SUPPLEMENTARY INFORMATION: A measurement and modelling investigation of the indoor air chemistry following cooking activities

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1 DOMESTIC Container Layout

Figure S1 shows the layout of the DOMESTIC container where cooking experiments took place.

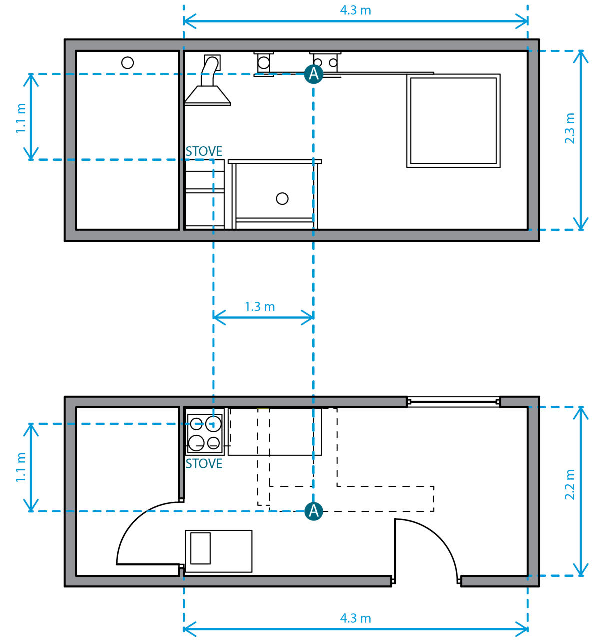

Figure S1: Section and plan views of the DOMESTIC facility. The SIFT-MS inlet position is marked by A. The door to the bathroom on the left side of the diagram was kept shut during experiments.

2 SIFT-MS

The VOCs measured using each reagent ion in the SIFT-MS method are shown in Table S1, along with the species molecular weights, product ions, rate coefficients and branching ratios. Whether or not detection of a species on that particular reagent ion was used for quantification is also shown in the 'Included in Analysis' column. Table S2 shows the species that were measured by the SIFT during the DOMESTIC campaign, their limits of detection (LOD), and whether or not the species are calibrated against a gas standard.

Table S1: The compounds measured by the SIFT-MS and their corresponding reagent ions, molecular masses (MM) and product ion chemical formulae

Reagent Ion	Compound	MM (g mol^{-1})	Product Ion	Reaction Rate (cm^3 molecules^{-1} s^{-1})	Branching ratio (%)	Included in Analysis?
H3O+	Formaldehyde	31	CH_3O^+	3.4×10^{-9}	100	✓
		49	$H_2HCO \cdot H^+ \cdot H_2O$	3.4×10^{-9}		
	Methanol	33	CH_5O^+	2.7×10^{-9}	100	✓
	Acetonitrile	42	$CH_3CN \cdot H^+$	5.1×10^{-9}	100	✓
		60	$CH_3CN \cdot H^+ \cdot H_2O$	5.1×10^{-9}		
	Acetaldehyde	45	$C_2H_4O \cdot H^+$	3.7×10^{-9}	100	✓
	Ethanol	47	$C_2H_7O^+$	2.7×10^{-9}	100	✓
	Nitrous Acid	48	$H_2NO_2{}^+$	2.7×10^{-9}	33	
	Propanal[a]	59	C3H7O+	3.6×10^{-9}	100	✓
	Acrylamide	72	$C_2H_3NH_2CO \cdot H^+$	2.1×10^{-9}	100	✓
	N-methylpyrrole	82	$C_5H_7N \cdot H^+$	3.0×10^{-9}	100	✓
	Hexanal	101	$C_6H_{13}O^+$	3.7×10^{-9}	95	✓
		119	$C_6H_{13}O^+ \cdot 2\,H_2O$	3.7×10^{-9}		
	2,5-dimethylpyrazine	109	$C_6N_2H_8 \cdot H^+$	3.4×10^{-9}	100	
	Heptanal	115	$C_7H_{15}O^+$	4.7×10^{-9}	80	✓
	Benzoic Acid	123	$C_7H_6O_2 \cdot H^+$	3.0×10^{-9}	100	✓
	Maltol	127	$C_6H_6O_3 \cdot H^+$	4.0×10^{-9}	100	✓
		145	$C_6H_6O_3 \cdot H_3O^+$	4.0×10^{-9}		
	Octanal	129	$C_8H_{17}O^+$	3.8×10^{-9}	85	✓
	Total Monoterpenes	137	$C_{10}H_{17}{}^+$	2.6×10^{-9}	30	
		155	$C_{10}H_{17} \cdot H_2O^+$	2.6×10^{-9}		
	Nonanal	143	$C_9H_{19}O^+$	2.5×10^{-9}	86	✓
	2,4-decadienal	153	$C_{10}H_{17}O^+$	4.9×10^{-9}	100	
		171	$C_{10}H_{17}{}^+ \cdot H_2O$	4.9×10^{-9}		
	Decane	161	$H_3O^+ \cdot C_{10}H_{22}$	1.6×10^{-9}	100	✓
	Methyl Cinnamate	163	$C_{10}H_{10}O_2 \cdot H^+$	3.4×10^{-9}	100	✓
		181	$C_{10}H_{10}O_2 \cdot H^+ \cdot H_2O$	3.4×10^{-9}		
	Total Sesquiterpenes	205	$C_{15}H_{25}{}^+$	2.5×10^{-9}	64	✓
NO+	Acrolein	55	$C_3H_3O^+$	1.6×10^{-9}	60	
	1-propanol	59	$C_3H_7O^+$	6.3×10^{-10}	59	✓
		77	$C_3H_7O \cdot H_2O^+$	6.3×10^{-10}		
	Furan/Isoprene	68	$C_4H_4O^+$	1.7×10^{-9}	100	✓
	Benzene	78	$C_6H_6{}^+$	1.5×10^{-9}	55	✓
	Acrolein	86	$C_3H_4O \cdot NO^+$	2.7×10^{-9}	40	✓
	Acetone[b]	88	NO+.C3H6O	1.0×10^{-9}	100	✓
	Acetic Acid	90	$NO^+ \cdot CH_3COOH$	9.0×10^{-10}	100	✓
	Toluene	92	$C_7H_8{}^+$	2.2×10^{-9}	100	✓

	Dimethyl Disulfide	94	$(CH_3)_2S_2{}^+$	2.4×10^{-9}	100	✓
	Hexanal	99	$C_6H_{11}O^+$	2.5×10^{-9}	100	
	Benzoic Acid	105	$C_7H_5O^+$	3.0×10^{-9}	60	
	m-Xylene (C2-alkyl benzenes)	106	$C_8H_{10}{}^+$	2.0×10^{-9}	100	✓
	2-heptenal	111	$C_7H_{11}O^+$	3.9×10^{-9}	85	✓
	Trimethylbenzene (C3-alkyl benzenes)	120	$C_9H_{12}{}^+$	1.9×10^{-9}	100	✓
	Maltol	126	$C_6H_6O_3{}^+$	2.5×10^{-9}	100	
	Cinnamaldehyde	132	$C_9H_8O^+$	2.0×10^{-9}	100	
	Total Monoterpenes	136	$C_{10}H_{16}{}^+$	2.2×10^{-9}	75	✓
	Diallyl Disulfide	146	$(C_3H_5)_2S_2{}^+$	2.4×10^{-9}	100	✓
	2,4-decadienal	151	$C_{10}H_{15}O^+$	4.2×10^{-9}	80	✓
	Eucalyptol	154	$C_{10}H_{18}O^+$	2.4×10^{-9}	94	✓
	Undecane	155	$C_{11}H_{23}{}^+$	3.8×10^{-9}	84	✓
	Methyl Cinnamate	162	$C_{10}H_{10}O_2{}^+$	1.4×10^{-9}	100	
		181	$C_{10}H_{12}O_2 \cdot H^+$	1.4×10^{-9}		
	Eugenol	164	$C_{10}H_{12}O_2{}^+$	2.4×10^{-9}	100	✓
	2-ethyl-2,5,-dimethylpyrazine	166	$C_8H_{12}N_2 \cdot NO^+$	3.0×10^{-9}	15	✓
	Cinnamyl Acetate	176	$C_{11}H_{12}O_2{}^+$	3.0×10^{-9}	100	✓
	Total Sesquiterpenes	204	$C_{15}H_{24}{}^+$	2.0×10^{-9}	38	
O2+	Nitrogen Dioxide	46	$NO_2{}^+$	6.2×10^{-10}	100	✓
	Furan/Isoprene	68	$C_4H_4O^+$	1.6×10^{-9}	100	
	Octane	85	$C_6H_{13}{}^+$	1.6×10^{-9}	50	✓
	Nonane	99	$C_7H_{15}{}^+$	2.1×10^{-9}	10	✓
	2,5-dimethylpyrazine	108	$C_6N_2H_8{}^+$	2.7×10^{-9}	100	✓
	Dimethyl Trisulfide	111	$CH_3S_3{}^+$	2.2×10^{-9}	15	✓
	Maltol	126	$C_6H_6O_3{}^+$	2.5×10^{-9}	100	
	Cinnamaldehyde	132	$C_9H_8O^+$	2.0×10^{-9}	100	✓
	Cinnamyl Acetate	134	$C_9H_{10}O^+$	1.5×10^{-9}	100	
	Undecane	156	$C_{11}H_{24}{}^+$	3.2×10^{-9}	31	✓
	Eugenol	164	$C_{10}H_{12}O_2{}^+$	1.9×10^{-9}	100	

[a] Propanal calculated by subtracting the acetone measured at NO^+ mass 88 from the combined propanal and acetone measured at H_3O^+ mass 59. [b] Acetone calculated by subtracting the limonene measured at NO^+ mass 136 from the combined acetone and monoterpenes measured at NO^+ mass 88.

Table S2: Species identified by the SIFT-MS, their limits of detection, and whether or not their concentrations were calibrated against a gas standard

Species	Limit of Detection (ppb)	Calibrated?
Acetaldehyde	1.80	
Propanal	1.10	
Acrolein	0.45	
Hexanal	0.75	
2-Heptenal	0.12	
Heptanal	0.48	
Octanal	0.24	
Nonanal	0.30	
Octane	1.20	✓
Nonane	4.40	✓
Decane	0.26	
Undecane	0.21	
Total monoterpenes	0.20	✓[a]
Total sesquiterpenes	0.92	
Methanol	1.20	✓
Ethanol	2.20	✓
1-Propanol	0.89	
Benzene	0.24	✓
Total C2 alkyl benzenes	0.20	✓[b]
Total C3 alkyl benzenes	0.33	✓[c]
Benzoic acid	0.22	
Dimethyl trisulfide	1.90	
Diallyl disulfide	0.12	
Dimethyl disulfide	0.19	
Acetonitrile	0.32	✓
Acetone	0.52	✓
Nitrogen dioxide	3.80	
Acetic acid	1.20	
Maltol	0.18	
Eucalyptol	0.12	
Furan + isoprene	0.11	✓[d]
N-methylpyrrole	0.07	
Acrylamide	0.19	
2,4-Decadienal	0.06	

[a] Limonene used as calibration gas. [b] m-xylene used as calibration gas. [c] 1,2,4-trimethylbenzene used as calibration gas. [d] Isoprene used as calibration gas.

3 SIFT Uncertainties

The uncertainties for uncalibrated species measured by the SIFT is taken to be ±35%, as recommended in Syft training resources [1], and [2]. However, for calibrated species, the uncertainties in the measurements are generally smaller, and are attributable to the calibration process.

For calibration, known concentrations of calibrant species are measured by the SIFT, and the relationship between the calibrant concentration and the measured concentration is assessed using linear regression, as follows:

$$C_{c,m} = \nabla C_m + y_i \tag{1}$$

where $C_{c,m}$ is the calibrated measured concentration, C_m is the raw measured concentration, ∇ is the gradient of the fitted line, which equates to the calibration coefficient, and y_i is the y-intercept which corresponds to concentration-independent shift in the measured data such that the measurement is 0 when no calibrant is being input.

The resulting overall uncertainty is then calculated by combining the standard errors of ∇ and y_i from the linear fit, and the uncertainty in the calibration equipment (gas standard concentration and the gas blenders). Therefore, total uncertainty is

$$U_s = \left[C_{c,m} \times \sqrt{\left(\frac{SE_\nabla}{\nabla}\right)^2 + RSE_{blend}^2 + RSE_{gas}^2} \right] + SE_{y_i} \tag{2}$$

Where $C_{c,m}$ is the calibrated measured concentration, SE_∇ is the standard error of ∇, RSE_{blend} is the relative uncertainty of the gas blenders (±2%), RSE_{gas} is the relative uncertainty of the gas standard (±5%), and SE_{y_i} is the standard error of the y-intercept. The relative errors are added in quadrature and multiplied by the measured concentration then summed with SE_{y_i} to give a total uncertainty in ppb. The absolute error on any measured point is, therefore, $C_{c,m} \pm U_s$.

Values for the calibration factors, y-intercepts and their associated errors for each of the calibrated species are given in Table S3.

Table S3: Calibrated SIFT species error analysis values

Species	∇ ($\pm SE_\nabla$)	y_i ($\pm SE_{y_i}$)
Octane	0.7513 (±0.0094)	-0.8698 (±0.0828)
Nonane	1.0749 (±0.0494)	-2.4658 (±0.3470)
Total monoterpenes	5.3003 (±0.0752)	0.2873 (±0.1522)
Methanol	1.3385 (±0.0239)	-1.5906 (±0.1275)
Ethanol	1.5542 (±0.0474)	-8.4624 (±0.4091)
Benzene	1.1375 (±0.0078)	-0.1230 (±0.0413)
Total C2 alkyl benzenes[a]	1.4639 (±0.0150)	0.0824 (±0.0603)
Total C3 alkyl benzenes	1.6809 (±0.0336)	0.3394 (±0.1106)
Acetonitrile	1.1942 (±0.0093)	-0.3278 (±0.0482)
Acetone	2.3656 (±0.0373)	-0.6599 (±0.1008)
Furan + isoprene	2.2139 (±0.0160)	0.0079 (±0.0429)

Errors are calculated from fits using calibration data from 4 days when cooking experiments were carried out. [a]Errors for total C2 alkyl benzenes were calculated using only 3 calibrations, as a C2 alkyl benzene calibration was not performed on one of the cooking days.

4 Air Change Rate

The air change rate in the DOMESTIC facility was measured during the campaign using acetonitrile tracer releases. An example fitting of an acetonitrile decay is shown in Figure S2 for one of the cooking-only days on the campaign. The background acetonitrile concentration was subtracted from the calibrated acetonitrile concentration $(C_t - C_b)$, then the natural log was taken on the resulting concentration. After the release, the initial 10 minute mixing period was discounted, then a decay curve was fitted to the next 2 hour period, shown by the green shaded area in Figure S2.

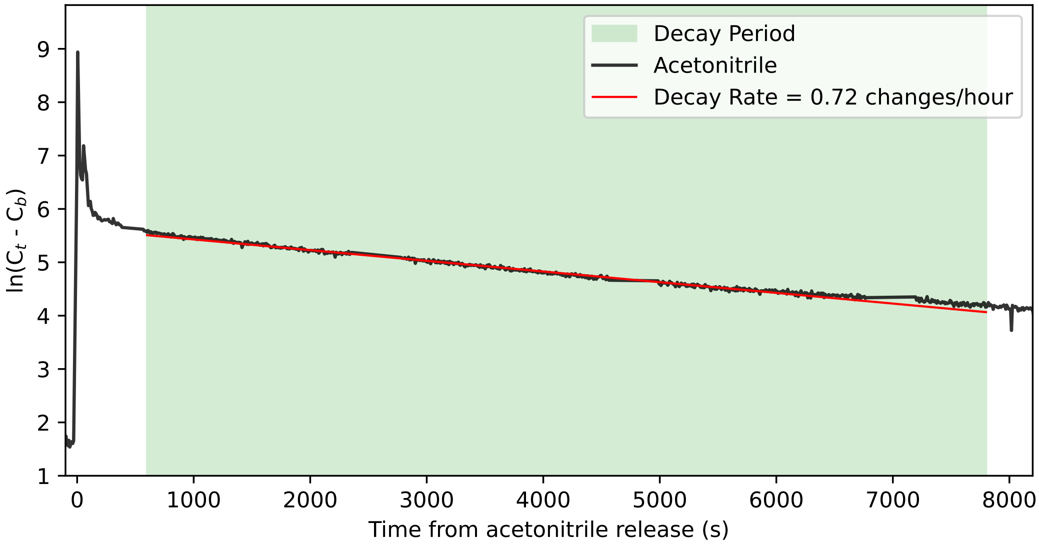

Figure S2: Fitting a log-linear decay curve to acetonitrile concentration data to determine air change rate (ACR) during the DOMESTIC campaign.

5 Experimental Reproducibility

Figure S3 shows the mixing ratios of three representative species that are emitted during the stir-fry cooking process, measured using SIFT-MS. The data from cooking-only days are shown by the red and orange markers, and data from layered days are shown by the light and dark blue markers. The green shaded area signifies the cooking period from t0 to the point when the pan was removed from the kitchen. In general, the data are very reproducible, with peaks appearing at the same time points in the cooking process for each of the days, even though the mixing ratios vary between repeats.

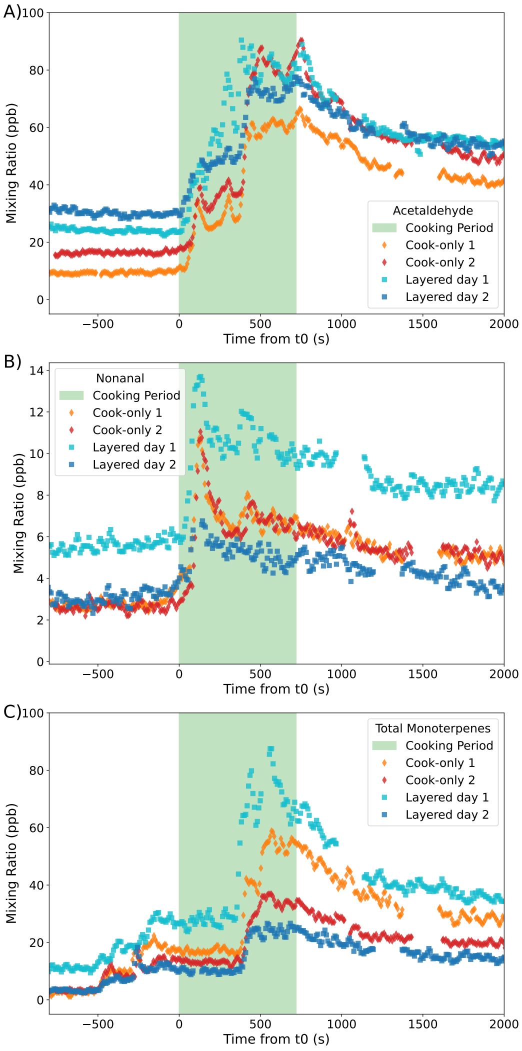

Figure S3: Mixing ratios of acetaldehyde (A), nonanal (B) and total monoterpenes (C) measured by SIFT-MS to assess the reproducibility of stir-fry emissions on multiple days. ‘Cook-only’ refers to days when only the stir-fry was cooked and the room was undisturbed for the rest of the day. ‘Layered day’ refers to days when the stir-fry was cooked as part of a series of sequential cooking and cleaning activities in the DOMESTIC container.

6 Obtaining Experimental Emission Rates

Emission rates are fitted manually to the SIFT-MS data, whereby the concentrations at the start and end of an emission are chosen, and the gradient calculated and taken to be the emission rate. Examples of fitted emissions are shown in Figure S4 for three representative VOCs emitted by the average stir-fry from the two cooking-only days, as described in the manuscript.

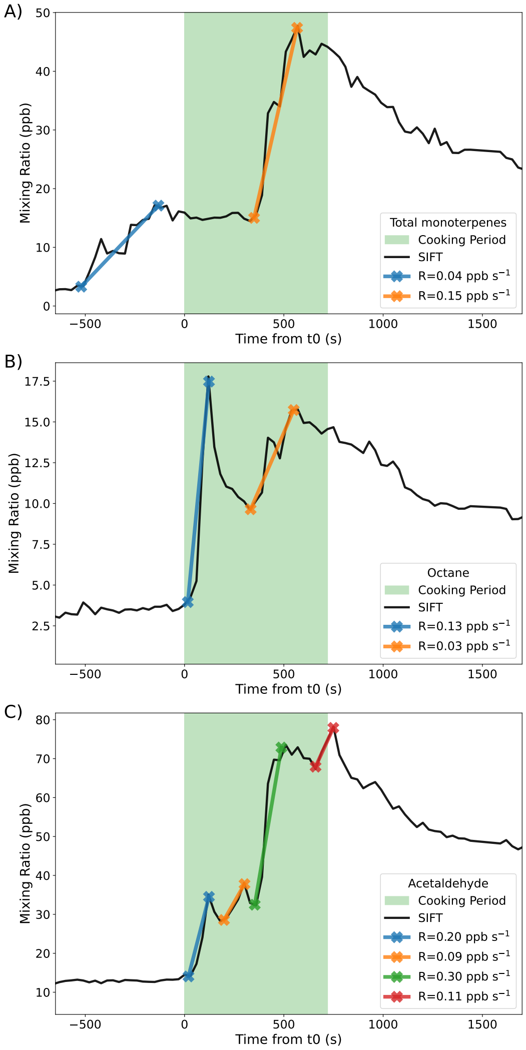

Figure S4: Concentrations of total monoterpenes (A), octane (B) and acetaldehyde (C) for the average stir-fry (average SIFT-MS data for the two cooking-only days). Emission rates (coloured lines) are determined from the gradient of peaks and are shown in the legend. This protocol was used for the other species emitted, but not shown here. The shaded green area shows the time period where cooking takes place.

7 Simulated Cooking Emissions

The species included as emissions are shown in Figure S5, which shows the changes in concentration of individual VOCs, and the total concentrations of the different chemical groups. The seven aldehydes measured by the SIFT-MS that increase in concentration during cooking are shown in two groups for clarity. The groups are ≤C3 aldehydes (acetaldehyde, propanal and acrolein) and >C3 aldehydes (hexanal, heptanal, octanal and nonanal). After the cooking period (shown by the purple shaded area), each of the VOCs decay away significantly in the 3 hour period post-cooking (green shaded area). The combination of these experimentally-derived emissions and the average kitchen properties constitutes the base case cooking scenario.

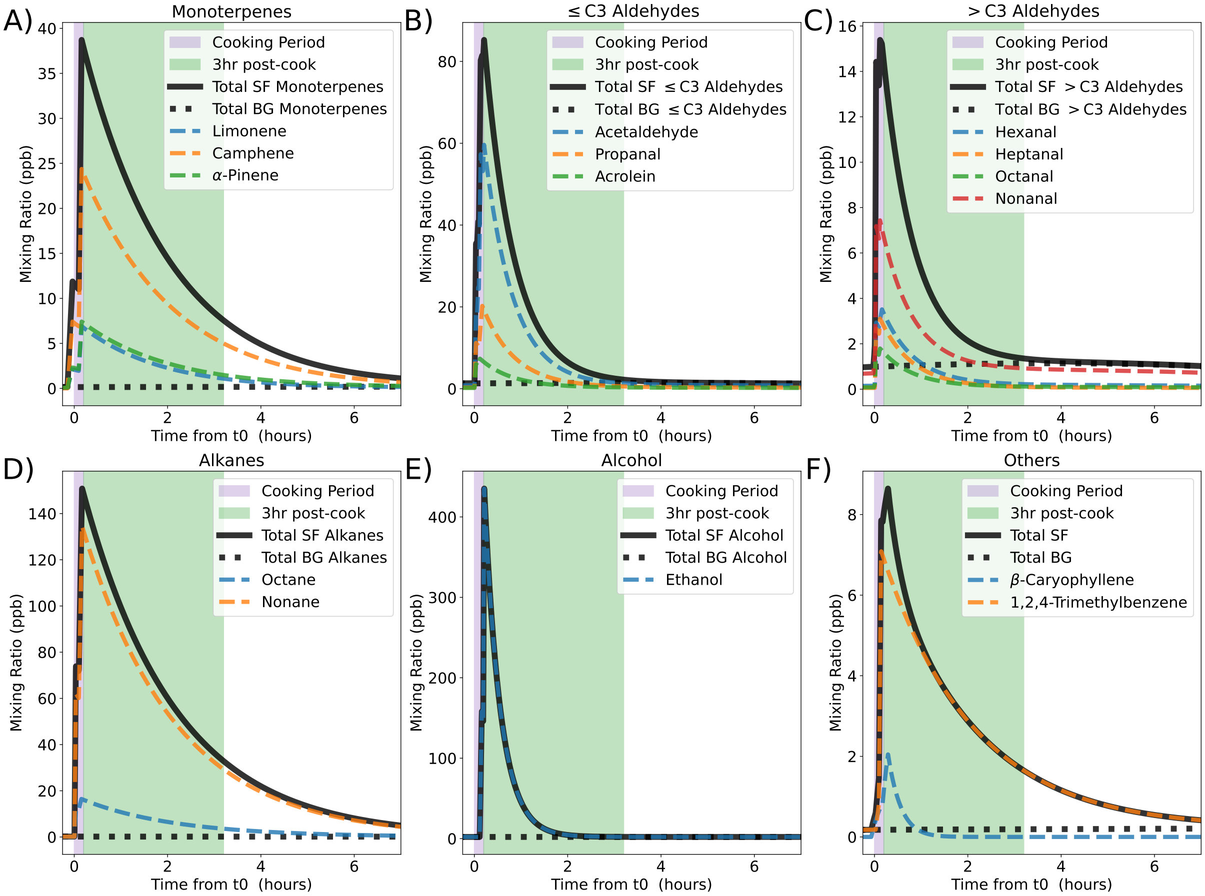

Figure S5: Mixing ratios versus time for the species used to simulate the cooking of an average stir-fry. Each plot shows a different group of species, where the black solid line shows the total mixing ratio of the group during cooking (total SF), and the black dotted line shows the total background mixing ratio when no cooking takes place (total BG). The coloured dashed lines show the individual species in each of the groups. ≤C3 and >C3 aldehydes are shown separately for clarity.

8 Methanol Emissions

As discussed in the paper, methanol concentrations were extremely high in the DOMESTIC facility during the campaign, likely due to off-gassing from the relatively new materials used in the container. Therefore, methanol was excluded from the simulations. This was acceptable as simulation tests where methanol emissions were included or not showed that the inclusion of methanol emissions made very little difference to the secondary chemistry, and the formation of secondary species.

The formation of various secondary products following cooking in the base case scenario (shown in Table 3, main article) when methanol emissions were included (Inc. CH_3OH) or excluded (Exc. CH_3OH) is shown in Figure S6. The small pink circles show the background mixing ratios when no cooking takes place, and the larger purple circles show the average species mixing ratios when cooking emissions are included.

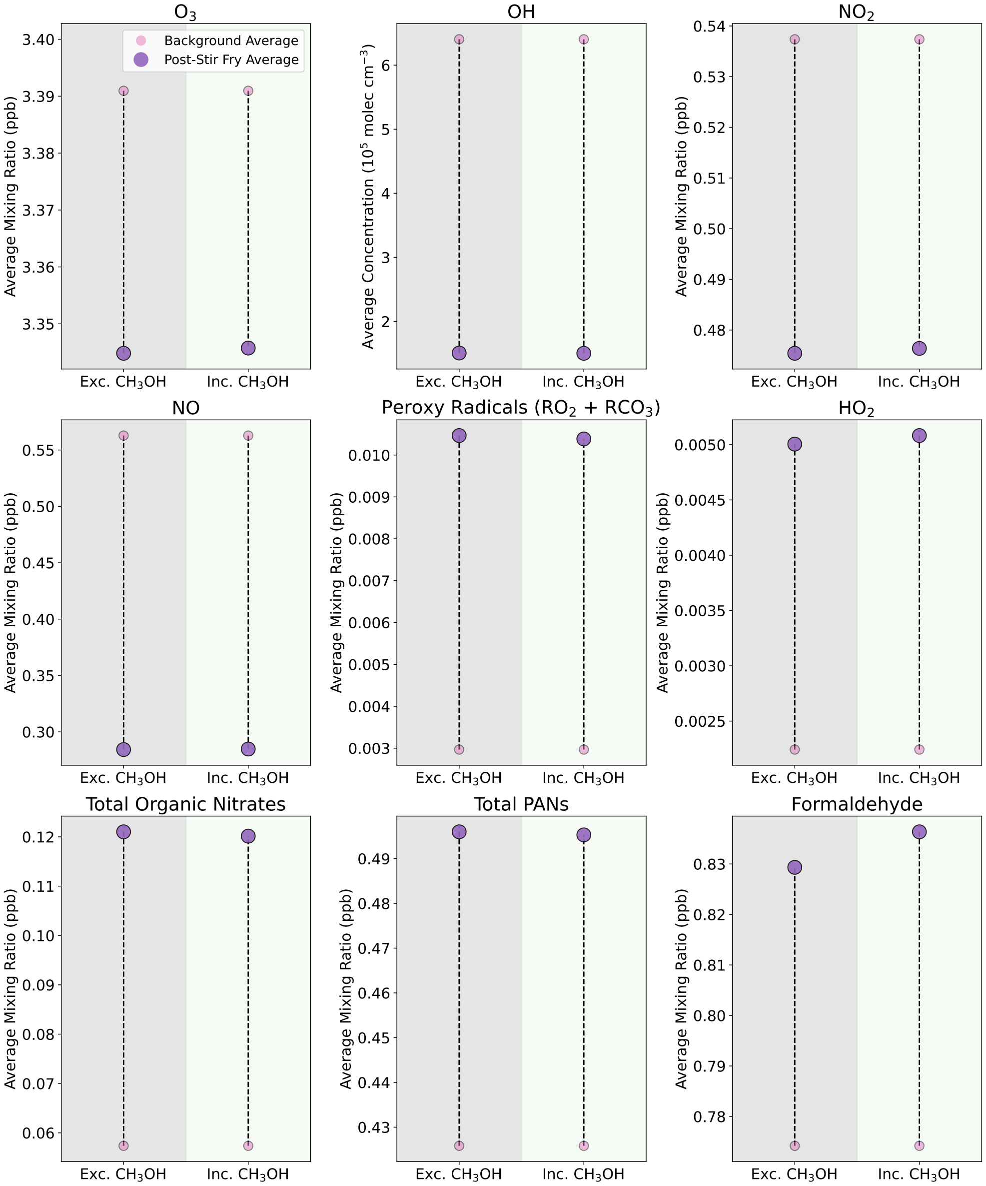

Figure S6: Effect of including or excluding methanol on the formation of secondary products. Smaller pink circles show the average background mixing ratio, and the larger purple circles show the average mixing ratio when a stir-fry is simulated. In both cases the average is calculated over a 3 hour period, starting 15 minutes before t0.

9 Relative Effects of Cooking on Primary and Secondary Species

To see the relative effect of cooking on species that are emitted directly or produced via secondary chemistry, the total concentration of the 15 modelled species emitted during cooking was calculated and normalised, so that the background concentration is 0, and the maximum total concentration is 1. A similar summing and normalising process was taken for the secondary species (total PANs, total organic nitrates and formaldehyde). The results are shown in Figure S7. Here it can be seen that the secondary species are relatively higher for longer than the directly emitted species. Following the initial peak, the primary emissions reach 50% and 10% of peak by $\approx$ 35 mins and 2 h 20 mins, respectively. In contrast, the secondaries reach 50% and 10% of post-cooking peak concentrations by $\approx$1 h 50 mins and 4 h 40 mins, respectively.

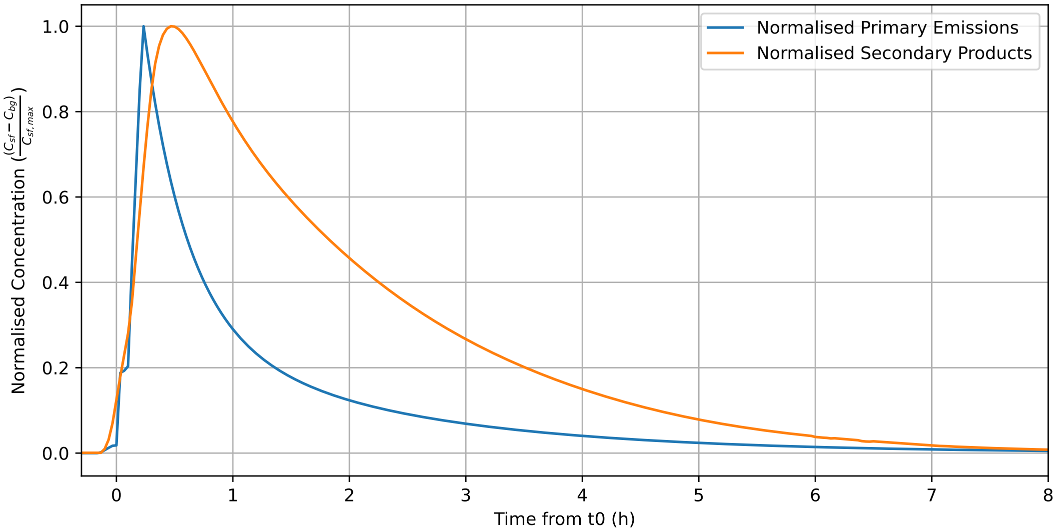

Figure S7: Normalised concentration of total directly emitted species (the 15 species modelled as cooking emissions) and total secondary products (total PANs, total organic nitrates and formaldehyde) over time, following t0.

10 Outdoor Oxidant Concentrations

The outdoor mixing ratios of O_3, NO and NO_2 used in the Base case and in the Polluted variation (Table 3, main text) are shown in Figure S8. Base case data is from the 'GB0586A, suburban London, 0.070766 51.45258' monitoring station (European Air Quality Database [3]), and the Polluted data is from measurements taken during a particularly polluted heatwave period in August 2003 in Milan [4].

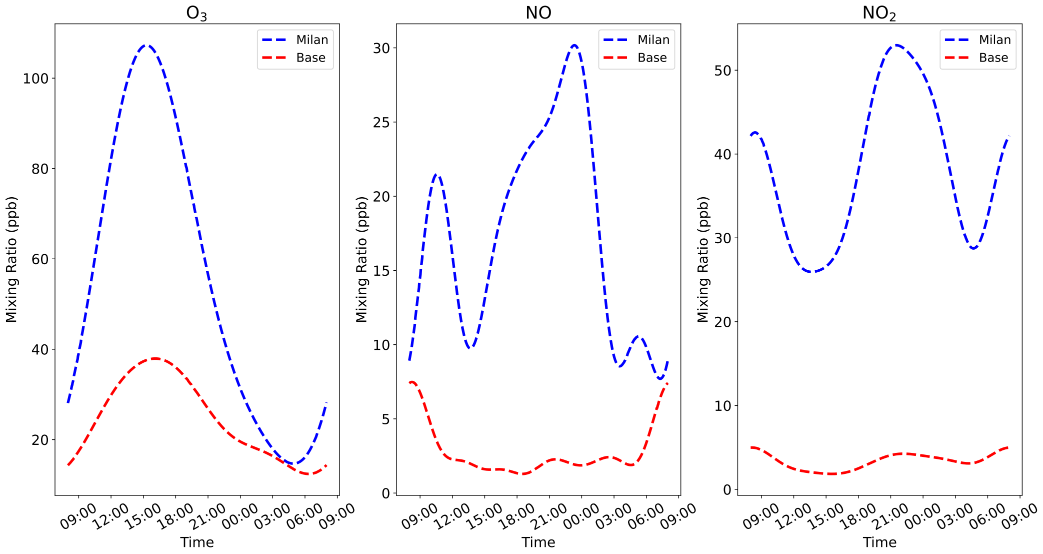

Figure S8: Mixing ratios of O_3, NO and NO_2 used in the Base case the Polluted variation simulations. Base case data comes from the European Air Quality Database [3], and the Milan data is from Terry et al., 2014 [4].

References

[1] Syft Technologies. Quantitation: SIFT-MS Calibration Principles. *Syft Technologies Training Materials*, 2014.

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