Supplemental Information

From the HOMEChem Frying Pan to the Outdoor Atmosphere: Chemical Composition, Volatility Distributions and Fate of Cooking Aerosol

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S0. Thermal Denuder Calibration

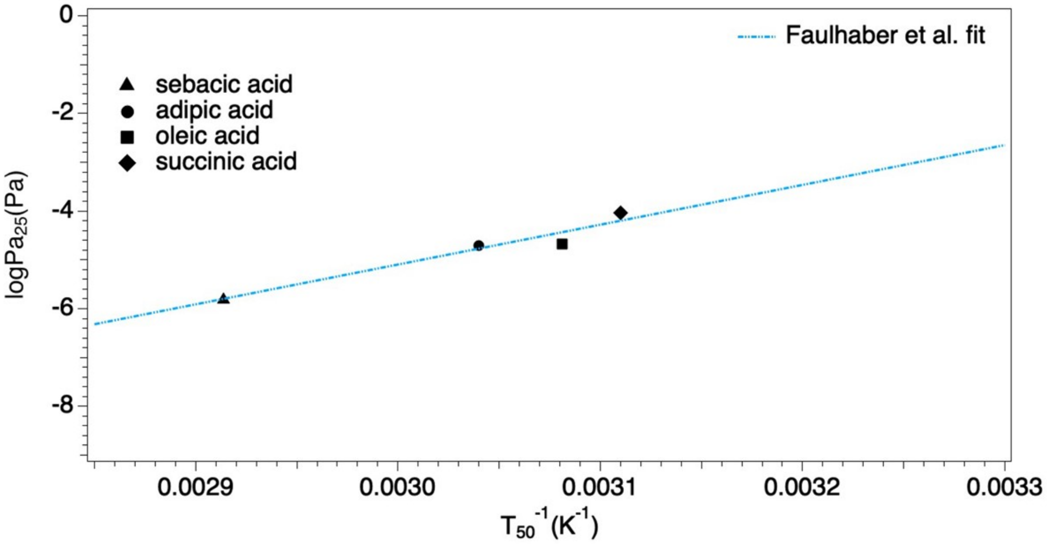

Figure S1: We calibrated the thermodenuder using four organic acids (black markers), with results in close agreement with Faulhaber et al.'s previous characterization of the same thermodenuder (blue line). Calibrations were conducted uniform across each organic acid: particle distribution mean diameter (200nm), mass loading (100 μg m^{-3}), flow rate (0.5 lpm), and residence time (15s)

Equations S1 – S3: Calibration equations used to relate thermodenuder T_{50} data to saturation vapor pressure (Pa_{25}). We relate Pa_{25} to saturation vapor concentration (C*) through Eqn S2 and S3 when constructing a volatility basis set. R is the gas constant, T is a given temperature, and MW is molecular weight. Here, we assume the molecular weight is that of oleic acid, 282.5 g mol^{-1}. Oleic acid is a major component of soybean oil, and one of the calibration compounds shown in Figure S1.

$$log\ Pa_{25} = \frac{8171}{T_{50}} - 29.61 \qquad \text{(Eqn. S1)}$$

$$C^* = \frac{MW \times Pa_{25} \times 10^6}{RT} \qquad \text{(Eqn. S2)}$$

$$T_{50} = \log\left(C^*\right) + \log\left(\frac{RT}{MW}\right) + 23.61 \qquad \text{(Eqn. S3)}$$

S1. Positive Matrix Factorization Analysis

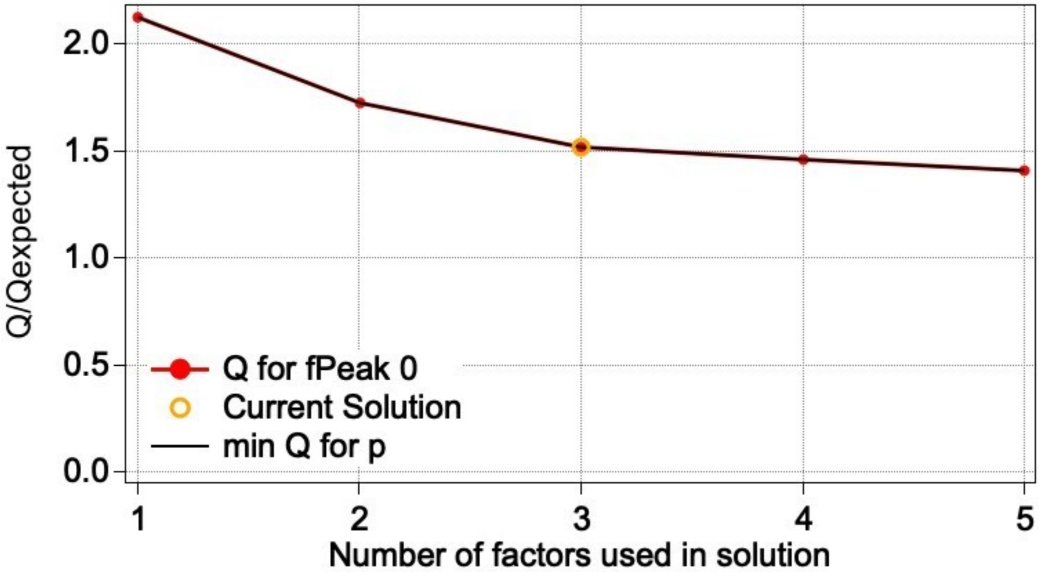

Figure S2: The Q/Q_{exp} value of the PMF solutions illustrates the actual model error (Q) compared to the expected error (Q_{exp}), as calculated from the solution residuals (Figure S3, below). The Q/Q_{exp} is shown as a function of number of factors used in solution, and highlights the stronger improvement in goodness of fit between a 2- and 3-factor solution than between a 3- and 4-factor solution.

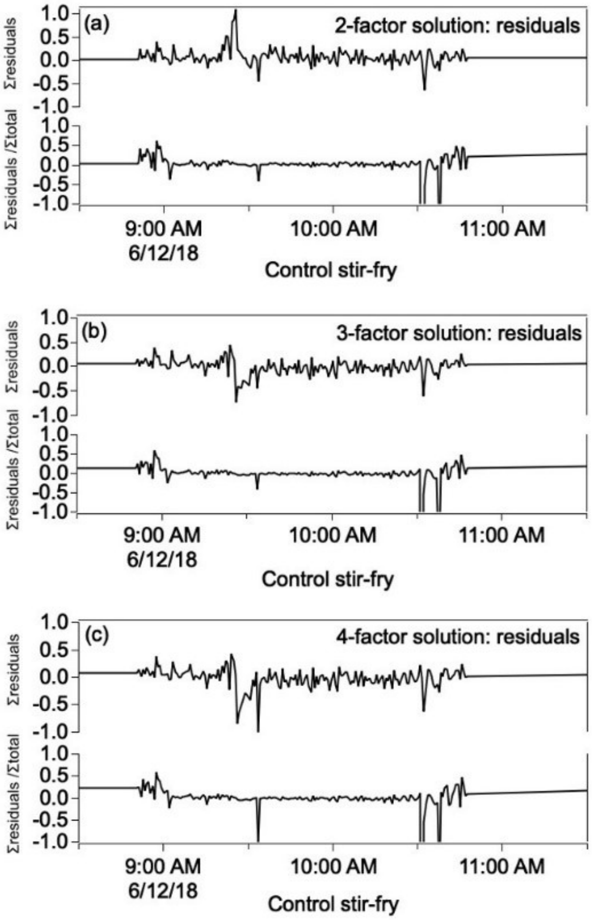

Figure S3: Timeseries residuals for the 3-factor solution during the Control stir-fry for the 2-factor (a), 3-factor (b), and 4-factor (c) solutions.

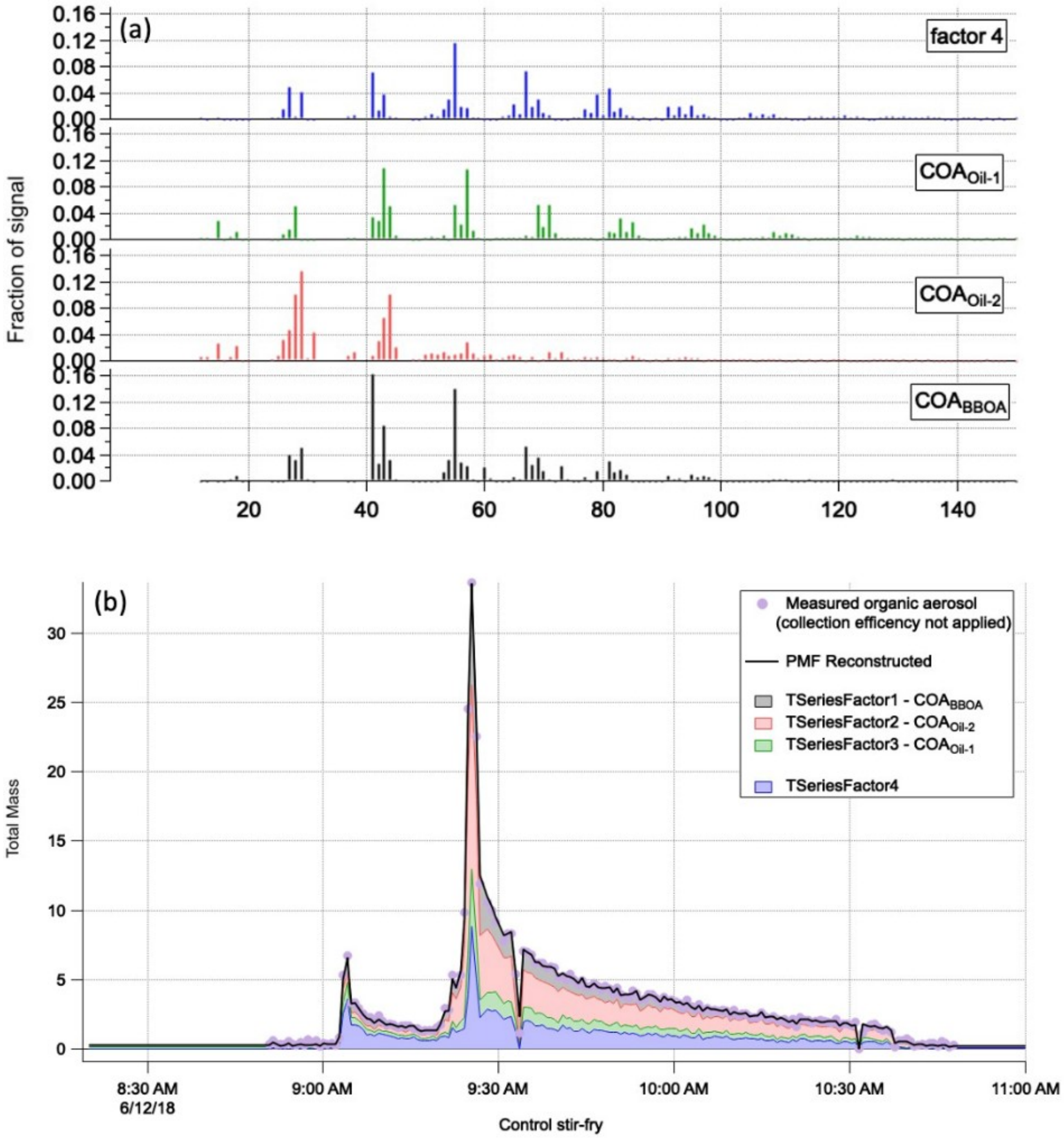

Figure S4: The output of the PMF 4-factor solution for the control stir-fry experiment includes (a) mass spectra for each factor and (b) timeseries of the four factors. The 4-factor solution appears to split the oil addition to create a fourth factor. Factors 1-3 are similar to the 3-factor solution.

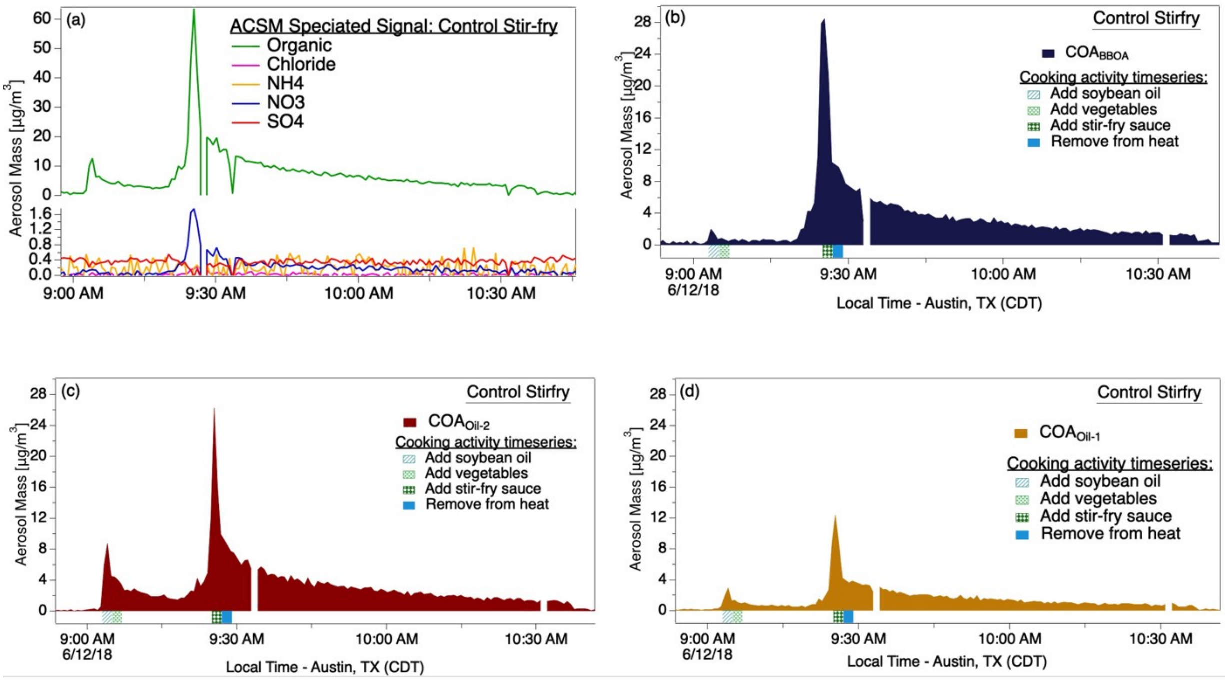

Figure S5: Aerosol mass from the control experiment (Figure 1), separated by **(a)** standard ACSM speciation of organic and inorganic components and **(b,c,d)** each cooking organic aerosol (COA) factor shown on its own.

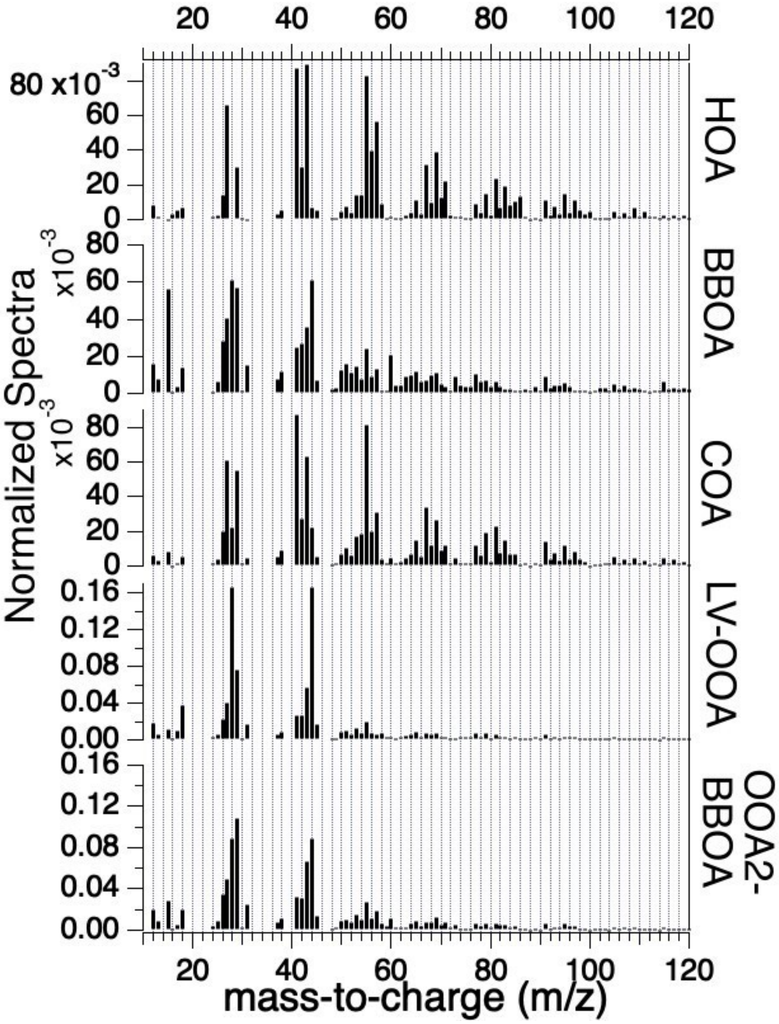

Figure S6: Positive matrix factorization (PMF) factors the MEGAPOLI campaign of urban organic aerosol (OA) measured in the winter of 2010 Paris, France, data from Crippa et al. (2013).

Table S1: Means and uncertainty of percentage of total COA mass for each of the three COA factors (columns), presented for each of the nine stir-fries (rows). We separate data into oil addition (left) and sauce addition (right) peaks. The uncertainty (σ) is calculated as the standard deviation of the timeseries during a given event. N/A indicates that data are unavailable due to the timing of the thermal scan. The campaign average represents the average of all nine experiments.

% Particulate mass in each COA factor						
	Oil addition			**Sauce addition**		
	COA_{BBOA}	$\text{COA}_{\text{Oil-2}}$	$\text{COA}_{\text{Oil-1}}$	COA_{BBOA}	$\text{COA}_{\text{Oil-2}}$	$\text{COA}_{\text{Oil-1}}$
Control Experiment (mean ± σ)						
9am, 12 June	0.18 ± 0.03	0.63 ± 0.04	0.19 ± 0.03	0.41 ± 0.02	0.39 ± 0.02	0.20 ± 0.02
Thermal Switch Experiments (mean ± σ)						
Switch 1 9pm, 12 June	0.10 ± 0.01	0.70 ± 0.01	0.20 ± 0.02	0.26 ± 0.03	0.52 ± 0.02	0.22 ± 0.02
Switch 2 9am, 17 June	0.22 ± 0.02	0.61 ± 0.01	0.17 ± 0.02	0.44 ± 0.04	0.34 ± 0.01	0.22 ± 0.02
Switch 3 12pm, 25 June	0.12 ± 0.04	0.61 ± 0.02	0.27 ± 0.02	0.29 ± 0.03	0.39 ± 0.03	0.32 ± 0.02
Thermal Scan Experiments (mean)						
Scan 1 9am, 6 June	0.20	0.51	0.29	0.31	0.44	0.25
Scan 2 12pm, 8 June	0.15	0.51	0.34	0.35	0.39	0.26
Scan 3 1pm, 12 June	N/A	N/A	N/A	0.39	0.36	0.25
Scan 4 5pm, 12 June	N/A	N/A	N/A	0.48	0.30	0.22
Scan 5 5pm, 17 June	N/A	N/A	N/A	0.32	0.47	0.21
Campaign average (mean ± σ)	0.16 ± 0.05	0.60 ± 0.07	0.24 ± 0.06	0.36 ± 0.07	0.40 ± 0.09	0.24 ± 0.03

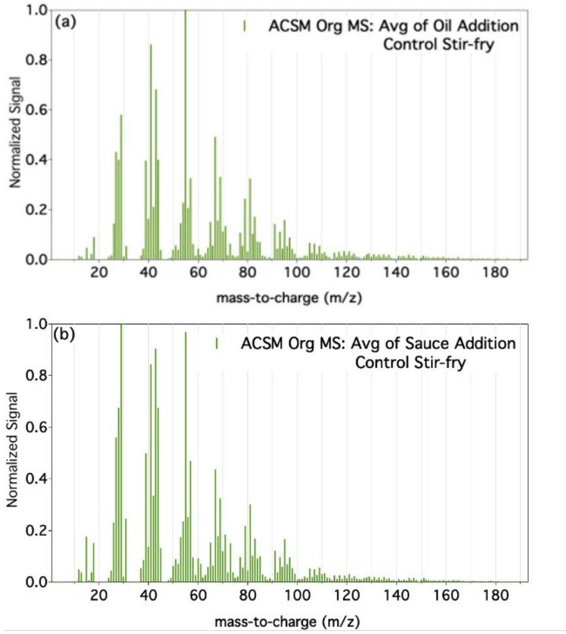

Figure S7: The mass spectrum of organic aerosol mass averaged following either **(a)** the oil addition (step 6) or **(b)** the sauce addition (step 8).

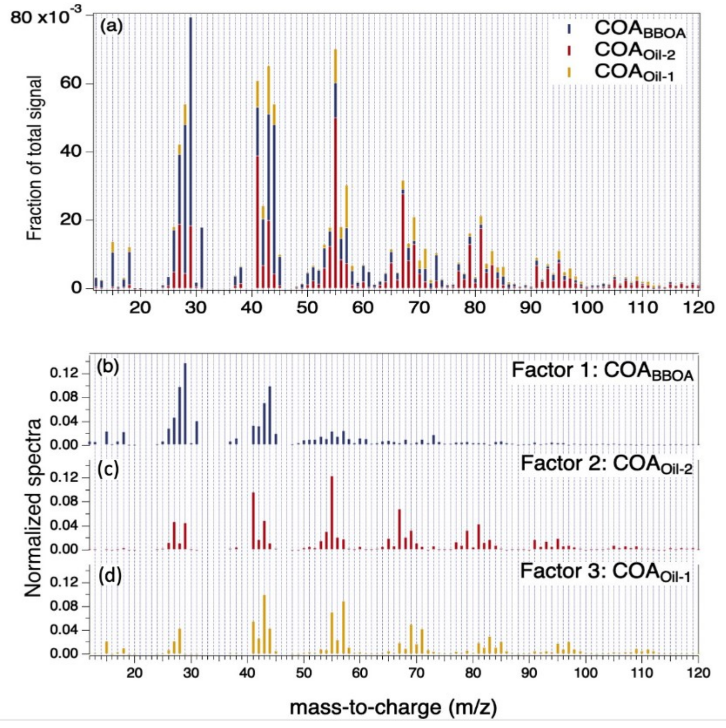

Figure S8: The mass spectrum of PMF-derived COA. The top panel **(a)** shows the contribution of the 3 factors at each m/z for the data following the sauce addition of the control experiment. The bottom panels **(b-d)** show the same data but separated by each factor.

S2. Estimating a reference spectrum for soybean oil

To further explore COA, we compare the two COA_{oil} factors to soybean oil related reference spectra. Spectra of commercial cooking oil are scarce, but both unhydrogenated and partially hydrogenated soybean oil are a mixture of ~five organic acids (with varying fractions;amounts - Figure S9a). We thus construct weighted reference spectra for two types of soybean oil using established electron ionization (EI) mass spectra of known components from the NIST EI mass spectral database (Figure S9b,c).

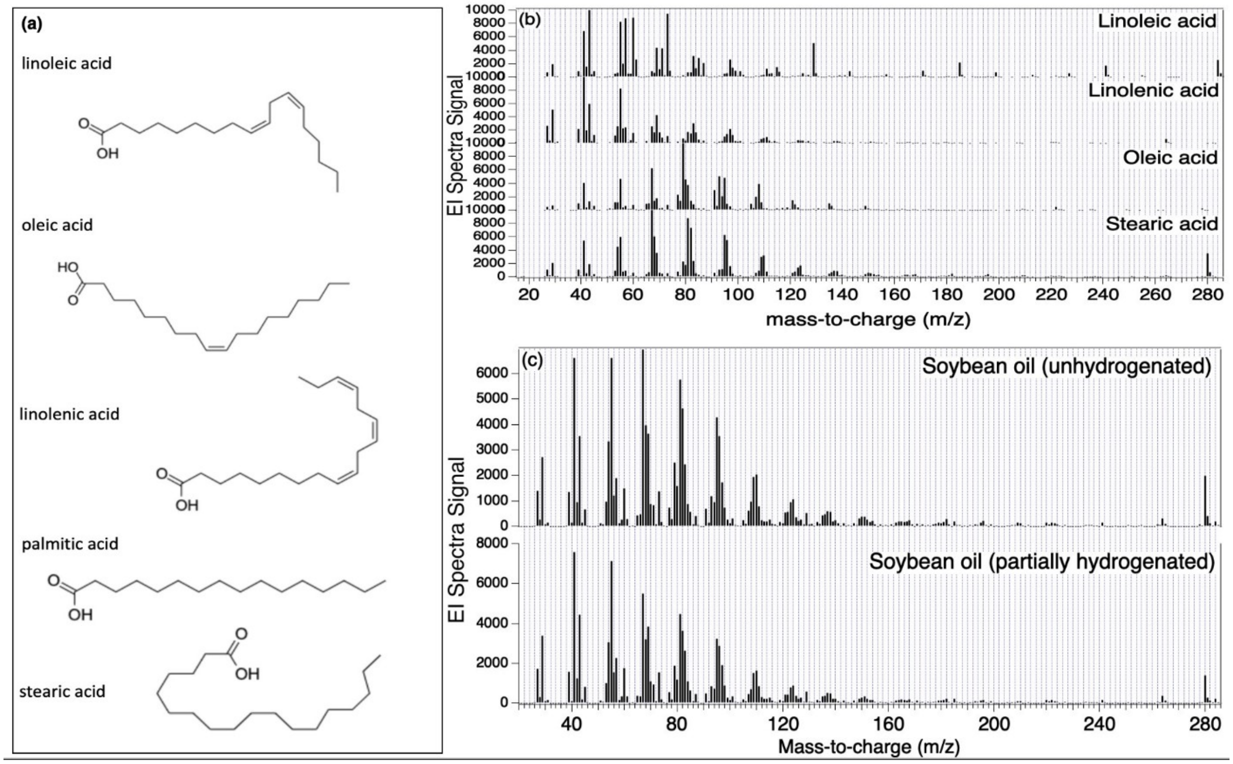

Figure S9: (a) The chemical structure of organic acids found in two types of soybean oil. **(b)** EI spectra for linoleic, linoleinic, oleic, and stearic acid from the NIST database. We note that EI mass spectra were unavailable for palmitic aid, but it only accounts for ~10% of the total mass of unhydrogenated or partially hydrogenated soybean oil. **(c)** The reconstructed mass weighted spectra from the four organic acids for unhydrogenated and partially hydrogenated soybean oil.

Table S2: The (Pearson's r, r^2), and [dot product] values for comparison of **(top row – measured MS)** the total organic mass spectrum of the first and second emission spikes from the control (i.e., no thermodenuder) experiment as well as the 3 PMF factors derived from all nine stir fry experiments; COA_{BBOA}, COA_{Oil-2}, and COA_{Oil-1}, as well as COA_{Oil} – the composition of COA_{Oil-2} and COA_{Oil-1}. We compare these mass spectra against **(left column – literature/reference MS)** literature reported PMF spectra from an urban aerosol measurements campaign as well as the reconstructed soybean oil mass spectrum.

Paris winter campaign 2010 & NIST spectra	**Control stir-fry emission: oil addition**	**Control stir-fry emission: sauce addition**	**COA_{BBOA}**	**COA_{Oil-2}**	**COA_{Oil-1}**	**Factor 2+3 – Cooking Oil OA (COA_{Oil})**
HOA	(0.88,0.77) [0.94]	(0.83,0.69) [0.94]	(0.48,0.23) [0.65]	(0.79,0.63) [0.88]	(0.91,0.83) [0.90]	(0.88,0.77) [0.94]
BBOA	(0.61,0.38) [0.82]	(0.79,0.62) [0.85]	(0.89,0.78) [0.81]	(0.42,0.17) [0.79]	(0.49,0.24) [0.67]	(0.46,0.21) [0.80]
COA	(0.97,0.95) [0.98]	(0.92,0.85) [0.97]	(0.60,0.36) [0.72]	(0.93,0.86) [0.96]	(0.74,0.55) [0.83]	(0.95,0.90) [0.98]
LV-OOA	(0.48,0.23) [0.85]	(0.69,0.48) [0.88]	(0.87,0.76) [0.81]	(0.25,0.06) [0.80]	(0.43,0.19) [0.73]	(0.31,0.10) [0.83]
OOA2-BBOA	(0.65,0.43) [0.93]	(0.85,0.73) [0.96]	(0.98,0.96) [0.84]	(0.43,0.19) [0.87]	(0.52,0.27) [0.85]	(0.48,0.23) [0.92]
partially hydrogenated soybean oil (calculated)	(0.85,0.72) [0.80]	(0.71,0.50) [0.80]	(0.28,0.08) [0.52]	(0.89,0.78) [0.73]	(0.64,0.41) [0.85]	(0.89,0.79) [0.80]
unhydrogenated soybean oil (calculated)	(0.76,0.58) [0.78]	(0.61,0.37) [0.78]	(0.19,0.04) [0.50]	(0.83,0.69) [0.73]	(0.54,0.29) [0.80]	(0.65,0.43) [0.79]

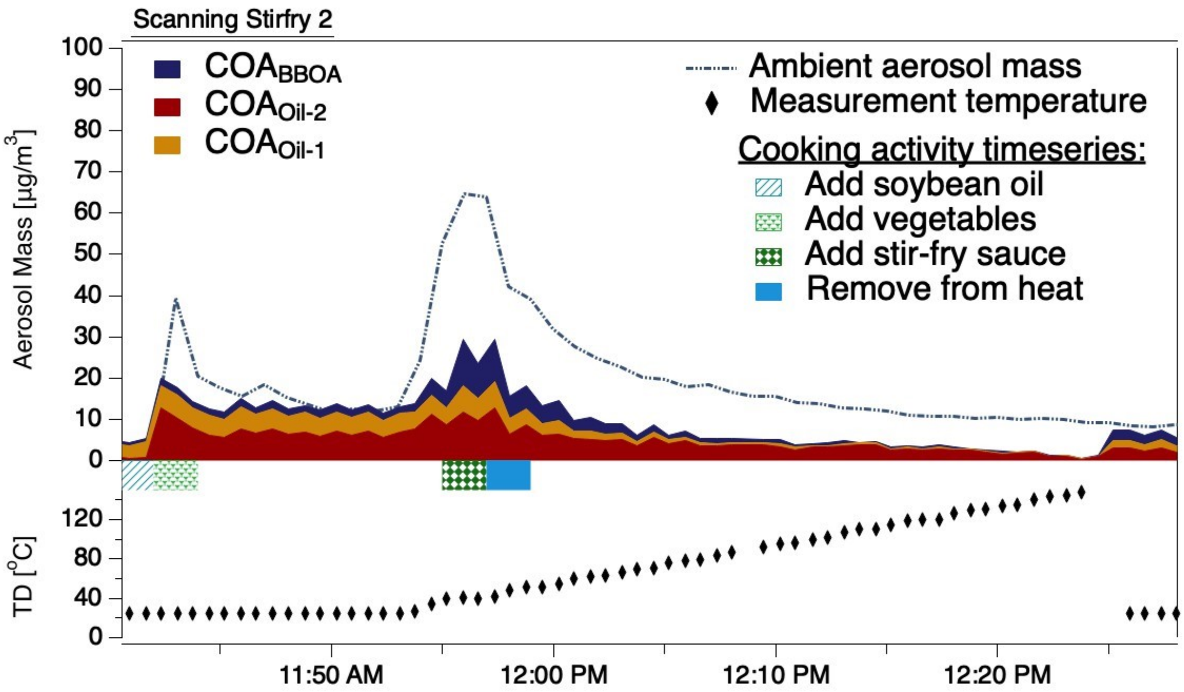

Figure S10: The timeseries of organic aerosol during a thermal scanning experiment (stir-fry 2); the dashed line shows the total sub-micron aerosol loading measured by the portable optical particle sampler (POPS). The lower figure shows the thermodenuder temperature.

S3. Verifying stability of factors during stir-fry experiments

Calculating Mass Fraction Remaining (MFR) for PMF factors requires understanding the ambient mass of each factor at each time point. In the ambient atmosphere, aerosol typically changes at a slow enough pace that measurements taken before or after a thermal denuder measurement are sufficient; however, aerosol mass loading often shifts an order(s) of magnitude during cooking experiments over the time required to complete one thermal scan (ambient → 150^{o}C). During the control and bypass line measurements, each COA factor maintained a stable fraction of the ambient loading (Table S1, Figure S11). We thus combine ambient mass loadings measured by the POPS with COA ratios from ambient/bypass data during the same emission period (either oil or sauce addition) to estimate the ambient mass of each COA factor. The temperature held in the thermodenuder during switching experiments (80^{o}C) had little effect on COA$_{Oil-2}$, allowing us to validate this method of estimating ambient mass loading of the factors.

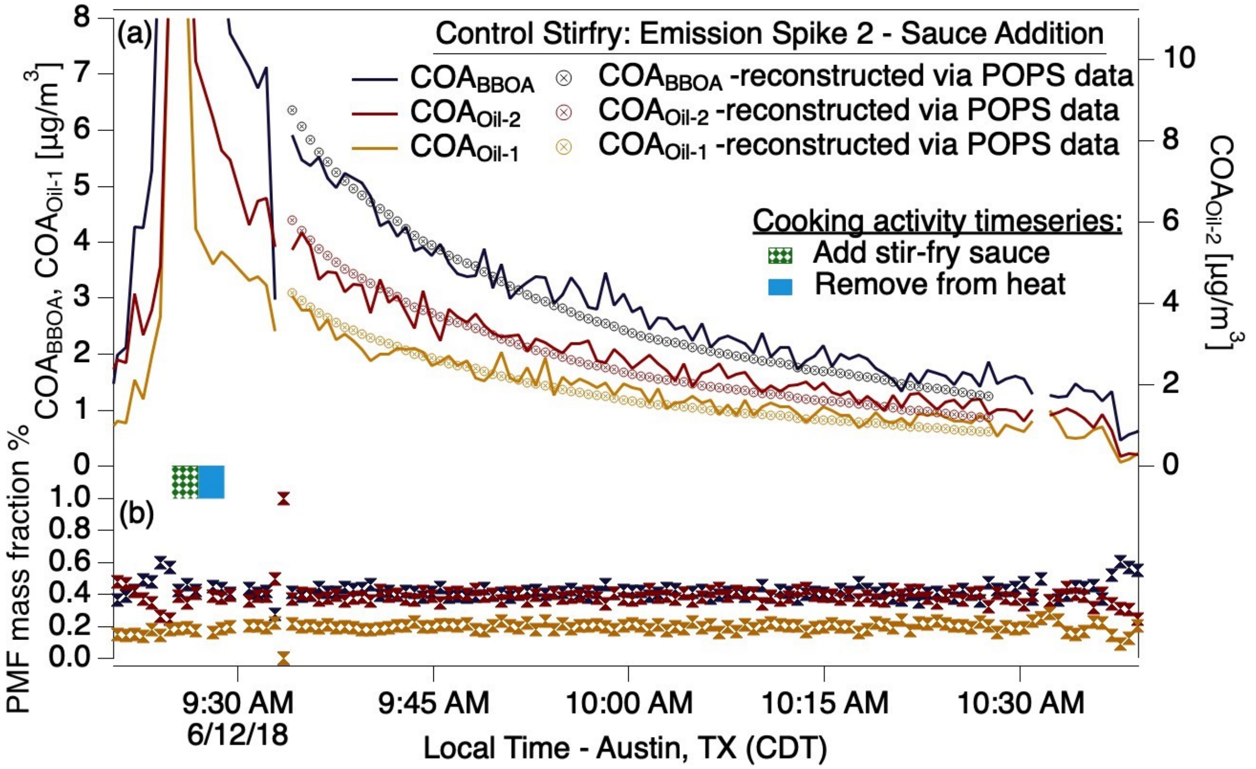

Figure S11: Aerosol mass loading of the three COA factors during the sauce addition period of the control experiment demonstrates that the POPS provides a reasonable proxy for total aerosol when the ACSM is connected to the heated thermal denuder. (a) Ambient ACSM data (solid lines) are similar to estimated ambient mass loadings calculated using POPS data (cross marker). Due to similar mass loading of COA$_{BBOA}$ and COA$_{Oil-2}$, COA$_{Oil-2}$ is on a separate y-axis. The lower panel (b) shows the timeseries of the mass fraction of each factor; these data are summarized in Table S1.

S4. VBS calculations

The analysis workflow to translate thermodenuder data (Figure 2a) to a VBS (Figure 2c) for COA$_{BBOA}$ is as follows:

1. The thermodenuder must be calibrated using a set of laboratory organic acid standards to empirically determine a calibration equation, shown in Figure S1 for this system (See Faulhaber et al. (2009) for a detailed description of this process). Equation S1 relates the evaporation dynamics inside the thermodenuder to a set of standards with known vapor pressures.

2. Creation of mass fraction remaining (MFR) curves requires the knowledge of each factor’s ambient mass loading during the thermodenuder ramp. If the ambient organic aerosol mass loading and composition are similar before and after the temperature ramp, one can assume the composition was stable and thus estimate the ambient mass for each factor. However, the organic aerosol mass loading changed significantly each stir-fry, and

we use other a priori knowledge of the system. See section **S3 Determining factor mass stability during stir-fry experiments** for details of our approach.

3. Calculate MFR using ambient factor mass and thermodenuder data **(Figure S12a; bottom axis)**.

4. Convert temperature data from Celsius to Kelvin **(Figure S12a; top axis)**.

5. Fit the mass fraction remaining data as a function of temperature **(Figure S12a; dashed line)**.

6. Convert temperature to inverse Kelvin **(Figure S12b; lower axis)**.

7. Calculate C^* bin limits for the range of C^*bins measured during the temperature ramp using Eqn. S3 **(results shown in Figure S12b; top figure and shading)**.

8. Calculate the fraction of mass in each C^* bin ($F_{p,initial}$) as the difference in mass fraction remaining between the fit line at each of the bin limits.

9. Calculate the particle and gas phase mass loading (µg m^{-3}) for each C^* bin ($C_{p,i}$ and $C_{g,i}$) using **Equations S4, S5, and S6** from Donahue et al. 2006 and Pankow 1994a, where f_i is the $F_{p,\ initial}$ calculated in step 8 and C_{OA} is the total ambient OA mass loading (µg m^{-3}) determined from independent measurements.

$$\frac{C_{p,i}}{C_{g,i}} = \frac{C_{OA}}{C_i{}^*} \qquad \textbf{(Eqn. S4)}$$

$$C_{p,i} = F_{p,initial} \times C_{OA} \qquad \textbf{(Eqn. S5)}$$

$$C_{g,i} = F_{p,initial} \times C_i{}^* \qquad \textbf{(Eqn. S6)}$$

10. If the sampling environment is not 25°C (standard conditions), the dataset must then be converted into standard conditions, but as the temperature of the UTest house was kept at 25°C the entire campaign, this conversion was unnecessary for this work.

11. Equations S7 and S8 are then iteratively solved, and the resulting set of values described by Equations S5 and S6 are used to create the VBS. The F_p values in **Figure S12b** are from the VBS solution for COA_{BBOA}. These F_p and C_{OA} values are later used as the VBS parameterization in modeling dilution-driven evaporation.

$$C_{OA} = \sum C_{p,i} \qquad \textbf{(Eq. S7)}$$

$$C_{p,i} = \frac{C_{OA} \times C_{tot,i}}{(C_i{}^{*} + C_{OA})}$$

(Eq. S8)

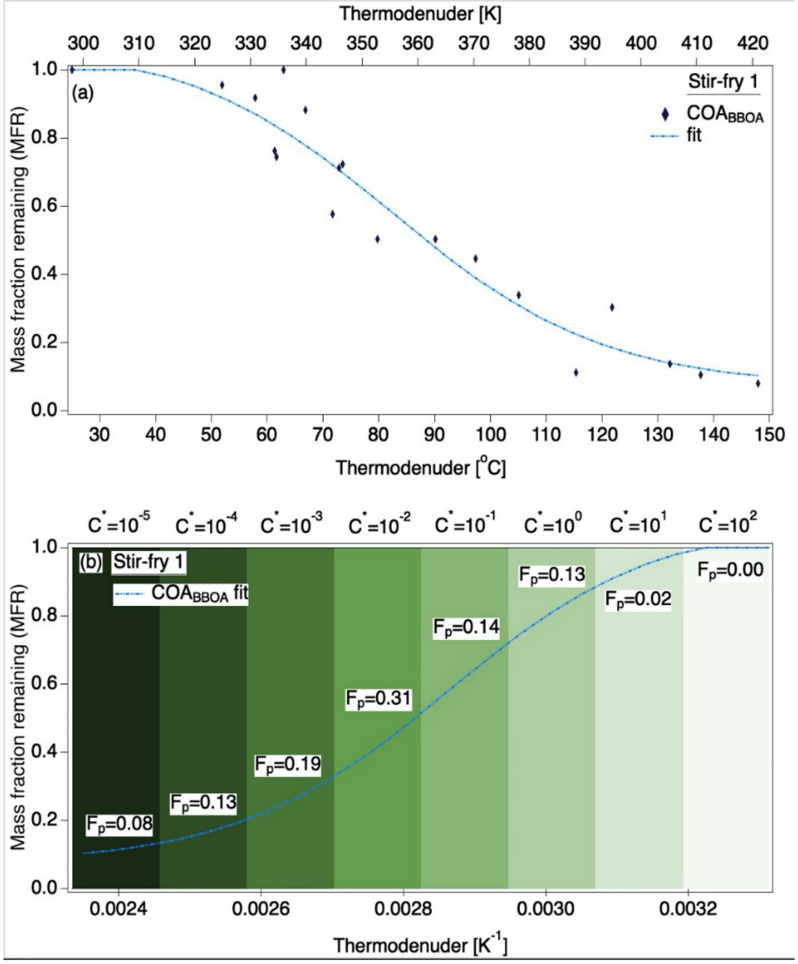

Figure S12: Mass fraction remaining (MFR) data for COA_{BBOA} from the sauce addition of stir-fry 1 (stir-fry 1) calculated using data from Figure 2a. **(a)** MFR data fit (shown in Figure 2b) used to calculate the VBS. Panel **(b)** shows the same data converted to K^{-1} **(bottom axis)**, with C^* bins determined using equation S3 **(top axis and shading)**. F_p values represent the portion of the OA mass that belongs to each C^* bin.

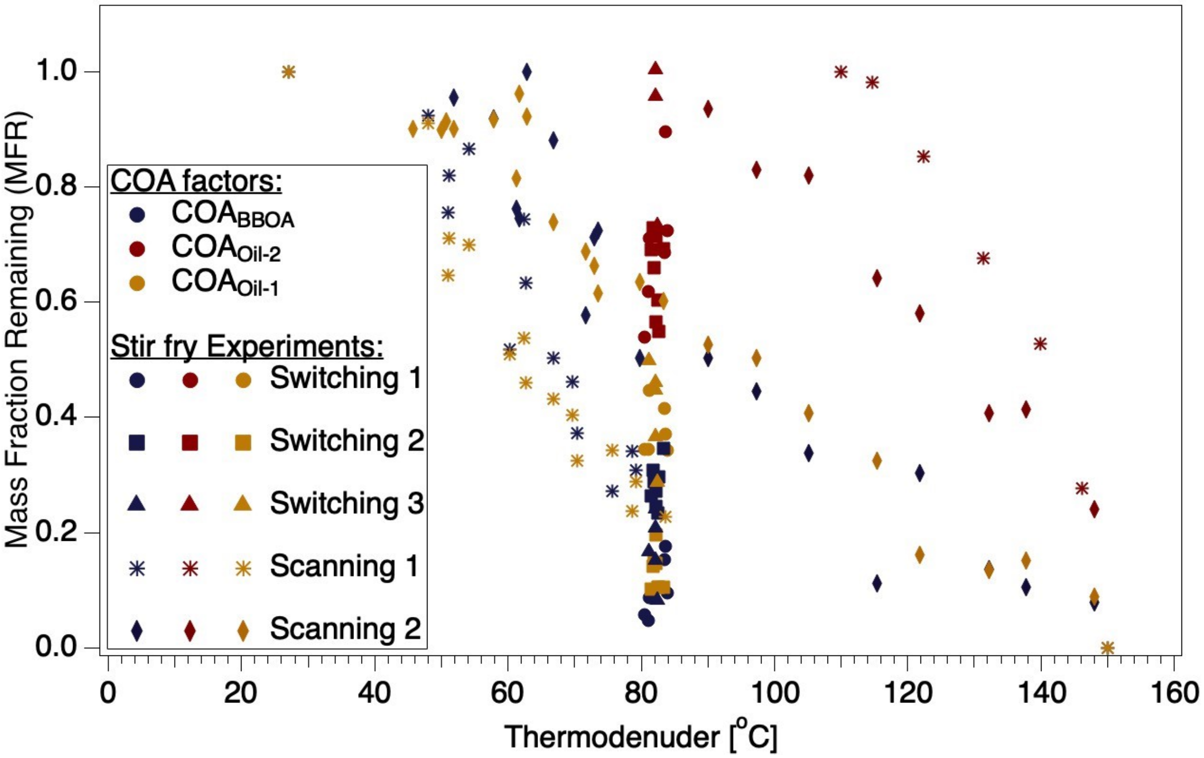

Figure S13: Thermal scan data for COA_{BBOA} (blue),COA_{Oil-2} (red), and COA_{Oil-1} (gold) for both thermal scan experiments presented in the main text (diamonds and stars) and a large clustering of datapoints ~80°C from data gathered during the switching experiments (circles, squares and triangles).

S5. Kinetic partitioning model

Our model calculates the time-dependent aerosol and gas phase mass and aerosol diameter change after a step change in temperature or concentration due to dilution. The model was originally developed by Riipinen et al.[53] to describe the evaporation dynamics and calculate timescales required to achieve thermodynamic equilibrium, within a thermodenuder. The method outlined in section **S4 VBS calculation** does not require the assumption that the measured aerosol mass has achieved thermodynamic equilibrium, what this type of model is typically used for. Instead, we choose to use the calculated VBS to validate the models ability to describe the partitioning dynamics of the aerosol mass (Figure S14 and accompanying Table S3). Finding good agreement between the results and measured aerosol mass, we then leveraged the model to understand shifts in the aerosols thermodynamic equilibrium as it dilutes throughout the house and eventually to the outdoor/ambient atmosphere (S5.1 – Model inputs and results).

Table S3: Model results showing the particle mass that has evaporated, the equilibrium value, and the difference from each volatility bin at a given temperature.

	$C^*=10^{-5}$	**$C^*=10^{-4}$**	**$C^*=10^{-3}$**	**$C^*=10^{-2}$**	**$C^* =10^{-1}$**	**$C^*=10^{0}$**	**$C^*=10^{1}$**	**$C^*=10^{2}$**	**Modeled mass total (Residence Time = 18s)**	**TD//ACSM measured (Residence Time = 18s)**
Thermodenuder temperature = 50.7°C										
Particle mass modeled [μg m^{-3}]	6.10	3.06	3.37	1.92	1.97	1.44	0.21	3x10^{-3}	18.07	16.34
Equilibrium particle mass [μg m^{-3}]	6.10	3.06	3.37	1.91	1.89	1.30	0.21	3 x10^{-3}		
Thermodenuder temperature = 63°C										
Particle mass modeled [μg m^{-3}]	4.07	2.04	2.23	1.22	0.93	0.23	0.03	6x10^{-4}	10.75	11.34
Equilibrium particle mass [μg m^{-3}]	4.07	2.04	2.21	1.14	0.70	0.70	0.03	6 x10^{-4}		
Thermodenuder temperature = 80°C										
Particle mass modeled [μg m^{-3}]	3.63	1.81	1.90	0.78	0.17	0.03	4x10^{-3}	1x10^{-4}	8.32	5.83
Equilibrium particle mass [μg m^{-3}]	3.62	1.78	1.71	0.52	0.13	0.03	4x10^{-3}	9x10^{-5}		
Thermodenuder temperature = 101°C										
Particle mass modeled [μg m^{-3}]	2.78	1.30	0.93	0.07	0.01	2x10^{-3}	4x10^{-4}	1x10^{-5}	5.09	3.82
Equilibrium particle mass [μg m^{-3}]	2.60	0.92	0.36	0.04	7x10^{-3}	2x10^{-3}	3x10^{-4}	8x10^{-6}		
Thermodenuder temperature = 120°C										
Particle mass modeled [μg m^{-3}]	2.27	0.72	0.09	5x10^{-3}	1x10^{-3}	2x10^{-4}	5x10^{-5}	2x10^{-6}	3.09	2.29
Equilibrium particle mass	0	0	0	0	0	0	0	0		

[µg m^{-3}]										
Thermodenuder temperature = 146^{o}C										
Particle mass modeled [µg m^{-3}]	0.51	5x10^{-3}	8x10^{-4}	8x10^{-5}	2x10^{-5}	5x10^{-5}	1x10^{-6}	4x10^{-7}	0.52	1
Equilibrium particle mass [µg m^{-3}]	0	0	0	0	0	0	0	0		

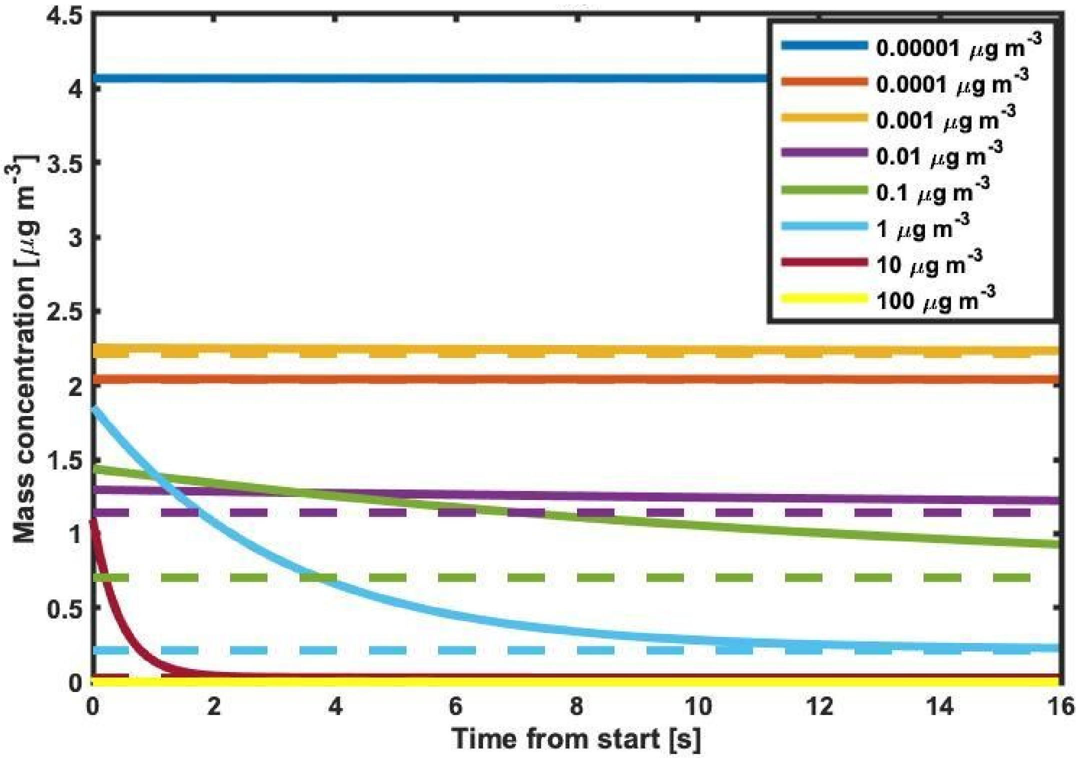

Figure S14: The decrease in mass concentration as a function of time shows the evaporation timescales required for each C* bin (solid lines, colored by saturation vapor pressure) to reach its thermodynamic equilibrium (dashed lines) when the thermodenuder was at ~85^{o}C.

S5.1. Model inputs and results

Required experimental parameters/general input include:

1 - ambient house temperature
2 - ambient house pressure

3 - organic aerosol mass concentration; we used the peak concentration after oil and sauce additions
4 - initial particle diameter; we used mean Dp of corresponding COA concentration peak; aerosol size distributions from HOMEChem are reported on extensively in Boedicker et al. 2021 and Patel et al. 2020)

Species specific inputs are required for each C^* bin used:

5 - H_{vap} heat of vaporization (used recommendation from Donahue et al. 2006; $C^* = 1$ µg m^{-3} bin is assigned an H_{vap} of 100 kJ mol^{-1} with each adjacent bin differing by 5.8 kJ mol^{-1} in H_{vap})

i.e., $C^* = 10$ µgm$^{-3} = H_{vap}$ of 94.2 kJ mol^{-1}
$C^* = 1$ µgm$^{-3} = H_{vap}$ of 100 kJ mol^{-1}
$C^* = 0.1$ µgm$^{-3} = H_{vap}$ of 105.8 kJ mol^{-1}
$C^* = 0.01$ µgm$^{-3} = H_{vap}$ of 111.6 kJ mol^{-1}

*****(variable is held constant across all C^* bins)

***6** - molecular weight (calculated the weighted average molecular weight of soybean oil components - 267.83 g mol^{1})

***7** - density (from Patel et al. (2020); 1050 kg m^{-3})

***8** - diffusivity (1x10^{-5} m^{2}s^{-1})

9 - We determine F_P fraction of organic aerosol mass belonging to a given C^* bin (calculated using mass fraction remaining data and thermodenuder calibration equation)

Fp index : $C^* = [10^{-5}, 10^{-4}, 10^{-3}, 10^{-2}, 10^{-1}, 10^{0}, 10^{1}, 10^{2}]$

stir-fry 1: oil addition (F_p) = [0.352, 0.170, 0.170, 0.0692, 0.0748, 0.0920, 0.0544, 0.00254]
stir-fry 1: sauce addition (F_p) = [0.2873, 0.1442, 0.1589, 0.0915, 0.1015, 0.1310, 0.0774, 0.0026]

stir-fry 2: oil addition (F_p) = [0.2661, 0.1347, 0.1521, 0.0942, 0.1072, 0.1439, 0.0850, 0.0020]
stir-fry 2: sauce addition (F_p) = [0.0867, 0.0555, 0.0682, 0.0904, 0.1551, 0.2451, 0.2153, 0.0729]

We use an accommodation coefficient: $\alpha = 1.0$.

Below are the model results using data from both the oil and sauce additions of the stir-fries presented in Figure 2a and Figure S10. Model results from ‘stir-fry 2: sauce addition’ are used to make Figure 3 in the main text.

Table S4 Stir-fry 1: oil addition

	Initial aerosol mass	Fraction of aerosol mass remaining after dilution to house	Fraction of aerosol mass remaining after dilution to 15 µg m^{-3} atmosphere	Fraction of aerosol mass remaining after dilution to 5 µg m^{-3} atmosphere
Total OA	61.40 µg m^{-3}	0.97	0.96	0.94
COA_{BBOA}	10.60 µg m^{-3}	0.91	0.88	0.83
COA_{Oil-2}	35.50 µg m^{-3}	1.0	1.0	1.0
COA_{Oil-1}	15.30 µg m^{-3}	0.93	0.90	0.87

Table S5 Stir-fry 1: sauce addition

	Initial aerosol mass	Fraction of aerosol mass remaining after dilution to house	Fraction of aerosol mass remaining after dilution to 15 µg m^{-3} atmosphere	Fraction of aerosol mass remaining after dilution to 5 µg m^{-3} atmosphere
Total OA	38.40 µg m^{-3}	0.93	0.91	0.88
COA_{BBOA}	14.05 µg m^{-3}	0.88	0.84	0.78
COA_{Oil-2}	15.06 µg m^{-3}	1.0	1.0	1.0
COA_{Oil-1}	9.29 µg m^{-3}	0.90	0.87	0.83

Table S6 Stir-fry 2: oil addition

	Initial aerosol mass	Fraction of aerosol mass remaining after dilution to house	Fraction of aerosol mass remaining after dilution to 15 µg m^{-3} atmosphere	Fraction of aerosol mass remaining after dilution to 5 µg m^{-3} atmosphere
Total OA	19.89 µg m^{-3}	0.91	0.88	0.85
COA_{BBOA}	1.27 µg m^{-3}	0.77	0.70	0.61
COA_{Oil-2}	15.21 µg m^{-3}	0.96	0.94	0.93
COA_{Oil-1}	3.41 µg m^{-3}	0.76	0.68	0.60

Table S7 Stir-fry 2: sauce addition

	Initial aerosol mass	**Fraction of aerosol mass remaining after dilution to house**	**Fraction of aerosol mass remaining after dilution to 15 µg m^{-3} atmosphere**	**Fraction of aerosol mass remaining after dilution to 5 µg m^{-3} atmosphere**
Total OA	64.70 µg m^{-3}	0.76	0.69	0.62
COA$_{BBOA}$	24.53 µg m^{-3}	0.71	0.62	0.52
COA$_{Oil-2}$	21.95 µg m^{-3}	0.91	0.88	0.85
COA$_{Oil-1}$	18.22 µg m^{-3}	0.65	0.56	0.47

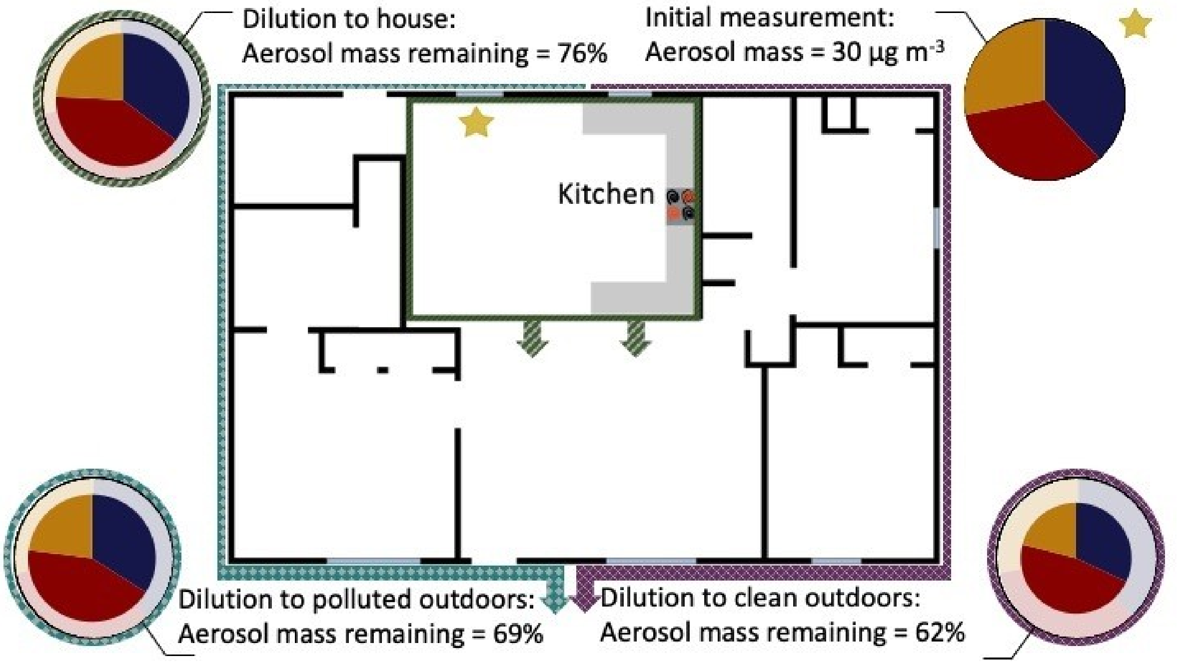

Figure S15: Results from a chemical kinetic partitioning model describing the dilution-driven evaporation of aerosol emitted during stir-fry experiment, superimposed on a diagram of the UTest house. The star indicates the inlet location for the TD-ACSM. Each pie chart shows the fraction of mass remaining for each factor (solid) relative to the initial emission (light color). Pie charts show the initial, emitted particulate mass of 30 µg m^{-3} (top right), particulate mass remaining upon mixing through the entire house air volume (top left), particulate mass remaining upon dilution outdoors under more polluted (bottom left) versus cleaner (bottom right) conditions. Polluted versus clean conditions were defined by outdoor measurements of ambient air in Austin, TX during the HOMEChem study.