



Supplement of

Seasonal characterization of submicron aerosol chemical composition and organic aerosol sources in the southeastern United States: Atlanta, Georgia, and Look Rock, Tennessee

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1 ACSM Analysis

| 2 | Table S1. Estimated dry density of PM ₁ and meteorological conditions at JST and LRK sites |
|---|---|
| 3 | during 2012 and 2013, respectively. |

| | Win | ter | Spr | ing | ; | Summer | | Fa | all |
|--------------------------------------|--------|--------------|-----------|----------|--------|--------|-------|-------|-------|
| | JST | LRK | JST | LRK | JST | JST-11 | LRK | JST | LRK |
| Dry density $(g \text{ cm}^{-3})$ | 1.53 | 1.59 | 1.58 | 1.5 | 1.53 | n.a. | 1.54 | 1.54 | 1.55 |
| | | Mete | eorologic | al condi | tions | | | | |
| Precipitation (mm) | 83.98 | <i>n.a</i> . | 216.94 | n.a. | 80.84 | 174.14 | n.a. | 76.46 | n.a. |
| Solar radiation (W m ⁻²) | 184.64 | n.a. | 329.91 | n.a. | 294.04 | 239.03 | n.a. | 216.4 | n.a. |
| Temperature (°C) | 12.29 | 4.15 | 21.26 | 12.83 | 26.02 | 25.89 | 20.97 | 14.49 | 11.1 |
| RH (%) | 66.01 | 71.48 | 65.15 | 67.26 | 69.05 | 68.45 | 80.15 | 67.56 | 75.18 |

n.a. is data unavailable. JST-11 is data from measurements at JST in 2011.

1 PMF Analysis of JST Winter 2012

The ratio of Q/Qexp for p of 1 to 10 was used to determine the optimum number of factors in 2 3 PMF analysis of JST winter 2012 (Fig. S1). Time series and mass spectra of O/Oexp suggest 4 that at least four factors are needed to describe the solution. The time series and mass spectrum of the four-factor solution were significantly lower than those of three factor 5 6 solution. Five factor solution resulted in splitting factors without substantial reduction of 7 Q/Qexp. Four factor solution was further diagnosed for time series and profile uncertainties as 8 well as examination of multiple random seeds. Seed analysis in Fig. S2 shows that changes in 9 mass fraction contribution of each factor were negligible (< 1%) over seed range. Similarly, 10 Q/Qexp values at different seed were nearly identical with very small changes (< 1%). All 11 four factors showed some uncertainties in their mass spectra and time series, which were nonetheless small compared to the general factor profile and contribution. Thus, four factor 12 13 solution is selected as the best solution and FPEAK = 0 is chosen based on correlation of 14 factor time series with external gas- and particle-phase tracers. Diagnostic plots and 15 correlations with external tracers and reference mass spectra are provided in Fig. S3 and Table 16 S2, respectively.



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2 Figure S1. Time series (a) and mass spectra (b) of Q/Qexp of factors in PMF analysis of JST 3 winter 2012.





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Figure S2. Multiple random seed analysis of (a) fractional contribution of OA factors for the 7 chosen number of factors, and (b) Q/Qexp as a function of seed of JST winter 2012. 8 Uncertainties of the candidate four factor solution time series (b) and mass spectra (c) are 9 shown in black line with $1-\sigma$ error bars in red.



Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK, (d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA 6 mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each 7 point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for 8 each m/z.

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1 PMF Analysis of JST Spring 2012

2 The ratio of O/Oexp for p of 1 to 10 was used to determine the optimum number of factors in PMF analysis of JST spring 2012 (Fig. S4). Time series and mass spectra of Q/Qexp suggest 3 4 that at least three factors are needed to describe the solution. The time series and mass 5 spectrum of the four-factor solution were lower than those of three factor solution. However, 6 a factor with overall low concentration showed noisy temporal variation, which might indicate 7 a split factor. Five factor solution resulted in factors without distinctive profiles and/or time 8 series as well as insignificant reduction of O/Oexp. Three factor solution was further 9 diagnosed for time series and profile uncertainties as well as examination of multiple random 10 seeds. Seed analysis in Fig. S5 shows that changes in mass fraction contribution of each factor 11 were negligible (< 1%) over seed range. Similarly, Q/Qexp values at different seed were 12 nearly identical with very small changes (< 1%). All three factors showed some uncertainties in their mass spectra and time series, which were nonetheless small compared to the general 13 14 factor profile and contribution. Thus, three factor solution is selected as the best solution and 15 FPEAK = 0 is chosen based on correlation of factor time series with external gas- and 16 particle-phase tracers. Diagnostic plot and correlations with external tracers and reference 17 mass spectra are provided in Fig. S6 and Table S2, respectively.



Figure S4. Time series (a) and mass spectra (b) of Q/Qexp of factors in PMF analysis of JST spring 2012.





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Figure S5. Multiple random seed analysis of (a) fractional contribution of OA factors for the chosen number of factors, and (b) Q/Qexp as a function of seed of JST spring 2012. 8 Uncertainties of the candidate three factor solution time series (b) and mass spectra (c) are





Figure S6. Diagnostic plots for PMF analysis of JST spring 2012 three factor solution: (a) Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK, 5 (d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA 6 mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each 7 point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for 8 each m/z.

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1 PMF Analysis of JST summer 2012

2 The ratio of O/Oexp for p of 1 to 10 was used to determine the optimum number of factors in PMF analysis of JST summer 2012 (Fig. S7). Time series and mass spectra of Q/Qexp 3 4 suggest that at least three factors are needed to describe the solution. The time series and mass 5 spectrum of the four-factor solution were lower than those of three factor solution. However, 6 a factor with overall low concentration showed noisy temporal variation, which might indicate 7 a split factor. Five factor solution resulted in factors without distinctive profiles and/or time 8 series as well as insignificant reduction of O/Oexp. Three factor solution was further 9 diagnosed for time series and profile uncertainties as well as examination of multiple random 10 seeds. Seed analysis in Fig. S8 shows that changes in mass fraction contribution of each factor 11 were negligible (< 1%) over seed range. Similarly, Q/Qexp values at different seed were 12 nearly identical with very small changes (< 1%). All three factors showed some uncertainties in their mass spectra and time series, which were nonetheless small compared to the general 13 factor profile and contribution. Thus, three factor solution is selected as the best solution and 14 15 FPEAK = 0 is chosen based on correlation of factor time series with external gas- and 16 particle-phase tracers. Diagnostic plots are provided in Fig. S9 and correlations of factor with 17 external tracers and reference mass spectra are provided in Table S2.



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Figure S7. Time series (a) and mass spectra (b) of Q/Qexp of factors in PMF analysis of JST summer 2012.



5 6 7 Figure S8. Multiple random seed analysis of (a) fractional contribution of OA factors for the chosen number of factors, and (b) Q/Qexp as a function of seed of JST spring 2012. Uncertainties of the candidate three factor solution time series (b) and mass spectra (c) are 9 shown in black line with $1-\sigma$ error bars in red.



Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected

for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK,

(d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA

mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each

point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for

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8 9 each m/z.

1 PMF Analysis of JST Fall 2012

2 The ratio of O/Oexp for p of 1 to 10 was used to determine the optimum number of factors in PMF analysis of JST fall 2012 (Fig. S10). Time series and mass spectra of Q/Qexp suggest 3 4 that at least four factors are needed to describe the solution. The time series and mass 5 spectrum of the four-factor solution were lower than those of three factor solution. Five factor 6 solution yielded splitting factors that profiles and/or time series could not be distinguished, 7 and minimum reduction of Q/Qexp. Four factor solution was further diagnosed for time series 8 and profile uncertainty as well as examination of multiple random seeds. Seed analysis in Fig. 9 S11 shows that changes in mass fraction contribution of each factor were negligible (< 1%) 10 over seed range. Similarly, Q/Qexp values at different seed were nearly identical with very 11 small changes (< 1%). All four factors showed some uncertainties in their mass spectra and 12 time series, which were nonetheless small compared to the general factor profile and contribution. Four factor is selected as the best solution and FPEAK = 0 is chosen based on 13 14 correlation of factor time series with external gas- and particle-phase tracers. Diagnostic plots 15 and factor correlations with external tracers and reference mass spectra are provided in Fig. 16 S12 and Table S2, respectively.



Figure S10. Time series (a) and mass spectra (b) of Q/Qexp of factors in PMF analysis of JST fall 2012.



5 6 Figure S11. Multiple random seed analysis of (a) fractional contribution of OA factors for the 7 chosen number of factors, and (b) Q/Qexp as a function of seed of JST spring 2012. 8 Uncertainties of the candidate four factor solution time series (b) and mass spectra (c) are 9 shown in black line with $1-\sigma$ error bars in red.





Figure S12. Diagnostic plots for PMF analysis of JST fall 2012 four factor solution: (a) Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected 4 for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK, 5 (d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA 6 mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each 7 point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for 8 each m/z.

1 PMF Analysis of LRK Winter 2013

The ratio of Q/Qexp for p of 1 to 5 was used to determine the optimum number of factors in 2 3 PMF analysis of LRK winter 2013 (Fig. S13). Time series and mass spectra of O/Oexp 4 suggest that at least two factors are needed to describe the solution. Three and four factor solutions resulted in lower Q/Qexp. However, time series and mass spectra of the three and 5 6 four factor solutions were correlated with each other. Tuning FPEAK of the three and four 7 factor solutions resulted in substantial change in factors profiles, which suggest splitting 8 factors. Thus, adding more factor than two factor solution might not yield additional 9 information from the LRK winter 2013. Two factor solution was further diagnosed for time 10 series and profile uncertainty as well as examination of multiple random seeds. Seed analysis 11 in Fig. S14 shows that changes in mass fraction contribution of each factor were negligible (< 1%) over seed range. Similarly, O/Oexp values at different seed were nearly identical with 12 13 very small changes (< 1%). All two factors showed some uncertainties in their mass spectra 14 and time series, which were nonetheless small compared to the general factor profile and 15 contribution. Thus, two factor is selected as the best solution and FPEAK = 0 is chosen based 16 on correlation of factor time series with external gas- and particle-phase tracers. Diagnostic plots and correlations with external tracers and reference mass spectra are provided in Fig. 17 18 S15 and Table S3, respectively.



5 Date and Time (Local) m/z6 **Figure S14.** Multiple random seed analysis of (a) fractional contribution of OA factors for the 7 chosen number of factors, and (b) Q/Qexp as a function of seed of LRK winter 2013. 8 Uncertainties of the candidate two factor solution time series (b) and mass spectra (c) are 9 shown in black line with 1- σ error bars in red.



Figure S15. Diagnostic plots for PMF analysis of LRK winter 2013 two factor solution: (a) Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected 4 for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK, 5 (d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA 6 mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each 7 point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for 8 each m/z.

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1 PMF Analysis of LRK Spring 2013

2 The ratio of O/Oexp for p of 1 to 5 was used to determine the optimum number of factors in PMF analysis of LRK spring 2013 (Fig. S16). Time series and mass spectra of Q/Qexp 3 4 suggest that at least two factors are needed to describe the solution. Three factor solution 5 vielded lower Q/Qexp with distinctive factor profiles and time series. Tuning FPEAK of the 6 three factor solution resulted in improved correlations of factor times series with external 7 tracers without significantly changed factors profiles. Four factor solution yielded splitting 8 factors that did not give additional information about LRK spring 2013. Three factor solution 9 was further diagnosed for time series and profile uncertainty as well as examination of 10 multiple random seeds. Seed analysis in Fig. S17 shows that changes in mass fraction 11 contribution of each factor were negligible (< 1%) over seed range. Similarly, Q/Qexp values 12 at different seed were nearly identical with very small changes (< 1%). All three factors showed some uncertainties in their mass spectra and time series, which were nonetheless 13 14 small compared to the general factor profile and contribution. Three factor solution is selected as the best solution and FPEAK = -0.15 is chosen based on correlation of factor time series 15 16 with external gas- and particle-phase tracers. Diagnostic plots and correlations with external 17 tracers and reference mass spectra are provided in Fig. S18 and Table S3, respectively.



Figure S16. Time series (a) and mass spectra (b) of Q/Qexp of factors in PMF analysis of LRK spring 2013.



5 **Figure S17.** Multiple random seed analysis of (a) fractional contribution of OA factors for the chosen number of factors, and (b) Q/Qexp as a function of seed of LRK spring 2013. Uncertainties of the candidate three factor solution time series (b) and mass spectra (c) are shown in black line with 1- σ error bars in red.



Figure S18. Diagnostic plots for PMF analysis of LRK spring 2013 three factor solution: (a) Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected 4 for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK, 5 (d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA 6 mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each 7 point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for 8 each m/z.

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1 PMF Analysis of LRK Summer 2013

2 The ratio of O/Oexp for p of 1 to 5 was used to determine the optimum number of factors in PMF analysis of LRK summer 2013 (Fig. S19). Time series and mass spectra of Q/Qexp 3 4 suggest that at least two factors are needed to describe the solution. Three factor solution 5 resulted in lower Q/Qexp and distinctive factor profiles and time series. Tuning FPEAK of the 6 three factor solution improved factors correlation with external tracers without changing the 7 mass spectra substantially. Adding the fourth factor resulted in splitting factors, and did not 8 provide additional information about LRK summer 2013. Three factor solution was further 9 diagnosed for time series and profile uncertainties as well as examination of multiple random 10 seeds. Seed analysis in Fig. S20 shows that changes in mass fraction contribution of each 11 factor were negligible (< 1%) over seed range. Similarly, Q/Qexp values at different seed 12 were nearly identical with very small changes (< 1%). All three factors showed some uncertainties in their mass spectra and time series, which were nonetheless small compared to 13 14 the general factor profile and contribution. Three factor is selected as the best solution and 15 FPEAK = -0.03 is chosen based on correlation of factor time series with external gas- and 16 particle-phase tracers. Diagnostic plots and correlations with external tracers and reference 17 mass spectra are provided in Fig. S21 and Table S3, respectively.



Figure S19. Time series (a) and mass spectra (b) of Q/Qexp of factors in PMF analysis of LRK summer 2013.



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Figure S20. Multiple random seed analysis of (a) fractional contribution of OA factors for the 7 chosen number of factors, and (b) Q/Qexp as a function of seed of LRK summer 2013. 8 Uncertainties of the candidate three factor solution time series (b) and mass spectra (c) are 9 shown in black line with $1-\sigma$ error bars in red.



Figure S21. Diagnostic plots for PMF analysis of LRK summer 2013 three factor solution: (a) Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK, (d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for each m/z.

1 PMF Analysis of LRK Fall 2013

2 The ratio of O/Oexp for p of 1 to 5 was used to determine the optimum number of factors in PMF analysis of LRK fall 2013 (Fig. S22). Time series and mass spectra of Q/Qexp suggest 3 4 that at least two factors are needed to describe the solution. Three factor solution resulted in 5 lower Q/Qexp and distinctive factors. Tuning FPEAK of the three factor solution improved 6 correlations of factor with external tracers without substantially changed the profiles. Fourth 7 factor solution resulted in splitting factors and did not give additional information about LRK 8 fall 2013. Three factor solution was further diagnosed for time series and profile uncertainties 9 as well as examination of multiple random seeds. Seed analysis in Fig. S23 shows that 10 changes in mass fraction contribution of each factor were negligible (< 1%) over seed range. 11 Similarly, Q/Qexp values at different seed were nearly identical with very small changes (< 12 1%). All three factors showed some uncertainties in their mass spectra and time series, which were nonetheless small compared to the general factor profile and contribution. Three factor 13 14 is selected as the best solution and FPEAK = 0 is chosen based on correlation of factor time 15 series with external gas- and particle-phase tracers. Diagnostic plots and correlations with 16 external tracers and reference mass spectra are provided in Fig. S24 and Table S3, 17 respectively.





Figure S23. Multiple random seed analysis of (a) fractional contribution of OA factors for the chosen number of factors, and (b) Q/Qexp as a function of seed of LRK fall 2013. Uncertainties of the candidate three factor solution time series (b) and mass spectra (c) are shown in black line with 1- σ error bars in red.



Q/Qexp as a function of number of factors (p), (b) Q/Qexp as a function of FPEAK selected

for the chosen number of factors, (c) fractional contribution of OA factors for each FPEAK,

(d) correlation among PMF factors based on factor TS and MS, (e) TS of the measured OA

mass and the reconstructed OA mass, (f) variation of the residual of the fit, Q/Qexp for each

point in time (g) and for each m/z (h), and the box and whisker plot of the scaled residuals for

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, 8 9

each *m/z*.

Table S2. Correlations of PMF factor temporal variations (r_{TS}^2) and mass spectra (r_{MS}^2) resolved from OA measurements at JST with external gas- and particle-phase measurements and reference mass spectra.

| | НОА | | | | | BB | OA | | | LVO | DOA | | | SVG | DOA | | IEPOXOA | | | |
|---|------|------|------|------|------|------|--------------|------|------|------|------|------|------|------|--------------|------|---------|------|------|--------------|
| | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall |
| r_{TS}^2 | | | | | | | | | | | | | | | | | | | | |
| BC | 0.70 | 0.58 | 0.64 | 0.75 | 0.49 | n.a. | n.a. | 0.40 | 0.13 | 0.04 | 0.00 | 0.01 | 0.25 | n.a. | n.a. | 0.41 | n.a. | 0.20 | 0.14 | <i>n.a</i> . |
| CO | 0.74 | 0.63 | 0.58 | 0.81 | 0.42 | n.a. | n.a. | 0.39 | 0.07 | 0.03 | 0.00 | 0.00 | 0.24 | n.a. | n.a. | 0.34 | n.a. | 0.18 | 0.16 | <i>n.a</i> . |
| NO _x (=NO+NO ₂) | 0.81 | 0.70 | 0.64 | 0.81 | 0.28 | n.a. | n.a. | 0.37 | 0.02 | 0.00 | 0.04 | 0.00 | 0.23 | n.a. | n.a. | 0.30 | n.a. | 0.07 | 0.01 | <i>n.a</i> . |
| NO_y | 0.80 | 0.70 | 0.65 | 0.80 | 0.27 | n.a. | n.a. | 0.36 | 0.01 | 0.00 | 0.02 | 0.00 | 0.22 | n.a. | n.a. | 0.30 | n.a. | 0.08 | 0.02 | <i>n.a</i> . |
| NOz | 0.32 | 0.19 | 0.07 | 0.32 | 0.06 | n.a. | n.a. | 0.10 | 0.00 | 0.09 | 0.20 | 0.02 | 0.01 | n.a. | n.a. | 0.17 | n.a. | 0.10 | 0.24 | <i>n.a</i> . |
| O_3 | 0.30 | 0.21 | 0.15 | 0.25 | 0.15 | n.a. | n.a. | 0.28 | 0.02 | 0.04 | 0.21 | 0.01 | 0.22 | n.a. | n.a. | 0.12 | n.a. | 0.01 | 0.05 | n.a. |
| O_x (=NO ₂ +O ₃) | 0.00 | 0.04 | 0.03 | 0.02 | 0.02 | n.a. | n.a. | 0.01 | 0.02 | 0.11 | 0.25 | 0.10 | 0.00 | n.a. | n.a. | 0.00 | n.a. | 0.01 | 0.11 | n.a. |
| SO2 | 0.27 | 0.21 | 0.00 | 0.33 | 0.05 | n.a. | n.a. | 0.16 | 0.00 | 0.01 | 0.01 | 0.00 | 0.06 | n.a. | n.a. | 0.15 | n.a. | 0.02 | 0.02 | n.a. |
| SO4 | 0.03 | 0.05 | 0.00 | 0.10 | 0.02 | n.a. | n.a. | 0.03 | 0.04 | 0.17 | 0.18 | 0.20 | 0.00 | n.a. | n.a. | 0.18 | n.a. | 0.15 | 0.19 | <i>n.a</i> . |
| ACSM SO ₄ | 0.00 | 0.01 | 0.00 | 0.00 | 0.00 | n.a. | n.a. | 0.00 | 0.05 | 0.10 | 0.17 | 0.21 | 0.02 | n.a. | n.a. | 0.08 | n.a. | 0.14 | 0.26 | <i>n.a</i> . |
| ACSM NO ₃ | 0.15 | 0.31 | 0.26 | 0.07 | 0.25 | n.a. | n.a. | 0.19 | 0.17 | 0.05 | 0.11 | 0.10 | 0.38 | n.a. | n.a. | 0.16 | n.a. | 0.28 | 0.45 | <i>n.a</i> . |
| ACSM NH ₄ | 0.02 | 0.06 | 0.02 | 0.18 | 0.04 | n.a. | n.a. | 0.20 | 0.12 | 0.15 | 0.20 | 0.20 | 0.08 | n.a. | n.a. | 0.29 | n.a. | 0.18 | 0.28 | n.a. |
| LWC | 0.02 | 0.04 | 0.04 | 0.10 | 0.03 | n.a. | n.a. | 0.05 | 0.02 | 0.00 | 0.17 | 0.00 | 0.03 | n.a. | n.a. | 0.10 | n.a. | 0.02 | 0.06 | <i>n.a</i> . |
| pН | 0.14 | 0.06 | 0.18 | 0.15 | 0.06 | n.a. | n.a. | 0.23 | 0.00 | 0.03 | 0.07 | 0.03 | 0.20 | n.a. | n.a. | 0.02 | n.a. | 0.00 | 0.01 | n.a. |
| r^2_{MS} | | | | | | | | | | | | | | | | | | | | |
| HOA | 0.96 | 0.90 | 0.68 | 0.91 | 0.41 | n.a. | n.a. | 0.12 | 0.04 | 0.04 | 0.03 | 0.03 | 0.10 | n.a. | n.a. | 0.32 | n.a. | 0.15 | 0.18 | <i>n.a</i> . |
| LV-OOA | 0.04 | 0.07 | 0.55 | 0.21 | 0.45 | n.a. | <i>n.a</i> . | 0.85 | 0.95 | 0.94 | 0.92 | 0.93 | 0.85 | n.a. | n.a. | 0.73 | n.a. | 0.91 | 0.93 | <i>n.a</i> . |
| SV-OOA | 0.41 | 0.58 | 0.85 | 0.60 | 0.88 | n.a. | n.a. | 0.52 | 0.35 | 0.36 | 0.34 | 0.34 | 0.48 | n.a. | n.a. | 0.87 | n.a. | 0.68 | 0.69 | <i>n.a</i> . |
| BBOA | 0.46 | 0.62 | 0.79 | 0.66 | 0.77 | n.a. | <i>n.a</i> . | 0.45 | 0.27 | 0.29 | 0.28 | 0.27 | 0.43 | n.a. | <i>n.a</i> . | 0.68 | n.a. | 0.52 | 0.55 | n.a. |
| 82Fac | 0.17 | 0.31 | 0.67 | 0.39 | 0.62 | n.a. | <i>n.a</i> . | 0.72 | 0.70 | 0.72 | 0.70 | 0.69 | 0.73 | n.a. | <i>n.a</i> . | 0.74 | n.a. | 0.80 | 0.86 | n.a. |
| 91Fac | 0.60 | 0.72 | 0.94 | 0.83 | 0.68 | n.a. | <i>n.a</i> . | 0.49 | 0.39 | 0.37 | 0.33 | 0.33 | 0.43 | n.a. | <i>n.a</i> . | 0.68 | n.a. | 0.55 | 0.61 | <i>n.a</i> . |

| | HOA | | | | BBOA | | | LVOOA | | | SVOOA | | | | IEPOXOA | | | | | |
|---------------|------|------|------|------|------|------|------|-------|------|------|-------|------|------|------|---------|------|------|------|------|--------------|
| | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall |
| ATL IEPOX-OA | 0.13 | 0.27 | 0.71 | 0.37 | 0.79 | n.a. | n.a. | 0.72 | 0.61 | 0.61 | 0.60 | 0.58 | 0.74 | n.a. | n.a. | 0.92 | n.a. | 0.89 | 0.92 | n.a. |
| Lab IEPOX SOA | 0.20 | 0.36 | 0.61 | 0.42 | 0.62 | n.a. | n.a. | 0.47 | 0.34 | 0.37 | 0.37 | 0.35 | 0.48 | n.a. | n.a. | 0.60 | n.a. | 0.53 | 0.59 | n.a. |
| SOAS IEPOX-OA | 0.04 | 0.08 | 0.57 | 0.23 | 0.48 | n.a. | n.a. | 0.88 | 0.93 | 0.94 | 0.94 | 0.93 | 0.89 | n.a. | n.a. | 0.74 | n.a. | 0.92 | 0.95 | <i>n.a</i> . |
| SOAS 91Fac | 0.12 | 0.20 | 0.72 | 0.35 | 0.66 | n.a. | n.a. | 0.84 | 0.81 | 0.80 | 0.78 | 0.79 | 0.80 | n.a. | n.a. | 0.88 | n.a. | 0.94 | 0.95 | <i>n.a</i> . |

Mas spectra references: Ng et al. (2011), Robinson et al. (2011), Budisulistiorini et al. (2013, 2015)

Table S3. Correlations of PMF factor temporal variations (r_{TS}^2) and mass spectra (r_{MS}^2) resolved from OA measurements at LRK with external gas- and particle-phase measurements and reference mass spectra.

| | | BB | OA | | | LVO | DOA | | 91Fac | | | | IEPOXOA | | | |
|---|------|------|--------------|--------------|------|------|------|------|--------------|------|------|------|---------|------|------|------|
| | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall |
| r_{TS}^2 | | | | | | | | | | | | | | | | |
| BC | 0.21 | n.a. | n.a. | n.a. | 0.72 | 0.33 | 0.33 | 0.68 | n.a. | 0.30 | 0.37 | 0.17 | n.a. | 0.28 | 0.33 | 0.37 |
| CO | 0.00 | n.a. | n.a. | <i>n.a</i> . | 0.01 | 0.04 | 0.36 | 0.03 | n.a. | 0.11 | 0.24 | 0.18 | n.a. | 0.06 | 0.31 | 0.04 |
| NO_x (=NO+NO ₂) | 0.00 | n.a. | n.a. | <i>n.a</i> . | 0.00 | 0.10 | 0.00 | 0.07 | n.a. | 0.00 | 0.01 | 0.03 | n.a. | 0.14 | 0.01 | 0.05 |
| NO_y | 0.00 | n.a. | n.a. | <i>n.a</i> . | 0.01 | 0.01 | 0.04 | 0.07 | n.a. | 0.04 | 0.09 | 0.09 | n.a. | 0.02 | 0.07 | 0.05 |
| NOz | 0.01 | n.a. | n.a. | <i>n.a</i> . | 0.04 | 0.04 | 0.10 | 0.03 | n.a. | 0.04 | 0.07 | 0.11 | n.a. | 0.00 | 0.14 | 0.02 |
| O_3 | 0.09 | n.a. | n.a. | n.a. | 0.30 | 0.11 | 0.26 | 0.24 | n.a. | 0.09 | 0.09 | 0.00 | n.a. | 0.01 | 0.16 | 0.07 |
| O_x (=NO ₂ +O ₃) | 0.10 | n.a. | n.a. | n.a. | 0.32 | 0.10 | 0.21 | 0.22 | n.a. | 0.10 | 0.04 | 0.01 | n.a. | 0.01 | 0.15 | 0.06 |
| SO2 | 0.02 | n.a. | n.a. | n.a. | 0.04 | 0.01 | 0.03 | 0.01 | n.a. | 0.01 | 0.03 | 0.08 | n.a. | 0.00 | 0.15 | 0.00 |
| SO4 | 0.01 | n.a. | n.a. | n.a. | 0.09 | 0.10 | 0.22 | 0.32 | n.a. | 0.00 | 0.10 | 0.00 | n.a. | 0.00 | 0.56 | 0.03 |
| ACSM SO ₄ | 0.01 | n.a. | n.a. | n.a. | 0.11 | 0.44 | 0.36 | 0.47 | n.a. | 0.04 | 0.13 | 0.03 | n.a. | 0.27 | 0.66 | 0.11 |
| ACSM NO ₃ | 0.04 | n.a. | n.a. | n.a. | 0.06 | 0.13 | 0.50 | 0.03 | n.a. | 0.12 | 0.49 | 0.18 | n.a. | 0.01 | 0.55 | 0.00 |
| ACSM NH ₄ | 0.04 | n.a. | n.a. | <i>n.a</i> . | 0.20 | 0.46 | 0.42 | 0.37 | n.a. | 0.08 | 0.19 | 0.11 | n.a. | 0.20 | 0.62 | 0.09 |
| LWC | 0.01 | n.a. | <i>n.a</i> . | n.a. | 0.01 | 0.01 | 0.04 | 0.01 | <i>n.a</i> . | 0.00 | 0.00 | 0.00 | n.a. | 0.00 | 0.00 | 0.01 |
| pН | 0.01 | n.a. | n.a. | n.a. | 0.10 | 0.05 | 0.08 | 0.02 | n.a. | 0.01 | 0.00 | 0.00 | n.a. | 0.13 | 0.09 | 0.01 |
| r^2_{MS} | | | | | | | | | | | | | | | | |
| HOA | 0.42 | n.a. | n.a. | n.a. | 0.03 | 0.02 | 0.05 | 0.06 | n.a. | 0.14 | 0.16 | 0.19 | n.a. | 0.14 | 0.09 | 0.16 |
| LV-OOA | 0.76 | n.a. | n.a. | n.a. | 0.94 | 0.92 | 0.97 | 0.98 | n.a. | 0.97 | 0.98 | 0.84 | n.a. | 0.99 | 0.97 | 0.97 |
| SV-OOA | 0.83 | n.a. | n.a. | <i>n.a</i> . | 0.33 | 0.30 | 0.41 | 0.42 | n.a. | 0.55 | 0.60 | 0.45 | n.a. | 0.61 | 0.51 | 0.65 |
| BBOA | 0.83 | n.a. | n.a. | n.a. | 0.21 | 0.18 | 0.28 | 0.30 | n.a. | 0.46 | 0.45 | 0.44 | n.a. | 0.45 | 0.43 | 0.47 |
| Borneo 82Fac | 0.84 | n.a. | n.a. | <i>n.a</i> . | 0.64 | 0.59 | 0.69 | 0.73 | n.a. | 0.84 | 0.81 | 0.68 | n.a. | 0.85 | 0.88 | 0.83 |
| Borneo 91Fac | 0.85 | n.a. | n.a. | <i>n.a</i> . | 0.35 | 0.32 | 0.41 | 0.45 | n.a. | 0.61 | 0.66 | 0.62 | n.a. | 0.60 | 0.52 | 0.63 |
| ATL IEPOX-OA | 0.85 | n.a. | n.a. | n.a. | 0.58 | 0.54 | 0.65 | 0.67 | n.a. | 0.76 | 0.76 | 0.60 | n.a. | 0.81 | 0.79 | 0.81 |
| Lab IEPOX SOA | 0.72 | n.a. | n.a. | n.a. | 0.26 | 0.23 | 0.30 | 0.33 | n.a. | 0.48 | 0.42 | 0.44 | n.a. | 0.47 | 0.53 | 0.46 |

| | | BB | OA | | LVOOA | | | | | 91 | Fac | | IEPOXOA | | | | |
|---------------|------|------|------|------|-------|------|------|------|------|------|------|------|---------|------|------|------|--|
| | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | Wtr | Spr | Smr | Fall | |
| SOAS IEPOX-OA | 0.81 | n.a. | n.a. | n.a. | 0.90 | 0.88 | 0.93 | 0.95 | n.a. | 0.98 | 0.95 | 0.86 | n.a. | 0.98 | 1.00 | 0.96 | |
| SOAS 91Fac | 0.88 | n.a. | n.a. | n.a. | 0.80 | 0.77 | 0.86 | 0.87 | n.a. | 0.92 | 0.97 | 0.79 | n.a. | 0.96 | 0.89 | 0.98 | |

Mass spectra references: Ng et al. (2011), Robinson et al. (2011), Budisulistiorini et al. (2013, 2015)

Table S4. Measurements of organic and inorganic species in JST site in 2012 as well as other sites in Atlanta, Georgia at different periods. Mass concentration is presented as average \pm standard deviation in unit of μ g m⁻³.

| 111400 0 | oneena anon | is presented | us average - | - Standard de | ideron in dine | or µg m i | | | | |
|-------------------|-----------------|---------------------|-----------------|---------------------|---------------------|-----------------|---------------------|----------------------|-----------------|---------------------|
| | Winter | Jan-13 ^a | Spring | May-12 ^b | Smr-11 ^c | Summer | Jul-12 ^d | Fall-11 ^e | Fall | Nov-12 ^f |
| OM | 6.94 ± 6.45 | 4.70 ± 3.60 | 3.24 ± 2.45 | 9.10 ± 4.30 | 11.19 ± 4.83 | 6.15 ± 3.35 | 9.60 ± 4.40 | 10.12 ± 8.89 | 8.22 ± 5.90 | 7.90 ± 5.10 |
| SO_4^{2-} | 1.35 ± 1.12 | 1.60 ± 1.20 | 1.51 ± 1.51 | 3.00 ± 1.50 | 2.66 ± 1.46 | 1.53 ± 1.08 | 4.0 ± 2.10 | 0.88 ± 0.60 | 1.98 ± 1.64 | 1.70 ± 0.90 |
| NO_3^- | 0.93 ± 0.86 | 1.40 ± 1.30 | 0.34 ± 0.31 | 0.40 ± 0.30 | 0.61 ± 0.33 | 0.40 ± 0.32 | 0.40 ± 0.40 | 1.15 ± 0.91 | 1.16 ± 1.02 | 1.20 ± 1.10 |
| $\mathrm{NH_4}^+$ | 0.80 ± 0.46 | 0.90 ± 0.60 | 0.51 ± 0.31 | 1.10 ± 0.50 | 1.23 ± 0.49 | 0.69 ± 0.35 | 1.20 ± 0.60 | 0.71 ± 0.39 | 1.08 ± 0.63 | 0.90 ± 0.60 |
| Cl | 0.03 ± 0.11 | 0.06 ± 0.11 | 0.01 ± 0.01 | 0.03 ± 0.03 | 0.02 ± 0.03 | 0.01 ± 0.04 | 0.02 ± 0.01 | 0.05 ± 0.24 | 0.04 ± 0.07 | 0.06 ± 0.07 |

 $\frac{C1}{a} \frac{0.03 \pm 0.11}{0.03 \pm 0.11} \frac{0.00 \pm 0.11}{0.01 \pm 0.01} \frac{0.03 \pm 0.11}{0.03 \pm 0.11} \frac{0.03 \pm 0.11}{0.03$

^f 6 November - 4 December 2012 at JST site (Xu et al. 2015)



Figure S25. Mass spectral comparisons of BBOA, LV-OOA, and IEPOX-OA resolved from both JST and LRK sites at different season.





1 2 3 4 5 6 Figure S26. Temporal variations of the m/z 82 fragment ion (C₅H₆O⁺) contribution to PMF factors resolved from OA measurements (a) at the JST site during spring and summer seasons, and (b) at the LRK site during spring, summer, and fall seasons.



- **Figure S27.** Scatter plots of m/z 91 of 91Fac factor versus (a) NO_x and (b) m/z 82 of IEPOX-OA factor at LRK site during spring (green) and summer (orange) seasons.
- 1 2 3 4