Source apportionment of carbonaceous aerosols in East Asia based on radiocarbon and molecular marker analyses

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Keywords: Source apportionment, Radiocarbon, Organic markers, East Asia.
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Carbonaceous materials, including organic and elemental carbons (OC and EC, respectively), are one of the most important constituents of aerosols and are ubiquitous across various atmospheric environments (e.g., Kanakidou et al., 2005). In particular, understanding the sources and secondary processes of carbonaceous aerosols is of great importance in considering their climatic and environmental impacts in the atmosphere. Source apportionment of carbonaceous aerosols has been conducted in a lot of previous studies. Molecular marker analysis of OC is recognized as one of the most promising methods to investigate the particular emission sources of carbonaceous aerosols (e.g., Simoneit et al., 2002). Radiocarbon (^{14}C) analysis can directly discriminate modern and fossil carbons and has been performed for the aerosol particles (e.g., Currie, 2000). Their combination enables us to perform the unambiguous characterization of carbonaceous aerosols.

We conducted ground-based sampling of aerosols using two high-volume air samplers (HV500F, Sibata co. Ltd., JP) at Fukue island (32.75ºN, 128.68ºE) in the spring of 2015 (one for the size-segregated sampling of aerosol particles (> and <2.5 μm) and another for the sampling of fine aerosol particles (<2.5 μm)). The pre-combusted (3h, 900ºC) quartz fiber (QF) filters were used for the sampling. Each filter where fine aerosol particles loaded was divided into pieces for the ion, EC/OC, molecular marker (GC-MS), and ^{14}C (Accelerator Mass Spectrometer) analyses. Online measurements of carbon monoxide (CO, model 48C Thermo Scientific, USA) were also performed to diagnose the impacts of continental outflow. Meteorological and environmental characterization during the sampling period was performed by the use of backward trajectories calculated using the NOAA HYSPLIT model, NCEP reanalysis (surface wind), COBE-SST2 (sea surface temperature) and MODIS data sets (Chlorophyll a).

Radiocarbon analysis was applied only for the total carbon (TC) content in aerosol samples collected on the QF filters. As we performed size-segregated sampling, we assume that TC does not include carbonate carbon or primary biogenic carbon such as pollen, which can significantly affect the ^{14}C concentrations. The ^{14}C concentrations are reported as f_M (fraction of modern carbon, e.g., Szidat et al., 2006). Contribution of biomass burning (BB) to modern carbon was analyzed using a molecular marker, levoglucosan (Lev), and the emission ratio of Lev to OC and EC. These emission ratios for the BB over the East Asian continent were estimated using the concentrations of Lev, EC, and OC observed at Rudong town (32.75ºN, 121.37ºE; Jiangsu province, the People's Republic of China) in the early summer of 2010 (e.g., Pan et al., 2012).

The values of f_M for TC varied from ~0.4 to ~0.9, depending on the impact of the continental outflow (i.e., CO enhancement). The estimated contributions of BB to EC were ~7% on average, which was lower than expected from the emission inventory in the year of 2008 (~25%, REAS ver. 2.1). The non-BB/non-fossil (i.e., biogenic) OC concentrations were comparable to and sometimes overwhelmed fossil OC. Note that the chemical instability of Lev can affect the interpretation of the results. In the presentation, we will discuss in detail the temporal variations of source-separated OC/EC concentrations with the discussion on uncertainties.

This work was supported in part by the Environment Research and Technology Development Funds (S7, 2-1403) of the Ministry of Environment, Japan. NCEP reanalysis and COBE-SST2 data sets were downloaded from the website (http://www.esrl.noaa.gov/psd/data/gridded). MODIS data sets were downloaded from the website http://neo.sci.gsfc.nasa.gov/.

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