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Supplement of

Aircraft observations of the chemical composition and aging of aerosol in the Manaus urban plume during GoAmazon 2014/5

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S1 Calculation of Δorg/ΔCO

As described in the main manuscript text, we find no change in Δorg/ΔCO, particularly on March 13 2014, as the Manaus plume is transported downwind and ages. Other aging metrics, such as the aerosol carbon oxidation state are observed to increase, indicating plume aging. In an effort to ensure this observation is robust, we calculated Δorg/ΔCO in several different ways. In order to test the sensitivity of the calculation to the chosen background organic aerosol and CO concentrations, we also vary these quantities. In total, we calculated Δorg/ΔCO for the March 13 data in 16 different ways, using a combination of different calculation methods and backgrounds. The results of five of these calculations, which span a range of calculation methods and backgrounds, are shown for the March 13, 2014 flight in Figure S1. As seen in Figure S1, none of these methods shows a trend in Δorg/ΔCO with plume aging. We next describe the details of the calculation methods, the different background values of org and CO that were used in calculations, and the rationale for choosing these backgrounds and end with some general conclusions regarding the calculations.

1) Method 1 (M1 on Figure S1) – The AMS-measured organic aerosol concentrations were plotted against the measured CO concentrations. All data for each flight leg perpendicular to the wind direction were included. Background values of org and CO were not subtracted from the data. Δorg/ΔCO was calculated as the slope of the linear regression that was not forced through the origin.

2) Method 2 (M2) – Uses the same methodology as M1 except a background value of org=0.33 μg/m^3 and CO=80.5 ppbv were subtracted from each OA and CO measurement. The linear regression was not forced through the origin.

3) Method 3 (M3) - Δorg/ΔCO calculated using the arithmetic mean as: $\frac{\Delta org}{\Delta CO} = \frac{(\overline{org}_{plume} - \overline{org}_{background})}{(\overline{CO}_{plume} - \overline{CO}_{background})}$. Background values of CO and org are different for each leg and chosen as the mean of the quantities at the beginning and end of each leg (i.e., furthest from the plume).

4) Method 4 (M4) – same as M3, but with fixed backgrounds of org=0.33 μg/m^3 and CO=80.5 ppbv subtracted from the mean of plume quantities for each leg.

5) Method 5 (M5) - same as M2 but with backgrounds of org=0.53 μg/m^3 and CO=87.7 ppbv subtracted from each OA and CO measurement.

The additional calculations not explicitly described are permutations of the linear regression method employing different background values for CO (80.5, 83, 87.7 ppbv) and organics (0.33, 0.53 μg/m^3) and either forcing or not forcing the regression through the origin.

Backgrounds of CO and org were chosen in several different ways, all of which could be considered reasonable. The organic and CO backgrounds of 0.33 μg/m^3 and 80.5 ppbv were the average values obtained when the G-1 was unambiguously above the boundary layer on the March 13 flight. These values are the lowest values observed during any portion of the Marth 13 flight. The organic and CO backgrounds of 0.53 μg/m^{3}and 87.7 ppbv were calculated by averaging the data when the G-1 was transitioning between plume transects (i.e., traveling parallel to the wind and spatially removed from the plume) at 500 m.

These values represent the lowest values measured at 500 m. The CO background of 83 ppbv is the average CO value at 1000 m when the G-1 was transitioning between plume transects.

Our general observations on the Δorg/ΔCO calculation methods are as follows:

1) We see very little sensitivity to using different background org and CO concentrations when using the linear regression method if the regression is not forced through zero. In Figure S1, M1, M2, and M5 return largely the same Δorg/ΔCO values despite using different background values.

2) For this data set, subtracting the background values was not necessary, provided the linear regression was not forced through zero.

3) The choice of background values had the largest influence on Δorg/ΔCO when linear regressions were forced through zero and backgrounds were subtracted.

4) Perhaps this is obvious, but data with smaller Δorg and ΔCO are more sensitive to background subtraction.

5) When linear regressions were not forced through zero, the Y intercept was generally not representative of the background CO concentration.

6) Calculations using the arithmetic mean (methods 3 and 4) were more sensitive than the linear regression method to choice of the background values.

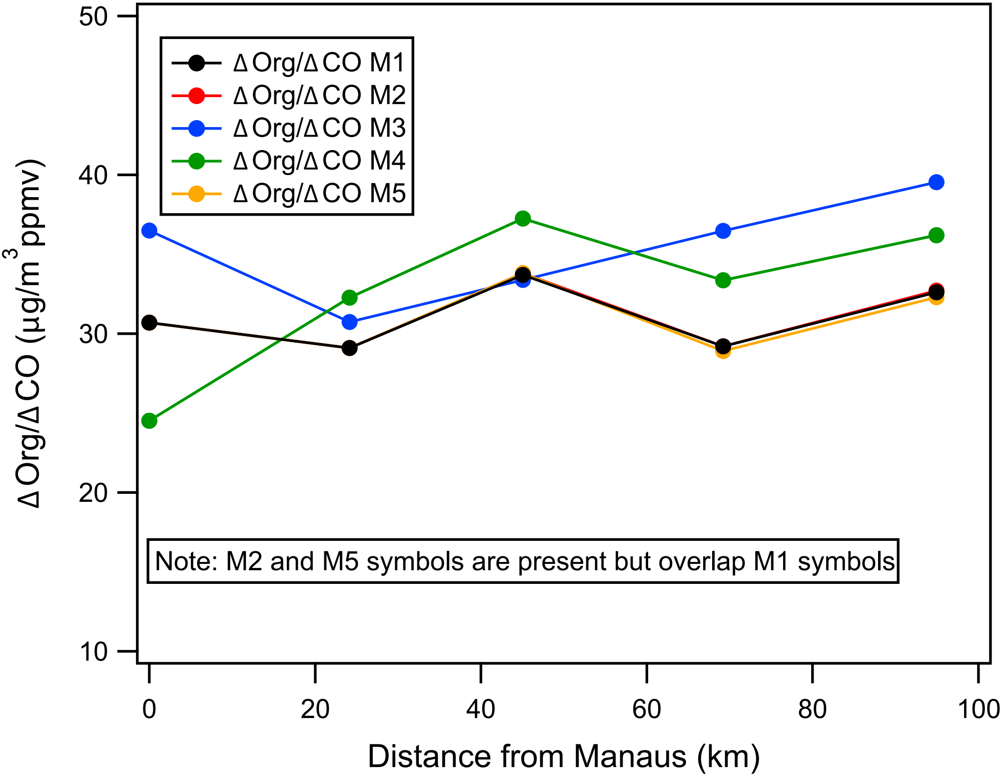

Figure S1. Evaluation of Δorg/ΔCO on March 13, 2014 as a function of Manaus plume age using several different methods. A total of 16 different methods were used to calculate Δorg/ΔCO, with a subset shown here. Note that no method shows a significant change in Δorg/ΔCO with plume age. The different methods for calculating Δorg/ΔCO are describe in the SI.

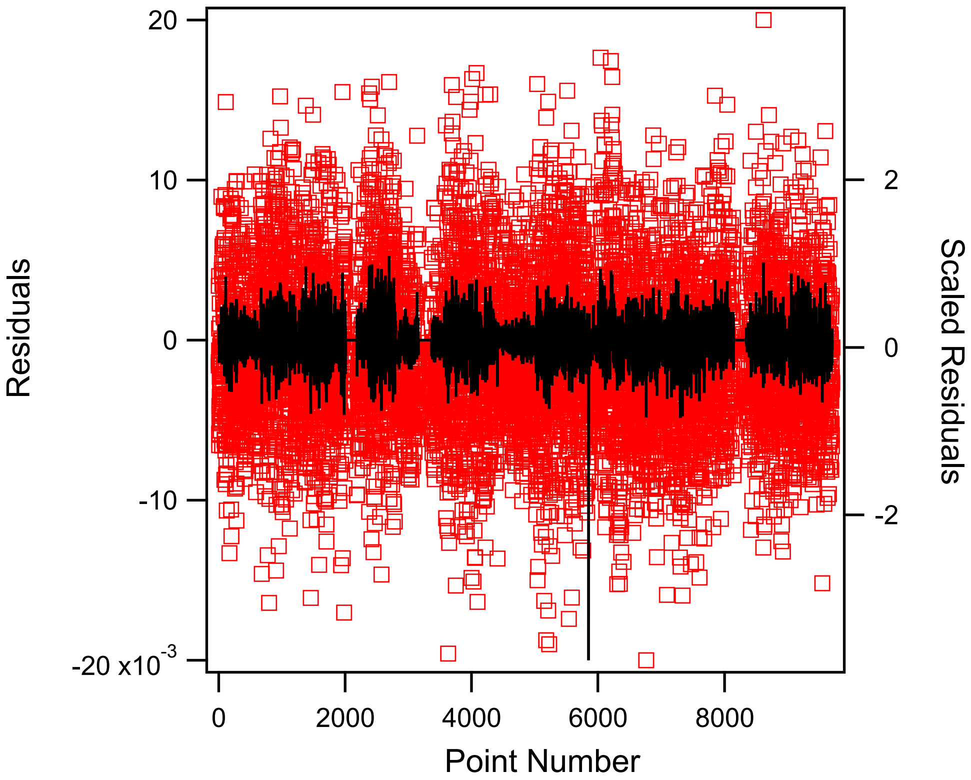

Figure S2. Absolute and scaled residuals for the wet season data matrix.

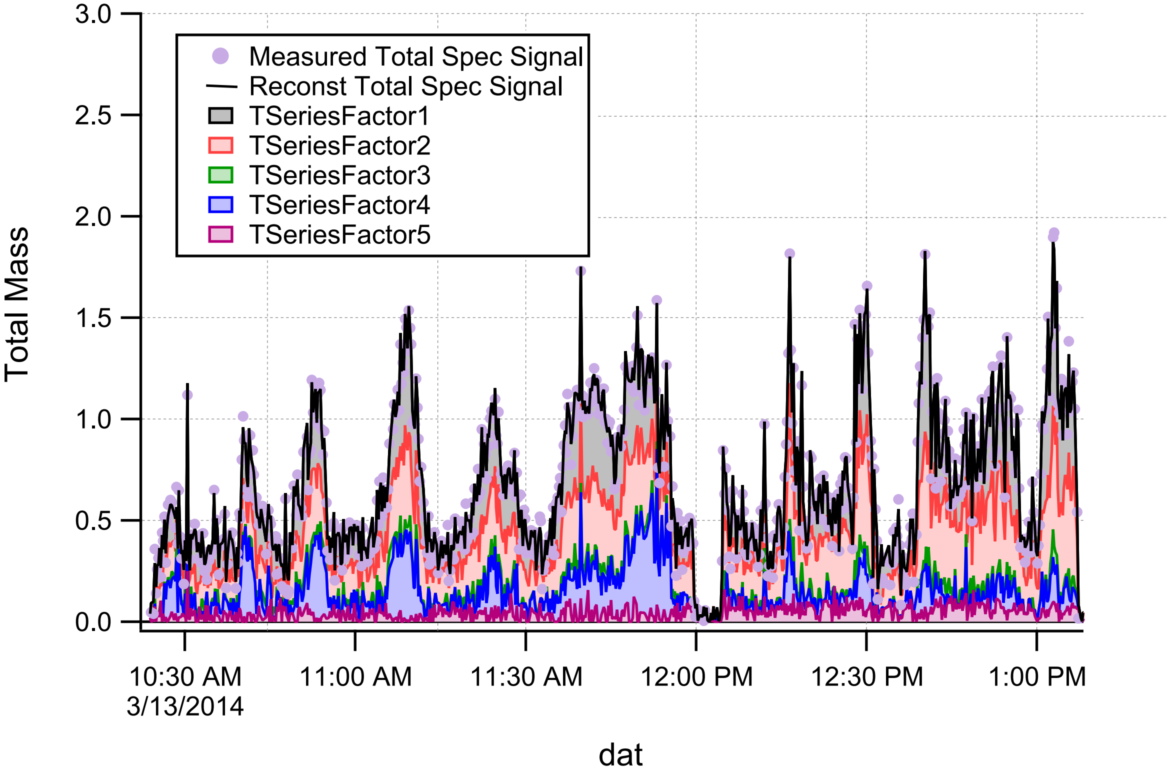

Figure S3. Reconstruction of the mass of the 5 Factor solution chosen for the wet season data set. The total measured mass is shown as the purple dots and the reconstructed mass is shown as a black line. Individual factors are color coded. To make the information legible, we have chosen to highlight the data for the March 13 flight presented in the main text. Factors 1, 2, and 3 were combined into a single OOA factor when reported in the main text as discussed in the appendix.

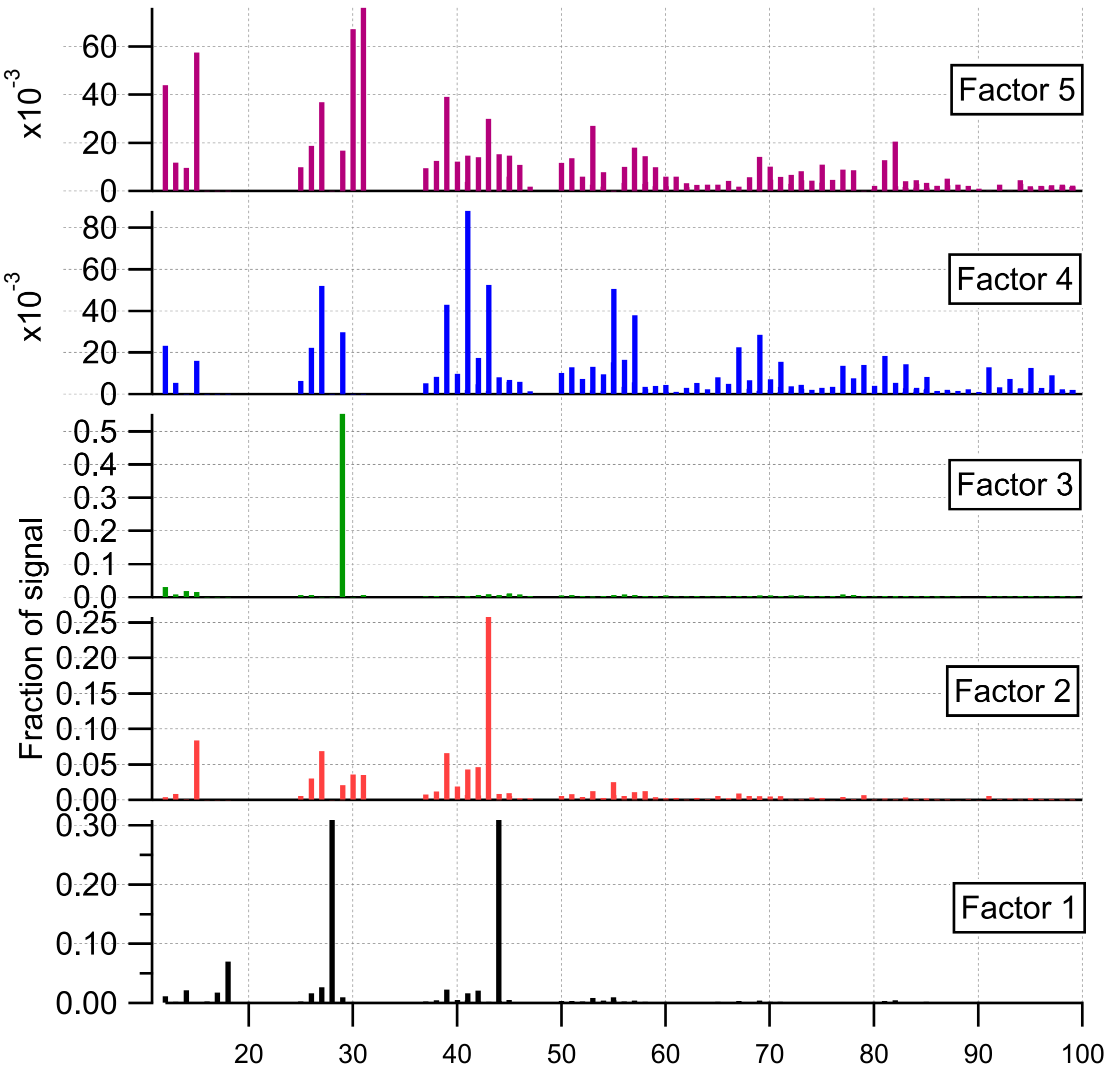

Figure S4. Mass spectral profiles of the 5 factor solution chosen for the wet season data. Factors 1, 2, and 3 were combined when reported in the text as discussed in the appendix.

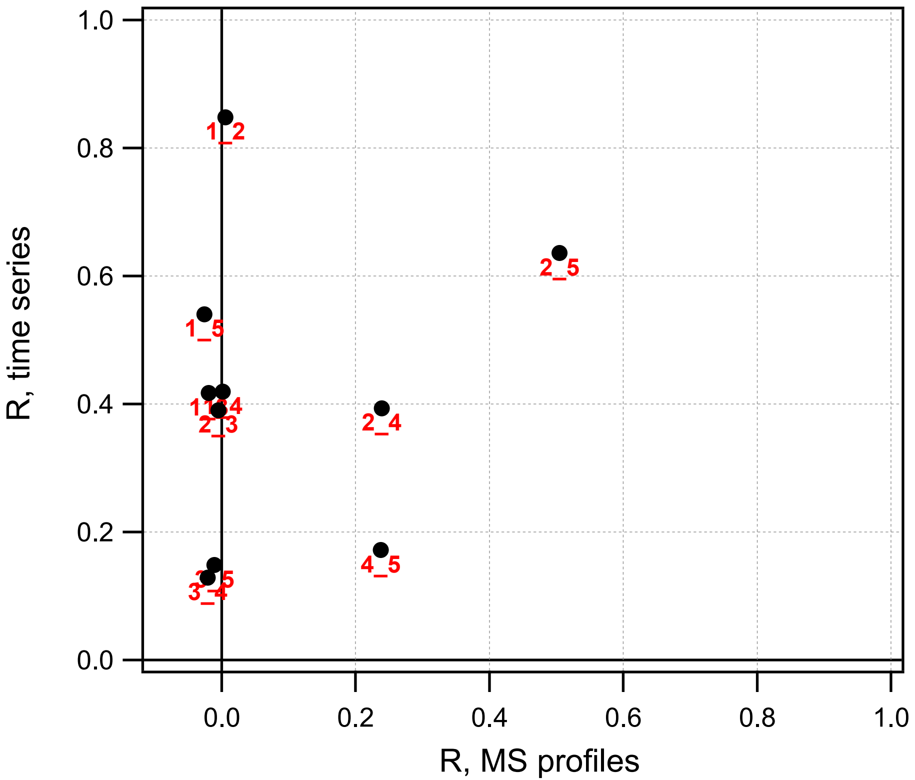

Figure S5. Plot of the cross correlation coefficients between factors for both the the time series and mass spectra of the 5 factor solution.

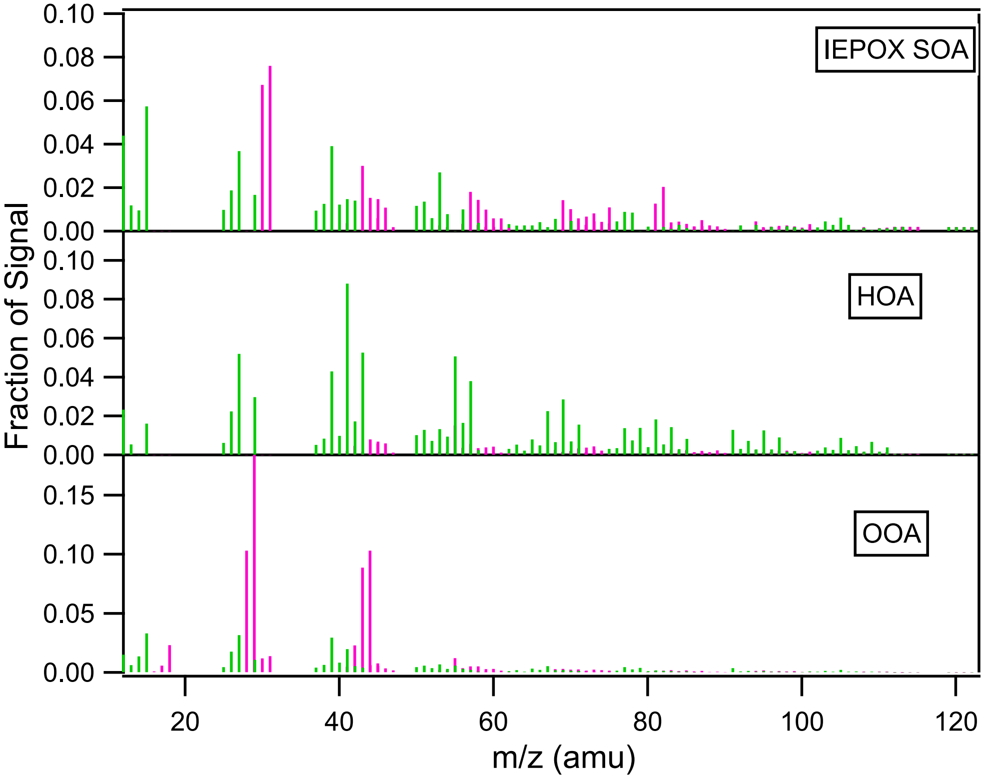

Figure S6. PMF spectra for the three factors reported in the manuscript. Factors 1, 2, and 3 were combined into the OOA factor. The analysis used high resolution AMS data, but the signals are represented as sticks for clarity. Sticks are colored by the chemical composition of the ion; ions in the CxHy family are green and those in the CxHyOz family are pink.