

1 **Supporting Information**

2 **Intercomparison of an Aerosol Chemical Speciation** 3 **Monitor (ACSM) with Ambient Fine Aerosol Measurements** 4 **in Downtown Atlanta, Georgia**

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19 20 **A. ACSM Data Analysis**

21 The ACSM has non-unit collection efficiency (CE) for sampled particles due to (i)
22 transmission losses in the aerodynamic lenses, (ii) broadening of the particle beam; and (iii)
23 particle bounce losses during impaction on the vaporizer (Huffman et al., 2005). These CE
24 constraints are identical for both the ACSM and AMS instruments. Previous measurements
25 have shown that an AMS CE of 0.5 reproduces ambient species mass concentrations to within
26 25% or better of measurements of collocated instruments (Canagaratna et al., 2007) and within
27 81-90% of fine aerosol volume or PILS measurements (Middlebrook et al., 2012). A

1 composition dependent CE parameterization account for higher CEs that are observed when the
2 sampled ambient aerosol is acidic, has a high nitrate content, or is sampled under very humid
3 conditions (Middlebrook et al., 2012). In this manuscript we use CE of 0.5 that was examined
4 against composition dependent collection efficiency ($CE_{estimate}$) based on Middlebrook et al.
5 (2012) parameterizations as follows:

6 a) Effect of high aerosol acidity:

$$7 \quad \frac{NH_4^+_{measured}}{NH_4^+_{predicted}} = \frac{NH_4/18}{(SO_4/96) \times 2 + (NO_3/62) + (Chl/35.5)}$$

$$8 \quad CE_{est,acidic} = 1.0 - 0.73 \times \left(NH_4^+_{meas} / NH_4^+_{predict} \right)$$

$$9 \quad CE_{dry} = \max(0.5, CE_{est,acidic})$$

10 b) Effect of high ammonium nitrate fraction (ANMF):

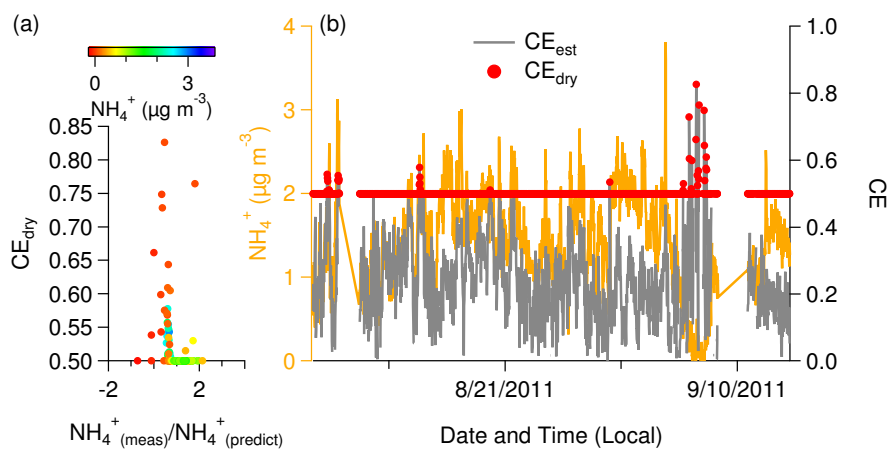
$$11 \quad ANMF = \frac{(80/62) \times NO_3}{(NH_4 + SO_4 + NO_3 + Chl + Org)}$$

$$12 \quad CE_{est,ANMF} = 0.0833 + 0.9167 \times ANMF$$

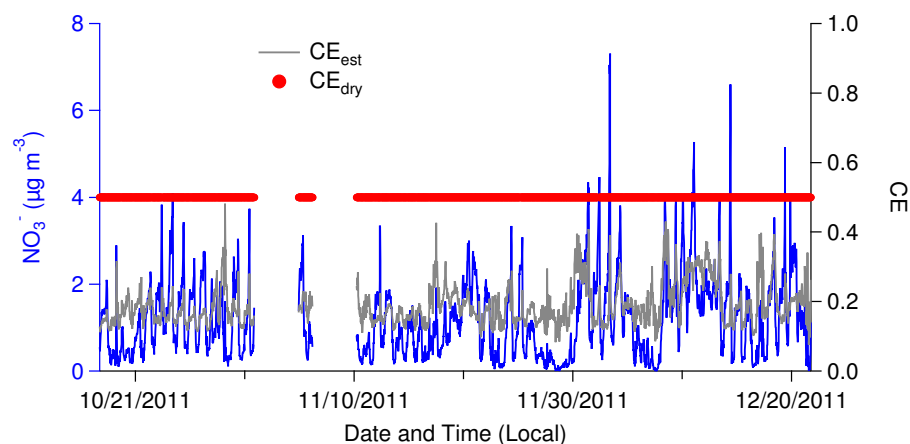
$$13 \quad CE_{dry} = \max(0.5, CE_{est,ANMF})$$

14 Observation of summer 2011 dataset suggests that only a few sporadic events were
15 influenced by high aerosol acidity (Figure S1) which are attributed to low ammonium loadings.
16 In addition, in fall 2011 where nitrate concentration was enhanced compared to summer
17 measurements, suggests that CE was not affected by high ammonium nitrate fraction (Figure
18 S2). Therefore, CE of 0.5 was used in analysis of all species for all dataset.

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1
2 **Figure S1.** (a) CE_{dry} against NH₄⁺_(meas)/NH₄⁺_(predict) color coded by concentration of ammonium,
3 and (b) relationship between ammonium loading and CE_{dry} in the summer 2011.



4
5 **Figure S2.** CE estimation based on nitrate loading during fall 2011.

6 Relative ionization efficiency (RIE) values were previously determined from laboratory
7 calibrations with nitrate, ammonium, sulfate, and organics particles (Alfarra et al., 2004). Since
8 the experiments for RIEs were conducted using AMS instrument, the values for ACSM might
9 be different. It was found that for RIE of ammonium (RIE_{NH4}) that was 3.5, the value obtained
10 from ACSM calibrations was between 5 and 6. RIE of sulfate might be estimated by fitting
11 measured sulfate and predicted sulfate values. Measured sulfate (SO_{4,meas}) is sulfate that is
12 measured by the ACSM, while predicted sulfate (SO_{4,pred}) is the estimated value of sulfate from
13 ion balance approach. SO_{4,pred} is derived from NH_{4,pred} equation (N. L. Ng, personal
14 communication, 2012) as follows:

$$NH_{4,pred} = 2 \left(\frac{MW_{NH_4}}{MW_{SO_4}} \right) SO_{4,meas} + \left(\frac{MW_{NH_4}}{MW_{NO_3}} \right) NO_{3,meas} + \left(\frac{MW_{NH_4}}{MW_{chl}} \right) chl_{meas} \quad (1)$$

$$SO_{4,pred} = \frac{NH_{4,meas} - \left(\frac{MW_{NH_4}}{MW_{NO_3}}\right)NO_{3,meas} - \left(\frac{MW_{NH_4}}{MW_{chl}}\right)chl_{meas}}{2\left(\frac{MW_{NH_4}}{MW_{SO_4}}\right)} \quad (2)$$

2 Previous value of RIE_{SO_4} 1.5 is then multiplied by slope obtained from fitting $SO_{4,pred}$
 3 versus $SO_{4,meas}$ and used as the RIE_{SO_4} value of this study.

4 Maintenance issues included periods of complete instrument shutdown for calibration
 5 and start-up following calibration. During such start-up periods, vaporizer temperature,
 6 naphthalene signal, which serves as internal calibration, and airbeam signal showed instability,
 7 indicating that the ACSM was adjusting to operating conditions. An irregular naphthalene
 8 signal during continuous operation was also indicative of a problem, probably temporary
 9 clogging of the 1- μ m pinhole of the naphthalene bath. Another operational issue encountered
 10 was a temporary disturbance in the electronic baseline (i.e., electronic zero value). Shifts in the
 11 electronic baseline might have occurred when there were short power outages at the JST site.
 12 Any of the described above were immediately reflected on the diagnostic panel, allowing
 13 precise determination of the sampling periods to be excluded from the data analysis.

1 **B. JST Site Measurements**

2 **B.1 Integrated particle measurements**

3 Integrated PM_{2.5} sampling and analysis are listed in Table 2 and described briefly below.
4 24-h integrated PM_{2.5} samples were collected using particle composition monitor (PCM) built
5 by ARA that was specifically designed to minimize and/or account for potential artefacts and
6 reactive gas interferences (Edgerton et al., 2005). PCM is a multichannel, sequential filter-based
7 sampler that each consists of a teflon-coated cyclone (URG) with 10-mm cut size as the inlet,
8 one or more denuder to remove gas interferences, Well Impactor Ninety-Six (WINS) with cut
9 size of 2.5-mm, and filter media. Flow through each PCM's channel was maintained at 16.7 L
10 min⁻¹ using mass flow controllers (MFCs). There were three PCM channels that were sampling
11 simultaneously. PCM1 channel was used for routine quantification of PM_{2.5} mass, major ions,
12 volatile nitrate (NO₃⁻), volatile ammonium (NH₄⁺), and trace elements. Series of denuders used
13 in PCM1 channel were sodium bicarbonate (NaHCO₃) followed by citric acid (C₆H₆O₇) that
14 remove HNO₃, SO₂, and NH₃. Filter media used in PCM1 are three stacks of filters comprising
15 of a 47-mm diameter Teflon filter, 47-mm diameter Nylon filter, and lastly 47-mm diameter
16 C₆H₆O₇-coated cellulose filter. PCM2 channel was used to quantify sulfate (SO₄²⁻), total NO₃⁻,
17 and NH₄⁺ but was discontinued in 2000 due to measurement redundancy to that of PCM1
18 channel. PCM3 channel was used for quantification of organic carbon (OC) and black carbon
19 (BC). A 100-mm long by 30-mm ID carbon honeycomb denuder (MAST Carbon, Ltd.,
20 Guildford, U.K.) was used to remove semi-volatile gaseous organics in PCM3. Filter media
21 used for PCM3 channel were two pre-baked 37-mm diameter quartz filters stacked together.
22 OC on the back filter was considered as volatilization loss from OC on the front filter, therefore,
23 the resulting value represent a lower limit for the actual OC concentrations.

24 Component mass loading from each filter was corrected by blank filter using SEARCH
25 network-wide average loadings from field blanks, then the corrected loading was normalized
26 by sampling volume (Edgerton et al., 2005). Blank correction had been shown to significantly
27 influence the overall mass and OC loadings. Moreover, species that were at or below instrument
28 detection limits, such as non-volatile NO₃⁻, black carbon (BC), and major metal oxides, were
29 found to have poor precisions for overall SEARCH measurements (Edgerton et al., 2005).

30 Mass is determined using best estimate (BE) method that attempts to calculate particle
31 compositions based on their actual loading in the atmosphere. PM_{2.5} mass is calculated from
32 blank-corrected mass from FRM, PCM1 or TEOM and adding volatile NO₃⁻ from PCM1 Nylon,

1 volatile NH_4^+ , and volatile OM from PCM back filter. NO_3^- is calculated as total NO_3^- from
2 PCM1 Teflon filter + PCM1 Nylon filter. SO_4^{2-} calculation for BE method is identical to FRM
3 method. NH_4^+ is calculated as total NH_4^+ that includes non-volatile NH_4^+ from PCM1 Teflon
4 and volatile NH_4^+ that is estimated as 0.29 times the volatile NO_3^- . OC is calculated as the sum
5 of front and back filters from PCM3. Since the back filter is assumed as volatilization product
6 of the front filter and only 10% of them are analyzed, it is estimated as a quarterly ratio of OC
7 from the back filter to OC from the front filter. These result in a formula for OC:

$$8 \quad OC = OC_{front}(1 + R_q) \quad (1)$$

$$9 \quad OM = OC \times 1.8 \quad (2)$$

10 where R_q is the estimated average ratio of volatile OC for quarter q . Particle
11 compositions resulted from BE method estimation are used in this study as it represents the
12 actual atmospheric loadings.

13 **B.2 Continuous particle measurements**

14 Details of continuous $\text{PM}_{2.5}$ sampling and analysis are provided in Edgerton et al. (2006)
15 and listed in Table 2. Briefly, $\text{PM}_{2.5}$ mass is measured continuously using an R & P Model 1400
16 a/b TEOM operated at 30 °C to reduce losses of semivolatile compounds and main flow of 3 L
17 min^{-1} . Sample air is pulled through PM_{10} inlet followed by $\text{PM}_{2.5}$ cyclone and goes inside the
18 trailer where a multitube Nafion drier (Perma Pure) is installed to dry the sample.

19 SO_4^{2-} is measured continuously using a modified Harvard School of Public Health
20 (HSPH). The method utilizes a stainless steel tube (300-mm section of 316 stainless steel)
21 heated to >850 °C in a Lindberg/Blue M horizontal tube furnace to reduce particulate sulfate to
22 gaseous sulfur dioxide (SO_2) that is detected by a Thermo-Environmental Instrument (TEI)
23 Model 43s or 43ctl high-sensitivity, pulsed ultraviolet fluorescence SO_2 analyzer. Sample air is
24 drawn through $\text{PM}_{2.5}$ cyclone (BGI) followed by two 30 mm of outer diameter (OD), 254 mm
25 long sodium carbonate and citric acid coated annular denuders (URG) and a 30 mmOD, 100
26 mm long activated carbon honeycomb denuder (Novacarb, Mast Carbon, Ltd.) that remove SO_2 ,
27 reduced sulfur gases, nitrogen oxides (NO_x) and volatile organic compounds (VOCs). Baseline
28 of the analyzer is zeroed every 90 min by diverting sample air through an inline filter upstream
29 the heated tube for 10 min to correct baseline drift.

30 NH_4^+ and NO_3^- were measured using a three-channel continuous differencing
31 approached developed by ARA (Edgerton et al., 2006). Sample air is coming from the same

1 inlet and denuders system as SO_4^{2-} and the denuded sample is divided into three analytical
2 channels. Channel 1 (CH1) provides instrument dark current and residual gas-phase NO_y that
3 represents baseline signal that will be used for the downstream analyzer. Channel 2 (CH2)
4 produces baseline NO_y signal and NO signal converted from particulate nitrogen species
5 assuming nitrate is the only species in the signal. Channel 3 (CH3) oxidizes NH_4^+ to NO and
6 NO_2 , and reduces NO_3^- , NO_2 , and residual NO_y to NO. TEI Model 42s or 42ctl NO-nitrogen
7 oxide (NO_x) analyzer is installed downstream of the three-channel converters to measure NO
8 from each converter by NO-ozone chemiluminescence method. Ammonium and nitrate are then
9 determined as CH3-CH2 and CH2-CH1, respectively. A caveat of this approach is other
10 particulate nitrogen compounds can be measured as ammonium and nitrate species as long as
11 they are convertible to NO (Edgerton et al., 2006).

12 Total carbon (TC) is measured using the Sunset OC/EC analyzer. Sample air is drawn
13 through a PM_{10} inlet at flow rate of 16.7 L min^{-1} followed by a $\text{PM}_{2.5}$ cyclone. Aerosol is first
14 collected on one of two metal plate impactors with cut size of 0.14 mm aerodynamic diameter
15 for 60-min period, and then sample is diverted to the second impactor while the first impactor
16 is heated. Particulate carbon is converted to CO_2 through two temperature plateaus, i.e., $275 \text{ }^\circ\text{C}$
17 and $750 \text{ }^\circ\text{C}$, and then detected by non-dispersive infrared absorption (NDIR). The instrument
18 is calibrated using CO_2 in zero air and is zeroed with CO_2 -free air ($<5 \text{ ppm}$). TC is defined as
19 the net carbon produced at the last temperature plateau ($750 \text{ }^\circ\text{C}$).

20 Black carbon (BC) or elemental carbon (EC) is measured using a Magee Scientific
21 Model AE-16 single-beam aethalometer that measures EC based on attenuation of light at a
22 wavelength of 880 nm. Sample air is pulled through a $\text{PM}_{2.5}$ cyclone at flow rate of $5\text{-}6 \text{ L min}^{-1}$
23 followed by a Nafion drier and a quartz tape filter. The instrument is zeroed with an in-line
24 ball valve and absolute filter for 15 min every day. The ambient concentration of EC is estimated
25 based on the rate of attenuation change, sample flow rate, and the default bulk absorption
26 coefficient from the manufacturer ($16.6 \text{ m}^2 \text{ g}^{-1}$).

27 Component mass concentrations from the continuous analyzers (hereafter referred as
28 Level1 data) were then adjusted to match the filter-based data since the continuous analyzers
29 had been shown to drift over time. The resulting filter-adjusted continuous data (hereafter
30 referred as Level_2 data) had been shown to agree within 1:1 line with the filter-based
31 measurements (Edgerton et al., 2006). With respect to carbon measurements, OC is calculated

- 1 as the difference between filter-adjusted TC and filter-adjusted EC, and OM is estimated
- 2 according to Eq. 2.

1 **C. Results of ACSM and collocated measurements at JST site**

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3 **Table S1.** Chemical characteristics of ambient aerosol mass and constituents at the JST site
4 measured by JST site instruments presented as average concentration \pm 1 standard deviation in
5 $\mu\text{g m}^{-3}$.

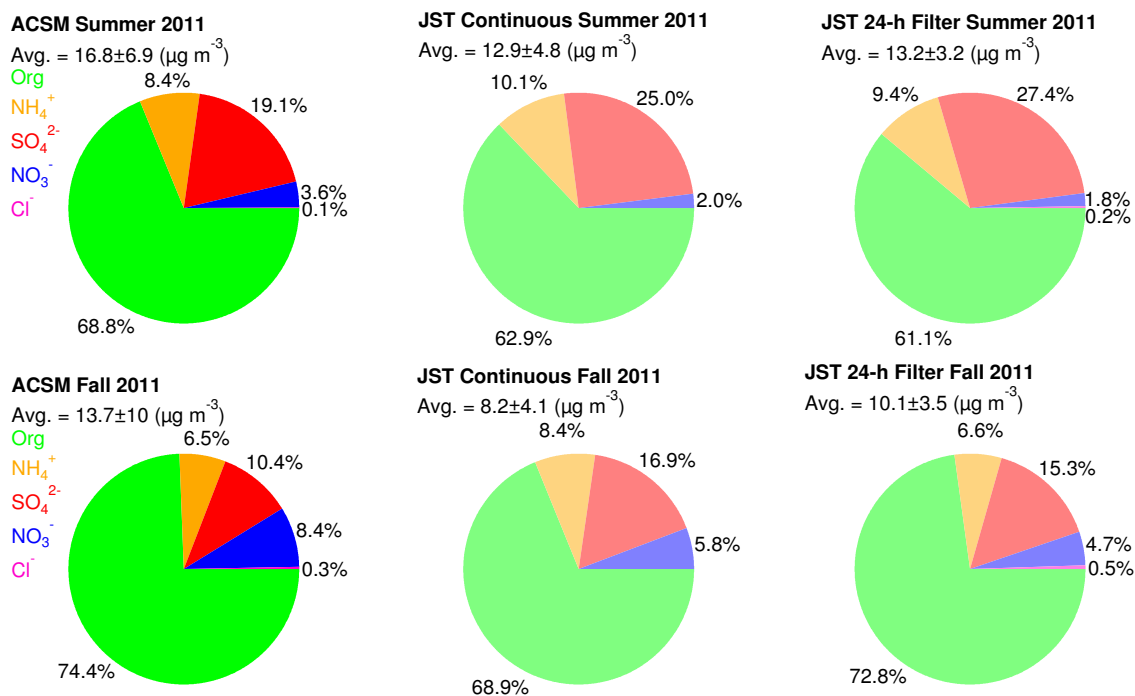
Methods	Mass	OC	SO ₄ ²⁻	NO ₃ ⁻	NH ₄ ⁺	Cl ⁻	EC
JST Continuous ^a							
Summer 2011	13.67 \pm 5.09	3.89 \pm 1.14	3.78 \pm 1.74	0.26 \pm 0.18	1.34 \pm 0.49	-	0.79 \pm 0.50
Fall 2011	9.11 \pm 5.58	3.34 \pm 2.38	1.59 \pm 1.27	0.67 \pm 0.54	0.69 \pm 0.34	-	0.99 \pm 1.07
JST Filter ^a							
Summer 2011	13.23 \pm 5.21	3.78 \pm 1.17	3.65 \pm 1.34	0.25 \pm 0.05	1.25 \pm 0.47	0.03 \pm 0.01	0.74 \pm 0.29
Fall 2011	9.85 \pm 4.40	3.86 \pm 1.55	1.57 \pm 0.74	0.48 \pm 0.36	0.68 \pm 0.30	0.05 \pm 0.05	0.98 \pm 0.78

6 ^a JST measures PM_{2.5} mass and chemical constituents.

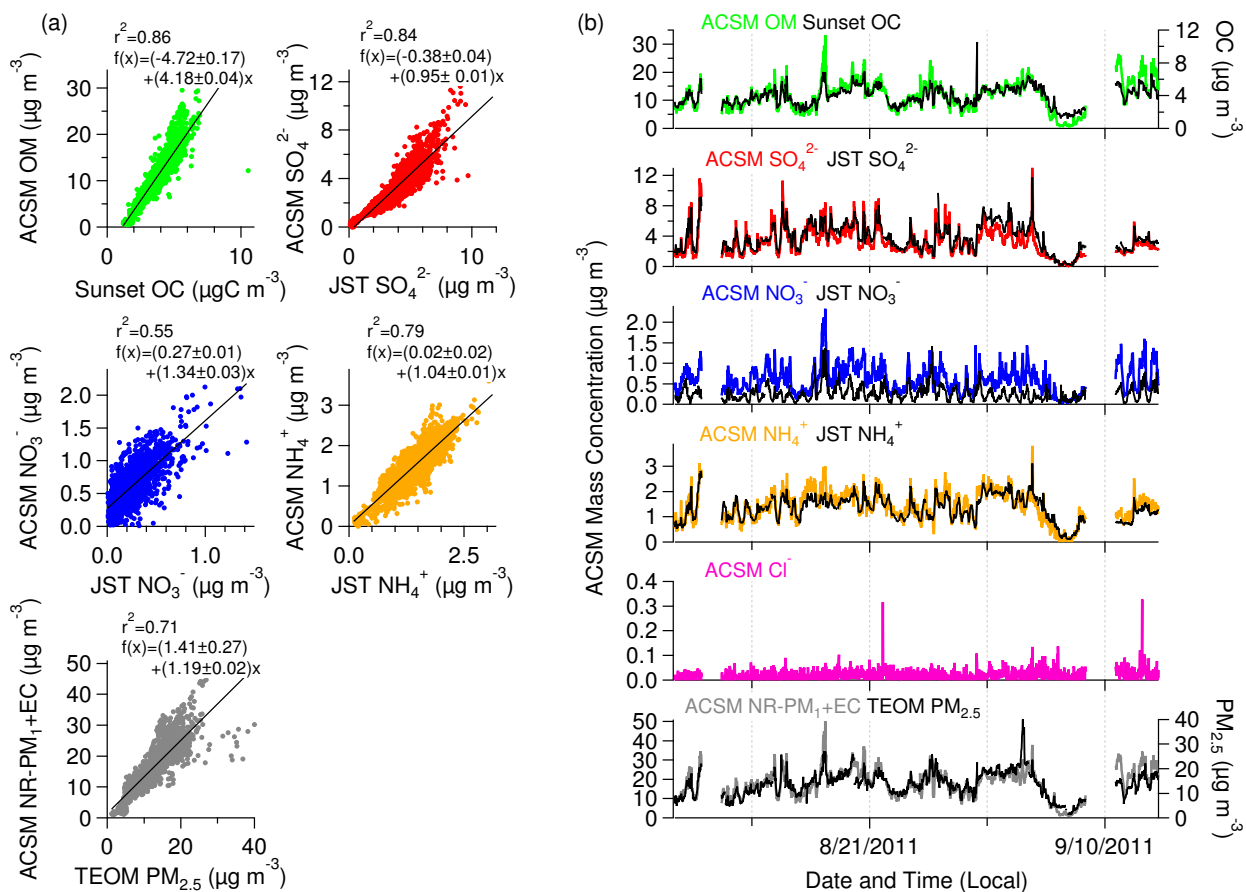
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8 **Table S2.** Ambient aerosol mass concentrations measured by the integrated FRM methods
9 presented in average concentration \pm 1 standard deviation.

	FRM Filters	
	PM ₁ ($\mu\text{g m}^{-3}$)	PM _{2.5} ($\mu\text{g m}^{-3}$)
Winter 2011	8.08 \pm 3.44	9.10 \pm 3.85
Spring 2012	8.58 \pm 2.67	9.71 \pm 3.01
Summer 2012	8.71 \pm 2.65	10.38 \pm 3.02

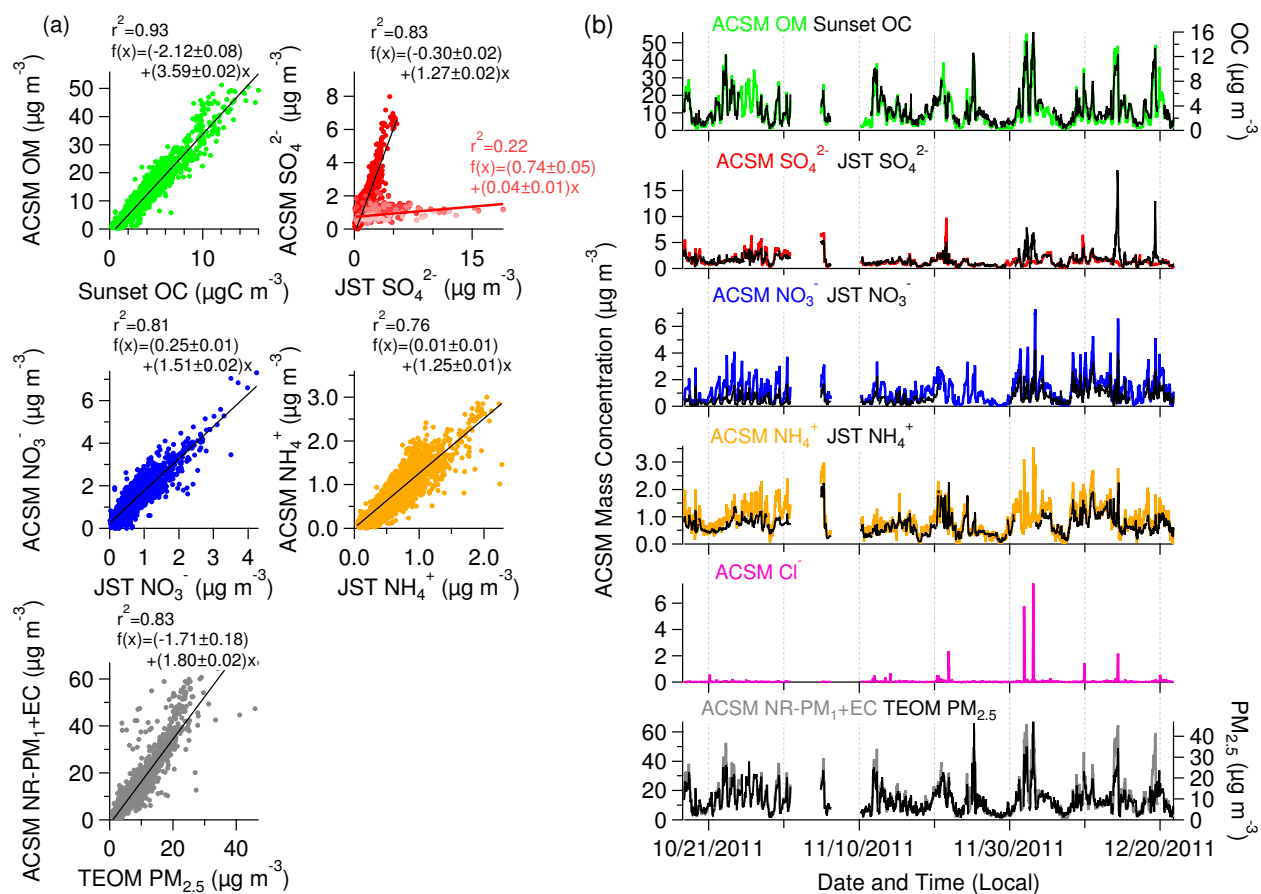
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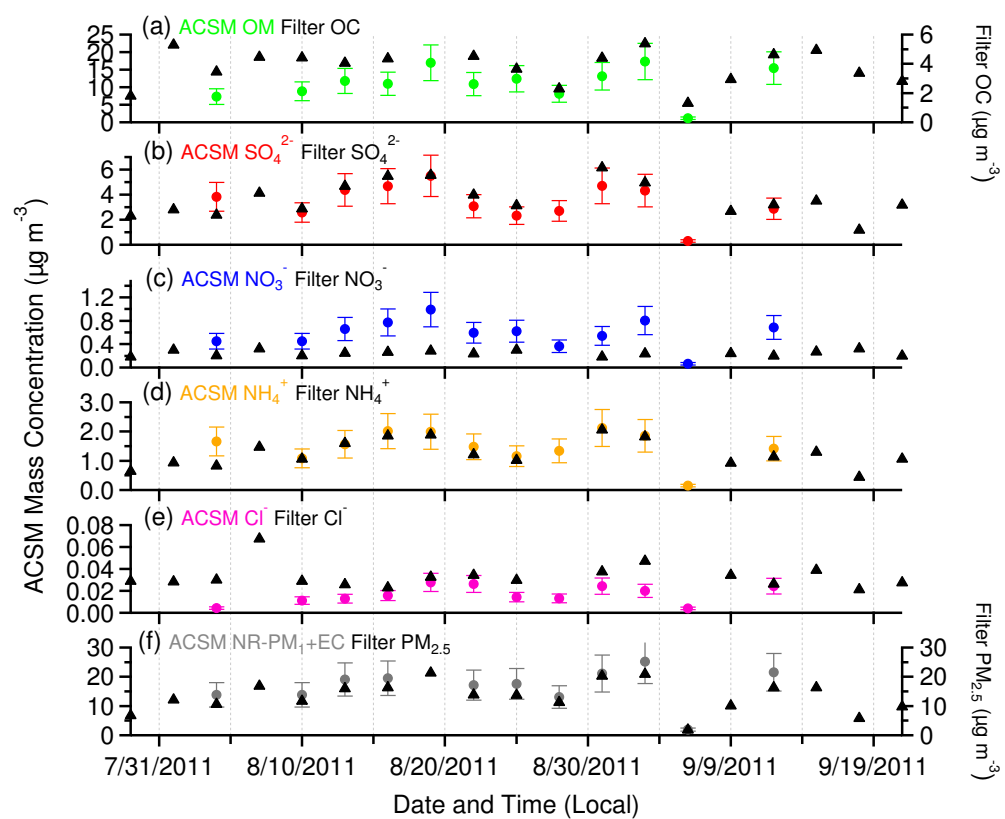
1 **Figure S3.** Pie charts of speciated aerosol measurements from the ACSM and JST. Pie charts
 2 in sharp colors are the ACSM, while the blur colors indicate other measurement techniques.
 3 Organic fraction of filter-adjusted continuous data was calculated from OC measurement
 4 multiplied by 1.8, which is ratio of OM/OC. For the 24-h filter-based data, it was calculated
 5 from OC measurement multiplied by 1.8 and correction values (SAF), that are 1.13 and 1.07
 6 for periods of July–September and October–December, respectively. ACSM measures PM_{10}
 7 while JST measures $\text{PM}_{2.5}$. Average $\text{PM}_{2.5}$ mass for 24-h filter based measurement was
 8 calculated from five species (i.e., OM, NH_4^+ , SO_4^{2-} , NO_3^- , and Cl^-), hence it excluded
 9 contribution from other anions such as Na^+ , Mg^+ , K^+ , and Ca^+ . Calculation of average $\text{PM}_{2.5}$
 10 mass for continuous measurement did not include chloride and other anions as they are not
 11 available.



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 2 **Figure S4.** (a) Linear regression correlation and (b) time series plots of organic and inorganic
 3 constituents measured by the UNC ACSM and collocated measurements at JST site during
 4 summer 2011.

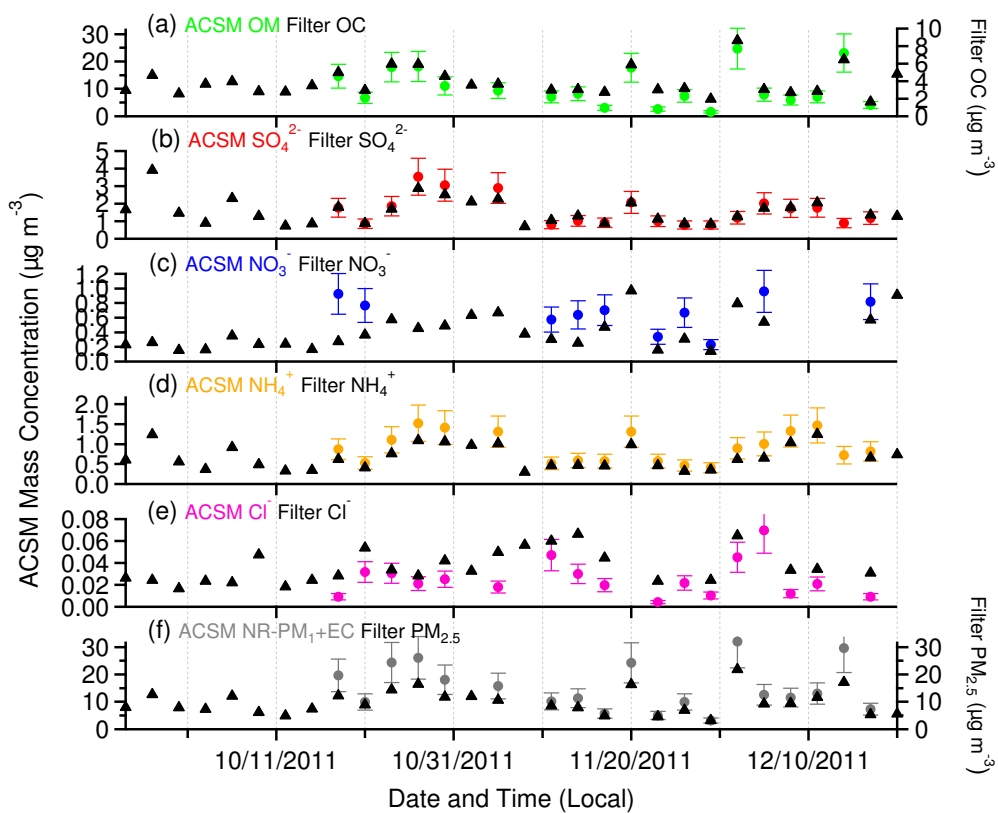


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 2 **Figure S5.** (a) Linear regression correlation and (b) time series plots of organic and inorganic
 3 constituents measured by the UNC ACSM and collocated measurements at JST site during fall
 4 2011.



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3 **Figure S6.** Time series plots with 30% of uncertainty of organic (OM vs. OC), inorganics
4 constituents (SO_4^{2-} , NO_3^- , NH_4^+ , and Cl^-), and mass concentrations measured by the UNC
5 ACSM and the JST 24-h filter measurement during summer period.



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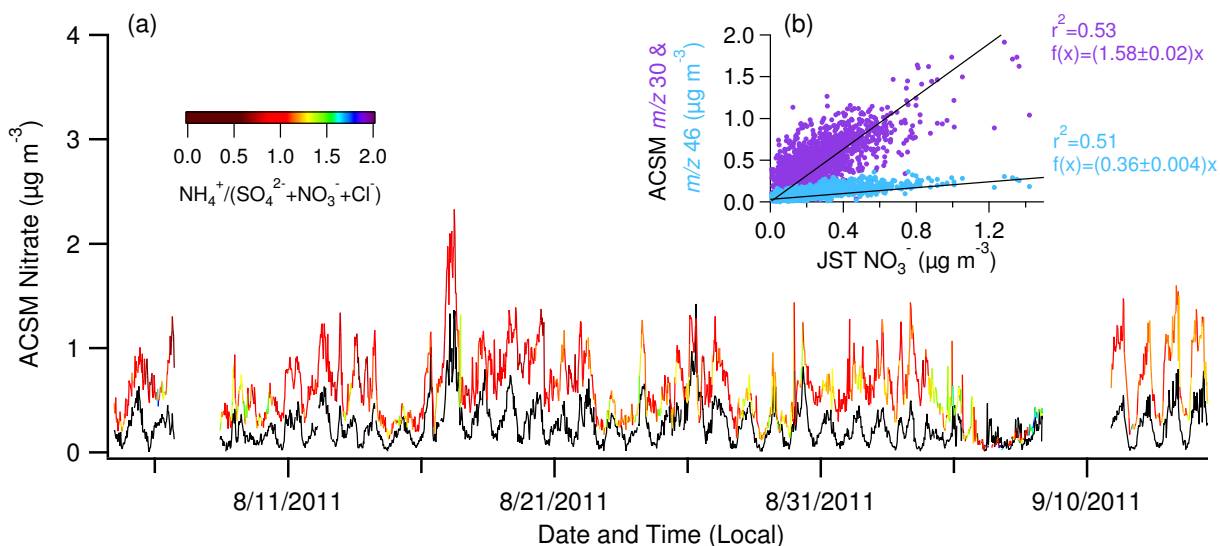
2 **Figure S7.** Time series plots with 30% of uncertainty of organic (OM vs. OC), inorganics
 3 constituents (SO_4^{2-} , NO_3^- , NH_4^+ , and Cl^-), and mass concentrations measured by the UNC
 4 ACSM and the JST 24-h filter measurement during fall period.

1 **D. Influence of organic nitrate component to ACSM NO₃⁻ signal**

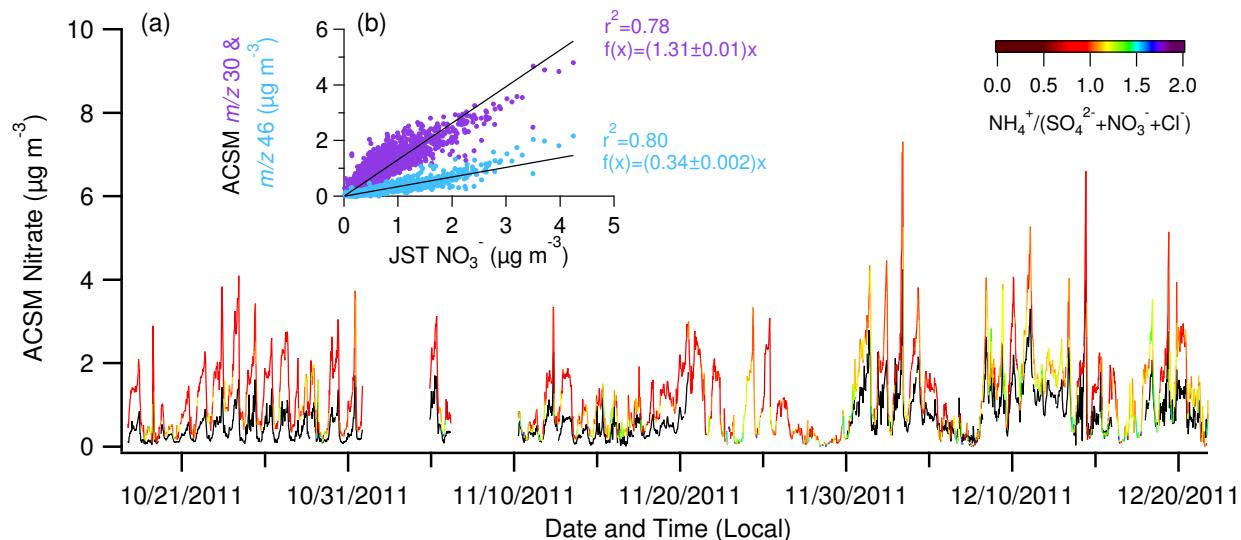
2 Discrepancies in the ACSM NO₃⁻ and the continuous measurements might also be
3 attributable to the overall low concentrations of NO₃⁻ in summer where both measurements are
4 near their detection limits. ACSM NO₃⁻ measurements are based on the measured *m/z* 30 and
5 *m/z* 46 ion signals. At low concentrations, small contributions to the *m/z* 30 signal can also
6 originate from organic nitrates (NO⁺), oxygenated organics (CH₂O⁺), and/or organic-nitrate
7 compounds (CH₄N⁺) that are not precisely accounted for. This *m/z* 30 interference can result in
8 higher reported values for inorganic nitrate (as measured from NO⁺ (*m/z* 30) and NO₂⁺ (*m/z* 46))
9 (Marcolli et al., 2006, Bae et al., 2007). Contribution of sum of organic and inorganic nitrate
10 can be significant to total secondary organic aerosol (SOA), although, Rastogi et al. (2011)
11 suggested that contribution of water soluble nitrogenous organic compounds is not significant
12 during summer in the southeastern U.S.

13 To investigate aerosol acidity influence to NO₃⁻ measurement, time series traces of
14 ACSM NO₃⁻ colour coded by degree of neutralization calculated according to Zhang et al.
15 (2007), and JST NO₃⁻ measured in summer and fall are presented in Figures S9 and S10,
16 respectively. Most of the time, the aerosol is slightly acidic (ratio of NH₄⁺ to SO₄²⁻ + NO₃⁻ + Cl⁻
17 < 1) in summer, suggesting that nitrate concentration in ambient acidic aerosol is usually low
18 due to HNO₃ displacement by H₂SO₄ (Zhang et al., 2005b). In contrast, during fall season the
19 aerosol is less acidic (ratio > 1) and the nitrate concentration is higher. The linear correlations
20 of *m/z* 30 and 46 from the ACSM NO₃⁻ with JST NO₃⁻ are moderate in summer (*r*² = 0.5), but
21 they are stronger in fall (*r*² ≥ 0.6). Interestingly, while linear regression slope of *m/z* 30 versus
22 JST NO₃⁻ was decreasing from summer to fall (1.58 to 1.31), slopes of *m/z* 46 were relatively
23 constant between these two seasons (0.36 to 0.34). These may suggest that the *m/z* 30 ion
24 measured by the ACSM is likely being influenced by fragments other than NO₃⁻ compared to
25 that of *m/z* 46. In order to investigate the influence of organic or oxygenated organic species to
26 *m/z* 30, time series traces of excess of *m/z* 30 signal ($\Delta m/z$ 30) calculated by formula provided
27 in Bae et al. (2007) in the summer and fall are presented in Figures S10 and S11, respectively.
28 The $\Delta m/z$ 30, which is suggested to be derived from organic-related or organic nitrate-related
29 *m/z* 30 (Bae et al., 2007), has positive values most of time, but the signal is lower in summer
30 (Figure S10a) than in fall (Figure S11a). During both seasons, the linear correlations between
31 $\Delta m/z$ 30 and the HOA factor (Figures S10b and S11b) derived from positive matrix factorization
32 (PMF) analysis of the ACSM organic fraction (Budisulistiorini et al., 2013) are weak (*r*² < 0.2),
33 but they are better for correlations with OOA factor (Figures S10c and S11c). HOA factor is

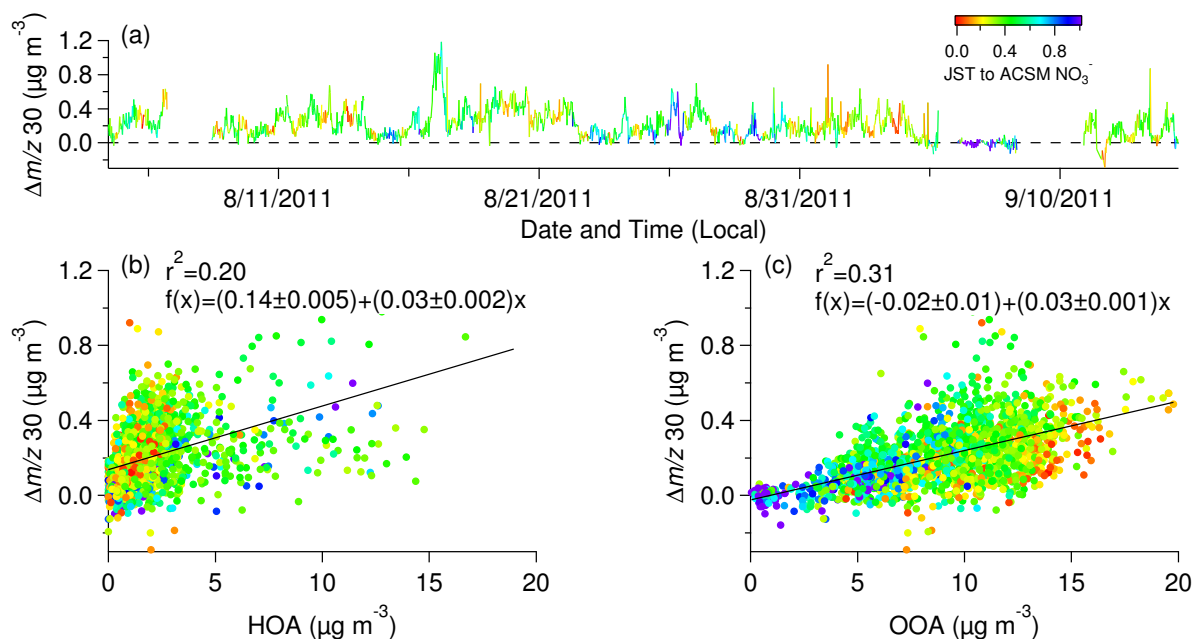
1 mainly attributed to organic aerosol formed from primary emissions and mass spectra are
 2 dominated by ions identical with hydrocarbons. On the other hand, OOA factor is characterized
 3 by mass spectra dominated by oxygenated ion fragments, and thus linked to SOA (Zhang et al.,
 4 2005a). The moderate correlation of $\Delta m/z$ 30 with OOA factor ($r^2 = 0.5$) in the fall might
 5 indicate that the m/z 30 signals measured by the ACSM are influenced by oxygenated organic
 6 species.



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 8 **Figure S8.** (a) Time series traces of the ACSM nitrate color coded by degree of neutralization
 9 and JST nitrate (black line), and (b) correlation scatterplot between the ACSM nitrate ion
 10 tracers, i.e., m/z 30 and 46, and JST nitrate for summer 2011 period.

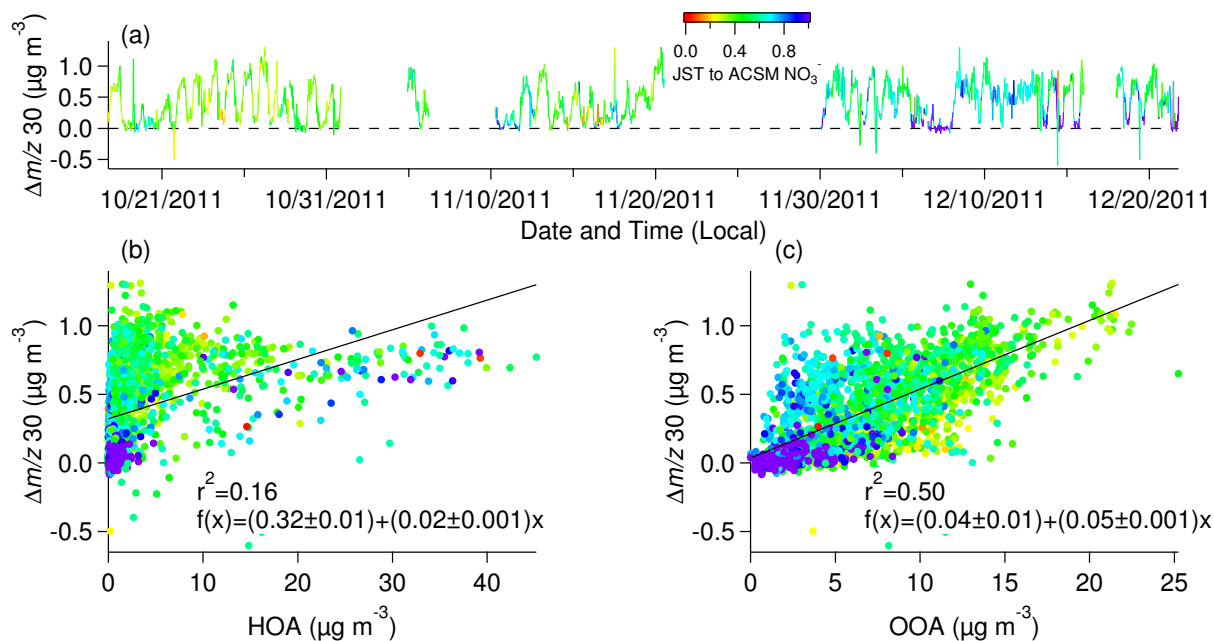


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 12 **Figure S9.** (a) Time series traces of the ACSM nitrate color coded by degree of neutralization
 13 and JST nitrate (black line), and (b) correlation scatterplot between the ACSM nitrate ion
 14 tracers, i.e., m/z 30 and 46, and JST nitrate for fall 2011 period.



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Figure S10. (a) Time series traces of the ACSM nitrate color coded by ratio of JST nitrate to ACSM nitrate, and correlation scatterplot between estimated $m/z\ 30$ signal excess attributed to organic-linked ($\Delta m/z\ 30$ mass) and (b) HOA and (c) OOA (= LV-OOA + SV-OOA + IEPOX-OA) factors from PMF analysis for summer 2011 period.



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Figure S11. (a) Time series traces of the ACSM nitrate color coded by ratio of JST nitrate to ACSM nitrate, and correlation scatterplot between estimated $m/z\ 30$ signal excess attributed to

1 organic-linked ($\Delta m/z$ 30 mass) and (b) HOA and (c) OOA (= LV-OOA + SV-OOA) factors
2 from PMF analysis for fall 2011 period.

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