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**Summary of Research for FY-1995; Progress report**

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The object of this proposal is to study the reduction in mid-latitude stratospheric ozone and to estimate the budget of tropospheric ozone.

The product of this proposal include: (1) the estimation of dilution of air masses processed by polar stratospheric clouds inside the polar vortex during winter; and (2) the destruction of ozone via heterogeneous reactions on the surface of aerosol particles which are present at all latitudes, especially after large volcanic eruptions such as Mt. Pinatubo; (3) to quantify photochemical production and destruction of  $O_3$  in the free troposphere; (4) to quantify export of ozone from "polluted" to remote regions, and (5) to quantify cross-tropopause exchanges of  $O_3$  and other species.

The approach of this proposal is to use and to improve our two-dimensional and three-dimensional global chemical/dynamical models including: (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

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acrosols 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 microphysical formation of PSCs to study the role of transport on the mid-latitude ozone reduction; (3) the tropospheric version of a three-dimensional off-line chemical-transport model (MOZART) with detailed 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 detailed chemical reactions but relatively lower resolution to study the sensitivities of natural and anthropogenic perturbations on tropospheric ozone.

In 1995, several studies have been completed and are summarized in the following.

### 1. Stratospheric ozone depletion

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 calculations suggest that, during the first year (July/1991 to June/1992) following the volcanic eruption, the observed changes in the ozone amount integrated between 65°S and 65°N 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 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  $N_2O_5$  and  $ClONO_2$  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 named 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.

We have used this model to study the effects of springtime "Antarctic ozone hole" on midlatitude and upper tropospheric ozone. The calculation shows a maximum total ozone depletion of 40% over Antarctica in October. After the breakdown of the polar vortex in December, air with depleted ozone is transported to Midlatitude in the Southern Hemisphere, resulting in a 2-4% ozone decrease at Midlatitude 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. The model results also show that heterogeneous conversion of bromine reservoirs has an important effect on the formation of reactive chlorine, which could result an important effect on the ozone concentration at midlatitude, especially after the large volcanic eruptions of 1980's and 1990's.

### 1. Tropospheric ozone budget

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.

#### Publications:

**Tie, X., G. P. Brasseur, B. Briegleb, and C. Granier,** Two-dimensional simulation of Pinatubo aerosol and its effect on stratospheric ozone, *J. Geophys. Res.*, 99, 20545-20562, 1994.

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**Muller, J.-F., and G. P. Brasseur,** IMAGES: A three-dimensional chemical transport model of the global troposphere, *J. Geophys. Res.*, 100, 16445-16490, 1996.

**Brasseur, G. P.,** Atmospheric Impact of NOx emissions by subsonic aircraft: A three-dimensional model study, *J. Geophys. Res.*, 101, 1423-1428, 1996.

Granier, C., J.-F. Muller, S. Madronich, and G. P. Brasseur, Possible causes for the 1990-1993 decrease in the global tropospheric CO abundance: a three-dimensional study, submitted to *Atmospheric Environment*.

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