1 Supplementary Information for

- 2 Primary emissions versus secondary formation of fine particulate matter in the top
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2 Fig. S1. PMF profiles of OA sources for 4-, 5-, and 6-factor solutions.

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Note: The resolved two secondary factors in the 6-factor solution, referred to as "OOA₁" and "OOA₂", have the similar contributions from oxygenated fragment-related m/z (m/z 44) and the strong correlation with each other ($R^2 = 0.72$). The sum of the contributions of OOA₁ and OOA₂ matches the OOA contribution from 5-factor solution ($R^2 = 0.92$ and slope = 1.1).



1 m/Z
2 Fig. S3. ME-2 profiles of OA sources. The COA profile is from that of Crippa et al. (2013), and the

- 3 HOA profile is from that of Ng et al. (2011b). The others are unconstrained factors.





Fig. S4. The relative contributions of OA factors to the m/z's.



Fig. S5. The maps of potential source contribution function (PSCF) analysis for BBOA, CCOA, and

3 00A.