



High contributions of fossil sources to more volatile organic carbon

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Radiocarbon analysis on carbonaceous aerosols has been very useful for source apportionment of organic carbon (OC) and elemental carbon (EC). It has been recognized that OC contains organic compounds covering a wide range of volatilities and modeling of organic aerosol is currently widely based on the volatility basis set approach. Due to difference in volatility of OC emitted by various primary sources, the source contributions to OC can be different for each volatility fraction. Moreover, newly formed secondary OC (SOC) and recently emitted primary OC are usually more volatile than OC that has undergone extensive photochemical processing in the atmosphere.

Sources of particulate OC with different volatility have rarely been investigated despite the significant importance for better understanding of the atmospheric processes of organic aerosols. In this work, wintertime PM_{2.5} samples from 6 Chinese megacities are studied. Fossil (e.g., vehicle emissions, coal burning etc.) and non-fossil (e.g., biomass burning, biogenic emissions, cooking etc.) contributions to OC and EC are unambiguously quantified using radiocarbon (¹⁴C) measurement. Further, we describe and evaluate a new method for ¹⁴C source apportionment in more volatile fraction of OC (mvOC) based on thermal desorption from filter samples. In all cities mvOC has a significantly higher contribution from fossil source than total OC. This is consistent with source studies indicating that primary emissions from vehicular exhausts are more volatile than primary emissions from biomass burning. However, the contribution of mvOC to OC in the ambient samples is much smaller than typical for primary emissions, suggesting active processing of the aerosol. Mass concentrations and source contributions of SOC are modelled based on ¹⁴C-apportioned OC and EC, and compared to concentrations and sources of mvOC. Correlation between mvOC and SOC from fossil sources and from non-fossil sources are examined to further explore sources and formation processes of mvOC and SOC.