Wet removal mechanisms of black carbon observed in Tokyo and Okinawa

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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 the atmosphere. There are two categories of wet removal mechanisms: nucleation scavenging and impaction scavenging. Theoretically, the removal efficiencies of both mechanisms depend on microphysical properties (size(D_{BC}), coating thickness, and hygroscopicity) of BC. However, the relative contribution of these mechanisms to the removal of BC and the key BC microphysical property controlling the wet removal efficiency of BC have never been tested by direct observation. In this study, we conducted ground-based observations of BC in air and in rainwater in Tokyo and Okinawa, using a new observational method (Mori et al., 2016). We compared the observed size-dependent wet removal efficiency of BC particles ($RE(D_{BC})$, defined as the ratio of the measured size distributions of BC in rainwater to that of BC in ambient air) with the size-dependent number fractions of BC particles scavenged by nucleation ($F_{ccn}(D_{BC})$) and impaction ($F_{imp}(D_{BC})$) mechanisms. The $F_{ccn}(D_{BC})$ and $F_{imp}(D_{BC})$ were estimated from the observed microphysical properties of BC in the air before precipitation starts and the observed droplet size and intensity of precipitation.
The size dependence of $RE(D_{BC})$ showed remarkable differences for the observed 42 precipitation events (31 events in Tokyo and 11 events in Okinawa). The size dependence of $RE(D_{BC})$ for BC-containing particles with D_{BC} > 100 nm was successfully explained by the size dependence of $F_{ccn}(D_{BC})$, whereas the contribution of impaction mechanism can be dominant for particles with D_{BC} < 100 nm. For D_{BC} > 100 nm, when BC particles have relatively thick coating (shell/core ratio > 1.2), the $RE(D_{BC})$ depended little on D_{BC} and the major fraction of BC were removed via nucleation scavenging. On the other hand, when BC particles are nearly bare or have less coating, the $RE(D_{BC})$ highly depended on D_{BC}, and RE (i.e. F_{ccn}) is more sensitive to the hygroscopiticy of coating materials and the maximum supersaturation of water vapor that BC particles would experience during moist convection process. Our results show that the coating thickness of BC is the key parameter controlling wet removal of BC, and indicate that for accurate simulation of vertical transport of BC from the boundary layer to the free troposphere, detailed modeling of microphysical properties of BC and atmospheric supersaturation is required especially for BC particles with thin coatings.

Keywords: Black carbon, Wet removal mechanism, Field observation

