



Supplement of

Secondary organic aerosol production from local emissions dominates the organic aerosol budget over Seoul, South Korea, during KORUS-AQ

Benjamin A. Nault et al.

Correspondence to: Jose L. Jimenez (jose.jimenez@colorado.edu)

The copyright of individual parts of the supplement might differ from the CC BY 4.0 License.

34 S1. KORUS-AQ Overview

35	Table S1. List of NASA DC-8 research flights and date of take-off. Unless noted, the take-off
36	dates are different than the local dates since the data was recorded in UTC. We document the
37	research flights with the UTC dates to correspond with the data repository (Aknan and Chen,
38	2018).

Research Flight	Date of Take-off	Regions Sampled	Number of Seoul
Number			Missed Approaches
01	01/May/2016	Jeju jetway (×2)	3
02	03/May/2016	Yellow Sea, Jeju jetway	3
03	04/May/2016	Jeju jetway	2
04	06/May/2016	Busan jetway (×2)	3
05	10/May/2016	Jeju jetway, other ^b	2
06	11/May/2016	Yellow Sea, other ^c	3
07	12/May/2016	Yellow Sea, other ^d	0
08	16/May/2016	Jeju jetway, Busan jetway	3
09	17/May/2016	Yellow Sea, Busan jetway	3
10	19/May/2016	Busan jetway (×2)	3
11	21/May/2016	Yellow Sea	3
12	24/May/2016	Yellow Sea	2
13	26/May/2016 ^a	Jeju Jetway	2
14	29/May/2016	Yellow Sea, Busan jetway	4
15	30/May/2016	Yellow Sea, Jeju jetway	3
16	01/June/2016	Busan jetway, Jeju jetway	3
17	02/June/2016	Busan jetway, Jeju jetway	3
18	04/June/2016	Yellow Sea, other ^e	5
19	08/June/2016	Busan jetway (×2)	3
20	09/June/2016	Jeju jetway, other ^b	2

^aFor RF13, the DC-8 took-off after 00:00 UTC, corresponding to the date in local time and UTC

40 time being the same.

41 ^bThe DC-8 sampled south of the Korean peninsula.

42 ^cThe DC-8 sampled east of Seoul to the Sea of Japan.

- 43 ^dThe DC-8 sampled the Sea of Japan.
- ^eThe DC-8 remained in the greater Seoul area to sample point sources.

46 **Table S2.** Description of the geographical locations used in Figure 1 and throughout the text, and

47 shown in Figure S1..

45

Location	Lat Min (°N)	Lat Max ($^{\circ}N$)	Lon Min (°E)	Lon Max ($^{\circ}E$)
Seoul	36.8	37.6	124.6 ^a	128.0
Yellow Sea			124.0	126.0
Jeju jetway	34.2	36.8	126	127
Busan jetway	35.2	36.8	128.6	129.9

48 ^aThis value was chosen to include the Seoul outflow observed during RF11 and RF18.



51 Figure S1. Geographical regions shown in SI Table 1. Note, the Seoul box is extended into the

- 52 Yellow Sea to capture the outflow of Seoul emissions for two flights (RF11 and RF18).

54 S2. CU-AMS Sampling and Calibration

55 After almost every flight, the ionization efficiency (IE) was calibrated (Figure S3) using the single particle technique. Briefly, air containing 150 - 250 particles/cm³ of NH₄NO₃, of 400 56 57 nm (mobility diameter, sized with a differential mobility analyzer, TSI model 3080, that was 58 installed in the same rack as the CU-AMS) was sampled by the AMS. Thresholds of 4 (m/z 30) or 59 3 (m/z 46) ions per event were selected to produce a low, but detectable background (typically ~7 60 events/cm³ background). An event would be recorded, after evaporation and ionization of NH₄NO₃ particle, if at least 4 (m/z 30) or 3 (m/z 46) ions were observed. These values were analyzed using 61 62 the ToF AMS Ionization Efficiency Calibration Panel for ET, v1.0.5F (http://cires1.colorado.edu/jimenez-63

64 <u>group/ToFAMSResources/ToFSoftware/index.html#ToF_IE_Cal</u>), to process the data and 65 calculate IE and IE/AB (AB is air beam). Typical values during KORUS-AQ, for 400 nm (mobility 66 diameter) NH₄NO₃ calibrations were the following: 10 baseline segments and minimum and 67 maximum ions per particle values of 1 and 200. During KORUS-AQ, the average IE/AB was 8.10(\pm 0.64)×10⁻¹³ ions/molecule of nitrate, which leads to an overall 10% variability for this value 69 during the whole campaign. Further details about using single particle technique for IE/AB 70 calibrations can be found in Nault et al. (2016).

These IE calibrations also provided relative ionization efficiency (RIE) calibrations of NH₄ after nearly every flight, as well (Figure S3), along with the NO⁺ and NO₂⁺ ratios of ammonium nitrate, which are useful to estimate particle organic nitrate concentrations, as detailed in Fry et al. (2013). The SO₄ and Chl RIEs were measured about once every week, and the interpolated values were used for the SO₄ and Chl concentrations. For the organic aerosol, we used an RIE of 1.4 (Jimenez et al., 2016; Xu et al., 2018). Finally, to test the effects of solution mixtures on RIE for 77 SO₄ and NH₄, we made calibration solutions ranging from 0 - 100% NH₄NO₃, with the balance 78 coming from (NH₄)₂SO₄. We find no effects, both on the calculated NH₄ balance (Figure S6), 79 when using the NH₄ and SO₄ RIE's from the pure calibration, and on the recalculated NH₄ (SO₄) 80 RIE when keeping a constant SO₄ (NH₄) RIE from the pure calibrations (Figure S6). The 81 consistency in the NH₄ balance, as observed in prior studies (Docherty et al., 2011; Jimenez et al., 82 2016), and the high precision (3% precision in all calculations) provides further confidence in the 83 stability of the RIEs for the species in calculating their mass in mixed particles, and indicates that 84 there are no effects on the RIE with changing composition, and, thus, CE (Jimenez et al., 2016).

Also, the IE calibrations performed after each flight provided an opportunity to calculate the effect of pNO₃ on producing a small artifact CO_2^+ signal, as detailed in Pieber et al. (2016), and of pNO₃ on producing small artifact HCl⁺ and Cl⁺ signal, as detailed in Hu et al. (2017a) (Figure S3). The CU-AMS data has been corrected for these small effects. The corrections were typically 1% of CO_2^+ and 0.8% Chl.

90 Three different lens transmission calibrations to characterize the high end of the AMS 91 transmission curve were performed: (1) comparing the NH4NO3 mass measured with the CPC and 92 the CU-AMS between 200 – 450 nm (mobility diameter, d_m); (2) comparing the number of 93 particles measured with the CPC and the CU-AMS between 300 - 450 nm (d_m) using the single 94 particle vaporization technique detailed above; and (3) comparing the (NH₄)₂SO₄ mass measured 95 with the CPC and the CU-AMS between 250 - 450 nm (d_m), normalizing to the value at 250 nm. 96 The NH₄NO₃ diameters were converted to vacuum aerodynamic diameters (d_{va}), as discussed in 97 DeCarlo et al. (2004). As seen in Figure S4, both techniques show good agreement for the particle 98 transmission, and this transmission is similar to the recommended transmission curve in the 99 literature (Knote et al., 2011; Hu et al., 2017b). For this curve, it is assumed that the transmission

linearly increases from 0 – 100% between 40 – 100 nm (d_{va}) (Zhang et al., 2004), remains 100% between 100 – 550 nm (aerodynamic diameter), and decreases linearly from 100 – 0% between 550 nm – 1500 nm (d_{va}). This leads to a 50% cut-off of ~900 nm (d_{va}) during KORUS-AQ.

103 The particle sizing in the AMS Particle Time-of-Flight (PToF) mode was calibrated with 104 PSLs, ranging from 70 - 700 nm (geometric diameter) (Figure S5). This calibration was compared 105 against the velocities calculated from data collected during the NH4NO3 lens transmission 106 measurements. As seen in Figure S5, these two different methods to calibrate the PToF velocity 107 show comparable results, falling within the 95% confidence interval of the PSL calibration. The 108 fact that both PToF calibrations agree, and that the SMPS used for the AN calibrations showed 109 less than 2 nm deviation from the nominal PSLs diameters at all sizes increases our confidence in 110 accuracy of the IE calibration described above, and in particular on lack of evaporation of NH4NO3 111 after its size selection in the DMA.

112 Finally, the vaporizer power, and thus, temperature, was calibrated by using monodisperse 113 NaNO₃ particles of $d_m = 350$ nm (Figure S8), as recommended by Williams (2010) and Hu et al. 114 (2017b). This method is more accurate than relying on the temperature reported by the 115 thermocouple on the AMS vaporizer, which can often be unreliable (Williams, 2010; Hu et al., 116 2017b). The general idea is to increase the vaporizer power between $\sim 1 - 7$ W and locate where 117 the NaNO₃ full-width half maximum nearly remains constant, indicating that the vaporizer 118 temperature is ~600°C and allowing for maximum peaks in OA, pNO₃, and SO₄ while minimizing 119 the influence of refractory species (Williams, 2010; Hu et al., 2017b).





Local Time

121	Figure S2.	Example time series	of the CU-AMS	sampling scheme	during KORUS-AQ.	Though
	1 1 1 0					

the final 8 s of each minute are dedicated to ePToF, some of the time is ued by the computer in

saving the 6 s of closed and 46 s of open signal and ePToF signal; therefore, only 3 - 5 s of ePToF

signal is actually recorded. The approximate saving time are shown as white spaces.



126

Figure S3. Time series of the (a) the Single Ion (SI) at take-off for each flight; (b) the air beam (AB, dark grey), ionization energy (IE, light grey), and IE/AB (middle grey) for each calibration;

129 (c) the relative ionization energies (RIE) for ammonium (NH₄), sulfate (SO₄), and chloride (Chl)

130 for each calibration; (d) the ratios of different ions for each calibration; and, (e) measured artifact

131 signal ratios for CO₂⁺/pNO₃ "Pieber effect" (Pieber et al., 2016) and (Cl⁺ + HCl⁺)/pNO₃ "Hu

132 efffect" (Hu et al., 2017a) effects from each calibration.



133

Aerodynamic Diameter (nm)

134 Figure S4. Measured transmission percentage of ammonium nitrate and ammonium sulfate versus vacuum aerodynamic diameters (nm) during KORUS-AQ. The green dashed-line is the expected 135 transmission curve for the CU-AMS from the literature (Knote et al., 2011; Hu et al., 2017b). The 136 137 black data represents the ammonium nitrate transmission curve using mass closure, from an 138 experiment conducted on 09/May/2016. The blue data represents the ammonium nitrate 139 transmission curve using single particle ("event trigger") number closure, from an experiment 140 conducted on 17May/2016. The red data represents the ammonium sulfate transmission curve 141 using mass, from an experiment conducted on 06/May/2016. Finally, the error bars represent 1σ 142 variability for the transmission at each size.



145 **Figure S5.** Particle velocity (m/s) inside the AMS vacuum chamber (after exiting the aerodynamic

lens) versus vacuum aerodynamic diameter (nm) calibrations for the ePToF mode, using PSLs(black). Solid red line is the fit to the PSLs. The ammonium nitrate measured for the mass closure

148 transmission curves (Figure S4) for comparison to the PSL values.







Figure S7. (a) Plot of NH₄ RIE, keeping SO₄ RIE constant, versus the molar fraction of pNO₃ measured in the solution, for calibration solutions of varying mixtures of NH₄NO₃ and (NH₄)₂SO₄. (b) Same as (a), but for SO₄ RIE and keeping NH₄ RIE constant. For both figures, the black dots are the values from the calibrations, the thick red line is the average of all the values, and the shaded red area is $\pm 1\sigma$.



Figure S8. (a) Full-width half-maximum (FWHM) of NaNO₃ size distributions in PToF mode (b)
 vs. different vaporizer power inputs. See text for further details.

166 S3. Application of Positive Matrix Factorization (PMF)

167 Positive matrix factorization analysis (PMF, performed using the CU-Boulder PMF 168 Evaluation Tool **PET-Panel** v3.00A, http://cires1.colorado.edu/jimenez-169 group/wiki/index.php/PMF-AMS_Analysis_Guide#PMF_Evaluation_Tool_Software) (Ulbrich 170 et al., 2009) was used to apportion the total OA aerosol into several components. PMF was run on 171 the combined CU-AMS 1 min organic ion matrix for all RFs together during KORUS-AQ. A 6-172 factor solution was derived with an FPEAK value of 0. Based on comparisons with reference mass 173 spectra from the AMS high-resolution spectral database (http://cires1.colorado.edu/jimenez-174 group/HRAMSsd/#Ambient), comparisons of time series (Figure S9), and correlations with other 175 trace species (Figure S11), the factors were recombined into more-oxidized, oxidized organic 176 aerosol (MO-OOA), less-oxidized, oxidized aerosol (LO-OOA), and hydrocarbon-like organic 177 aerosol (HOA) (Figure S10). HOA correlated with primary emissions (e.g., NO_x , various 178 hydrocarbons) whereas LO-OOA and MO-OOA correlated with secondary photochemical 179 products (e.g., CH₂O, PAN, pNO₃, SO₄). Here, primary OA is defined as the HOA factor and total 180 oxidized OA (OOA) as the LO-OOA plus MO-OOA factors. OOA is assumed to be dominantly 181 composed of secondary organic aerosol, which is supported by its strong correlation with other 182 secondary photochemical products as documented in the paper, as well as by many prior studies 183 (e.g., Jimenez et al., 2009; and references therein).



185

Figure S9. Example time series of the 3 PMF ((a) HOA, (b) LO-OOA, and (c) MO-OOA) results (left axes) and some species that correlate with the corresponding PMF results (right axes) from

188 RF14. The morning and afternoon overpasses over Seoul, South Korea, are shown.



Figure S10. Mass spectra for PMF solution (a) HOA, (b) LO-OOA, and (c) MO-OOA for all of

192 KORUS-AQ.



194 Figure S11. Pearson correlation coefficients for HOA (grey, top), LO-OOA (light pink, middle), 195 and MO-OOA (dark pink, bottom) factors versus species listed in x-axis. The background colors 196 represent the dominant group of sources of the correlating species. The yellow in the far right

197 indicates other PM₁ components measured by the CU-AMS.

199 S4. Oxidation Flow Reactor (OFR) Sampling



Figure S12. Schematic of the OFR sampling during KORUS-AQ. UV lamp is represented by the purple light in the OFR. Size and type of tubing is represented in figure, where ID is internal diameter and SS is stainless steel. Tubing distances were always as short as feasible and often shorter than represented, but they are stretched in this drawing for clarity



Figure S13. 3D rendition of the computer model of the ¹/₂" press fitted stainless steel inlet, coated in SilcoNert (SilcoTek Co, Bellefonte, PA), used in the inlet of the OFR during KORUS-AQ, to

avoid "short-circuiting" between the inlet and outlet of the OFR.



209

Figure S14. (left) Measured OH_{exp} from the decay of CO in ambient air (measured by the DACOM instrument, see text) and OFR output air (measured by the Picarro instrument) and (right) measured OH_{exp} from the decay of CO from a calibration cylinder versus calculated OH_{exp} using the predictive expression in Peng et al. (2015). The calibration factor determined by this analysis was similar to past studies (Palm et al., 2016) and was applied to all data shown in this paper.



Figure S15. (left) Frequency distribution of water vapor below 2 km during KORUS-AQ. (right)
Normalized histogram of measured OH reactivity (OHR) below 2 km during KORUS-AQ.

216



220

Figure S16. Frequency distribution of the ambient (black), OFR (blue), and difference between OFR and ambient temperature (grey) (°C). Vertical lines show the mean (long-dashed) and median

223 (short-dashed) temperatures for the ambient, OFR, and difference between the two.



Figure S17. Comparison of organic (green), sulfate (red), nitrate (blue), and ammonium (orange) aerosol sampled through the OFR, with lights off, versus ambient aerosol. Under these conditions the OFR is just acting as a thermal denuder (e.g. Huffman et al., 2009), leading to evaporation of some aerosols due to increased temperature in the aircraft cabin vs. outside. In addition, small particle losses in lines and the OFR are observed for sulfate, which is generally non-volatile. See text for further details and discussion.





Figure S18. (a) Scatter plot of OFR SO₄ Enhancement (OFR – Ambient) versus calculated SO₄
 produced, using SO₂ observations, estimated OH_{exp}, and condensation fate correction. (b) Time
 series of OFR SO₄ (black squares), ambient SO₄ (light grey), and calculated SO₄ (dark red) for the
 RF11 flight.

237 Analysis of CS Values for KORUS-AQ

238 If we used the condensational sink from just the ambient data, which neglects the added particle 239 surface area formed in the OFR as described in Section 2.4 and Eq. 1 (Ortega et al., 2016; Palm et 240 al., 2016, 2017, 2018), the agreement between calculated and measured SO₄ enhancement decreases to a slope of 0.74 ($R^2 = 0.28$), indicating that the condensational sink is likely too low. 241 242 This suggests that, to first order, the aerosol surface area, estimated from observations and Eq. 1 243 (in the main paper), provides a reasonable estimate of the condensational sink within the OFR 244 during KORUS-AQ. Thus, similar to other studies (Ortega et al., 2016; Palm et al., 2016, 2017, 245 2018), we find, at the typical OH_{exp} in the OFR, that 50 - 60% of the oxidized condensable organic 246 gases are condensing onto aerosol, with 20 - 25% undergoing further reactions with OH leading 247 to highly volatile compounds, 8 – 13% exiting the OFR prior to condensing on aerosol, and 12%

248 condensing to the wall (Figure S19). Note that the further reactions with OH are not relevant for





Figure S19. (a) Observed normalized frequency and cumulative frequency of OH Exposure observed during KORUS-AQ in the OFR. (b) Calculated fate of the SO₂ oxidized in the OFR

versus OFR OH Exposure. (c) Calculated fate of low-volatility condensable vapors (formed from
VOC oxidation) versus OFR OH Exposure. For (b) and (c), the losses include flowing through the
OFR without condensing onto aerosol (black), condensing onto the wall (red), condensing onto
the aerosol (assuming a median value of 85.8 s, green), and reacting with OH enough to make it
too volatile to condense onto aerosol (blue).

259 S5. Calculation of Photochemical Age over Seoul, South Korea

The photochemical clock calculations used throughout this work are described here. The rate constants used for these clocks are located in SI Table 3. For the NO_x/NO_y photochemical clock (e.g., Kleinman et al., 2007) (herein referred to as the NO_x photochemical clock), Eq. S1 is used, with the updated rate constant from Mollner et al. (2010).

$$264 t = \frac{ln\left(\frac{NO_X}{NO_y}\right)}{k_{OH+NO_2}[OH]} (S1)$$

where *t* is the time, in days, [OH] is assumed to be 1.5×10^6 molecules/cm³ (for standarization), and NO_x and NO_y are the chemiluminescence measurements. The NO_x clock is used for photochemical ages less than 1 day to (1) reduce the effect of loss of HNO₃ and other oxidized reservoirs due to deposition (lifetime ~6 hours) (Neuman et al., 2004; Nguyen et al., 2015; Romer et al., 2016) and (2) to ensure that t was still sensitive (and precise) to the NO_x and NO_y concentrations (~20% of NO_x still remaining at *t* = 1 day).

For the aromatic photochemical clock over Seoul, the more reactive aromatics (ethylbenzene in the denominator) are utilized, which should be more sensitive to the short photochemical aging observed over Seoul (Parrish et al., 2007), along with Eq. SS2.

274
$$t = -\frac{1}{[OH] \times \left(k_{aromatic_i} - k_{ethylbenzene}\right)} \times \left(ln\left(\frac{aromatic_i(t)}{ethylbenzene(t)}\right) - ln\left(\frac{aromatic_i(0)}{ethylbenzene(0)}\right)\right)$$
(S2)

where *t* is the time, in hours, the *k*'s are the corresponding OH rate constants for each aromatic compound (SI Table 3), and the third term $\left(\ln\left(\frac{\operatorname{aromatic}_{i}(0)}{\operatorname{ethylbenzene}(0)}\right)\right)$ corresponds to the emission ratios for those two aromatic compounds. Similar to the NO_x clock, we assume [OH] = 1.5×10^{6} molecules/cm³ for standardization. The aromatics measurements used in this calculation are from WAS. 280 To evaluate which aromatic compounds to use in the clock, the behavior of the ratios of 281 each aromatic compound with ethylbenzene versus the three missed approaches (morning, noon, 282 and afternoon) over Seoul during KORUS-AQ (Figure S20) are compared. The idea is that if the 283 2 aromatic compounds are co-emitted, the ratios should be removed proportionally to their OH 284 rate constants. E.g. for faster reacting compounds (e.g., o-xylene), the ratio to ethylbenzene should 285 decrease with time as more o-xylene was consumed, compared to ethylbenzene, by OH (de Gouw 286 et al., 2017). On the other hand, for slower reacting compounds (e.g., toluene), the ratio to 287 ethylbenzene should increase with time as more ethylbenzene was consumed by OH. Also, this 288 analysis provides an indication of which ratios would provide meaningful results throughout the 289 entire day (de Gouw et al., 2017). Ideally, there should be a decrease with each later missed 290 approach, and not a leveling off after two missed approaches (e.g., the trimethylbenzenes and 291 ethyltoluenes). Only the m+p-xylene/ethylbenzene and o-xylene/ethylbenzene ratios meet this 292 criterion. Finally, to determine the emission ratios, we calculated what the m+p-293 xylene/ethylbenzene and o-xylene/ethylbenzene ratio was for observations where the NO_x 294 photochemical was less than 0.07 days (corresponding to less than 10% of either species being 295 consumed). Comparing these two aromatic clocks to the NO_x photochemical clock (Figure S21), 296 a similar agreement between the two aromatic clocks with the NO_x photochemical clock was 297 observed, providing confidence in using all three clocks to calculate photochemical age to evaluate 298 OA production over Seoul. For the remainder of the paper, we mainly use the NO_x photochemical 299 clock to eliminate the uncertainty of the emission ratios, unless otherwise noted.

Finally, for observations over the Yellow Sea, the aromatic clock (Eq. SS2) was used, but benzene and toluene were used since these air masses are more photochemically processed (Parrish et al., 2007). For the emission ratios, values reported by Yuan et al (2013) were used.



Figure S20. Comparison of various aromatic compounds/ethylbenzene ratios sampled over Seoul,

306 South Korea, during KORUS-AQ. The ratios are normalized by the morning ratios.



Figure S21. Binned scatter plot of the aromatic photochemical clock ages versus NO_x photochemical clock ages for all observations over Seoul. All ages are normalized to $OH = 1.5 \times 10^6$ molecules/cm³.

Reaction	Rate Constant (cm ³ /molecules/s)	Reference	
Inorganic			
СО	2.28×10 ^{-13,a}	Sander et al. (2011)	
NO ₂	$1.23 \times 10^{-11,a}$	Mollner et al. (2010)	
SO_2	$8.94 \times 10^{-13,a}$	Atkinson et al. (2004)	
Alkanes			
Ethane	$6.9 \times 10^{-12} \times \exp(-1000/T)$	Atkinson et al. (2006)	
Propane	$7.6 \times 10^{-12} \times \exp(-585/T)$	Atkinson et al. (2006)	
n-Butane	$9.8 \times 10^{-12} \times \exp(-425/T)$	Atkinson et al. (2006)	
i-Butane	$1.17 \times 10^{-17} \times T^2 \times exp(213/T)$	Atkinson (2003)	
n-Pentane	$2.52 \times 10^{-17} \times T^2 \times exp(158/T)$	Atkinson (2003)	
i-Pentane	3.6×10 ⁻¹²	Atkinson (2003)	
n-Hexane	$2.54 \times 10^{-14} \times T \times exp(-112/T)$	Atkinson (2003)	
Methyl-cyclopentane	7.65×10^{-12}	Sprengnether et al. (2009)	
Cyclohexane	$3.26 \times 10^{-17} \times T^2 \times exp(262/T)$	Atkinson (2003)	
Methyl-cyclohexane	9.43×10 ⁻¹²	Sprengnether et al. (2009)	
n-Heptane	$1.95 \times 10^{-17} \times T^2 \times exp(406/T)$	Atkinson (2003)	
n-Octane	$2.72 \times 10^{-17} \times T^2 \times exp(361/T)$	Atkinson (2003)	
n-Nonane	$2.53 \times 10^{-17} \times T^2 \times exp(436/T)$	Atkinson (2003)	
n-Decane	$3.17 \times 10^{-17} \times T^2 \times exp(406/T)$	Atkinson (2003)	
Alkenes	•		
Ethylene	7.84×10 ^{-12,a}	Atkinson et al. (2006)	
Propene	$2.86 \times 10^{-11,a}$	Atkinson et al. (2006)	
1-butene	$6.6 \times 10^{-12} \times \exp(465/T)$	Atkinson et al. (2006)	
i-butene	$9.4 \times 10^{-12} \times \exp(505/T)$	Atkinson et al. (2006)	
cis-butene	$1.1 \times 10^{-11} \times \exp(485/T)$	Atkinson et al. (2006)	
trans-butene	$1.0 \times 10^{-11} \times \exp(553/T)$	Atkinson et al. (2006)	
1,3-butadiene	$1.48 \times 10^{-11} \times exp(448/T)$	Atkinson and Arey (2003)	
Aromatics			
Benzene	$2.3 \times 10^{-12} \times \exp(-190/\text{T})$	Atkinson et al. (2006)	
Toluene	$1.8 \times 10^{-12} \times \exp(340/T)$	Atkinson et al. (2006)	
Ethylbenzene	7×10^{-12}	Atkinson and Arey (2003)	
Isopropylbenzene	6.3×10^{-12}	Atkinson and Arey (2003)	
n-propylbenzene	5.8×10^{-12}	Atkinson and Arey (2003)	
Styrene	5.8×10^{-11}	Atkinson and Arey (2003)	
m+p-xylene	$1.87 \times 10^{-11,b}$	Atkinson and Arey (2003)	
o-xylene	1.36×10^{-11}	Atkinson and Arey (2003)	
1,3,5-trimethylbenzene	$1.32 \times 10^{-11} \times \exp(450/T)$	Bohn and Zetzsch (2012)	
1,2,3-trimethylbenzene	$3.61 \times 10^{-12} \times \exp(620/T)$	Bohn and Zetzsch (2012)	
1,2,4-trimethylbenzene	$2.73 \times 10^{-12} \times \exp(730/T)$	Bohn and Zetzsch (2012)	
3-Ethyltoluene	1.2×10^{-11}	Atkinson and Arey (2003)	
4-Ethyltoluene	1.2×10^{-11}	Atkinson and Arey (2003)	
S/IVOCs			

Table S3. Rate constants used throughout this study. Unless noted otherwise, rate constants
 without temperature dependence only have a value measured at 298 K.

S/IVOC	2×10^{-11}	Ma et al. (2017)
Biogenics		
Isoprene	$2.7 \times 10^{-11} \times \exp(390/T)$	Atkinson et al. (2006)
α-pinene	$1.2 \times 10^{-11} \times \exp(440/T)$	Atkinson et al. (2006)
β-pinene	$1.55 \times 10^{-11} \times \exp(467/T)$	Atkinson and Arey (2003)
Radicals		
$NO + RO_2$	$2.8 \times 10^{-12} \times \exp(300/\text{T})$	Sander et al. (2011)
$HO_2 + RO_2$	$4.1 \times 10^{-13} \times \exp(750/T)$	Sander et al. (2011)
$RO_2 + RO_2$	$9.5 \times 10^{-14} \times \exp(390/T)$	Sander et al. (2011)

^aShowing the rate constant at 298 K, 1013 hPa. However, for this study, we used the temperature

and pressure dependent formulation listed in each respective reference.

³¹⁵ ^bThis is the average of m-xylene and p-xylene rate constants.

316

317 S6. Potential SOA Calculations

318 To determine the amount of SOA produced from the observed precursors, Eq. S3 was used, 319 where Y is the stoichiometric aerosol yield for each hydrocarbon (RH) species *i*, similar to other 320 studies (e.g., Zhao et al., 2014). The updated yields from Ma et al. (2017) were used, which 321 incorporate a correction for the gas-phase partitioning of semi-volatile compounds to chamber 322 walls (Krechmer et al., 2016). Since there were no direct measurements of S/IVOC concentrations, 323 an estimated (Robinson et al., 2007; Dzepina et al., 2009) relationship between the amount of gas-324 phase S/IVOC co-emitted with POA at the typical temperatures (~20°C) and OA mass 325 concentrations (~10 μ g sm⁻³) observed over Seoul were used. The POA is taken from Figure 5b 326 and is within the range of values observed in other urban environments (Zhang et al., 2005; Hayes et al., 2013; Ait-Helal et al., 2014; Kim et al., 2018) (13 μ g sm⁻³ ppmv⁻¹ in Seoul versus 4.5 – 28.8 327 328 $\mu g \text{ sm}^{-3} \text{ ppmv}^{-1}$ in other studies).

329
$$P(OA) = \sum_{i} Y_i \times \Delta R H_i$$
(S3)

330
$$\Delta RH = \frac{RH(t)}{e^{(-k[OH]t)}} - RH(t)$$
(S4)

331 The hydrocarbons measured on the DC-8 were the concentrations at time, *t*; thus, Eq. SS4 332 was used, which takes into account the amount of OH that oxidized the hydrocarbon ($OH_{exp} =$ [OH]*t*) between emissions and measurement, and *k* is the OH rate constant for each specifichydrocarbon (SI Table 3).

335 Finally, to determine the fate of the RO₂ radical in the reactions over Seoul (high/low NO_x 336 regime), and thus, what aerosol yields to use, the RO₂ lifetime with reaction of NO, HO₂, and RO₂ 337 versus photochemical age was calculated (Figure S22). The measured NO and HO₂ was used in 338 the calculations, we assumed RO₂ was approximately the same concentration as HO₂ in this 339 calculation (Thornton et al., 2002), and the rate constants in SI Table 3 were used to calculated the 340 lifetime and fractional fate of RO₂. The fate of RO₂ with autoxidation is not included as the rate is 341 still uncertain (Crounse et al., 2013) and it should be less important in highly polluted 342 environments such as Seoul, especially at the lower photochemical ages (< 0.5 eq. days) where 343 most SOA is observed to be formed. The dominant sink of RO2 over Seoul during KORUS-AQ is 344 the reaction with NO, suggesting that the SOA yields for "high NO" conditions should be used to 345 describe the production of SOA.





Figure S22. (left) Lifetime of RO₂ due to reactions with NO (red), HO₂ (blue), and RO₂ (grey) versus NO_x photochemical clock, normalized by OH = 1.5×10^6 molecules/cm³. (right) Fraction of RO₂ reacting with NO (red), HO₂ (blue), or RO₂ (red) versus NO_x photochemical clock, normalized by OH = 1.5×10^6 molecules/cm³. Values are calculated using observations over Seoul, South Korea, during KORUS-AQ, and RO₂ is assumed to be approximately equal to HO₂

353 (Thornton et al., 2002).

354 S7. FLEXPART Source Analysis

355 Source contributions have been estimated using Lagrangian backtrajectory calculations with the 356 FLEXPART-WRF model (Brioude et al., 2013) in version 3.3.1, driven by meteorological output 357 from NCEP GFS (NCEP) analyses downscaled to 5 km horizontal resolution using the Weather 358 Research and Forecasting (WRF) model (Skamarock et al., 2008) in combination with 359 the CREATE emission inventory (Woo et al., 2013). Approximately 20,000 parcels are released 360 in 1 min intervals from the then-current location of the DC-8 during its research flights and parcel 361 trajectories are followed back in time for 24 hours. The total time parcels spent in the lowermost 362 100 m—as surrogate for air having contact with an emission source at the ground—is recorded (residence time, [s kg⁻¹ m³]) and then folded with the emission fluxes ([kg m⁻² s⁻¹]) given by the 363 364 CREATE inventory for different compounds and source regions. This delivers an estimate of the 365 source contribution (as increment in volume mixing ratio at the receptor, i.e., the DC-8 location) 366 of the emissions of a given compound from a given region, assuming a perfect transport simulation 367 and an inert compound.

368 S8. Intercomparisons of CU-AMS with Other Measurements on the NASA DC-8

369 We evaluate the measurement comparisons of the CU-AMS versus other aerosol measurements on-board the DC-8 during KORUS-AO. We start with the mist chamber / ion 370 371 chromatograph instrument (MC/IC), which has a comparable size cut as the AMS. The comparisons for SO₄ show good correlation ($R^2 = 0.76$) and slope close to 1 (0.95) (Figure S23). 372 373 The higher scatter for the MC/IC is thought to arise from the lag and smearing in the measurements 374 that has been observed in prior studies (TAbMET, 2009). For example, the correlation between instruments without lag and smearing have R^2 of 0.87 - 0.91 (CU-AMS versus extinction and CU-375 376 AMS vs K-AMS for certain RFs). If the MC/IC and CU-AMS SO₄ measurements are averaged to 377 the sampling frequency of the University of New Hampshire filters (not shown), the R^2 improves 378 (0.82) with no impact on the slope.

The comparison between the UNH filters and CU-AMS SO₄ shows higher R^2 (0.86) but 379 lower slope (0.80), compared to MC/IC vs. CU-AMS. The higher R² is likely due to longer 380 381 averaging time and lack of smearing that occurred with the MC/IC. As a comparison, the R^2 382 between MC/IC and filters are 0.84. The lower slope for the filters than the MC/IC is thought to 383 be due to the different size cut-offs for the two measurements. For the filters, the upper size cut-384 off is ~4 µm (McNaughton et al., 2007); whereas, the upper size cut-off for the MC/IC is 385 comparable to the AMS aerosol size cut-off (~ 1 µm aerodynamic). This means that the filter samples may include SO4²⁻ from sea salt (sodium and calcium) and dust (calcium) (Heo et al., 386 387 2009; Kim et al., 2016; Heim et al., 2018). This is shown in Figure S24 and described in detail in 388 Heim et al. (2018). Heim et al. (2018) found that dust dominated supermicron aerosol for approximately half of the campaign, and during these periods, supermicron SO4²⁻ accounted for 389 390 ~50% of the total SO_4^{2-} (sub plus supermicrong). Taken together, the comparisons of SO_4 mass

concentrations from the CU-AMS from these two different methods (filter and MC/IC) indicate
that the CU-AMS quantitatively captures the concentrations of SO₄.

0,7

393 Next, we compare the non-refractory species concentrations measured by the CU- and K-394 AMS. Intercomparisons between these two measurements for a few flights have been presented in 395 prior publications (Hu et al., 2018a, 2018b). The K-AMS used a capture vaporizer, which leads to 396 CE of ~1 for all ambient species (Hu et al., 2017a, 2017b; Xu et al., 2017). Here, we investigate the entire campaign. As shown in Figure S25, $R^2 > 0.80$ for all five species, and all slopes fall 397 398 within $\pm 20\%$ of unity, which is within the combined uncertainty of both AMSs (~27\%). However, 399 at high concentrations (greater than $\sim 5 - 10 \ \mu g \ sm^{-3}$), the scatter between the two measurements 400 increases, and for some species (e.g., SO₄), there is a slight curvature in the comparisons, where 401 CU-AMS is greater than K-AMS. We believe this discrepancy originated from differences in 402 transmission vs. particle size through the aerosol inlet and focusing lens (Figure S26). In-field 403 calibrations showed that The K-AMS had 50% transmission at 615 nm (vacuum aerodynamic 404 diameter; DeCarlo et al. (2004)), compared to the CU-AMS 50% transmission occurring at 900 405 nm. The reasons for the smaller transmission of the K-AMS are likely related to the PCI design 406 (Bahreini et al., 2008, 2009) or possibly an underperforming aerodynamic lens in K-AMS (Liu et 407 al., 2007). It was found that, in general, the RFs could be split between RFs generally below the 408 K-AMS size cut-off (RFs 1 - 9, 11, 15, and 19) and above the size cut-off (RFs 10, 12 - 14, 16 - 14, 18, 20) (Figure S27). The slopes and R^2 greatly improves for the observations below the K-AMS 409 cut-off versus above (for slopes, 1.02 versus 0.84 and for \mathbb{R}^2 , 0.91 versus 0.82). 410

Finally, the ratios of the total AMS PM_1 masses measured by CU-AMS and K-AMS remain nearly constant about one (within ±11%) for the entire campaign and show no trend with estimated CE (for the standard vaporizer only) using the Middlebrook et al. (2012) algorithm (Figure S28). Thus, when accounting for transmission effects, the two AMSs agree to within 10%, and the CU-AMS agrees to within 20% with the other co-located aerosol mass concentration measurements (filters and MC/IC) on the DC-8. This provides overall confidence in the calculated CE for the standard vaporizer (Middlebrook et al., 2012), RIE, and transmission of PM₁ for the CU-AMS measurements.

419 Besides directly comparing species mass, another well-established method to investigate 420 aerosol instrument quantification is to compare the measured PM1 mass (CU-AMS plus BC from 421 SP2) versus the submicron extinction measured using methods described in Section 2.3.2 422 (nephlometer for scattering and absorption by PSAP) (e.g., DeCarlo et al., 2008). During KORUS-AQ, the slope between mass and extinction is 6.00 m² g⁻¹ (Figure S29) with an R² of 0.87. The 423 424 high correlation and similar slope compared to prior comparisons (Hand and Malm, 2007; DeCarlo 425 et al., 2008; Dunlea et al., 2009; Shinozuka et al., 2009; Liu et al., 2017) indicates that the CU-426 AMS was not substantially impacted by the aerosol transmission effects discussed above. Also, the strong correlation ($R^2 = 0.87$) between the two instruments, which both have comparable, very 427 428 high time resolution, indicate that the CU-AMS did not experience any plume recovery artifacts 429 that were observed with the MC/IC or artifacts in measuring highly concentrated plumes.

Finally, we compare the PM₁ volume concentrations estimated from the LAS PM₁ versus the CU-AMS plus SP2. For this comparison, we use the calibrated AMS transmission curve during this campaign (Figure S30), which is consistent with those from recent studies (Knote et al., 2011; Hu et al., 2017b), to correct for particle transmission differences between the instruments. The LAS diameters were corrected by a factor of 1.115 from the PSL-calibrated values, to account for the lower refractive index of ambient particles, similar to Liu et al. (2017). To estimate the volume concentration from the combined AMS and BC measurements, we assume additive species

volumes (DeCarlo et al., 2004). Species densities of 1.78 g cm⁻³ for NH₄, pNO₃, and SO₄ (Lide, 437 1991; Salcedo et al., 2006), 1.52 g cm⁻³ for Chl (Lide, 1991; Salcedo et al., 2006), 1.77 g cm⁻³ for 438 439 BC (Park et al., 2004), and the OA density is estimated from the CU-AMS O/C and H/C ratios of 440 OA using the parameterization of Kuwata et al. (2012). The comparison between total PM₁ volume 441 estimated from the CU-AMS plus BC vs. versus LAS shows a correlation (R²) of 0.86. However, 442 the volume from AMS plus SP2 is higher (slope of 1.56) when comparing all of KORUS-AQ. We 443 hypothesize that this may be due to saturation of the LAS detector at high particle concentrations that were frequently observed in this campaign (greater than 1800 particles cm⁻³ or total CU-AMS 444 plus SP2 mass greater than 40 µg sm⁻³), as has been observed in prior comparisons (Liu et al., 445 446 2017), or a change in the refractive index when OA becomes dominant at these high concentrations 447 (Moise et al., 2015). Different filters are tested and shown in Figure S30 and Figure S31, using 448 both values reported in literature and values that represent a stable ratio between LAS and calculated CU-AMS plus SP2 volume. If we filter for data when there is less than 20 μ g sm⁻³, the 449 slope drops to 1.00, showing agreement between within the combined uncertainties ($R^2 = 0.79$), 450 451 and providing strong evidence that LAS saturation at higher concentrations is the main reason for 452 the apparent disagreement when analyzing the entire campaign.

We further investigate (Figure S31) whether the slope could be due to LAS saturation or a bias in RIE_{OA}, or in CE, vs. the values used in our analyses (Jimenez et al., 2016; Xu et al., 2018). There is a slight increase in the ratio of AMS plus SP2 to LAS volumes versus OA/total CU-AMS mass at high fractions of OA, although still within the combined measurement uncertainties. With filtered data (less than 1600 particles cm⁻³ or total CU-AMS mass less than 20 μ g sm⁻³), the volume ratios remain nearly flat, even at high *f*(OA). This confirms that LAS saturation is the most likely cause for the differences. Finally, a recent study (Xu et al., 2018) has reported new laboratory 460 measurements of $RIE_{OA} = 1.6\pm0.5$, although these authors indicated that it was unclear whether 461 this value was applicable to ambient particles, and the value of $RIE_{OA} = 1.4$ used in this study is 462 well within their reported uncertainty. When using $RIE_{OA} = 1.6$ in our analysis (not shown) the 463 slope for the entire dataset decrease by only 6% (1.56 to 1.47), indicating that RIE uncertainties 464 cannot explain the bulk of the observed difference.

465



467 Figure S23. (top) Time series of mist-chamber (dark red line) and CU-AMS (red line) SO₄ for one
468 flight (RF17). (bottom) Scatter plot of CU-AMS SO₄ versus mist-chamber ion-chromatograph
469 (MC/IC) SO₄ for entire KORUS-AQ campaign.



470

471 **Figure S24.** (top) Time series of filter (black squares) and CU-AMS (red line) SO₄ for one flight 472 (RF17). The CU-AMS data has been averaged to the filter sampling time. (bottom) Scatter plot of 473 CU-AMS SO₄ versus filter SO₄ for entire KORUS-AQ campaign. The points are colored by the 474 total sodium (Na⁺) and calcium (Ca²⁺) measured by the filters, as indicators of sea salt and dust, 475 respectively.



Figure S25. Scatter plot of not transmission corrected K-AMS versus CU-AMS mass
concentrations for all of KORUS-AQ for (a) Chl, (b) SO₄, (c) pNO₃, (d) NH₄, (e) OA, and (f) total
AMS mass. The slopes and R² for all comparisons are shown in each scatter plot.



483

Figure S26. (left axis) Transmission curve for CU-AMS (black circle and dark grey square) and K-AMS (purple diamond). The curve from literature (Knote et al., 2011; Hu et al., 2017b), which describes the CU-AMS, is shown (grey dotted line). The fit for K-AMS transmission is shown with the purple dotted line. (right axis) Average mass distributions for OA (green), pNO₃ (blue), SO₄ (red), and NH₄ (orange) measured by CU-AMS in the boundary layer during KORUS-AQ. Note that some of the apparent signal at larger particle sizes is caused by the limited time response of the AMS detection system.



492

493 **Figure S27.** Scatter plot of K-AMS versus CU-AMS total mass concentrations (a) RFs 1 - 9, 11, 494 15, and 19 and (b) RFs 10, 12 - 14, 16 - 18, and 20. These are flights where the average sizes were 495 found below (a) and above (b) the K-AMS size cut-off (Figure S26). The slopes and R² for all 496 comparisons are shown in each scatter plot.



Figure S28. Binned total PM₁ AMS mass ratios, normalized by the average ratio, versus the calculated CE used for the CU-AMS measurements. The error bars are the standard error about the mean, and the shaded grey area is the combined uncertainty of the two AMS measurements $(\pm 27\%)$. The data is only for flights where the PM₁ sizes were typically below the K-AMS size cut-off (RFs 1 – 9, 11, 15, and 19).



Figure S29. (top) Time series of total submicron mass (black, left axis) and 532 nm extinction (green, right axis) for one flight (RF19). (bottom) Scatter plot of 532 nm extinction versus total

507 submicron mass (black carbon + CU-AMS species) for the entire KORUS-AQ campaign.

508





Figure S30. (top) Time series of total submicron volume from LAS (grey dashed line) and CU-510 511 AMS plus black carbon (black) for one flight (RF11). (bottom) Scatter plot of total submicron 512 volume (black carbon + CU-AMS species) versus LAS volume for entire KORUS-AQ campaign. 513 The data is colored by total CU-AMS mass. Pink line is a fit to all data, the purple line is a fit to 514 data where the particle number concentration is less than 1800 particles scm⁻³. The red line is a fit to the data where the CU-AMS plus SP2 total mass is less than 40 μ g sm⁻³. The black line is a fit 515 to the data where the particle number concentration is less than 1600 particles scm⁻³ and Ca²⁺ 516 concentration is less than 0.35 μ g sm⁻³. Finally, the grey line is a fit to the data where the CU-517 518 AMS plus BC total mass is less than 20 μ g sm⁻³.



520

Figure S31. (a) Binned volume ratio (CU-AMS plus black carbon volume/LAS Volume) versus fraction of organic aerosol (OA) to total CU-AMS mass. (b) Binned volume ratio versus LAS particle number concentration. (c) Binned volume ratio versus CU-AMS total mass. In all figures, the black data is for all data whereas the blue data is for the volume ratio where the particle number concentration is less than 1600 particles cm⁻³ and the orange data is for the volume ratio where the CU-AMS total mass concentration is less than 20 μ g sm⁻³. Also, the shaded area represents the combined uncertainty in both measurements (Bahreini et al., 2009).



S9. Fractional PM1 Contribution to Vertical Profile

Figure S32. Fractional contribution of PM₁ contribution vertical profile for all of KORUS-AQ.

534 S10. Observed Aerosol Production over Seoul, South Korea



Figure S33. Scatter plot of OA versus CO, observed over Seoul, during KORUS-AQ. The points
 are colored by the NO_x photochemical clock. The fit is for the decile binned data.



Figure S34. Comparison of $\Delta OA/\Delta CO$ observed over Seoul with different CO backgrounds.

Location	Slope (ppmv CO/ppmv CO ₂)	Study
Mexico City	0.045	Vay et al. (2009)
Los Angeles	0.009	Peischl et al., (2013)
Beijing	0.02	Wang et al. (2010)
		Silva et al. (2013)
		Tohjima et al. (2014)
Outflow China	0.02	Wang et al. (2010)
		Silva et al. (2013)
		Tohjima et al. (2014)
Seoul	0.01	Silva et al. (2013)
		Tang et al. (2018)

Table S4. Compilation of slopes used to convert from $\Delta OA/\Delta CO$ to $\Delta OA/\Delta CO_2$ used in this study.



Figure S35. Same as **Figure 4**(a), but comparing results using three different photochemical 545 clocks (Figure S21).



550 **Figure S36.** Same as **Figure 6**b, but speciated for MO-OOA, LO-OOA, and HOA. (a) is over the 551 Yellow Sea (RF12) and (b) is over Seoul.

First, we briefly discuss how the AMS OA source tracers typically used to investigate OA chemistry evolved over Seoul (Figure S37). During KORUS-AQ, there was no appreciable influence from isoprene production of IEPOX-SOA (Hu et al., 2015), as the ion indicative of isoprene IEPOX-SOA ($C_5H_6O^+$) remained at background values typical of air without isoprene SOA influence.

Similarly, biomass burning OA (BBOA) appeared to be present but dilute in its contribution to OA. Most of the OA had $f_{C_2H_4O_2}$, an ion indicative of biomass burning and levoglucosan (Schneider et al., 2006; Aiken et al., 2010), below 0.1 over Seoul, and the PMF factors fall near the limit of detection for BBOA (Cubison et al., 2011) and lower than the values that typically indicate ambient and laboratory BBOA emissions at various stages of chemical evolution (Cubison et al., 2011; Ortega et al., 2013). We speculate that the limited BBOA is highly 563 mixed into the OA from the numerous, small agricultural fires that were observed during the 564 campaign and have been observed during this time period, during other years, in South Korea 565 (Kang et al., 2006). However, the amount of fresher BBOA was not high enough, nor as strong of 566 a feature as observed in prior studies (Aiken et al., 2010; DeCarlo et al., 2010; Cubison et al., 2011; 567 Hu et al., 2016), to reliably resolve a separate BBOA PMF factor. As shown in Figure S11, typical 568 gas-phase biomass burning tracers (CO, NO_x, acetonitrile, HCN, and black carbon) do not show a 569 consistent strong correlation with any of the PMF factors, further suggesting that BBOA is not a 570 major contributor, and any BBOA present is highly mixed with HOA and the oxidized OA. 571 Consistent with our results, Kim et al. (2017) did not resolve a BBOA factor from a ground site in 572 Seoul during the KORUS campaign.

573 Similar to other studies over urban areas or for chamber studies oxidizing urban VOCs 574 (e.g., benzene, xylenes, etc.) (Ng et al., 2010; Freney et al., 2014; Ortega et al., 2016), marked 575 chemical evolution was observed as tracked by the $C_2H_3O^+$ and CO_2^+ ions. The evolution of these 576 two ions, as a fraction of total OA, fall in the same space as has been observed in these prior 577 studies, indicating consistent photochemical evolution of SOA over urban locations.

578 Finally, unlike Kim et al. (2017), we did not observe clear indication for cooking organic 579 aerosol (COA) in our PMF results. The COA was at a minimum (less than 1 μ g m⁻³) at the surface 580 in Seoul during the times the DC-8 overpassed (Kim et al., 2018); thus, we speculate the amount 581 of COA sampled was a small fraction of OA and was mostly lumped into the HOA factor. This 582 does not affect our characterization of HOA as POA, since COA is also a primary aerosol emission. 583



Figure S37. Plots of (a) f_{CO_2} versus $f_{C_2H_3O}$, (b) f_{CO_2} versus $f_{C_2H_4O_2}$, and (c) f_{CO_2} versus $f_{C_5H_6O}$. 585 Points highlighted in color refer to observations over Seoul, South Korea, during KORUS-AQ. In 586 (a), the triangle is from Ng et al. (2010); in (b), the triangle is from Cubison et al. (2011), and the 587 vertical line is the typical "background" values for $f_{C_2H_4O_2}$ from Cubison et al. (2011); and, in (c), 588 589 the vertical line is the typical "background" values for $f_{C_5H_6O}$ from Hu et al. (2015). The PMF 590 results for each triangle plot are shown in squares, where grey is HOA, light green is LO-OOA, 591 and dark green is MO-OOA. The light orange dots in (b) are the observations from ARCTAS forest fires (Cubison et al., 2011), as an example for data strongly impacted by biomass burning. The 592 593 quantile average values (averaged the x variables according to quantiles of the y variables) for 594 each comparison are shown in light red circles.

595



599 Figure S38. Time series of the fractional contribution of organic nitrates (pRONO₂) to the total

600 pNO₃ signal during KORUS-AQ. (b) Fractional contribution of organic nitrates versus pNO₃

601 during KORUS-AQ.

603 **References**

- 604
- Aiken, A. C., de Foy, B., Wiedinmyer, C., DeCarlo, P. F., Ulbrich, I. M., Wehrli, M. N., Szidat,
- 606 S., Prévôt, A. S. H., Noda, J., Wacker, L., Volkamer, R., Fortner, E., Wang, J., Laskin, A.,
- 607 Shutthanandan, V., Zheng, J., Zhang, R., Paredes-Miranda, G., Arnott, W. P., Molina, L. T., Sosa,
- 608 G., Querol, X. and Jimenez, J. L.: Mexico City aerosol analysis during MILAGRO using high
- resolution aerosol mass spectrometry at the urban supersite (T0)-Part 2: Analysis of the biomass
- burning contribution and the non-fossil carbon fraction, Atmos. Chem. Phys., 10(12), 5315–5341,
- 611 doi:10.5194/acp-10-5315-2010, 2010.
- 612 Ait-Helal, W., Borbon, A., Sauvage, S., De Gouw, J. A., Colomb, A., Gros, V., Freutel, F., Crippa,
- 613 M., Afif, C., Baltensperger, U., Beekmann, M., Doussin, J.-F. F., Durand-Jolibois, R., Fronval, I.,
- Grand, N., Leonardis, T., Lopez, M., Michoud, V., Miet, K., Perrier, S., Prévôt, A. S. H.,
- 615 Schneider, J., Siour, G., Zapf, P. and Locoge, N.: Volatile and intermediate volatility organic 616 compounds in suburban Paris: Variability, origin and importance for SOA formation, Atmos.
- 617 Chem. Phys., 14(19), 10439–10464, doi:10.5194/acp-14-10439-2014, 2014.
- Aknan, A. and Chen, G.: KORUS-AQ DC-8 Aircraft Dataset, [online] Available from:
 https://www-air.larc.nasa.gov/cgi-bin/ArcView/korusaq (Accessed 6 December 2018), 2018.
- Atkinson, R.: Kinetics of the gas-phase reactions of OH radicals with alkanes and cycloalkanes,
 Atmos. Chem. Phys., 3(6), 2233–2307, doi:10.5194/acp-3-2233-2003, 2003.
- Atkinson, R. and Arey, J.: Atmospheric Degradation of Volatile Organic Compounds, Chem. Rev.,
 103, 4605–4638, doi:10.1021/CR0206420, 2003.
- 624 Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M.
- 625 E., Rossi, M. J. and Troe, J.: Evaluated kinetic and photochemical data for atmospheric chemistry:
- 626 Volume I gas phase reactions of Ox, HO_x, NO_x and SO_x species, Atmos. Chem. Phys., 4(6), 627 1461 1728 doi:10.5104/com 4.1461.2004.2004
- 627 1461–1738, doi:10.5194/acp-4-1461-2004, 2004.
- 628 Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M.
- 629 E., Rossi, M. J., Troe, J. and IUPAC Subcommittee: Evaluated kinetic and photochemical data for
- 630 atmospheric chemistry: Volume II gas phase reactions of organic species, Atmos. Chem. Phys.,
- 631 6(11), 3625–4055, doi:10.5194/acp-6-3625-2006, 2006.
- Bahreini, R., Dunlea, E. J., Matthew, B. M., Simons, C., Docherty, K. S., DeCarlo, P. F., Jimenez,
- 633 J. L., Brock, C. A. and Middlebrook, A. M.: Design and Operation of a Pressure-Controlled Inlet
- 634 for Airborne Sampling with an Aerodynamic Aerosol Lens, Aerosol Sci. Technol., 42(6), 465–
- 635 471, doi:10.1080/02786820802178514, 2008.
- 636 Bahreini, R., Ervens, B., Middlebrook, A. M., Warneke, C., de Gouw, J. A., DeCarlo, P. F.,
- 637 Jimenez, J. L., Brock, C. A., Neuman, J. A., Ryerson, T. B., Stark, H., Atlas, E., Brioude, J., Fried,
- 638 A., Holloway, J. S., Peischl, J., Richter, D., Walega, J., Weibring, P., Wollny, A. G. and
- 639 Fehsenfeid, F. C.: Organic aerosol formation in urban and industrial plumes near Houston and
- 640 Dallas, Texas, J. Geophys. Res., 114(16), D00F16, doi:10.1029/2008JD011493, 2009.
- 641 Bohn, B. and Zetzsch, C.: Kinetics and mechanism of the reaction of OH with the
- 642 trimethylbenzenes experimental evidence for the formation of adduct isomers, Phys. Chem.
- 643 Chem. Phys., 14(40), 13933, doi:10.1039/c2cp42434g, 2012.
- Brioude, J., Arnold, D., Stohl, A., Cassiani, M., Morton, D., Seibert, P., Angevine, W., Evan, S.,

- 645 Dingwell, A., Fast, J. D., Easter, R. C., Pisso, I., Burkhart, J. and Wotawa, G.: The Lagrangian
- particle dispersion model FLEXPART-WRF version 3.1, Geosci. Model Dev., 6(6), 1889–1904,
 doi:10.5194/gmd-6-1889-2013, 2013.
- 648 Crounse, J. D., Nielsen, L. B., Jørgensen, S., Kjaergaard, H. G. and Wennberg, P. O.: Autoxidation
- 649 of Organic Compounds in the Atmosphere, J. Phys. Chem. Lett., 4(20), 3513–3520, 650 doi:10.1021/jz4019207, 2013.
- 651 Cubison, M. J., Ortega, A. M., Hayes, P. L., Farmer, D. K., Day, D. A., Lechner, M. J., Brune, W.
- H., Apel, E., Diskin, G. S., Fisher, J. A., Fuelberg, H. E., Hecobian, A., Knapp, D. J., Mikoviny,
- T., Riemer, D., Sachse, G. W., Sessions, W., Weber, R. J., Weinheimer, A. J., Wisthaler, A. and Jimenez, J. L.: Effects of aging on organic aerosol from open biomass burning smoke in aircraft
- and laboratory studies, Atmos. Chem. Phys., 11(23), 12049–12064, doi:10.5194/acp-11-12049-
- 656 2011, 2011.
- de Gouw, J. A., Gilman, J. B., Kim, S.-W., Lerner, B. M., Isaacman-VanWertz, G., McDonald, B.
- 658 C., Warneke, C., Kuster, W. C., Lefer, B. L., Griffith, S. M., Dusanter, S., Stevens, P. S. and Stutz,
- 659 J.: Chemistry of Volatile Organic Compounds in the Los Angeles basin: Nighttime Removal of
- Alkenes and Determination of Emission Ratios, J. Geophys. Res., 122(21), 11,843-11,861,
- 661 doi:10.1002/2017JD027459, 2017.
- DeCarlo, P. F., Slowik, J. G., Worsnop, D. R., Davidovits, P., Jimenez, J. L., Stainken, K.,
 Williams, L., Jayne, J., Kolb, C. and Rudich, Y.: Particle Morphology and Density
 Characterization by Combined Mobility and Aerodynamic Diameter Measurements. Part 1:
 Theory, Aerosol Sci. Technol., 38(12), 1185–1205, doi:10.1080/027868290903907, 2004.
- 666 DeCarlo, P. F., Dunlea, E. J., Kimmel, J. R., Aiken, A. C., Sueper, D., Crounse, J., Wennberg, P.
- 667 O., Emmons, L., Shinozuka, Y., Clarke, A., Zhou, J., Tomlinson, J., Collins, D. R., Knapp, D.,
- 668 Weinheimer, A. J., Montzka, D. D., Campos, T. and Jimenez, J. L.: Fast airborne aerosol size and
- 669 chemistry measurements above Mexico City and Central Mexico during the MILAGRO campaign,
- 670 Atmos. Chem. Phys., 8(14), 4027–4048, doi:10.5194/acp-8-4027-2008, 2008.
- 671 DeCarlo, P. F., Ulbrich, I. M., Crounse, J., de Foy, B., Dunlea, E. J., Aiken, A. C., Knapp, D.,
- 672 Weinheimer, A. J., Campos, T., Wennberg, P. O. and Jimenez, J. L.: Investigation of the sources
- and processing of organic aerosol over the Central Mexican Plateau from aircraft measurements
- 674 during MILAGRO, Atmos. Chem. Phys., 10(12), 5257–5280, doi:10.5194/acp-10-5257-2010,
- 675 2010.
- 676 Docherty, K. S., Aiken, A. C., Huffman, J. A., Ulbrich, I. M., DeCarlo, P. F., Sueper, D., Worsnop,
- D. R., Snyder, D. C., Peltier, R. E., Weber, R. J., Grover, B. D., Eatough, D. J., Williams, B. J.,
- 678 Goldstein, A. H., Ziemann, P. J. and Jimenez, J. L.: The 2005 Study of Organic Aerosols at
- 679 Riverside (SOAR-1): instrumental intercomparisons and fine particle composition, Atmos. Chem.
- 680 Phys., 11(23), 12387–12420, doi:DOI 10.5194/acp-11-12387-2011, 2011.
- 681 Dunlea, E. J., DeCarlo, P. F., Aiken, A. C., Kimmel, J. R., Peltier, R. E., Weber, R. J., Tomlison,
- 582 J., Collins, D. R., Shinozuka, Y., McNaugthon, C. S., Howell, S. G., Clarke, A. D., Emmons, L.
- 683 K., Apel, E. C., Pfister, G. G., van Donkelaar, A., Martin, R. V., Millett, D. B., Heald, C. L. and
- 584 Jimenez, J. L.: Evolution of Asian aerosols during transpacific transport in INTEX-B, Atmos.
- 685 Chem. Phys., 9(19), 7257–7287, doi:10.5194/acp-9-7257-2009, 2009.
- Dzepina, K., Volkamer, R. M. R., Madronich, S., Tulet, P., Ulbrich, I. M., Zhang, Q., Cappa, C.
 D., Ziemann, P. J. and Jimenez, J. L.: Evaluation of recently-proposed secondary organic aerosol

- models for a case study in Mexico City, Atmos. Chem. Phys., 9(15), 5681–5709, doi:10.5194/acp9-5681-2009, 2009.
- 690 Freney, E. J., Sellegri, K., Canonaco, F., Colomb, A., Borbon, A., Michoud, V., Doussin, J.-F.,
- 691 Crumeyrolle, S., Amarouche, N., Pichon, J.-M., Bourianne, T., Gomes, L., Prevot, A. S. H.,
- 692 Beekmann, M. and Schwarzenböeck, A.: Characterizing the impact of urban emissions on regional
- 693 aerosol particles: airborne measurements during the MEGAPOLI experiment, Atmos. Chem.
- 694 Phys., 14(3), 1397–1412, doi:10.5194/acp-14-1397-2014, 2014.
- Fry, J. L., Draper, D. C., Zarzana, K. J., Campuzano-Jost, P., Day, D. A., Jimenez, J. L., Brown,
- 696 S. S., Cohen, R. C., Kaser, L., Hansel, A., Cappellin, L., Karl, T., Hodzic Roux, A., Turnipseed, 697 A., Cantrell, C., Lefer, B. L. and Grossberg, N.: Observations of gas- and aerosol-phase organic
- 698 nitrates at BEACHON-RoMBAS 2011, Atmos. Chem. Phys., 13(17), 8585–8605, 699 doi:10.5194/acp-13-8585-2013, 2013.
- 700 Hand, J. L. and Malm, W. C.: Review of aerosol mass scattering efficiencies from ground-based
- 701 measurements since 1990, J. Geophys. Res., 112(D16), D16203, doi:10.1029/2007JD008484,
- 702 2007.
- 703 Hayes, P. L., Ortega, A. M., Cubison, M. J., Froyd, K. D., Zhao, Y., Cliff, S. S., Hu, W. W.,
- Toohey, D. W., Flynn, J. H., Lefer, B. L., Grossberg, N., Alvarez, S., Rappenglück, B., Taylor, J.
- 705 W., Allan, J. D., Holloway, J. S., Gilman, J. B., Kuster, W. C., de Gouw, J. A., Massoli, P., Zhang,
- 706 X., Liu, J., Weber, R. J., Corrigan, A. L., Russell, L. M., Isaacman, G., Worton, D. R., Kreisberg,
- N. M., Goldstein, A. H., Thalman, R., Waxman, E. M., Volkamer, R., Lin, Y. H., Surratt, J. D.,
 Kleindienst, T. E., Offenberg, J. H., Dusanter, S., Griffith, S., Stevens, P. S., Brioude, J., Angevine,
- Kleindienst, T. E., Offenberg, J. H., Dusanter, S., Griffith, S., Stevens, P. S., Brioude, J., Angevine,
 W. M. and Jimenez, J. L.: Organic aerosol composition and sources in Pasadena, California, during
- 710 the 2010 CalNex campaign, J. Geophys. Res., 118(16), 9233–9257, doi:10.1002/jgrd.50530, 2013.
- 711 Heim, E., Dibb, J., Scheuer, E., Campuzano-Jost, P., Nault, B. A., Jimenez, J. L., Peterson, D.,
- 712 Knote, C., Fenn, M., Hair, J., Beyersdorf, A. J. and Anderson, B. E.: Asian Dust Observed during
- 713 KORUS-AQ Facilitates the Uptake and Incorporation of Soluble Pollutants during Transport to
- 714 South Korea: The Hwangsa Anthropogenic Model, J. Geophys. Res. Atmos., submitted, 2018.
- Heo, J.-B., Hopke, P. K. and Yi, S.-M.: Source apportionment of PM2.5 in Seoul, Korea, Atmos.
 Chem. Phys., 9(14), 4957–4971, doi:10.5194/acp-9-4957-2009, 2009.
- 717 Hu, W., Hu, M., Hu, W., Jimenez, J. L., Yuan, B., Chen, W., Wang, M., Wu, Y., Chen, C., Wang,
- 718 Z., Peng, J., Zeng, L. and Shao, M.: Chemical composition, sources and aging process of sub-
- 719 micron aerosols in Beijing: contrast between summer and winter, J. Geophys. Res. Atmos.,
- 720 doi:10.1002/2015JD024020, 2016.
- Hu, W., Campuzano-Jost, P., Day, D. A., Croteau, P., Canagaratna, M. R., Jayne, J. T., Worsnop,
- 722 D. R. and Jimenez, J. L.: Evaluation of the new capture vaporizer for aerosol mass spectrometers
- 723 (AMS) through field studies of inorganic species, Aerosol Sci. Technol., 51(6), 735-754,
- 724 doi:10.1080/02786826.2017.1296104, 2017a.
- Hu, W., Campuzano-Jost, P., Day, D. A., Croteau, P., Canagaratna, M. R., Jayne, J. T., Worsnop,
- 726 D. R. and Jimenez, J. L.: Evaluation of the new capture vapourizer for aerosol mass spectrometers
- 727 (AMS) through laboratory studies of inorganic species, Atmos. Meas. Tech., 10(6), 2897–2921,
- 728 doi:10.5194/amt-10-2897-2017, 2017b.
- Hu, W., Day, D. A., Campuzano-Jost, P., Nault, B. A., Park, T., Lee, T., Croteau, P., Canagaratna,

- M. R., Jayne, J. T., Worsnop, D. R. and Jimenez, J. L.: Evaluation of the new capture vaporizer for Aerosol Mass Spectrometers: Characterization of organic aerosol mass spectra, Aerosol Sci.
- 732 Technol., 52(7), 752–739, doi:10.1080/02786826.2018.1454584, 2018a.
- 733 Hu, W., Day, D. A., Campuzano-Jost, P., Nault, B. A., Park, T., Lee, T., Croteau, P., Canagaratna, 734 M. R., Jayne, J. T., Worsnop, D. R. and Jimenez, J. L.: Evaluation of the New Capture Vaporizer 735 for Aerosol Mass Spectrometers (AMS): Elemental Composition and Source Apportionment of 736 Aerosols (OA), ACS Earth Sp. Chem., 410-421, Organic 2(4),doi:10.1021/acsearthspacechem.8b00002, 2018b. 737
- 738 Hu, W. W., Campuzano-Jost, P., Palm, B. B., Day, D. A., Ortega, A. M., Hayes, P. L., Krechmer, 739 J. E., Chen, O., Kuwata, M., Liu, Y. J., de Sá, S. S., McKinney, K., Martin, S. T., Hu, M., 740 Budisulistiorini, S. H., Riva, M., Surratt, J. D., St. Clair, J. M., Isaacman-Van Wertz, G., Yee, L. 741 D., Goldstein, A. H., Carbone, S., Brito, J., Artaxo, P., de Gouw, J. A., Koss, A., Wisthaler, A., 742 Mikoviny, T., Karl, T., Kaser, L., Jud, W., Hansel, A., Docherty, K. S., Alexander, M. L., 743 Robinson, N. H., Coe, H., Allan, J. D., Canagaratna, M. R., Paulot, F. and Jimenez, J. L.: 744 Characterization of a real-time tracer for Isoprene Epoxydiols-derived Secondary Organic Aerosol 745 (IEPOX-SOA) from aerosol mass spectrometer measurements, Atmos. Chem. Phys., 15(8),
- 746 11807–11833, doi:10.5194/acp-15-11807-2015, 2015.
- 747 Huffman, J. A., Docherty, K. S., Aiken, A. C., Cubison, M. J., Ulbrich, I. M., DeCarlo, P. F.,
- Sueper, D., Jayne, J. T., Worsnop, D. R., Ziemann, P. J. and Jimenez, J. L.: Chemically-resolved
 aerosol volatility measurements from two megacity field studies, Atmos. Chem. Phys., 9(1), 7161–
- 750 7182, doi:doi:10.5194/acp-9-7161-2009, 2009.
- 751 Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., 752 DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., 753 Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, 754 C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., 755 Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, E. J., Huffman, J. A., 756 Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., 757 758 Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., 759 Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., 760 Middlebrook, A. M., Kolb, C. E., Baltensperger, U., Worsnop, D. R. and Worsnop, D. R.: 761 Evolution of organic aerosols in the atmosphere., Science, 326(5959), 1525–1529, 762 doi:10.1126/science.1180353, 2009.
- Jimenez, J. L., Canagaratna, M. R., Drewnick, F., Allan, J. D., Alfarra, M. R., Middlebrook, A.
 M., Slowik, J. G., Zhang, Q., Coe, H., Jayne, J. T. and Worsnop, D. R.: Comment on "The effects of molecular weight and thermal decomposition on the sensitivity of a thermal desorption aerosol mass spectrometer," Aerosol Sci. Technol., 50(9), i–xv, doi:10.1080/02786826.2016.1205728, 2016.
- Kang, C. M., Kang, B. W. and Lee, H. S.: Source identification and trends in concentrations ofgaseous and fine particulate principal species in Seoul, South Korea, J. Air Waste Manag. Assoc.,
- 770 56(7), 911–921, doi:10.1080/10473289.2006.10464506, 2006.
- Kim, B. M., Seo, J., Kim, J. Y., Lee, J. Y. and Kim, Y.: Transported vs. local contributions from
 secondary and biomass burning sources to PM2.5, Atmos. Environ., 144, 24–36,

- 773 doi:10.1016/j.atmosenv.2016.08.072, 2016.
- Kim, H., Zhang, Q. and Heo, J.: Influence of Intense secondary aerosol formation and long-range
- transport on aerosol chemistry and properties in the Seoul Metropolitan Area during spring time :
- Results from KORUS-AQ, Atmos. Chem. Phys., 18, 7149–7168, doi:10.51944/acp-2017-947,
 2018.
- 778 Kleinman, L. I., Springston, S. R., Daum, P. H., Lee, Y.-N., Nunnermacker, L. J., Senum, G. I.,
- 779 Wang, J., Weinstein-Lloyd, J., Alexander, M. L., Hubbe, J., Ortega, J., Canagaratna, M. R. and
- Jayne, J.: The time evolution of aerosol composition over the Mexico City plateau, Atmos. Chem.
- 781 Phys., 8(6), 1559–1575, doi:10.5194/acp-8-1559-2008, 2008.
- 782 Knote, C., Brunner, D., Vogel, H., Allan, J., Asmi, A., Äijälä, M., Carbone, S., van der Gon, H.
- 783 D., Jimenez, J. L., Kiendler-Scharr, A., Mohr, C., Poulain, L., Prévôt, A. S. H., Swietlicki, E. and
- 784 Vogel, B.: Towards an online-coupled chemistry-climate model: evaluation of trace gases and
- aerosols in COSMO-ART, Geosci. Model Dev., 4(4), 1077–1102, doi:10.5194/gmd-4-1077-2011,
 2011.
- Krechmer, J. E., Pagonis, D., Ziemann, P. J. and Jimenez, J. L.: Quantification of Gas-Wall
 Partitioning in Teflon Environmental Chambers Using Rapid Bursts of Low-Volatility Oxidized
 Species Generated in Situ, Environ. Sci. Technol., 50(11), 5757–5765,
- 790 doi:10.1021/acs.est.6b00606, 2016.
- 791 Kuwata, M., Zorn, S. R. and Martin, S. T.: Using Elemental Ratios to Predict the Density of
- Organic Material Composed of Carbon, Hydrogen, and Oxygen, Environ. Sci. Technol., 46(2),
 787–794, doi:10.1021/es202525q, 2012.
- Lide, D. R.: CRC Handbook of Chemistry and Physics, CRC Press Inc., USA., 1991.
- Liu, P. S. K., Deng, R., Smith, K. A., Williams, L. R., Jayne, J. T., Canagaratna, M. R., Moore,
- K., Onasch, T. B., Worsnop, D. R. and Deshler, T.: Transmission Efficiency of an Aerodynamic
 Focusing Lens System: Comparison of Model Calculations and Laboratory Measurements for the
 Aerodyne Aerosol Mass Spectrometer, Aerosol Sci. Technol., 41(8), 721–733,
- 799 doi:10.1080/02786820701422278, 2007.
- 800 Liu, X., Huey, L. G., Yokelson, R. J., Selimovic, V., Simpson, I. J., Müller, M., Jimenez, J. L.,
- 801 Campuzano-Jost, P., Beyersdorf, A. J., Blake, D. R., Butterfield, Z., Choi, Y., Crounse, J. D., Day,
- 802 D. A., Diskin, G. S., Dubey, M. K., Fortner, E., Hanisco, T. F., Hu, W., King, L. E., Kleinman, L.,
- 803 Meinardi, S., Mikoviny, T., Onasch, T. B., Palm, B. B., Peischl, J., Pollack, I. B., Ryerson, T. B.,
- 804 Sachse, G. W., Sedlacek, A. J., Shilling, J. E., Springston, S., St. Clair, J. M., Tanner, D. J., Teng,
- A. P., Wennberg, P. O., Wisthaler, A. and Wolfe, G. M.: Airborne measurements of western U.S.
- 806 wildfire emissions: Comparison with prescribed burning and air quality implications, J. Geophys.
- 807 Res., 122(11), 6108–6129, doi:10.1002/2016JD026315, 2017.
- 808 Ma, P. K., Zhao, Y., Robinson, A. L., Worton, D. R., Goldstein, A. H., Ortega, A. M., Jimenez, J.-
- 809 L., Zotter, P., Prévôt, A. S. H., Szidat, S. and Hayes, P. L.: Evaluating the impact of new
- 810 observational constraints on P-S/IVOC emissions, multi-generation oxidation, and chamber wall
- 811 losses on SOA modeling for Los Angeles, CA, Atmos. Chem. Phys., 17(15), 9237-9259,
- 812 doi:10.5194/acp-17-9237-2017, 2017.
- 813 McNaughton, C. S., Clarke, A. D., Howell, S. G., Pinkerton, M., Anderson, B., Thornhill, L.,
- 814 Hudgins, C., Winstead, E., Dibb, J. E., Scheuer, E. and Maring, H.: Results from the DC-8 Inlet

- 815 Characterization Experiment (DICE): Airborne versus surface sampling of mineral dust and sea 816 salt aerosols, Aerosol Sci. Technol., 41(2), 136–159, doi:10.1080/02786820601118406, 2007.
- 817 Middlebrook, A. M., Bahreini, R., Jimenez, J. L. and Canagaratna, M. R.: Evaluation of
- 818 Composition-Dependent Collection Efficiencies for the Aerodyne Aerosol Mass Spectrometer
- 819 using Field Data, Aerosol Sci. Technol., 46(3), 258–271, doi:10.1080/02786826.2011.620041,
- 820 2012.
- 821 Moise, T., Flores, J. M. and Rudich, Y.: Optical Properties of Secondary Organic Aerosols and
- 822 Their Changes by Chemical Processes, Chem. Rev., 115(10), 4400–4439, doi:10.1021/cr5005259,
 823 2015.
- Mollner, A. K., Valluvadasan, S., Feng, L., Sprague, M. K., Okumura, M., Milligan, D. B., Bloss,
 W. J., Sander, S. P., Martien, P. T., Harley, R. a, McCoy, A. B. and Carter, W. P. L.: Rate of Gas
 Phase Association of Hydroxyl Radical and Nitrogen Dioxide, Science (80-.)., 330(6004), 646–
 9, doi:10.1126/science.1193030, 2010.
- Murphy, D. M.: The effects of molecular weight and thermal decomposition on the sensitivity of a thermal desorption aerosol mass spectrometer, Aerosol Sci. Technol., 50(2), 118–125, doi:10.1080/02786826.2015.1136403, 2016.
- 831 Nault, B. A., Campuzano-Jost, P., Schroder, J. C., Sueper, D. and Jimenez, J. L.: Using Event
- 832 Trigger Panel for IE/AB and Transmission Curve Calibrations, in 17th AMS Users' Meeting,
 833 Portland. [online] Available from: http://cires1.colorado.edu/jimenez834 group/wiki/index.php/AMSUsrMtgs#17th_AMS_Users_Meeting.2C_Portland.2C_Oregon,
- 835 2016.
- NCEP: National Centers for Environmental Prediction, [online] Available from:
 http://www.emc.ncep.noaa.gov/GFS/doc.php (Accessed 16 November 2017), n.d.
- Neuman, J. A., Parrish, D. D., Ryerson, T. B., Brock, C. A., Wiedinmyer, C., Frost, G. J.,
 Holloway, J. S. and Fehsenfeld, F. C.: Nitric acid loss rates measured in power plant plumes, J.
- 840 Geophys. Res., 109(23), 1–13, doi:10.1029/2004JD005092, 2004.
- 841 Ng, N. L., Canagaratna, M. R., Zhang, Q., Jimenez, J. L., Tian, J., Ulbrich, I. M., Kroll, J. H.,
- 842 Docherty, K. S., Chhabra, P. S., Bahreini, R., Murphy, S. M., Seinfeld, J. H., Hildebrandt, L.,
- 843 Donahue, N. M., Decarlo, P. F., Lanz, V. A., Prévôt, A. S. H., Dinar, E., Rudich, Y. and Worsnop,
- 844 D. R.: Organic aerosol components observed in Northern Hemispheric datasets from Aerosol Mass
- 845 Spectrometry, Atmos. Chem. Phys., 10(10), 4625–4641, doi:10.5194/acp-10-4625-2010, 2010.
- 846 Nguyen, T. B., Crounse, J. D., Teng, A. P., St. Clair, J. M., Paulot, F., Wolfe, G. M. and Wennberg,
- 847 P. O.: Rapid deposition of oxidized biogenic compounds to a temperate forest, Proc. Natl. Acad.
- 848 Sci., 112(5), E392–E401, doi:10.1073/pnas.1418702112, 2015.
- 849 Ortega, A. M., Day, D. A., Cubison, M. J., Brune, W. H., Bon, D., de Gouw, J. A. and Jimenez, J.
- 850 L.: Secondary organic aerosol formation and primary organic aerosol oxidation from biomass-
- burning smoke in a flow reactor during FLAME-3, Atmos. Chem. Phys., 13(22), 11551–11571,
- doi:10.5194/acp-13-11551-2013, 2013.
- 853 Ortega, A. M., Hayes, P. L., Peng, Z., Palm, B. B., Hu, W., Day, D. A., Li, R., Cubison, M. J.,
- Brune, W. H., Graus, M., Warneke, C., Gilman, J. B., Kuster, W. C., de Gouw, J. A., Gutiérrez-
- 855 Montes, C. and Jimenez, J. L.: Real-time measurements of secondary organic aerosol formation
- and aging from ambient air in an oxidation flow reactor in the Los Angeles area, Atmos. Chem.

- 857 Phys., 16(11), 7411–7433, doi:10.5194/acp-16-7411-2016, 2016.
- 858 Palm, B. B., Campuzano-Jost, P., Ortega, A. M., Day, D. A., Kaser, L., Jud, W., Karl, T., Hansel,
- 859 A., Hunter, J. F., Cross, E. S., Kroll, J. H., Peng, Z., Brune, W. H. and Jimenez, J. L.: In situ
- 860 secondary organic aerosol formation from ambient pine forest air using an oxidation flow reactor,
- 861 Atmos. Chem. Phys., 16(5), 2943–2970, doi:10.5194/acp-16-2943-2016, 2016.
- 862 Palm, B. B., Campuzano-Jost, P., Day, D. A., Ortega, A. M., Fry, J. L., Brown, S. S., Zarzana, K.
- 863 J., Dube, W., Wagner, N. L., Draper, D. C., Kaser, L., Jud, W., Karl, T., Hansel, A., Gutiérrez-
- 864 Montes, C. and Jimenez, J. L.: Secondary organic aerosol formation from in situ OH, O3, and NO3
- oxidation of ambient forest air in an oxidation flow reactor, Atmos. Chem. Phys., 17(8), 5331–
- 866 5354, doi:10.5194/acp-17-5331-2017, 2017.
- 867 Palm, B. B., de Sá, S. S., Day, D. A., Campuzano-Jost, P., Hu, W., Seco, R., Sjostedt, S. J., Park,
- 868 J.-H., Guenther, A. B., Kim, S., Brito, J., Wurm, F., Artaxo, P., Thalman, R., Wang, J., Yee, L. D.,
- 869 Wernis, R., Isaacman-VanWertz, G., Goldstein, A. H., Liu, Y., Springston, S. R., Souza, R.,
- 870 Newburn, M. K., Alexander, M. L., Martin, S. T. and Jimenez, J. L.: Secondary organic aerosol
- 871 formation from ambient air in an oxidation flow reactor in central Amazonia, Atmos. Chem. Phys.,
- 872 18(1), 467–493, doi:10.5194/acp-18-467-2018, 2018.
- Park, K., Kittelson, D. B., Zachariah, M. R. and McMurry, P. H.: Measurement of Inherent
 Material Density of Nanoparticle Agglomerates, J. Nanoparticle Res., 6(2/3), 267–272,
 doi:10.1023/B:NANO.0000034657.71309.e6, 2004.
- Parrish, D. D., Stohl, A., Forster, C., Atlas, E. L., Blake, D. R., Goldan, P. D., Kuster, W. C. and
 de Gouw, J. A.: Effects of mixing on evolution of hydrocarbon ratios in the troposphere, J.
- 878 Geophys. Res., 112(D10), D10S34, doi:10.1029/2006JD007583, 2007.
- Peischl, J., Ryerson, T. B., Brioude, J., Aikin, K. C., Andrews, A. E., Atlas, E., Blake, D., Daube,
 B. C., de Gouw, J. A., Dlugokencky, E., Frost, G. J., Gentner, D. R., Gilman, J. B., Goldstein, A.
- H., Harley, R. A., Holloway, J. S., Kofler, J., Kuster, W. C., Lang, P. M., Novelli, P. C., Santoni,
- 6. W., Trainer, M., Wofsy, S. C. and Parrish, D. D.: Quantifying sources of methane using light
- alkanes in the Los Angeles basin, California, J. Geophys. Res. Atmos., 118(10), 4974–4990,
 doi:10.1002/jgrd.50413, 2013.
- Peng, Z., Day, D. A., Stark, H., Li, R., Lee-Taylor, J., Palm, B. B., Brune, W. H. and Jimenez, J.
 L.: HO_x radical chemistry in oxidation flow reactors with low-pressure mercury lamps
 systematically examined by modeling, Atmos. Meas. Tech., 8(11), 4863–4890, doi:10.5194/amt8-4863-2015, 2015.
- 889 Pieber, S. M., El Haddad, I., Slowik, J. G., Canagaratna, M. R., Jayne, J. T., Platt, S. M., Bozzetti,
- C., Daellenbach, K. R., Fröhlich, R., Vlachou, A., Klein, F., Dommen, J., Miljevic, B., Jiménez,
 J. L., Worsnop, D. R., Baltensperger, U. and Prévôt, A. S. H.: Inorganic Salt Interference on CO2+
- in Aerodyne AMS and ACSM Organic Aerosol Composition Studies, Environ. Sci. Technol.,
- 893 50(19), 10494–10503, doi:10.1021/acs.est.6b01035, 2016.
 - Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop,
 A. P., Lane, T. E., Pierce, J. R. and Pandis, S. N.: Rethinking Organic Aerosols: Semivolatile
 Emissions and Photochemical Aging, Science (80-.)., 315(5816), 1259–1262,
 doi:10.1126/science.1133061, 2007.
 - 898 Romer, P. S., Duffey, K. C., Wooldridge, P. J., Allen, H. M., Ayres, B. R., Brown, S. S., Brune,

- 899 W. H., Crounse, J. D., de Gouw, J., Draper, D. C., Feiner, P. A., Fry, J. L., Goldstein, A. H., Koss,
- 900 A., Misztal, P. K., Nguyen, T. B., Olson, K., Teng, A. P., Wennberg, P. O., Wild, R. J., Zhang, L.
- and Cohen, R. C.: The lifetime of nitrogen oxides in an isoprene-dominated forest, Atmos. Chem.
- 902 Phys., 16(12), 7623–7637, doi:10.5194/acp-16-7623-2016, 2016.
- 903 Salcedo, D., Onasch, T. B., Dzepina, K., Canagaratna, M. R., Zhang, Q., Huffman, J. A., Decarlo,
- 904 P. F., Jayne, J. T., Mortimer, P., Worsnop, D. R., Kolb, C. E., Johnson, K. S., Zuberi, B., Marr, L.
- 905 C., Volkamer, R., Molina, L. T., Molina, M. J., Cardenas, B., Bernabe, R. M., Marquez, C.,
- Gaffney, J. S., Marley, N. A., Laskin, A., Shutthanandan, V., Xie, Y., Brune, W., Lesher, R.,
 Shirley, T. and Jimenez, J. L.: Characterization of ambient aerosols in Mexico City during the
- 907 Shirley, T. and Jimenez, J. L.: Characterization of ambient aerosols in Mexico City during the 908 MCMA-2003 campaign with Aerosol Mass Spectrometry: results from the CENICA Supersite,
- 909 Atmos. Chem. Phys., 6(4), 925–946, doi:10.5194/acp-6-925-2006, 2006.
- Sander, S. P., Abbatt, J. P. D., Barker, J. R., Burkholder, J. B., Friedl, R. R., Golden, D. M., Huie,
- R. E., Kolb, C. E., Kurylo, M. J., Moortgat, G. K., Orkin, V. L. and Wine, P. H.: Chemical Kinetics
 and Photochemical Data for Use in Atmospheric Studies, Evaluation No. 17, JPL Publ. 10-6, Jet
- 912 and Photochemical Data for Use in Atmospheric Studies, Evaluation No. .
 - 913 Propuls. Lab. Pasadena, 2011.
 - 914 Schneider, J., Hings, S. S., Nele Hock, B., Weimer, S., Borrmann, S., Fiebig, M., Petzold, A.,
 - Busen, R. and Kärcher, B.: Aircraft-based operation of an aerosol mass spectrometer:
 Measurements of tropospheric aerosol composition, J. Aerosol Sci., 37(7), 839–857,
 doi:10.1016/j.jaerosci.2005.07.002, 2006.
 - 918 Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Dunlea, E. J., Roberts, G. C.,
 - 919 Tomlinson, J. M., Collins, D. R., Howell, S. G., Kapustin, V. N., McNaughton, C. S. and Zhou, J.:
 - 920 Aerosol optical properties relevant to regional remote sensing of CCN activity and links to their
 - 921 organic mass fraction: airborne observations over Central Mexico and the US West Coast during
 - 922 MILAGRO/INTEX-B, Atmos. Chem. Phys., 9(18), 6727–6742, 2009.
 - Silva, S. J., Arellano, A. F. and Worden, H. M.: Toward anthropogenic combustion emission
 constraints from space-based analysis of urban CO₂/CO sensitivity, Geophys. Res. Lett., 40(18),
 4971–4976, doi:10.1002/grl.50954, 2013.
 - 926 Skamarock, C., Klemp, B., Dudhia, J., Gill, O., Barker, D., Duda, G., Huang, X., Wang, W. and
 - 927 Powers, G.: A Description of the Advanced Research WRF Version 3, , doi:10.5065/D68S4MVH,
 - 928 2008.
 - 929 Sprengnether, M. M., Demerjian, K. L., Dransfield, T. J., Clarke, J. S., Anderson, J. G. and
 - 930 Donahue, N. M.: Rate Constants of Nine C6-C9 Alkanes with OH from 230 to 379 K: Chemical
 - 931 Tracers for [OH], J. Phys. Chem. A2, 113, 5030–5038, doi:10.1021/jp810412m, 2009.
 - 932 TAbMET: TAbMEP Assessment: POLARCAT HNO3 Summary, **TAbMEP** in 933 POLARCAT/ICARTT Analysis, edited by G. Chen, https://www-934 air.larc.nasa.gov/TAbMEP2_polarcat.html, Boulder., 2009.
 - 935 Tang, W., Arellano, A. F., DiGangi, J. P., Choi, Y., Diskin, G. S., Agustí-Panareda, A., Parrington,
 - 936 M., Massart, S., Gaubert, B., Lee, Y., Kim, D., Jung, J., Hong, J., Hong, J.-W., Kanaya, Y., Lee,
 - 937 M., Stauffer, R. M., Thompson, A. M., Flynn, J. H. and Woo, J.-H.: Evaluating High-Resolution
 - 938 Forecasts of Atmospheric CO and CO₂ from a Global Prediction System during KORUS-AQ Field
 - 939 Campaign, Atmos. Chem. Phys. Discuss., 1–41, doi:10.5194/acp-2018-71, 2018.
 - 940 Thornton, J. A., Wooldridge, P. J., Cohen, R. C., Martinez, M., Harder, H., Brune, W. H.,

- 941 Williams, E. J. J., Roberts, J. M. M., Fehsenfeld, F. C., Hall, S. R., Shetter, R. E. E., Wert, B. P.
- and Fried, A.: Ozone production rates as a function of NO_x abundances and HO_x production rates in the Nashville urban plume, J. Geophys. Res., 107(D12), 1-17, doi:10.1029/2001JD000932,
- 944 2002.
- 945 Tohjima, Y., Kubo, M., Minejima, C., Mukai, H., Tanimoto, H., Ganshin, A., Maksyutov, S.,
- 946 Katsumata, K., Machida, T. and Kita, K.: Temporal changes in the emissions of CH4 and CO from
- 947 China estimated from CH4 / CO2 and CO / CO2 correlations observed at Hateruma Island, Atmos.
- 948 Chem. Phys., 14(3), 1663–1677, doi:10.5194/acp-14-1663-2014, 2014.
- Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R. and Jimenez, J. L.: Interpretation
 of organic components from Positive Matrix Factorization of aerosol mass spectrometric data,
 Atmos. Chem. Phys., 9(9), 2891–2918, doi:10.5194/acp-9-2891-2009, 2009.
- yor ranks. Chem. (1993, 9(9), 2091 2910, doi:10.319 //dep 9 2091 2009, 2009.
- Vay, S. A., Tyler, S. C., Choi, Y., Blake, D. R., Blake, N. J., Sachse, G. W., Diskin, G. S. and
- 953 Singh, H. B.: Sources and transport of $\Delta 14$ C in CO 2 within the Mexico City Basin and vicinity,
- Atmos. Chem. Phys., 9, 4973–4985, doi:10.159/acp-9-4973-2009, 2009.
- 955 Wang, Y., Munger, J. W., Xu, S., McElroy, M. B., Hao, J., Nielsen, C. P. and Ma, H.: CO₂ and its
- 956 correlation with CO at a rural site near Beijing: implications for combustion efficiency in China,
- 957 Atmos. Chem. Phys., 10(18), 8881–8897, doi:10.5194/acp-10-8881-2010, 2010.
- Williams, L.: What is My Vaporizer Temperature? Vaporizer Temperature Power Curve for
 Several Systems, [online] Available from: http://cires1.colorado.edu/jimenezgroup/UsrMtgs/UsersMtg11/WilliamsAMSUsersMtg_2010_VapT.pdf (Accessed 12 March
 2018), 2010.
- Woo, J.-H., An, S.-M., Kim, D.-Y., Kim, H.-K., Choi, K.-C. and Kim, Y.-H.: Development of the
 Asia Emission Inventory in Support of Integrated Modeling of Climate and Air Quality (III),
 Incheon, Korea., 2013.
- Xu, W., Croteau, P., Williams, L., Canagaratna, M., Onasch, T., Cross, E., Zhang, X., Robinson,
 W., Worsnop, D. and Jayne, J.: Laboratory characterization of an aerosol chemical speciation
 monitor with PM_{2.5} measurement capability, Aerosol Sci. Technol., 51(1), 69–83,
 doi:10.1080/02786826.2016.1241859, 2017.
- 969 Xu, W., Lambe, A., Silva, P., Hu, W., Onasch, T., Williams, L., Croteau, P., Zhang, X., Renbaum-
- 970 Wolff, L., Fortner, E., Jimenez, J. L., Jayne, J., Worsnop, D. and Canagaratna, M.: Laboratory
- 971 evaluation of species-dependent relative ionization efficiencies in the Aerodyne Aerosol Mass
- 972 Spectrometer, Aerosol Sci. Technol., 1–16, doi:10.1080/02786826.2018.1439570, 2018.
- Yuan, B., Hu, W. W., Shao, M., Wang, M., Chen, W. T., Lu, S. H., Zeng, L. M. and Hu, M.: VOC
 emissions, evolutions and contributions to SOA formation at a receptor site in eastern China,
- 975 Atmos. Chem. Phys., 13(17), 8815–8832, doi:10.5194/acp-13-8815-2013, 2013.
- Zhang, Q., Stanier, C. O., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., Pandis, S. N. and
 Jimenez, J. L.: Insights into the chemistry of new particle formation and growth events in
 Pittsburgh based on aerosol mass spectrometry, Environ. Sci. Technol., 38(18), 4797–4809,
 doi:Doi 10.1021/Es035417u, 2004.
- 280 Zhang, Q., Alfarra, M. R., Worsnop, D. R., James, D., Coe, H., Canagaratna, M. R. and Jimenez,
- 981 J. L.: Deconvolution and Quantification of Hydrocarbon-like and Oxygenated Organic Aerosols
- 982 Based on Aerosol Mass Spectrometry Deconvolution and Quantification of Hydrocarbon-like and

983 Oxygenated Organic Aerosols Based on Aerosol Mass Spectrometry, Environ. Sci. Technol.,
984 39(13), 4938–4952, doi:10.1021/es0485681, 2005.

Zhao, Y., Hennigan, C. J., May, A. A., Tkacik, D. S., de Gouw, J. A., Gilman, J. B., Kuster, W.
C., Borbon, A. and Robinson, A. L.: Intermediate-volatility organic compounds: a large source of
secondary organic aerosol, Environ. Sci. Technol., 48(23), 13743–50, doi:10.1021/es5035188,
2014.

989