

1 **Supplemental Material for Secondary Organic Aerosol Production from Local Emissions**
2 **Dominates the Organic Aerosol Budget over Seoul, South Korea, during KORUS-AQ**

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34 **SI 1. KORUS-AQ Overview**

35 **SI Table 1.** 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).

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

39 ^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.

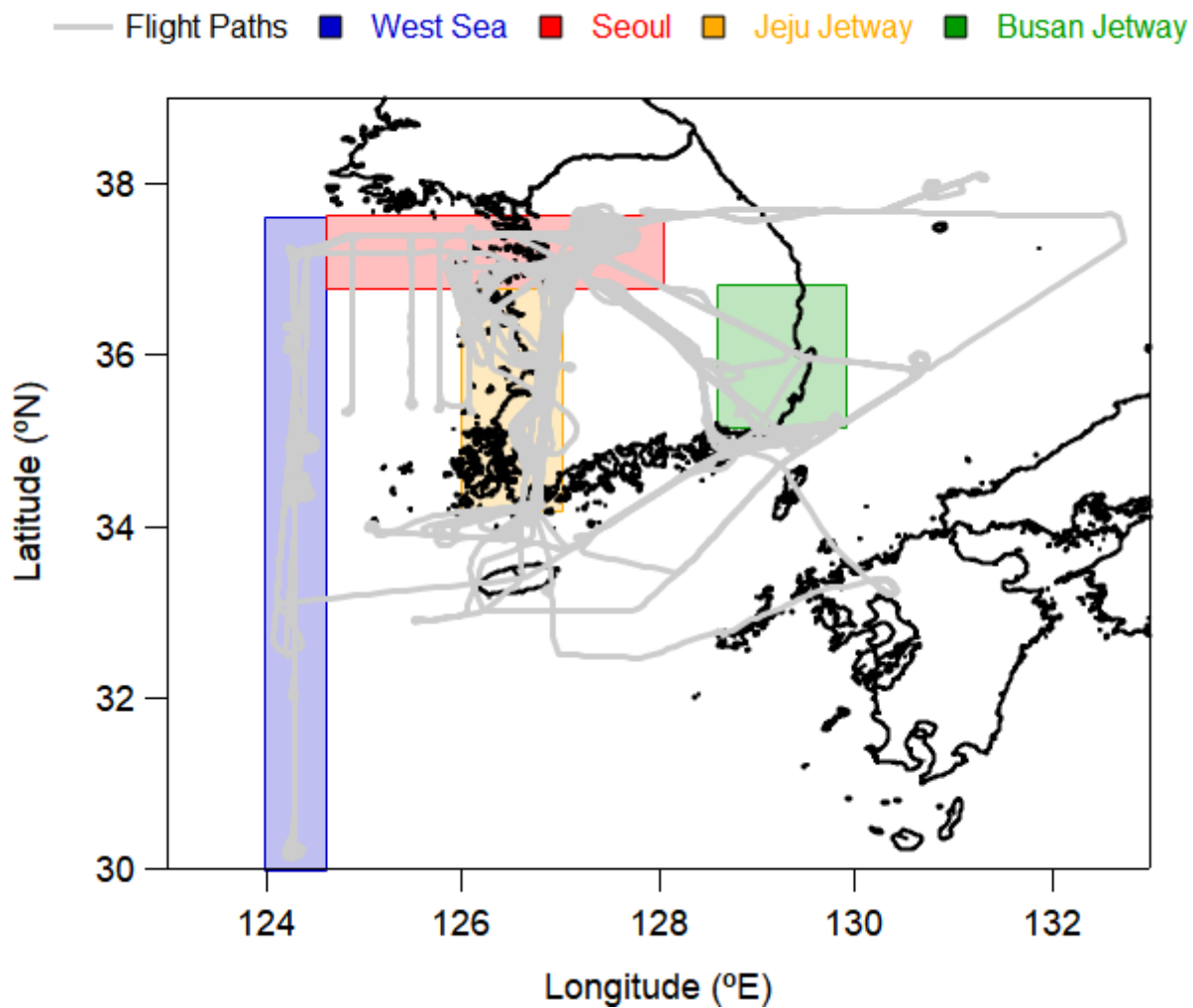
44 ^eThe DC-8 remained in the greater Seoul area to sample point sources.

45

46 **SI Table 2.** Description of the geographical locations used in Figure 1 and throughout the text,
 47 and shown in SI Figure 1..

<i>Location</i>	<i>Lat Min (°N)</i>	<i>Lat Max (°N)</i>	<i>Lon Min (°E)</i>	<i>Lon Max (°E)</i>
Seoul	36.8	37.6	124.6 ^a	128.0
West 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.



50

51 **SI Figure 1.** Geographical regions shown in SI Table 1. Note, the Seoul box is extended into the
 52 West Sea to capture the outflow of Seoul emissions for two flights (RF11 and RF18).

53

54 SI 2. CU-AMS Sampling and Calibration

55 After almost every flight, the ionization efficiency (IE) was calibrated (SI Figure 3) using
56 the single particle technique. Briefly, air containing 150 – 250 particles/cm³ of NH₄NO₃, of 400
57 nm (mobility diameter, sized with a built-in differential mobility analyzer, DMA, TSI model 3080)
58 was sampled by the AMS. Thresholds of 4 (*m/z* 30) or 3 (*m/z* 46) ions per event were selected to
59 produce a low, but detectable background (typically ~7 events/cm³ background). An event would
60 be recorded, after evaporation and ionization of NH₄NO₃ particle, if at least 4 (*m/z* 30) or 3 (*m/z*
61 46) ions were observed. These values were analyzed using the ToF AMS Ionization Efficiency
62 Calibration Panel for ET, v1.0.5F ([http://cires1.colorado.edu/jimenez-](http://cires1.colorado.edu/jimenez-group/ToFAMSResources/ToFSoftware/index.html#ToF_IE_Cal)
63 [group/ToFAMSResources/ToFSoftware/index.html#ToF_IE_Cal](http://cires1.colorado.edu/jimenez-group/ToFAMSResources/ToFSoftware/index.html#ToF_IE_Cal)), to process the data and
64 calculate IE and IE/AB (AB is air beam). Typical values during KORUS-AQ, for 400 nm
65 (geometric diameter) NH₄NO₃ calibrations were the following: 10 baseline segments and
66 minimum and maximum ions per particle values of 1 and 200. During KORUS-AQ, the average
67 IE/AB was $8.10(\pm 0.64) \times 10^{-13}$ ions/molecule of nitrate, which leads to an overall 10% variability
68 for this value during the whole campaign. Further details about using single particle technique for
69 IE/AB calibrations can be found in Nault et al. (2016).

70 These IE calibrations also provided relative ionization efficiency (RIE) calibrations of NH₄
71 after nearly every flight, as well (SI Figure 3), along with the NO⁺ and NO₂⁺ ratios of ammonium
72 nitrate, which are useful to estimate particle organic nitrate concentrations, as detailed in Fry et al.
73 (2013). The SO₄ and Chl RIEs were measured about once every week, and the interpolated values
74 were used for the SO₄ and Chl concentrations. For the organic aerosol, we used an RIE of 1.4
75 (Jimenez et al., 2016; Xu et al., 2018). Finally, to test the effects of solution mixtures on RIE for
76 SO₄ and NH₄, we made calibration solutions ranging from 0 – 100% NH₄NO₃, with the balance

77 coming from $(\text{NH}_4)_2\text{SO}_4$. We find no effects, both on the calculated NH_4 balance (SI Figure 6),
78 when using the NH_4 and SO_4 RIE's from the pure calibration, and on the recalculated NH_4 (SO_4)
79 RIE when keeping a constant SO_4 (NH_4) RIE from the pure calibrations (SI Figure 6). The
80 consistency in the NH_4 balance, as observed in prior studies (Docherty et al., 2011; Jimenez et al.,
81 2016), and the high precision (3% precision in all calculations) provides further confidence in the
82 stability of the RIEs for the species in calculating their mass in mixed particles, and indicates that
83 there are no effects on the RIE with changing composition, and, thus, CE (Jimenez et al., 2016).

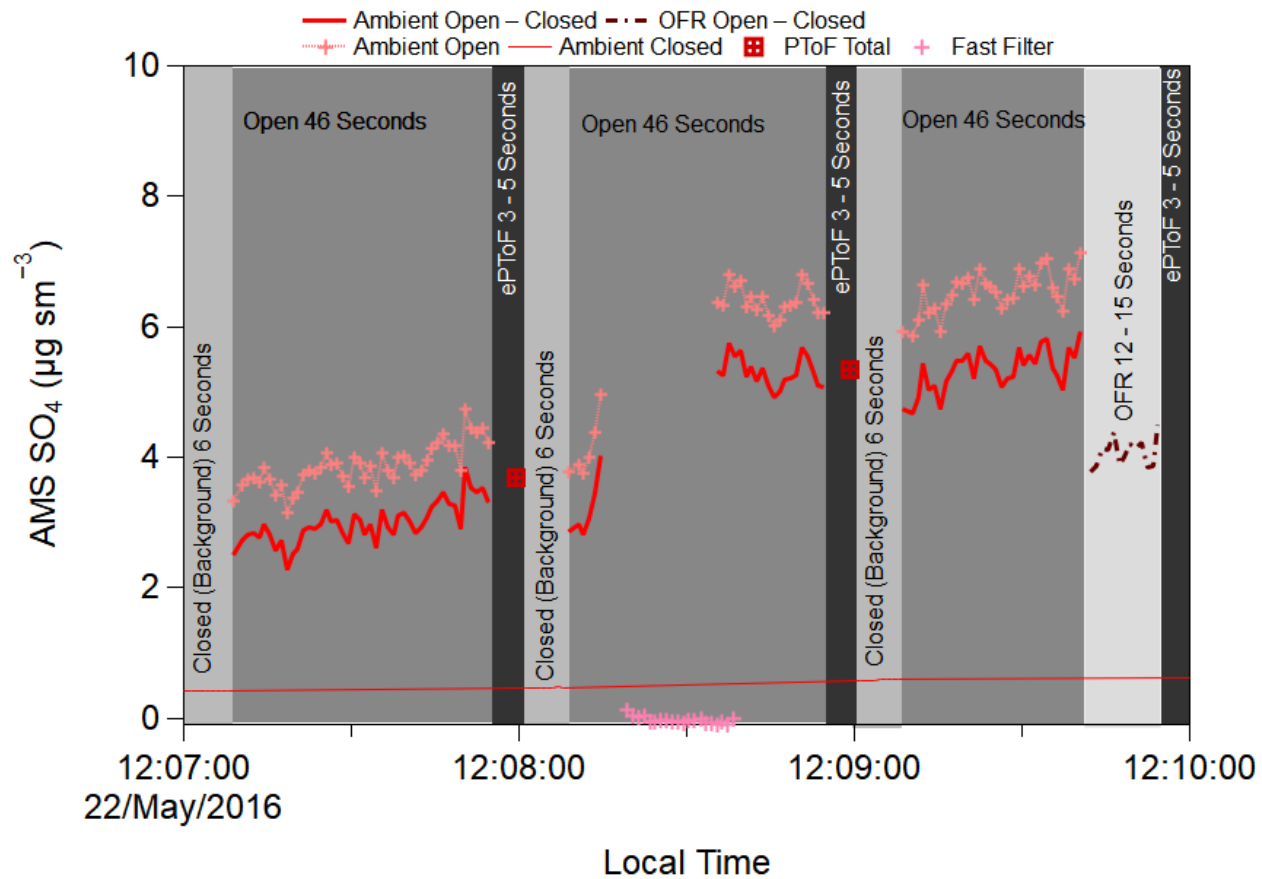
84 Also, the IE calibrations performed after each flight provided an opportunity to calculate
85 the effect of pNO_3 on producing a small artifact CO_2^+ signal, as detailed in Pieber et al. (2016),
86 and of pNO_3 on producing small artifact HCl^+ and Cl^+ signal, as detailed in Hu et al. (2017a) (SI
87 Figure 3). The CU-AMS data has been corrected for these small effects. The corrections were
88 typically 1% of CO_2^+ and 0.8% Chl.

89 Three different lens transmission calibrations to characterize the high end of the AMS
90 transmission curve were performed: (1) comparing the NH_4NO_3 mass measured with the CPC and
91 the CU-AMS between 200 – 450 nm (mobility diameter, d_m); (2) comparing the number of
92 particles measured with the CPC and the CU-AMS between 300 – 450 nm (d_m) using the single
93 particle vaporization technique detailed above; and (3) comparing the $(\text{NH}_4)_2\text{SO}_4$ mass measured
94 with the CPC and the CU-AMS between 250 – 450 nm (d_m), normalizing to the value at 250 nm.
95 The NH_4NO_3 diameters were converted to vacuum aerodynamic diameters (d_{va}), as discussed in
96 DeCarlo et al. (2004). As seen in SI Figure 4, both techniques show good agreement for the particle
97 transmission, and this transmission is similar to the recommended transmission curve in the
98 literature (Knote et al., 2011; Hu et al., 2017b). For this curve, it is assumed that the transmission
99 linearly increases from 0 – 100% between 40 – 100 nm (d_{va}) (Q. Zhang et al., 2004), remains 100%

100 between 100 – 550 nm (aerodynamic diameter), and decreases linearly from 100 – 0% between
101 550 nm – 1500 nm (d_{va}). This leads to a 50% cut-off of ~900 nm (d_{va}) during KORUS-AQ.

102 The particle sizing in the AMS Particle Time-of-Flight (PToF) mode was calibrated with
103 PSLs, ranging from 70 – 700 nm (geometric diameter) (SI Figure 5). This calibration was
104 compared against the velocities calculated from data collected during the NH_4NO_3 lens
105 transmission measurements. As seen in SI Figure 5, these two different methods to calibrate the
106 PToF velocity show comparable results, falling within the 95% confidence interval of the PSL
107 calibration. The fact that both PToF calibrations agree, and that the SMPS used for the AN
108 calibrations showed less than 2 nm deviation from the nominal PSLs diameters at all sizes
109 increases our confidence in accuracy of the IE calibration described above, and in particular on
110 lack of evaporation of NH_4NO_3 after its size selection in the DMA.

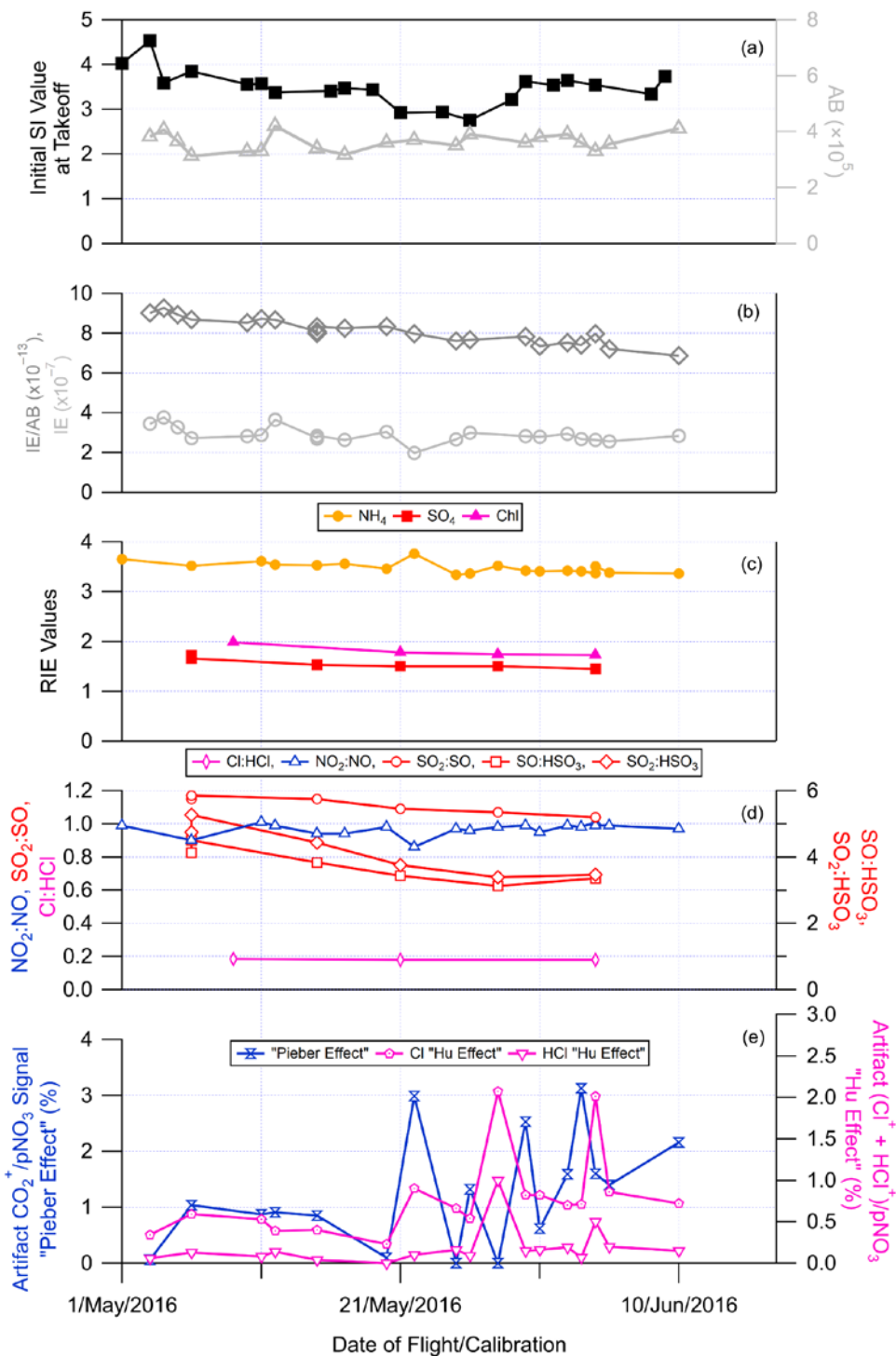
111 Finally, the vaporizer power, and thus, temperature, was calibrated by using monodisperse
112 NaNO_3 particles of $d_m = 350$ nm (SI Figure 8), as recommended by Williams (2010) and Hu et al.
113 (2017b). This method is more accurate than relying on the temperature reported by the
114 thermocouple on the AMS vaporizer, which can often be unreliable (Williams, 2010; Hu et al.,
115 2017b). The general idea is to increase the vaporizer power between ~1 – 7 W and locate where
116 the NaNO_3 full-width half maximum nearly remains constant, indicating that the vaporizer
117 temperature is ~600°C and allowing for maximum peaks in OA, pNO_3 , and SO_4 while minimizing
118 the influence of refractory species (Williams, 2010; Hu et al., 2017b).



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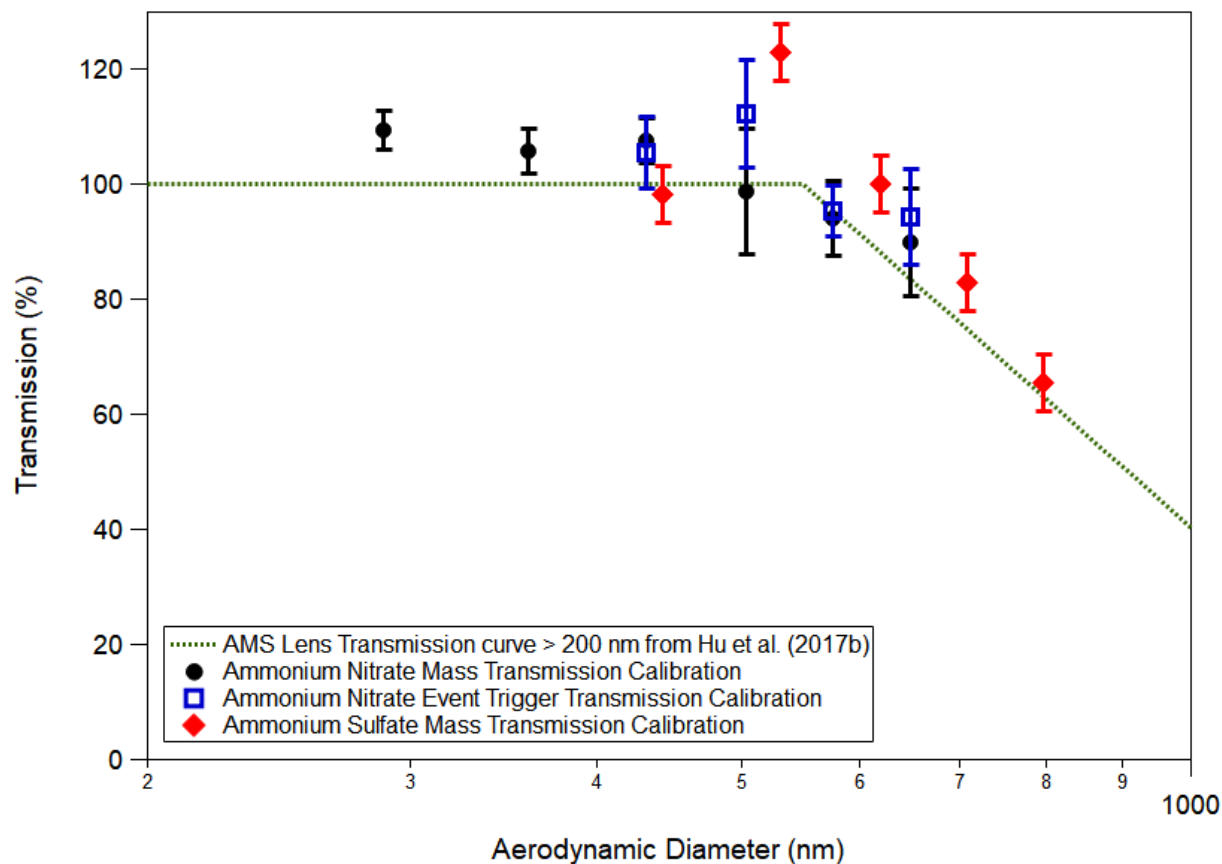
120 **SI Figure 2.** Example time series of the CU-AMS sampling scheme during KORUS-AQ.

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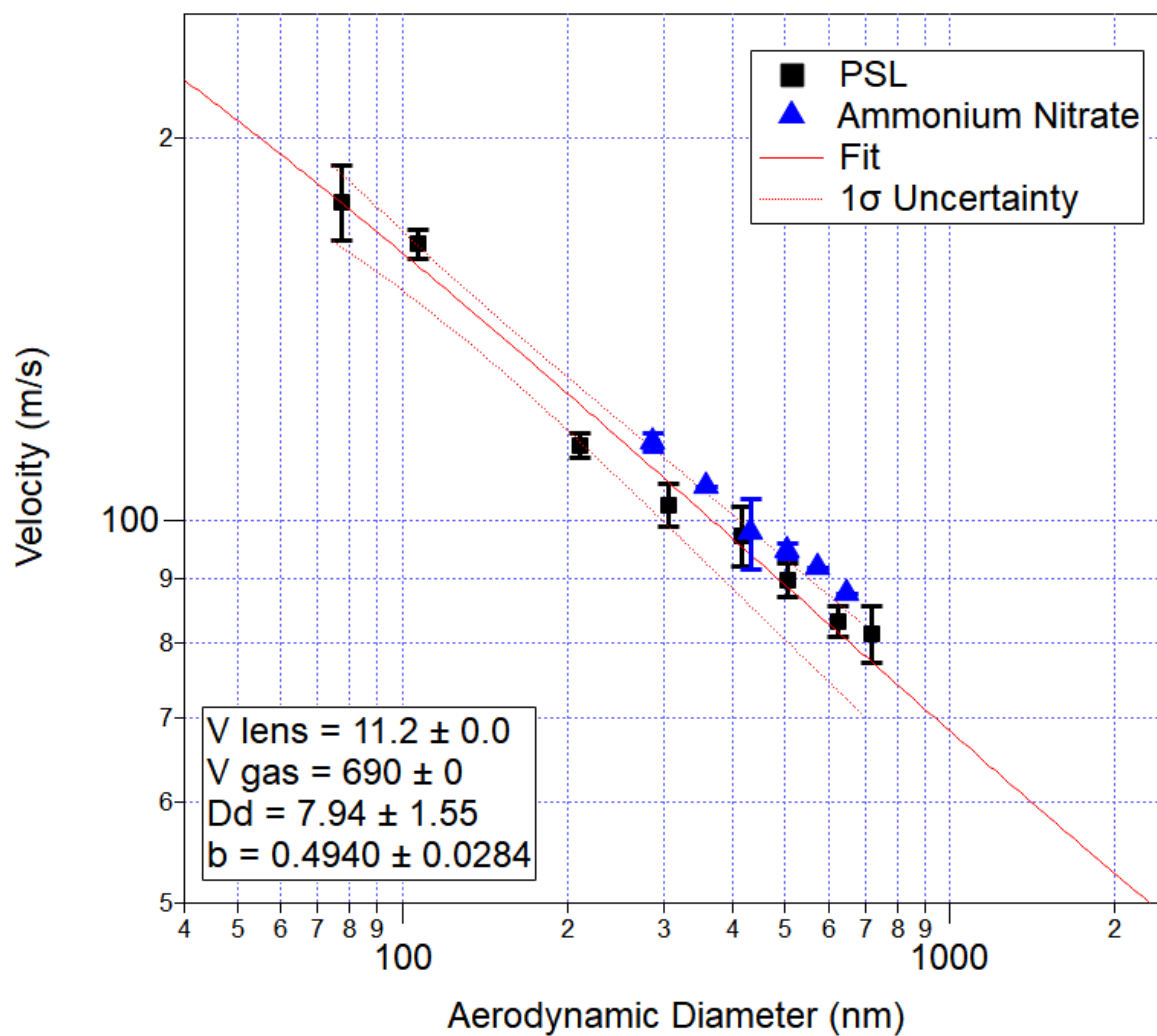
123 **SI Figure 3.** Time series of the (a) the Single Ion (SI) at take-off for each flight; (b) the air beam
 124 (AB, dark grey), ionization energy (IE, light grey), and IE/AB (middle grey) for each calibration;
 125 (c) the relative ionization energies (RIE) for ammonium (NH₄), sulfate (SO₄), and chloride (Cl)
 126 for each calibration; (d) the ratios of different ions for each calibration; and, (e) measured artifact
 127 signal ratios for CO₂⁺/pNO₃ “Pieber effect” (Pieber et al., 2016) and (Cl⁺ + HCl⁺)/pNO₃ “Hu
 128 effect” (Hu et al., 2017a) effects from each calibration.



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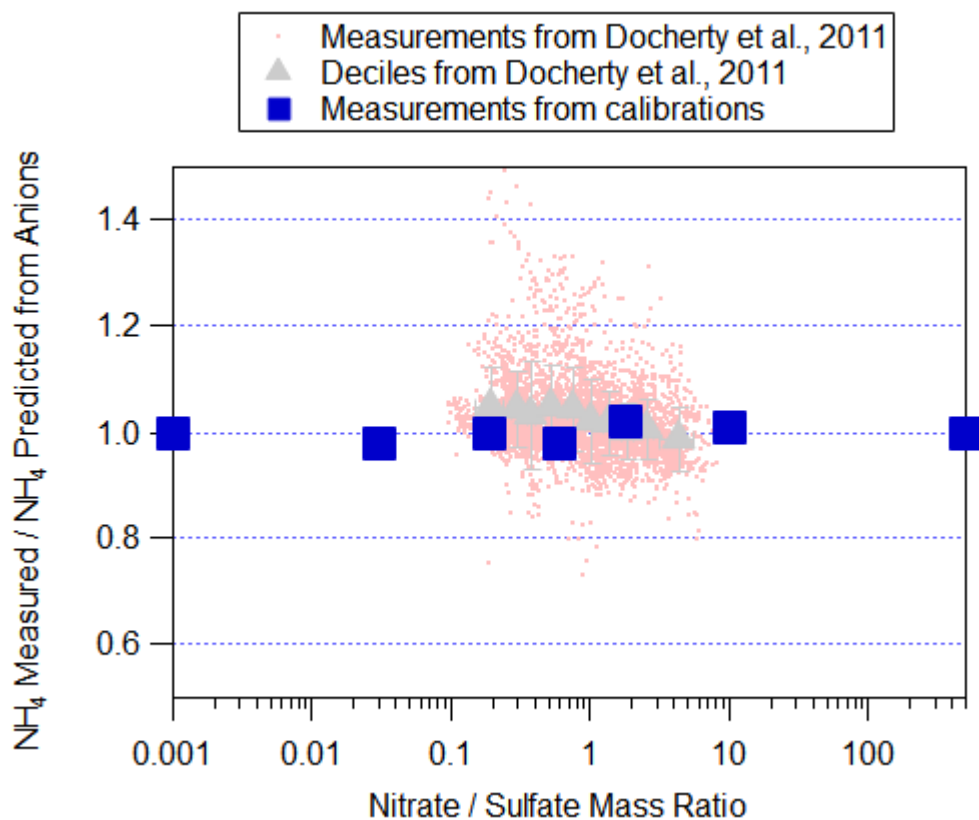
130 **SI Figure 4.** Measured transmission percentage of ammonium nitrate and ammonium sulfate
 131 versus vacuum aerodynamic diameters (nm) during KORUS-AQ. The green dashed-line is the
 132 expected transmission curve for the CU-AMS from the literature (Knote et al., 2011; Hu et al.,
 133 2017b). The black data represents the ammonium nitrate transmission curve using mass closure,
 134 from an experiment conducted on 09/May/2016. The blue data represents the ammonium nitrate
 135 transmission curve using single particle (“event trigger”) number closure, from an experiment
 136 conducted on 17May/2016. The red data represents the ammonium sulfate transmission curve
 137 using mass, from an experiment conducted on 06/May/2016. Finally, the error bars represent 1σ
 138 variability for the transmission at each size.

139



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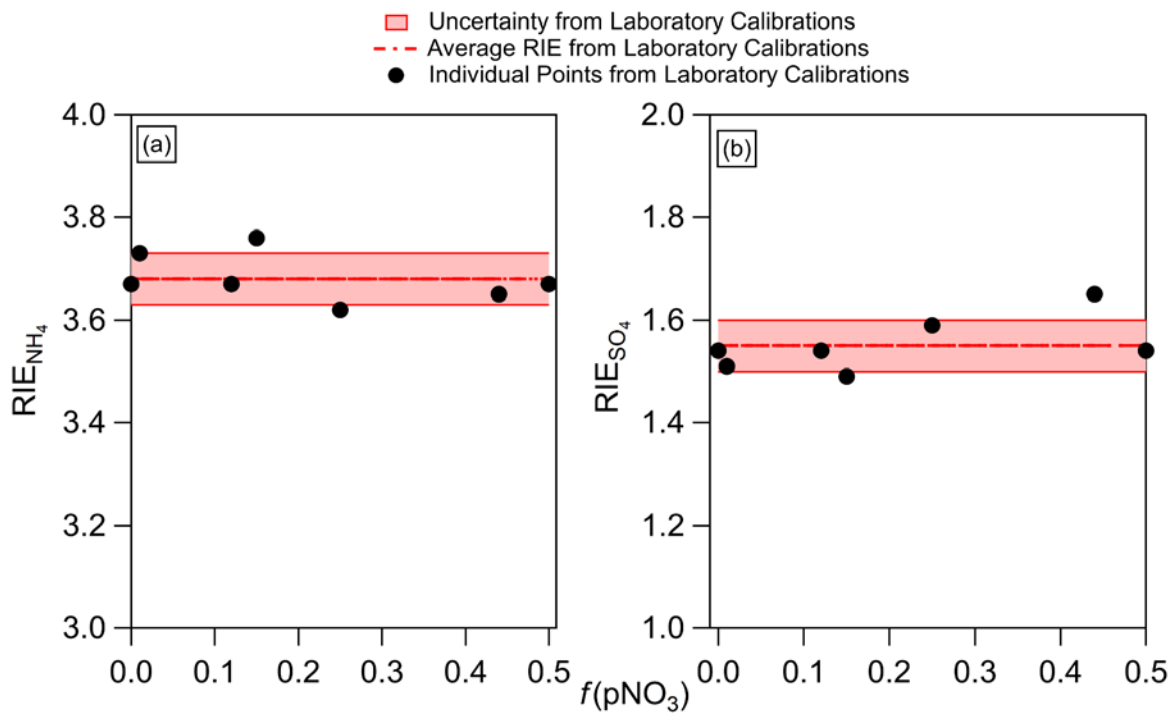
141 **SI Figure 5.** Particle velocity (m/s) inside the AMS vacuum chamber (after exiting the
 142 aerodynamic lens) versus vacuum aerodynamic diameter (nm) calibrations for the ePTof mode,
 143 using PSLs (black). Solid red line is the fit to the PSLs. The ammonium nitrate measured for the
 144 mass closure transmission curves (SI Figure 4) for comparison to the PSL values.



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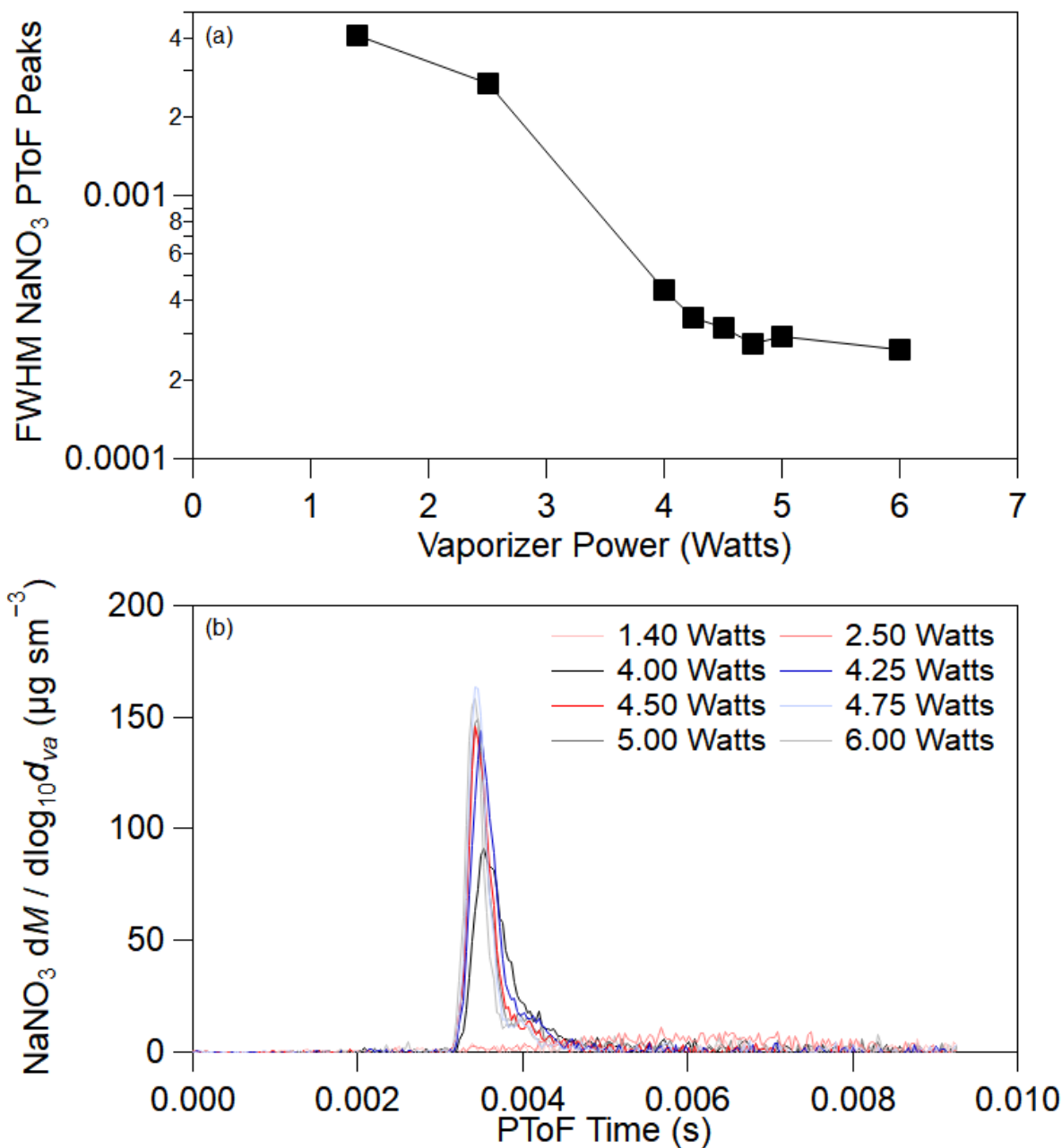
146 **SI Figure 6.** Ratio of measured and predicted NH₄ from anions versus ratio of nitrate to sulfate
 147 mass. Red points are from Docherty et al. (2011), grey triangles are deciles of the data from
 148 Docherty et al., and blue points are measurements from calibration solutions of varying mixtures
 149 of NH₄NO₃ and (NH₄)₂SO₄. Such consistency would be unexpected if a major fraction of the
 150 particle NH₄⁺ evaporated as intact salts, as suggested by Murphy (2016) (Hu et al., 2017b) .

151



152

153 **SI Figure 7.** (a) Plot of NH_4 RIE, keeping SO_4 RIE constant, versus the molar fraction of pNO_3
 154 measured in the solution, for calibration solutions of varying mixtures of NH_4NO_3 and $(\text{NH}_4)_2\text{SO}_4$.
 155 (b) Same as (a), but for SO_4 RIE and keeping NH_4 RIE constant. For both figures, the black dots
 156 are the values from the calibrations, the thick red line is the average of all the values, and the
 157 shaded red area is $\pm 1\sigma$.



158

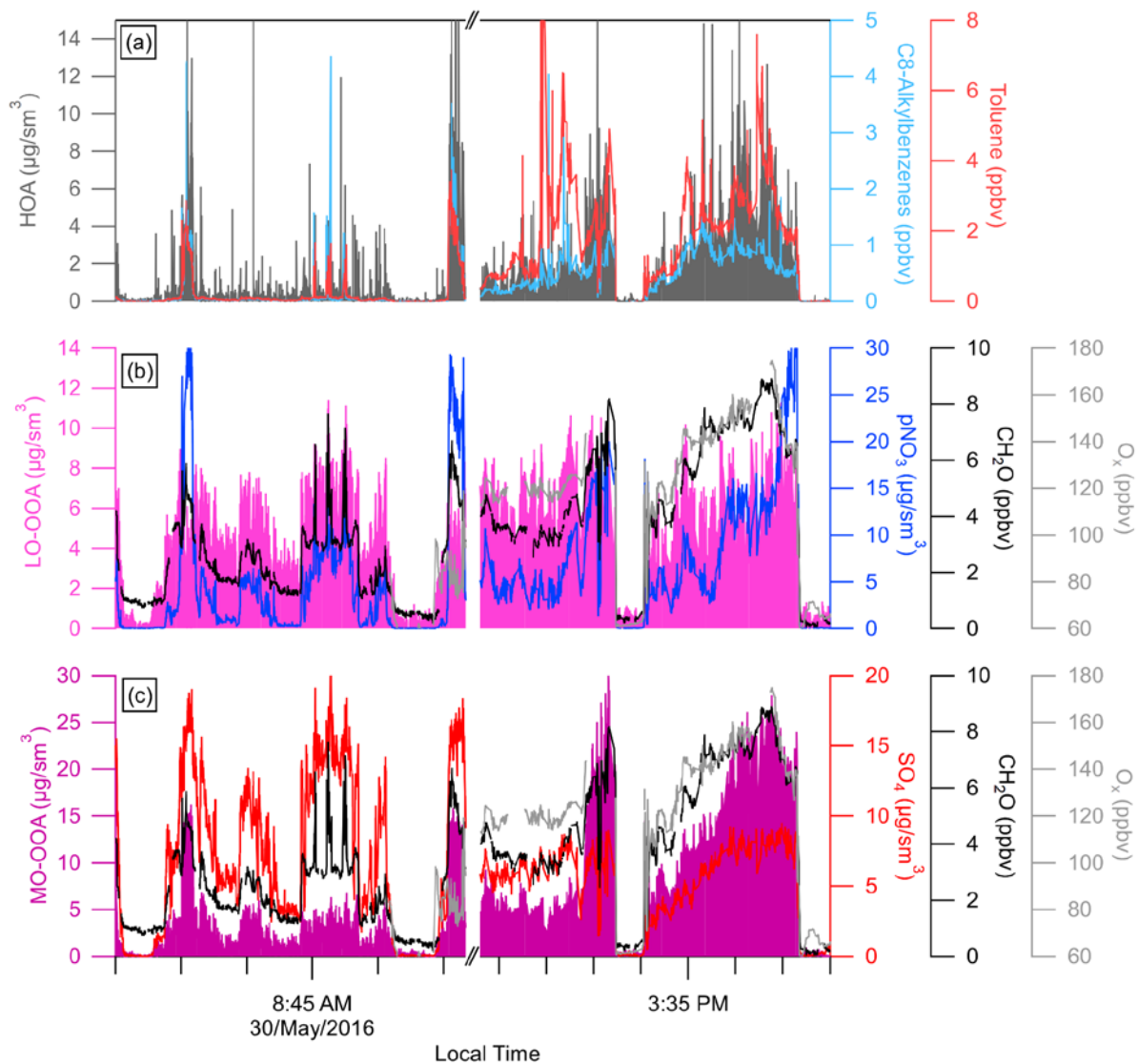
159 **SI Figure 8.** (a) Full-width half-maximum (FWHM) of NaNO_3 size distributions in PToF mode
 160 (b) vs. different vaporizer power inputs. See text for further details.

161

162 **SI 3. Application of Positive Matrix Factorization (PMF)**

163 Positive matrix factorization analysis (PMF, performed using the CU-Boulder PMF
164 Evaluation Tool PET-Panel v3.00A, [http://cires1.colorado.edu/jimenez-](http://cires1.colorado.edu/jimenez-group/wiki/index.php/PMF-AMS_Analysis_Guide#PMF_Evaluation_Tool_Software)
165 [group/wiki/index.php/PMF-AMS_Analysis_Guide#PMF_Evaluation_Tool_Software](http://cires1.colorado.edu/jimenez-group/wiki/index.php/PMF-AMS_Analysis_Guide#PMF_Evaluation_Tool_Software)) (Ulbrich
166 et al., 2009) was used to apportion the total OA aerosol into several components. PMF was run on
167 the combined CU-AMS 1 min organic ion matrix for all RFs together during KORUS-AQ. A 6-
168 factor solution was derived with an FPEAK value of 0. Based on comparisons with reference mass
169 spectra from the AMS high-resolution spectral database ([http://cires1.colorado.edu/jimenez-](http://cires1.colorado.edu/jimenez-group/HRAMSsd/#Ambient)
170 [group/HRAMSsd/#Ambient](http://cires1.colorado.edu/jimenez-group/HRAMSsd/#Ambient)), comparisons of time series (SI Figure 9), and correlations with other
171 trace species (SI Figure 11), the factors were recombined into more-oxidized, oxidized organic
172 aerosol (MO-OOA), less-oxidized, oxidized aerosol (LO-OOA), and hydrocarbon-like organic
173 aerosol (HOA) (SI Figure 10). HOA correlated with primary emissions (e.g., NO_x, various
174 hydrocarbons) whereas LO-OOA and MO-OOA correlated with secondary photochemical
175 products (e.g., CH₂O, PAN, pNO₃, SO₄). Here, primary OA is defined as the HOA factor and total
176 oxidized OA (OOA) as the LO-OOA plus MO-OOA factors. OOA is assumed to be dominantly
177 composed of secondary organic aerosol, which is supported by its strong correlation with other
178 secondary photochemical products as documented in the paper, as well as by many prior studies
179 (e.g., Jimenez et al., 2009; and references therein).

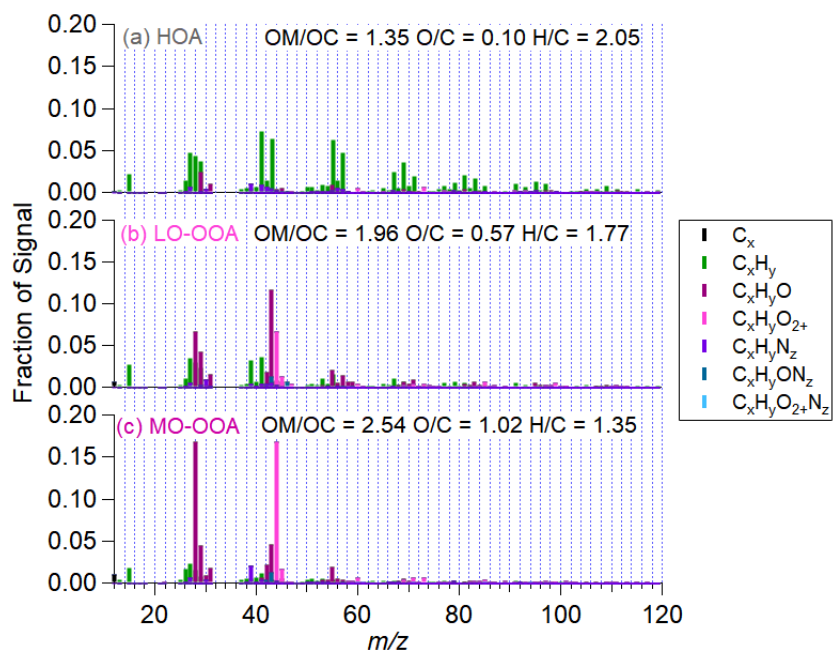
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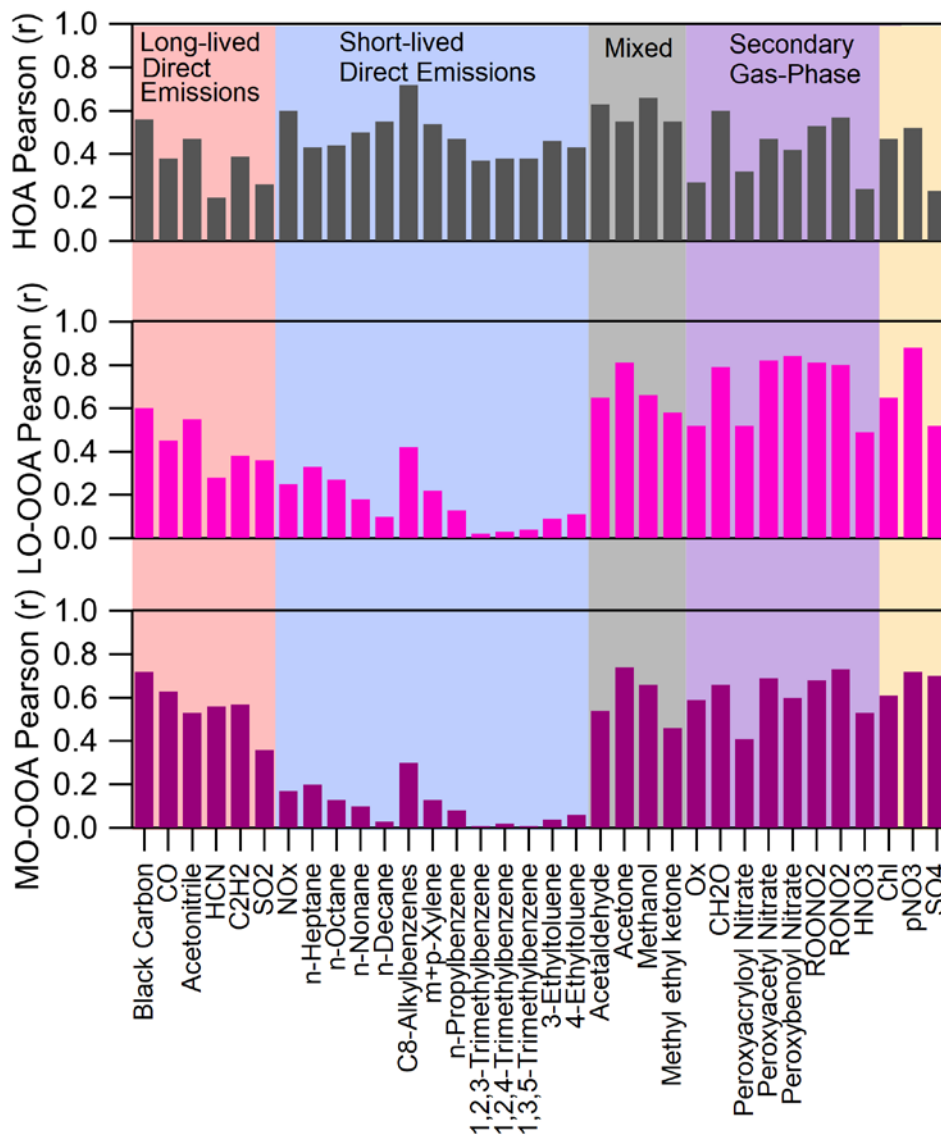
182 **SI Figure 9.** Example time series of the 3 PMF ((a) HOA, (b) LO-OOA, and (c) MO-OOA) results
 183 (left axes) and some species that correlate with the corresponding PMF results (right axes) from
 184 RF14. The morning and afternoon overpasses over Seoul, South Korea, are shown.

185



186

187 **SI Figure 10.** Mass spectra for PMF solution (a) HOA, (b) LO-OOA, and (c) MO-OOA for all of
 188 KORUS-AQ.

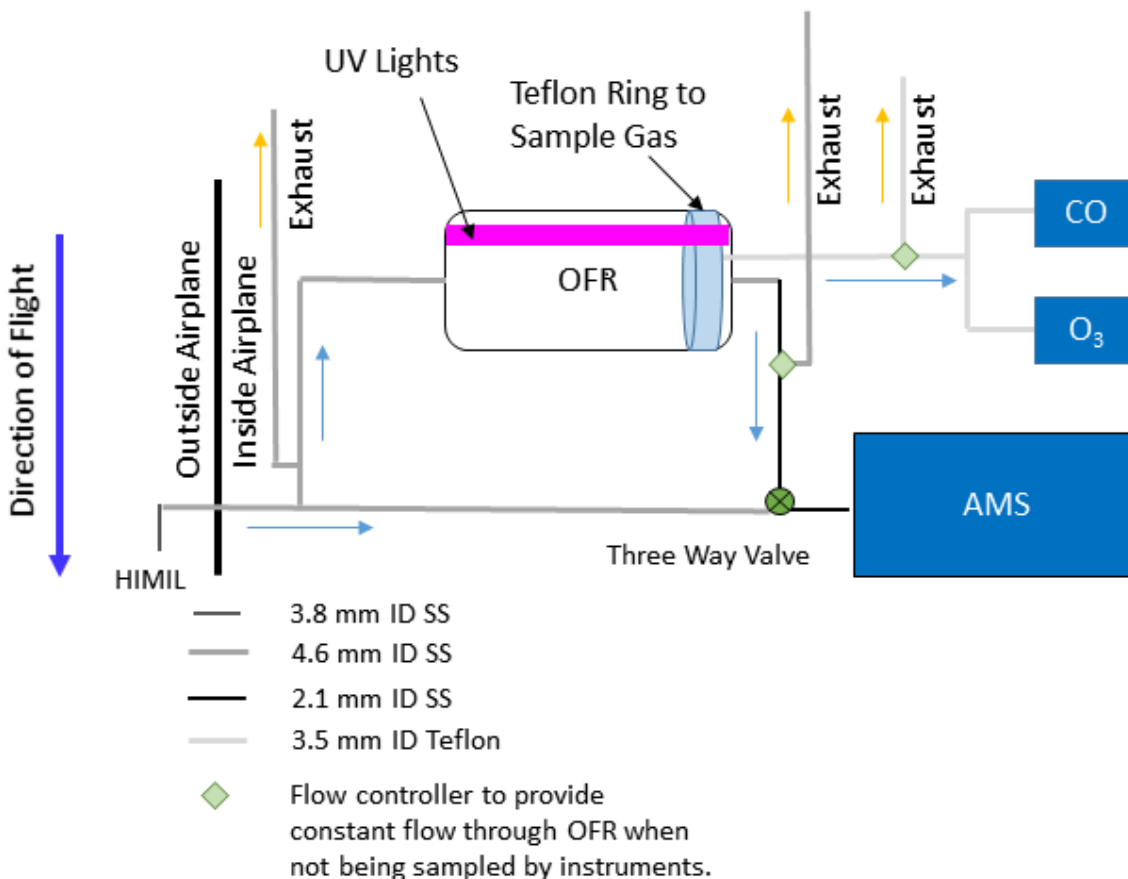


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190 **SI Figure 11.** Pearson correlation coefficients for HOA (grey, top), LO-OOA (light pink, middle),
 191 and MO-OOA (dark pink, bottom) factors versus species listed in x-axis. The background colors
 192 represent the dominant group of sources of the correlating species. The yellow in the far right
 193 indicates other PM₁ components measured by the CU-AMS.

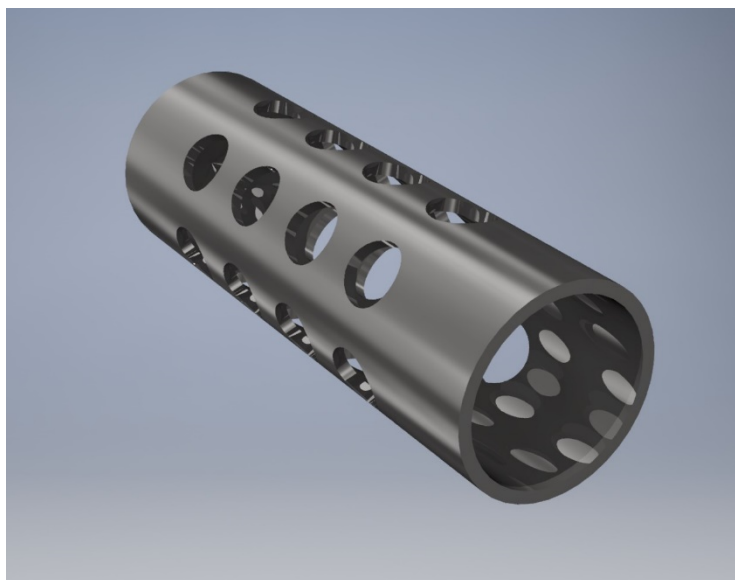
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195 **SI 4. Oxidation Flow Reactor (OFR) Sampling**

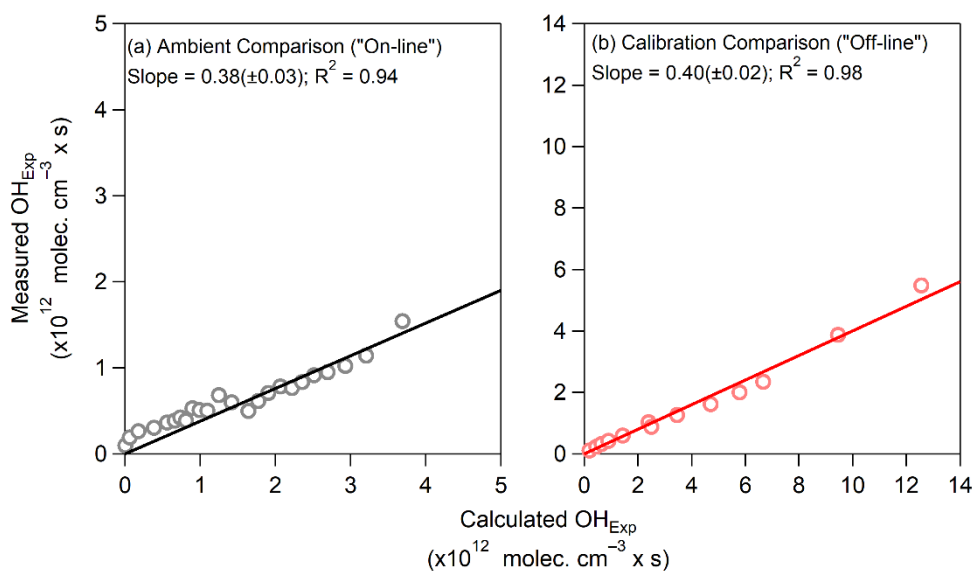


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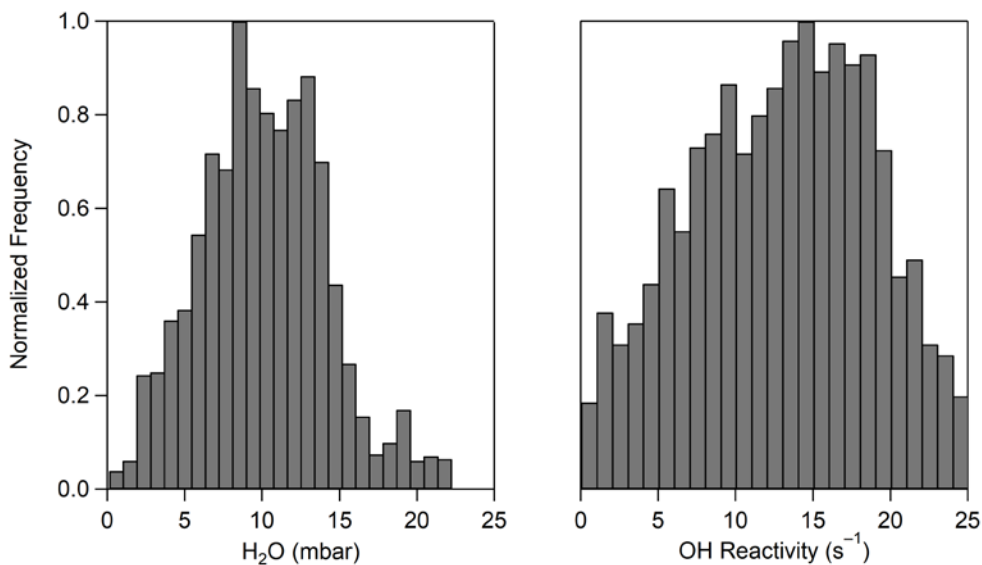
197 **SI Figure 12.** Schematic of the OFR sampling during KORUS-AQ. UV lamp is represented by
 198 the purple light in the OFR. Size and type of tubing is represented in figure, where ID is internal
 199 diameter and SS is stainless steel. Tubing distances were always as short as feasible and often
 200 shorter than represented, but they are stretched in this drawing for clarity



201
 202 **SI Figure 13.** 3D rendition of the computer model of the 1/2" press fitted stainless steel inlet, coated
 203 in SilcoNert (SilcoTek Co, Bellefonte, PA), used in the inlet of the OFR during KORUS-AQ, to
 204 avoid "short-circuiting" between the inlet and outlet of the OFR.



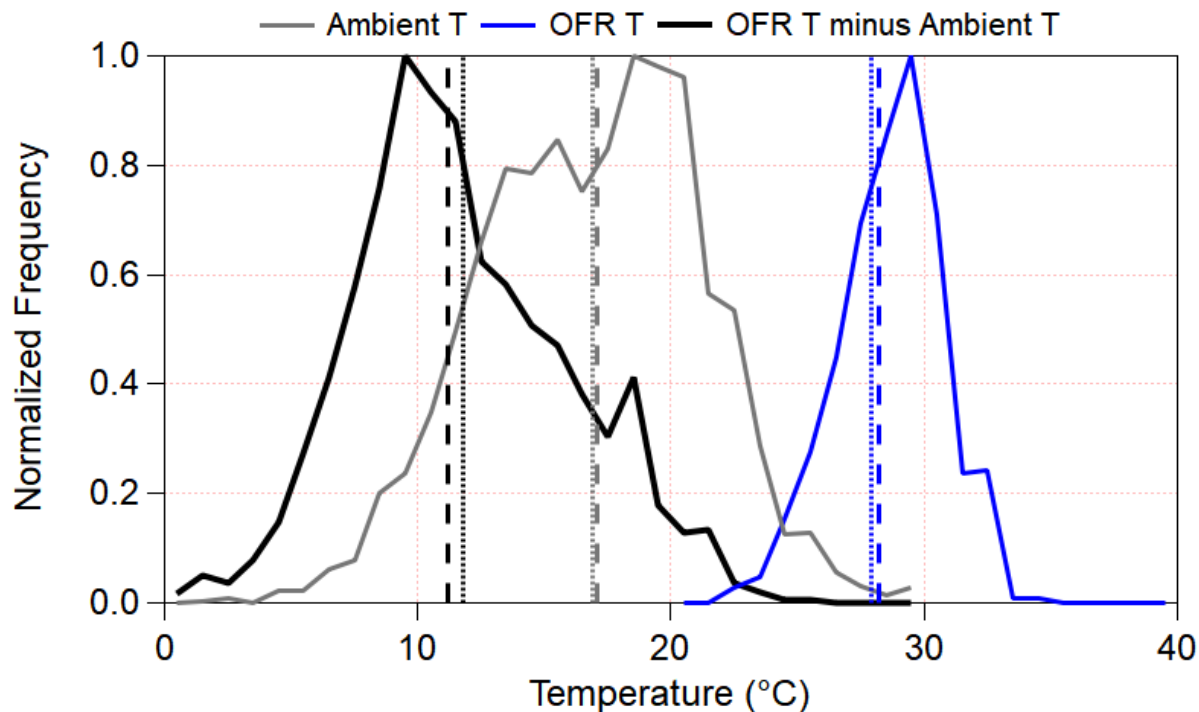
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 206 **SI Figure 14.** (left) Measured OH_{Exp} from the decay of CO in ambient air (measured by the
 207 DACOM instrument, see text) and OFR output air (measured by the Picarro instrument) and (right)
 208 measured OH_{Exp} from the decay of CO from a calibration cylinder versus calculated OH_{Exp} using
 209 the predictive expression in Peng et al. (2015). The calibration factor determined by this analysis
 210 was similar to past studies (Palm et al., 2016) and was applied to all data shown in this paper.



212

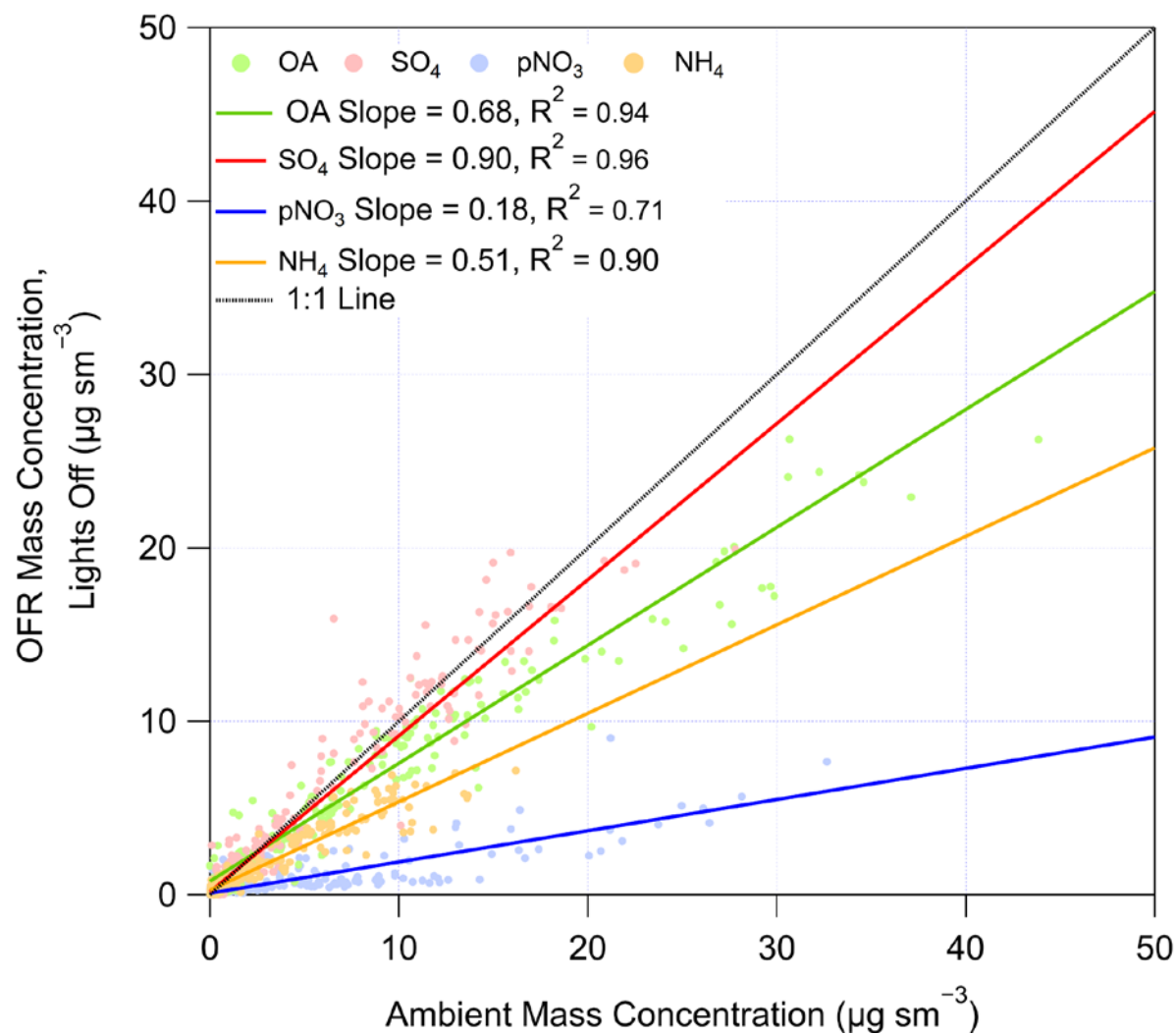
213 **SI Figure 15.** (left) Frequency distribution of water vapor below 2 km during KORUS-AQ. (right)
 214 Normalized histogram of measured OH reactivity (OHR) below 2 km during KORUS-AQ.

215



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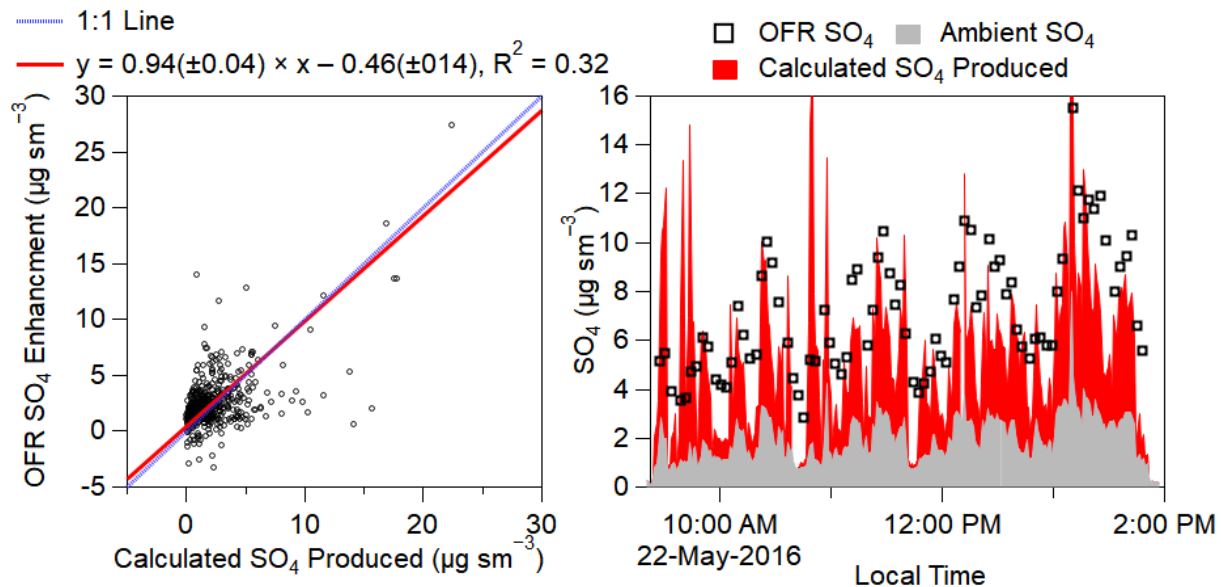
217 **SI Figure 16.** Frequency distribution of the ambient (black), OFR (blue), and difference between
 218 OFR and ambient temperature (grey) (°C). Vertical lines show the mean (long-dashed) and median
 219 (short-dashed) temperatures for the ambient, OFR, and difference between the two.



220

221 **SI Figure 17.** Comparison of organic (green), sulfate (red), nitrate (blue), and ammonium (orange)
 222 aerosol sampled through the OFR, with lights off, versus ambient aerosol. Under these conditions
 223 the OFR is just acting as a thermal denuder (e.g. Huffman et al., 2009), leading to evaporation of
 224 some aerosols due to increased temperature in the aircraft cabin vs. outside. In addition, small
 225 particle losses in lines and the OFR are observed for sulfate, which is generally non-volatile. Figure
 226 is adapted from Heim et al. (2018). See text for further details and discussion.

227



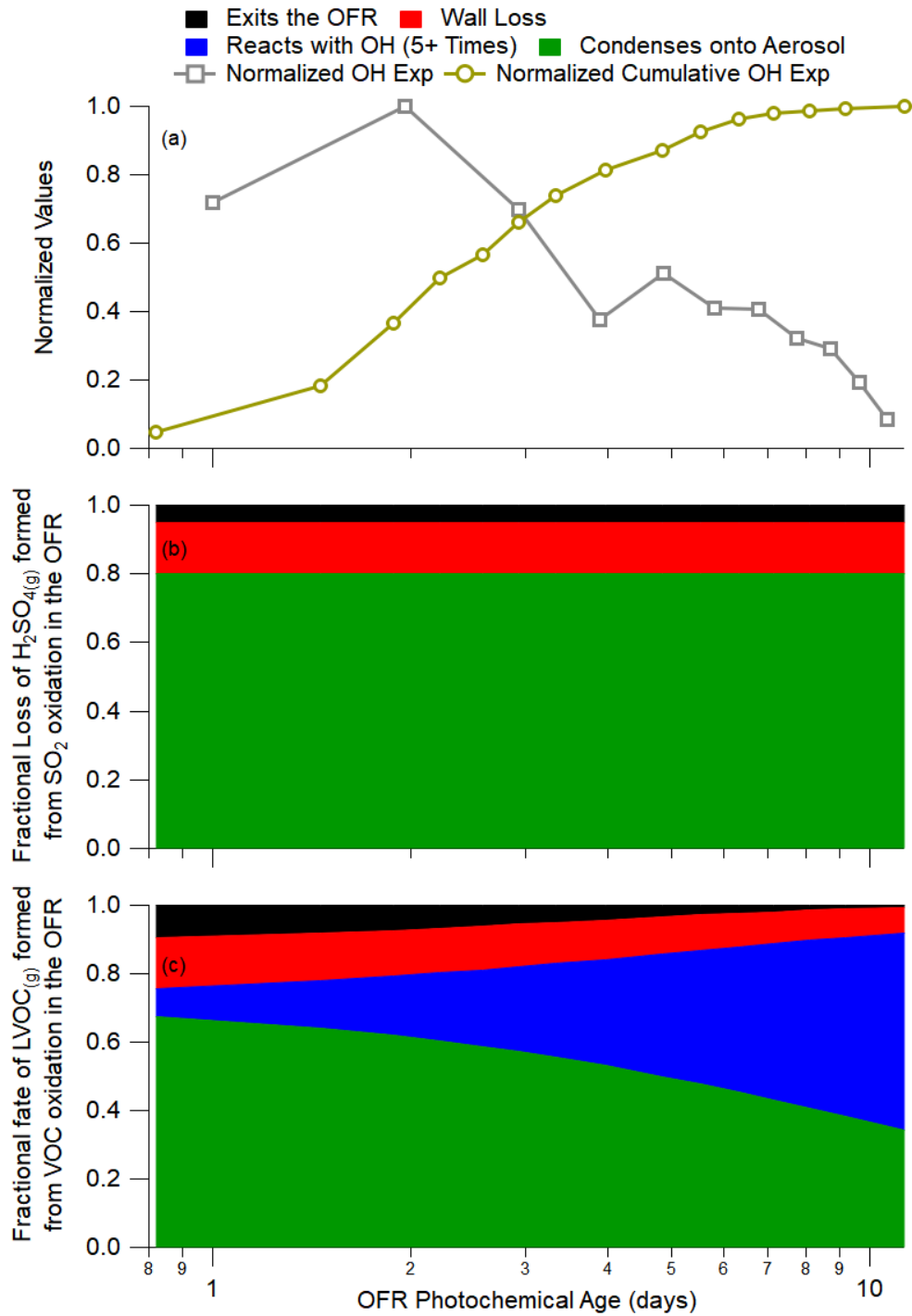
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229 **SI Figure 18.** (a) Scatter plot of OFR SO₄ Enhancement (OFR – Ambient) versus calculated SO₄
 230 produced, using SO₂ observations, estimated OH_{exp}, and condensation fate correction. (b) Time
 231 series of OFR SO₄ (black squares), ambient SO₄ (light grey), and calculated SO₄ (dark red) for the
 232 RF11 flight.

233 *Analysis of CS Values for KORUS-AQ*

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

244 condensing to the wall (SI Figure 19). Note that the further reactions with OH are not relevant for
 245 H₂SO₄, and thus they have not been included in the analysis shown in in Fig. SI-18.



246

247 **SI Figure 19.** (a) Observed normalized frequency and cumulative frequency of OH Exposure
 248 observed during KORUS-AQ in the OFR. (b) Calculated fate of the SO₂ oxidized in the OFR

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

254

255 SI 5. Calculation of Photochemical Age over Seoul, South Korea

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

$$260 \quad t = \frac{\ln\left(\frac{NO_x}{NO_y}\right)}{k_{OH+NO_2}[OH]} \quad (S1)$$

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

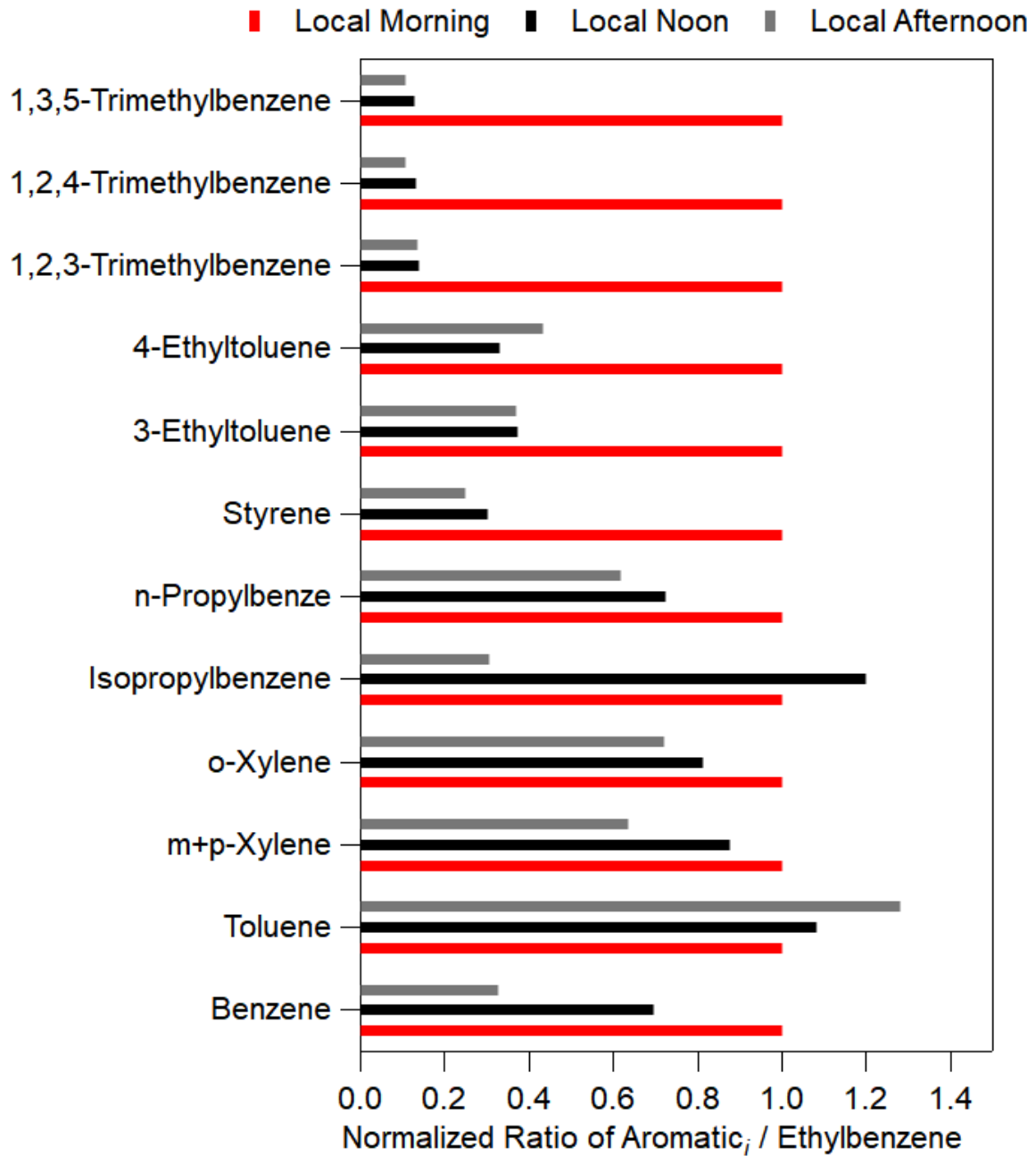
267 For the aromatic photochemical clock over Seoul, the more reactive aromatics
268 (ethylbenzene in the denominator) are utilized, which should be more sensitive to the short
269 photochemical aging observed over Seoul (Parrish et al., 2007), along with Eq. SS2.

$$270 \quad t = -\frac{1}{[OH] \times (k_{aromatic_i} - k_{ethylbenzene})} \times \left(\ln\left(\frac{aromatic_i(t)}{ethylbenzene(t)}\right) - \ln\left(\frac{aromatic_i(0)}{ethylbenzene(0)}\right) \right) \quad (S2)$$

271 where t is the time, in hours, the k 's are the corresponding OH rate constants for each aromatic
272 compound (SI Table 3), and the third term $\left(\ln\left(\frac{aromatic_i(0)}{ethylbenzene(0)}\right)\right)$ corresponds to the emission ratios
273 for those two aromatic compounds. Similar to the NO_x clock, we assume [OH] = 1.5×10^6
274 molecules/cm³ for standardization. The aromatics measurements used in this calculation are from
275 WAS.

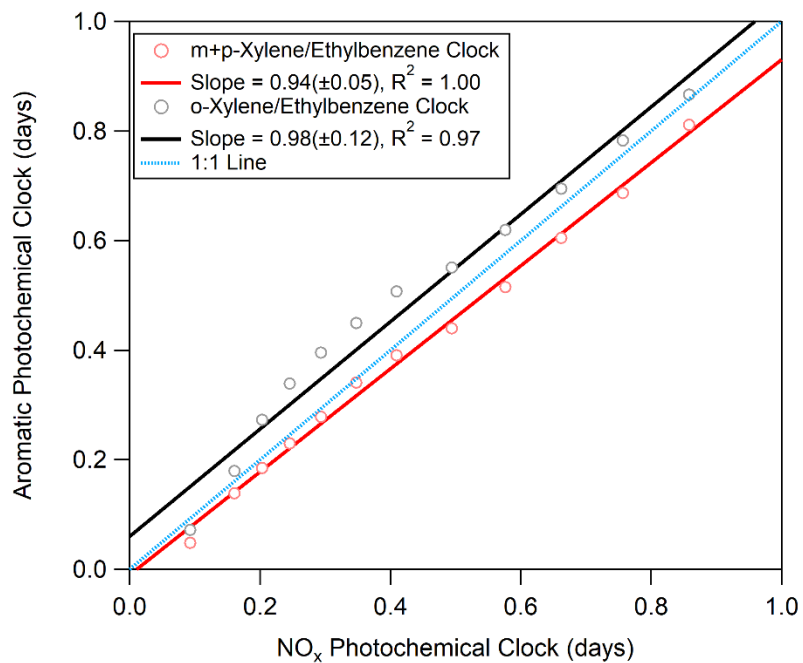
276 To evaluate which aromatic compounds to use in the clock, the behavior of the ratios of
277 each aromatic compound with ethylbenzene versus the three missed approaches (morning, noon,
278 and afternoon) over Seoul during KORUS-AQ (SI Figure 20) are compared. The idea is that if the
279 2 aromatic compounds are co-emitted, the ratios should be removed proportionally to their OH
280 rate constants. E.g. for faster reacting compounds (e.g., o-xylene), the ratio to ethylbenzene should
281 decrease with time as more o-xylene was consumed, compared to ethylbenzene, by OH (de Gouw
282 et al., 2017). On the other hand, for slower reacting compounds (e.g., toluene), the ratio to
283 ethylbenzene should increase with time as more ethylbenzene was consumed by OH. Also, this
284 analysis provides an indication of which ratios would provide meaningful results throughout the
285 entire day (de Gouw et al., 2017). Ideally, there should be a decrease with each later missed
286 approach, and not a leveling off after two missed approaches (e.g., the trimethylbenzenes and
287 ethyltoluenes). Only the m+p-xylene/ethylbenzene and o-xylene/ethylbenzene ratios meet this
288 criterion. Finally, to determine the emission ratios, we calculated what the m+p-
289 xylene/ethylbenzene and o-xylene/ethylbenzene ratio was for observations where the NO_x
290 photochemical was less than 0.07 days (corresponding to less than 10% of either species being
291 consumed). Comparing these two aromatic clocks to the NO_x photochemical clock (SI Figure 21),
292 a similar agreement between the two aromatic clocks with the NO_x photochemical clock was
293 observed, providing confidence in using all three clocks to calculate photochemical age to evaluate
294 OA production over Seoul. For the remainder of the paper, we mainly use the NO_x photochemical
295 clock to eliminate the uncertainty of the emission ratios, unless otherwise noted.

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



300

301 **SI Figure 20.** Comparison of various aromatic compounds/ethylbenzene ratios sampled over
 302 Seoul, South Korea, during KORUS-AQ. The ratios are normalized by the morning ratios.



303

304 **SI Figure 21.** Binned scatter plot of the aromatic photochemical clock ages versus NO_x
 305 photochemical clock ages for all observations over Seoul. All ages are normalized to OH = 1.5×10⁶
 306 molecules/cm³.

307 **SI Table 3.** Rate constants used throughout this study. Unless noted otherwise, rate constants
 308 without temperature dependence only have a value measured at 298 K.

Reaction	Rate Constant (cm ³ /molecules/s)	Reference
<i>Inorganic</i>		
CO	$2.28 \times 10^{-13,a}$	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)
<i>Alkanes</i>		
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)
<i>Alkenes</i>		
Ethylene	$7.84 \times 10^{-12,a}$	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)
<i>Aromatics</i>		
Benzene	$2.3 \times 10^{-12} \times \exp(-190/T)$	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)
<i>S/IVOCs</i>		

S/IVOC	2×10^{-11}	Ma et al. (2017)
<i>Biogenics</i>		
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)
<i>Radicals</i>		
NO + RO ₂	$2.8 \times 10^{-12} \times \exp(300/T)$	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)

309 ^aShowing the rate constant at 298 K, 1013 hPa. However, for this study, we used the temperature
310 and pressure dependent formulation listed in each respective reference.

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

312

313 **SI 6. Potential SOA Calculations**

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

$$325 \quad P(OA) = \sum_i Y_i \times \Delta RH_i \quad (\text{S3})$$

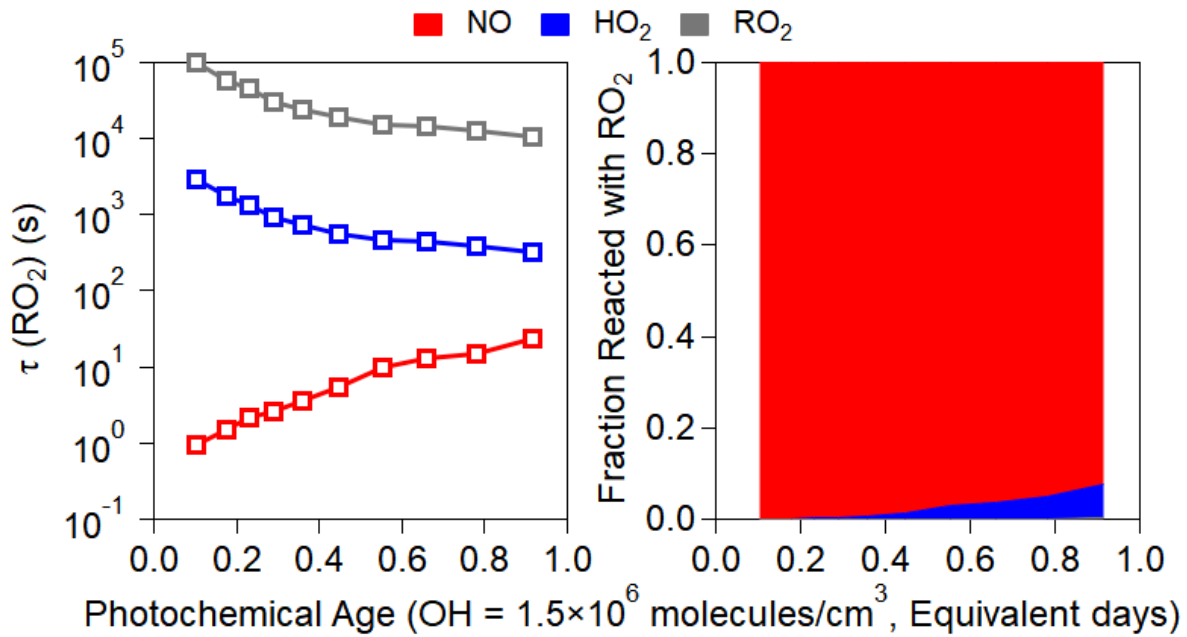
$$326 \quad \Delta RH = \frac{RH(t)}{e^{(-k[\text{OH}]t)}} - RH(t) \quad (\text{S4})$$

327 The hydrocarbons measured on the DC-8 were the concentrations at time, t ; thus, Eq. SS4
328 was used, which takes into account the amount of OH that oxidized the hydrocarbon ($\text{OH}_{\text{exp}} =$

329 $[\text{OH}]t$) between emissions and measurement, and k is the OH rate constant for each specific
330 hydrocarbon (SI Table 3).

331 Finally, to determine the fate of the RO_2 radical in the reactions over Seoul (high/low NO_x
332 regime), and thus, what aerosol yields to use, the RO_2 lifetime with reaction of NO, HO_2 , and RO_2
333 versus photochemical age was calculated (SI Figure 22). The measured NO and HO_2 was used in
334 the calculations, we assumed RO_2 was approximately the same concentration as HO_2 in this
335 calculation (Thornton et al., 2002), and the rate constants in SI Table 3 were used to calculate the
336 lifetime and fractional fate of RO_2 . The fate of RO_2 with autoxidation is not included as the rate is
337 still uncertain (Crouse et al., 2013) and it should be less important in highly polluted
338 environments such as Seoul, especially at the lower photochemical ages (< 0.5 eq. days) where
339 most SOA is observed to be formed. The dominant sink of RO_2 over Seoul during KORUS-AQ is
340 the reaction with NO, suggesting that the SOA yields for “high NO” conditions should be used to
341 describe the production of SOA.

342



343

344 **SI Figure 22.** (left) Lifetime of RO₂ due to reactions with NO (red), HO₂ (blue), and RO₂ (grey)
345 versus NO_x photochemical clock, normalized by OH = 1.5×10^6 molecules/cm³. (right) Fraction of
346 RO₂ reacting with NO (red), HO₂ (blue), or RO₂ (red) versus NO_x photochemical clock,
347 normalized by OH = 1.5×10^6 molecules/cm³. Values are calculated using observations over Seoul,
348 South Korea, during KORUS-AQ, and RO₂ is assumed to be approximately equal to HO₂
349 (Thornton et al., 2002).

350 **SI 7. FLEXPART Source Analysis**

351 Source contributions have been estimated using Lagrangian backtrajectory calculations with the
352 FLEXPART-WRF model (Brioude et al., 2013) in version 3.3.1, driven by meteorological output
353 from NCEP GFS (NCEP) analyses downscaled to 5 km horizontal resolution using the Weather
354 Research and Forecasting (WRF) model (Skamarock et al., 2008) in combination with
355 the CREATE emission inventory (Woo et al., 2013). Approximately 20,000 parcels are released
356 in 1 min intervals from the then-current location of the DC-8 during its research flights and parcel
357 trajectories are followed back in time for 24 hours. The total time parcels spent in the lowermost
358 100 m—as surrogate for air having contact with an emission source at the ground—is recorded
359 (residence time, [$\text{s kg}^{-1} \text{m}^3$]) and then folded with the emission fluxes ($[\text{kg m}^{-2} \text{s}^{-1}]$) given by the
360 CREATE inventory for different compounds and source regions. This delivers an estimate of the
361 source contribution (as increment in volume mixing ratio at the receptor, i.e., the DC-8 location)
362 of the emissions of a given compound from a given region, assuming a perfect transport simulation
363 and an inert compound.

364 **SI 8. Intercomparisons of CU-AMS with Other Measurements on the NASA DC-8**

365 We evaluate the measurement comparisons of the CU-AMS versus other aerosol
366 measurements on-board the DC-8 during KORUS-AQ. We start with the mist chamber / ion
367 chromatograph instrument (MC/IC), which has a comparable size cut as the AMS. The
368 comparisons for SO₄ show good correlation ($R^2 = 0.76$) and slope close to 1 (0.95) (SI Figure 23).
369 The higher scatter for the MC/IC is thought to arise from the lag and smearing in the measurements
370 that has been observed in prior studies (TAbMET, 2009). For example, the correlation between
371 instruments without lag and smearing have R^2 of 0.87 – 0.91 (CU-AMS versus extinction and CU-
372 AMS vs K-AMS for certain RFs). If the MC/IC and CU-AMS SO₄ measurements are averaged to
373 the sampling frequency of the University of New Hampshire filters (not shown), the R^2 improves
374 (0.82) with no impact on the slope.

375 The comparison between the UNH filters and CU-AMS SO₄ shows higher R^2 (0.86) but
376 lower slope (0.80), compared to MC/IC vs. CU-AMS. The higher R^2 is likely due to longer
377 averaging time and lack of smearing that occurred with the MC/IC. As a comparison, the R^2
378 between MC/IC and filters are 0.84. The lower slope for the filters than the MC/IC is thought to
379 be due to the different size cut-offs for the two measurements. For the filters, the upper size cut-
380 off is $\sim 4 \mu\text{m}$ (McNaughton et al., 2007); whereas, the upper size cut-off for the MC/IC is
381 comparable to the AMS aerosol size cut-off ($\sim 1 \mu\text{m}$ aerodynamic). This means that the filter
382 samples may include SO₄²⁻ from sea salt (sodium and calcium) and dust (calcium) (Heo et al.,
383 2009; Kim et al., 2016; Heim et al., 2018). This is shown in SI Figure 24 and described in detail
384 in Heim et al. (2018). Heim et al. (2018) found that dust dominated supermicron aerosol for
385 approximately half of the campaign, and during these periods, supermicron SO₄²⁻ accounted for
386 $\sim 50\%$ of the total SO₄²⁻ (sub plus supermicron). Taken together, the comparisons of SO₄ mass

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

389 Next, we compare the non-refractory species concentrations measured by the CU- and K-
390 AMS. Intercomparisons between these two measurements for a few flights have been presented in
391 prior publications (Hu et al., 2018a, 2018b). The K-AMS used a capture vaporizer, which leads to
392 CE of ~1 for all ambient species (Hu et al., 2017a, 2017b; Xu et al., 2017). Here, we investigate
393 the entire campaign. As shown in SI Figure 25, R² > 0.80 for all five species, and all slopes fall
394 within ±20% of unity, which is within the combined uncertainty of both AMSs (~27%). However,
395 at high concentrations (greater than ~5 – 10 μg sm⁻³), the scatter between the two measurements
396 increases, and for some species (e.g., SO₄), there is a slight curvature in the comparisons, where
397 CU-AMS is greater than K-AMS. We believe this discrepancy originated from differences in
398 transmission vs. particle size through the aerosol inlet and focusing lens (SI Figure 26). In-field
399 calibrations showed that The K-AMS had 50% transmission at 615 nm (vacuum aerodynamic
400 diameter; DeCarlo et al. (2004)), compared to the CU-AMS 50% transmission occurring at 900
401 nm. The reasons for the smaller transmission of the K-AMS are likely related to the PCI design
402 (Bahreini et al., 2008, 2009) or possibly an underperforming aerodynamic lens in K-AMS (Liu et
403 al., 2007). It was found that, in general, the RFs could be split between RFs generally below the
404 K-AMS size cut-off (RFs 1 – 9, 11, 15, and 19) and above the size cut-off (RFs 10, 12 – 14, 16 –
405 18, 20) (SI Figure 27). The slopes and R² greatly improves for the observations below the K-AMS
406 cut-off versus above (for slopes, 1.02 versus 0.84 and for R², 0.91 versus 0.82).

407 Finally, the ratios of the total AMS PM₁ masses measured by CU-AMS and K-AMS remain
408 nearly constant about one (within ±11%) for the entire campaign and show no trend with estimated
409 CE (for the standard vaporizer only) using the Middlebrook et al. (2012) algorithm (SI Figure 28).

410 Thus, when accounting for transmission effects, the two AMSs agree to within 10%, and the CU-
411 AMS agrees to within 20% with the other co-located aerosol mass concentration measurements
412 (filters and MC/IC) on the DC-8. This provides overall confidence in the calculated CE for the
413 standard vaporizer (Middlebrook et al., 2012), RIE, and transmission of PM₁ for the CU-AMS
414 measurements.

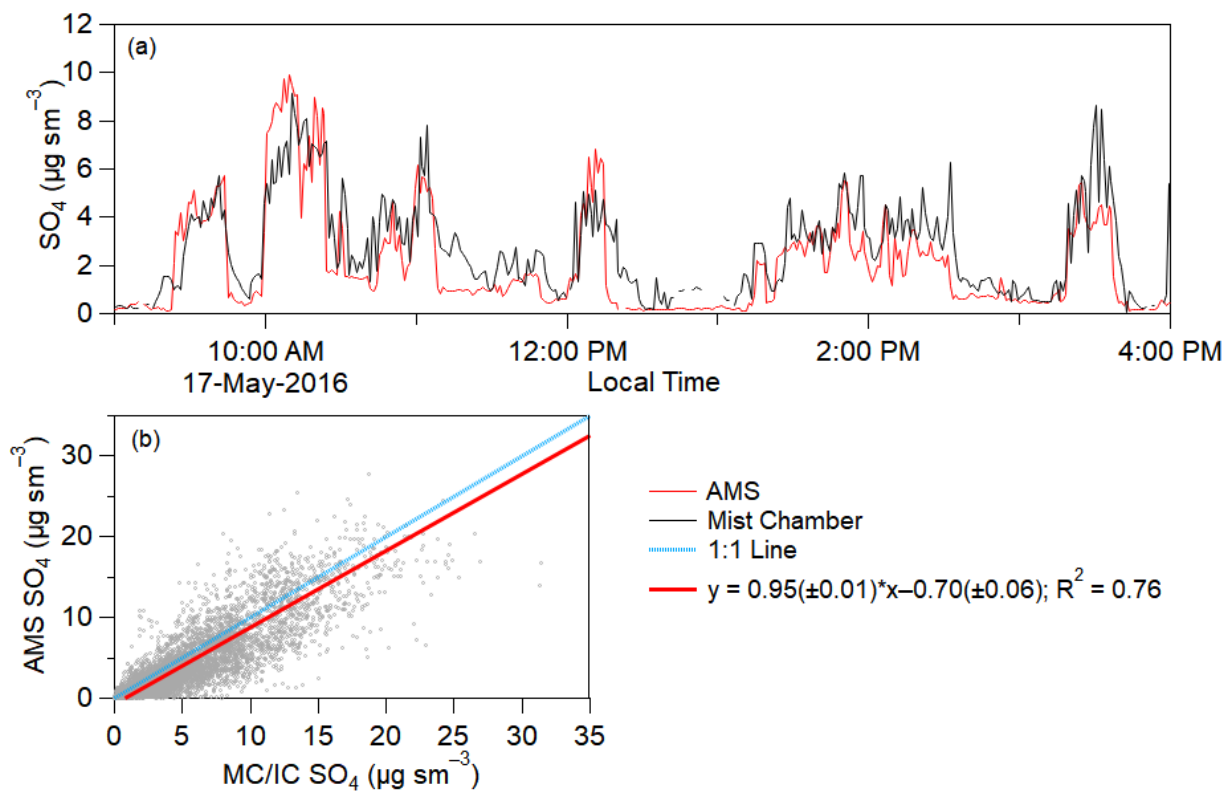
415 Besides directly comparing species mass, another well-established method to investigate
416 aerosol instrument quantification is to compare the measured PM₁ mass (CU-AMS plus BC from
417 SP2) versus the submicron extinction measured using methods described in Section 2.3.2
418 (nephelometer for scattering and absorption by PSAP) (e.g., DeCarlo et al., 2008). During KORUS-
419 AQ, the slope between mass and extinction is 6.00 m² g⁻¹ (SI Figure 29) with an R² of 0.87. The
420 high correlation and similar slope compared to prior comparisons (Hand and Malm, 2007; DeCarlo
421 et al., 2008; Dunlea et al., 2009; Shinozuka et al., 2009; Liu et al., 2017) indicates that the CU-
422 AMS was not substantially impacted by the aerosol transmission effects discussed above. Also,
423 the strong correlation (R² = 0.87) between the two instruments, which both have comparable, very
424 high time resolution, indicate that the CU-AMS did not experience any plume recovery artifacts
425 that were observed with the MC/IC or artifacts in measuring highly concentrated plumes.

426 Finally, we compare the PM₁ volume concentrations estimated from the LAS PM₁ versus
427 the CU-AMS plus SP2. For this comparison, we use the calibrated AMS transmission curve during
428 this campaign (SI Figure 30), which is consistent with those from recent studies (Knote et al.,
429 2011; Hu et al., 2017b), to correct for particle transmission differences between the instruments.
430 The LAS diameters were corrected by a factor of 1.115 from the PSL-calibrated values, to account
431 for the lower refractive index of ambient particles, similar to Liu et al. (2017). To estimate the
432 volume concentration from the combined AMS and BC measurements, we assume additive species

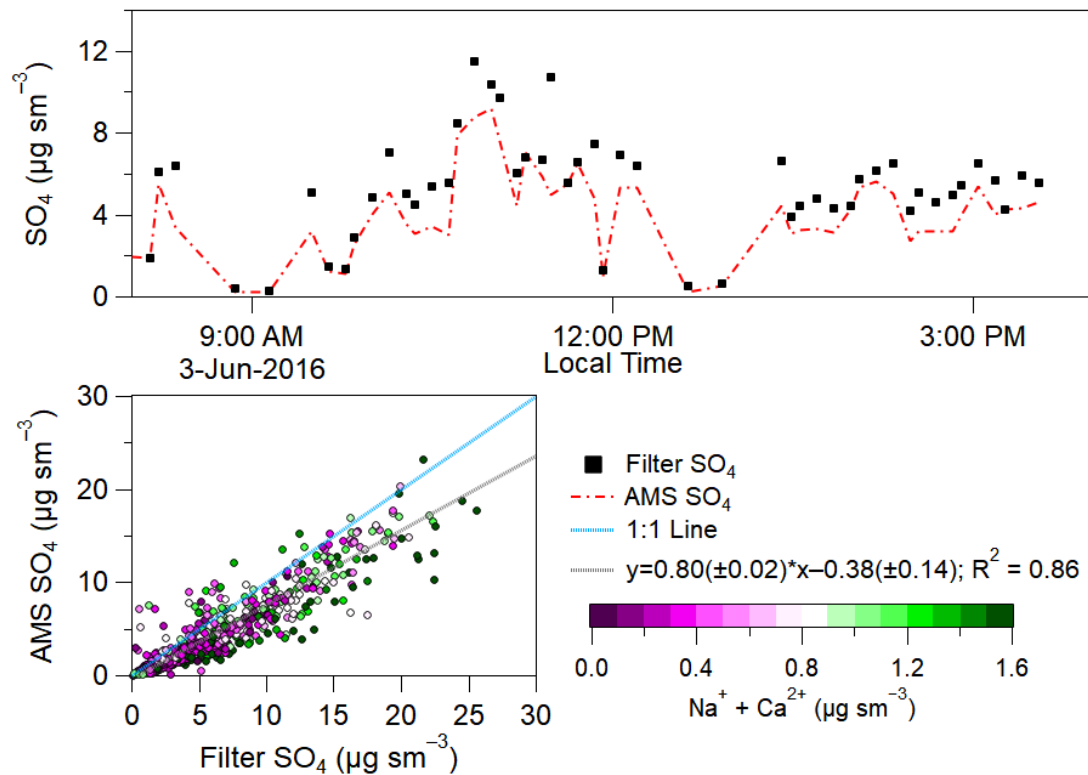
433 volumes (DeCarlo et al., 2004). Species densities of 1.78 g cm^{-3} for NH_4 , pNO_3 , and SO_4 (Lide,
434 1991; Salcedo et al., 2006), 1.52 g cm^{-3} for Chl (Lide, 1991; Salcedo et al., 2006), 1.77 g cm^{-3} for
435 BC (Park et al., 2004), and the OA density is estimated from the CU-AMS O/C and H/C ratios of
436 OA using the parameterization of Kuwata et al. (2012). The comparison between total PM_{10} volume
437 estimated from the CU-AMS plus BC vs. versus LAS shows a correlation (R^2) of 0.86. However,
438 the volume from AMS plus SP2 is higher (slope of 1.56) when comparing all of KORUS-AQ. We
439 hypothesize that this may be due to saturation of the LAS detector at high particle concentrations
440 that were frequently observed in this campaign (greater than $1800 \text{ particles cm}^{-3}$ or total CU-AMS
441 plus SP2 mass greater than $40 \mu\text{g sm}^{-3}$), as has been observed in prior comparisons (Liu et al.,
442 2017), or a change in the refractive index when OA becomes dominant at these high concentrations
443 (Moise et al., 2015). Different filters are tested and shown in SI Figure 30 and SI **Figure 31**, using
444 both values reported in literature and values that represent a stable ratio between LAS and
445 calculated CU-AMS plus SP2 volume. If we filter for data when there is less than $20 \mu\text{g sm}^{-3}$, the
446 slope drops to 1.00, showing agreement between within the combined uncertainties ($R^2 = 0.79$),
447 and providing strong evidence that LAS saturation at higher concentrations is the main reason for
448 the apparent disagreement when analyzing the entire campaign.

449 We further investigate (SI Figure 31) whether the slope could be due to LAS saturation or
450 a bias in RIE_{OA} , or in CE, vs. the values used in our analyses (Jimenez et al., 2016; Xu et al., 2018).
451 There is a slight increase in the ratio of AMS plus SP2 to LAS volumes versus OA/total CU-AMS
452 mass at high fractions of OA, although still within the combined measurement uncertainties. With
453 filtered data (less than $1600 \text{ particles cm}^{-3}$ or total CU-AMS mass less than $20 \mu\text{g sm}^{-3}$), the
454 volume ratios remain nearly flat, even at high $f(\text{OA})$. This confirms that LAS saturation is the most
455 likely cause for the differences. Finally, a recent study (Xu et al., 2018) has reported new laboratory

456 measurements of $RIE_{OA} = 1.6 \pm 0.5$, although these authors indicated that it was unclear whether
 457 this value was applicable to ambient particles, and the value of $RIE_{OA} = 1.4$ used in this study is
 458 well within their reported uncertainty. When using $RIE_{OA} = 1.6$ in our analysis (not shown) the
 459 slope for the entire dataset decrease by only 6% (1.56 to 1.47), indicating that RIE uncertainties
 460 cannot explain the bulk of the observed difference.
 461



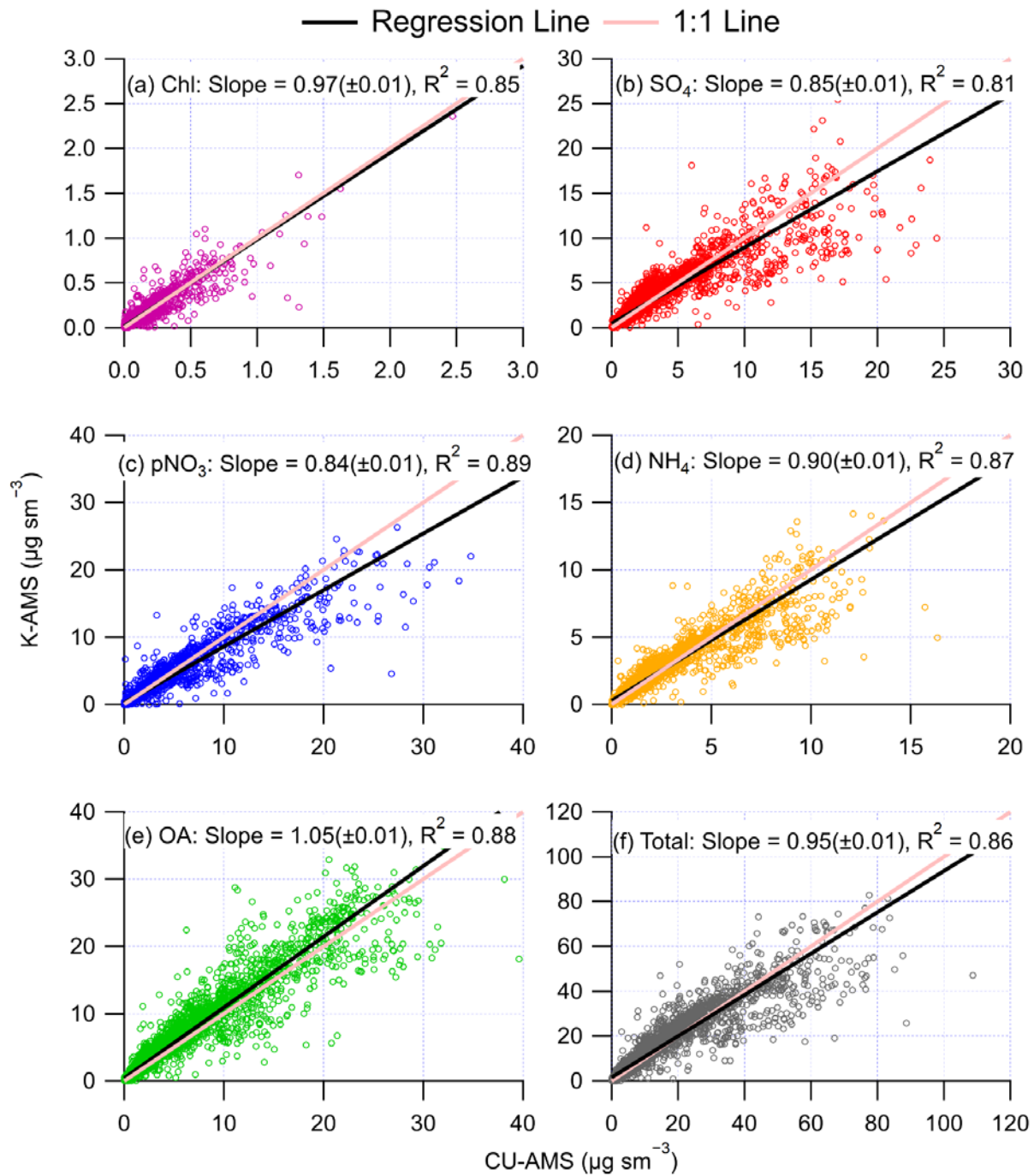
462
 463 **SI Figure 23.** (top) Time series of mist-chamber (dark red line) and CU-AMS (red line) SO_4 for
 464 one flight (RF17). (bottom) Scatter plot of CU-AMS SO_4 versus mist-chamber ion-chromatograph
 465 (MC/IC) SO_4 for entire KORUS-AQ campaign.



466

467 **SI Figure 24.** (top) Time series of filter (black squares) and CU-AMS (red line) SO₄ for one flight
 468 (RF17). The CU-AMS data has been averaged to the filter sampling time. (bottom) Scatter plot of
 469 CU-AMS SO₄ versus filter SO₄ for entire KORUS-AQ campaign. The points are colored by the
 470 total sodium (Na⁺) and calcium (Ca²⁺) measured by the filters, as indicators of sea salt and dust,
 471 respectively.

472

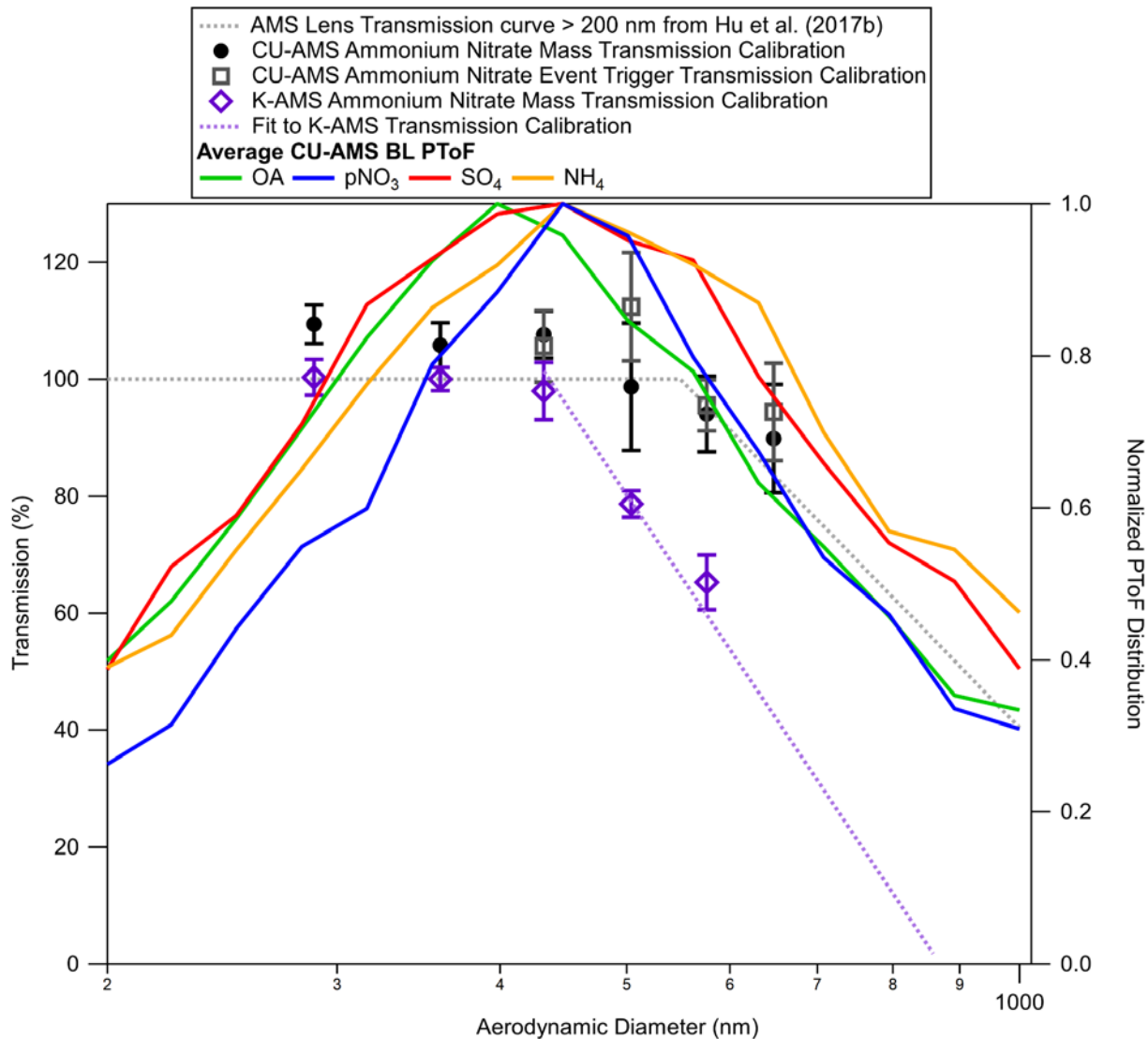


473

474 **SI Figure 25.** Scatter plot of not transmission corrected K-AMS versus CU-AMS mass
 475 concentrations for all of KORUS-AQ for (a) Chl, (b) SO_4 , (c) pNO_3 , (d) NH_4 , (e) OA, and (f) total
 476 AMS mass. The slopes and R^2 for all comparisons are shown in each scatter plot.

477

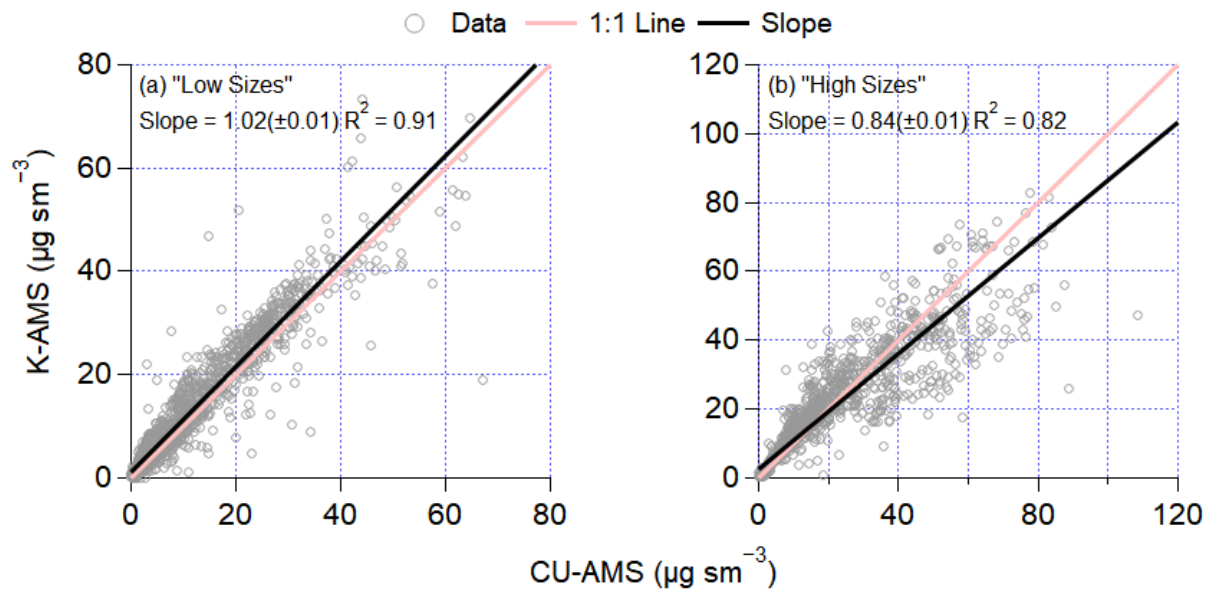
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479

480 **SI Figure 26.** (left axis) Transmission curve for CU-AMS (black circle and dark grey square) and
 481 K-AMS (purple diamond). The curve from literature (Knote et al., 2011; Hu et al., 2017b), which
 482 describes the CU-AMS, is shown (grey dotted line). The fit for K-AMS transmission is shown
 483 with the purple dotted line. (right axis) Average mass distributions for OA (green), pNO₃ (blue),
 484 SO₄ (red), and NH₄ (orange) measured by CU-AMS in the boundary layer during KORUS-AQ.
 485 Note that some of the apparent signal at larger particle sizes is caused by the limited time response
 486 of the AMS detection system.

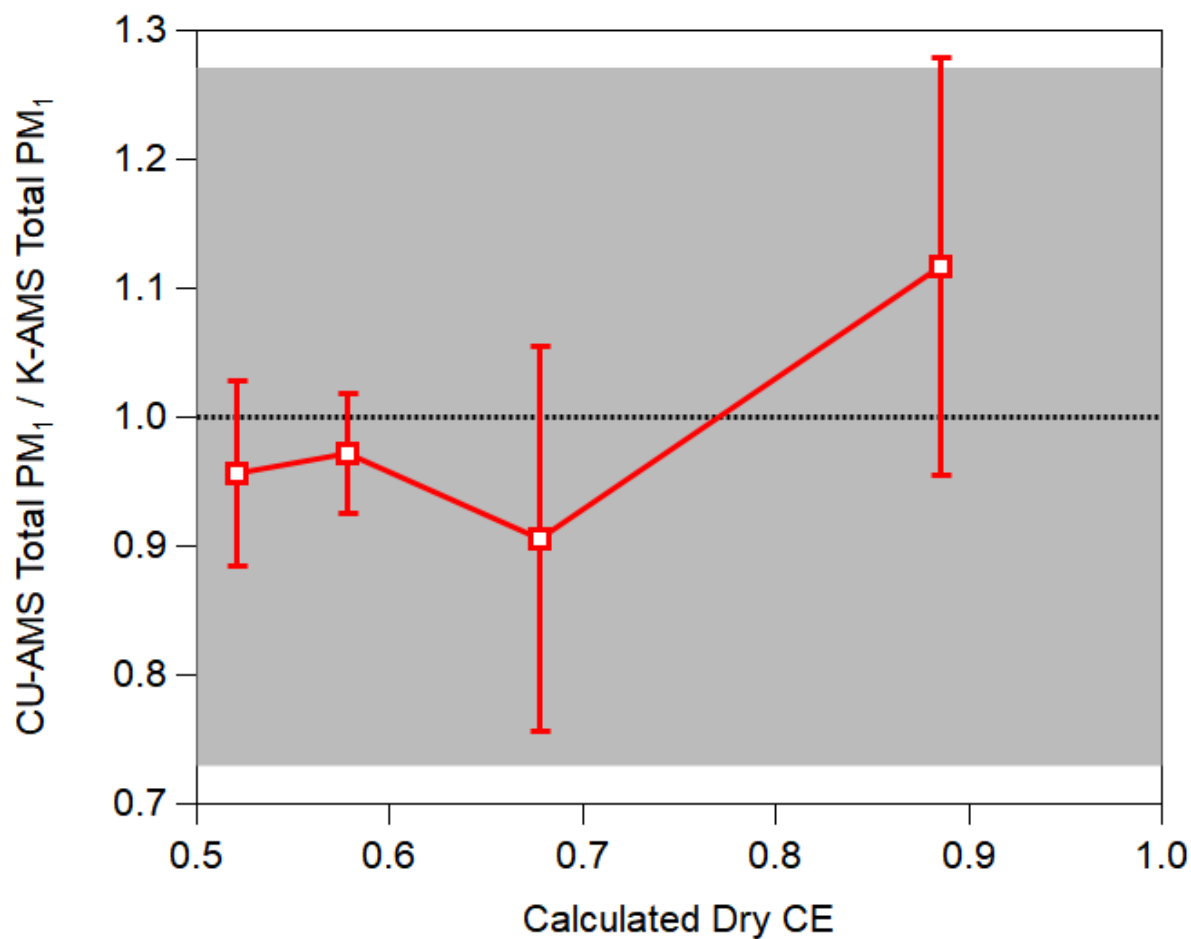
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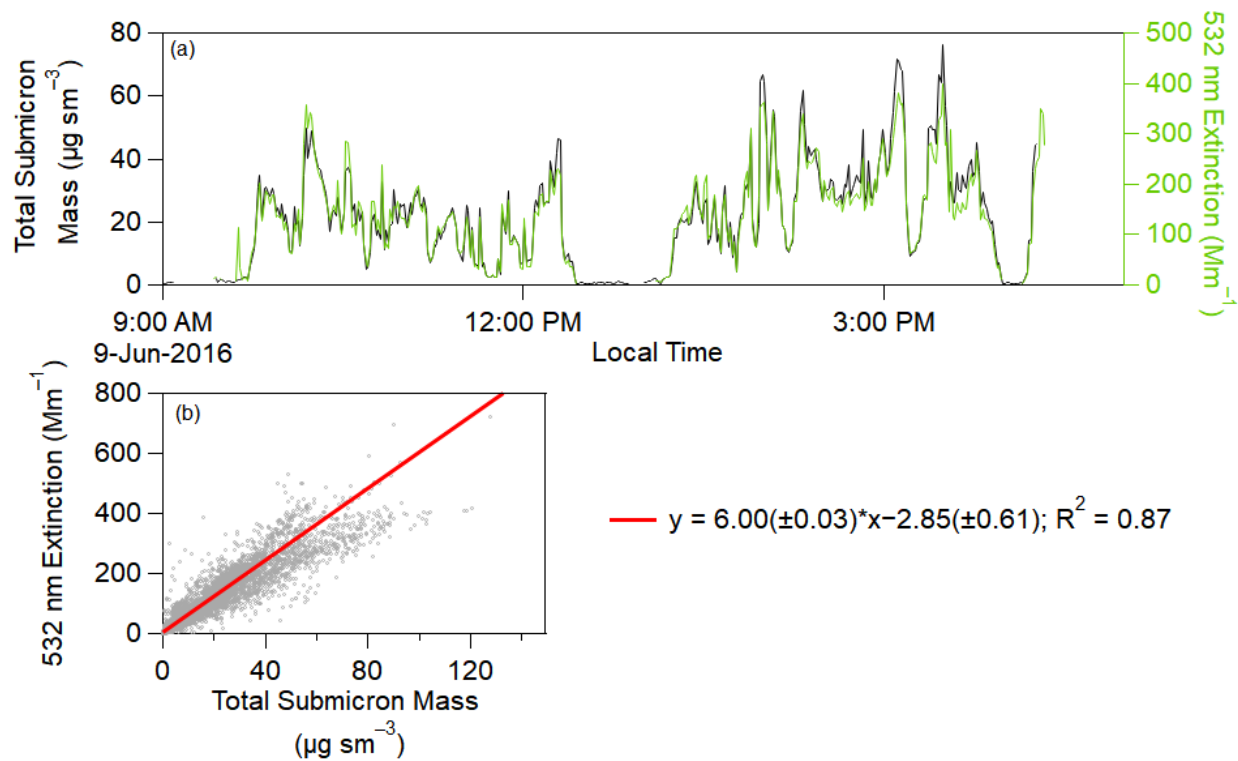
489 **SI Figure 27.** Scatter plot of K-AMS versus CU-AMS total mass concentrations (a) RFs 1 – 9, 11,
 490 15, and 19 and (b) RFs 10, 12 – 14, 16 – 18, and 20. These are flights where the average sizes were
 491 found below (a) and above (b) the K-AMS size cut-off (SI Figure 26). The slopes and R^2 for all
 492 comparisons are shown in each scatter plot.

493



494

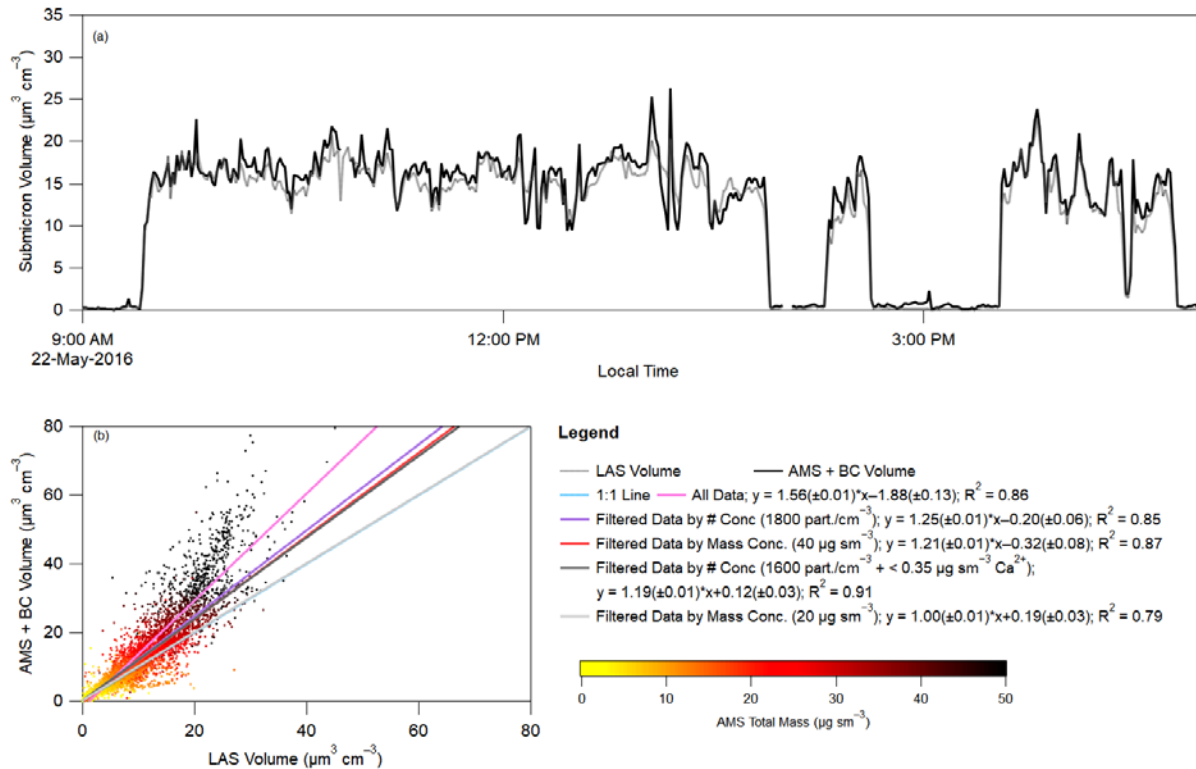
495 **SI Figure 28.** Binned total PM₁ AMS mass ratios, normalized by the average ratio, versus the
 496 calculated CE used for the CU-AMS measurements. The error bars are the standard error about
 497 the mean, and the shaded grey area is the combined uncertainty of the two AMS measurements
 498 ($\pm 27\%$). The data is only for flights where the PM₁ sizes were typically below the K-AMS size
 499 cut-off (RFs 1 – 9, 11, 15, and 19).



500

501 **SI Figure 29.** (top) Time series of total submicron mass (black, left axis) and 532 nm extinction
 502 (green, right axis) for one flight (RF19). (bottom) Scatter plot of 532 nm extinction versus total
 503 submicron mass (black carbon + CU-AMS species) for the entire KORUS-AQ campaign.

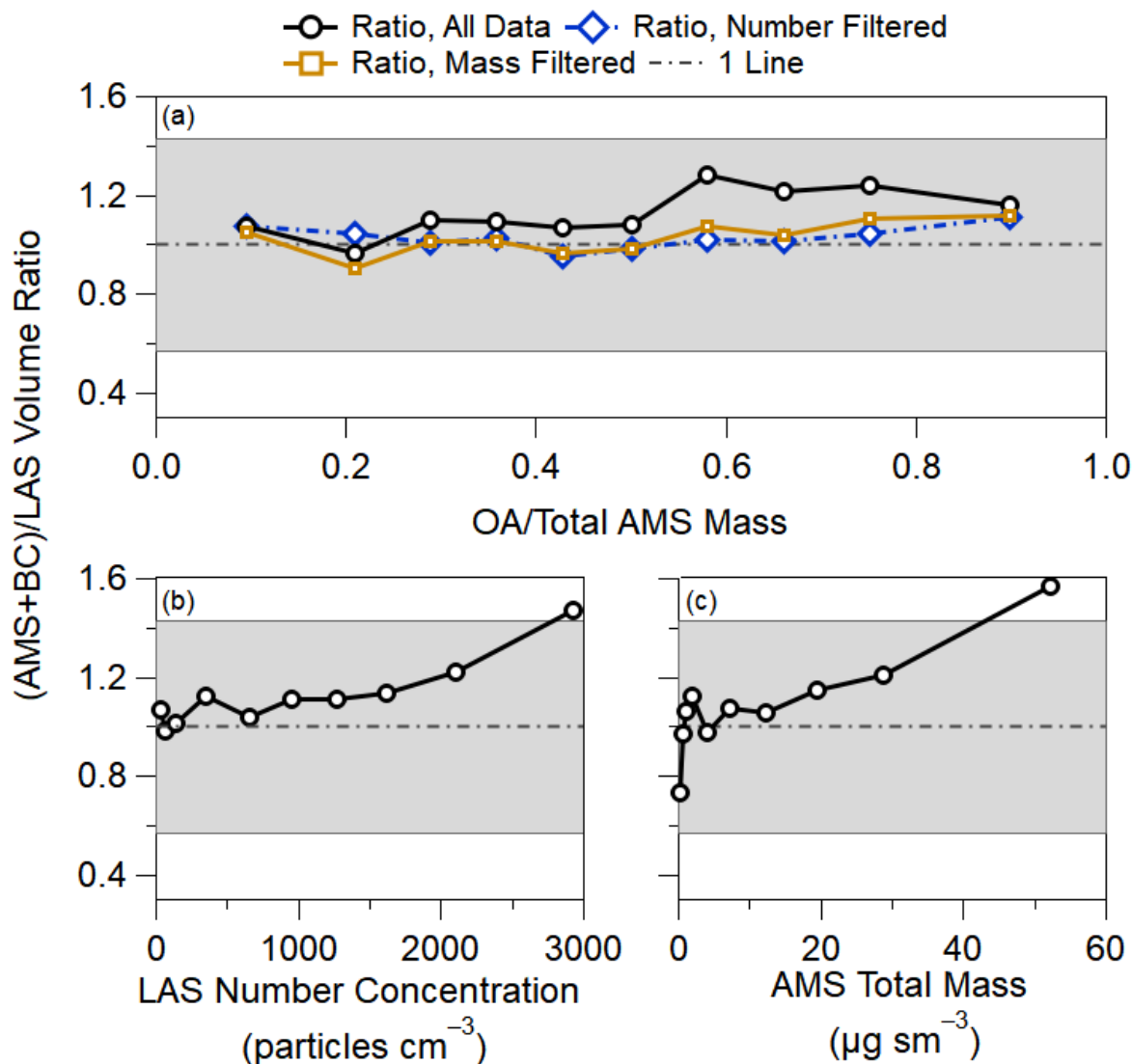
504



505

506 **SI Figure 30.** (top) Time series of total submicron volume from LAS (grey dashed line) and CU-
 507 AMS plus black carbon (black) for one flight (RF11). (bottom) Scatter plot of total submicron
 508 volume (black carbon + CU-AMS species) versus LAS volume for entire KORUS-AQ campaign.
 509 The data is colored by total CU-AMS mass. Pink line is a fit to all data, the purple line is a fit to
 510 data where the particle number concentration is less than $1800 \text{ particles cm}^{-3}$. The red line is a fit
 511 to the data where the CU-AMS plus SP2 total mass is less than $40 \mu\text{g sm}^{-3}$. The black line is a fit
 512 to the data where the particle number concentration is less than $1600 \text{ particles cm}^{-3}$ and Ca^{2+}
 513 concentration is less than $0.35 \mu\text{g sm}^{-3}$. Finally, the grey line is a fit to the data where the CU-
 514 AMS plus BC total mass is less than $20 \mu\text{g sm}^{-3}$.

515

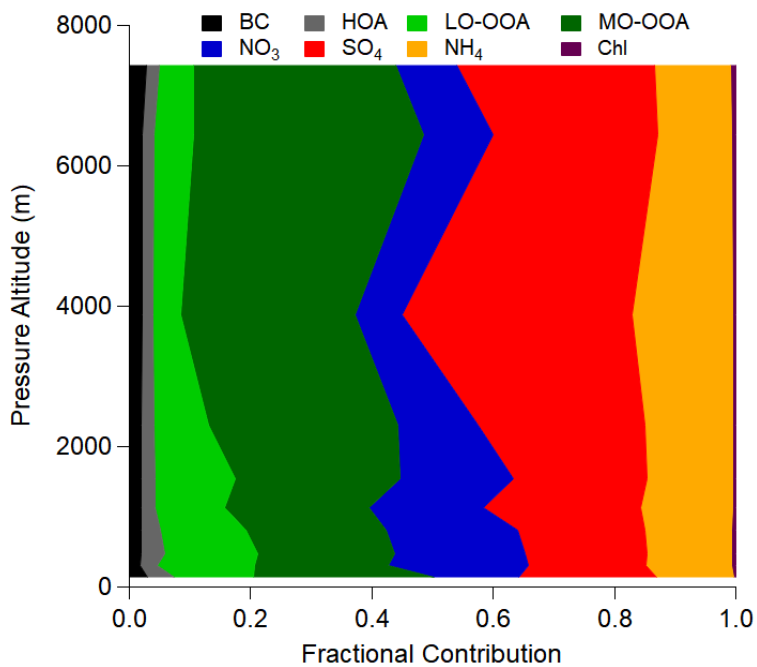


516

517 **SI Figure 31.** (a) Binned volume ratio (CU-AMS plus black carbon volume/LAS Volume) versus
 518 fraction of organic aerosol (OA) to total CU-AMS mass. (b) Binned volume ratio versus LAS
 519 particle number concentration. (c) Binned volume ratio versus CU-AMS total mass. In all figures,
 520 the black data is for all data whereas the blue data is for the volume ratio where the particle number
 521 concentration is less than 1600 particles cm^{-3} and the orange data is for the volume ratio where the
 522 CU-AMS total mass concentration is less than 20 $\mu\text{g sm}^{-3}$. Also, the shaded area represents the
 523 combined uncertainty in both measurements (Bahreini et al., 2009).

524

525 **SI 9. Fractional PM₁ Contribution to Vertical Profile**



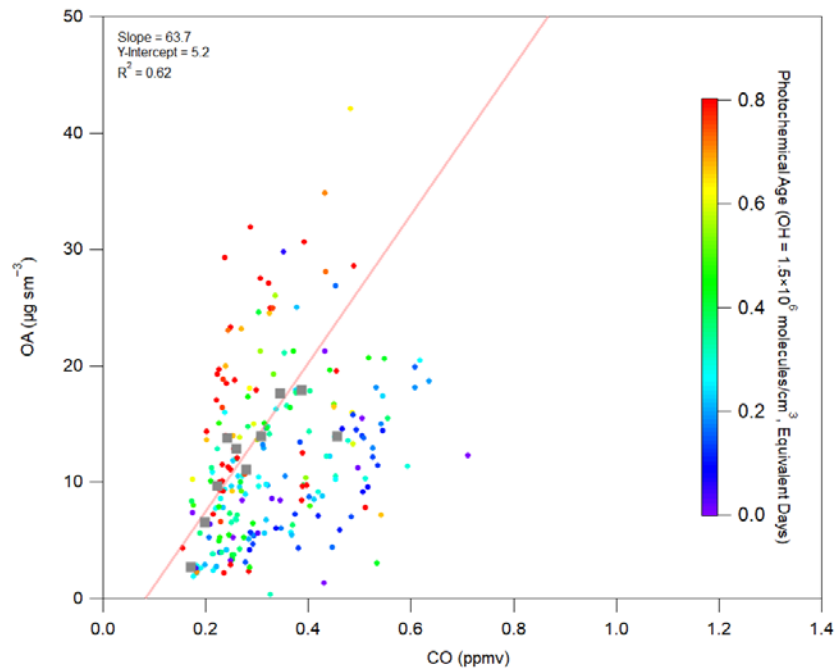
526

527 **SI Figure 32.** Fractional contribution of PM₁ contribution vertical profile for all of KORUS-AQ.

528

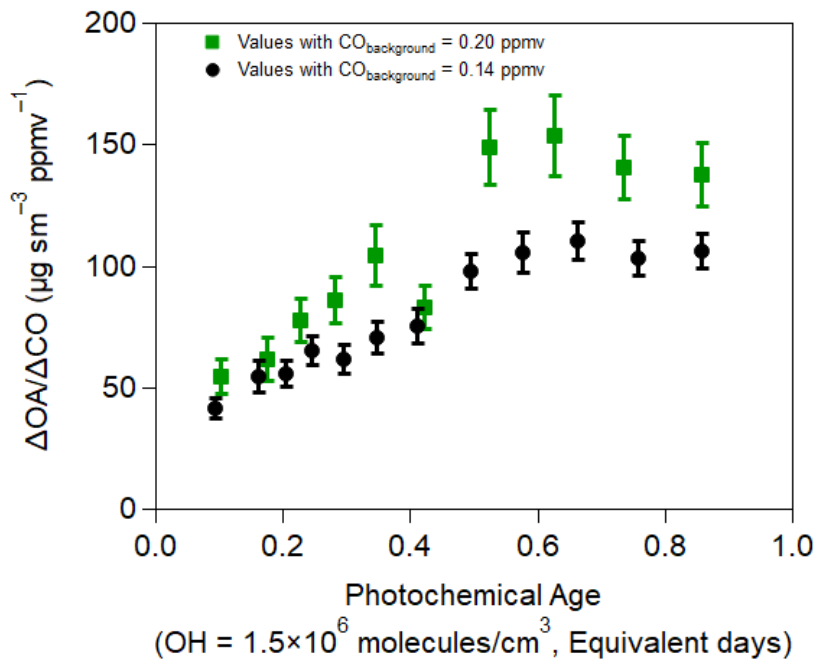
529

530 **SI 10. Observed Aerosol Production over Seoul, South Korea**



531
532 **SI Figure 33.** Scatter plot of OA versus CO, observed over Seoul, during KORUS-AQ. The points
533 are colored by the NO_x photochemical clock. The fit is for the decile binned data.

534

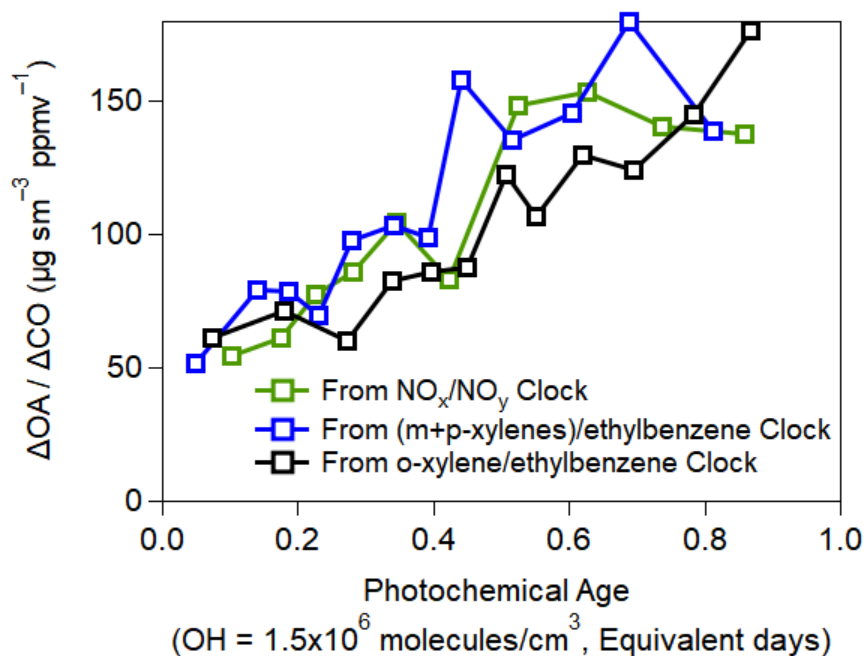


535
536 **SI Figure 34.** Comparison of $\Delta\text{OA}/\Delta\text{CO}$ observed over Seoul with different CO backgrounds.

537 **SI Table 4.** Compilation of slopes used to convert from $\Delta\text{OA}/\Delta\text{CO}$ to $\Delta\text{OA}/\Delta\text{CO}_2$ used in this
 538 study.

<i>Location</i>	<i>Slope (ppmv CO/ppmv CO₂)</i>	<i>Study</i>
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)

539



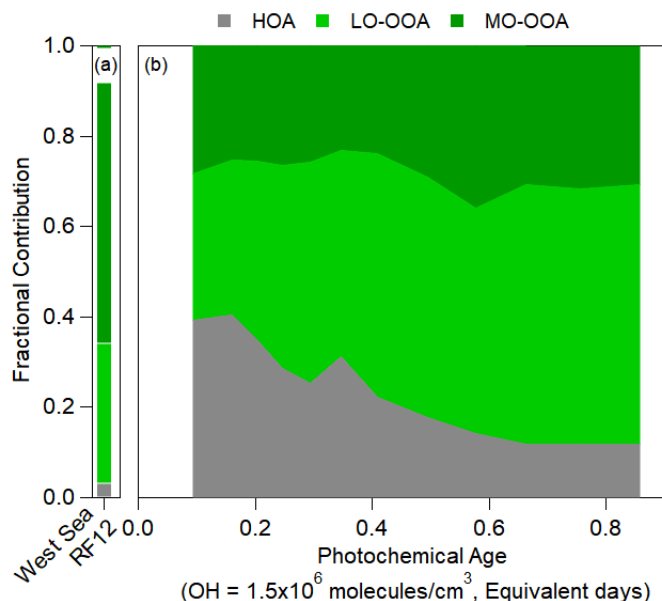
540

541 **SI Figure 35.** Same as **Figure 4(a)**, but comparing results using three different photochemical
 542 clocks (SI Figure 21).

543

544

545 **SI 11. Oxidation of OA**



546

547 **SI Figure 36.** Same as **Figure 6b**, but speciated for MO-OOA, LO-OOA, and HOA. (a) is over
 548 the West Sea (RF12) and (b) is over Seoul.

549 First, we briefly discuss how the AMS OA source tracers typically used to investigate OA
 550 chemistry evolved over Seoul (SI Figure 37). During KORUS-AQ, there was no appreciable
 551 influence from isoprene production of IEPOX-SOA (Hu et al., 2015), as the ion indicative of
 552 isoprene IEPOX-SOA ($C_3H_6O^+$) remained at background values typical of air without isoprene
 553 SOA influence.

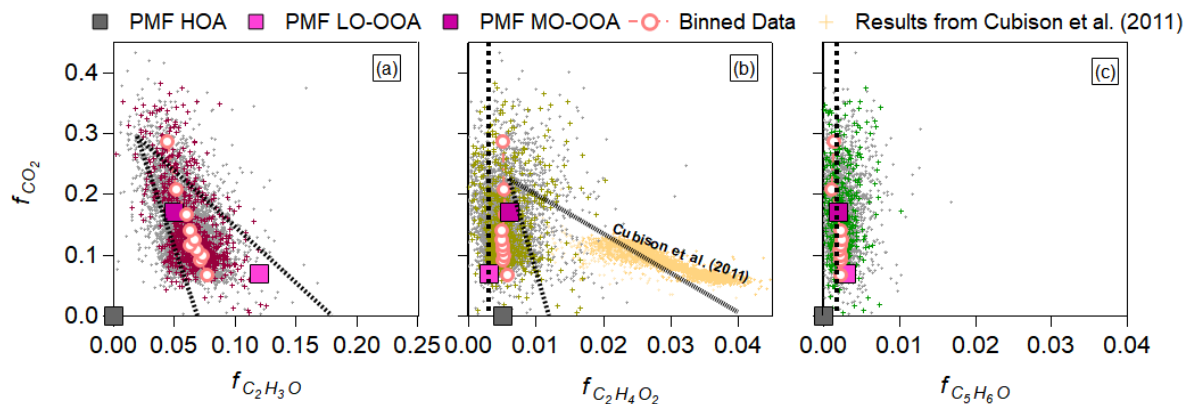
554 Similarly, biomass burning OA (BBOA) appeared to be present but dilute in its
 555 contribution to OA. Most of the OA had $f_{C_2H_4O_2}$, an ion indicative of biomass burning and
 556 levoglucosan (Schneider et al., 2006; Aiken et al., 2010), below 0.1 over Seoul, and the PMF
 557 factors fall near the limit of detection for BBOA (Cubison et al., 2011) and lower than the values
 558 that typically indicate ambient and laboratory BBOA emissions at various stages of chemical
 559 evolution (Cubison et al., 2011; Ortega et al., 2013). We speculate that the limited BBOA is highly

560 mixed into the OA from the numerous, small agricultural fires that were observed during the
561 campaign and have been observed during this time period, during other years, in South Korea
562 (Kang et al., 2006). However, the amount of fresher BBOA was not high enough, nor as strong of
563 a feature as observed in prior studies (Aiken et al., 2010; DeCarlo et al., 2010; Cubison et al., 2011;
564 Hu et al., 2016), to reliably resolve a separate BBOA PMF factor. As shown in SI Figure 11,
565 typical gas-phase biomass burning tracers (CO, NO_x, acetonitrile, HCN, and black carbon) do not
566 show a consistent strong correlation with any of the PMF factors, further suggesting that BBOA
567 is not a major contributor, and any BBOA present is highly mixed with HOA and the oxidized
568 OA. Consistent with our results, Kim et al. (2017) did not resolve a BBOA factor from a ground
569 site in Seoul during the KORUS campaign.

570 Similar to other studies over urban areas or for chamber studies oxidizing urban VOCs
571 (e.g., benzene, xylenes, etc.) (Ng et al., 2010; Freney et al., 2014a; Ortega et al., 2016), marked
572 chemical evolution was observed as tracked by the C₂H₃O⁺ and CO₂⁺ ions. The evolution of these
573 two ions, as a fraction of total OA, fall in the same space as has been observed in these prior
574 studies, indicating consistent photochemical evolution of SOA over urban locations.

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

580



581
 582 **SI Figure 37.** 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 $f_{C_5H_6O}$.
 583 Points highlighted in color refer to observations over Seoul, South Korea, during KORUS-AQ. In
 584 (a), the triangle is from Ng et al. (2010); in (b), the triangle is from Cubison et al. (2011), and the
 585 vertical line is the typical “background” values for $f_{C_2H_4O_2}$ from Cubison et al. (2011); and, in (c),
 586 the vertical line is the typical “background” values for $f_{C_5H_6O}$ from Hu et al. (2015). The PMF
 587 results for each triangle plot are shown in squares, where grey is HOA, light pink is LO-OOA, and
 588 dark pink is MO-OOA. The light orange dots in (b) are the observations from ARCTAS forest
 589 fires (Cubison et al., 2011), as an example for data strongly impacted by biomass burning. The
 590 binned values for each comparison are shown in light red crosses.

591

592

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