## Seasonal variation of wet deposition fluxes of black carbon in East Asia: long-term measurements

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Quantitative understanding of wet removal process of black carbon (BC) is important because it controls temporal and spatial distribution of BC in atmosphere. Majority of BC-containing particles in a planetary boundary layer are mainly active as cloud condensation nuclei and removed by precipitation. To understand the wet removal process of BC, it is essential to measure BC mass concentration in air ( $M_{BC}$ ) and in rainwater ( $C_{BC}$ ) for a long-term period, simultaneously. In this study, a simultaneous measurement of  $M_{BC}$  and  $C_{BC}$  was made at Cape Hedo in Okinawa prefecture during 2010-2016, at Cape Echizen in Fukui prefecture during 2012-2016, and at Happo in Nagano prefecture during 2012-2016. Seasonal variations of both mass concentrations and the wet deposition fluxes ( $F_{BC}$ ) of BC were presented.

 $M_{\rm BC}$  was measured by a Continuous Soot Monitoring System (COSMOS). Rainwater samples were collected on a daily basis by using an automated wet-only sampler and  $C_{\rm BC}$  was measured by consisting of a pneumatic nebulizer (Marin-5) and a single-particle soot photometer (SP2). The monthly averaged  $M_{\rm BC}$ and  $C_{\rm BC}$  at Cape Hedo showed a marked seasonal variation, which was highest in spring (0.25±0.11  $\mu$ g m <sup>-3</sup> and 66.1±70.0  $\mu$ g L<sup>-1</sup>, respectively) and lowest in summer (0.06±0.03  $\mu$ g m<sup>-3</sup> and 5.2±4.2  $\mu$ g L<sup>-1</sup>, respectively). The high  $M_{\rm BC}$  and  $C_{\rm BC}$  in spring were associated with transport of air masses from the Asian continent by northwesterly winds. At Cape Echizen and Happo, both monthly averaged M<sub>BC</sub> also showed a distinct seasonal variation, which was highest in spring (0.31±0.05  $\mu$ g m<sup>-3</sup> and 0.17±0.08  $\mu$ g m<sup>-3</sup>, respectively). On the other hand, both monthly averaged  $C_{\rm BC}$  were highest in winter (19.6±14.1  $\mu$ g L<sup>-1</sup> and 14.4±10.8  $\mu$ g L<sup>-1</sup>, respectively). The high  $C_{BC}$  in winter were not always associated with  $M_{BC}$  in a planetary boundary layer.  $F_{BC}$ , estimated as the product of  $C_{BC}$  and precipitation, also showed a distinct seasonal variation. The monthly  $F_{BC}$  at Cape Hedo was highest in spring (11.8 mg m<sup>-2</sup> month<sup>-1</sup>) and about 80% of annual  $F_{\rm BC}$  occurred in this season on average. Monthly  $F_{\rm BC}$  at Cape Echizen and Happo were highest in winter (4.08 mg m<sup>-2</sup> month<sup>-1</sup> and 2.05 mg m<sup>-2</sup> month<sup>-1</sup>, respectively) and about 40-50% of annual  $F_{\rm BC}$  occurred in this season on average. These  $F_{\rm BC}$  observed in this study were very useful data to varify  $F_{\rm BC}$  calculated by climate models.

Keywords: Black carbon, Wet deposition, Field observation, East Asia