Source apportionment of carbonaceous PM2.5 with ^{14}C analysis in Nagoya, Japan

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Radiocarbon (^{14}C) is a powerful tracer for estimating the source apportionment of carbonaceous compounds because ^{14}C can be used to discriminate between modern and fossil carbon. There are many studies of PM using ^{14}C in Japan, however studies with ^{14}C analysis for annual and long-range transport from Asian continent are few. We measured ^{14}C concentrations of total carbon (TC) in PM2.5 in Nagoya to investigate the source apportionment of carbonaceous compounds and the long-range transport from Asian continent.

Sampling of PM2.5 was conducted in Nagoya, Japan. Nagoya is located almost at the center of Japan (latitude 35°18'N, longitude 136°91'E), has a temperate climate (average temperature of 16.6 °C), over 2.2 million inhabitants and is one of the major cities in Japan. We carried out daily sampling of PM2.5 for 23.5 h using FRM-2000 from April 2003 to March 2004 at an urban site in Nagoya. 173 samples were collected for a year. Organic carbon (OC) and elemental carbon (EC) were analyzed by the IMPROVE thermal/optical reflectance (TOR) method with carbon analyzer (Sunset Lab). $^{14}C/^{12}C$ ratio was measured with an AMS ^{14}C system at Nagoya University. The samples for 3-11 days were pooled, because it required more than 1 mgC per 1 measurement in order to measure $^{14}C/^{12}C$ ratio with sufficient accuracy. Biomass carbon (BC) and fossil fuel carbon (FC) were calculated by the previous method (Takahashi *et al*, 2007).

The changes of pMC and OC/EC are shown in Fig. 1. Especially in spring, pMC and OC/EC show similar seasonal variations. It is suggested that the change of pMC is not influenced by primary sources (e.g. diesel, industry, etc.) but by other sources (e.g. secondary generation from volatile organic compounds (VOC), long-range transport, etc.).

The concentrations of FC and BC are shown in Fig. 2. BC concentration shows variations similar to OC in spring. Backward air mass trajectories were calculated during the sampling period in spring to identify the source region of high BC concentrations because the air mass flows from the Asian continent in spring. The air mass came from Siberia when OC and OC/EC have high values especially 5-1 and 6-1 (Fig. 2). However the result of 6-2 that has lower OC and BC concentration than 5-1 or 6-1 showed that the air mass came from various regions. In Siberia, large forest burning happened in 2003. High BC concentration in 5-1 and 6-1 is influenced by this forest burning in Siberia.

BC and FC show high concentrations in fall to winter. Some reports say that open burning of agricultural biomass and dry vegetation might be the dominant sources of biomass-derived aerosols. It is considered that biomass burning is important contributor to total carbon (TC) in fall to winter, because daily concentrations of K^+ which is a known tracer for biomass burning s how good correlation with BC. In addition, it is considered that fossil-origin carbon from primary sources is important contributor to TC, because daily concentrations of NO_3^-, NO_x and CO which are known the tracers for primary sources (e.g. diesel, industry, etc.) show good correlation with FC. The high BC and FC concentrations were due to atmospheric stability, because the velocity of the wind showed reverse variations for BC and FC.

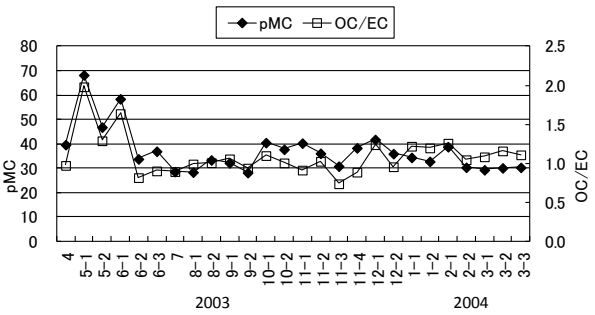

Figure 1. The changes of pMC to OC/EC.

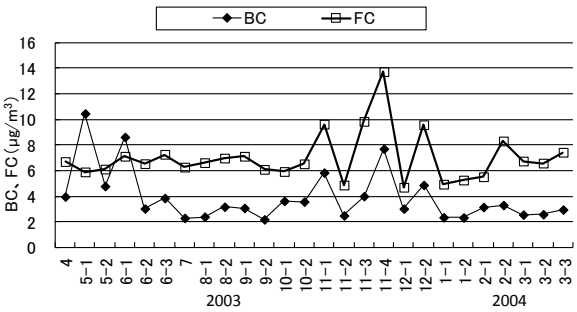

Figure 2. The variations of BC and FC concentration.

Takahashi, K., Hirabayashi, M., Tanabe, K., Shibata, Y., Nishikawa, M., Sakamoto, K., (2007) *Water, Air, Soil Pollut*. **185**, 305–310.