



Decadal trend of black carbon and refractory carbonaceous aerosol in the western rim of the North Pacific Ocean: atmospheric concentration and the retrieved record of deposition flux

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The long-term trend of light absorbing carbonaceous aerosols (or black carbon: BC) or refractory carbonaceous aerosol (or elemental carbon: EC) concentration is reported at European background sites such as Mace Head, and that of aerosol absorption coefficient are monitored in many GAW sites. On the contrary, such long-term data are relatively scarce at around the western part of the North Pacific Ocean.

Thus, to understand the long-term variation of in the area, BC in fine aerosol fraction has been measured at Chichijima Islands, Japan. Chichijima Island is located 1000 km south of the Japanese mainland, and 1800 km west of the coast line of the Asian continent. BC has been measured with an Aethalometer (Magee, AE-16 and AE-30) since December, 1998 with 1 hr time resolution. Mass flowmeter embedded inside the Aethalometer is calibrated with a rotational dry gas-meter once a year.

Monthly averaged BC concentration shows an obvious seasonal variation, i.e. high concentration during late autumn-winter-spring period resulting from the transport from East Asia, with maximum daily concentration above 500 ng m^{-3} . In summer, daily concentration was usually less than 20 ng m^{-3} , due to the clean background air mass originating from the North Pacific Anticyclone. Decadal trend of the annual averaged BC concentration showed an increasing trend from 2000 to 2007 and started to decrease after 2008, which roughly coincides with the reported emission trend of SO_2 in China (Lu et al., 2010).

In addition, total (i.e., wet + dry) deposition record of refractory carbon at two sites in the northern Japan (Rishiri Island: a remote island site, and Sapporo City: an urban site) are retrieved. At these sites, the local government have been measuring the chemical components in precipitation water collected by deposition gauges. In the deposition gauge, a membrane filter made of cellulose-acetate is fixed at the bottom of the funnel to remove water-insoluble particles from the precipitated water. Those "used" membrane filters have been archived for almost two decades at Sapporo without interruption, and for 13 years at Rishiri Island intermittently. We developed a procedure to analyse particulate refractory carbon retained on the membrane filter. The analysis was conducted by thermal-optical transmittance (TOT) method with a Sunset Model-4 carbon analyser (IMPROVE protocol). In this study, we call the resultant analytical quantity as TOT-EC(I).

In Sapporo, the annual total deposition of TOT-EC(I) in 2011 ($80 \text{ mg m}^{-2} \text{ yr}^{-1}$) decreased greatly from that in 2001 ($208 \text{ mg m}^{-2} \text{ yr}^{-1}$) probably due to the strict emission control regulation for the exhaust of diesel powered vehicles that took effect in the 2000s in Japan. In Rishiri Island, total deposition in 2011 ($47 \text{ mg m}^{-2} \text{ yr}^{-1}$) also decreased from that of 2001 ($141 \text{ mg m}^{-2} \text{ yr}^{-1}$). However, the monthly variation of the total deposition in 2001 had pronounced springtime increase while that in 2011 is not obvious. Thus, there is a possibility that the data in 2001 at Rishiri Island were affected by the smoke from Siberian forest fire which becomes active typically during the spring to summer season.

Reference Lu, Z. et al., *ACP*, **10**, 6311-6331 (2010).