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GLOBAL CHANGE

ATMOSPHERIC CHEMISTRY PROGRAM

GLOBAL 3-D MODELLING OF ATMOSPHERIC OZONE IN THE FREE TROPOSPHERE AND THE STRATOSPHERE WITH EMPHASIS ON MIDLATITUDE REGIONS

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Objects: To use global chemical-transport models to study the chemical and dynamical processes that affect midlatitude stratospheric ozone and to quantify the budget of tropospheric ozone.

Approach: Four models will be improved and used: (1) a new version of our two-dimensional chemical-radiative-dynamical model with microphysical process of sulfate aerosols and polar stratospheric clouds (PSCs), and heterogeneous conversions on the surfaces of sulfate aerosols and PSCs; (2) the stratospheric version of three-dimensional off-line chemical-transport model (STARS) with a relatively high horizontal resolution (2.8 degree in latitudes) with a microphysical formation of PSCs; (3) the tropospheric version of three-dimensional off-line chemical-transport model (MOZART) with a details in the surface emissions and hydro-carbon reactions to estimate the tropospheric ozone budget and perturbations; (4) the intermediate model of the global and annual evolution of species (IMAGES) with a detailed chemical reactions but relatively lower resolutions. Model results will be compared with available data.

Results: The coupled chemical radiative, dynamical and microphysical 2-D model is used to assess the response of stratospheric ozone to the injection of sulfur following the eruption of Mt Pinatubo. Model calculation suggests that, during the first year (July/1991 to June/1992) following the volcanic eruption, the observed changes in the ozone amount integrated between 65oS and 65oN have been caused primarily by changes in the meridional circulation (associated with heating by the volcanic cloud in the tropics) and in the photolysis rate of molecules such as ozone (associated with backscattering of light by the cloud). During the second year after the

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eruption, as the aerosol has been dispersed at all latitudes and, in particular, has reached the polar region, the largest contribution to ozone reduction results from the heterogeneous chemical conversion of N2O5 and ClONO2 on the surface of the aerosol particles. Model calculations also suggest that the ozone decrease observed a few years after the eruptions of Mt. Pinatubo and El Chichon may have been unique in the Earth's history, and is directly linked to the emission in the atmosphere of industrially manufactured chlorofluorocarbons. For chlorine loadings typical of the pre-1980 period, the ozone column abundance should have increased after a large volcanic eruption. After 1980, as a result of growth in chlorine loading, the response of ozone became negative in winter at mid- and high latitudes. In the future, the response of ozone is expected to become positive again, if the production of chlorofluorocarbons is sufficiently reduced.

We have developed a global three-dimensional transport/chemical model of the stratosphere call ed STARS (Study of Transport And Chemical Reactions in the Stratosphere), which includes a representation of the formation of polar stratospheric clouds (PSCs) and detailed heterogeneous reactions on the surface of PSCs and sulfate aerosols. The formation of the observed springtime "Antarctic ozone hole" is well reproduced by the model. A maximum of 40% total ozone depletion occurs in October. Calculated ozone and chlorine concentrations are consistent with satellite observations. After the breakdown of the polar vortex in December, air with depleted ozone is transported to mid-latitudes in the Southern Hemisphere, resulting in a 2-4% ozone decrease at 50oS in December and a 1% decrease in the tropics. Ozone-poor airmasses are also transported to the troposphere, and produce a significant decrease (20-30%) in upper tropospheric ozone. Ice particles (Type II PSCs) sediment into the troposphere, producing a large decrease in the concentrations of stratospheric HNO3 and NO2. As a result, the conversion of ClO into ClONO2 after the evaporation of PSCs is reduced. The model shows that this process could enhance the ozone depletion by 20% during the month of October. The model is also able to reproduce the ozone minimum observed in Antarctica when the chlorine loading was as low as 0.6 pptv. Under these conditions, the polar ozone depletion caused by chemical processes is very small (maximum of 3%) in October. In November, the ozone concentration even increases above 22 km in response to PSC processes.

We have implemented in our 2-D model a chemical scheme describing more accurately the methane oxidation chain in the troposphere, and have estimated the change in the tropospheric ozone abundance due to a doubling in the methane concentration.

We have also completed the development of the IMAGES model, which describes the three-dimensional distribution of approximately 50 chemical compounds from the surface to the 50 mbar level. The model has been used to assess the impact of aircraft emissions on tropospheric ozone and to investigate potential causes for the recently observed decrease in CO abundances.

Most recently, we have completed the development of a global three-dimensional model (MOZART) which simulates the distribution of ozone and its precursors in the troposphere and lower stratosphere. The model, which includes approximately 40 chemical species and 120 chemical and photochemical reactions, is driven by winds and temperatures provided by the NCAR Community Climate Model (CCM-2). The spatial resolution is 2.8 degrees in longitude and latitude, with 18 levels in the vertical from the surface to 1 mb. Boundary layer exchanges, cloud convection, and aqueous phase chemistry are included in the model. Surface emissions are based on pre-established inventories.

Collaborations: We plan to collaborate with Lon Hood of University of Arizona to evaluate further the relative importance of chemistry and dynamics in producing midlatitudinal ozone trends.

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Dr. Marv Wesely
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Dear Dr. Wesely

According to your request during the 1996 annual meeting, we are pleased to send you several copies of recent published papers as well as the list of papers. The copies of our posters and their abstracts have been already given to you during the meeting.

The list of recent publications:

(1) **Tie, X.**, and **G. P. Brasseur,** The response of stratospheric ozone to volcanic eruptions: Sensitivity to atmospheric chlorine load, *Geophys. Res. lett., 22,* 3035-3038, 1995.

(2) De Rudder, A, N. Larsen, **X. Tie**, C. Granier, and **G. P. Brasseur,** Model study of polar stratospheric clouds and their effects on stratospheric ozone: Part I- Model description, *J. Geophys. Res.*, 101, 12567-12574, 1996.

(3) **Tie, X., G. P. Brasseur,** C. Granier, A. De Rudder, N. Larsen, Model study of polar stratospheric clouds and their effects on stratospheric ozone: Part II- Model results, *J. Geophys. Res.*, 101, 12575-12584, 1996.

(4) **Tie, X.**, and **G. P. Brasseur,** The importance of heterogeneous bromine chemistry in the lower stratosphere, *Geophys. Res. lett., 23,* 2505-2508, 1996.

(5) **Brasseur, P. G., X. Tie**, P. Rasch, and F. Lefevre, A three-dimensional simulation of

the Antarctic ozone hole: The impact of anthropogenic chlorine on the lower stratosphere and upper troposphere, Accepted by *J. Geophys. Res.* 1996.

Thanks.

Sincerely,

Guy Brasseur and Xuexi Tie

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