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TITLE A 3-D Dynamical/Chemical GCM for Simulating the Anthropogenical Effects on Ozone

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# **A 3-D Dynamical/Chemical GCM for Simulating the Anthropogenic Effects on Ozone**

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## **1. Introduction**

In spite of the significantly increased attention atmospheric ozone has received over the last two decades or so, a fully quantitative understanding of the three-dimensional ozone structure remains unavailable. In the past, one-dimensional (vertical column) models have provided most of the quantitative simulations of atmospheric chemical systems, because these models can treat large ensembles of chemical process economically. More recently, two-dimensional (meridional cross section) models have become popular, in part because of increasing computer capabilities. There has been little work using a sophisticated 3-D GCM coupled with a comprehensive photochemical model to study the chemical-dynamical interactions involving the ozone abundance. Namely, either the dynamics or the chemistry is too simplified in most of the existing models (see a review in Kaye and Rood, 1989). Relatively more advanced photochemistry has been included in the stratospheric models (dealing the vertical domain from 20 to 70 km above the surface) at the expense of tropospheric details (Kaye and Rood, 1989; Grose *et al.*, 1987). An important motivation for this research has been the desire to estimate the magnitude of changes in the ozone layer induced by anthropogenic perturbations such as the injection of oxides of nitrogen and chlorine compounds. Accordingly, the attempt has been to develop a complete but feasible photochemical scheme and to combine it with the Los Alamos National Laboratory (LANL) 3-D GCM (Malone *et al.*, 1986).

## **2. The Model**

We have utilized the NCAR CCM0A (Malone *et al.*, 1983) modified to predict the effects of smoke on the global atmosphere. Particular modifications which benefit this current research include: a) stability dependent vertical diffusion, b) subsurface storage and diffusion of heat and

moisture, c) annual and diurnal cycles of sunlight, d) finer zoning in stratosphere, e) Lagrangian transport of aerosols, and f) improved 3-D transport scheme for chemical species.

The model results for the unperturbed atmosphere have also been validated against the long-term climate analyzed by ECMWF (European Center for Medium-Range Weather Forecast). The model history tape for a summertime condition has provided meteorological conditions to the PNL's (Pacific Northwest Laboratory) Global Chemistry Model to evaluate the role of fair weather cumulus clouds in the long-range transport and chemistry of methane (Berkowitz *et al.*, 1989).

The complete photochemical system in the stratosphere includes several dozen trace species and hundreds of photochemical processes. Our approach toward simplifying the photochemical calculations utilizes a "family" scheme (e.g., see Turco and Whitten, 1977) in which the key chemical constituents are organized into groups that have relatively short inter-species equilibration times (compared to the computational time steps used in the GCM). Within a family, the constituent members can thus be assumed to reach a quasi-equilibrium state or to have simple time-dependent behaviors that are amenable to analytical representation. On the other hand, the family itself is selected to have a photochemical time constant that is longer than (or equivalent to) the computational time step. The families and species considered are summarized in Table 1. The essential photochemical processes and rates are adopted from DeMore *et al.* (1987) and Turco (1975). The present chemistry set constitutes an initial representation for the principal atmospheric chemical families.

The fundamental equations that determine the time evolution of the family concentrations can be derived from the complete set of photochemical continuity equations by making significant simplifications. For example, reactions of secondary importance in controlling the chemical transformation (production and loss) rates of families are neglected; less important families in stratosphere (e.g., sulfur compounds) are omitted; and continuity equations for individual family members are summed over the family to eliminate reactions that simply cycle family constituents (i.e., intra family conversion processes). Care must be taken in any simplifying approach to identify all of the key rate limiting reactions (for example, those which control the odd oxygen loss rate) and dominant cycling processes that fix the relationship between family members, in Table 2.

### 3. Model Performance

To assure ourselves that the "family" photochemical scheme is numerically stable and computationally feasible, the photochemical scheme described above has been implemented in the LANL GCM to test the effects of  $\text{NO}_x$  deposited by nuclear fireballs on ozone depletion under a post-war condition by Kao *et al.*, 1989. In essence, the large amount of instantaneous injection of NO during the explosions rapidly converts into  $\text{NO}_x$  which, in turn, produces a large amount of  $\text{N}_2\text{O}_5$  and  $\text{HNO}_3$ .  $\text{N}_2\text{O}_5$  is later involved with the transfer of  $\text{NO}_x$ , and  $\text{HNO}_3$  can be photodissociated into  $\text{HO}_x$ . Both  $\text{NO}_x$  and  $\text{HO}_x$  can react with odd-oxygen family members to deplete ozone. The net results on ozone abundance are illustrated in Figs. 1 and 2. During this highly nonlinear process, the modified circulation due to the solar heating of smoke contributes to push the  $\text{NO}_x$  rapidly upward into the levels where the maximum ozone concentration resides. The heating of the stratosphere by rising smoke also speeds up several catalytic chemical reactions. Furthermore, this chemical/dynamical model is run with an "outline" fusion so that the diurnal cycle of the sunlight associated with photodissociation is fully implemented and the effects of ozone abundance on the radiative transfer can be included. In contrast, most of the existing model are run in an "off-line" fashion in which the chemical module is driven by either the analyzed data of winds and temperature or the data set generated from a GCM.

#### 4. Future Work

We will extend the top of the vertical domain from the present 33 km to about 70 km to cover the whole stratosphere. The photochemical processes and their interactions with the transport mechanism will then be better resolved. In particular, the photodissociation of  $\text{O}_2$ , which is the dominant source term for odd-oxygen family, will appropriately be included. Secondly, the numerical difficulties due to the wave reflection from the rigid top boundary can be reduced. We will also add families for chlorine, bromine, and hydrocarbon species, bringing the total number of constituents to roughly 65; i.e., a complete photochemical set and we will formulate a comprehensive set of photochemical processes for the GCM using the recommendation of the Panel of Chemical Kinetics and Photochemical Data Evaluation (Demore *et al.*, 1987).

#### Acknowledgements

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**TABLE 1. PHOTOCHEMICAL CONSTITUENTS**

<b>Family</b>	<b>Active Species</b>	<b>Tracer Species</b>	<b>Fixed/Source Species</b>
Odd-Oxygen (O <sub>x</sub> )	O, O <sub>3</sub> , O( <sup>1</sup> D)	O <sub>3</sub>	O <sub>2</sub>
Nitrogen Oxides (NO <sub>x</sub> )	NO, NO <sub>2</sub> , NO <sub>3</sub>	NO <sub>x</sub>	N <sub>2</sub> /N <sub>2</sub> O
Dinitrogen Pentoxide	N <sub>2</sub> O <sub>5</sub>	N <sub>2</sub> O <sub>5</sub>	—
Nitric Acid	HNO <sub>3</sub>	HNO <sub>3</sub>	—
Water Vapor	H <sub>2</sub> O	H <sub>2</sub> O	H <sub>2</sub> , CH <sub>4</sub>
Hydrogen Radicals (HO <sub>x</sub> )	OH, HO <sub>2</sub>	HO <sub>x</sub>	—/H <sub>2</sub> O
Hydrogen Peroxide	H <sub>2</sub> O <sub>2</sub>	H <sub>2</sub> O <sub>2</sub>	—

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### Figure Captions

Fig. 1. (a) Initial zonal-averaged ozone concentrations in units of molecules  $\text{cm}^{-3}$ . The contour lines are scaled by  $10^{-10}$ . (b) the same as (a) except at 20 days after the initial condition. The decreased concentrations in the Northern Hemisphere are due to both the chemical reactions associated with  $\text{NO}_x$  and the transport mechanism associated with the heating anomalies due to the absorption of sunlight by smoke. The increased concentrations in the Southern Hemisphere are due to the transport.

Fig. 2. (a) Initial ozone column number density in units of molecules  $\text{cm}^{-2}$ . The contour lines are scaled by  $1 \times 10^{-18}$ , (b) the same as (a) except at 20 days after the initial condition. The readers are referred to the caption of Fig. 1.

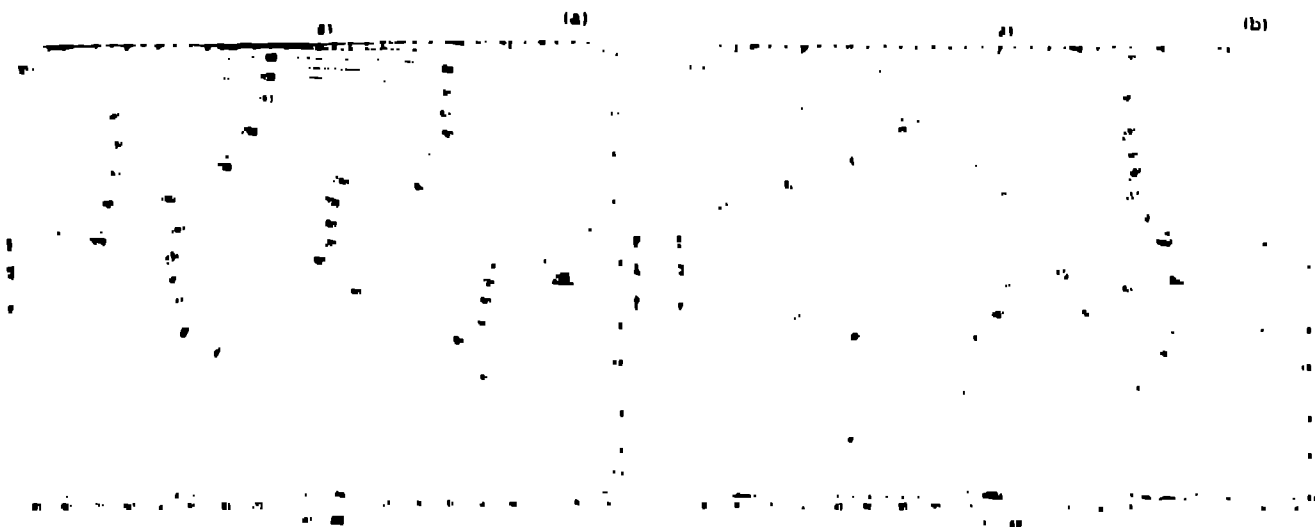


Fig. 1.

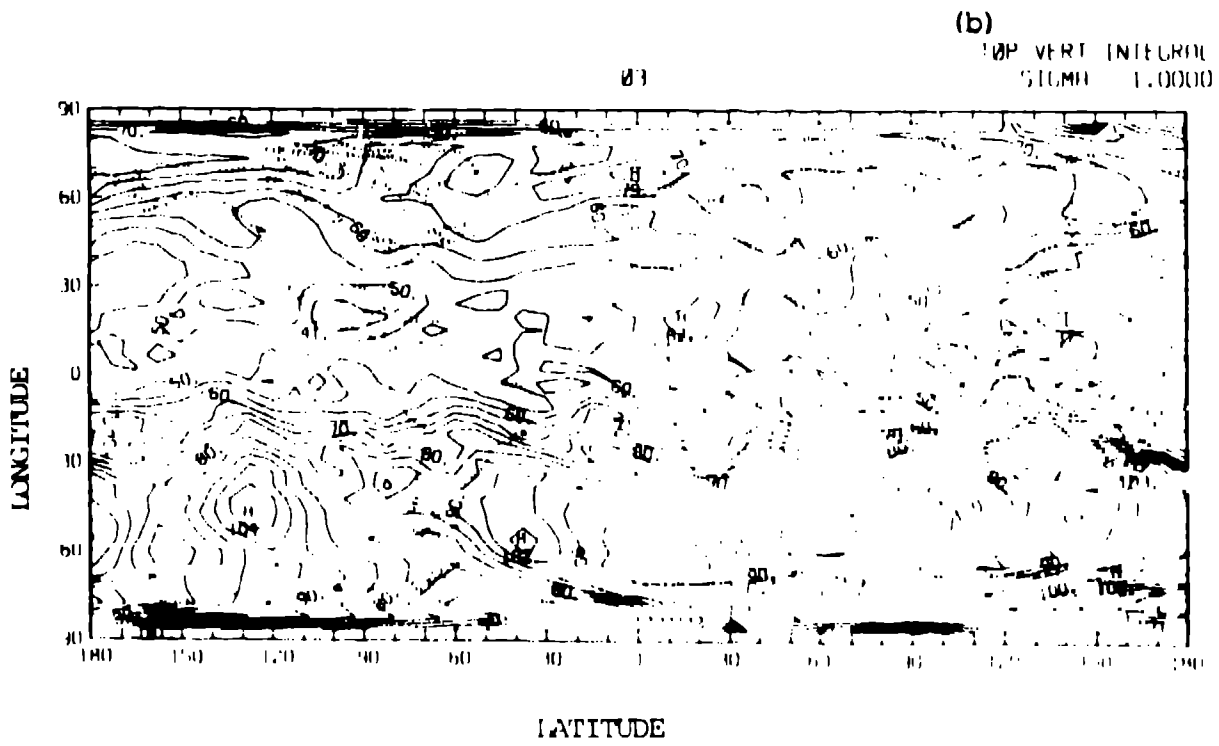
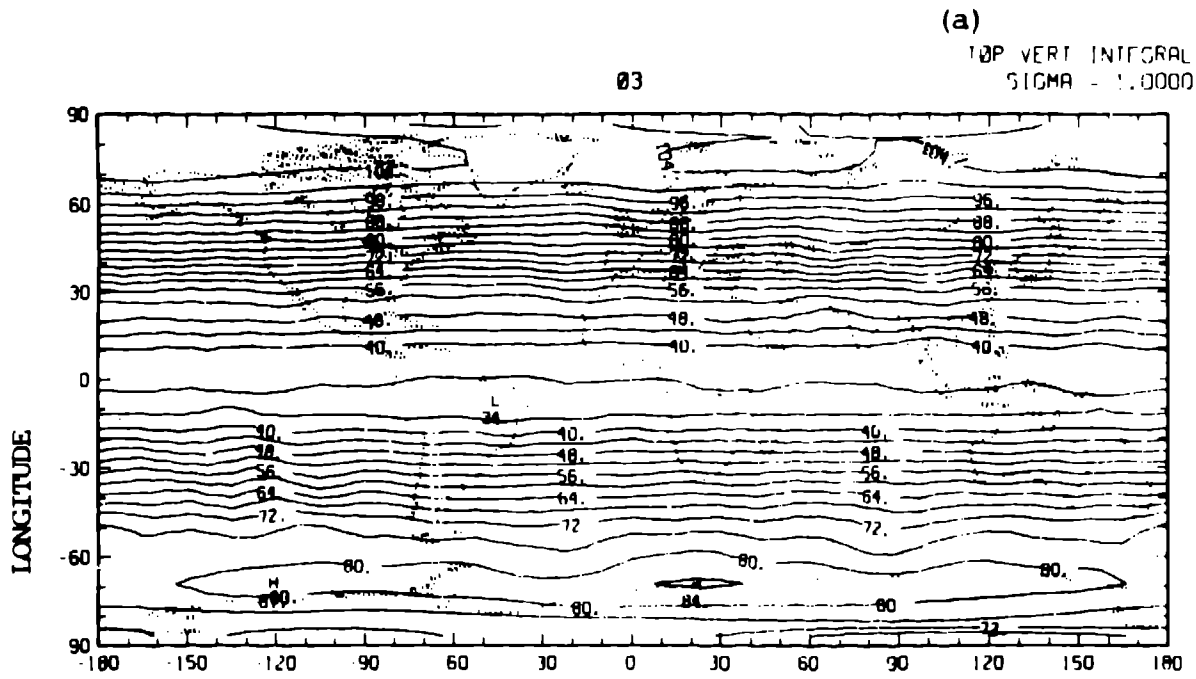


Fig. 2.