Black carbon content of traffic emissions impacts significantly on black carbon mass size distributions and mixing states

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1. Particle number size distributions inverted from DMA-SP2 measurements and multiple charging corrections

The aerosol number concentrations at mobility diameter Dp measured by the SP2 represent the number concentration of aerosols in a diameter range with half width of the electrical mobility as $\Delta Z_p = Z_p \frac{Q_a}{Qsh}$, where $Z_p$is the electrical mobility corresponding to Dp, Q_a is the SP2 sample flow (0.1 L/min) and Qsh is the DMA sheath flow (2 L/min). Therefore, if we term the measured aerosol number concentration as ΔN（Dp）, the corresponding $\Delta\log(Dp)$ can be calculated based on ΔZ_p, with the relationship between Z_p and Dp as $Z_p = \frac{neC(Dp)}{3\pi\mu Dp}$, where e is the elementary charge, μ is the gas viscosity coefficient, and $C(Dp)$ is the Cunningham slip correction factor.

The size distributions with only transmission efficiency of single charge particles accounted for can be formulated as:

$$\frac{dN(Dp)}{dlog(Dp)} = \frac{\Delta N(Dp)}{\Delta\log(Dp)}/R,$$

where R is the known transfer efficiency ratio of mobility diameter. Then, multiple charging correction of one-dimensional size distribution can be conducted as described in Zhao et al. (2021).

One the basis of this, the particle number size distributions (PNSD) of BC-free aerosols and BC-containing aerosols are also be derived. Following Zhao et al. (2021), multiple charging corrections were conducted separately for BC-containing and BC-free aerosols. The distribution of BC-containing aerosols could be described using a two-variable function $\frac{\partial N}{\partial\log(Dp)\partial\log(Dc)}$, where Dc is the BC core diameter. The Dc was divided into 30 different bins from 80 to 500 nm, where the Δ log Dc was the same for different bins. For each Dc bin, there was only Dp dimension for the size distribution, therefore, the multiple charging correction can be applied.

2. PMF analysis

An improved source apportionment technique called Multilinear Engine (ME-2) were used to deconvolve organic aerosol (OA) spectra measured by the Q-ACSM into OA factors. Different from traditional PMF, ME-2 offers a coefficient called a-value to constrain the spectral variation extent of OA factor with given priori mass spectra. The unconstrained runs with PMF technique were firstly performed with a possible factor number of 2-8, and diagnostics analysis were shown in Fig.S1, and the three factors solution seems the best solution, and the spectral and time series analysis of factors

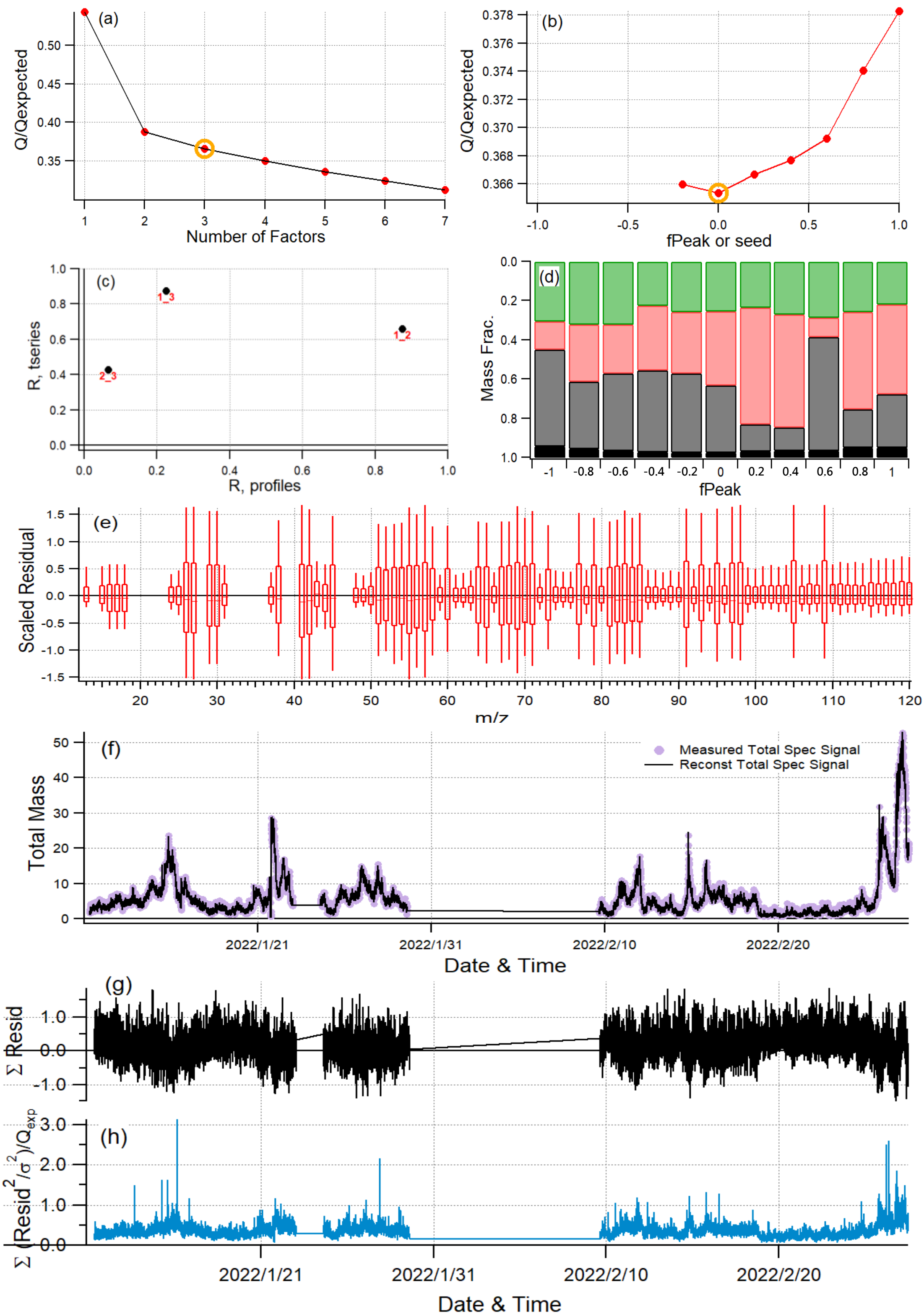

Figure S1. Diagnostic plots of the 3-factor solution in the unconstrained PMF.

are shown in Fig. S2. It shows clearly a primary organic aerosol factor (POA), s less-oxidized oxygenated organic aerosol factor (OOA) and a more-oxidized oxygenated organic aerosol factor (MOOA). However, three factors solution does not split two major primary OA factors of cookinglike OA (COA) and hydrocarbon-like OA (HOA) in urban area. Therefore, we had chosen 4 factors for ME-2 analysis and constrained the COA profile with the *a* value ranging from 0.1 to 0.5. The COA profile reported in Liu et al. (2022) as a proxy was used considering the following three reasons: (1) The used instrument of this study is the same one of Liu et al. (2022); (2) the COA profile reported in Liu et al. (2022) was determined during the period when both COVID-19 silence-action and festival spring occurred when cooking activities grew and traffic activities almost vanished thus COA shall dominated over HOA. More details regarding the method can be referred to Liu et al. (2022); (3) Resolved variations of HOA and COA are well explained by external datasets such as correlations of HOA with black carbon whose correlation coefficient could reach 0.88. The four-factor solution using the ME-2 technique with *a*=0.1 was obtained and was shown in Fig. S3.

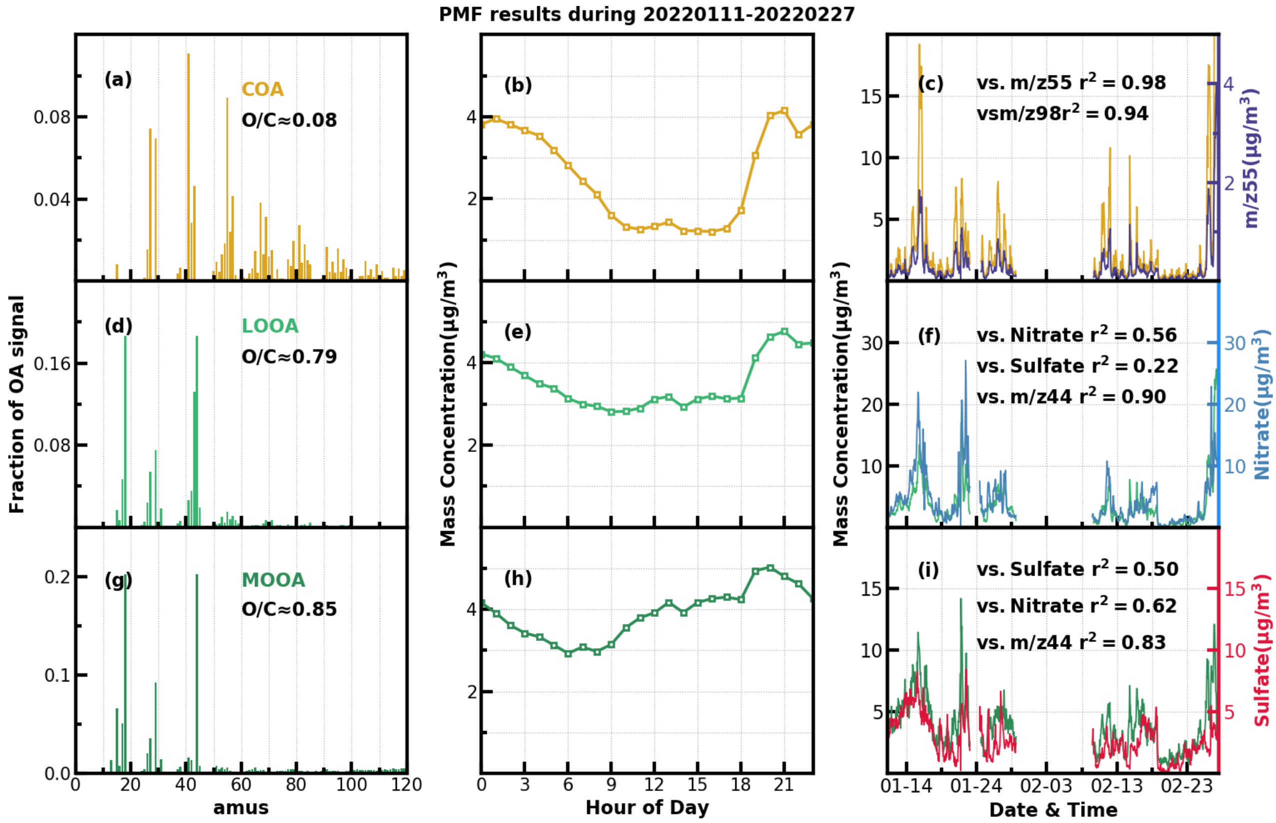

Figure S2. The spectral characteristics, diurnal variations and time series analysis of three factors resolved by the PMF

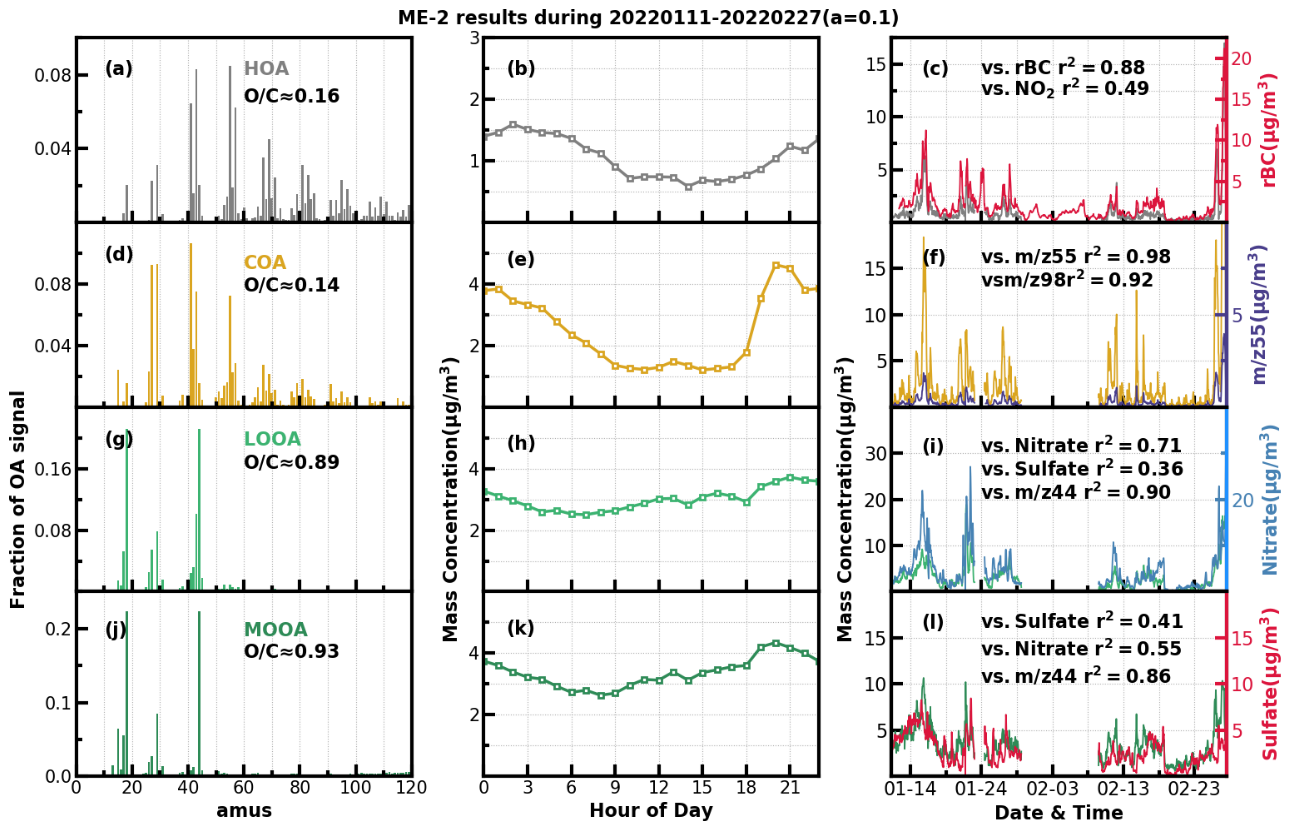

Figure S3. Mass spectral profiles, diurnal cycles and correlations with external data of COA(a-c), HOA(d-f), LOOA(g-i) and MOOA(j-l) from ME2-ACSM analysis.

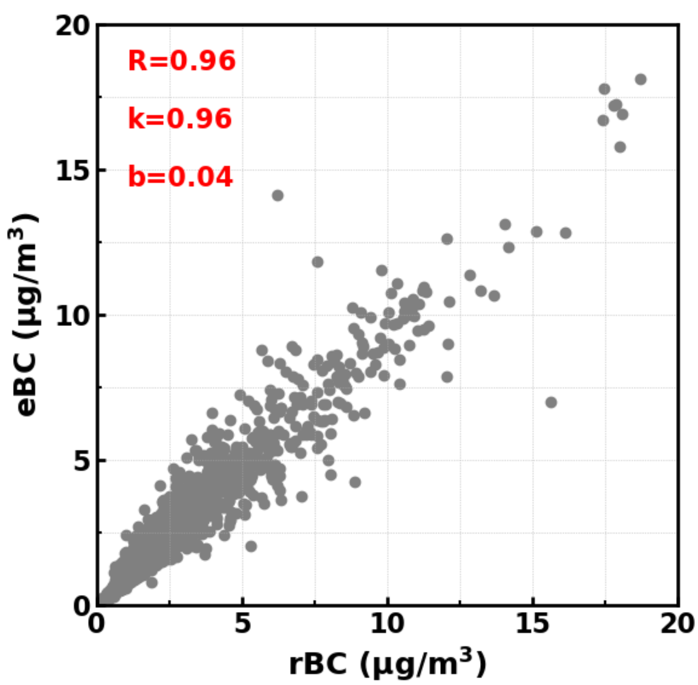

Figure S4. Comparison between rBC derived from DMA-SP2 measurements and optical equivalent BC (eBC) derived from AE33 measurements.

Fiiting form of BCMSD:

$$\frac{dM_{BC}}{dlogDp} = \frac{M_{BC}}{\sqrt{2\pi}\log(\sigma_g)} \cdot \exp\left(-\frac{\left[\log(D_p)-\log(D_{g,BC})\right]^2}{2\log(\sigma_g)^2}\right) \quad (1)$$

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