- 1 Supplement of
- 2 Formation and Evolution of Secondary Organic Aerosol Derived from Urban Lifestyle Sources:

3 Vehicle Exhaust and Cooking Emission

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- 33 List of the supporting information:
- 34 Section S1. Details about vehicle and cooking lab experiments
- 35 Section S2. Go: PAM conditions
- 36 **Table S1.** Comparison of results between blank and experimental groups.
- Figure S1. Comparison of measured and estimated OH exposures during off-line OH exposure calibration using SO₂ for
 vehicle experiment.
- Figure S2. Comparison of measured and estimated OH exposures during off-line OH exposure calibration using SO₂ for
 cooking experiment.
- 41 **Figure S3.** Previous performance tests for Go: PAM.
- 42 Figures S4-S7. Mass spectra of PMF-resolved POA and SOA factors for cooking experiment.
- 43 Figures S8-S11. Diagnostic plots of the PMF analysis for cooking experiment.
- 44
- 45 Section S1: Details about vehicle and cooking lab experiments.

The vehicle experiment was conducted from July to October in 2019, at Department of Automotive Engineering, Tsinghua 46 University. For all experiments, the gasoline direct injection (GDI) engine ran in a single room, its exhaust was drawn into 47 pipeline and then entered the Go: PAM at a 30 fold dilution where aerosols and gases reacted at a stable temperature and 48 49 relative humidity. The cooking experiment was conducted from November 2019 to January 2020, at Langfang Branch, 50 Institute of Process Engineering, Chinese Academy of Sciences. The cooking time and oil temperature were different due to 51 the inherent features of ingredients. For all experiments, the closed kitchen was full of fumes where the vision was blurred 52 and the air was choky after a long time of cooking process. Subsequently, the cooking fumes were drawn into pipeline from 53 kitchen to lab and then entered the Go: PAM at an 8 fold dilution where aerosols and gases reacted at a stable temperature and 54 relative humidity. Besides, a temperature controller and heat insulation cotton were wrapped around the sampling pipelines 55 with the purpose of preventing freshly warm gas from condensing on the pipes wall. Silicon tubes were used to dry the emissions before they entered measuring instruments. The particle densities were measured through the determination of 56 DMA-CPMA-CPC system (DMA- Differential Mobility Analyzer; CPMA- Centrifugal Particle Mass Analyzer; CPC-57 Condensation Particle Counter) in our study. Prior to each experiment, all pipelines and the Go: PAM chamber were 58 continuously flushed with purified dry air until the concentrations of gases and particles were minimal. Furthermore, blank 59 experiments were separately designed in the presence of boiling water emissions or dilution air under the same condition. The 60 results of blank groups can be found in Table S1. When the OH exposure was zero, OA concentrations derived from dilution 61 air were so low that they couldn't be quantified correctly. On the whole, the OA concentrations of blank groups were far below 62 63 those of experimental groups. The field study was deployed at the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (39°58'N; 116°22'E) in autumn and winter (Autumn: Oct. 1st, 2018 - Nov. 15th, 2018; Winter: Jan. 5th, 2019 -64 65 Jan. 31st, 2019) (Li et al., 2020a). The sample site is located in the south of Beitucheng West Road and west of Beijing 66 Chengde expressway in Beijing, which is a typical urban site affected by local emissions (Li et al., 2020b).

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68 Section S2: Go: PAM conditions

Before the start of experiments, the consumption of SO₂ in the Go: PAM was used to calculate the OH exposure (Lambe et al., 2011). As shown in equation (1), K_{OH-SO_2} is the reaction rate constant of OH radical and SO₂ (9.0×10⁻¹³ molecule⁻¹·cm³·s⁻ ¹). The SO_{2, f} and SO_{2, i} are the SO₂ concentrations (ppb) under the conditions of UV lamp on or off respectively. The photochemical age (days) can be calculated in equation (2) when assuming the OH concentration is 1.5×10^6 molecules·cm⁻³ in the atmosphere (Mao et al., 2009).

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$$OH \ exposure = \frac{-1}{K_{OH-SO_2}} \times ln(\frac{SO_{2,f}}{SO_{2,i}})$$
 (1)

75 Photochemical age = $\frac{OH \ exposure}{24 \times 3600 \times 1.5 \times 10^6}$ (2)

Except for the off-line calibration based on the decay of SO₂, a flow reactor exposure estimator was also used in this study 76 77 (Peng et al., 2016). The OH exposures calculated by both methods showed a good correlation (Figure S1&S2). This estimator could also evaluate the potential non-OH reactions in flow reactor such as the photolysis of VOCs, the reactions with O(¹D), 78 79 $O(^{3}P)$ and O_{3} . Our results showed that non-OH reactions were not significant except for the photolysis of acetylacetone. But 80 there is no acetylacetone from vehicle exhaust or cooking emission according to our measurements and previous studies. The acetylacetone was usually considered as a kind of VOCs emitted from industrial production (Ji et al., 2020). Therefore, its 81 potential photolysis wouldn't take place during our cooking conditions, and OH reactions still played the dominant role. 82 Overall, our Go: PAM could reasonably simulate the oxidation process of cooking OA in ambient. 83

84 Furthermore, the external OH reactivity and OH exposure were both influenced by external OH reactants, such as NOx and 85 VOCs during experiments. The NOx concentration was measured by a NO-NO2-NOx Analyzer (Model 42i, Thermo Electron Corporation, USA). As for VOCs, we have divided them into 5 types including alkane, alkene, aromatic, O-VOCs (Oxidized 86 87 VOCs, mainly included aldehyde and ketone) and X-VOCs (halogenated-VOCs) using the measurement of GC-MS (Gas Chromatography-Mass Spectrometry, GC-7890, MS-5977, Agilent Technologies Inc). The compounds with relatively high 88 proportion were regarded as surrogate species for each type of VOCs. The total concentrations of VOCs were determined by 89 a portable TVOC Analyzer (PGM-7340, RAE SYSTEMS). The external OH reactivities for different vehicle experiments 90 $(10.4 \sim 20.2 \text{ s}^{-1})$ were all comparable to that of off-line calibration result (15.8 s⁻¹), and the external OH reactivities for different 91 92 cooking experiments (21.7~25.7 s⁻¹) were also comparable to that of off-line calibration result (24.0 s⁻¹). Besides, the ratio of 93 OH exposure calculated by the estimator to that calculated by the decay of SO₂ ranged from 83% to 119% for vehicle 94 experiments and 97% to 111% for cooking experiments, which means that our off-line OH exposure could be a representative value to all experiments. 95

- 96 The mixing and wall loss tests have already conducted in previous work using the same Go: PAM according to Li et al.(Li et
- al., 2019) and Watne et al. (Watne et al., 2018). In Figure S3(a), SO₂ was continually injected to the "4 Humidified oxidant

flow" and "5 Sample flow", and was measured through "3 Processed sample flow" (Watne et al., 2018). As shown in Figure 98 S3(b), there was nearly no difference when using different inlets, which demonstrated a great mixing of the sample and 99 oxidant flow in the Go: PAM (Watne et al., 2018). Figure S3(c) modeled the wall loss of LVOC (low-volatility VOC) following 100 the method of Palm et al., 2018;Palm et al., 2016). The results indicated that most LVOC tended to react with 101 OH or condensate on particles rather than exit or cause loss to the wall (Li et al., 2019). Figure S3(d) tested the particle wall 102 103 loss using nebulized ammonium sulfate particles. Results showed that the particle losses with size above 22.1 nm were nearly smaller than 10% which would only make a negligible effect in Go: PAM (Watne et al., 2018), while in this study, we still 104 corrected the wall loss of particle in each size bin measured by two synchronous SMPS (two SMPS run before and after Go: 105 PAM respectively). 106

Table S1. Comparison of results between blank and experimental groups (Dilution air and boiled water are two kinds of blank
 groups. The others are experimental groups.).

Experiment	OH Exposure (×10 ¹⁰ molecules·cm ⁻³ ·s)	OA Concentration ($\mu g/m3$)	Standard Deviation	Relative Standard Deviation
Dilution Air	0	-	-	-
	9.6	0.37	0.04	12%
Boiled Water	0	0.04	0.02	44%
	9.6	0.36	0.12	32%
Deep-fried Chicken	0	12.30	0.49	4%
	9.6	28.29	2.55	9%
Shallow-fried Tofu	0	13.56	0.68	5%
	9.6	21.70	1.08	5%
Stir-fried Cabbage	0	10.75	0.65	6%
	9.6	18.38	1.65	9%
Kung Pao Chicken	0	6.47	0.52	8%
	9.6	11.39	1.25	11%
GDI 20 km/h	0	0.40	0.01	3%
	7.8	19.68	1.48	8%
GDI 40 km/h	0	0.41	0.01	3%
	7.8	15.24	0.62	4%
GDI 60 km/h	0	0.42	0.02	5%
	7.8	23.23	4.00	17%

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Figure S1. Comparison of measured and estimated OH exposures during off-line OH exposure calibration using SO₂ for
 vehicle experiment (Temp:19~22°C, RH: 44~49%).



Figure S2. Comparison of measured and estimated OH exposures during off-line OH exposure calibration using SO₂ for
 cooking experiment (Temp:16~19°C, RH: 18~23%).



119

Figure S3. Previous performance tests for Go: PAM: (a) The schematic diagram of the Go: PAM reactor. (1) Quartz glass 120 flow reactor; (2) Exhaust flow; (3) Processed sample flow; (4) Humidified oxidant flow; (5) Sample flow; (6) Gas distributor 121 plate (Watne et al., 2018). (b) SO₂ added in turn in the "sample flow" (flow 5) and the "oxidant flow" (flow 4), and sampled 122 from "processed sample flow" (Watne et al., 2018). (c) Modeled fractional fates of LVOCs loss as a function of the equivalent 123 photochemical age in the Go: PAM (Li et al., 2019). (d) The particle penetration (Pp) as a function of the particle mobility 124 125 diameter (Dp) in Go: PAM. The solid line and shaded area represent the average and one standard deviation of the five different mass loadings of the nebulized ammonium sulfate particles (39-258 µg/m3), respectively. The dashed black line 126 represents 100% of particle penetration. The values for the first two size bins (6.04 and 6.98 nm) were extrapolated due to 127 128 low signal to noise ratio (Watne et al., 2018).



130 Figure S4. Mass spectra of PMF- resolved POA and SOA factors for deep-fried chicken groups.









Figure S6. Mass spectra of PMF- resolved POA and SOA factors for stir-fried cabbage vegetable groups.



Figure S7. Mass spectra of PMF- resolved POA and SOA factors for Kung Pao chicken groups.



Figure S8. Diagnostic plots of the PMF analysis for deep-fried chicken groups. The following plots are shown (a) Q/Qexp vs
number of factors; (b) Q/Qexp vs. fpeak for the solution with optimal number of factors; (c) mass fraction of PMF factors vs.
fpeak; (d) the distribution of scaled residuals for each m/z; (e) comparison of the reconstructed and measured total organic
mass; (f) time series of the residual of PMF solutions; (g) time series of Q/Qexp; (h) the Q/Qexp vs. m/z.



Figure S9. Diagnostic plots of the PMF analysis for shallow-fried tofu groups. The following plots are shown (a) Q/Qexp vs
number of factors; (b) Q/Qexp vs. fpeak for the solution with optimal number of factors; (c) mass fraction of PMF factors vs.
fpeak; (d) the distribution of scaled residuals for each m/z; (e) comparison of the reconstructed and measured total organic
mass; (f) time series of the residual of PMF solutions; (g) time series of Q/Qexp; (h) the Q/Qexp vs. m/z.



151

Figure S10. Diagnostic plots of the PMF analysis for stir-fried cabbage groups. The following plots are shown (a) Q/Qexp vs number of factors; (b) Q/Qexp vs. fpeak for the solution with optimal number of factors; (c) mass fraction of PMF factors vs. fpeak; (d) the distribution of scaled residuals for each m/z; (e) comparison of the reconstructed and measured total organic mass; (f) time series of the residual of PMF solutions; (g) time series of Q/Qexp; (h) the Q/Qexp vs. m/z.



157

Figure S11. Diagnostic plots of the PMF analysis for Kung Pao chicken groups. The following plots are shown (a) Q/Qexp vs number of factors; (b) Q/Qexp vs. fpeak for the solution with optimal number of factors; (c) mass fraction of PMF factors vs. fpeak; (d) the distribution of scaled residuals for each m/z; (e) comparison of the reconstructed and measured total organic mass; (f) time series of the residual of PMF solutions; (g) time series of Q/Qexp; (h) the Q/Qexp vs. m/z.

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