Time evolution of size distribution and mixing state of black carbon and cloud scavenging process

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Black Carbon (BC) is ubiquitous aerosol in troposphere emitted mainly from fossil fuel combustion and biomass burning. BC is an effective visible light absorber so that it brings strong positive radiative forcing and be considered to be responsible for local climate change. Size distribution and mixing state of BC affect strongly on its mass absorption coefficient of visible light. Measuring size and mixing state of BC along mass concentration are important to estimate radiative forcing. In this study, Single Particle Soot Photometer (SP2) fabricated by Droplet Measurement Technologies Inc. is introduced to measure size resolved mass concentration of single particle BC and non-light absorbing aerosols. BC introduced into intense laser beam in SP2 begins heats up to over 3000 K and emits incandescent light along with elastically scattered light. SP2 detects incandescent and scattered light simultaneously with respect to single particle BC. The mass of BC particle is a function of intensity of incandescent light. Relationship of BC mass and incandescent intensity is determined by our laboratory experiment which adopts Aerosol Particle Mass analyzer (APM) to sort known mass BC. At this point, combination of SP2 and APM is the most effective and reliable method to measure size resolved mass concentration of BC in high time resolution suited to aircraft observation. Schwartz et al., (2006, JGR) suggests based on experimental results that the time delay of scattering to incandescence of BC detection signal (t) can be used to measure qualitatively whether BC is coated by non-light absorbing aerosol or not. In this study, using originally developed aerosol coating equipment and simulation model, we independently demonstrate the validity this t based mixing state classification method. In March 2004, we participated in the PEACE-C (Pacific Exploration of Asian Continental Emission -C) aircraft observation campaign conducted in over Pacific Ocean off the Tokai area, Japan and measured BC by SP2 and absorption coefficient of aerosols by filter-based light absorption method (by PSAP). By the analysis of backward trajectory of plume and observed aerosol data, time evolution of size distribution, mixing state, mass absorption efficiency of BC was derived in the temporal scale of 7 hours in boundary layer. Hour 0 is defined as the time when the air mass passed above urban area (emission area). Size distribution of BC was well fitted by log-normal distribution. Peak diameter of volume distribution significantly changed: 180 nm to 220 nm, during 7 hours transport. Coagulation of BC included aerosols is thought to be responsible for this enlargement of size. t was used to classify mixing state of BC of specific volume equivalent diameter. The nomenclatures of Type1 (degree of internal mixing is low) and Type2 (degree of internal mixing is high) are used in this analysis. With respect to BC of d=180 nm, number fraction of Type 2 was significantly increased (0.3 to 0.6) during 7 hours. This is the first study that demonstrates the time scale of internal mixing of BC. Then, mass absorption coefficient derived by combination of mass concentration of BC and absorption coefficient was found to increase factor 1.4 during 7 hours. Internal mixing with non-light absorbing aerosol and BC is thought to be mainly responsible for this enhancement of BC mass absorption coefficient. We also estimated cloud scavenging efficiency of BC by comparing the number concentration of BC just under the cloud and in the cloud at same horizontal location. In two cases during PEACE-C campaign, cloud scavenging efficiency of BC strongly depended on mixing state. In these cases, Type 2 BC is more efficiently scavenged in cloud than Type 1 BC. The dependence of cloud scavenging efficiency of BC on its mixing state is demonstrated for the first time in this study.