学 位 論 文 内 容 の 要 旨/Dissertation Abstract

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Study on the origin and atmospheric emission process of secondary fatty alcohols in aerosols in cool-temperate forests
（冷温帯林におけるエアロゾル中の脂肪族第二級アルコールの起源と大気放出に関する研究）

Fatty alcohols (FAs) are major components of surface lipids in plant leaves and can be emitted into the atmosphere as primary biological aerosol particles (PBAPs), which serve as surface-active organic aerosols. Furthermore, FAs in aerosols can act as ice nucleating particles to form cloud particles, which is thus important for radiative forcing, precipitation, affecting climate change. Among FAs, secondary FAs (SFAs) are less affected by atmospheric photochemical reactions than primary FAs due to their chemical structures. Therefore, SFAs can serve as tracers of specific sources of aerosols. Despite the importance of aerosol SFAs in atmospheric chemistry and climate, studies on SFAs in ambient aerosols are limited, due to a lack of aerosol measurements particularly in the vicinity of their specific source regions. The objective of this thesis is to elucidate the origin and emission process of SFAs in atmospheric aerosols, based on size-segregated aerosol and leaf samples of different plant species collected at two forest sites.

Size-segregated aerosol samplings were performed at Sapporo Forest meteorology research site and Tomakomai (TMK) experimental forest site of Hokkaido University in different seasons. Leaf samples from seven different plant species were simultaneously collected at TMK site. SFAs in both aerosol and leaf samples were measured by using a gas chromatograph coupled to a mass spectrometer (GC-MS).

At both forest sites, SFAs in aerosols showed the highest concentration in spring (growing season), followed by autumn. Among SFAs identified, *n*-nonacosan-10-ol was the most abundant compound. The mass of *n*-nonacosan-10-ol was mostly in the supermicrometer size range, whereas the peak of the size range shifted to smaller sizes from spring to autumn. At Sapporo Forest meteorology research site, the mass size distributions were highly correlated with those of bulk water-insoluble organic carbon (WIOC) in spring, indicating that SFA is an important factor controlling the WIOC mass. These results suggest that SFAs originate mostly from plant waxes, and that leaf senescence status is likely an important factor controlling the size distribution of SFAs.

At TMK site, among the leaf samples of all the plant species collected at the study site, *n*-nonacosan-10-ol was identified only in the conifer leaf samples. *n*-nonacosan-10-ol was mostly (94%) in the non-esterified (free) form in the conifer leaf. The mass of *n*-nonacosan-10-ol per one needle leaf was largest in spring, followed by autumn. This seasonal trend was similar to that of SFA concentrations in aerosols. The seasonal changes in the

mass of *n*-nonacosan-10-ol in the aerosols and conifer leaves corresponded well with those of the magnitude of local wind speeds. The overall results suggest that plant waxes in the conifer leaves can act as source of aerosol SFAs, and that the emission strength is controlled by the phenology of the conifer leaf and the magnitude of local wind speeds. This study suggested that *n*-nonacosan-10-ol can be a new tracer compound of PBAPs which originate from conifer leaves.

The atmospheric emission flux of plant leaf wax from coniferous trees during the growing season at TMK site corresponds to approximately ~18% of the total PBAP flux from the forest shown in literature. The current study demonstrated the importance of conifer leaves as a source of SFAs in PBAPs, which are widely distributed on a global scale. This study also implied that the growth and senescent status of plants should be taken into account as factors controlling the amount of SFAs in PBAPs. These PBAPs can affect the ice nucleating activity of aerosols and are thus important for the cloud formation.