Trends in cloud water sulfate and nitrate as measured at two mountain sites in the Eastern United States:

Regional contributions and temporal changes compared with regional changes in emissions, 1986–1999

James B. Anderson [a], Ralph E. Baumgardner, Jr. [b], Sandra E. Grenville [c]

[a] Center for Water and Wetlands Resources, The University of Mississippi Field Station, 15 Road 2078, Abbeville, MS. 38601, USA

[b] National Exposure Research Laboratory, US Environmental Protection Agency, Research Triangle Park, NC 27709, USA

[c] Air Quality Services, Inc., 1604 Brookwood Road, Jacksonville, FL 32207, USA

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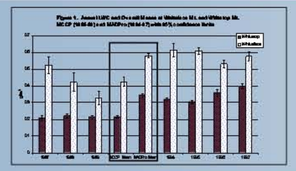
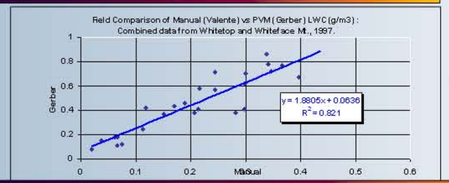
BACKGROUND

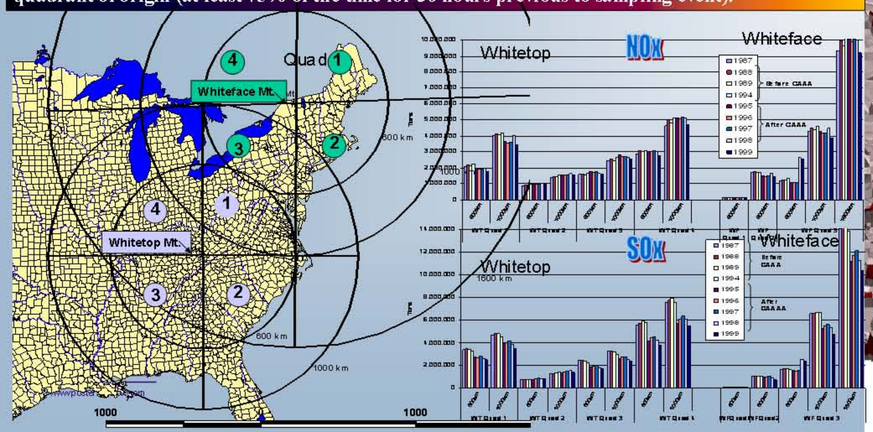
Air pollutant emissions in the US generally peaked in 1970, the year that the Clean Air Act was passed, and have declined since, except for oxides of nitrogen (NOx), which have remained steady or slowly increased. In 1995 Phase 1 of the Clean Air Act Amendments (CAAA) of 1990 went into effect resulting in a sharp drop in sulfur dioxide (SO_2) emissions in most areas. Pollutant concentrations measured in precipitation and ambient air generally reflected the changes in emissions in most areas of the eastern US. Only the southern Appalachian Mountain region did not see an improvement in precipitation acidity over the last decade

MADPro and MCCP Comparability: LWC method problem - MCCP corrected





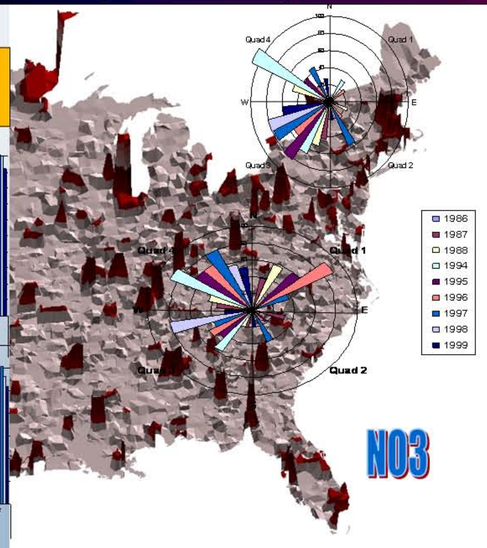
Cumulative annual county emissions are compared with average annual concentrations on Mts. Below are shown the emissions data, out to 600 and 1000 meters, segregated by back-trajectory quadrant of origin (at least 75% of the time for 36 hours previous to sampling event).

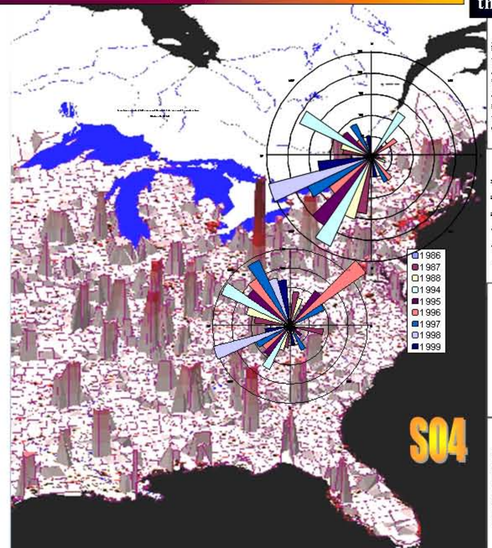


MATERIALS AND METHODS

Previous studies of cloud chemistry in the eastern US found no pattern of temporal trends, possibly because of large year-to-year variation in meteorology. In this paper, we present spatial and temporal trends in SO_4^{-2} and NO_3^- concentrations of cloud water samples collected in warm seasons only at two mountain sites (Whiteface Mountain, NY and Whitetop Mountain, VA). This analysis is based on a combined cloud chemistry data-set collected by EPA's Mountain Cloud Chemistry Program (MCCP) (1986–1989) and Mountain Acid Deposition Program (MADPro) (1994–1999). After MCCP Liquid Water Content values were corrected (see previous), sample concentrations were (1) normalized by Liquid Water Content (to reduce within-cloud variation) and (2) segregated into 90° arrival quadrants based on 36 h back trajectory analysis (to diminish between-cloud or meteorological variation). For each quadrant at the two sites, annual (12 month) county emissions of SO_2 and NOx were accumulated for all counties out to 600, 1000, and 1600 km, and these values were compared with the annual means (warm season only) of normalized SO_4^{-2} and NO_3^- concentrations in hourly samples of cloud water (segregated by back-trajectory) collected at each site.

Below are depicted annual average NO_3^- and $SO_4^{=}$ concentrations sampled at each mountaintop and segregated into back trajectory quadrants plotted as pollution roses on a map of the eastern US showing annual cumulative county emissions of NOx and SO2 (1989) as elevation.





RESULTS

For the period 1987–1999, Quadrant 3 (SW) for Whiteface Mt. and Quadrants 3 (SW) and 4 (NW) for Whitetop Mt. had the highest SO_2 emissions and showed the largest decline in SO_2 emissions after the CAAA was implemented. These same quadrants which had the largest decrease in emissions showed significant declines in cloud water SO_4^{-2} over the time period. NOx emissions were highest in Quadrant 3 for Whiteface Mt. and in Quadrants 1 and 4 for Whitetop Mt. Only in Quadrant 1 at Whitetop Mt. did NOx emissions decrease during the study period (1987–1999). Cloud water NO_3^- showed no consistent pattern at either mountain site with some quadrants having higher cloud water NO_3^- values after Phase 1 of the CAAA and other quadrants having little change in cloud water NO_3^- values.

Below shows cumulative annual emissions (tons of NOx and SO_2, respectively) out to 600 and 1000 km from each site compared with average yearly concentrations of nitrate and sulfate in cloud water sampled on each mountain. The last figure contrasts the average emissions and cloud conc. before (blue) and after (red) the activation of the Clean Air Amendments in 1994, for both N and S compounds.