

### **3.049 Long-term decline of stable carbon isotopic composition of oxalic acid in marine aerosols from the western North Pacific: Atmospheric implication for an enhanced emission of terrestrial organic matter.**

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Abstract:

Oxalic, malonic and other dicarboxylic acids comprise an important fraction of water-soluble organic aerosols in various environments. A rapid industrial development in China and East Asian countries for last two decades may have caused a serious change in the air quality and atmospheric composition over the North Pacific. To better understand long-term atmospheric changes in the western North Pacific, we collected marine aerosol samples in 2001-2013 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 distributions of dicarboxylic acids (diacids, C<sub>2</sub>-C<sub>11</sub>) and stable carbon isotopic composition (δ<sup>13</sup>C) of oxalic acid using a GC/combustion/isotope ratio monitoring MS (GC/IR/MS) technique.

We found a strong seasonal change in δ<sup>13</sup>C values of oxalic acid with summer maxima and winter minima, e.g., seasonal shift from -22‰ in winter to -4‰ in summer in 2006. The significant enrichment of <sup>13</sup>C in oxalic acid with higher values in summer should be associated with photochemical aging of organic aerosols in the marine atmosphere; that is, kinetic isotopic fractionation during the photo degradation of oxalic acid in the presence of Fe (III), and/or gas/particle partitioning reaction of glyoxal and glyoxylic acid, potential precursors of C<sub>2</sub>.

We also found a decadal decrease in the stable carbon composition of oxalic acid from 2001 to 2013 by ca. 3‰. The decadal decrease can be most likely explained by an enhanced input of continental organic matter (isoprene and biomass burning) from East Asia over the western North Pacific. An increased emission of CO<sub>2</sub> by fossil fuel combustion may potentially contribute to enhance the emission of isoprene, whereas the corresponding decline of δ<sup>13</sup>C (0.2‰) due to fossil fuel CO<sub>2</sub> may contribute to decrease in δ<sup>13</sup>C of terrestrial plants and their isoprene emissions.