

## Seasonal and decadal variations in stable carbon isotope ratios of low molecular weight dicarboxylic acids and related compounds in the remote marine aerosols from the western North Pacific

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Oxalic and other dicarboxylic acids comprise an important fraction of water-soluble organic aerosols in various environments. To better understand long-term atmospheric changes in the western North Pacific, we collected marine aerosol samples in 2001-2014 on weekly basis at a remote island, Chichijima (27° 04'E; 142°13'N), which is located in the boundary of westerly and easterly wind regimes. Here we present seasonal and decadal trends in stable carbon isotopic compositions ( $\delta^{13}\text{C}$ ) of diacids dicarboxylic acids ( $\text{C}_2\text{-C}_{11}$ ), oxoacids ( $\text{C}_2\text{-C}_9$ ) and  $\alpha$ -dicarbonyls ( $\text{C}_2\text{-C}_3$ ) in the Chichijima aerosols. These compounds were determined using gas chromatography (GC) and GC/MS techniques after  $\text{BF}_3/\text{n-butanol}$  derivatization and using a GC/isotope ratio/MS technique.

Concentrations of total diacids fluctuated in a range of  $10\text{-}600\text{ ngm}^{-3}$  with winter/spring maxima and summer minima. The maximum concentrations in winter/spring can be explained by a combination of enhanced emissions of polluted aerosols and their precursors in the Asian Continent and enhanced atmospheric transport to the North Pacific due to the intensified westerly winds in winter/spring. The concentrations of diacids seemed to increase from 2001 to 2008 and then decrease toward recent years, as supported by major ion analyses. Recent decline of diacids concentrations may be associated with the changes in the anthropogenic emissions due to the air quality improvement in Asian countries.

Stable carbon isotopic compositions determined for 2006 showed relatively high  $\delta^{13}\text{C}$  values of oxalic acid ( $-22\text{‰}$  to  $-4\text{‰}$ ). The isotopic ratios were found to increase from winter to summer. Significant enrichment of  $^{13}\text{C}$  in oxalic acid in summer should be associated with photochemical aging of organic aerosols in the marine atmosphere, caused by kinetic isotopic fractionation during the photo degradation of oxalic acid. We will discuss the decadal changes in the stable carbon composition of oxalic acid in relation to the atmospheric oxidation capability in the western North Pacific.

Keywords: Marine aerosols, Oxalic acid, Stable carbon isotope ratio, Seasonal and decadal trends