

4.045 Decadal and seasonal distributions of low molecular weight dicarboxylic acids and related compounds, and their stable carbon isotope ratios in the remote marine aerosols from the western North Pacific.

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Abstract:

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 distributions of dicarboxylic acids (C₂-C₁₁), oxoacids (C₂-C₉) and α-dicarbonyls (C₂-C₃), which were determined using gas chromatography (GC) and GC/MS techniques after BF₃/n-butanol derivatization. We also measured stable carbon isotopic composition (δ¹³C) of diacids and related compounds using a GC/isotope ratio/MS technique. Concentrations of total diacids fluctuated in a range of 10-600 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 δ¹³C values of oxalic acid (-22‰ to -4‰). The isotopic ratios were found to increase from winter to summer. Significant enrichment of ¹³C 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.