



Supplement of

Fine-mode organic mass concentrations and sources in the Amazonian wet season (AMAZE-08)

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Supplementary Material For

Fine-Mode Organic Mass Concentrations and Sources in the Amazonian Wet Season (AMAZE-08)

A. AMS sampling and data processing

Chen et al. (2009) and Martin et al. (2010) described the deployment of the AMS. Aerosol particles above the forest canopy were drawn through a stainless steel tube (1/2" OD; 10.9 mm ID) in a turbulent flow of 40 to 80 L min⁻¹ to the roof of the instrument container. The flow was then split in three: a 3-m line for filter sampling (20 L min⁻¹), a 2-m line (1/4" OD; 4.8 mm ID) into an Aerodyne high-resolution Aerosol Mass Spectrometer (HR-AMS) (4 L min⁻¹), and a by-pass line. Calculations for a Reynolds number of 5000 to 10000 for the turbulent-flow inlet suggested nearly 100% transmission of particles for diameters of 17 nm to 3.1 µm (Farmer et al., 2013)(Farmer et al., 2013)(Farmer et al., 2013). The flow in the AMS sampling line passed through a Nafion dryer just prior to entering the instrument container. The sampling RH measured at the AMS inlet ranged from 40% to 70%. Sampling temperature and pressure were also measured inline prior to entering the AMS. A second AMS operated by MPI-C sampled on a laminar-flow line (20 to 35% RH). The major results reported in this paper were confirmed by co-analysis of the MPI-C data set (Schneider et al., 2011).

The AMS data were saved in 150-s intervals in alternating medium- (V) and highresolution (W) modes. For the V-mode, the instrument was operated in "mass spectrum" and "particle-time-of-flight" submodes for equal time periods. The spectra were analyzed using the software toolkits Sequential Igor Data Retrieval (SQUIRREL), Peak Integration by Key Analysis (PIKA), and Analytic Procedure for Elemental Separation (APES). Standard relative ionization efficiencies (RIE) were used in the analysis, including 1.1 for nitrate, 1.2 for sulfate, 1.4 for

organic molecules, 4.0 for ammonium, 1.3 for chloride, and 2.0 for water (DeCarlo et al., 2006; Mensah et al., 2011).

Several updates were made to the fragmentation table (Allan et al., 2004). Specifically, the fragmentation coefficients at m/z 16, 17, 18, 29, 30, 37, 38, 39, 40, 44, and 46 were adjusted to account for the variability of gas-phase contributions and for the interference of ions having the same nominal m/z. The signals of NH₂⁺ at m/z 16, NH₃⁺ at m/z 17, NO⁺ at m/z 30, and NO₂⁺ at m/z 46 were calculated as time-dependent fractions of the signals at unit resolution. The organic signals of $[CO^+]_{org}$ at m/z 28 and $[H_xO^+]_{org}$ at m/z 16, 17, and 18 were adjusted using the approach of Aiken et al. (2008). The organic signals of C_3H^+ at m/z 37, $C_3H_2^+$ at m/z 38, $C_3H_3^+$ at m/z 39, and $C_3H_4^+$ at m/z 40, which made up 5-8% of the total organic signal, were calculated time-dependently based on the ratio of them to $C_2H_2^+$ at m/z 26. Air contributions (e.g., ¹⁵NN⁺ at m/z 29) were subtracted as a constant fraction of the N₂⁺ signal based on the data recorded with a HEPA filter in-line, with the remaining signal at m/z 29 assigned to CHO⁺. Variations of the $[CO_2^+]_{air}$ signal at m/z 44 were corrected using the measured gas-phase CO₂ concentrations and the data recorded with a HEPA filter in-line. For the periods when there were no gas-phase CO₂ measurements, mean diel CO₂ concentrations were used, which corresponds to 10% error for the organic particle mass concentration of 0.2 μ g m⁻³. For comparison, using only the data recorded with a HEPA filter for this correction caused 20% error at similar mass concentration.

Mass concentrations were calculated from the V-mode data and were adjusted to standard temperature and pressure (noted as STP; 273 K and 10⁵ Pa), which were approximately 10% greater than those at calibration conditions (299.3 K and 100591.7 Pa). Occasionally the sampling site was influenced by the exhaust plumes from the site power source, which was a

diesel generator located 0.72 km from TT34 and typically downwind. Abrupt increases in AMSmeasured sulfate mass concentrations, even greater than the organic concentrations, were indicators of influence by the local pollution source. These pollution events were excluded from the data sets analyzed herein. In the present study particle-phase water was not included in the calculations of species mass concentrations.

Organosulfate species have been observed both for laboratory-generated biogenic secondary organic particles under acidic conditions as well as for ambient particles sampled in the southeastern USA, Germany, and Hungary (Gomez-Gonzalez et al., 2008; Surratt et al., 2008; Iinuma et al., 2009). The technique employed in these studies was the analysis of filter samples by electrospray-ionization mass spectrometry coupled to pre-separation by liquid chromatography. In the AMS, the organosulfate species can fragment to organic ions ($C_xH_yO_z^+$), organosulfur ions ($C_xH_yO_zS^+$), and ions with a pattern indistinguishable from inorganic sulfate (e.g., SO_2^+) (Farmer et al., 2010). For the AMAZE-08 data set, signal intensities for $C_xH_yO_zS^+$ ions were not above noise in the collected high-resolution mass spectra. The agreement among AMS, IC, and PIXE sulfate mass concentrations, as well as the absence of organosulfur ions in the high-resolution mass spectra, did not provide evidence for substantial contributions of organosulfate species during AMAZE-08, at least at concentrations above uncertainty levels.

The AMS detection limits, calculated as three times the standard deviation of mass concentrations for filtered air obtained at 150-s intervals, were 0.06, 0.02, 0.001, 0.006, 0.002 μ g m⁻³ for organic material, sulfate, ammonium, chloride, and nitrate, respectively. The AMS is capable of focusing particles with 30-1000 nm with size-dependent particle transmission efficiency (Liu et al., 2007). As described in Gunthe et al. (2009), we operated the AMS at sampling pressures of 867–907 hPa. Under these conditions, the transmission efficiency is close

to 100% for particles with vacuum aerodynamic diameter d_{va} of 100–400 nm and is greater than 20% for particles with d_{va} of 50-1000 nm. For organic measurements, the estimated uncertainty is 30% at concentrations of 1 µg m⁻³ to 40% at concentrations of 0.5 µg m⁻³. It can increase to 100% for low organic concentrations (0.1 µg m⁻³). For sulfate measurements, the uncertainty is <10% for high concentrations (0.5 µg m⁻³) and about 40% for low concentrations (0.05 µg m⁻³).

The mass-diameter distributions reported herein for the AMS represented the average of 74 measurements. The distributions were selected for time periods having nearly identical mass concentrations of sulfate. For comparison, nephelometer data at 550 nm were averaged for the same time periods. The mass-diameter distributions were multiplied by diameter-dependent mass extinction efficiencies (m² g⁻¹) to estimate light scattering coefficients. The mass extinction efficiencies were calculated at 550 nm using Mie theory for a refractive index of 1.42 - 0.006 I (Guyon et al., 2003). An agreement was found between calculated and measured aerosol scattering coefficients, particularly for periods free of influence of long-range advection of mineral dust (see further in Sect. B and Fig. S2). Comparisons were also made between the AMS mass-diameter distributions and volume-diameter distributions measured by a Scanning Mobility Particle Sizer (Lund SMPS), consisting of a differential mobility analyzer attached to a condensation particle counter (Roldin, 2008). The combined data sets were used to estimate the particle effective density ρ_{eff} (kg m⁻³) based on an in-common mode diameter (Katrib et al., 2005) and the organic density $\rho_{\rm org}$ (see further in Sect. B and Fig. S3). The analysis obtained $\rho_{\rm eff}$ of 1390 ± 150 kg m $^{\text{-3}}$ and ρ_{org} of 1270 ± 110 kg m $^{\text{-3}}$ for the AMAZE-08 data set.

Atomic ratios of oxygen-to-carbon (O:C), hydrogen-to-carbon (H:C), and nitrogen-tocarbon (N:C), as well as the mass ratios of organic material to organic carbon (OM:OC), were calculated from the W-mode data following previously described methods (Aiken et al., 2008). A recent study shows that organic aerosol with mixed keto-, hydroxyl-, and acid-functionalities readily undergo thermally-induced dehydration and decarboxylation on the AMS vaporizer (Canagaratna et al., 2013 in preparation). Such dehydration and decarboxylation can lead much greater $(CO^+)_{org}$: $(CO_2^+)_{org}$ and $(H_2O^+)_{org}$: $(CO_2^+)_{org}$ ratios than the ones that have been empirically used in the "general" elemental analysis described by Aiken et al. (2008). A correction of 34% increase in O:C and 17% increase in H:C was applied based on Canagaratna's correction formula (2013). The contributions of organonitrates and organosulfates, detected as inorganic nitrate or sulfate ions, to the elemental ratios were negligible because their low mass concentrations.

B. Other concurrent measurements and comparisons among measurements

Instruments making measurements during AMAZE-08 at the TT34 site are listed in Martin et al. (2010). The size distribution of particles between 0.010 and 0.48 μm (mobility diameter) was measured every 5 min by a Scanning Mobility Particle Sizer (Lund SMPS) (Roldin, 2008). Particle volume concentrations were calculated from the SMPS size distributions for an assumption of spherical particles. The total number concentration for particles greater than 0.010 μm was measured every 3 s by a Condensation Particle Counter (CPC, TSI 3010). Particle scattering coefficients at multiple wavelengths were measured every 1 min by a nephelometer (TSI 3563) and averaged to 10 min. The light absorption at 637 nm of deposited particles was measured every 1 min by the Multiangle Absorption Photometer (MAAP, Thermo 5012). These several instruments sampled through a laminar-flow line (i.e., separate sampling from the AMS line) that was characterized by lower and upper limits of transmission for particle diameters of 0.004 and 7 μm, respectively (Martin et al., 2010).

Several particle filter samples were collected (Artaxo et al., 2013). Total-particle filters (TPF; PM₃) were collected in-line with the turbulent inlet used by the AMS. Stacked filter units

(SFU) were installed separately at 10 m to sample fine- (PM_2) and coarse-mode particles (PM_{2-10}). The two types of filters show reasonable agreement. The fine-mode data from SFU are reported herein. Filter samples were analyzed by ion chromatography (IC) for water-soluble ionic components, including sulfate, nitrate, and ammonium, among other components. The filters were also analyzed by particle-induced X-ray emission (PIXE) for elemental composition. Concentrations were adjusted to STP conditions.

The AMS data can be compared to other concurrent measurements of AMAZE-08. The mass ratio of NR-PM₁ measured by the AMS to PM₂ by filter assays was 0.65 as a campaign average (Table S2). The ratio was less than unity because PM₂ included contributions by black carbon and mineral dust (Sect. 3.1) as well as organic material in the diameter range of 1 to 2 μ m (P öschl et al., 2010). Particle mass-diameter distributions obtained from gravimetric analysis of stages of a Multi-Orifice Uniform Deposit Impactor (MOUDI) showed that an average of 30% of the particle mass concentration was associated with diameter range from 1 to 2 μ m (cf. Fig. 16 in Martin et al. (2010)).

Figure S2a shows a line of slope *m* of 1.24 and correlation R^2 of 0.81 (cf. also Figure S5 for temporal series) in a scatter plot between the AMS-calculated and the SMPS-derived particle volume concentrations. Figure S2b shows the scatter plot of the number concentrations obtained by integrating the SMPS measurements and those directly measured by the CPC. The slope of 0.6 ($R^2 = 0.90$) indicates that the CPC measured more particles than the corresponding SMPS-derived quantity. The SMPS bias to particle undercounting can explain m > 1 in the scatter plot of Fig. S2a. The scatter plot between sulfate particle mass concentrations measured by the AMS and those measured by IC analysis of the filters is fit by a line of m = 0.90 and $R^2 = 0.50$ (Fig. S2c).

The combined AMS and SMPS data sets were used to estimate the particle effective density ρ_{eff} (kg m⁻³) based on an in-common mode diameter (Katrib et al., 2005). For nonporous spherical particles, material density ρ has the same values as ρ_{eff} , and this condition was assumed to hold in the performed data analysis. The organic material density ρ_{org} was then derived by assuming volume additivity and by using ρ_{inorg} of 1780 kg m⁻³ as ammonium bisulfate for the inorganic components. The estimated campaign-average value of ρ_{eff} for submicron Amazonian particles is 1390 ± 150 kg m⁻³. Figure S3 shows one example of the mass-diameter distribution measured by the AMS compared to that derived from the SMPS measurements. Assuming that the chemical components either do not mix or alternatively have a numerically small excess volume of mixing, we can derive ρ_{org} of 1270 ± 110 kg m⁻³ based on the campaign-average chemical composition and a density of 1770 kg m⁻³ for all inorganic components (Cross et al., 2007). The value of ρ_{org} is consistent with the density of 1200-1500 kg m⁻³ observed for laboratory-generated biogenic secondary organic material (Bahreini et al., 2005; Shilling et al., 2009; Chen et al., 2012).

Figure S2d shows the linear regression of the light scattering derived from the AMS (PM₁) and the nephelometer measurements (PM₇), all for 550-nm wavelength. During the period of 22 February to 3 March 2008, the ratio of the AMS volume concentration to the nephelometer scattering is high (Fig. S5b). Elevated mass concentrations of mineral dust are observed by the lidar measurements (Baars et al., 2011) and the filter-based PIXE analysis (Prenni et al., 2009). Furthermore, local wind and Hysplit back trajectories showed a Manaus plume on March 1, 2008.The elevated scattering is, therefore, plausibly a combination of African advection and Manaus plume influence although the coarse-mode contribution from mineral dust is the major driven force of the a weak correlation ($R^2 = 0.21$) between the nephelometer and AMS dataset. In

contrast, a strong correlation (m = 0.62; $R^2 = 0.82$) of the two data sets is shown for other periods, suggesting a dominant contribution of the non-refractory submicron volume to the total particle scattering. This non-refractory submicron volume is mainly organic material. The scattering coefficients related to the submicron organic material can go up to 6 Mm⁻¹ at 550 nm.

C. Positive-Matrix Factorization

Positive-matrix factorization (PMF) is a receptor-based model using a weighted least squares method to identify patterns in data. With caveats, it can be a useful tool to derive the source profiles of organic components from AMS data sets (Ulbrich et al., 2009). In this study, the PMF analysis was conducted on the V-mode organic UMR spectra (m/z 12 to 220). The spectra were analyzed using the SQUIRREL tookit. Prior to PMF analysis, the data set was prefiltered to remove inorganic contributions, and the analysis was carried out only on the residual data set of the organic component. Fifteen m/z values were omitted because of the absence of organic ions. The time periods associated with random spikes, abrupt increase in sulfate mass concentrations, and little temporal variation caused by the instrument adjustments (Fig. 1) were removed. The error values were calculated using the method described by Ulbrich et al. (2009). Fragments having signal-to-noise ratio less than 2 and fragments set proportionally to m/z 44 were downweighted by increasing their error estimates (Ulbrich et al., 2009). $C_x F_y$ ions contributed significantly to the signals at m/z 69, 119, 131, 169, 181, and 219, indicating the contamination of Fomblin lubricating oil, possibly from instrument pumps at the site (Cross et al., 2009). These signals appeared always as one statistical factor, with a spiky time series for the loading of that factor. These fragments were downgraded by increasing their error by 100 times. The PMF analysis was conducted with (1) different model error and (2) different seed number. The former was introduced to add modeling uncertainty to the instrumental uncertainty,

reflecting the errors that may occur when the true factors do not have constant mass spectra. The latter represents the pseudo-random starting values. Unless otherwise noted, results are presented for both the model error and the seed number of zero. PMF produces a fit to the data, which is called a solution. The solution contains a set of factors and concentrations. For AMAZE-08, four statistical factors were identified and labeled as HOA, OOA-1, OOA-2, and OOA-3 (Fig. 4). The four factors HOA, OOA-1, OOA-2, and OOA-3 respectively accounted for 2%, 18%, 14%, and 66% of the variance in the data matrix, implying a residual variance of <1%.

Number p of factors

Several mathematical metrics were used to set the number *p* of factors. The ratio $Q:Q_{exp}$ of the sum of the squares of the uncertainty-weighted residuals to the expected values decreased by 16%, 8%, 3%, and 3% for *p* increasing from 2 to 5. Three or more factors therefore significantly account for the variance of data. The residual was 1% for *p* = 2 or 3 and < 0.3% for *p* = 4. Structure in the residual was significantly reduced by increasing from *p* of 3 to 4 (Fig. S11). For these reasons, a choice of *p* = 4 was made for the PMF analysis.

The choice of p = 4 was also evaluated with respect to factor similarity and correlations of the time series of the factors. Increasing the p from 4 to 5 resulted in strongly correlation ($R^2 =$ 0.96) among the factors (Fig. S12). No sufficient information from the correlations with other tracers exists to anticipate this correlation; correlation among factors for p of 5 is believed to arise from a splitting of real factors.

Rotational ambiguity of solutions (FPEAK).

FPEAK is the rotational parameter. For simplicity, FPEAK = 0 was used as the best representation of the PMF solution for this study. The PMF solution was evaluated for uniqueness under linear transformations ("rotations") by varying the *FPEAK* parameter (Ulbrich

et al., 2009). Solutions with *FPEAK* between -0.6 and 0.6 increase $Q:Q_{exp}$ by 1%. Figure S13 shows the time series of factor concentrations over this *FPEAK* range. Changes in time series are relatively small compared to the changes in the features of the factors. The largest change is for the HOA factor. This factor accounts for a low fraction of the total signal and hence its features can change without causing a great increase in the residual. The rotational uncertainty causes no conflicts in the interpretation of the PMF factors (Sect. 3.2).

Uncertainty of the solutions.

The results of running the PMF analysis for different pseudo-random starting values (i.e., seeds of 0 to 10) show negligible changes in the factors ($R^2 > 0.999$; m > 0.995) and the time series of the concentrations ($R^2 > 0.999$; m > 0.95). Testing a "model error" of 5% in the PMF analysis leads to changes in the factor profiles ($R^2 > 0.80$; m > 0.90) and in the time series of concentrations ($R^2 > 0.95$; m > 0.75) that are close to tolerance.

Quantitative assessment of the uncertainty of the factors is also made by 100 bootstrapping runs (Ulbrich et al., 2009). The results show that the uncertainties in the time series of the concentrations are 15% for the OOA-2 and OOA-3 factors and 30% for the OOA-1 and HOA factors. The uncertainties in the factor spectra are <4% for OOA-2 and OOA-3 and <9% for OOA-1 and HOA. The mass spectrum of the HOA factor has the largest uncertainty (Fig. S14).

D. Mass spectral markers of biogenic secondary organic material

In the framework of semivolatile partitioning, changes in the chemical composition of secondary organic material with particle-phase organic mass concentration $M_{\rm org}$ are expected: only the least volatile oxidation products can effectively condense to the particle phase at low $M_{\rm org}$ (Donahue et al., 2006). The mass spectra of secondary organic material (SOM) are hence

loading-dependent. For the purpose of comparing the mass spectra of chamber data with the AMAZE-08 data, SOM production from the oxidation of a C₅-C₁₀-C₁₅ terpene sequence, including the photooxidation of isoprene, the dark ozonolysis of α -pinene, and the dark ozonolysis of β -caryopyhllene, has been studied systematically over a range of M_{org} in the Harvard Environmental Chamber at low NO_x and moderate RH conditions (Chen et al., 2011).

Figure 6 shows the loading-dependent mass spectra of the three types of SOM. The fragmentation pattern extends to higher m/z for the increasing carbon skeleton of precursor VOCs. Signals at m/z < 60 account for 93-97%, 80-84%, and 71-79% of the total signal intensity for the isoprene, α -pinene, and β -caryophyllene SOM, respectively. For all three types of SOA, the relative intensities of major C_xH_y ions such as CH_3^+ at m/z 15, $C_2H_3^+$ at m/z 27, $C_3H_3^+$ at m/z 39, and $C_3H_5^+$ at m/z 41 are typically smaller at lower M_{org} . In contrast, the relative intensity of CO_2^+ at m/z 44 increases with decreasing M_{org} . Moreover, the relative intensity of $C_2H_4O^+$ (colored as purple in Fig. 6) increases with increasing M_{org} , accounting for about 10 to 50%, 3 to 5%, and 5 to 10% of the signal at m/z 44 for the isoprene, α -pinene, and β -caryophyllenethe SOM, respectively.

The mass spectra of isoprene SOM is consistent with the spectra collected in a batchmode chamber experiments under low-NO_x and dry conditions (Chhabra et al., 2010), showing a unique pattern compared to the spectra of the other two SOMs. The spectra of the isoprene SOM are dominated by CHO⁺ (m/z 29) and C₂H₃O⁺ (m/z 43). The two ions both show a decreasing trend for decreasing M_{org} . The relative intensities of CH₂O⁺ at m/z 30 and CH₃O⁺ at m/z 31 are small in the spectra of the α -pinene and β -caryophyllene SOM, and however, are five times greater in the spectra of the isoprene SOM. Moreover, $I_{44}:I_{\text{org}}$ has been used as a surrogate of O:C, where $I_{m/z}$ represents the absolute signal intensity at a m/z value (Aiken et al., 2008; Ng et al., 2010). Our data show that the isoprene SOM does not follow the empirical relationship between O:C and I_{44} : I_{org} (Fig. S10). Deriving O:C from I_{44} : I_{org} requires careful judgments on the contribution of isoprene SOM.

A general marker for the three biogenic SOM is identified as $C_7H_7^+$ at m/z 91 distinct from adjacent ions. The prominent $C_7H_7^+$ companied with a negligible signal of $C_6H_{13}^+$ at m/z 85 is also unique for the three biogenic SOA studied in the chamber. In urban environment, ions $C_xH_y^+$ adjacent to $C_7H_7^+$ typically have greater intensities and $C_6H_{13}^+$ has greater relative intensity than $C_7H_7^+$ (cf. Figure 9 in Zhang et al. (2005)). Moreover, $C_5H_6O^+$ at m/z 82 is more prominent compared to the adjacent peaks in the spectra of the isoprene SOM than in the spectra of other two types of biogenic SOM. This ion is suggested as a marker for the SOM originated from isoprene (Robinson et al., 2011). **Table S1.** Summary of the regression coefficients m of instrument comparisons. Expected m values are shown in parentheses. These values are estimated on the basis of the diameter domain of the various instruments and an assumed AMS collection efficiency of 1.0, in conjunction with typical mass distributions obtained by MOUDI measurements during the wet season in the Amazon basin (Martin et al., 2010; Pöschl et al., 2010).

	Volume concentration	Number concentration	Sulfate mass concentration	Particle mass concentration	Light scattering at 550 nm
	SMPS	CPC	Filter-based	Filter-based	Nephelometer
AMS	1.24 (1.0)	-	0.90 (1.0)	0.65 (0.7)	0.62 (< 1.0)
SMPS	-	0.59 (< 1.0)	-	-	

Table S2. Summary of the particle mass concentration ($\mu g m^{-3}$, STP) measured by the stacked filter units on the 10-m inlet (SFU), by the total-particle filter on the 38-m turbulent inlet (TPF), and by the AMS during AMAZE-08.

Sampling Periods (MM/DD/YY)	SFU: PM ₂	TPF: PM ₃	AMS: NR-PM ₁	AMS/SFU	AMS/TPF
02/10/08 - 02/14/08	2.51	n.a.	1.75	0.70	n.a.
02/14/08 - 02/16/08	1.41	1.20	1.37	0.97	0.87
02/16/08 - 02/19/08	0.87	1.50	0.89	1.02	
02/19/08 - 02/22/08	1.14	1.48	0.85	0.75	0.57
02/22/08 - 02/26/08	2.44		0.85	0.35	
02/26/08 - 02/29/08	3.20	2.86	0.96	0.30	0.33
02/29/08 - 03/04/08	3.38		0.99	0.29	
03/04/08 - 03/08/08	1.02	1.02	0.73	0.72	0.72
03/08/08 - 03/12/08	1.01	1.09	0.74	0.73	0.68
Average	1.89	1.55	1.02	0.65	0.63

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List of Figures

Figure S1. Map of the sampling site.

- Figure S2. Scatter plots among data sets. (a) AMS and SMPS volume concentrations for a particle material density calculated by using component densities of 1270, 1780, 1720, and 1520 kg m⁻³ for organic material, ammonium bisulfate, ammonium nitrate, and ammonium chloride respectively and assuming volume additivity. The AMS data in this plot were averaged to the SMPS timebase. (b) SMPS and CPC total number concentrations. The CPC data were averaged to the SMPS timebase. (c) AMS and filter-based IC/PIXE sulfate mass concentrations. The AMS data were averaged to the periods of filter collection. The filter data include SFU PM₂ and TPF PM₃. (d) AMS-derived and nephelometry-measured light scattering. For the AMS analysis, the measured size distributions were used as input to Mie calculations (see main text). The nephelometer data were averaged to the same periods as the AMS mass-diameter distributions. Valued in all panels are normalized to STP.
- **Figure S3.** Example of the mass-diameter distribution measured by the AMS compared to that derived from the SMPS measurements. The effective particle density ρ_{eff} is determined by the mode diameters. The SMPS mass-diameter distributions were derived by multiplying the SMPS volume-diameter distributions by ρ_{eff} . Data were sampled on March 11, 2008.
- **Figure S4.** Campaign-average pie chart of the composition of submicron particles, including the estimated contribution by mineral dust.

Figure S5. (a) Time series of the particle volume concentrations obtained by the AMS and the

SMPS measurements. (**b**) The ratio of the particle volume concentrations derived from the AMS measurements to the PM_7 light scattering coefficients measured by the nephelometer at 550 nm. The AMS data were averaged to the nephelometer timebase. Gray areas represent the periods that were influenced by the generator exhaust plumes.

- **Figure S6.** Scatter plot of the mass concentrations (STP) of components derived from the AMS measurements.
- **Figure S7.** Example of the variations of component concentrations and O:C ratios before and after a rain event.
- **Figure S8.** Time series of the relative intensity of m/z 60 in the organic mass spectra. The peak at m/z 60 is a fragment of levoglucosan and related species and can serve as a marker of biomass burning. The red line represents the threshold value of 0.35% that corresponds to natural background (Docherty et al., 2008).
- Figure S9. Mass spectrum of OOA-3 compared to the synthetic mass spectrum obtained from a linear combination of the mass spectra (cf. Fig. 6) of laboratory-generated biogenic SOM (30% α-pinene SOM, 20% β-caryophylene SOM, and 50% isoprene SOM).
- **Figure S10.** Scatter plot of O:C versus I_{44} : I_{org} for biogenic secondary organic material produced in the Harvard Environmental Chamber. Also shown is the empirical relationship between O:C and I_{44} : I_{org} presented in Aiken et al. (2008) for Mexico City. Marker size corresponds to mass concentration for the laboratory measurements.
- **Figure S11.** Time series of the model residuals e_{ij} for the PMF analysis with *FPEAK* of zero. Terms include factor *j*, time *i*, and error σ .

Figure S12. Pearson's R for the correlations between the time series and the mass spectra of any

two factors for the PMF solutions for different number of factors.

Figure S13. Four-factor solutions for selected *FPEAK* values.

Figure S14. The mass spectrum of HOA with bootstrapping analysis.



Figure S1



Figure S2



Vacuum Aerodynamic Diameter, d_{va} [nm]





Figure S5



Figure S6



Figure S7







Ambient

- Mexico City Airplane
 Mexico City Ground (Aiken et al., 2008)
 12-h average AMAZE

Chamber

- α -pinene ozonolysis
- β -caryophyllene ozonolysis isoprene photooxidation

Figure S10



Figure S11



Figure S12



Figure S13

