

Heterogeneous Chemistry in the Middle Stratosphere : Theory and Observation

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The heterogeneous hydrolysis of N_2O_5 on the surface of concentrated sulfuric acid aerosols exerts a strong influence on the chemical composition of the stratosphere at mid-latitudes by dramatically shifting the balance among key radical families, depleting abundances of NO and NO_2 , and increasing concentrations of ClO, OH, and HO_2 (e.g., Cadle et al., 1975; McElroy et al., 1992; Wennberg et al., 1994). The heterogeneous hydrolysis of $BrONO_2$ proceeds rapidly on the surface of concentrated sulfuric acid aerosols and provides an additional sink for NO_x under conditions of highly perturbed aerosol loading (Hanson and Ravishankara, 1995).

Large enhancements in sulfate aerosol loading due to the eruption of Mt. Pinatubo provide an excellent test of our understanding of heterogeneous processes on the abundance of gases that affect ozone. Good agreement between predicted and observed changes in NO_x due to enhanced loading of aerosols has been reported at 20 km (Fahey et al., 1993) and a single measurement of NO_2 and HNO_3 at 24 km (Webster et al., 1994). However, models underestimate, by nearly a factor of 2, the observed reduction in column NO_2 and enhancement in column HNO_3 observed over Lauder, NZ (45°S) following the eruption of Mt. Pinatubo (Koike et al., 1994). Observed reductions in column ozone following the eruption appear to be underestimated by 2D models that allow for changes in circulation induced by the aerosol (Kinnison et al., 1994).

We will present a comparison between theory and observation for a variety of satellite (ATMOS, SAGE II), balloon-borne (PHOS, FIRS, MarkIV, Nagoya Univ chemiluminescence, SLS), and in situ (MER-2 compliment of instruments) observations of nitrogen, hydrogen, and chlorine radicals in the lower and middle stratosphere. Our study uses a constrained photochemical model previously used to examine a variety of in situ and remote measurements (e.g., McElroy et al., 1992; Salawitch et al., 1994). We will define, as a function of altitude, latitude, and level of aerosol loading, our understanding of perturbations to the major gases thought to regulate the abundance of ozone.

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