論文の内容の要旨

Abstract of Dissertation

Title: Study on black carbon aerosols at urban and remote sites in East Asia

(東アジアの都市域及びリモート域におけるブラックカーボンエアロゾルの研究)

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Black carbon (BC) aerosols are produced from incomplete combustion of fossils fuels and biomass burning. The concerns for the BC aerosols at the global levels have been increased in recent past due to its active involvement in climate change. The radiative forcing (a measure of climate change) caused by BC is about 55% of that of carbon dioxide (CO_2). Also, BC interferes in microphysical properties of clouds by changing the radius and lifetime of clouds. BC emissions are increasing in Asia due to rapid commercial and economic developments. Asian BC is not only

impacting Asian climate, but also being transported to Arctic region, causing decreasing of snow albedo and increasing of Arctic haze. Climatic impacts of BC depends on the concentrations exist in atmosphere. The variations of ambient BC levels depends on the strength of emissions and local meteorology at source regions, while, transport and level of removal at remote places. These aspects of BC are poorly understood for Asian regions. For the first time, in this study, we addressed some of these issues by making continuous in situ measurements of mass concentrations of BC and mixing ratios of carbon monoxide (CO) and carbon dioxide (CO_2) at urban and remote sites in East Asia.

The urban site was situated in the main city of Guangzhou, Pear River Delta (PRD) region of South China. PRD is among the main industrial regions of China, where dominant BC emissions are from transport, industries, and domestic sectors. To study the relationships of BC with CO and CO_2 in light of local meteorology and emissions, in situ measurements of BC, CO, and CO_2 were made in July 2006. The observed average ± standard deviation (SD) concentrations of BC, CO, and CO_2 were 4.7±2.3 μgC m^{-3}, 798±459 ppbv and 400±13 ppmv, respectively, during entire campaign. The trends of these species were mainly controlled by synoptic-scale changes in meteorology during the campaign. Based on back trajectories, data are analyzed separately for two different air mass types representing northerly and southerly flows. Northerly air masses, constituting about 25% of the campaign, were mostly originated within the PRD and represent the observations of regional levels. On the other hand, southerly air masses measured during most of the campaign were continuously diluted by clean marine air. The diurnal patterns of BC, CO, and CO_2

exhibited peak concentrations during the morning and evening hours coinciding with rush-hour traffic. The ratios of OC/BC were lower during morning peak hours indicating dominance of fresh emissions. The diurnal variations of BC of southerly air masses closely followed the traffic pattern of heavy-duty vehicles (HDV). The observed slopes of $\Delta EC/\Delta CO$, $\Delta EC/\Delta CO_2$, and $\Delta CO/\Delta CO_2$ during northerly air masses were 0.0045 μgC m^{-3}/ppbv, 0.13 μgC m^{-3}/ppmv, and 49.4 ppbv/ppmv, respectively, agreeing reasonably well with their respective emission ratios derived from regional emission inventories.

To study the transport pattern, role of wet removal and the transport of BC relative to CO, in situ measurements of BC and CO were made at Hedo from March 2008 to May 2009. Hedo is remote observation site situated at the Okinawa Island of Japan, in Pacific Ocean. BC was measured using Continuous Soot Monitoring System (COSMOS), an improved version of filter-based instrument. CO was measured using non-dispersive infrared absorption (NDIR) based instrument. Hedo received East Asian outflow in winter and spring during which average concentrations of BC and CO were increases to 0.41 μgC m^{-3}, and 183 ppbv from the level of 0.19 μgC m^{-3}, and 101 ppbv, respectively, observed during summer and fall when mostly marine outflows were prevailed. Based on back trajectory analysis, the air masses have been classified as North China (NC), South China (SC), Korea (KR), Japan (JP), marine (MA), and free troposphere (FT). Characterizations of air masses arriving from each categories were made for BC and CO considering their residence time of at least 24 hours (RT > 24 hours). Based on the criteria of RT (>24 hours), about 44 – 52% of air masses were received from Chinese region during spring and winter, while about

75% MA from Pacific Ocean in summer. The observed average BC concentrations in the air masses from source regions were ranges from 0.40 to 0.79, 0.27 to 1.12, 0.22 to 0.45, and 0.16 to 0.29, in NC, SC, KR, and JP, respectively, in all five seasons, expressed in μgC m^{-3}. The levels of wet removal of BC were estimated by deriving wet deposition efficiency (WDE). WDE is the percentage removal of BC, estimated using BC simulated by Community Multiscale Air Quality (CMAQ) system. A good dependency of the slopes of ΔBC/ΔCO against WDE was observed. The observed slopes of ΔBC/ΔCO in the air mass categories of NC, SC, KR, JP, MA, and FT with RT > 24 hours and WDE < 20%, were 0.0065, 0.0080, 0.0062, 0.0050, 0.0016, and 0.0030, respectively, expressed in μgC m^{-3}/ppbv. Region-specific (of NC, SC, KR, and JP) ΔBC/ΔCO slopes are compared with the emissions ratios derived from recent inventory of *Zhang et al*. [2009]. We found there could be an over estimation of BC by a factor of two in East Asia regions.