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Evaluation of a method for measurement of black carbon particles suspended in rainwater

Sho Ohata[1*], Nobuhiro Moteki[1], Takuma Miyakawa[1], Yutaka Kondo[1]

[1]RCAST, The University of Tokyo

Black carbon (BC) aerosols are produced by incomplete combustion of fossil fuels and biomass. They contribute to global warming due to their strong absorption of solar radiation. The distribution of BC is controlled by emission, transport, and wet deposition during transport. BC concentration in rainwater is an important parameter for understanding of the detailed processes of wet deposition of BC. Reliable data of wet deposition flux of BC is also useful in validating representation of the removal processes by three dimensional models used for assessing the impacts of BC on climate.

In previous studies, total mass concentrations of BC in rainwater were mainly measured by a thermal-optical transmittance technique applied to BC particles collected on filters. However, this method is not practical for weak rain or high time-resolved measurements during each rain event because we need more than 100 mL of rainwater to determine the BC mass concentration by this method. It also cannot provide BC size distribution in rainwater, which are important for understanding of the CCN activities of BC. In this study, we evaluated a new method to measure BC particles suspended in rainwater. The new method utilizes an ultrasonic nebulizer and a laser-induced incandescence technique for BC detection (Single Particle Soot Photometer: SP2). We demonstrate that this method is practical for measurements of mass concentrations and size distribution of BC in a small amount (several mL) of rainwater sample.

The rainwater sample is transferred to an ultrasonic nebulizer by a peristaltic pump at a constant flow rate. In the ultrasonic nebulizer, some fraction of the introduced rainwater is converted to small droplets in air flow, and they are heated at 140 C and dried at the downstream. Remaining non-volatile cores of individual droplets in the air flow are introduced into the SP2 for single-particle detections of BC.

In this method, we need laboratory experiments to determine the fraction of BC mass in rainwater sample transferred into SP2. We used a commercially available carbon black AquaBlack001 and AquaBlack162 (Tokai Carbon Co. Ltd.) as laboratory standards of BC aqueous solution for the experiments. Under the optimized operating condition of the nebulizer, we determined the fraction R to be about 9% by using samples of various BC concentrations. The R value was almost independent of BC mass concentration and concentrations of other co-existing solutes. For the given value of R, we can determine the BC mass concentrations in rainwater samples.

Second, we tested the reproducibility of the measurement system by repeated measurements of one rainwater sample. This test also showed that the volume of rainwater required for reliable BC measurement is less than 5 mL. We also tested the temporal stability of a rainwater sample over a few months. The measured mass concentrations of a rainwater sample just after sampling and the same one after a storage in refrigerator over several months agreed to within 14%, indicating a relatively small effect of the degradation of rainwater samples over time.

The size distribution of BC in rainwater can change if multiple BC particles are contained in some of droplets generated by the nebulizer, because of coagulation associated with the evaporation of water droplet. Actually, it was found that the size distribution of BC particles shifted to larger sizes as increase of BC concentration. In order to minimize this artifact, we diluted the rainwater sample with pure water before measurements of size distribution. We derived size distribution of BC in a rainwater sample collected in Tokyo in December 2010 following the method described above. For this sample, the BC size distribution can be approximated by a lognormal function with count and mass median diameters of 98 and 190 nm, respectively.

Keywords: black carbon, aerosol, wet deposition