food preparation and dishwashing could not be quantitatively differentiated.[26] Arata *et al.*[26] estimates an approximate 19% contribution to ethanol concentrations from dishwashing, which was also identified as the highest emitted VOC from their stir-fry cooking. Their results however, enabled them to conclude that VOC emissions from cooking are a highly complex mixture and are likely to influence both indoor and outdoor air chemistry.[26,28] Davies *et al.*[14] studied the speciated VOC emissions and their impact on indoor air quality from stir-fry cooking in a semi-realistic environment. Their results showed that the VOC emission rates and compositions were highly dependent on the different stages of the cooking process and the ingredients cooked, and that these emissions were also an important source of indoor secondary pollutants.

In this study, we build upon the work carried out by Davies *et al.*[14] and our previous study (Kumar *et al.*[11]), and present real-time measurements of 39 VOCs during the cooking of three typical UK meals: chilli, curry, and stir-fry.[29] A vegetarian and non-vegetarian version of each meal allowed us to ascertain any differences in the emissions due to changes in the ingredients. The impact of these emissions on indoor air chemistry was further investigated *via* the INCHEM-Py model.

2 Methods

2.1 Site description

Experiments were performed at the DOMESTIC kitchen laboratory at the University of York. The facility comprises two shipping containers (Fig. 1) modified to mimic a "real-world" experimental facility for studying indoor air science and in particular the quantification of gaseous phase emissions from cooking, cleaning, and personal care product usage. The facility has been described in detail in previous works.[14,30] Container #1 consists of a kitchen (4.3 m × 2.2 m × 2.3 m, volume = 22.0 m^3) and bathroom (1.5 m × 2.2 m × 2.3 m, volume = 7.1 m^3), while container #2 houses the instruments. The indoor air from container 1 is drawn continuously to container 2 through a main sample line (∼20 m long, 1/2″ OD opaque PTFE tubing) at ∼30 L min^{-1} using a diaphragm pump (Model MPC 301 Z, Welch, Germany). The sample inlet in container #1 was stationed at a height of ∼1.2 m above the cooking hob (Fig. 1) and had an inline particle filter (filter diameter 47 mm; pore size 1 μm). The main sample line is insulated and heated at ∼60 °C using heating tape to minimize any condensation and potential losses. The air from the main line is subsampled by the instruments at their respective working flow rates. Another inlet line for the measurement of ambient air was stationed outside container #2 at ∼2 m above ground level and connected to the main sample line and pump. The air sampled by the instruments was manually switched between the indoor air (coming from container #1) and outdoor air (coming from the ambient inlet) *via* a three-way valve (1/4 inch, stainless steel, Swagelok). The ambient measurements were conducted for roughly 15 min prior to every cooking experiment.

2.2 Cooking experiments

The cooking experiments were carried out during two separate periods, one between 08 Feb 2023 and 03 March 2023, and the second between 13 May 2023 and 08 June 2023. Two experiments were performed in a day, one in the morning (∼10:00 local time) and one in the afternoon (∼16:00 local time), and the extractor fan was automatically run between 13:00 to 14:00 and 22:00 to 04:00 local time. Continuous VOC measurements were carried throughout the experiment, which allowed us to capture the periods corresponding to before cooking (room background), during cooking, and after cooking. To capture the repeatability and cook-to-cook variability, each recipe was cooked at least six times under low ventilation scenario (no extractor fan on). The cooking experiments comprised six different scripted recipes, namely chicken stir fry, tofu stir fry, beef chilli, non-meat chilli, paneer curry, and chicken curry. The recipes (Tables S1–S6) were developed to represent general cooking patterns in UK households and were based on the survey results from 300 households being studied as part of the INGENIOUS project[31,32] and previous works.[11,14,33] The raw cooking materials for all of the experiments were procured from the same manufacturer/supermarket to maintain consistency. The perishable items like meat were stored in the refrigerator at 4 °C while the canned and dry ingredients (oil and spices) were
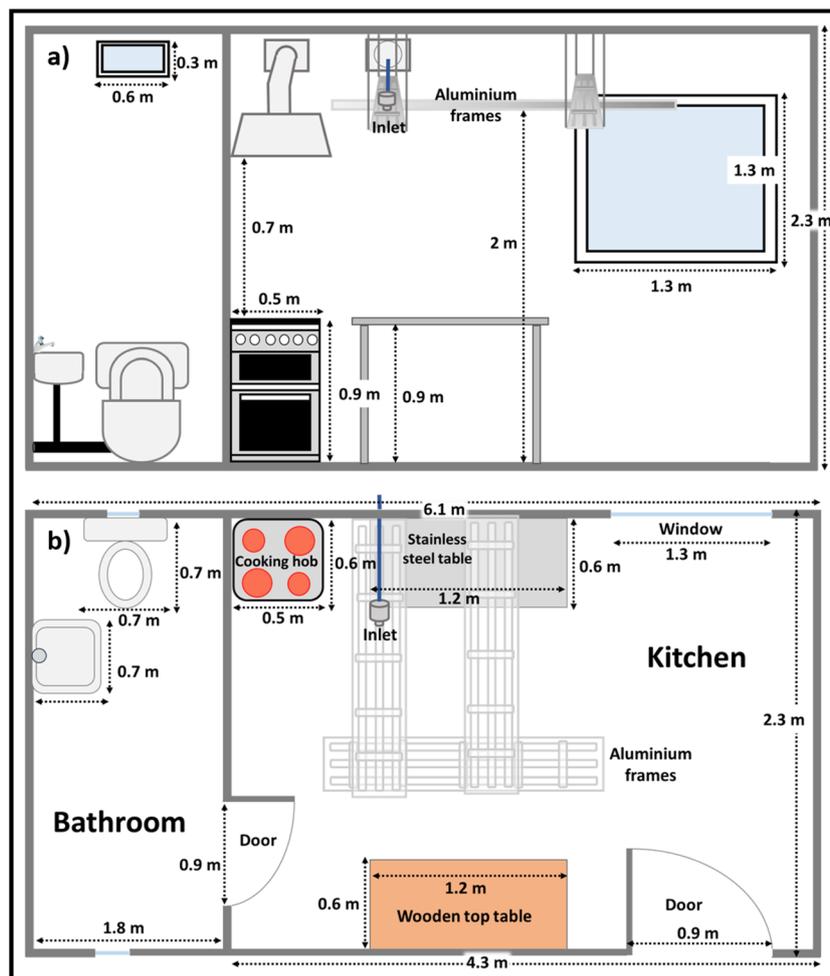
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Fig. 1 (a) Sectional and (b) plan view of the DOMESTIC kitchen laboratory (container 1). The sample inlet is positioned closer to the cooking station and marked in the figure. During the experiments, all the doors (including the bathroom door) and windows were shut.

kept at room temperature. The perishable ingredients were used within three days from the date of purchase. A fresh batch of non-perishable items like oils and spices was used for each of the experimental campaigns in February and May.

The protocols for the cooking experiments were based on the previous study by Davies *et al.*,[14] where a single person or chef entered the kitchen along with the required ingredients, and cooked the meal. Once the cooking was finished as per the recipe timings, the chef covered the pan containing the cooked food and immediately removed it from the kitchen and left the room. This ensured that the VOC emissions that were measured reflected only the active cooking, and not the period afterwards. As per the designed experiments, the start of the cooking period was determined by the addition of oil into the hot pan (as $t = 0$ s in Fig. 3) and the end of the cooking was determined by the removal of the pan from the cooking hob and covering it with the lid. The chef entered the kitchen roughly 10 min before $t = 0$ s, did pre-cooking preparations, cooked the food, and exited the kitchen with covered food within 1 min of the end of the cooking period. All the food preparations (like chopping) were carried out in the period before $t = 0$ s. The cooking

experiments #1–3 (for beef chili, chicken stir-fry and paneer curry), and experiments #1–4 (for non-meat chili, tofu stir-fry and chicken curry) were performed by the same chefs (one each for each recipe type of curry, chili and stir-fry), while for cooking experiments #4–7 across all recipes, different chefs were asked to prepare the food following the same protocol to ascertain any differences in emissions due to changes in cooking behaviours like the magnitude and frequency of stirring the contents of the pan.

The food quantities were weighed using a digital electronic kitchen scale (model 835/4150; Argos, UK) while a mini-chopper (model CH180; Kenwood, UK) was used to chop ginger, garlic and chili. All cooking was performed in a stainless-steel fry pan (24 cm diameter; model 97 000, Morphy Richards, UK) over an electric hob on the back right element (1.7 kW; model KDC5422A, Beko, Turkey). The temperature of the pan surface was measured using a digital thermometer (model RS41, RS Pro, UK). During cooking, the pan was covered by a lid only if stated in the recipe protocols. The extractor fan was switched off during the cooking. Prior to evacuating the kitchen with cooked food, the hob was switched off, and dry-wiped to remove any
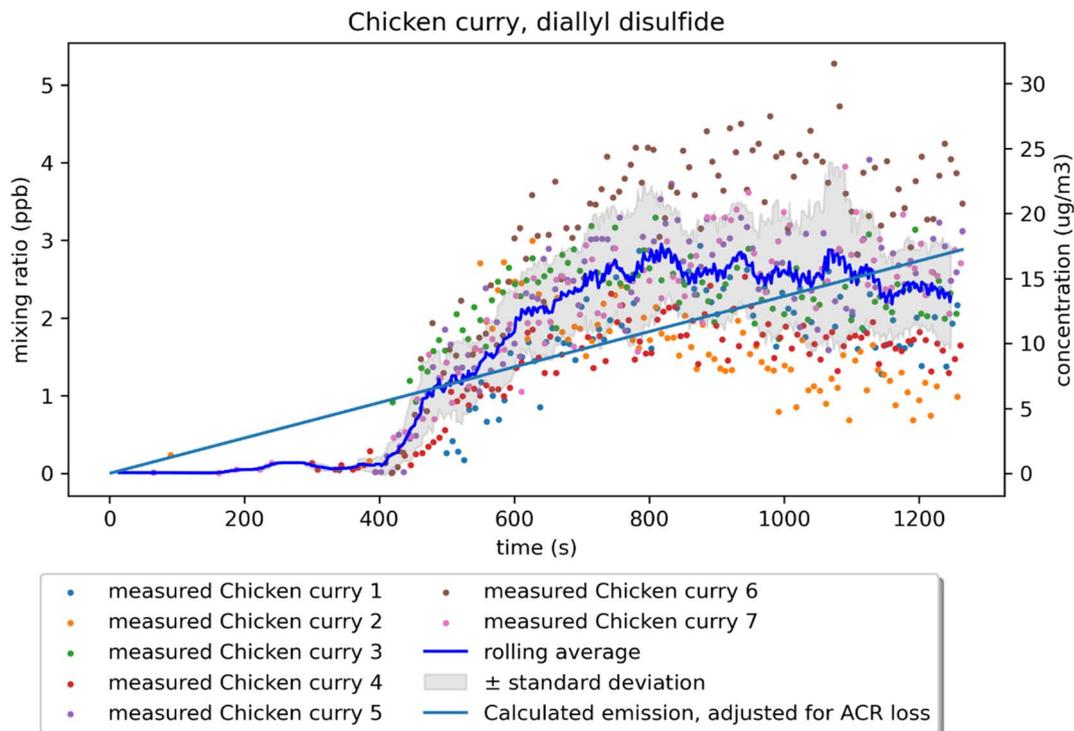
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Fig. 2 Measured mixing ratios of diallyl disulfide from seven cooks of a chicken curry. A 15-point rolling average is shown with the standard deviation of the measured concentrations. The calculated emission has been adjusted for the average 0.73 h^{-1} air change rate.

spills or food remains. Each cooking episode lasted for 12–26 min, and the post-cooking indoor VOC measurements were continued for at least 3 hours before the extractor fan was switched on automatically to ventilate the room and reduce the concentration levels to the original room background. The pans and cooking materials were thoroughly cleaned with a standard dishwashing soap and warm water and dried before each experiment.

2.3 Sampling and instrumentation

VOCs in the sample air were measured using selected ion flow tube mass spectrometry (SIFT-MS; Voice200 Ultra; Syft Technologies, New Zealand). Comprehensive details of the instrumental technique, operation settings, calibration procedures, and data analysis protocols are provided elsewhere.[14,34–36] Briefly, the SIFT-MS generates three reagent ions (H$_3$O$^+$, NO$^+$ and O$_2^+$) using a microwave discharge of air/water mixture and uses them to carry out the soft chemical ionisation of the analyte molecules in the sample air. The ionised analyte molecules are then separated by a quadrupole mass analyser and detected at their respective mass-to-charge ratios (m/z) with a dwell time of 0.1 s. The instrument was maintained and operated throughout the measurement period using the following settings: microwave ion source current at 40 mW and 300 mTorr pressure, flow tube temperature and voltage of 120 °C and 25 V respectively, 460 mTorr pressure, 1 Torr L^{-1} s^{-1} flow rate of carrier gas (nitrogen, research grade; BOC, UK). 100 mL min^{-1} of sample air was sub-sampled from the main sample line by SIFT-MS at atmospheric pressure and then analyzed in

a selected ion monitoring (SIM) mode where the compounds to be measured (listed in Table S7) were chosen based upon their occurrence in food materials and cooking processes as reported in the literature.[11] Identification and assignment of the masses to respective compounds were carried out using the instrument Labsyft software. The SIFT-MS calculates the mixing ratios of analyte compounds using the gas-phase chemical kinetics of ion–molecule reactions that take place over a well-defined reaction time.[34,35] The number density of the analyte molecule in the flow tube is determined via eqn (1). This is then converted to concentration units (ppb) using eqn (2) by considering the dilution of the sample flow in the carrier gas.

$$[\mathbf{M}] = \gamma \times \frac{[\mathbf{P}^+]}{[\mathbf{R}^+]t_{\mathrm{r}} \cdot k} \tag{1}$$

$$C_{\mathrm{M}}(\mathrm{ppb}) = 1.035 \times 10^{-10}\,[\mathbf{M}]\,\frac{T_{\mathrm{g}}}{P_{\mathrm{g}}}\left(\frac{\varphi_{\mathrm{c}}}{\varphi_{\mathrm{s}}} + 1\right) \tag{2}$$

where [M] is the number density of the analyte molecule M, γ is the instrument calibration factor (ICF), [P$^+$] is the product ion count (cps), [R$^+$] is the reagent ion count (cps), t_{r} is the reaction time (in seconds), and k is the rate constant of gas-phase reactions of reagent ions with the analyte molecule (cm^3 s^{-1}), C_{M} is the concentration of analyte M in ppb, T_{g} is the gas temperature in the flow tube (K), P_{g} is the gas pressure in the flow tube (in Torr), φ_{c} is the carrier gas flow rate, φ_{s} is the sample gas flow rate. The ICF is a factor that accounts for the ion transmission efficiency in SIFT-MS and is determined empirically during the instrument's performance check sequence using a designated
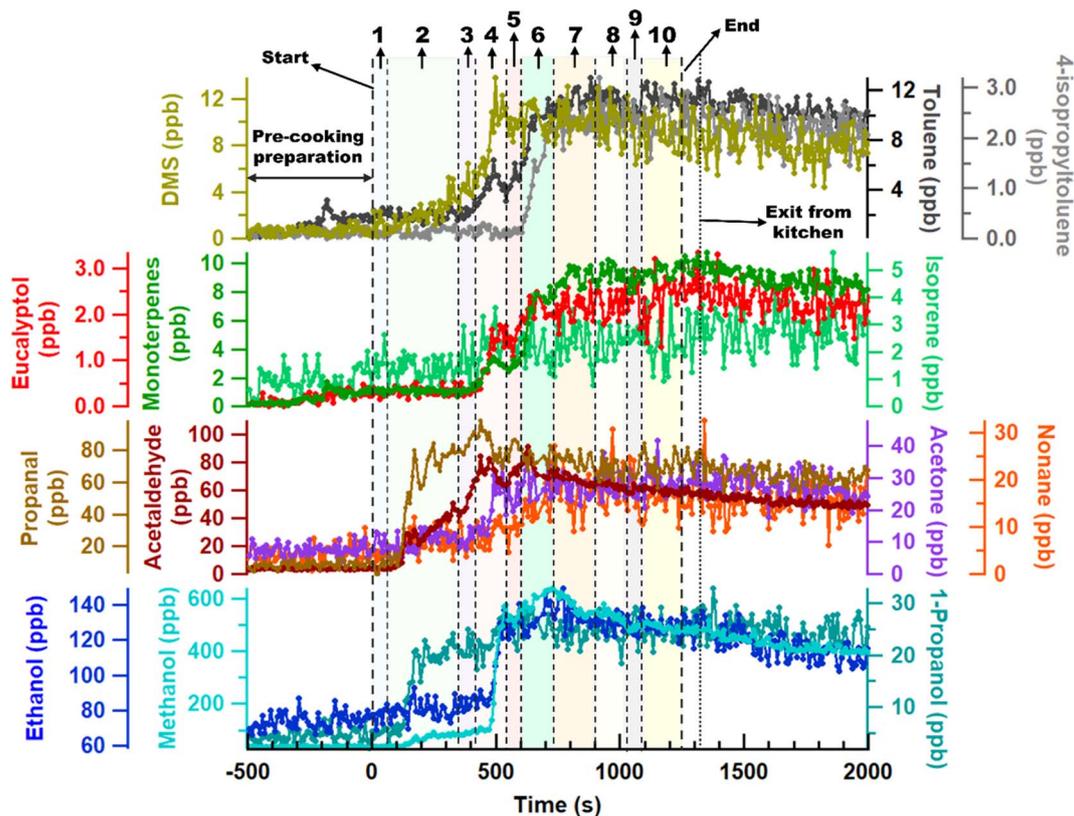
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Fig. 3 Time series of mixing ratios of selected VOCs measured during a chicken curry cooking experiment. Time = 0 s represents the time when oil was added to the hot pan and marks the start of the cooking experiment. At the end of the cooking, pan was covered with the lid and removed from the heat. Following this, the hob was switched off, and dry-wiped to remove any spills or food remains before the kitchen was exited. The shaded regions indicate the different steps of the chicken curry cooking process as following: 1: Added oil to the heated pan; 2: added onions; 3: added ginger, garlic and chili; 4: added tomatoes; 5: added spices (turmeric, cumin, coriander and cayenne); 6: added chicken; 7: added water and cooked in open pan; 8: cooked with lid closed and low heat; 9: added garam masala and salt, and stirred in open pan; 10: simmer with lid closed and occasional stirring.

validation standard. The calculation of ICF and t_r is based on the various operational parameters of the SIFT-MS such as the sample flow rate, carrier gas flow rate, rate coefficient for reaction of an analyte with reagent ion, and temperature and pressure in the flow tube reactor. The algorithm used by the SIFT-MS software (LabSyft) for reporting the concentrations is discussed in detail by Langford *et al.*[34] Isobaric compounds such as furan and isoprene (m/z 68 with NO^+ reagent ion) and monoterpenes (m/z 136 with NO^+ reagent ion) could not be differentiated in SIFT-MS. Therefore, they were reported as summed groups (furan + isoprene and total monoterpenes respectively) and are referred to as isoprene and monoterpenes henceforth. For the compounds where interferences have been reported in the literature but the exact contribution to the respective m/z channel is unknown, a correction could not be applied (example: hexanal, heptanal, octanal, nonanal, eucalyptol *etc*). Such compounds are reported with the name of the assigned compound (Table S7), however, the contributions owing to interferences are possible. The instrument was calibrated for methanol, acetonitrile, ethanol, acetaldehyde, acetone, isoprene, toluene and limonene in a calibration experiment performed prior to the measurement campaigns *via* dynamic

dilution of a custom VOC gas mixture (Table S8, cross-referenced to a primary NPL gas standard mixture (NPL, UK)). The total measurement uncertainty for these calibrated VOCs was <26%, while for the uncalibrated compounds that are reported in this work, a total measurement uncertainty of 35% can be assumed.[11,14]

Additionally, one pre and one post-cooking indoor air sample was collected per recipe in a pre-evacuated 3 L SilcoCan air sampling steel canisters (Restek, USA), which were then analysed within 6 h using a whole air canister sampling and pre-concentration GC-MS analysis (TD: TT24-7 Series 2, Markes International; GC-MS: 6850/5975C quadrupole, Agilent Technologies), for qualitatively determining the monoterpene speciation in the cooking emissions. Previous studies have shown that the VOCs remain stable within the sampling canisters for >24 h and it is expected that any loss of VOCs would be negligible.[37–41] At first, the sample gas was passed through an in-house moisture trap held at −30 °C to remove moisture. It was then preconcentrated at 0 °C for 21 min on sample traps (Tenax-TA and Carbopack-B). The traps were then thermally desorbed by heating them rapidly to 200 °C, and holding at this temperature for 5 min. The desorbed VOCs were
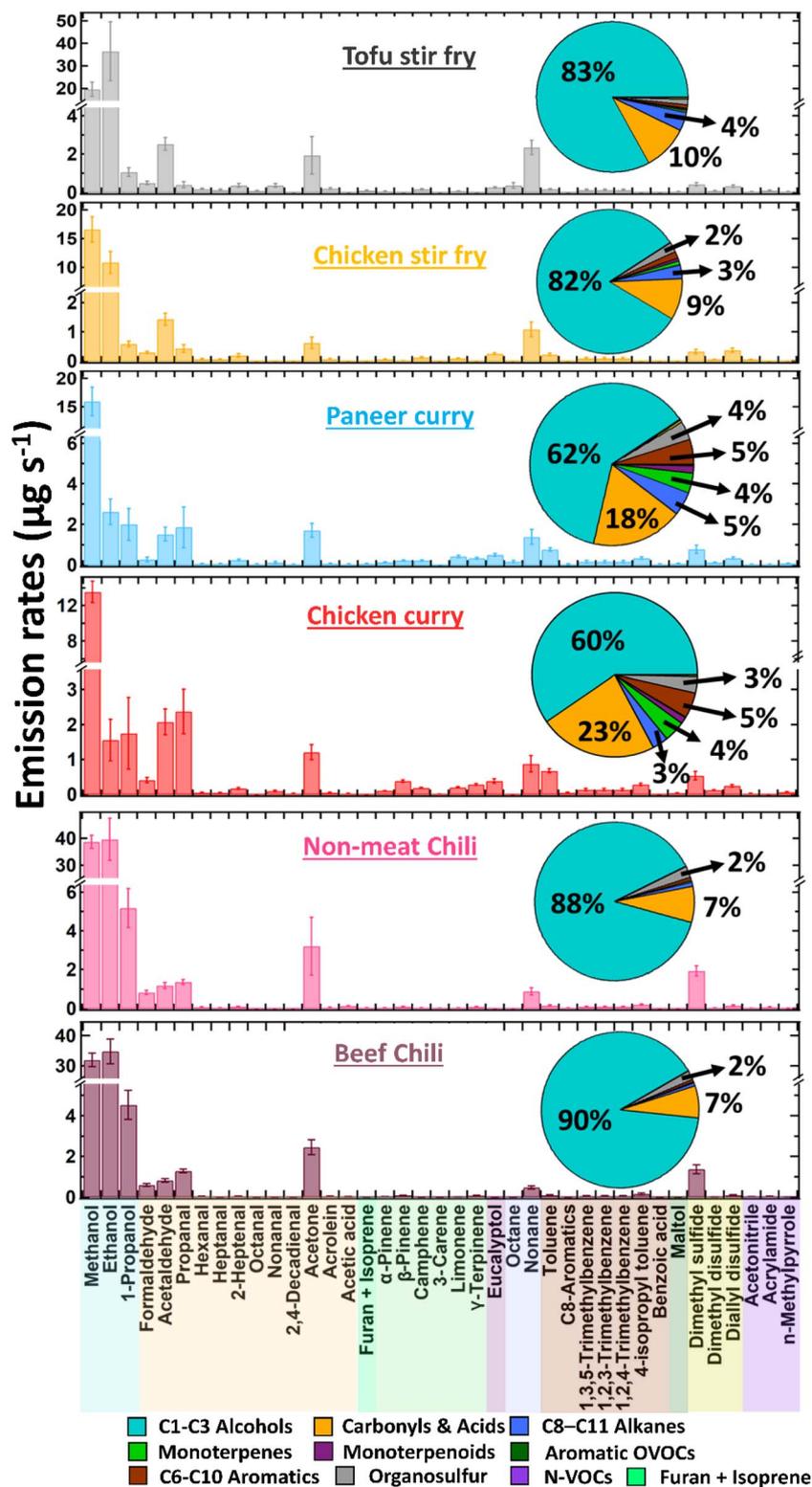
Fig. 4 Averaged constant VOC emission profiles of beef chili, non-meat chili, chicken curry, paneer curry, chicken stir-fry and tofu stir-fry derived from the scripted cooking experiments. Error bars represent the standard deviation of averaged rates (μg s^{-1}).

injected into the GC-MS at a 20 : 1 split *via* a heated inlet (130 °C) line. The GC system consisted of a single column (RTX-5, (5%-phenyl)-methylpolysiloxane, 10 m × 180 μm × 0.2 μm,

Restek) through which the analyte molecules eluted at a column flowrate of 1.5 mL min^{-1} of helium carrier gas. The GC oven temperature was programmed at 30 °C (2 min hold), then

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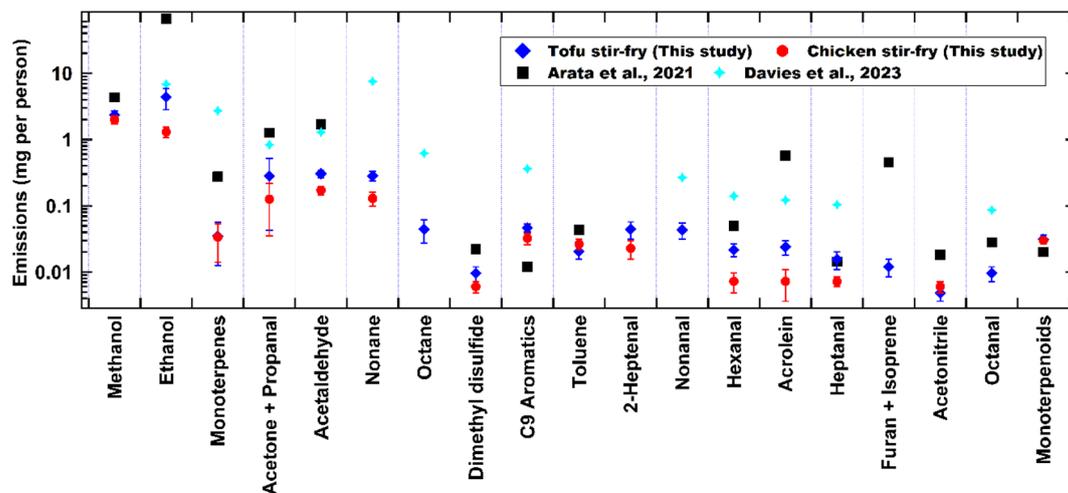This journal is © The Royal Society of Chemistry 2025

Fig. 5 Comparison of the emissions (mg per person) of VOCs from stir-fry cooking in this work and previously reported studies in literature.

ramped at 5 °C min^{-1} to 60 °C, and then at 45 °C min^{-1} to 200 °C. The identification of monoterpenes was carried out using a terpene mixture gas standard (NPL, UK). CH$_4$ was measured in real-time using Ultra-portable Greenhouse Gas Analysers (UGGA; Los Gatos Research Inc., USA) which subsampled the air from the main sample line at approximately 0.5 L min^{-1}.

2.4 Calculations

2.4.1 Air change rates (ACR). The mean air change rate was determined experimentally by the tracer gas concentration decay method.[42,43] Methane (CH$_4$) was used as the tracer gas due to its low reactivity.[44] The doors and windows were closed and the extractor fan was also switched off during the

measurements to create a "worst-case scenario" where the pollutants would accumulate indoors. The background mixing ratio of CH$_4$ in the kitchen was (1.8 ± 0.2) ppm, which was then spiked to 7–50 ppm by releasing CH$_4$ (natural gas; BOC, UK). The resultant decay in the concentration of CH$_4$ was then fitted according to eqn (3).

$$C(t) = (C_p - C_b)e^{-kt} + C_b \tag{3}$$

where, C_p and C_b are the peak and background CH$_4$ concentrations, respectively, t is the duration of decay (h), and k is the decay rate or the air change rate (h^{-1}). The average air change rate determined from these experiments was 0.73 h^{-1} (±0.10 standard error h^{-1}) (Table S9), which is similar to the average
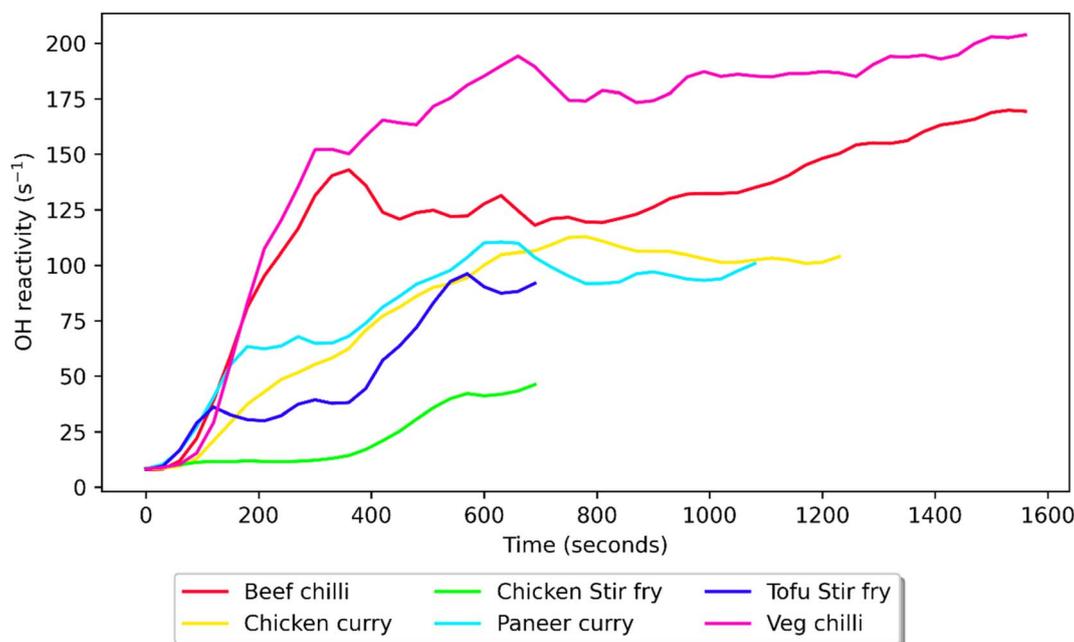

Fig. 6 OH reactivity (s^{-1}) variation with time during the cooking of each meal simulated by INCHEM-Py.
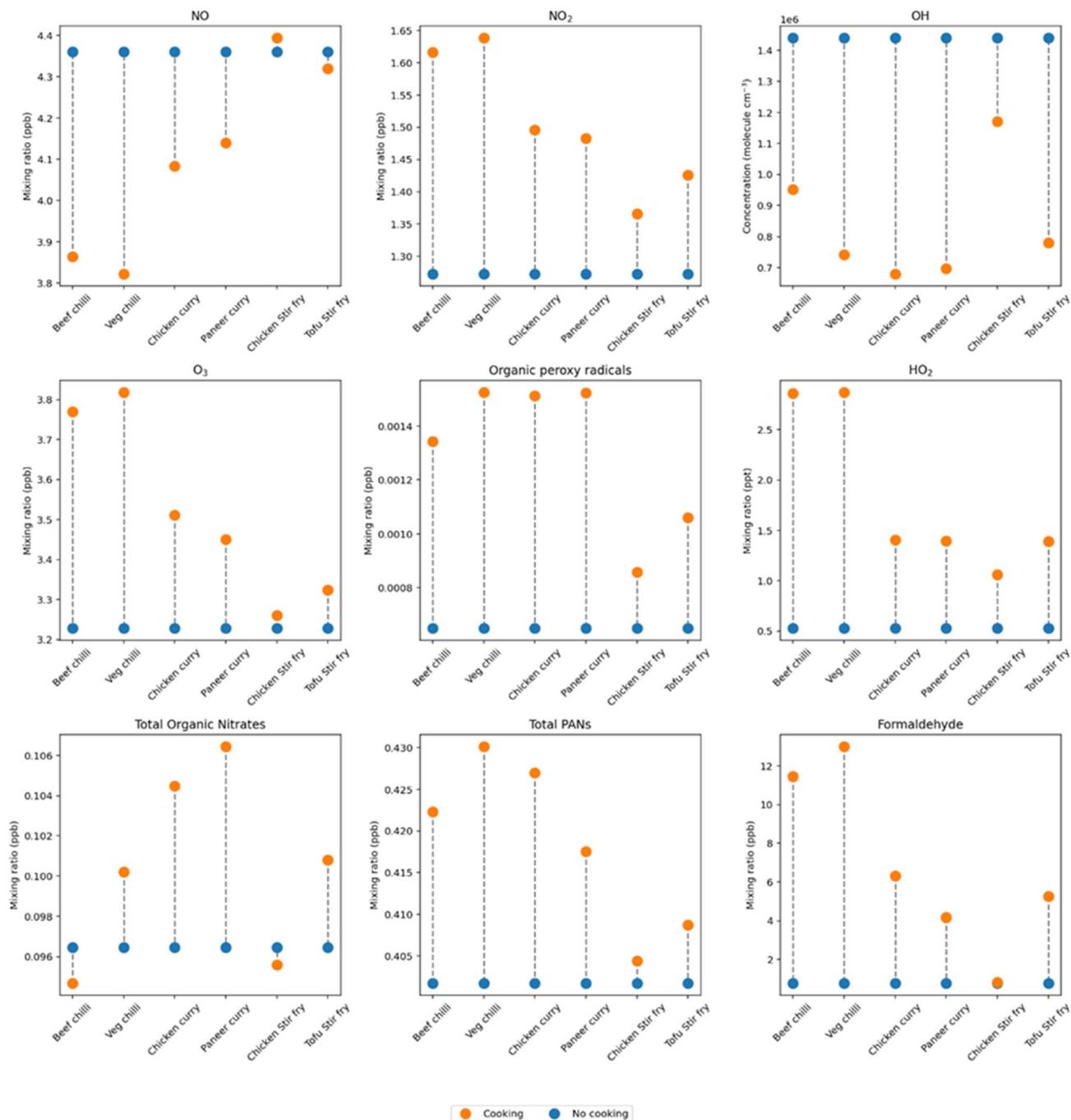
Fig. 7   The change in average simulated mixing ratios/concentrations of radical species and secondary species ozone during the cooking of the six different recipes. The background concentration for each species was simulated with no cooking activity (blue), compared to the simulated concentration during the cooking activity (orange).

air change rate for typical UK homes,[42,45] and the measured ACR of DOMESTIC kitchen reported previously with acetonitrile as the tracer molecule.[14]

2.4.2   Emission rates. The emission rates (ER, $\mu g\ s^{-1}$) for VOCs emitted from the cooking of stir fries, curries, and chillies were calculated using the total mass emission method, eqn (4).[26,46]

$$\mathrm{ER} = V \left(\frac{\left(\left(\frac{C_{\mathrm{final}}\ \Delta t k_{\mathrm{ACR}}}{3600} \right) + C_{\mathrm{final}} \right) - C_{\mathrm{initial}}}{\Delta t} \right) \quad (4)$$

where V is the volume of the DOMESTIC kitchen laboratory (21.8 m^3), C_{final} ($\mu g\ m^{-3}$) is the concentration of the measured VOC at the end of the cooking period, C_{initial} ($\mu g\ m^{-3}$), is the concentration of the VOC prior to adding oil to the pan, and Δt

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is the time difference (s) between the start ($t_{initial}$) and end (t_{final}) of the cooking period. The factor of 3600 converts k_{ACR} to s^{-1}.

Instead of individual emission rates for each experiment, an averaged emission rate was derived from multiple cooking experiments of each meal. The mixing ratios (ppb) measured by SIFT-MS were first converted into mass concentrations (μg m^{-3}) assuming 1 atm pressure and 298 K temperature. Then a background concentration of individual VOCs was calculated for the period immediately prior to commencing cooking (eqn (5)).

$$\text{LoD} = C_{avg} + 3.2\sigma \tag{5}$$

where LoD is the calculated limit of detection (μg m^{-3}), below which all measured values during the cooking period were considered to be background values, C_{avg} is the average VOC concentration during the background period immediately prior to the cooking experiment, and σ is the standard deviation of this background, which accounts for any noise in the background measurements. It is assumed that the indoor background concentration is at steady state and the indoor concentrations prior to cooking includes a constant contribution from outdoors. The LoD was then subtracted from the measurement of each VOC species' concentration and the time cropped to the period of Δt, leaving individual emission profiles starting at $C_{initial} = 0$. Use of background measurements immediately prior to the cooking period allowed us to account for any differences in the room background between experiments, given the cooking activities were conducted on different days and at different times. A VOC was considered to be emitted only if the measured concentration was above the LoD. If >75% of measured concentrations of a VOC during an experiment were below the LoD, then the emission rates for those VOC species were assumed to be negligible. The individual emission profiles for each VOC species, for each meal, were then combined into a single time series. For this, a 15-point rolling average (corresponding to roughly 2 min) was calculated from the combined measurements from each meal to determine an average emission profile across all individual experiments for the same meal. Using this rolling average, the final emission rates were then calculated according to eqn (6), which is a simplified version of eqn (4).

$$\text{ER} = V\overline{C}_{final} \frac{\left(\frac{\Delta t k_{ACR}}{3600}\right) + 1}{\Delta t} \tag{6}$$

where, $\overline{C}_{final}$ is the concentration of the rolling average at the end of the individual cooking experiments (at t_{final}).

This method of emission rate calculation makes two broad assumptions. Firstly, it assumes that the air samples being analysed are well-mixed, which is a reasonably valid assumption considering the small size of kitchen (22 m^3), the air change rate of 0.73 h^{-1}, the heat induced flow during the cooking experiment, and the location of the sample inlet. Secondly, this method assumes that the emission source strength and the air change rate primarily drive the indoor concentrations of emitted gaseous species. Given the location of the sample inlet in relation to the cooking station and that any surface emissions

will be temperature dependent and therefore consistent between cooking experiments, it is safe to assume that the indoor concentrations of the VOCs during these experiments were primarily controlled by the balance between input from cooking emissions and loss from air change with outdoors. Nevertheless, the emission rates presented here should be considered unique to the DOMESTIC kitchen and represent constant emission rates over the cooking period of each meal. As the SIFT cannot distinguish between isomers, whole air canister sampling and pre-concentration GC-MS analysis was used to speciate the monoterpenes, and the SIFT emission rate of total monoterpenes was then split fractionally into these species according to the GCMS data. The emission rate for trimethylbenzene was split equally between its three isomers. Fig. 2 shows a representative example of this methodology for the calculation of the emission rate of diallyl disulfide in chicken curry.

2.4.3 OH reactivity: model simulations and theoretical calculations. The hydroxyl radical (OH) reactivity of indoor air during recipe preparation was estimated using INCHEM-Py v1.2 which is an open-source indoor chemistry box model. The model has already been described in detail in previous studies.[14,47–50] Briefly, INCHEM-Py v1.2 formulates and solves ordinary differential equations to predict the temporal evolution of indoor species' concentrations, based on the assumption of a well-mixed environment and incorporates the master chemical mechanism (MCM)[51–54] for the atmospheric oxidation of VOCs. The model also includes additional reaction mechanisms of compounds that are specific to the indoor environments but are absent from the MCM, including for linalool, 2,5-dimethylbenzaldehyde, 2-nonenal, octanal, nonanal, decanal, chlorine, 3-carene, α-terpineol, γ-terpinene, citronellol, geraniol, geranial, dihydromyrcenol, α-terpinene, α-phellandrene and terpinolene.[49,50] The model was parameterised to the DOMESTIC kitchen laboratory, including the air change rate (0.73 h^{-1}), surface areas,[48] lighting conditions, and volume of the space (22 m^3), all detailed in Table S10. A background simulation was run where no cooking was taking place which allowed background chemical species to equilibrate in the simulated space. To simulate the cooking, starting at 1 pm, the model concentrations, for all measured chemical species available in the INCHEM-Py chemical mechanism, were constrained to the median measurement values from each time step plus the appropriate background concentration calculated as described above. The non-constrained species were able to react and vary according to the model mechanism. Concentrations of species and their rates of reaction were extracted for each time step of the model and used to calculate a total OH reactivity and the reactivity per group of species (*e.g.* aromatics, monoterpenes *etc.*).

2.4.4 Model simulations for cooking in a realistic kitchen. The production of secondary species and radicals from the cooking of different meals in a realistic kitchen was simulated using INCHEM-Py. The model was parameterized for a kitchen in a typical apartment located in urban London where outdoor pollution is relatively high in comparison to the DOMESTIC kitchen laboratory. The parameters for the kitchen surfaces and

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volume (assumed 25 m³) were adopted from previous studies to include varied surfaces that are found in typical kitchens according to literature[48] with an air change rate of 0.5 h⁻¹ and one occupant (of ~2 m² skin surfaces) to account for O_3 and H_2O_2 skin reactions and breath emissions.[14,48] In these simulations, the calculated emission rates were used after scaling to the new room volume. All model files from these model runs are included in the data attached to this publication, and further model parameters are given in Table S10.

3 Results and discussion

3.1 Emission timing and trends during cooking

A total of 39 VOCs were measured during the cooking of six recipes by SIFT-MS. Fig. 3 shows a representative real-time measurement of selected VOCs (ethanol, methanol, 1-propanol, acetaldehyde, propanal, acetone, nonane, monoterpenes, eucalyptol, isoprene, dimethyl sulfide, toluene, and 4-isopropyl toluene) measured during a chicken curry cooking experiment. Similar time series for the other recipes are shown in Fig. S1–S5. The start of the cooking period ($t = 0$ s) was determined by the addition of oil to the hot pan and the end of cooking was determined by the removal of the pan from the cooking hob.

Prior to the start of cooking, the mixing ratios of compounds like acetone, isoprene, methanol, and ethanol exhibited a minor increase at a consistent rate. These compounds have been reported in human breath emissions previously.[55] During this time, the kitchen was occupied by a single person, and the cooking ingredients were prepared. This included the slicing and chopping of fresh ginger, garlic and chilli pepper. A minor increase in the mixing ratios of dimethyl sulphide, monoterpenes and eucalyptol during this time was likely due to these pre-cooking preparations of spices, although the contributions from the occupant cannot be ruled out. Once the cooking began with the addition of oil to the hot pan, the measured concentrations for most of the VOCs rose an order of magnitude or more during the course of cooking. The concentration gradient of isoprene however was unperturbed and it increased at roughly the same rate as during the pre-cooking period, thereby suggesting that cooking of these recipes is not a major source of indoor isoprene. This was consistent with the observations of our previous work.[11] Isoprene is a common human emission[56–58] and its constant increase is consistent with presence of the cook throughout the experiment. Acetone, however, increased rapidly during cooking. Previous studies have shown significant amounts of acetone is emitted during the cooking processes.[59,60]

While different VOCs were emitted at different stages of the cooking, methanol and ethanol were emitted at the highest rates across all of the cooking experiments. During the cooking of chicken curry, the emissions of alcohols occurred at two points. First, when the onions were fried in the oil with ginger garlic, and chilli (steps 2 and 3 in Fig. 3) and secondly when the tomatoes were added to the pan (step 4). Large emissions of alcohols like ethanol and methanol have previously been reported during the cooking of vegetables[11,12,14,26] and is likely the source of these compounds here too. Emissions of alcohols were noticeably higher for tomatoes compared to onions.

During the cooking of onions in the heated oil during the initial five minutes (between 60 and 360 s), there was a gradual increase in the mixing ratios of carbonyls (acetaldehyde, propanal, and acetone), toluene, C9-aromatics, and nonane. These mixing ratios were slightly reduced at the time of the addition of the tomatoes, probably due to their high-water content, which may have reduced the temperature and arrested the emissions. While monoterpene mixing ratios increased gradually during the initial stages of cooking (until ~8 min or ~500 s), they started increasing rapidly when the spices were added (one at step 3 and the other at step 5) and only showed a minor decrease upon the addition of tomatoes and chicken. The increase in monoterpenes also coincided with the increase in the mixing ratios of eucalyptol (+ other monoterpenoids, $C_{10}H_{16}O$) and 4-isopropyl toluene, both of which have been reported to be emitted during the cooking of herbs and spices such as ginger.[11,13,61]

Amongst the organosulfur compounds, dimethyl sulfide (DMS) was the largest emitted VOC and showed its maximum increase upon the addition of garlic, consistent with previous studies.[11,62,63] A similar DMS emission trend was also observed during the cooking of paneer curry (Fig. S1). The replacement of chicken with paneer did not have any noticeable effect on the measured emissions. Another interesting observation was the difference in the levels of ethanol concentrations when onions were cooked (Fig. S6). In the chicken curry experiments #5–7, high ethanol concentrations (>2000 ppb) were observed when the onions were cooked. This was in stark contrast to chicken curry experiments #1–4 when the ethanol was only about 200 ppb during the same cooking step. This difference was most likely owing to the types of onions used. In cooking experiments #1–4, freshly chopped onions were used, while in the experiments #5–7, store bought packaged diced onions were used. Assuming that there was a span of couple of days between the packaging of the diced onions by the manufacturer and then using them for our cooking experiments, the onions had likely undergone fermentation, resulting in high ethanol emissions. To test this hypothesis, we performed an experiment of aging the onions and then measuring the emissions from cooking them 3 times over a 3 week period. The details pertaining to this experiment has been described in the SI. Fig. S7 shows the result from these aging experiments and indicates that the emissions of methanol and ethanol increased by 23 and 800 times respectively when the onions stored over for three weeks were cooked. The increased emissions of ethanol over three orders of magnitude during this duration, likely indicates that the increased ethanol emissions for the chicken curry experiments #5–7 was in fact due to the use of the packaged onions that would likely have been undergoing *in situ* fermentation from the day of their packaging. Recent studies show that the use of such ready-to-use already chopped vegetables (including onions) is on the rise because of the end-use convenience[64] and the difference in emission rates due to their usage demonstrates the cook-to-cook variability that can be induced by changes in the ingredients.

The cooking of the beef and non-meat chillies (Fig. S2 and S3) exhibited a different trend compared to curries. Methanol

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and ethanol concentrations started increasing with the addition of onions, and methanol exhibited another major increase upon the addition of tomatoes. During the chilli cooking, packaged diced onions were used and therefore the ethanol mixing ratios were higher than those observed during the preparation of the curries. Interestingly, large emissions of dimethyl sulfide and monoterpenes were observed at step 5 (in the beef chilli recipe) and step 4 (in the non-meat chilli recipe) of cooking, which was likely due to the addition of dried oregano. Unlike curries, no emissions of eucalyptol (and other monoterpenoids) were observed here, likely because ginger was not used in the chilli recipe. The carbonyls (acetaldehyde and acetone), toluene, C9 aromatics, and 4-isopropyl toluene showed similar concentration profiles in both chilli recipes and were mostly emitted during frying (steps 2 and 3). Adding the tomatoes and vegetables likely dampened further emissions of these species. Additionally, no major change in emissions was observed between the beef chilli and non-meat chilli. The cooking of beef mince (step 4 in beef chilli recipe), arrested the emissions of all measured compounds initially (likely due to a change in pan temperature and increased moisture content). However, as the beef got cooked, it started releasing small amounts of alcohols (ethanol, 1-propanol and methanol). No such trend was observed for the other VOCs.

During the cooking of stir-fries, again the maximum increase in the mixing ratios was observed for methanol and ethanol (see Fig. S4 and S5). These both increased when the vegetable mixture was added to the pan, consistent with previous observations.[12,14] Ethanol (and to a lesser extent methanol) exhibited a second increase in the mixing ratios when the sauce was added to the pan (step 8), likely arising from the sauce ingredients. Carbonyls and alkanes such as nonane increased during the heating of oil, but this increase was less pronounced than the observed by Davies *et al.*[14] Post addition of spices (ginger, garlic and chilli), several compounds like monoterpenes, dimethyl sulfide, diallyl disulfide, toluene, and 4-isopropyltoluene increased immediately, however no such increment was observed upon addition of the sauce which is also consistent with the observations of Davies *et al.*[14] High oil temperature at step 4, is likely to release more monoterpenes from the storage pools of fresh spices, which are likely depleted in the sauce during the manufacturing processes.[14] Fascinatingly, one key difference between the chicken and tofu stir fry was the emission trend of ethanol. In chicken stir fry, ethanol increased significantly upon the addition of vegetables only, however, in tofu stir fry, this increase in ethanol happened during the process of shallow frying tofu in hot oil (step 2). Shallow-frying chicken in step 2 of the stir fry recipe did not emit large amounts of ethanol compared to tofu. Although the tofu used in this study was not a fermented product, *in situ* fermentation within the packaging may have resulted in high ethanol emissions during the cooking process.

For each repeated cooking of a recipe, care was taken to maintain the consistency of the ingredients used. However, owing to the unavailability of certain items, the ingredients had to be altered from the original recipe. For example, in the stir fry experiments #5–7, the noodles used were a different brand to experiments #1–3, however, the labelled ingredients of both the noodles were the same. Similarly, instead of the store-bought mushroom stir-fry vegetable mixture, a similar sort of vegetable stir-fry mixture was prepared manually by mixing 230 g mixed pepper stir-fry mixture and 70 g sliced mushrooms. While the resulting vegetable mixture looked similar, there were some minor differences in the composition, such as the presence of red and yellow bell peppers, red cabbage, and carrots. These changes in the ingredients are likely to have a minor impact on the emissions, as the overall trend and composition of the emissions were fairly similar.

However, the major change in emissions was induced by changes in cooking behaviours. During cooking experiments #4–7 across all recipes, different people were asked to prepare the food following the same protocol. However, there were noticeable behavioural differences in the cooking processes like the magnitude and frequency of stirring the contents of the pan. Since the VOC emissions are dependent on the heating of the pan contents, the variable frequency and magnitude of the stirring likely induce differences in the cooking process and therefore the emissions. There were no noticeable differences in the temporal profile of concentrations, but the magnitude of these concentrations was however different, especially in experiments #4–7. Changes in air exchange rates of the kitchen during these experiments (arising from different ambient wind conditions) could also be a potentially significant factor. These differences are likely to arise in real-world conditions too, when the cooking is carried out using different styles, techniques and ingredients based on personal preferences. Therefore, the data presented in this work should be considered representative of real-world cooking scenarios, with the range of concentrations observed over several repeated experiments accounting for differences due to ingredients and cooking behaviours.

Fig. S8 shows the average outdoor concentrations measured for the VOCs over a period of 15 min prior to cooking. Furthermore, the statistical differences in the average outdoor concentrations between the different cooking experiments for all VOCs were ascertained using the Tukey's pairwise honestly significant difference (HSD) test, and the summary for this information is provided in Table S12. Based on the statistical test, it could be concluded with more than 95% confidence that the outdoor concentrations for almost all VOCs (except ethanol) were similar for all cooking experiments. Fig. S9 shows the average ratio between the indoor (I) and outdoor (O) mixing ratios of measured VOCs during the cooking and room background prior to cooking. In the absence of any major emission source, the concentration of the measured VOCs in the room background were similar to the outdoor ambient concentrations (a resultant I/O ratio of ~1) The I/O ratio of methanol was however consistently >1 at all times, suggesting an *in situ* emission source within the container 1 itself. A similar observation of higher methanol levels in the background of the same container was also reported by Davies *et al.*[14] and was attributed to the off-gassing from building materials. Additionally, since the experiments were carried out on consecutive days, residual emissions from previous days may have remained on surfaces within the room before off-gassing to contribute to the

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marginally higher I/O ratio of certain VOCs such as mono-terpenes and cinnamyl acetate ($C_{11}H_{12}O_2$). Previous studies have shown that terpene emissions from cooking have a complex multiphase chemistry and can partition between the gas-phase and indoor surfaces.[2] In contrast, the I/O ratio during the cooking episodes was >1 for the majority of the measured VOCs, therefore suggesting that cooking greatly perturbed the indoor concentration levels and was a major indoor source of several VOCs measured in this study.

3.2 Emission rates and composition of emissions

Fig. 4 shows the emission profiles of 39 VOCs measured during the cooking of six recipes. The pie charts represent the percentage contribution of each VOC group to the total emission rate of all measured VOC groups. The total VOC emission rates from different recipes were: non-meat chilli (94.5 ± 8.4 µg s^{-1}), beef chilli (79.1 ± 4.8 µg s^{-1}), chicken stir-fry (34.1 ± 3.0 µg s^{-1}), tofu stir-fry (68.9 ± 13.5 µg s^{-1}), paneer curry (33.1 ± 3.0 µg s^{-1}), and chicken curry (28.2 ± 1.9 µg s^{-1}). As discussed in the previous section, the emissions of ethanol were highly influenced by the type of onions used in chicken curry. Therefore, the emission rates were calculated separately for both scenarios: with packaged onions (chicken curry cooking experiments #5–7) and fresh onions (chicken curry cooking experiments #1–4) and presented separately in Fig. S10. In Fig. 4, the emissions of chicken curry are shown for experiments #1–4, *i.e.*, for the fresh onions only. All the calculated emission rates of speciated VOCs for each recipe are provided in Table S11.

Methanol and ethanol were the largest emitted VOCs across all the recipes and contributed >80% of total VOC emissions in the chilli and stir-fry recipes, and >50% of total VOC emissions in the curry recipes. While methanol was usually higher than ethanol, in tofu-stir fry and chicken curries cooked with pack-aged onions, higher ethanol emissions were measured compared to methanol. When the packaged diced onions were used in the chicken curry preparation, the fractional contribution of ethanol to the total emissions was ~60%, compared to ~6% when freshly chopped onions were used. This observation was further supported by paneer cooking experiments, where only the freshly diced onions were used and the ethanol accounted for ~8% of total VOC emissions. Interestingly, even though packaged diced onions were also used in the chilli recipes, the fractional contribution of methanol and ethanol were fairly equal (40–44%). This might be because of the larger quantity of tomatoes and beans used in chilli recipes compared to the curries, which upon cooking released larger amounts of methanol as suggested in previous studies.[14]

Carbonyl compounds like aldehydes, ketones and acids were the second largest class of VOCs measured across all recipes. These accounted for 7–23% of total measured VOC emissions, predominantly acetaldehyde (16–46% of total carbonyl emissions), acetone (18–46% of total carbonyl emissions), and propanal (6–36% of total carbonyl emissions). Hexanal, hepta-nal, octanal, nonanal, 2,4-decadienal and 2-heptenal, accounted for a total of 1–3%, 6–9% and 10–17% of total carbonyl emissions in chilli, curry and stir-fry recipes respectively. The

emissions of these compounds in stir fries (0.8–1.4 µg s^{-1}) and curries (0.8–1.0 µg s^{-1}) were comparatively higher than chili (0.1–0.3 µg s^{-1}), possibly because of the higher cooking temperatures. These compounds are known to be emitted from the heating of the oils[11,12] and higher cooking temperatures likely accelerated their emissions from stir fry and curry compared to chilli.

Monoterpenes are an important class of compounds for indoor environments because of their high reactivity and secondary aerosol formation potential.[13,65,66] The emission of monoterpenes during cooking is primarily because of the use of herbs and spices.[11,13] In this work, different rates of mono-terpene emissions were observed in different recipes. Curries emitted the highest rates of monoterpenes (1.2–1.3 µg s^{-1}), which accounted for ~4% of total VOC emissions, while chillies (0.2–0.3 µg s^{-1}) and stir-fries (~0.3 µg s^{-1}) had comparatively lower emissions (≤1% of total measured VOC emissions). The maximum quantity of spices was used in the curries (~49.5 g), compared to chili (~22.5 g) and stir-fry (~30 g), indicating that the monoterpene emissions rates were also dependent on the quantity of spices used. However, it is also to be noted that the type of spices used is important as each spice has its own monoterpene emission signature.[11] The monoterpene emission rates from chili and stir-fry were roughly the same despite the stir-fries having ~1.3 times more spices by weight compared to chili. To speciate the monoterpenes measured by SIFT-MS, whole air samples were collected offline, post-cooking experi-ments #1–4 and analysed *via* pre-concentration GC-MS. In total, seven monoterpenes were identified in the offline samples, namely limonene, α-pinene, β-pinene, camphene, 3-carene, myrcene, and γ-terpinene. Using the fractional composition of the identified monoterpenes from GC, the emission rates of total monoterpenes calculated by SIFT-MS measurements were apportioned to individual monoterpenes (Fig. S11). In the beef chilli and non-meat chilli, β-pinene (43% and 34% of total monoterpenes respectively) and γ-terpinene (42% and 29% of total monoterpenes respectively) produced the largest emission rates of monoterpenes. During the cooking of non-meat chilli, the spices (chili powder, paprika and ground cumin) were cooked for 5 minutes (~300 seconds) more than for the beef chilli (in step 2, see Fig. S2 and S3), leading to higher (~27%) total monoterpene emission rates. Monoterpene emissions from beef chilli were also arrested after the addition of beef. 3-Carene, limonene, and camphene, were nearly 4–20 times higher in non-meat chilli compared to beef chilli. The use of the same spices and cooking protocols resulted in a similar composition and magnitude of monoterpene emissions from the curries. The highest individual monoterpene emission rates in the chicken and paneer curries were: β-pinene (33% and 17% of total monoterpenes respectively), γ-terpinene (24% and 25% of total monoterpenes respectively), limonene (17% and 32% of total monoterpenes respectively) and camphene (16% and 17% of total monoterpenes respectively). In the stir-fries, the use of spices and cooking protocol was consistent between the meat and non-meat versions, and resulted in a similar monoterpene emission profile of camphene (42% in chicken stir-fry and 55% in tofu stir-fry), limonene (32% in chicken stir-fry and 25% in

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tofu stir-fry), and α-pinene (22% in chicken stir-fry and 20% in tofu stir-fry). Previous studies have reported camphene emitted in the headspace of fresh and dried ginger[10,67] and from its use in cooking of curries and heating in oils.[11] Use of ginger in curries and stir-fry likely contributed to higher camphene emissions in both recipes compared to chilli. Similarly, γ-terpinene and β-pinene were the dominant monoterpene emissions from chillies and curries, while negligible in the stir-fries. Previous studies have shown that both β-pinene and γ-terpinene are a major VOC emission from cumin[11,68,69] and the use of cumin in the chilli and curry recipes most likely contributed to their emissions.

It should be noted that the whole air sample was collected in the canister at the end of the cooking period, and is therefore a subset of the entire cooking period. The monoterpene emission signature could vary at the different cooking stages and consequently, the apportionment of the monoterpene emission rates based on the fraction measured at the end of cooking period is a limitation to this analysis. Nevertheless, this unique emission signature, particularly of reactive compounds like monoterpenes from each recipe is particularly interesting, because it provides valuable insights into the variable effects that cooking different recipes may have on indoor air chemistry (see Section 3.4).

3.3 Comparison of calculated emission rates to previous literature reports

VOC emissions from cooking are specific to the recipe or experimental conditions and difficult to generalise.[26] The emission rates can be highly dependent on the cuisine cooked, ingredients used, cooking behaviour and the quantity of food cooked, all of which need to be considered while comparing results from different studies.

Fig. 5 shows a comparison of the total emitted individual VOC mass per person during the cooking of stir-fries in this work compared to previous studies[14,26] The total measured VOC emission rates in this work was 34.1 ± 3.0 μg s^{-1} for chicken stir-fry and 68.9 ± 13.5 μg s^{-1} for tofu stir-fry, which is roughly equal to 12.3 ± 1.1 and 24.8 ± 4.9 mg per person meal respectively. In comparison, during the HOMEChem experiments, Arata et al.[26] reported a total measured VOC emission of 84.0 ± 15.0 mg per person meal from stir fries, 80% of which was comprised of ethanol (67 ± 15 mg per person meal). Ethanol was up to 17 times lower (~4 mg per person meal in chicken stir-fry and ~13 mg per person meal in tofu stir-fry) in this work. It is reported that during the HOMEChem experiments, nearly 19% of total ethanol emissions from cooking can be attributed to the dishwashing activity, which could not be differentiated from the cooking emissions due to the experimental conditions.[26] Despite accounting for these secondary contributions, the reported ethanol emissions during HOMEChem are significantly higher. This is likely due to the differences in the ingredients of stir-fry used in both studies. High ethanol emissions during the HOMEChem experiments are suspected to originate from the stir-fry sauce, in which ethanol was an active ingredient,[26] while the stir fry sauce used in this

work did not have alcohol as a labelled ingredient. Another noticeable difference between the two studies was the emission of acetic acid (3.5 mg in HOMEChem and a negligible emission in this work based on the methodology described in Section 2.4.2), which was again likely due to the differences in the composition of sauce. Interestingly, the emissions of methanol (4.3 ± 1.0 mg in HOMEChem and ~6–7 mg in this work) and monoterpenes (0.27 ± 0.06 mg in HOMEChem and ~0.1 mg in this work) were comparable between the two studies, suggesting that these emissions are strongly linked to the cooking of vegetables and spices in general rather than the sauce itself.

Lower ethanol emission rates from stir-fries compared to HOMEChem was also found by Davies et al.[14] who cooked the chicken stir-fry using similar ingredients and cooking protocols as this work. The measured emission rates in this work were similar to Davies et al.[14] except for nonane and monoterpenes which were up to 21 times higher in the Davies et al.[14] study. In Section 3.1 we noted that the cooking emissions are highly complex and susceptible to variation, despite the identical cooking protocols and ingredients. Changes in cooking behaviours may have induced these differences in the emissions of selected VOCs. Nonane and monoterpene emissions are particularly linked to the oil and spices,[11] and variable cooking behaviours during their use in the recipe might have produced the differences. Additionally, the difference in the methodology for calculating the emission rates between both studies could also contribute marginally to such differences.

3.4 Impact of cooking emissions on indoor air quality

Fig. 6 shows the temporal evolution of OH reactivity during the cooking of each recipe as simulated by the INCHEM-Py model for the conditions described in Section 2.4.3. The cooking activity significantly perturbs the OH reactivity inside the kitchen. The average simulated OH reactivity before the cooking experiments was in the range of 19.6–25.3 s^{-1}, which is approximately 3–8 times lower than the maximum simulated OH reactivity (50–200 s^{-1}) during the cooking period. A Crawford & Howell t-test analysis[70,71] shows that this increase in the OH reactivity was significant at >99% confidence ($p < 0.003$) across all recipes. The OH reactivity was estimated to be the highest during the cooking of chilli recipes, with the non-meat chilli producing the largest OH reactivity at almost all time points during the cooking. Interestingly, the OH reactivity during the cooking of chilli recipes exhibited a general increasing trend over the course of cooking, while in curries, the OH reactivity reached a plateau once the maxima (~100 s^{-1}) had been reached around 10–12 min (~600–720 s) after the cooking started. Water was added to the curries at around 12 min (~720 s) cooking time, which may have lowered the cooking temperature and arrested the emissions of VOCs. Another possible reason for this variable trend of OH reactivity across the recipes could be due to the use of the pan lid. During the cooking of curries, the pan was covered with the lid except for occasional stirring, while for stir-fries and chillies, the pan was left uncovered. In the case of stir-fries, the OH reactivity increased rapidly after 6 min (~360 s) into the cooking activity when the

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spices (ginger, garlic, and chilli) were added to the pan and continued to increase until cooking was complete.

Fig. S12 shows the fractional contribution of the different chemical classes of VOCs to the OH reactivity estimated by the INCHEM-Py. In chili recipes, alcohols were the largest contributors to the OH reactivity (~60%), followed by the carbonyls (~16%). In contrast, carbonyls contributed 33–35% and 25–33% of total OH reactivity in the curries and stir-fries respectively. This is likely because carbonyl emissions are lower from cooking chilli, but they are generally more reactive towards OH radicals than the alcohols which dominated chilli emissions. However, the contribution of alcohols to OH reactivity in tofu stir-fry was still quite high (~35%), because of their high total emissions. Compared to a steady-state calculation of OH reactivity, INCHEM-Py accounts for the changes in the OH concentration during the course of time, however, it should also be noted that the model is limited by the available chemical mechanisms (only 30 of the 39 measured species are represented in INCHEM-Py).

Fig. 7 illustrates the formation and depletion of the oxidants, radicals, and secondary products during the cooking of each meal in a realistic kitchen in an urban London house, as simulated by INCHEM-Py. For comparison, a no-cooking scenario is also shown (in blue dots). The OH radicals were consumed during all the cooking events and most prominently during the cooking of curries. As expected, the oxidation of VOCs initiated by the OH radicals also resulted in the formation of hydroperoxy radicals (HO_2) and organic peroxy radicals (RO_2). The RO_2 formation was influenced equally by curries and chilli cooking, but HO_2 formation was more pronounced for the chilli recipes. These radicals can further lead to the formation of secondary species in the presence of nitrogen oxides (NO and NO_2) that usually originate outdoors. The model simulations show that simulated NO mixing ratios were reduced (or in the case of chicken stir fry, essentially unchanged) during cooking, while NO_2 increased. This increase in NO_2 was also accompanied by an overall increase in secondary pollutants like O_3, PAN, and formaldehyde, and was the highest for chilli cooking. Interestingly, the organic nitrates decreased for beef chili and chicken stir-fry. This can possibly be explained by the complex chemistry of the radicals and oxidants. RO_2 is known to react with NO to form alkoxy radicals (RO), and organic nitrates (RNO_3) depending on the type of VOC species. This formation of RNO_3 occurs *via* a minor pathway where only about 20% of RO_2 is consumed.[14] This is further complicated by the variable fraction of reactive VOCs (such as monoterpenes) in each recipe. It has been shown before that the levels of oxidants and secondary species are highly dependent on the type and concentration of the VOCs due to the complex degradation pathways,[14,31,49] and likely resulted in the variable patterns observed in this study too. The depletion and formation of secondary products and radicals is of smaller magnitude (few ppt-ppb) compared to the primary emissions from cooking (several hundred ppb). However, the relative increase in secondary species persists for a significantly longer period-ranging from two to three times longer-than that of primary emissions.[14]

Formaldehyde in indoor environments poses significant health risks, ranging from respiratory irritation to long-term chronic effects.[72,73] The increment in formaldehyde (up to 12 ppb above background) estimated from the cooking under low ventilation conditions in this study, is not only higher than the reference concentration (RfC, 7 μg m^{-3} or 5.7 ppb) for sensory irritation suggested by the United States Environmental Protection Agency (US EPA),[74] but also is higher than the 8 hour reference exposure levels (REL, 9 μg m^{-3} or ~7.3 ppb) for non-cancerous impacts established by the California Office of Environmental Health Hazard Assessment (OEHHA).[75] At high concentrations, other carbonyl compounds like aldehydes are also known to irritate the eyes and respiratory tracts.[12,76] IARC categorizes acetaldehyde as a group 2b carcinogen with a 0.003 mg m^{-3} of chronic exposure limit.[12,77] During the cooking of the recipes discussed here, the average acetaldehyde concentration during the cooking period was 0.028–0.090 mg m^{-3}, exceeding the prescribed limit by a factor of 9–30. It is worth noting though that these exceedances are only for a short duration (few minutes to hours), and the reference values are established for a constant exposure over a prolonged period of time. Similarly, VOCs like hexanal, and 2,4-decadienal, and secondary species like PAN and organic nitrates, all have been suggested to be harmful to humans.[12,15,78,79] Unfortunately, prescribed exposure limits for many of these compounds and a comprehensive assessment of their exposure to human health are rarely studied.

4 Conclusion

In summary, this study presents detailed VOC emission rates for the full recipe cooking of common recipes like chilli, curry, and stir-fry from a total of 39 cooking experiments. 39 VOCs were quantified in real-time using SIFT-MS, which also allowed us to capture the dynamic emission signatures due to individual ingredients and the cooking process. Significant emissions of alcohols (methanol and ethanol) were observed in all the recipes primarily due to the cooking of vegetables like onions and tomatoes. The use of packaged diced onions was observed to cause high emissions of ethanol likely due to *in situ* fermentation within the packaging. Unique emission signatures were observed for each recipe based on the composition of alcohols, aldehydes, and monoterpenes. There were however, no significant changes in the emission patterns between a vegetarian and a non-vegetarian version of each recipe, apart from more ethanol observed in tofu stir-fry compared to chicken stir-fry. Substantial emissions of aldehydes and monoterpenes across all recipes had a profound effect on indoor air chemistry and secondary pollutant formation. Notable emission of monoterpenes and their distinct emission patterns for each recipe influenced the indoor air quality diversely depending upon the recipe cooked and the existing levels of oxidant species in the indoor environment. Use of more spices in curries likely resulted in higher production of secondary species compared to chili and stir-fry. Under higher ventilation conditions, secondary species are generally expected to be lower in concentration, although it may depend on the

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ambient background concentrations as well. These findings are also relevant to occupational culinary environments, including commercial restaurant kitchens, where the extensive use of spices and large-scale cooking operations may elevate concentrations of secondary pollutants, thereby indicating the requirement of suitable mitigation strategies like higher ventilation. Furthermore, the outflux of these indoor cooking emissions to the outdoor environments also may affect the ambient air quality. The results from this study will be a valuable addition to the existing database of VOC emissions from cooking processes and also boost future modelling studies that investigate the various dynamic processes influencing indoor air quality and design mitigating strategies to lower health exposures.

Author contributions

Ashish Kumar: methodology, investigation, formal analysis, data curation, writing – original draft, writing – review and editing, visualisation. Catherine O'Leary: methodology, investigation, formal analysis, data curation, writing – review and editing. Ruth Winkless: methodology, investigation, writing-review & editing. Wael Dighriri: methodology, investigation, formal analysis, writing-review & editing. Marvin Shaw: methodology, investigation, writing – review and editing. David Shaw: methodology, software, investigation, data curation, formal analysis, validation, visualisation, writing-original draft, writing-review & editing. Terry Dillon: conceptualisation, methodology, investigation, writing-original draft, writing-review and editing, supervision. Nicola Carslaw: conceptualisation, methodology, investigation, visualisation, writing-original draft, writing-review & editing, project administration, funding acquisition.

Conflicts of interest

There are no conflicts to declare.

Data availability

The INCHEM-Py model described in the work is available as an open-source model. All the INCHEM-Py model data files used in this work are available for download from the research data repository of the University of York at https://doi.org/10.15124/c5585507-4c30-4f22-85b4-1b46fb738822.

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