

## **Long-term (2001-2012) trends in carbonaceous aerosols from Chichijima Island in the western North Pacific: an outflow region of Asian pollutants and dust**

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### **Abstract**

The present study reports on long-term trends of carbonaceous aerosols in total suspended particulate (TSP) samples collected at Chichijima island in the western North Pacific during 2001-2012. Seasonal concentrations of elemental, organic, and water-soluble organic carbon (EC, OC and WSOC) showed maxima in winter to spring and minima in summer. These seasonal differences in the concentrations of carbonaceous aerosols are associated with a long range transport of polluted air masses from East Asia, which are clearly distinguishable from pristine air masses originated from the central Pacific. The higher concentration of carbonaceous aerosols during winter to spring are associated with long-range atmospheric transport of east Asian polluted air masses while lower concentrations may be due to pristine air masses from the central Pacific in summer. The annual trends of WSOC, and OC/EC and WSOC/OC ratios showed significant ( $p < 0.05$ ) increases during the period of 2001-2012, suggesting that an enhanced secondary formation of organic aerosols via photochemical oxidation of anthropogenic and biogenic volatile organic carbons (VOCs) during long-range atmospheric transport. We found a significant increase in  $nss-K^+/EC$  ratios, demonstrating that biomass burning-derived organic aerosols are increased, whereas combustion derived anthropogenic sources are decreasing over the western North Pacific. Further, secondary biogenic emissions are also important over the western North Pacific as inferred from a significant increase in the concentrations of methanesulfonate ( $MSA^-$ , a tracer for biogenic source). We also found significant increases in OC/TC and WSOC/TC ratios, which suggest that contribution of secondary organic aerosols to total carbon (TC) has significantly increased over the western North Pacific under the influence of long-range atmospheric transport.

**Keywords:** Carbonaceous aerosols, long-term trends, the western North Pacific, East Asia, biomass burning, biogenic emissions, long-range atmospheric transport, photochemical oxidation.