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主 論 文 の 要 旨

論文題目 Sources of urban organic aerosol and their relationship with hygroscopicity:
Insights from fractionation analysis

(都市有機エアロゾルの起源およびその吸湿性との関係：分画分析に基づく知見)

氏 名 周 瑞辰 (ZHOU Ruichen)

論 文 内 容 の 要 旨

Atmospheric organic aerosols (OA) play an important role in the Earth's radiative balance and affect human health. However, our understandings of the sources, compositions, and properties of OA are still poor. The environmental impacts of OA depend on their diverse properties, which are related to the composition, are further governed by the sources. Understanding how the sources of atmospheric OA govern its burden is crucial to assess its impact on environments and adopt proper control strategies. Further understanding the relationships between the sources and properties would facilitate the prediction of environmental impacts of OA. In this study, the sources of OA over Beijing were assessed year-around based on the combination of two separation approaches for OA, one from chemical fractionation into the high-polarity fraction of water-soluble organic matter (HP-WSOM), humic-like substances (HULIS), and water-insoluble organic matter (WISOM), and the other from statistical grouping using positive matrix factorization (PMF) of high-resolution aerosol mass spectra. HULIS, a class of unresolved organic compounds with limited influence from inorganics, are suitable material for hygroscopic study of OA as compared with HP-WSOM, which has strong interference from inorganics, and non-hygroscopic WISOM. Understanding the chemical structures and sources that control the hygroscopicity of HULIS is important for our prediction of their environmental impacts. Here, the hygroscopicity parameter of HULIS (κ_{HULIS}) was determined. The relationship between κ_{HULIS} and the structural characteristics represented by O/C ratio is discussed. Further, the contribution of the sources of HULIS to their hygroscopicity are quantified.

Among the three OA fractions, HP-WSOM has the highest O/C ratio (1.36), followed by HULIS (0.56) and WISOM (0.17). Five factors were identified from the PMF analysis, which are fossil-fuel OA (FFOA), cooking-like OA (COA), biomass burning OA (BBOA), less oxidized oxygenated OA (LO-OOA) and more oxidized oxygenated OA (MO-OOA); they contributed 24%, 12%, 6%, 8%, 50% to total organics, respectively. The major sources of different OA fractions were distinct: HP-WSOM was dominated by MO-OOA (96%); HULIS by COA (40%), LO-OOA (27%), and BBOA (21%); and WISOM by FFOA (77%). In addition, the results provide evidence that mass spectral-based PMF factors are associated with specific sub-structures in molecules. These structures are further discussed in the context of the FT-IR results.

The κ_{HULIS} was determined to be 0.06 ± 0.03 and was found to correlate positively with O/C ratio and f_{44} from offline aerosol mass spectrometry. The κ_{HULIS} also correlated positively with OOA, and further analysis resolved substantial κ values of 0.09 and 0.22 for LO-OOA and MO-OOA, respectively, in contrast to the low hygroscopicity of COA and BBOA. Extension of this parameterization well estimates the hygroscopicity of total OA with a moderate contribution from HULIS. The obtained source contributions to the κ values are similar to those from the online analysis of WSOM or total OA in different environments, suggesting their wide applicability.

This study, for the first time, presents an overall relationship of OA groups monitored by chemical and statistical approaches. The bridges between sources and hygroscopicity of OA would provide a basis to facilitate the modeling work of aerosol budget and related atmospheric chemistry and radiative forcing. The approach used in this study may be applicable for further studies of the other aerosol properties to provide better understanding and prediction of environmental impacts of atmospheric aerosol.