

NASA-CR-205073

*11-97-012
0-11
11-97-012*

Semi-Annual Report on
Coupling Processes
between
Atmospheric Chemistry and Climate
NAS5-97039

by

M. K. W. Ko, Debra Weisenstein, Run-Li Shia, and N. D. Sze

Atmospheric and Environmental Research, Inc.

840 Memorial Drive

Cambridge, MA 02139

Prepared for

NASA Atmospheric Chemistry Modeling and Data Analysis Program

July 15, 1997

Table of Contents

Abstract.....	2
Progress in the Past Six Months.....	3
(I) Two-dimensional Chemistry-Transport Model	3
(I.1) Model Enhancements	3
(I.1.1) Temperature distribution.....	3
(I.1.2) Incorporation of heterogeneous reactions on polar stratospheric clouds	3
(I.1.3) Mixing rate across the tropical barrier in the 2-D CTM	4
(I.1.4) Update of reaction rate constants	5
(I.2) Studies and Publications related to the 2-D CTM	5
(I.2.1) ODP of CH ₃ Br	5
(I.2.2) Chlorine loading of short-lived species	6
(I.2.3) Mixing rates in the 2-D CTM.....	7
(II) 2-D Sulfate model	7
(II.1) Model Enhancement	7
(II.2) Related studies and publications.....	8
(II.B.1) Documentation of the AER 2-D Sulfate Model	8
(II.B.2) Effect of Sulfur Emissions from HSCT on the Global Sulfate Layer	8
(III) 2-D Interactive model	10
(III.1) Model Enhancement	10
(III.2) Related Studies and Publications.....	10
(III.2.1) Analysis of wave structure using UARS observed N ₂ O and CH ₄	10
(III.2.2) The AER 3-wave model	11
(IV) 2-D Interactive Chemistry-Climate model (ICCM).....	11
(IV.1) Model Description.....	11
(IV.2) Studies and Publications	12
(IV.2.1) Effect of ozone depletion on climate.....	12
(IV.2.2) Manuscript documenting The 2-D Interactive Climate Chemistry Model	12
(V) Assessment Related Activities.....	13
(V.1) The Second Model and Measurement Workshop	13
(V.2) Ozone Assessment Report	14
(V.3) IPCC Report on Effect of Aviation	14
(VI) The POLARIS Campaign.....	14
Anticipated Activities in the next six months.....	15
References.....	16

Abstract

This is the first semi-annual report for NAS5-97039 summarizing work performed for January 1997 through June 1997. Work in this project is related to NAS1-20666, also funded by NASA ACMAP. The work funded in this project also benefits from work at AER associated with the AER three-dimensional isentropic transport model funded by NASA AEAP and the AER two-dimensional climate-chemistry model (co-funded by Department of Energy).

The overall objective of this project is to improve the understanding of coupling processes between atmospheric chemistry and climate. Model predictions of the future distributions of trace gases in the atmosphere constitute an important component of the input necessary for quantitative assessments of global change. We will concentrate on the changes in ozone and stratospheric sulfate aerosol, with emphasis on how ozone in the lower stratosphere would respond to natural or anthropogenic changes. The key modeling tools for this work are the AER two-dimensional chemistry-transport model, the AER two-dimensional stratospheric sulfate model, and the AER three-wave interactive model with full chemistry.

Progress in the Past Six Months

(I) Two-dimensional Chemistry-Transport Model

(I.1) Model Enhancements

(I.1.1) Temperature distribution

In previous versions of the AER 2-D model, temperature-dependent reaction rate constants were computed using $k(\langle T(\theta, z, t) \rangle)$ where $\langle T(\theta, z, t) \rangle$ is the zonal-mean temperature at latitude θ , altitude z , and time of the year t obtained by interpolating monthly zonal-mean temperature files. To account for the effect of longitudinal and temporal variations in temperature on photochemical reaction rates, monthly zonal mean temperature statistics were compiled for the 1979-1986 time period based on data from the NCEP/NCAR reanalysis project (Kalnay et al., 1996). Probability distribution functions, $P_{(\theta, z, M)}(T)$, defined to represent the probability of encountering the temperature T at latitude θ , altitude z , during the month M were created by combining daily data for each month and in each zonal band for the 8 year period. These distributions were found to be much broader in winter than in summer at mid and high latitudes. Temperature-dependent reaction rates are now calculated in the model by integrating the rate times the temperature probability for each 1 K of the distribution

$$\langle k_{\text{eff}} \rangle(\theta, z, t) = \int_0^{\infty} k(T) P_{(\theta, z, M)}(T) dT$$

$\langle k_{\text{eff}} \rangle(\theta, z, t)$ is now used in the photochemistry calculation instead of $k(\langle T(\theta, z, t) \rangle)$. The rates which are most sensitive to temperature variations are the heterogeneous reactions such as $\text{ClNO}_3 + \text{HCl}$ and $\text{HOCl} + \text{HCl}$ that occur on sulfate aerosols.

(I.1.2) Incorporation of heterogeneous reactions on polar stratospheric clouds

A parameterization for calculating the surface areas of type I and type II polar stratospheric clouds (PSCs) has been added to the AER 2-D model (Weissenstein et al., 1997b). The parameterization is based on equilibrium vapor pressure calculations rather than microphysics and is independent of the sulfate aerosol density. The local temperature determines the equilibrium vapor concentration of HNO_3 and H_2O . Excess HNO_3 and H_2O are assumed to form nitric acid trihydrate (NAT) particles with $0.5 \mu\text{m}$ radius. At lower temperature, excess water condenses to form ice particles. The radius of ice particles is calculated based on the quantity of water available for condensation and the number of

NAT particles per cubic centimeter. The temperature distribution described above is applied such that we calculated a NAT and ICE surface area at each temperature in the distribution, multiply by the appropriate heterogeneous chemical rate at the same temperature, and sum over the distribution. Our preliminary results with PSCs indicate reasonable calculated surface area. However, since the transport circulation in the 2-D model does not isolate air completely in the southern polar vortex, the Antarctic ozone hole is not well-represented. HSCT assessment results show greater ozone loss due to aircraft in the northern hemisphere when PSCs are included.

(I.1.3) Mixing rate across the tropical barrier in the 2-D CTM

The tropical barrier reduces the mixing between the tropics and mid-latitudes in the lower stratosphere and, thus has significant effects on tracer distribution in the stratosphere. Therefore, it is important for the 2-D CTM to have realistic exchange rates between the tropics and mid-latitudes. The exchange rate is determined by the eddy diffusion coefficient K_{yy} in the model.

In an earlier study, we introduced a tropical pipe by reducing the K_{yy} in the tropics from 0.3 to 0.03 (10^6 m²/sec) (Weissenstein, et al. 1996) in order to better simulate the model calculated NO_y/O_3 ratio at the tropics. Recently, in situ measurements of chemical species with a wide range of local lifetimes have been used to quantify the air exchange rate between the tropics and mid-latitudes in the lower stratosphere from the tropopause to around 21 km. It is found that the mid-latitude air is entrained into the tropical lower stratosphere with a replacement time scale of 10-18 months (Minschwaner et al., 1996; Volk et al., 1996; Greco, et al., 1996). Meanwhile, Schoeberl et al. (1997) estimate the mixing time between the tropics and mid-latitudes in the 20-28 km region to be at least 18 months using the QBO signals in $\text{N}_2\text{O}/\text{CH}_4$ ratio and tropical winds from UARS measurements. These results demonstrate that the tropical barrier allows a moderate penetration.

The horizontal diffusion fluxes and mixing ratios of CCl_4 , CF_2Cl_2 , CFCl_3 , CH_3CCl_3 , CH_4 , HNO_3 , N_2O , NO_y , and O_3 calculated in the AER 2-D model are used to derive the exchange rates between the tropical and mid-latitude lower stratosphere in the model. The exchange rate for the original model (global diffuser model) with a K_{yy} of 0.3×10^6 m²/sec in the lower tropical stratosphere is calculated to be about 5-6 months. The tropical pipe model results show a much slower exchange rate of 38-60 months. Our studies showed that values for K_{yy} of 0.13×10^6 m²/sec from the tropopause to 21 km and 0.07×10^6 m²/sec from 21 km to 35 km in the model (leaky pipe model) will generate exchange rates

around 14 months below 20 km and 18 months in the 20-30 km region, in agreement with estimates from measurements.

The model calculated lifetimes for species like N₂O and CFCs in the leaky pipe model are reduced by about 10% compared to the global diffuser model. Our model results show that the calculated ozone response to HSCT engine emissions is also smaller in the leaky pipe model. Some of these results were presented at the 1996 Winter AGU Meeting and the 1997 NASA/AEAP annual meeting and we are writing a paper for publication.

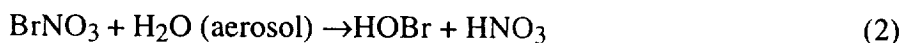
(I.1.4) Update of reaction rate constants

During May and June, chemical rates used in the 2-D CTM were updated according to JPL-97 (DeMore et al., 1997). There were major changes in photolysis cross-sections for HOBr, BrNO₃, ClONO₂, and BrCl. The JPL-97 changes resulted in more ozone from 20 to 35 km and less ozone elsewhere, though differences are less than 3% at any point. Bromine partitioning was changed significantly, with less HOBr and more BrO and BrNO₃ in the stratosphere.

(I.2) Studies and Publications related to the 2-D CTM

(I.2.1) ODP of CH₃Br

The AER 2-D CTM was used to compute the Ozone Depletion Potential (ODP) and the Bromine Efficiency Factor (BEF) of CH₃Br. These calculations, carried out before March 1997 have incorporated reaction rates from JPL-94 (DeMore et. al., 1994) along with new laboratory data which have been published concerning the following reactions:



Sander et al. (1997) reported a rate constant for reaction (1), the rate limiting step of the BrO_x/HO_x cycle, which is effectively a factor of two smaller at stratospheric temperatures than the present JPL-94 rate. A new expression for the sticking coefficient for heterogeneous reaction (2) was given by Hanson et al. (1996). The sticking coefficient of

reaction (2) is now dependent on temperature and water vapor, with a maximum value of 0.8 in cold polar regions, replacing a constant sticking coefficient of 0.4 (Hanson and Ravishankara, 1995). Photolysis cross sections for reaction (3), recommended by JPL-94, are based on measurements by Spencer and Rowland (1978) and assume a 100% yield via channel (3b). Nickolaisen and Sander (1997) indicate a probable product branching for reaction (3) with 74% proceeding along branch (3a) and 26% along (3b). Note that in contrast to (3b), reaction (3a) does not lead to ozone loss (i.e. a null cycle). We used the AER 2-D chemistry transport model (CTM) to assess the implications of the above reaction rate changes on the ODP and BEF of CH₃Br. The calculated CH₃Br ODP and BEF, based on an atmospheric lifetime of 0.84 years, from the AER 2-D CTM using JPL-94 are 0.36 and 44, respectively. Calculated values of the ODP and BEF with the updated rate constants are 0.40 and 49, respectively, showing a 9.9% increase from the JPL-94 calculated ODP and BEF. Detailed comparisons of the two cases indicate that changes in the BrO and HOBr concentrations are the primary cause for the 9.9% increase. These changes are dominated by reaction (2), while reactions (1) and (3) combine to reduce reaction (2)'s impact. During the summer months, a 5 -10% increase in BrO and a greater than 30% decrease of HOBr for altitudes greater than 24 km are seen. Winter values show similar results. JPL (1997) has adapted the rates discussed above for reactions (1), (2) and (3). Thus, the most up-to-date calculated CH₃Br ODP and BEF is from the combination of reactions (1), (2) and (3) which gives values of 0.40 and 49, respectively. We plan to recalculate the ODP and BEF with the full set of rate coefficients from JPL-97 (DeMore et al., 1997). Note that an additional recommendation in JPL-97 concerns the photolysis cross-section of HOBr, which was not included the analysis reported here.

A manuscript will be prepared to discuss the implications from the JPL-97 rate recommendations.

(I.2.2) Chlorine loading of short-lived species

Current methods for estimating the concentrations of inorganic chlorine/bromine species (Cl_y/Br_y) in the stratosphere due to decomposition of tropospheric source gases assume that the Cl_y/Br_y concentration in the stratosphere is determined mainly by the balance between production from in situ oxidation of the source gases in the stratosphere and removal by transport of Cl_y/Br_y out of the stratosphere. The rationale being that, for source gases whose lifetime is of order several months or longer, the concentration of Cl_y/Br_y in the troposphere is small because they are produced at a relatively slow rate and also removed efficiently by washout processes. As a result of the small concentration, the rate at which Cl_y/Br_y are transported to the stratosphere is expected to be small compared to

the in situ stratospheric production. Thus, the transport of Cl_y/Br_y from the troposphere contributes little to the stratospheric concentration. In contrast, the origin of stratospheric Cl_y/Br_y from reactive source gases with tropospheric lifetimes comparable to the washout lifetime of Cl_y/Br_y (of order 10-30 days) in the troposphere is distinctly different. The in situ source in the stratosphere is expected to be significantly smaller because only a small portion of the source gas is expected to survive the troposphere to be transported into this region. At the same time, these short-lived source gases produce appreciable amounts of Cl_y/Br_y in the troposphere such that transport to the stratosphere offers a larger source for stratospheric Cl_y/Br_y than in situ production. Thus, for reactive source species, simple methods of estimating the concentration of stratospheric Cl_y/Br_y that ignore the tropospheric contribution will seriously underestimate the loading. Therefore, estimation of the stratospheric Cl_y/Br_y loading requires not only measurements of tropospheric source gases but also measurements of Cl_y/Br_y at the tropopause. We have further examined this mechanism by using results from a two-dimensional chemistry-transport model. However, in view of the importance of tropospheric transport on stratospheric loading, the detailed values should be further evaluated using a three-dimensional model with appropriate treatment of convective transport. A manuscript (Ko et al., 1997) was submitted to Journal of Geophysical Research for review and is currently being revised in response to the reviewers' comments.

(I.2.3) Mixing rates in the 2-D CTM

This work was discussed above in section (I.1.3). A manuscript was submitted to Journal of Geophysical Research (Shia et al., 1997b).

(II) 2-D Sulfate model

(II.1) Model Enhancement

Temperature probability statistics, as described above in (I.1.1), have also been applied in the AER 2-D sulfate model to the calculation of homogeneous nucleation rates. Nucleation rates are very sensitive to temperature. Using only zonal mean temperatures, nucleation is calculated to occur only in the tropical lower stratosphere/upper troposphere and in a narrow region near the tropopause at all latitudes. With a temperature distribution, nucleation is calculated to occur in a much wider area, including polar regions in winter from 5 to 45 km. The global nucleation rate is enhanced by a factor of 10 and the stratospheric nucleation rate by a factor of 3 with inclusion of a temperature distribution. Aerosol surface area is also enhanced by 20% in the lower stratosphere.

(II.2) Related studies and publications

(II.B.1) Documentation of the AER 2-D Sulfate Model

A paper describing the AER 2-D sulfate model appeared in JGR in June of 1997. This paper (Weisenstein et al., 1997a) provides both a detailed description of the chemical and microphysical features of the model and shows the calculated aerosol concentrations and properties under background and volcanically-perturbed conditions. Sulfate precursor species included in the model are H_2S , CS_2 , DMS, OCS, and SO_2 . Aerosol particles in 40 size bins from 0.39 nm to 3.2 μm are modeled, assuming that all aerosols are strictly binary water-sulfuric acid solutions without solid cores. Microphysical processes included are homogeneous nucleation, condensation and evaporation, coagulation, and sedimentation. The main source of condensation nuclei for the stratosphere is new particle formation by homogeneous nucleation in the upper tropical troposphere. Though most stratospheric aerosol models have assumed OCS oxidation to be the main source of stratospheric sulfur, we have found this source to be inadequate to explain observed background aerosol loading. We find that rapid convective transport of surface-produced sulfur dioxide to the middle and upper troposphere can provide an additional source of stratospheric sulfur, resulting in better agreement between the model and observations. Our model is also successful at reproducing the magnitude of the stratospheric aerosol loading following the Mt. Pinatubo eruption, but the calculated rate of decay of aerosols from the stratosphere is faster than that derived from observations.

(II.B.2) Effect of Sulfur Emissions from HSCT on the Global Sulfate Layer

One application of the AER sulfate model has been to evaluate the possible impact of emission of sulfur from high-speed civil transport (HSCT) aircraft operating in the stratosphere. A hierarchy of models was used to investigate the impact of processes in the wake of an aircraft on the calculated global ozone response to sulfur emissions. The core model for this study was a version of the AER box model modified for calculation in the far wake, including a wake dilution module and the microphysical module from the 2-D sulfate model. We follow the evolution of aircraft emissions from the nozzle plane using three numerical models: the Standard Plume Flowfield-II/Plume Nucleation and Condensation model (SPF-II/PNC) used from the nozzle plane to 8 seconds after emission, the far wake model used from 8 seconds to one week after emission, and the AER global 2-D chemistry-transport/sulfate model run to steady state. Particle measurements in the wake of the Concorde (Fahey et al., 1995) are used to place constraints on sulfur oxidation processes in the engine and wake. Assuming similar characteristics for the proposed HSCT fleet, the global ozone response is then calculated by the 2-D model. To explain the Concorde

measurements, we consider scenarios with different fractions of SO_3 (2, 20 and 40%) in the sulfur emissions at the nozzle plane, and also the possibility of other unknown heterogeneous or homogeneous oxidation of SO_2 in the wake. We adopt an HSCT scenario with emission indices (EI) of NO_x and sulfur equal to 5 and 0.4, respectively, and a cruise speed of Mach 2.4 (Stolarski and Wesoky, 1993). Using the model-calculated wake processing of sulfur emissions under the above assumptions and constrained by the Concorde particle measurements, we calculate a range of ozone depletion at 40-50°N of 0.75 - 1.0%, a narrower range than that calculated by Weisenstein et al. (1996). Our analysis shows that the global ozone response is more sensitive to the assumed partitioning of sulfur emissions between SO_2 and SO_3 at the nozzle plane than to the wake dilution rate. This study, by Danilin et al. (1997) is accepted for publication in JGR.

Another paper dealing with the impact of sulfur emissions from HSCT aircraft on global ozone intercompares results from four independently-formulated sulfate aerosol models (Weisenstein et al., 1997b). These models are the AER model, the Novosibirsk State University model (in Russia), the Universita degli Studi-L'Aquila model (in Italy) and the University of Cambridge model (in the U.K.). Emission scenarios are from Stolarski et al. (1995) and assumptions regarding the form of emitted sulfur are similar to Weisenstein et al. (1996). Rather than modeling chemical and microphysical processes in the aircraft wake, the approach here is to assume that either 0%, 10%, or 100% of the emitted sulfur is in the form of aerosol particles of 10 nm radius at the point when the emitted material is diluted to grid-box size (a time period of approximately one week). All models show much greater changes in aerosol surface area when emissions are assumed to be 100% particles rather than gas phase SO_2 . Differences between models are deemed to be due mainly to differences in model transport, though differences due to aerosol size resolution are also explored. Ozone perturbations with $\text{EI}(\text{NO}_x)=5$, $\text{EI}(\text{H}_2\text{O})=1230$, and $\text{EI}(\text{SO}_2)=0.4$ are compared between the models. Though model differences are significant, the two models which include a temperature distribution to calculate heterogeneous reaction rates calculate significantly greater ozone depletion. Sensitivities to polar stratospheric clouds, background chlorine amount, additional heterogeneous reactions, and background aerosol loading are also explored. This paper has been submitted to JGR, been reviewed, and a revised manuscript submitted.

(III) 2-D Interactive model

(III.1) Model Enhancement

The current version of the AER 3-wave interactive model has a coarse vertical resolution of 3.5 km. To meet the future requirement for assessment studies, the resolution is being refined to match the fine resolution of 1.2 km for the AER 2-D CTM. The model with fine resolution is running on a SGI computer which is 8 time faster per CPU than the computer used for calculations with the low resolution version. The results show some differences in the temperature fields compared with that calculated in the model with coarse resolution. We are examining the causes of the differences.

(III.2) Related Studies and Publications

(III.2.1) Analysis of wave structure using UARS observed N₂O and CH₄

The derivation of eddy diffusion coefficients for chemical species in the AER 3-wave interactive model is based on the linear eddy equation for tracers with a crucial assumption that the eddy tracer concentration satisfies the linear wave equation so that the time and the longitudinal dependence of the eddy tracer wave is exactly the same as that of the eddy meridional velocity (See, e.g. Strobel, 1981; Shia, et al., 1997a). If the equation is valid, the ratio of the amplitude of wave in the tracer distribution divided by local latitudinal gradient of zonal mean mixing ratio should be the same for different long-lived species. We used UARS/CLAES N₂O and CH₄ data to examine whether this is valid. The UARS 3B data supplies the time series describing the waves for different tracers for wave number $m = 0$ to 6, at the selected latitudes and altitudes. We have examined several time periods in the lower stratosphere in the northern hemisphere for wave numbers 1 to 3. In the regions where the wave amplitudes are sufficiently large, the wave amplitudes scaled by the local latitudinal gradient of zonal mean mixing ratio for N₂O and CH₄ are approximately equal. This means that in those regions the assumption for deriving the eddy diffusion coefficient is valid.

(III.2.2) The AER 3-wave model

The original manuscript for the 3-wave model was submitted to Journal of Atmospheric Science 18 months ago. It was not accepted for publication after one revision. We submitted a revised version of the manuscript to the Journal of Geophysical Research (Shia et al., 1997a).

(IV) 2-D Interactive Chemistry-Climate model (ICCM)

(IV.1) Model Description

The 2-D ICCM is based on coupling two existing models (or ICCM modules) developed at AER: the refined seasonal-radiative-dynamical climate model (Wang et al., 1990; Molnar et al., 1994, and MacKay et al., 1997a) and the interactive chemistry-dynamics model (Ko et al., 1993 and Schneider et al., 1993).

The climate model and the chemistry model have the same meridional resolution, but different vertical coordinates. The climate module has better representation of the troposphere with finer tropospheric vertical resolution, more detailed atmospheric and surface physics, and separate land and ocean sectors in each zone. The chemistry-dynamics module has better representation of the stratosphere with finer stratospheric vertical resolution, and explicit calculation of heat transport and mean residual circulation. Because the physics and numerics of the two modules are different, we use a "modular" method to couple them. This method allows the two modules to run in parallel, and to exchange information continuously during long time integrations. Specifically, we use the chemistry module stratospheric temperatures and trace gas concentrations as input for the climate module during the time integration by replacing the stratosphere temperature field in the climate module by the temperature calculated from the chemistry-dynamics module. We also use the climate module tropospheric temperatures as input for the chemistry module. Since Newtonian cooling is used in the troposphere of the S93 chemistry-dynamics module, with climatological temperature (Cunnold et al., 1975) used as the "target" temperature, we use the temperature calculated from the climate module to replace the climatological "target" temperature. This is equivalent to forcing the tropospheric temperature gradually toward that calculated by the climate module. This data exchanging procedure provides the coupling of the two modules.

(IV.2) Studies and Publications

(IV.2.1) Effect of ozone depletion on climate

In order to study the potential climatic effects of the ozone hole more directly and to assess the validity of previous lower resolution model results, the latest high spatial resolution version of the AER seasonal radiative dynamical climate model is used to simulate the climatic effects of ozone changes relative to the other greenhouse gases. The steady state climatic effect of a sustained decrease in lower stratospheric ozone, similar in magnitude to the observed 1979 to 1990 decrease, is estimated by comparing three steady state climate simulations: I) 1979 greenhouse gas concentrations and 1979 ozone; II) 1990 greenhouse gas concentrations with 1979 ozone; and III) 1990 greenhouse gas concentrations with 1990 ozone. The simulated increase in surface air temperature resulting from non-ozone greenhouse gases is 0.272 K. When changes in lower stratospheric ozone are included, the greenhouse warming is 0.165 K which is approximately 39% lower than when ozone is fixed at the 1979 concentrations. Ozone perturbations at high latitudes result in a cooling of the surface-troposphere system that is greater (by a factor of 2.8) than that estimated from the change in radiative forcing resulting from ozone depletion and the model's 2xCO₂ climate sensitivity. Our results suggest that changes in meridional heat transport from low to high latitudes combined with the decrease in the infrared opacity of the lower stratosphere are very important in determining the steady state response to high latitude ozone losses. The 39% compensation in greenhouse warming resulting from lower stratospheric ozone losses is also larger than the 28% compensation simulated previously by the lower resolution model. The higher resolution model is able to resolve the high latitude features of the assumed ozone perturbation which are important in determining the overall climate sensitivity to these perturbations.

This work appeared in the *Journal of Climate* (MacKay et al., 1997a).

(IV.2.2) Manuscript documenting the 2-D Interactive Climate Chemistry Model

The Atmospheric and Environmental Research (AER) two-dimensional (2-D) Interactive Climate Chemistry Model (ICCM) is used to explore the interactions between changes in atmospheric composition, temperature, and dynamics. The ICCM is composed of two modules: 1) a 2-D seasonal radiative dynamical climate model with a mixed layer ocean, thermodynamic sea ice, detailed radiation code for both solar and terrestrial radiation, land and ocean sectors, and parameterized atmospheric and oceanic meridional heat transport, and 2) a chemistry/dynamics model with a comprehensive gas phase photochemistry (O_x, HO_x, BR_y, Cl_y, NO_y, and methyl families), parameterized eddy

transport of heat, momentum, and atmospheric tracers, and interactive calculation of mean transport. The primary difference between the ICCM and the chemistry/dynamics model is that tropospheric temperatures are calculated interactively in the ICCM and are prescribed in the chemistry/dynamics model. The results of a double CO₂ experiment from the 2-D ICCM are analyzed to illustrate its performance and to assess the differences, if any, between it and the AER chemistry/dynamics model. The 2xCO₂ increase in column ozone is slightly greater in the ICCM than in the chemistry/dynamics model (3.5% relative to 3.1%). Also, the ICCM predicts an increase in stratospheric mean mass residual circulation of up to 10%, where the chemistry/dynamics model did not. Both of these differences result from a slightly different temperature structure in the lower stratosphere between the two models. Overall the differences between the two simulations are relatively small. One of the primary reasons for this is that both models presently have a relatively weak exchange of energy, momentum, and chemical composition between the troposphere and stratosphere. Thus, calculating tropospheric temperatures interactively has a small influence in our present model framework. It is suggested that incorporating a three-wave model structure into the ICCM will provide a more realistic simulation of stratospheric wave activity and possibly an improved representation of stratosphere-troposphere interaction.

The manuscript describing this work was submitted to Journal of Geophysical Research (MacKay et al., 1997b).

(V) Assessment Related Activities

(V.1) The Second Model and Measurement Workshop

We are now in the process of gearing up for the Second Models and Measurements (M&M II) exercise. AER will submit model results for each of the defined exercise. Malcolm Ko was involved in defining two of the modeling exercises; one is the transport run A5 which is the simulation of the SF₆ distribution, the other is the photochemistry B3 experiment for which each modeling group will calculate the ozone distribution using prescribed ozone production and loss fields so that differences in transport of ozone can be compared among models. AER is providing the production and loss fields for this exercise to the other modelers. Malcolm Ko and staff at AER will also lead the analysis effort on the source gases.

(V.2) Ozone Assessment Report

Malcolm Ko was asked by Ron Prinn and Rudy Zander to be a co-author in chapter 1 of the 1998 Ozone Assessment Report. He will contribute a discussion of the atmospheric lifetimes of long-lived species as calculated by numerical models and how they compared to lifetimes derived from observations.

Jose Rodriguez was invited to be co-lead-author of chapter 2 with Michael Kurylo. The chapter deals with the behavior of shorter-lived source gases that affect ozone.

(V.3) IPCC Report on Effect of Aviation

AER will be performing calculations for the ozone assessment of the supersonic fleet to be reported in the IPCC Special Report on Aviation and the Global Atmosphere. The modeling exercise includes a set of 12 experiments to assess the effects of HSCTs in the stratosphere. Updated emission scenarios have been prepared for the 1997 assessment. Sulfate aerosol surface area increases due to aircraft will be included in the scenarios. AER has contributed these fields from calculations with our sulfate aerosol model. The results will be reported in chapter 4 of the IPCC report. Malcolm Ko was invited by Charles Jackman and Ivar Isaksen to be a co-author in the chapter.

Michael Danilin was invited by David Fahey to be a co-author in chapter 2. He will work on a section related to how particles evolve in the aircraft plume.

(VI) The POLARIS Campaign

We were selected to join the Theory Team of the Photochemistry of Ozone Loss in the Arctic Region in Summer (POLARIS) campaign. One of our tasks is to identify how the data can be used to improve assessment models. Malcolm Ko joined the team at Fort Wainright, Alaska during the May Deployment.

Anticipated Activities in the next six months

We anticipate that we will spend most of July and August generating model results for the Model and Measurement Workshop II and the IPCC report. This will be followed by our contributions to the analyses of the results. We also hope to put the finishing touch on several manuscripts to get them ready for publications.

The coupling between bromine and chlorine chemistries in the atmosphere (see e.g. Danilin et al., 1996) makes it difficult to use bromine loading and ODP for CH₃Br to predict how ozone would respond to change in emissions of CH₃Br. We plan to do scenario calculations using the AER 2-D CTM to compute the ozone responses.

We will continue to participate in the POLARIS campaign. Malcolm Ko will join the July deployment while Jose Rodriguez is planning to participate in the September deployment.

References

- Cunnold, D., F. Alyea, N. Phillips, and R.N. Prinn, A three-dimensional dynamical chemical model of atmospheric ozone, *J. Atmos. Sci.*, **32**, 170-194, 1975.
- Danilin, M.Y., N.D Sze, M.K.W. Ko, J.M. Rodriguez, and M.J. Prather, Bromine-chlorine coupling in the Antarctic ozone hole, *Geophys. Res. Lett.*, **23**, 153-156, 1996.
- Danilin, M.Y., J.M. Rodriguez, M.K.W. Ko, D.K. Weisenstein, R.C. Brown, R.C. Miake-Lye, and M.R. Anderson, Aerosol particle evolution in an aircraft wake: Implications for the high-speed civil transport fleet impact on ozone, *J. Geophys. Res.*, in press, 1997.
- DeMore, W.B., et al., Chemical Kinetics and Photochemical Data for use in Stratospheric Modeling, Evaluation Number 12, *JPL Publication 97-4*, 1997.
- DeMore, W.B., et al., Chemical Kinetics and Photochemical Data for use in Stratospheric Modeling, Evaluation Number 11, *JPL Publication 94-26*, 1994.
- Fahey, D. W., et al., Emission measurements of the Concorde supersonic aircraft in the lower stratosphere, *Science*, **270**, 70-74, 1995.
- Greco, J.C., M. Loewenstein, J.R. Podolske, Correlations of nitrous oxide with ozone and reactive nitrogen species in the lower stratosphere, AGU Fall Meeting, 1996.
- Hanson, D.R., A.R. Ravishankara, and E.R. Lovejoy, Reactions of BrONO₂ with H₂O on submicron sulfuric acid aerosol and the implications for the lower stratosphere, *J. Geophys. Res.*, **101**, 9063-9069, 1996.
- Hanson, D.R. and A.R. Ravishankara, Heterogeneous chemistry of bromine species in sulfuric acid under stratospheric conditions, *Geophys. Res. Lett.*, **22**, 385-388, 1995.
- Held, I.M., and A.Y. Hou, Nonlinear axially symmetric circulations in a nearly inviscid atmosphere, *J. Atmos. Sci.*, **37**, 515-533, 1980.
- Houghton, J.T., *The Physics of Atmospheres*, 2d ed., Cambridge University Press, 1986.
- Kalnay, E. et al., The NCEP/NCAR 40-year reanalysis project, *Bull. Am. Meteorol. Soc.*, **77**, 437-471, 1996.

Ko, M.K.W., N.D. Sze, M. Livshits, M.B. McElroy, and J.A. Pyle, The seasonal and latitudinal behavior of trace gases and O₃ as simulated by a two-dimensional model of the atmosphere, *J. Atmos. Sci.*, **41**, 2381-2408, 1984.

Ko, M.K.W., K.K. Tung, D.K. Weisenstein, and N.D. Sze, A zonal mean model of stratospheric tracer transport in isentropic coordinates: Numerical simulations for nitrous oxide and nitric acid, *J. Geophys. Res.*, **90**, 2313-2329, 1985.

Ko, M.K.W., N.D. Sze, and D.K. Weisenstein, The roles of dynamical and chemical processes in determining the stratospheric concentration of ozone in one-dimensional and two-dimensional models, *J. Geophys. Res.*, **94**, 9889-9896, 1989.

Ko, M.K.W., H.R. Schneider, R.-L. Shia, D.K. Weisenstein, and N.-D. Sze, A two-dimensional model with coupled dynamics, radiation, and photochemistry, 1, Simulation of the middle atmosphere, *J. Geophys. Res.*, **98**, 20,429-20,440, 1993.

Ko, M.K.W., N.D. Sze, C.J. Scott, and D.K. Weisenstein, On the relation between stratospheric chlorine/bromine loading and short-lived tropospheric source gases, *J. Geophys. Res.*, submitted, 1997.

Lacis, A., and J. Hansen, A parameterization for the absorption of solar radiation in the earth's atmosphere, *J. Atmos. Sci.*, **31**, 118-133, 1974.

MacKay, R.M., M.K.W. Ko, R.-L. Shia, Y. Yang, S Zhou, and G. Molnar, An estimation of the climatic effects of stratospheric ozone losses during the 1980s, *Journal of Climate*, **10**, 774-788, 1997a.

MacKay, R.M., M.K.W. Ko, R.-L. Shia, S Zhou, and Y. Yang, The response of a coupled climate-chemistry model to greenhouse forcing, *J. Geophys. Res.*, submitted, 1997b.

Molnar, G., M.K.W. Ko, S. Zhou, and N.D. Sze, Climate consequences of the observed ozone loss in the 1980's: Relevance to the greenhouse problem, *J. Geophys. Res.*, **99**, 25,755-25,760, 1994.

Newman, P.A., M.R. Schoeberl, R.A. Plumb, and J.E. Rosenfield, Mixing rates calculated from potential vorticity, *J. Geophys. Res.*, **93**, 5221-5240, 1988.

Nickolaisen and Sander, manuscript in preparation, 1997.

Peng, L., M.-D. Chou, and A. Arking, Climate studies with a multi-layer energy balance model. Part I: Model description and sensitivity to the solar constant. *J. Atmos. Sci.*, **39**, 2639-2656, 1982.

Peng, L., M.-D. Chou, and A. Arking, Climate warming due to increasing atmospheric CO₂: Simulations with a multilayer coupled atmosphere-ocean seasonal energy balance model, *J. Geophys. Res.*, **92**, 5505-5520, 1987.

Rennick, M.A., The parameterization of tropospheric lapse rates in terms of surface temperature, *J. Atmos. Sci.*, **34**, 854-862, 1977.

Schneider, H.R., M.K.W. Ko, N.D. Sze, G.-Y. Shi, and W.-C. Wang, An evaluation of the role of eddy diffusion in stratospheric interactive two dimensional models, *J. Atmos., Sci.*, **46**, 2079-2093, 1989.

Schneider, H.R., M.K.W. Ko, R.L. Shia, and N.D. Sze, A two-dimensional model with coupled dynamics, radiative transfer, and photochemistry, 2, Assessment of the response of stratospheric ozone to increased levels of CO₂, N₂O, CH₄, and CFC, *J. Geophys. Res.*, **98**, 20,41-20,449, 1993.

Schoeberl, M. R., et al., An estimation of the dynamical isolation of the tropical lower stratosphere using UARS wind and trace gas observations of the quasi-biennial oscillation, *Geophys. Res. Lett.*, **24**, pp. 53-56, 1997.

Shia, R.-L., S. Zhou, M.K.W. Ko, N.-D. Sze, D. Salstein, and K. Cady-Pereira, A three-wave model of the stratosphere with coupled dynamics, radiation, and photochemistry, *J. Geophys. Res.*, submitted, 1997a.

Shia, R.-L., M. K. W. Ko, D. K. Weisenstein, C. Scott and J. Rodriguez, Transport between the tropical and mid-latitude lower stratosphere: Implications for ozone response to HSCT emissions, *J. Geophys. Res.*, submitted, 1997b.

Spencer, J.E., Rowland, F.S., Bromine Nitrate and Its Stratospheric Significance, *J. Phys. Chem.*, **82**, 7-10, 1978.

Strobel, D. F., 1981: Parameterization of linear wave chemical transport in planetary atmospheres by eddy diffusion. *J. Geophys. Res.*, **86**, 9806-9810.

Stolarski, R.S., and H.L. Wesoky (Eds.), The atmospheric effects of stratospheric aircraft: A third program report, *NASA Ref. Publ.*, 1313, 413 pp., 1993.

Stolarski, R.S., et al., 1995 Scientific Assessment of the Atmospheric Effects of Stratospheric Aircraft, *NASA Reference Publication 1381*, NASA, Washington, D.C., 1995.

Shia, Run-Lie, Shuntai Zhou, Malcolm K. W. Ko, Nien-Dak Sze, David Salstein and Karen Cady-Pereira, 1997: A Three-Wave Model of the Stratosphere With Coupled Dynamics, Radiation and Photochemistry, Submitted to *J. Geophys. Res.*

Stone, P.H., The meridional variation of the eddy heat fluxes by baroclinic waves and their parameterization, *J. Atmos. Sci.*, **31**, 444-456, 1974.

Stone, P.H., and J.H. Carlson, Atmospheric lapse rate regimes and their parameterization, *J. Atmos. Sci.*, **36**, 415-423, 1979.

Volk, C.M., et al., Quantifying transport between the tropical and mid-latitude lower stratosphere, *Science*, **272**, 1763-1768, 1996.

Wang, W.-C., and P.B. Ryan, Overlapping effect of atmospheric H₂O, CO₂, and O₃ on the CO₂ radiative effect, *Tellus*, **35B**, 81-91, 1985.

Wang, W.-C., G. Molnar, M.K.W. Ko, S. Goldenberg, and N.D. Sze, Atmospheric trace gases and global climate: A seasonal model study, *Tellus*, **42B**, 149-161, 1990.

Weisenstein, D. K., M. K. W. Ko, N.-D. Sze, and J. M. Rodriguez, Potential impact of SO₂ emissions from stratospheric aircraft on ozone, *Geophys. Res. Lett.*, **23**, pp. 161-164, 1996

Weisenstein, D.K., G.K. Yue, M.K.W. Ko, N.D. Sze, J.M. Rodriguez, and C.J. Scott, A two-dimensional model of sulfur species and aerosols, *J. Geophys. Res.*, **102**, 13,019-13,035, 1997a.

Weisenstein, D.K., M.K.W. Ko, I.G. Dyominov, G. Pitari, L. Ricciardulli, G. Visconti, and S. Bekki, The effects of sulfur emissions from HSCT aircraft: A 2-D model intercomparison, *J. Geophys. Res.*, submitted, 1997b.

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE July 25, 1997	3. REPORT TYPE AND DATES COVERED Bi-annual Jan. 1997 - June 1997	
4. TITLE AND SUBTITLE Coupling Processes between Atmospheric Chemistry and Climate		5. FUNDING NUMBERS NAS5-97039	
6. AUTHOR(S) M.K.W. Ko, Debra Weisenstein, Run-Li Shia, and N.D. Sze		8. PERFORMING ORGANIZATION REPORT NUMBER P-698	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Atmospheric and Environmental Research, Inc. 840 Memorial Drive Cambridge, MA 02139		10. SPONSORING / MONITORING AGENCY REPORT NUMBER	
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) NASA Goddard Space Flight Center Earth Sciences Procurement Office Greenbelt, MD 20771		11. SUPPLEMENTARY NOTES	
12a. DISTRIBUTION / AVAILABILITY STATEMENT		12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) <p>This is the first semi-annual report for NAS5-97039 summarizing work performed for January 1997 through June 1997. Work in this project is related to NAS1-20666, also funded by NASA ACMAP. The work funded in this project also benefits from work at AER associated with the AER three-dimensional isentropic transport model funded by NASA AEAP and the AER two-dimensional climate-chemistry model (co-funded by Department of Energy).</p> <p>The overall objective of this project is to improve the understanding of coupling processes between atmospheric chemistry and climate. Model predictions of the future distributions of trace gases in the atmosphere constitute an important component of the input necessary for quantitative assessments of global change. We will concentrate on the changes in ozone and stratospheric sulfate aerosol, with emphasis on how ozone in the lower stratosphere would respond to natural or anthropogenic changes. The key modeling tools for this work are the AER two-dimensional chemistry-transport model, the AER two-dimensional stratospheric sulfate model, and the AER three-wave interactive model with full chemistry.</p>			
14. SUBJECT TERMS		15. NUMBER OF PAGES	
17. SECURITY CLASSIFICATION OF REPORT Unclassified		16. PRICE CODE	
18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT	

