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Assessing the role of anthropogenic and biogenic sources on PM_1 over southern West Africa using aircraft measurements

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Mass concentration comparison between C-ToF-AMS and collocated instrumentation

Aerosol loadings from the C-ToF-AMS were compared against volume integration using a Scanning Mobility Particle Sizer (SMPS) and Black Carbon (BC) from a Single Particle Soot Photometer (SP2), yielding good agreement (slope: 0.87, R2: 0.83, Fig. S1). The density used for each species was 1.78, 1.72, 1.72, 1.52, and 1.77 g cm^{-3} for sulphate, nitrate, ammonium, chloride, and BC, respectively (Holden and Lide, 1991; Park et al., 2004). The density of organics was estimated based on the oxygen-to-carbon (O : C) and hydrogen-to-carbon (H : C) ratios (Canagaratna et al., 2015; Kuwata et al., 2012), yielding a campaign average of 1.67 g cm^{-3}.

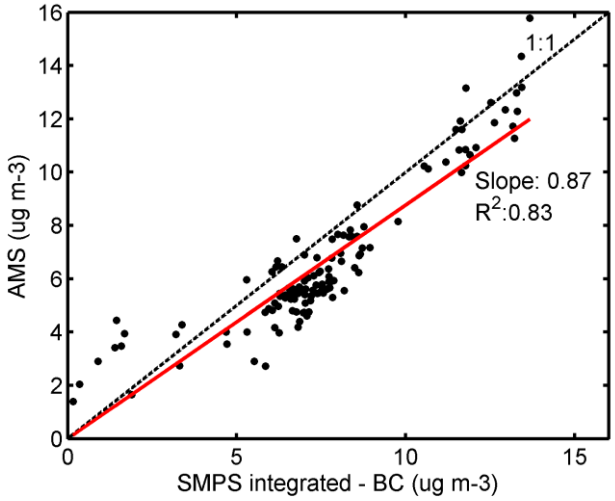

Fig. S1. Sum of AMS species versus integrated SMPS mass minus BC.

Positive Matrix Factorization

Positive Matrix Factorization (PMF) has been conducted on unit mass resolution spectra of organic species for source apportionments. Organic data matrix and error matrix are generated from Squirrel software version 1.57. The PMF Evaluation Toolkit (PET) software is utilized to process the data (Ulbrich et al., 2009). Any “weak” m/z’s (signal-to-noise ratio between 0.2 and 2) are downweighted by a factor of 2, and “bad” m/z’s (SNR smaller than 0.2) are removed (Ulbrich et al., 2009). The PMF solutions for the dataset has been obtained following the detailed procedure described in Zhang et al., (2011).

For the dataset analyzed here, a 3-factor solution is chosen after carefully checking the quality of the fit parameter (Q/Qexp) (Fig. S2 and S3). Solutions with more than 3 factors depict no significant improvement resolving individual m/z's (Fig. S2), as well as display splitting behavior of existing factors instead of providing new factors (Zhang et al., 2011). The rotational ambiguity of the 3-factor solution is examined by varying the FPEAK parameter, displaying an improved correlations with external tracers and reference spectra for FPEAK=-0.4. Combining the distribution of scaled residuals for each m/z (Fig. S2), key diagnostic plots (Fig. S3), PMF solutions with characteristic mass spectral signature (main text Fig. 3), and correlation with external tracers (Fig. S4), we find the 3-factor solution with FPEAK=-0.4 to be the most reasonable and meaningful solution.

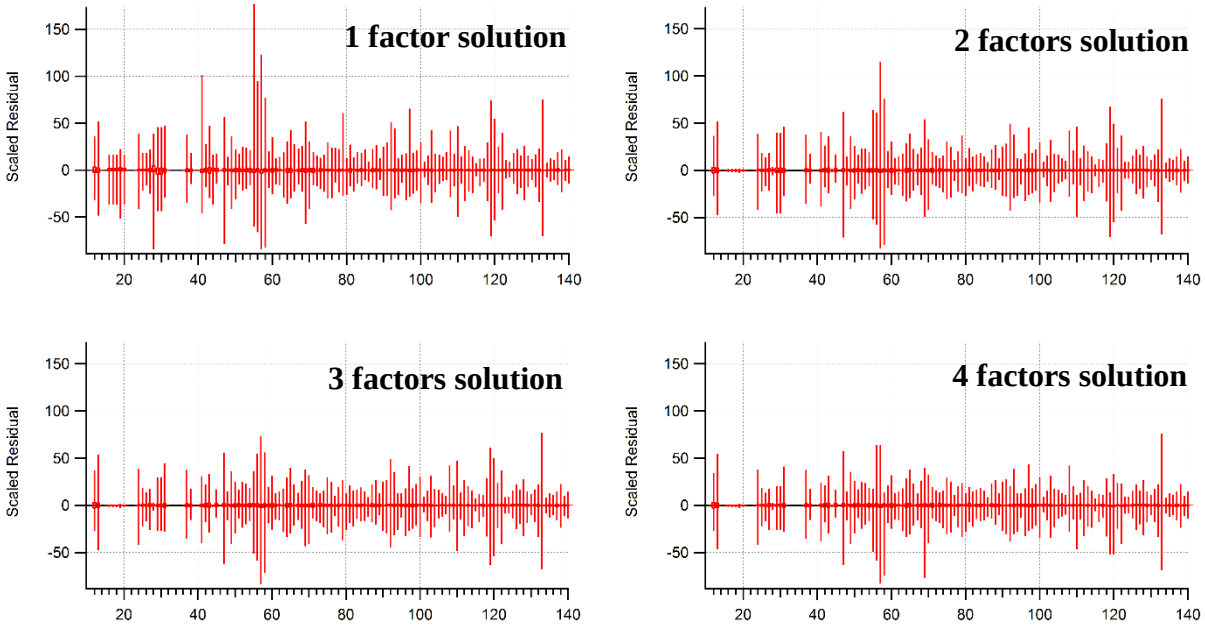

Fig. S2. The distribution of scaled residuals for each m/z according to the number of solutions, indicating no improvement between 3 and 4 factors solution.

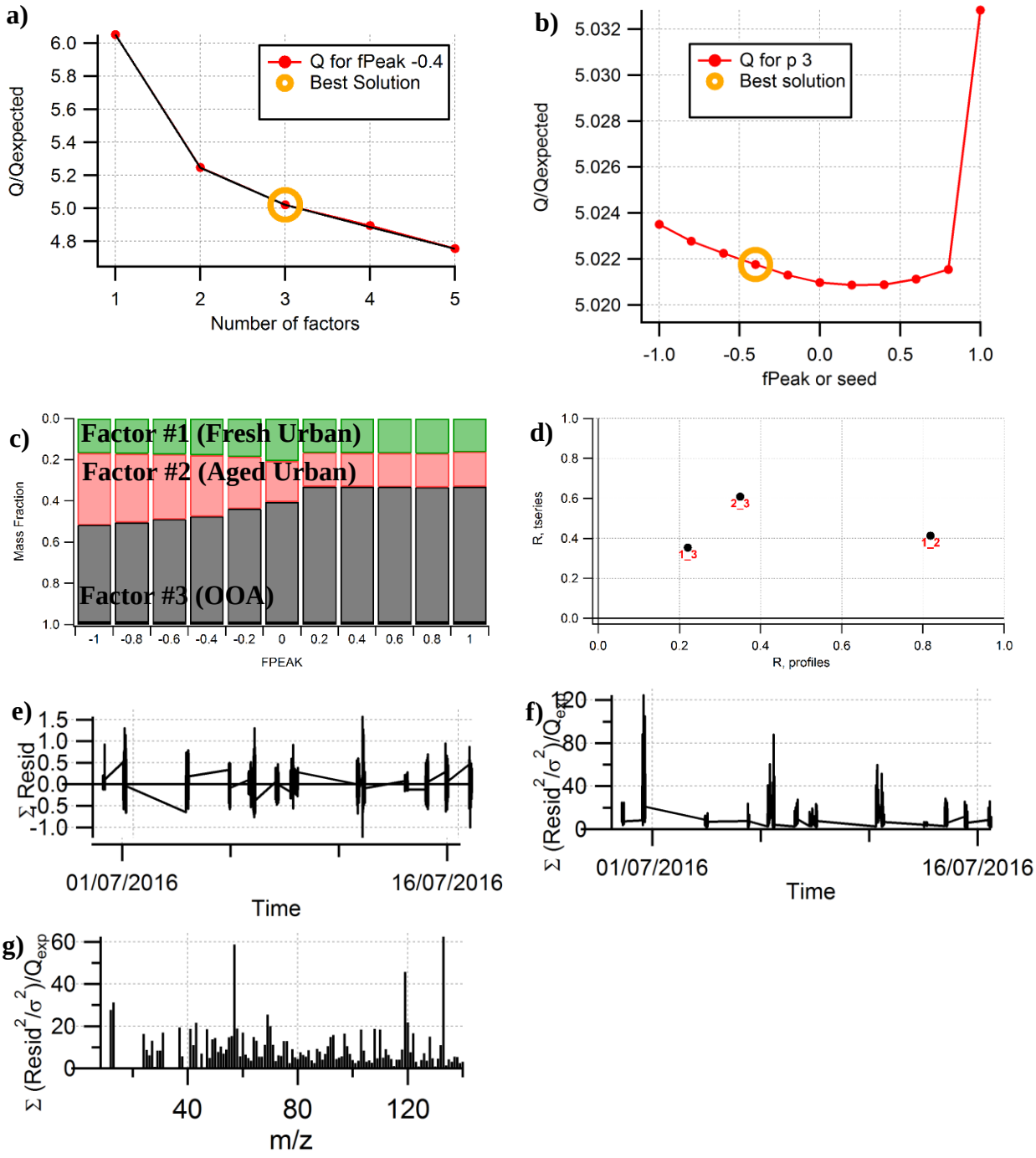

Fig. S3. Summary of key diagnostic plots of the PMF results. (a) Q/Qexp as a function of number of factors. (b) Q/Qexp as a function of FPEAK for the 3-factor solution. (c) Mass fraction of PMF factors as a function of FPEAK. (d) Correlations of time series and mass spectra among PMF factors. (e) Variations of the residual (= measured - reconstructed) of the

least-square-fit as a function of time. (f) The Q/Qexp for each point as a function of time. (g) The Q/Qexp values for each m/z.

Fig. S4. Correlation with external tracers for the 3 factor solution. FPEAK was chosen as -0.4 to maximize correlation with external tracers and reference spectra (referring to Fig. 3 of main text).

Back-trajectories analysis of the case-study of 06 July 2016

Figure S5 shows the back-trajectory analysis of the three flight transects. The back-trajectory was calculated using the ECMWF analysis dataset of the wind field.

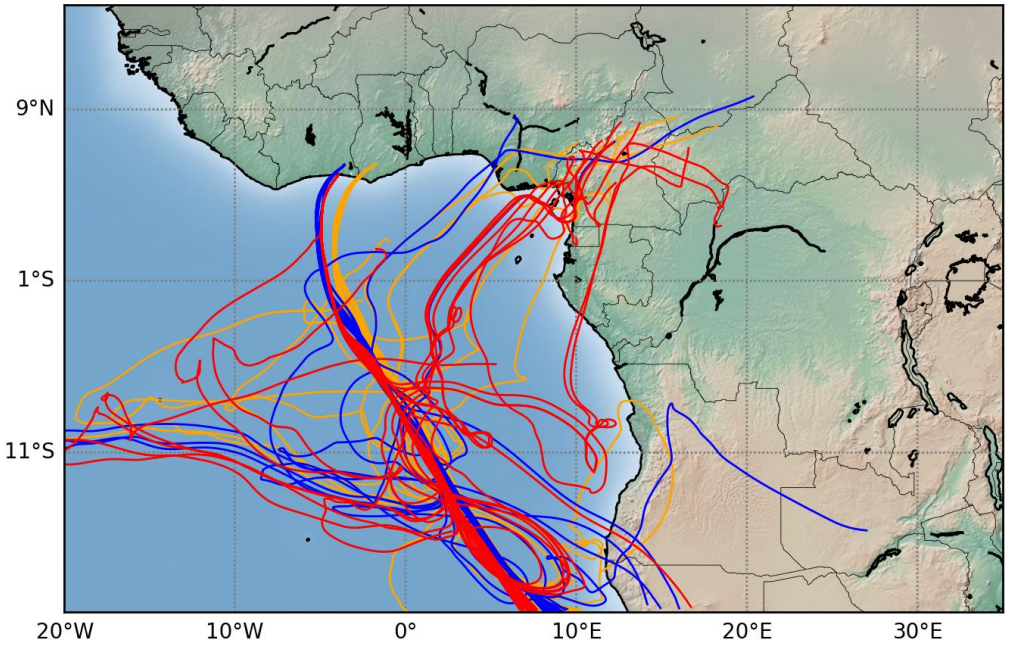

Fig. S5. Back-trajectory analysis of the sampling points of the case study of 06 July 2016. The blue, red and yellow lines represent Upwind Abidjan, within the plume and continental air mass, respectively.

Plume enhancement of BC, CO and NH4 during the case study of 06 July 2016

Figure S6 depicts data for BC, NH4 and CO in a similar fashion as Figure 5 of the main text, complementing the discussion of Section 3.2.

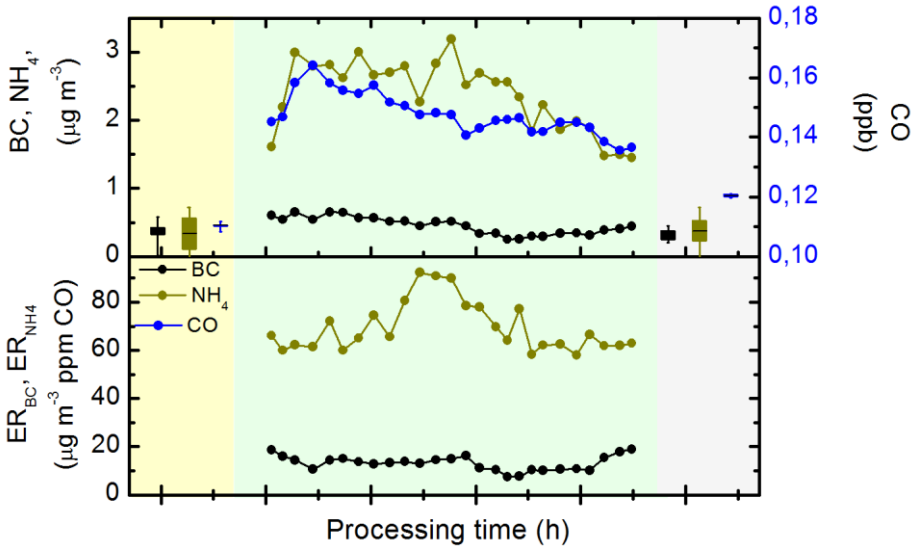

Fig. S6. Concentration (top) and enhancement ratios (bottom) for upwind of Abidjan (yellow), within the plume (green) and sampling regional aerosol (grey). Aircraft

measurements were carried out first sampling Abidjan plume around 09:30, upwind of Abidjan around 10:00, and regional aerosol at 13:30 UTC (identical to local time).

Systematic plume identification using ATR42 data

Figure S7 shows the location of measurement points classified as in-plume according to the systematic analysis described in Section 3.3. Distance between emitting city and sampling point has been calculated by identifying the emitting city according to sampling location.

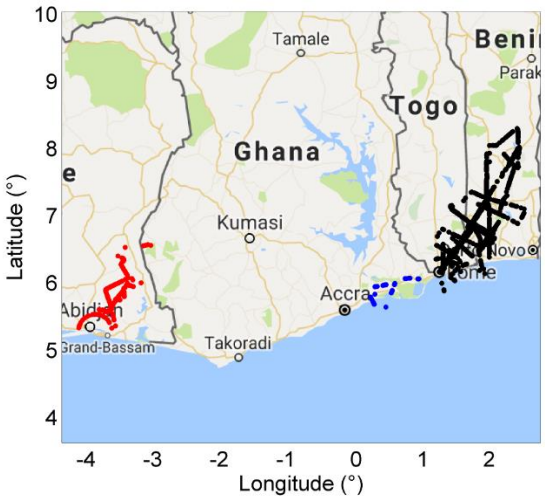

Fig. S7. Location of sampling points identified as in-plume according to systematic analysis. Red, blue and black points refer to Abidjan, Accra and Lomé, respectively.