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学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Studies on the effects of cold terrestrial biogenic emissions of organics on the cloud forming potential of atmospheric aerosols

（寒冷陸域植生から放出される有機物が大気エアロゾルの雲生成ポテンシャルに及ぼす影響に関する研究）

Terrestrial biogenic emissions of organics can affect the cloud condensation nuclei (CCN) activity and optical properties of atmospheric aerosols, and subsequently impact climate change. Large uncertainties exist in how the difference in types and abundance of biogenic sources affect the formation of organic aerosol and its subsequent CCN activity. In order to investigate the effects of biogenic organic compounds on the CCN properties, two field experiments were made at suburban (Sapporo) and cool-temperate forest (Tomakomai) sites to measure the chemical composition, particle hygroscopicity, and CCN properties of submicron aerosols.

The time-resolved online measurements of submicron aerosols at the suburban site in summer showed that temporal variation in the CCN activity, described by the hygroscopicity parameter κ, was closely related to changes in the chemical composition and mixing state of aerosols. The temporal variation of water-soluble organic matter (WSOM)-to-sulfate ratio was closely linked to that of κ, where WSOM was likely dominated by the influence of biogenic sources. From a two year-long measurement of water-soluble aerosols at the forest site, κ derived from CCN measurements (κ_{CCN}) exhibited a distinct seasonal trend with a minimum in autumn. The results also showed that κ_{CCN} depends on the WSOM/sulfate ratio, where the significant reduction in the κ_{CCN} values in autumn was linked to the increase of WSOM. Positive matrix factorization analysis indicates that α-pinene-derived secondary organic aerosols (SOA) substantially contributed to the WSOM mass (~75%) in autumn, the majority of which was attributable to emissions from litter/soil near the forest floor. These findings suggest that WSOM, most likely originated from the forest floor, can significantly suppress the CCN activity of aerosols in cool-temperate forests. The reduction of the CCN activity can be explained by α-pinene derived SOA coating pre-existing hygroscopic particles, which was estimated to reduce the number of activated CCN particles by ~30% in autumn. Considering the atmospheric lifetime of α-pinene derived SOA, emissions of α-pinene from the forest floor can affect the activation of CCN within the spatial scale of ~800 km.

Comparison of the filter-based aerosol measurements with aerosol optical properties retrieved from a Sky Radiometer at the forest site showed that the increased aerosol mass in the forest canopy corresponded to increase in the aerosol optical depth of fine mode particles (diameter < 1 μm) in autumn. In summer, the increased mass fraction of sulfate was linked to the enhanced properties of aerosol scattering. In contrast, the increase in the mass fraction of WSOM in autumn was found to be mainly associated with more absorbing characteristics of aerosols. This suggests that the biogenic organic aerosols at the forest site can modify the aerosol optical properties on a regional spatial scale. The overall results demonstrate that changes in types and amounts of biogenic organic emissions in cool-temperate forests can control the cloud forming potential and can affect the aerosol optical properties. This study has important implications for predicting regional climate effects by changes in biogenic emissions of organics in the future.