

## Origin of water-soluble organic nitrogen in marine atmospheric aerosols in the subtropical North Pacific

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Marine atmospheric aerosol is one of the most important factors that determine the climate change, because it absorbs and reflects the sunlight which affects radiative balance of the atmosphere. In particular, water-soluble organic nitrogen (WSON) in aerosol affect physicochemical properties of aerosol such as water-solubility characteristics, acidity, light-absorption properties of the particles. However, little is known about the origin of WSON in marine atmospheric aerosols. The purpose of this study is to elucidate the origin of WSON by cruise observation in subtropical North Pacific where very limited data are available.

Size-segregated aerosol samples were collected on board the R/V *Hakuho Maru* (KH-17-4) over the subtropical North Pacific, mainly along 23°N, during August–October in 2017. Water-soluble organic carbon (WSOC) and water-soluble total nitrogen (WSTN) were measured by a total organic carbon analyzer with a TN unit. The concentrations of inorganic nitrogen (IN) were measured by ion chromatograph. The WSON concentration was derived from the difference between WSTN and IN. The stable carbon isotope ratio ( $\delta^{13}\text{C}$ ) was measured by elemental analyzer-isotope ratio mass spectrometer (EA-IRMS). Longitudinal distributions of WSON at 23°N showed that the submicrometer WSON concentration (av.  $5.9 \pm 6.0 \text{ ng N m}^{-3}$ ) to the east of 160°W was significantly larger than that to the west (av.  $2.2 \pm 2.0 \text{ ng N m}^{-3}$ ). Additionally, the mass fraction of WSON to WSTN in the eastern oceanic region reached as large as 40% with an average of 14.1%, suggesting the importance of WSON in this oceanic region. Backward trajectory, the chlorophyll *a* concentration in seawater, and the  $\delta^{13}\text{C}$  value suggested that the majority of the observed WSON was derived from microbial activity in the sea surface. WSON did not show any significant correlation with a tracer of primary emission (i.e.,  $\text{Na}^+$ ), indicating that the majority of WSON were not directly emitted as aerosol particles from the ocean surface, but were produced from N-containing gas species released into the atmosphere. Furthermore, a significant correlation between the concentrations of WSON and the nitrogen fixation rate in the sea surface indicates that reactive nitrogen, produced by nitrogen fixing organisms (e.g., cyanobacteria), are emitted from sea surface to the atmosphere, which significantly contributed to the formation of aerosol WSON.

Keywords: Organic nitrogen, Marine atmospheric aerosol, Biogeochemical linkage between atmosphere and ocean, Nitrogen fixation