

Origin of marine atmospheric water-soluble organic aerosols at a high-altitude observatory, Reunion Island in the tropical Indian Ocean

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Ocean-derived atmospheric aerosols can affect radiative forcing via formation of cloud droplets and ice nuclei as well as biogeochemical cycle of bio-elements. Moreover, oxygenated volatile organic compounds (OVOCs) give significant impact on the atmospheric oxidative capacity and climate, which serve as key precursors of climatically active secondary organic aerosol (SOA) in the marine boundary layer. Atmospheric reactions of OVOCs and aerosols are closely linked, and emissions of these are significant in the tropical oceans. However, current atmospheric models significantly underestimate the budget of SOA especially over tropical oceans, primarily due to poor knowledge of sources and paucity of observations of these parameters in tropical remote regions.

To investigate the role of tropical oceans in the formation of atmospheric organic aerosols, size-segregated aerosol sampling was conducted for the first time at the Maïdo observatory (2,200m a.s.l.) in Reunion Island in the Indian Ocean. Aerosol samples were continuously collected in daytime and nighttime, which corresponded to ambient conditions of the marine boundary layer (MBL) and free troposphere (FT), respectively, during March 15–May 24, 2018. Off-line chemical analysis of submicrometer and supermicrometer water-soluble aerosols, which are important for cloud formation, was made using the aerosol samples. In particular, stable carbon isotope ratio ($\delta^{13}\text{C}$) of water-soluble organic carbon (WSOC) ($\delta^{13}\text{C}_{\text{WSOC}}$) was measured for the source apportionment.

The analysis showed that organic matter was the dominant component of the submicrometer aerosol ($46\pm 23\%$ in MBL) during the first half of the study period (March 15–April 24; wet season), whereas sulfate dominated ($60\pm 19\%$ in MBL) during the latter half (April 24–May 24; dry season) in the absence of volcanic-plume influence. $\delta^{13}\text{C}_{\text{WSOC}}$ together with significant positive correlations of WSOC with methanesulfonic acid (MSA) and ozone suggested that most of WSOC in the submicrometer aerosols were secondary products originated from marine biogenic sources under the MBL condition during the wet season. On the other hand, WSOC under the FT conditions were dominated by the influence of terrestrial biogenic sources throughout the entire period, which was suggested by $\delta^{13}\text{C}_{\text{WSOC}}$ and biogenic tracer compounds. Overall, the impact of organic matter originated from marine surface is suggested to be confined within the MBL, which might be important for the formation of low-level cloud particularly during the wet season in this oceanic region.

Keywords: marine atmospheric aerosols, water-soluble organic carbon, marine boundary layer, free troposphere, OVOC