

## Seasonal Variations of High Time-Resolved Chemical Compositions, Sources and Evolution for Atmospheric Submicron Aerosols in the Megacity of Beijing

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This study aims to investigate aerosol secondary formation and aging process in the megacity of Beijing. Seasonal intensive campaigns were conducted from March 2012 to March 2013 at an urban site located at the campus of Peking University (116.31°E, 37.99°N). An Aerodyne high-resolution time-of-flight aerosol mass spectrometry (HR-ToF-AMS) and other relevant instrumentations for gaseous and particulate pollutants were deployed. The average submicron aerosol (PM<sub>1</sub>) mass concentrations were  $45.1 \pm 45.8$ ,  $37.5 \pm 31.0$ ,  $41.3 \pm 42.7$  and  $81.7 \pm 72.4$   $\mu\text{g m}^{-3}$  in spring, summer, autumn and winter, respectively. Organic matter was the most abundant component, accounting for 31%, 33%, 44% and 36% in PM<sub>1</sub> correspondingly, followed by sulfate and nitrate. Distinct seasonal and diurnal patterns of the components of PM<sub>1</sub> tracking primary sources (e.g., BC and HOA) and secondary formation (e.g., sulfate, nitrate, ammonium, LV-OOA and SV-OOA) were significantly influenced by primary emissions and mesoscale meteorology.

Combining positive matrix factorization (PMF) analysis with the mass spectrometry of organics measured by AMS, the contributions of primary and secondary sources to submicron organic aerosols (OA) were apportioned. In spring and summer, the primary sources were hydrocarbon-like OA (HOA) and cooking OA (COA), and the secondary components were low volatility (LV-OOA) and semi-volatile oxygenated OA (SV-OOA). In winter biomass burning OA (BBOA) was also resolved. In autumn, four factors were resolved, that is, OOA, HOA, COA and BBOA. In general, OOA (sum of LV-OOA and SV-OOA) was important in OA in four seasons, accounting for about 63%, 70%, 47% and 50%, respectively. SV-OOA dominated OA in summer (44%) due to the fresh secondary formation from strong photochemical oxidations; whereas, LV-OOA was dominant in OA in winter (33%), maybe because the transported air masses were more aged in heavily polluted days. The POA (sum of HOA, COA and BBOA) in OA was dominant in autumn because primary emissions, such as biomass burning, strongly influenced Beijing and surrounding areas. The evolution processes of OA in the atmosphere are illustrated according to the organic mass to organic carbon ratio (OM/OC), the elemental ratios (O/C and H/C), the average carbon oxidation state, as well as the van Krevelen triangle diagram in detail. Therefore, to prevent regional PM<sub>2.5</sub> and haze pollution effectively, further strengthening the control of primary particulate emissions is expected; in addition the emissions of secondary species' precursors must be reduced, especially in adverse meteorological conditions.

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