"Stratospheric Heterogeneous Chemistry and Microphysics: Model Development, Validation and Applications"

Summary Report—July 1, 1994 to June 30, 1996

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Research Objectives: The objectives of this project were to define the chemical and physical processes leading to stratospheric ozone change that involve polar stratospheric clouds (PSCs) and the reactions occurring on the surfaces of PSC particles; to study the formation processes, and the physical and chemical properties of PSCs, that are relevant to atmospheric chemistry and to the interpretation of field measurements taken during polar stratosphere missions; to develop quantitative models describing PSC microphysics and heterogeneous chemical processes; to assimilate laboratory and field data into these models; to calculate the extent of chemical processing on PSCs and the impact of specific microphysical processes on polar composition and ozone depletion.

Summary of Research: During the course of the project, a new coupled microphysics/physical-chemistry/ photochemistry model for stratospheric sulfate aerosols and nitric acid and ice PSCs was developed and applied to analyze data collected during NASA's Arctic Airborne Stratospheric Expedition-II (AASE-II) and other missions (Drdla et al., 1992, 1993; Drdla, 1996). In this model, detailed treatments of multicomponent sulfate aerosol physical chemistry (see below), sulfate aerosol microphysics, polar stratospheric cloud microphysics, PSC ice surface chemistry (Tabazadeh and Turco, 1993a), as well as homogeneous gas-phase chemistry were included for the first time. In recent studies focusing on AASE measurements, the PSC model was used to analyze specific measurements from an aircraft deployment of an aerosol impactor, FSSP, and NOy detector (Drdla, 1996; Drdla et al., 1994). The calculated results are in excellent agreement with observations for particle volumes as well as NOy concentrations, thus confirming the importance of supercooled sulfate/nitrate droplets in PSC formation. The same model has been applied to perform a statistical study of PSC properties in the Northern Hemisphere using several hundred high-latitude air parcel trajectories obtained from Goddard. The rates of ozone depletion along trajectories with different meteorological histories are presently being systematically evaluated to identify the principal relationships between ozone loss and aerosol state.

Under this project, we formulated a detailed quantitative model that predicts the multicomponent composition of sulfate aerosols under stratospheric conditions, including sulfuric, nitric, hydrochloric, hydrofluoric and hydrobromic acids (Tabazadeh et al., 1994a,b). This work defined for the first time the behavior of liquid ternary-system type-1b PSCs. The model also allows the compositions and reactivities of sulfate aerosols to be calculated over the entire range of environmental conditions encountered in the stratosphere (and has been incorporated into a trajectory/microphysics model—see above). Important conclusions that derived from this work over the last few years include the following: the HNO_3 content of liquid-state aerosols dominate PSCs below about 195 K; the freezing of nitric acid ice from sulfate aerosol solutions is likely to occur within a few degrees K of the water vapor frost point; the uptake and reactions of HCl in liquid aerosols is a critical component of PSC heterogeneous chemistry (Turco and Hamill, 1982; Drdla, 1996). In a related application of this work, the inefficiency of chlorine injection into the stratosphere during major volcanic eruptions was explained on the basis of nucleation of sulfuric acid aerosols in rising volcanic plumes leading to the formation of supercooled water droplets on these aerosols, which efficiently scavenges HCl via precipitation (Tabazadeh and Turco, 1993b).

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