Study on inorganic halogens and other related ions in the lower atmospheric aerosols

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Aerosol chemistry and cycling of three inorganic halogen species (bromide, chloride and fluoride) in the troposphere are studied with necessary reference to other ionic precursors (ammonium, calcium, magnesium, nitrate, potassium, sodium and sulphate). Aerosols (size segregated and bulk) over the Atlantic Ocean were collected at sea during two interhemispheric research cruises in September-October 2001 and May-June 2003 between around 50°N and 50°S, and at Bermuda during May-September 2003. Ambient continental aerosols were also collected over short campaigns from 2002 to 2004 in Edinburgh, London, Merlewood and Norwich to study continental influence in the open ocean aerosols. Samples were analysed by ion chromatography.

Size distribution of non sea salt fraction of the ions of interest, air parcel back trajectories and interspecies correlations (especially with pollution tracer ions) were used to identify the processes regulating depletion and enrichment of halogen ions of interest, as well as to characterise the sources of these halogens aerosols over the Atlantic Ocean. Both depletion (partial and complete) and enrichment of bromide and chloride with respect to sea salt are evident in open ocean and continental aerosols. Fluoride in open ocean aerosols is found to be below detection limit, whereas mainly enriched in continental aerosols.

In general, signatures of industrial and urban emission sources from Western Europe, British Isles, North Africa, North America, and South America, mineral dust blown off the Saharan, sub-Saharan and Patagonian arid region, biomass burning from Southern Africa, and marine biogenic emission have been shown to influence halides and other aerosol ions. The effects of interhemispheric differences in emission pattern are evident.

This thesis suggests that (1) halogen cycling in the troposphere is not simply governed by halogen depletion mechanisms (such as acid displacement) as revealed from the existing literatures (2) there are some significant sources other than sea salt even above the open ocean (3) depletion of bromide is not unequivocally larger compared with Cl-, as predicted by acid-catalysed autocatalytic cycle.