Cloud and Aerosol Ammonium Concentrations on Mount Washington, NH (1,540 m)

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Atmospheric acidity and sulfate concentrations have declined steeply in the US due to the 1990 Clean Air Act Amendments (CAAA) and other more recent CAA emission regulations. From 1990 to 2011, national sulfur dioxide emissions declined by 70%, nitrogen oxide emissions by 48%, Yet ammonium emissions have changed little since 1990 (Source: National Emissions Inventory*). A multi-decade summertime cloud and aerosol chemistry dataset from the Lakes of the Clouds (LOC) site on Mount Washington, NH (1,540 m asl) reflects these emission changes with declines in sulfate in aqueous and aerosol phases. Acidity has also declined, however the relative amount of ammonium has increased with associated measured sulfate. Regression analysis of samples from pre and post 1995, e.g. implementation of the 1990 Clean Air Act Amendments (CAAA), showed that LOC site molar ratio of ammonium to sulfate in cloud water has increased from 0.77 to 1.08 while aerosol ratios have changed from 0.98 to 1.22. Changes were also observed in lower elevation aerosol records pre and post CAAA.

Cloud and rain water summer annual median concentrations, from 1996 -2010, also show changes in ammonium levels relative to other ions. Median cloud water ammonium concentrations transition over the record from relatively lower (1996-1999), to near equal (2000-2004), and then exceeding (2005-2010) hydrogen concentrations. In rainfall median ammonium and nitrate concentrations are similar at LOC from 1996 to 2004 and then ammonium increases while nitrate does not from 2005 to 2008. All ions dipped dramatically in 2009, which was a very wet summer, but then rebound upward in 2010.

A comparison of 72-hour back trajectories, using NOAA's HYSPLIT and North American Regional Reanalysis data, between relatively acidic vs. neutralized cloud and aerosol events measured at LOC will be presented. Initial results demonstrate that high sulfate-ammonium dominated occur when air masses have relatively longer flow paths from ammonia emission source regions.

* NEI 2008v2 at the Tier 1 level http://www.epa.gov/ttn/chief/trends/

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