A. Title of Research Task:

Tropospheric Trace Gas Interactions with Aerosols

B. Investigators and Institutions:

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C. Abstract of Research Objectives

Tropospheric aerosols are of considerable environmental importance. They modify the radiative budget of Earth by scattering and absorbing radiation, and by providing nuclei for cloud formation. Additionally, they provide surfaces for heterogeneous and multiphase reactions that affect tropospheric chemistry. For example, Dentener and Crutzen (1993) showed that reactions of N₂O₅ and NO₃ with sulfate aerosols may significantly alter the tropospheric concentrations of NOₓ, O₃, and OH by converting NOₓ to HNO₃ which is rapidly removed by precipitation. Zhang et al. (1994) assumed these same reactions would occur on dust aerosols and showed that dust outbreaks may reduce NOₓ levels by up to 50%. Dentener et al. (1996) studied the possible effect of reactions on dust on sulfate, nitrate, and O₃ concentration. Heterogeneous and multiphase reactions on aerosols may also perturb the sulfur cycle the chlorine cycle and the bromine cycle. Because these reactions can release free chlorine and free bromine they might lead to the destruction of ozone in the marine boundary layer that may be important to include in models of tropospheric chemistry. The goal of our proposed work is to examine the role of heterogeneous and multiphase reactions in the tropospheric cycles of reactive nitrogen and sulfur.

D. Summary of Progress and Results (covering work from April, 1998, through 1999)

We initially examined the coupling between HNO₃, NH₃, and sulfate aerosol using a box-model simulation. We used the multi-component, size-resolved aerosol chemistry box model described by Jacobson (1996) and updated in Jacobson (1999). This model simulates inorganic chemistry in a system consisting of sulfate, nitrate, ammonium, chloride, sodium, organic carbon, elemental carbon, dust, and water. We applied the model under the assumption that sulfate, nitrate and ammonium are internally
mixed. The major assumption in this application is that the aerosol is at thermodynamic equilibrium with the gas phase. We first specified the size distribution of aerosol sulfate into 10 size bins. Then the aerosol chemistry model was used to calculate the gas/aerosol partitioning for nitrate and ammonium, so that chemical equilibrium was maintained in each size section. The take-up of water by each size section as a function of relative humidity was also modeled.

To examine the effect of different conditions more fully, we initialized a global scale version of the aerosol chemistry model using the three-dimensional monthly average distributions of aerosol sulfate [Chuang et al., 1997; Penner et al., 1994] and the predicted HNO₃ [Penner et al., 1994] from previous simulations of the GRANTOUR model [Walton et al., 1988; Penner et al., 1991]. We also obtained monthly average concentrations of NH₄⁺ plus NH₃ from the model of Dentener and Crutzen [1993]. We examined the amount of nitrate in aerosol under conditions of chemical equilibrium within \( \text{H}_2\text{SO}_4 + \text{NH}_3 + \text{HNO}_3 \) system and also within the \( \text{H}_2\text{SO}_4 + \text{NH}_3 + \text{HNO}_3 + \text{Dust} \) system and the \( \text{H}_2\text{SO}_4 + \text{NH}_3 + \text{HNO}_3 + \text{Dust} + \text{Sea Salt} \) system. The percentage of nitrate in aerosol increased from 11% to 33% and 63%, respectively, at the surface in the 3 systems. This shows the importance of including a fully coupled system of the reactive nitrogen cycle with dust and sea salt in order to calculate the effects of NOx on ozone, for example.

We also examined the effects of reactions of \( \text{SO}_2 \) on dust and sea salt. \( \text{SO}_4^{2-} \), production on dust and sea salt accounts for approximately 4.1% and 4.4% of the total annual average concentration of \( \text{SO}_4^{2-} \), respectively.

These results point to the importance of coupling tropospheric chemistry models with aerosol models. Nitrate partitioned to the aerosol phase will undergo different scavenging and deposition processes than nitrate partitioned only in the gas phase. Thus, it is important to continue to develop the capability to treat the coupling between aerosol and gas phase chemistry.

E. Journal Publications (Includes publications from our previous NASA-ACMAP sponsored research: Global Studies of the Sulfur Cycle Including the Influence of DMS and Fossil Fuel Sulfur on Climate and Climate Change which was funded during 1997 ). We also have two publications in progress from our research under the current project (listed below)


Penner, J.E., Chuang, C.C., and K. Grant, 1999: Climate Change and Radiative Forcing by Anthropogenic Aerosols: Research Findings During the Last 5 Years, La Jolla International School of Science, The Institute for Advanced Physics Studies, La Jolla, CA 92038-2946, March 29-30, 1999.


