学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Study on the origin of atmospheric water-soluble organic aerosols at the high-altitude observatory, Réunion island in the tropical Indian Ocean
（熱帯インド洋レユニオン島の高高度観測所における大気水溶性有機エアロゾルの起源に関する研究）

The ocean is a major source of submicrometer aerosols, which play an important role in the atmospheric radiative budget because they determine the number of cloud condensation nuclei (CCN) and ice nuclei (IN). Marine-derived submicrometer organic aerosols (OAs) can affect the marine aerosol optical depth (AOD) as well as CCN and IN concentrations. These are particularly important over remote oceans, as these areas experience minimal influence from anthropogenic emissions originating from terrestrial sources. The tropical Indian Ocean (IO) is expected to be a significant source of water-soluble organic aerosols (WSOAs) which are important factors relevant to cloud formation of aerosol particles, because of high marine primary productivity over this oceanic region. Current atmospheric numerical models significantly underestimate the budget of organic aerosols and their precursors, especially over tropical oceans. This is primarily due to poor knowledge of sources and the paucity of observations of these parameters considering spatial and temporal variation over the tropical open ocean.

To evaluate the contribution of sources to WSOA as well as their formation processes, submicrometer aerosol sampling was conducted at the high-altitude Maïdo observatory (21.1°S, 55.4°E, 2,160 m a.s.l), located on the remote island of La Réunion in the southwest IO. The sampling was made within the framework of an international project, OCTAVE (Oxygenated Compounds in the Tropical Atmosphere: Variability and Exchanges). The aerosol samples were continuously collected during local daytime and nighttime, which corresponded to the ambient conditions of the marine boundary layer (MBL) and free troposphere (FT), respectively, from March 15 to May 24, 2018.

Chemical analysis showed that organic matter was the dominant component of submicrometer water-soluble aerosol (~45 ± 17%) during the wet season (March 15–April 23). On the other hand, sulfate dominated (~77 ± 17%) during the dry season (April 24–May 24), most of which was attributable to the effect of volcanic eruption. Measurements of the stable carbon isotope ratio of water-soluble organic carbon (WSOC) suggested that marine sources contributed significantly (~70%) to the observed WSOC mass in both

the MBL and the FT in the wet season, whereas a mixture of marine and terrestrial sources dominantly contributed to WSOC in the dry season. The distinct seasonal changes in the dominant source of WSOC were also supported by Lagrangian trajectory analysis.

Positive matrix factorization analysis suggested that marine secondary OA dominantly contributed to the observed WSOC mass (~70%) during the wet season, whereas mixtures of marine and terrestrial sources contributed during the dry season in both MBL and FT. Overall, this study demonstrates that secondary formation of marine-derived aerosol is likely important up to the FT in the wet season, when marine biological activity and vertical transport are more significant. The formation of marine secondary OA is a process generally missing in current climate models. Current models typically consider only marine primary OA (i.e., sea spray aerosols) from the sea surface to represent the OA burden in tropical "pristine" oceanic regions. The impacts of marine secondary OA up to FT aerosols lead to changes in the microphysical and optical properties of aerosol particles. The findings in this study may have important implications for understanding the climate effects of aerosols in these oceanic regions.