論文内容の要旨

論文題目

Cloud condensation nuclei properties of atmospheric aerosol particles

(実大気エアロゾルの雲凝結核特性)

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This thesis focuses on the cloud condensation nuclei (CCN) activity of atmospheric particles. According to Köhler theory, the CCN activity of atmospheric particles is determined by particle size, mixing state, and chemical composition. However, only a few studies have addressed how size and chemical composition affect the CCN activity of atmospheric particles, and no investigation has been conducted for the influence of CCN activity on mixing state. In this thesis, I study (1) how the activation diameter of size-resolved CCN spectra is determined when particles are internally mixed (Chapter 2), (2) how the mixing state of non-volatile cores influences size-resolved CCN spectra in urban areas (Chapter 3), and (3) how the CCN activity of less volatile particles is determined (Chapters 4 and 5).

Size-resolved CCN spectra of internally-mixed particles were observed at Gosan, Jeju Island (Korea), during March and April of 2005. In addition, chemical composition (inorganic ions,

water-soluble organic carbon, organic carbon, and elemental carbon) was observed at $PM_{2.5}$. Number fractions of CCN-active particles were almost equal to zero at small particle diameter (100 nm for supersaturation (S) = 0.1%) and increased with increasing diameter, approaching unity at 200 nm (S = 0.1%). The, 50% activation diameter is defined as D_{50}. The temporal variation of D_{50} was similar to that of the mass fraction of water-soluble compounds (summation of inorganic ions and water-soluble organic carbon), demonstrating that D_{50} is controlled by chemical composition. In addition, the activation diameter was calculated using the observed data of chemical composition. Although calculated values were correlated with D_{50}, they were larger than D_{50}. It is demonstrated that the possible influence of the size dependence of chemical composition or surface tension depletion can account for the discrepancy.

The influence of the mixing state of non-volatile cores on size-resolved CCN spectra was investigated by conducting simultaneous measurements of size-resolved CCN spectra and volatility tandem differential mobility analyzer (VTDMA). The VTDMA measurement was conducted at a heater temperature of 400 °C. The size distributions of the non-volatile cores of size-selected particles measured by the VTDMA were bimodal; one mode showed relatively small changes (< 10%) in peak diameter by volatilization, and the other showed significant changes in diameter (> 10% in peak diameter). The former mode is referred to as less-volatile (LV), and the latter mode is called more-volatile (MV). The main component of non-volatile cores in Tokyo is known to be black carbon (BC). Although a stepwise increase in CCN-active particle fraction was observed as in the case of Gosan, the fractions were smaller than unity after the increase. A CCN-LV correlation analysis shows that for small (< 80 nm) particles, the slopes of the correlations are smaller than unity, although the correlations are significant. This indicates that CCN-inactive fractions are explained by LV particles and particles co-emitted with LV particles (likely primary organic aerosol particles). For larger particles (> 100 nm), the CCN-inactive fractions are close to the LV particle fractions, suggesting that CCN-inactive particles were composed of fresh soot, although some LV particles were found to be CCN active especially during daytime.

To investigate the CCN activity of LV particles, I developed a volatility tandem aerosol particle mass analyzer (VTAPM), which measures the mass of condensed compounds on non-volatile particles. The VTAPM was operated downstream of a differential mobility analyzer (DMA) to investigate particles selected by their size and effective density (ρ_{eff}). The ρ_{eff} distribution of atmospheric particles in Tokyo was bimodal: a light mode (ρ_{eff} ~ 0.8 g cm^{-3}) and a heavy mode (ρ_{eff} ~ 1.4 g cm^{-3}). The VTAPM measurements have shown that light particles mainly consisted (70 ~ 80% in weight) of non-volatile compound (BC). A comparison with CCN measurements demonstrates 0.18 fg (supersaturation (SS) = 0.9%) and 0.07 fg (SS = 1.3%) of volatile mass are required for CCN activation of light particles. As the light particles are likely fresh soot, these values give a measure of the criterion for separating hydrophobic and hydrophilic soot particles.