Model Calculations of the Atmospheric Effects of Supersonic Aviation Which Were Used in the IPCC Special Report on Aviation and the Global Atmosphere

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Rationale

This report is intended to expand upon the IPCC Special Report on Aviation and the Global Atmosphere. It provides additional detail regarding the supersonic emission scenario calculations presented in Chapter 4—Modeling the Future Atmosphere. Due to the space limitations in that document, detailed plots and discussion for a majority of the model calculations were omitted. This document is intended to fill that gap and has been compiled for two reasons: (1) to aid those called upon to review the IPCC Special Report, and (2) to archive and document the calculations performed by nine modeling groups for up to 50 scenarios each. This document does not attempt to provide a synthesis of the modeling results or draw conclusions regarding the impact of aircraft on the atmosphere. That function is provided by the IPCC report.

The model descriptions and model results presented in this document represent the state of those models in 1997 when the calculations were performed. Because models are continually evolving and undergoing further development, these results should be considered as a historical snapshot. Please do not cite results presented herein without first obtaining permission from the group which performed the calculations.

Contents

Chapter 1

Model Descriptions

Descriptions of the nine numerical models of the stratosphere used to predict perturbations to ozone due to a future fleet of supersonic aircraft are presented in this chapter. These are six two-dimensional (2-D) models and three three-dimensional (3-D) models as listed in Table 1.1. The 2-D models are the Atmospheric and Environmental Research (AER) model, the Commonwealth Scientific and Industrial Organization (CSIRO) model, and NASA Goddard Space Flight Center (GSFC) model, the Lawrence Livermore National Laboratory (LLNL) model, the University of Edinburgh (THINAIR) model, and the University of L'Aquila (UNIVAQ) model. The 3-D models are the NASA Langley Research Center (LARC) model, the University of Oslo (SCTM1) model, and the University of Cambridge (SLIMCAT) model. The scientists who comprise the research team for each model, as well as the person to contact regarding results in this report, are listed in Table 1.1.

These model descriptions refer to the models as they were in 1997 when the IPCC scenario calculations were performed. As models are continually evolving and undergoing improvements and enhancements, the following descriptions do not necessarily represent the present state of the models described, but merely a historical snapshot. For current information regarding any of these models, please contact the research team directly.

We will discuss the transport and chemical treatments of the models below with emphasis on the differences that may explain the different model predictions of aircraft impact on ozone. Model transport affects the residence time and concentration of emitted materials in the lower stratosphere. Treatment of chemistry in the model determines how the emitted pollutants will modify radical concentrations and ozone.

1.1 Transport Formulation

1.1.1 Domain and Resolution

All 2-D models have the lower boundary at the ground and most use log-pressure coordinates in the vertical with equal resolution in the troposphere and the stratosphere. The THINAIR model uses potential temperature (θ) as the vertical coordinate. Key parameters for model grid and resolution are summarized in Table 1.2. Horizontal resolution is 10^o for the AER, GSFC, THINAIR, and UNIVAQ models and 5^o for the CSIRO and LLNL models. Vertical resolution varies from 1.2 km for the AER model, 1.5 km for the LLNL model, 2 km for the CSIRO and GSFC models, 2.8 km for the UNIVAQ model, and 3.5 km for the THINAIR model. The model top boundary varies from 60 km (AER and THINAIR) to 90 km (GSFC). THINAIR has a model top boundary of 100 km for dynamics, but chemistry is explicitly calculated only up to 60 km.

Among the 3-D models, the SLIMCAT and SCTM1 models are grid point models. The LARC 3-D chemistry transport model (CTM) uses a spectral representation in the horizontal dimension with triangular truncation to zonal wavenumber 16 (T16). Constituent chemistry calculations are done in grid space using a 5.5^ox5.5^o grid that ensures alias-free evaluation of all inverse transforms (Eckman et al., 1995). Winds and temperatures are truncated from a higher resolution (T32) version of the LARC spectral GCM for input to the CTM. Sigma coordinates are used in the vertical dimension with ten layers in the troposphere and 14

Table 1.1: List of Models that Contributed Results to this Report

Model name	Institution	Model Team	contact person for model	contact person for results in this report
2-D Models				
AER	Atmospheric and Environmental Research, Inc., U.S.A.	Malcolm Ko, Debra Weisenstein, Courtney Scott, Jose Rodriguez, Run-Lie Shia, N.D. Sze	Malcolm Ko	Debra Weisenstein
CSIRO	Commonwealth Scientific and Industrial Research Organization (CSIRO), Telecommunications and Industrial Physics, Australia	Keith Ryan, Ian Plumb, Peter Vohralik, Lakshman Randeniya	Keith Ryan	Peter Vohralik
GSFC	NASA Goddard Space Flight Center, U.S.A.	Charles Jackman, David Considine, Eric Fleming	Charles Jackman	Charles Jackman
LLNL	Lawrence Livermore National Laboratory, U.S.A.	Peter Connell, Keith Grant, Douglas Kinnison, Douglas Rotman	Douglas Kinnison	Douglas Kinnison
THINAIR	University of Edinburgh, United Kingdom	Robert Harwood, Vicky West	Robert Harwood	Vicky West
UNIVAQ	University of L'Aquila, Italy	Giovanni Pitari, Barbara Grassi, Lucrezia Ricciardulli, Guido Visconti	Giovanni Pitari	Giovanni Pitari
3-D Models				
LARC	NASA Langley Research Center, U.S.A.	William Grose, Richard Eckman	William Grose	Richard Eckman
SCTM1	University of Oslo, Norway	Ivar Isaksen, Michael Gauss	Ivar Isaksen	Michael Gauss
SLIMCAT	University of Cambridge, United Kingdom	Martyn Chipperfield, Helen Rogers	Martyn Chipperfield	Helen Rogers

layers in the stratosphere. The vertical resolution is approximately 3 km in the stratosphere and decreases in the troposphere to less than 1 km near the surface.

The SCTM1 model has a horizontal resolution of 7.8^o latitude x 10^o longitude, with 29 vertical layers from the surface to 90 km. Sigma coordinates are used in the layers below 100 hPa and constant pressure coordinates above. The vertical resolution is approximately 1.5 km between 350 hPa and 21 hPa. Above 21 hPa, the vertical resolution is 5 km. The SLIMCAT model uses a horizontal resolution of 7.5^o latitude x 7.5^o longitude. The lower boundary of this model is at θ=335 K (approximately 8 km) and the upper boundary at θ=2700 K (about 60 km). There are 12 vertical layers in θ (potential temperature) coordinates, with vertical resolution in the lower stratosphere about 3 km.

Most models use temperature lapse rate to define the location of the tropopause, which changes with season and latitude. The 2-D models use temperature from climatology, except for the THINAIR model which calculates temperature interactively. The 3-D models employ a single year of temperature data. The LARC 3-D model uses temperature calculated by a T32 version of the Langley spectral GCM. The SCTM1 model uses NMC temperature data from 1990, and the SLIMCAT model uses 1992 data from the U.K. Met Office. The SCTM1 model uses a different approach to identify the model tropopause. In this model, tropospheric air is defined by an ozone mixing ratio less than 0.25 ppmv. The tropopause is constrained, however, and not allowed to rise above the 46 hPa level.

In 2-D models, the troposphere is distinguished from the stratosphere by assigning large values of K_{yy} (typically 1-1.5x10^6 m^2/sec) and K_{zz} (4-10 m^2/sec). Studies indicate that strat-trop exchange may be dominated by transport from the middle-world to the troposphere (Eluszkiewicz, 1996). In a 2-D model, this will manifest itself as eddy flux along isentropic surfaces across the boundary (Shia et al., 1993). If this is true, the model calculated residence time of aircraft emissions in the lower stratosphere would be sensitive to the horizontal and vertical resolutions of the models that determine the location and seasonal variation of the tropopause.

1.1.2 Temperature and Transport Parameters

Most results in this report are generated by the models in the chemistry-transport (CTM) mode, i.e. results are obtained using pre-calculated temperature and transport fields. The temperature is used in the models to compute temperature-dependent reaction rate constants, and, in some models, to predict the surface area of polar stratospheric clouds (PSCs). In addition, the temperature is also used in computing the winds and eddy diffusion coefficients in some of the 2-D models. The THINAIR 2-D model is the exception and calculates temperature and winds interactively along with chemical perturbations. Model characteristics related to temperature, winds, and parameterization of eddy diffusion are summarized in Tables 1.3 and 1.4. A more detailed description is given below.

The UNIVAQ 2-D model uses the zonal mean temperature and statistics for zonal asymmetry in temperature from the twelve year NMC average (1979-1990) in the chemistry calculations, but the transport parameters are computed (Pitari, 1993a) from the output of a spectral quasi-geostrophic 3-D model (Pitari et al., 1992). The residual circulation is calculated directly from velocity and temperature data of the GCM. It calculates K_{yy} as the quasi-geostrophic potential vorticity flux to gradient ratio (see Newman et al., 1986), with the dynamical data taken from the GCM. Typical values for K_{yy} in the tropical lower stratosphere are 0.1-0.5x10^5 m^2/sec. A minimum value of 2x10^5 m^2/sec is adopted outside the tropics. K_{yz} values are obtained by projecting the K_{yy} from isentropic to pressure surfaces.

The AER model uses the zonal mean temperature and statistics for zonal asymmetry in temperature from the eight year NMC average (1979-1986). It computes the vertical velocity as the ratio of the heating rate (based loosely on published results of Dopplick [1979]) to the temperature lapse rate (see e.g. Tung [1982] and references therein). The vertical velocity field is integrated to obtain a stream function with adjustments made to the heating rate to assure mass conservation (see Ko et al. [1985] for a description of the procedure). Values for K_{yy} are assigned to simulate the tropical barrier (Shia et al., 1998) guided by observed exchange time constants between midlatitudes and the tropics (Minschwaner et al., 1996; Volk et al., 1996; Schoeberl et al., 1997). Values chosen are 0.7-1.3x10^5 m^2/s for the tropics and 3-10x10^5 m^2/s elsewhere. Values of 0.1 m^2/sec are adopted for K_{zz} from the tropopause to 40 km, increasing to 1 m^2/sec above 50 km. Values of K_{yz} are obtained by projecting the K_{yy} from isentropic surfaces to pressure surfaces.

The CSIRO model uses the zonal mean temperature from the eight year NMC average (1979-1986). The

Table 1.2: Model Grid and Resolution

Model	Horizontal Resolution (lat x lon)	Vertical Resolution, Coordinate	Model Top Boundary	Tropopause
2-D Models				
AER	10^o	1.2 km log-pressure	60 km 0.2 hPa	temperature gradient from climatology
CSIRO	5^o	2 km log-pressure	80 km 0.01 hPa	temperature gradient from climatology
GSFC	10^o	2 km log-pressure	90 km 0.0024 hPa	temperature gradient from climatology
LLNL	5^o	1.5 km log-pressure	84 km 0.01 hPa	temperature gradient from climatology
THINAIR	10^o	3.5 km θ coordinates	60 km 0.2 hPa	temperature gradient from climatology
UNIVAQ	10^o	2.8 km log-pressure	70 km 0.04 hPa	temperature gradient from climatology
3-D Models				
LARC	5.5^o x 5.5^o (T16 spectral)	100 hPa interval below 100 hPa; 3 km interval above 100 hPa, sigma coordinates	65 km 0.1 hPa	temperature gradient from T32 GCM
SCTM1	7.8^o x 10^o	1.5 km between 350 hPa and 21 hPa, 5 km above 21 hPa, sigma/pressure	90 km 0.002 hPa	defined as O_3=0.25 ppmv but always below 46 hPa
SLIMCAT	7.5^o x 7.5^o	12 levels, $\sim$3 km θ coordinates	$\sim$60 km θ=2700 K	model lower boundary at θ=335 K ($\sim$8 km)

Table 1.3: Temperature, Winds and K_{yy} Used in the Models

Model	zonal mean temperature	zonal asymmetry in temperature	winds	K_{yy}
2-D Models				
AER	8 year average (1979-1986) monthly mean T from NMC	probability distribution based on statistics from NCEP (1979-1986), used in chemistry calculation only	$w=Q/\Gamma$ Heating rate based loosely on Dopplick, adjusted to conserve mass	assigned values to match exchange time constant across the tropical barrier
CSIRO	8 year average (1979-1986) monthly mean T from NMC	zonal asymmetry not used.	iterative procedure to solve TEM energy equation (include dT/dt term); heating rates taken from M&M augmented by latent heat in troposphere, computed for mesosphere	computed from $\frac{EPFlux}{gradient\ of\ PV}$
GSFC	17 year average (1979-1995) monthly mean T from NCEP	probability distribution based on statistics from NCEP (1979-1995)	solve stream function using Garcia and Solomon (1983); heating rate from climatological T, O_3, and H_2O plus latent heat; forcing from 6 planetary waves and gravity wave breaking	computed from $\frac{EPFlux}{gradient\ of\ PV}$
LLNL	Fleming (private communication)	Fleming (private comm.), used in chemistry calculations and in determination of winds and K_{yy}	solve stream function using Garcia and Solomon (1983); heating rate from climatological T and calculated trace gases; forcing from 2 planetary waves, gravity wave breaking, and Rayleigh friction	computed from $\frac{EPFlux}{gradient\ of\ PV}$
THINAIR	calculated interactively	zonal asymmetry base on wave 1 in temperature	calculated interactively using 3 planetary waves and spectrum of gravity waves	computed from $\frac{EPFlux}{gradient\ of\ PV}$
UNIVAQ	12 year average (1979-1990) monthly mean T from NMC	zonal asymmetry based on statistics, used in chemistry calculations only	calculated from velocity and temperature data of 3-D GCM	computed from $\frac{EPFlux}{gradient\ of\ PV}$
3-D Models				
LARC	from T32 GCM	explicit	from T32 GCM	none
SCTM1	1990 NMC	explicit	horizontal winds from GISS GCM, vertical winds from continuity	in troposphere only, associated with convection
SLIMCAT	UKMO analysis (1992)	explicit	UKMO analysis (1992)	none

Table 1.4: Values of Eddy Diffusion Coefficients Used in the 2-D Models

Model	K_{yy}, trop (10^5 m^2/sec)	K_{zz}, trop (m^2/sec)	K_{yy}, strat (10^5 m^2/sec)	K_{zz}, strat (m^2/sec)	K_{yz}, strat (m^2/sec)
2-D Models					
AER	15	10	0.7-1.3 in tropics, 3-10 elsewhere	0.1-1	projected, typical values $\pm$100
CSIRO	10	4	calculated diagnostically, set to 0.1 when wind is easterly, always between 0.1 and 30	0.25 from 21 km-42 km; 0.1 elsewhere	projected, magnitude can be as large as 1000
GSFC	calculated (6 waves), minimum value of 15	tropopause: 1; at ground: 50 in tropics, 25 at poles	calculated (6 waves), minimum value 0.1	0.03 at tropopause to 0.3 at stratopause, computed in mesosphere	0
LLNL	10	4	calculated (2 waves), minimum value 0.1	calculated, with minimum value of 0.2	0
THINAIR	1	5	calculated (3 waves)	0	0
UNIVAQ	5-15	10-30	calculated from 3-D output, typically 0.1-0.5 in tropics, $\geq$2 elsewhere	calculated from 3-D output, 0.1-1.0 in stratosphere	projected, typical values $\pm$100

heating rates (a combination of archived stratospheric values in the M&M report [Prather and Remsberg, 1993], calculated values in the mesosphere based on climatological ozone and temperature, and latent heat in the troposphere from Dopplick) are used along with the adopted climatological zonal-mean temperature field to compute the velocities from the zonal-mean energy equation (see Solomon et al. [1986]). K_{yy} values are calculated as the ratio of the E-P flux divergence to the meridional gradient of potential vorticity where the E-P flux is determined diagnostically from the zonal-mean momentum equation using the calculated velocities and the zonal wind determined from the zonal-mean temperature (see Randeniya et. al. [1997] and Vohralik et al. [1998] for details). Values for K_{zz} in the stratosphere are taken to be 0.1 to 0.25 m^2/sec. Values of K_{yz} are obtained by projecting the K_{yy} from isentropic surfaces to pressure surfaces.

The GSFC and LLNL models use the seventeen year average (1979-1995) temperature from NCEP. Both models use the climatological 3-D temperature field to compute the zonal-mean winds and K_{yy} in a self-consistent way. The GSFC model (see Jackman et al., 1996) uses observed ozone, H_2O, and temperature to compute the diabatic heating which is augmented by the latent heat to provide the net heating in the energy equation. Mechanical forcing from planetary waves (calculated using the 3-D temperature database using six waves) and effects of gravity wave breaking provide the wave driving for the momentum equation. The stream function is obtained by solving an elliptic equation obtained by combining the zonal-mean momentum and energy equations (see e.g., Garcia and Solomon, 1983). The LLNL model solves a similar set of equations but employs different methods to compute the heating and the forcing terms. The heating rate for the LLNL model is computed from climatological temperatures and model calculated trace gases. Mechanical forcing employs 2 planetary waves, gravity wave breaking, and Rayleigh friction. Both models use the ratio of the E-P flux divergence to the gradient of potential vorticity computed using the 3-D temperature database to calculate K_{yy} (see e.g. Garcia, 1991; Randel and Garcia, 1994), and use a gravity wave breaking parameterization to calculate K_{zz}. The K_{yz} is zero in these models.

The THINAIR model is the only model which calculated temperature and winds interactively, ie. the temperature and circulation differ in each scenario based on variations in model-calculated ozone, which is itself controlled by temperature and circulation. Three planetary waves and a spectrum of gravity waves are employed in the calculation of model winds and stratospheric K_{yy} (see Kinnersley and Harwood, 1993). K_{zz} is not used in the stratosphere. The THINAIR model utilizes the "wave" approach to accounting for zonal asymmetry in temperature [see De Rudder et al., 1996; Portmann et al., 1996; Smyshlaev et al., 1998]. A wavenumber 1 modulation to the zonal mean temperature is employed to approximate the temperature fluctuations a circulating atmospheric parcel might experience as it moves around the globe. The amplitude of the modulation varies in space and time and is proportional to the calculated planetary wave amplitudes. The period of the modulation is also a spatially and temporally varying quantity, similar to the circumnavigation time of an air parcel in the atmosphere. Thus, the model temperature at some grid point can vary dramatically on short time scales and be very far from the zonal mean temperature of the grid point.

It should be noted that K_{yy} values calculated using observed temperature are sometimes negative (see e.g. Newman et al., 1986). In those cases, the models reassign a minimum value, typically about 0.1×10^5 m^2/sec. CSIRO also assigns K_{yy} values of 0.1×10^5 m^2/sec whenever the zonal wind (derived from temperature data) is easterly and resets values larger than 30×10^5 m^2/sec to that value.

Temperatures and wind fields for the LARC 3-D model are taken from the LARC T32 spectral GCM output. The SCTM1 and SLIMCAT 3-D models both employ observed temperature fields, SCTM1 using the 1990 NMC temperature field and SLIMCAT the UKMO analysis from 1992. SLIMCAT also uses horizontal winds from the 1992 UKMO analysis (Swinbank and O'Neill, 1994). SCTM1 uses horizontal winds from the GISS GCM and vertical winds from continuity. SLIMCAT generates vertical winds diagnostically using the MIDRAD radiation scheme (Shine, 1987). These 3-D models do not require eddy diffusion coefficients, though the SCTM1 models does employ K_{yy} diffusion in the troposphere in association with convective events.

While no specific study has been performed to confirm this, it is reasonable to assume that each model has checked on their own that their velocities and numerical schemes are mass conserving. Judging from the minimum value chosen for K_{yy} in the models, one would assume that numerical diffusion should not be an issue.

Given the different methods used in deriving the transport parameters in the models, it is not surprisingly that there are large differences in the calculated distributions of the trace gases. Large differences in model simulated distributions of chemically inert tracers such as sulfur hexafluoride point to transport differences as a major contributor. The community is trying to identify a climatological database of zonal-mean distributions of trace gases that can be used to diagnose the transport parameters.

1.2 Chemistry Treatment

1.2.1 Gas-phase Chemistry

The kinetic reaction rate constants are from JPL-97 (DeMore, 1997) except SCTM1 which uses JPL-94. CSIRO, LARC and SLIMCAT assumes a 5% yield of HCl from OH+ClO. The 3-D models use the temperature at the 3-D grid points to compute the reaction rate constants. Among the 2-D models, gas phase rate constants are computed using the zonal mean temperature in the CSIRO, GSFC, and UNIVAQ models. The AER and LLNL models use rate constants weighted by the probability distribution of the zonally asymmetric temperature structure.

1.2.2 Heterogeneous Chemistry

The following seven heterogeneous reactions were identified in JPL-97 as possible reactions on sulfate aerosol and/or polar stratospheric cloud (PSC) particles.

$$N_2O_5 + H_2O(solid) \rightarrow 2HNO_3 \quad \text{(A)}$$
$$ClONO_2 + H_2O(solid) \rightarrow HNO_3 + HOCl \quad \text{(B)}$$
$$BrONO_2 + H_2O(solid) \rightarrow HNO_3 + HOBr \quad \text{(C)}$$
$$ClONO_2 + HCl(solid) \rightarrow HNO_3 + Cl_2 \quad \text{(D)}$$
$$HOCl + HCl(solid) \rightarrow Cl_2 + H_2O \quad \text{(E)}$$
$$HOBr + HCl(solid) \rightarrow BrCl + H_2O \quad \text{(F)}$$
$$N_2O_5 + HCl(solid) \rightarrow HNO_3 + ClNO_2 \quad \text{(G)}$$

Most models assume that the rate constant is in the form γvA/4, where γ is the sticking coefficient, v is the thermal velocity of the reactant, and A is the surface area of the sulfate or PSC.

1.2.2.1 Reactions on Sulfate Aerosol

For reactions on sulfate, AER, GSFC, LLNL, and SCTM1 include the first six reactions. UNIVAQ includes the first 5, CSIRO the first 3, while THINAIR and LARC includes only the first two. All models use γ=0.1 for reaction A. There are slight variations in the temperature dependence of γ for the other reactions on sulfate particles. Previous analyses (Murphy and Ravishankara, 1994; Borrmann et al., 1997; Michelsen et al., 1997) have shown that the γ for some of the reactions are very sensitive to temperature. Ignoring the zonal asymmetry in temperature by using the zonal mean temperature in calculations may underestimate the ozone impact from HSCT (Weisenstein et al., 1998). Since the sulfate surface area is specified in the calculations, the temperature dependence comes from γ and v. The AER and LLNL models use the 3-D temperature distribution to calculate a weighted γv. UNIVAQ uses the temperature distribution to calculate a weighted γ. CSIRO and GSFC use zonal mean temperature. The 3-D models explicitly employ 3-D temperature fields and so handle the temperature-dependent reaction rates more appropriately than 2-D models. A summary of the treatment of heterogeneous chemistry in the models is given in Table 1.5.

The model calculated ozone responses to HSCT will depend on the sulfate surface area density (SAD) in the atmosphere. In this study, the SAD is prescribed rather than calculated by the individual models. In the background atmosphere, the SAD from WMO (1992) corresponding to the clean background with no volcanic influence is used. Sulfur in the fuel will result in SO_2 emission from the engine, with the potential to perturb the SAD. The change in SAD depends on how much of the emitted SO_2 is converted in the plume to sulfate particles (Weisenstein et al., 1996, Danilin et al., 1997). Most results in this study use SAD changes calculated from the AER 2-D sulfate model (Weisenstein et al., 1997) with different assumptions on

Table 1.5: Treatment of Heterogeneous Chemistry in the Models

Model	Sulfate Reactions	Sulfate Reaction Rates	PSC Reactions	PSC Reaction Rates	PSC Parameterization
2-D Models					
AER	A-F	$\overline{\gamma(T)v(T)}A/4$	A,B,D,E,G on NAT A-G on ice	$\overline{\gamma(T)v(T)A(T)}/4$	thermodynamic equilibrium, no supersaturation
CSIRO	A-C	$\gamma(\overline{T})v(\overline{T})A/4$	none	NA	NA
GSFC	A-F	$\gamma(\overline{T})v(\overline{T})A/4$	A,B,D,E on NAT A-G on ice	$\gamma(\overline{T})v(\overline{T})\overline{A(T)}/4$	thermodynamic equilibrium, supersaturation ratios: 10 for NAT, 1.4 for ice
LLNL	A-F	$\overline{\gamma(T)v(T)}A/4$	A-F on STS	$\overline{\gamma(T)v(T)}A/4$	PSC surface area fixed from climatology
THINAIR	A-B	$\gamma(\overline{T})v(\overline{T})A/4$	A,B,D,G	$\gamma(\overline{T})v(\overline{T})\overline{A(T)}/4$	PSC reactions for T<195 K assume gas lifetimes of 9 hours
UNIVAQ	A-E	$\overline{\gamma(T)}v(\overline{T})A/4$	A,B,D,E on NAT A-E on ice	$\overline{\gamma(T)}v(\overline{T})\overline{A(T)}/4$	explicit microphysics with nucleation and condensation
3-D Models					
LARC	A-B	$\gamma(T)v(T)A/4$	A,B,D,E,G on NAT and ice	$\gamma(T)v(T)A(T)/4$	thermodynamic equilibrium
SCTM1	A-F	$\gamma(T)v(T)A/4$	A,B,D,E,G on NAT none on ice	$\gamma(T)v(T)A(T)/4$	thermodynamic equilibrium
SLIMCAT	A-F	$\gamma(T)v(T)A/4$	A,B,D,E,G on NAT A-G on ice	$\gamma(T)v(T)A(T)/4$	thermodynamic equilibrium

conversion in the plume. The changes in SAD from the AER model were found to be in general agreement with the results of other sulfate aerosol models (Weisenstein et al., 1998).

Both the AER and UNIVAQ models have the capability to calculate sulfate surface area interactively with a microphysics code. This capability was utilized in several scenarios, so we will describe here the microphysical processes and methodologies employed. Both models calculate aerosol size distributions by employing discrete size binning. The AER model uses 40 size bins, from 0.0004 μm to 3.2 μm by volume doubling. The UNIVAQ model uses 11 size bins, from 0.01 μm to 10.25 μm by radius doubling. Both models include the processes of homogeneous nucleation, coagulation, condensational growth, sedimentation, and evaporation. The UNIVAQ model also includes heterogeneous nucleation. A more detailed description of the AER microphysical model can be found in Weisenstein et al. (1997) and of the UNIVAQ microphysical model in Pitari et al. (1993b). A comparison of the two models is given by Weisenstein et al. (1998). The main differences between the calculated sulfate aerosol in these models result from different wind fields and the different aerosol size resolutions.

1.2.2.2 Reactions on PSCs and Cold Sulfate Aerosol

The reaction rate constants for reactions on PSCs are also in the form of γvA/4, where A is the surface area for type I PSC (assumed to be NAT) or type II PSC (assumed to be ice) or ternary sulfate particles. A summary of the PSC parameterization scheme and PSC reactions employed by each model is given in

Table 1.5. The CSIRO model does not include any reactions on PSCs or ternary sulfate. UNIVAQ includes the first 5 reactions on ice and reactions A, B, D and E on NAT. The microphysics model described in Pitari et al. (1993b) is used to compute the sulfate surface area and PSC surface area in the UNIVAQ model. The model keeps track of homogeneous nucleation, heterogeneous nucleation (H_2O and H_2SO_4 on Aiken nuclei; HNO_3 and H_2O on sulfate particles; H_2O on NAT particles); condensation (H_2O and H_2SO_4 on sulfate; HNO_3 and H_2O on NAT; H_2O on ice); coagulation (only for sulfate particles), evaporation and gravitational settling to calculate the particle size distributions for the sulfate, NAT and ice. For this report, the model calculated sulfate surface area is replaced by the assigned surface area in the calculations. Since the model explicitly calculates type I and type II PSC, transport of solid HNO_3 and H_2O are included.

GSFC and AER treat all seven reactions as surface reactions on ice and reactions A, B, D and E on NAT using the sticking coefficients as stated in JPL-97. The GSFC model adjusts the rates for reactions B, D and E to account for the dependence of these reaction rate constants on relative humidity, as suggested by Tabazadeh and Turco (1993) and Hanson and Ravishankara (1994). Both models compute the PSC surface area following the approach of Considine et al. (1994). AER assumes local thermodynamical equilibrium between gas and condensed phases for HNO_3 and H_2O with no supersaturation requirement. If the temperature is colder that the critical temperature for NAT, but warmer than the critical temperature for ice, the excess gas-phase HNO_3 and H_2O (enough to form the trihydrate) will be put into NAT particles with 0.5 μm radius and bulk density of 1.6 gm/cm^3. For temperatures colder than the critical temperature of ice, it is assumed that some of the NAT that is formed previously will be coated by ice. After the excess gas-phase HNO_3 and some of the excess H_2O is put into 0.5 μm NAT particles, the left-over excess H_2O is used to cover some of the NAT particles to form 7 μm radius type II PSC (considered as ice as far as assigning γ is concerned) with a bulk density of 0.93 gm/cm^3. As a result, both type I and type II PSCs are present in this case.

The GSFC model calculations employ supersaturation ratios of 10 for NAT and 1.4 for ice, effectively reducing the nucleation temperatures (i.e. the temperature threshold for particle formation) by 3K and 2K for type I and type II PSCs, respectively. In the GSFC model, NAT is assumed to have a bulk density of 1.6 gm/cm^3, lognormal size distribution with a mode radius of 1 μm and standard deviation of 1.8. The corresponding parameters for Type II PSC are 1.0 gm/cm^3, 10 μm radius and 1.8. Both AER and GSFC transport solid HNO_3 and solid H_2O separately with sedimentation velocities at each time step. The GSFC model takes temperature asymmetry into account when calculating surface area. The AER model uses the temperature asymmetry to calculate a weighted product for γvA.

LLNL simulates polar processing of radicals by allowing the six reactions to occur on sulfate ternary droplets. The rates are obtained from various published literature and have not been reviewed by JPL-97. A surface area density of 1 μm^2/cm^3 is imposed within 25^o of the poles when the PSC climatology of Poole and Pitts (1994) indicates a PSC frequency of occurrence exceeding 0.08. Dehydration and denitrification are represented globally (independent of the PSC surface area parameterization) by assuming that the partial pressure in excess of the saturation vapor pressure over ice (calculated using zonal mean temperatures) is removed permanently with first order time constants of 1 day for H_2O and 0.5 days for HNO_3.

The THINAIR model employs wave 1 zonal asymmetry in temperature to determine the fraction of each latitude circle which contains PSCs. Reactions A, B, D, and G are turned on for temperatures below 195 K. These reactions are assumed to remove the gas phase species with a local time constant of 9 hours.

LARC treats reactions A, B, D, E and G on PSCs using a routine supplied by Martyn Chipperfield (Univ. of Cambridge) and employing a code by Ken Carslaw to calculate rates. The parameterization calculates surface area for type I PSCs from the amount of HNO_3 condensed assuming that there are 10 NAT particles cm^{-3} and for type II PSCs from the amount of H_2O condensed assuming the radius of ice particles is 10 μm. Denitrification is triggered by the presence of type II PSCs and simulated by adopting a first order removal rate constant to remove solid phase HNO_3. Solid HNO_3 is turned back into gas-phase HNO_3 with a time constant of 1 day when the temperature is above 195 K.

The SCTM1 model employs cold heterogeneous chemistry on NAT only, using reactions A, B, D, E, and G. The occurrence of PSCs is calculated based on temperature, but is confined to latitudes higher than 47^o. Between 100 and 47 hPa, the threshold temperature is 197 K; between 47 and 22 hPa it is 193 K. The effects of PSCs are modeled in terms of the effective lifetimes of the gaseous reactants in the presence of polar stratospheric clouds. The lifetimes are calculated based on mass transfer rates taken from Turco et al. (1989). The SLIMCAT model employs cold heterogeneous chemistry on both NAT and ice particles using the PSC scheme of Chipperfield, which is also used by the LARC model. An equilibrium calculation for NAT and ice formation is used to obtain rates for reactions A, B, D, E, and G on NAT and reactions A-G on ice.

Accounting for temperature deviations away from the zonal mean is essential to correctly representing polar heterogeneous chemistry in a 2D model. A quantitative investigation of the differences between the temperature distribution (used by AER, GSFC, LLNL, and UNIVAQ) and the temperature wave (used by THINAIR) approaches has been performed by Smyshlaev et al. (1998). They found that the wave approach utilizing at least 3 waves accurately represents 3-D temperature fluctuations. Models which use the probability distribution approach based on monthly statistics are likely to predict too long a duration for PSCs. Model which use only 1 wave are likely to underpredict PSC duration.

The above discussion shows that PSC treatments in models have not converged and are different among the models. For example, the chlorine activation rate on PSCs in the LLNL and SCTM1 models does not respond to changes in concentration of HNO_3 and H_2O due to aircraft emissions. Temperature asymmetry in 2-D models is handled differently in different models. Different sets of heterogeneous reactions are employed. Different PSC surface areas are calculated even for the same conditions.

1.2.3 Chemistry Solvers

Finally, we would like to comment on the chemistry solvers used in the 2-D models. Previous model intercomparison exercises have shown that solvers in most models calculate the same partitioning of the radicals under the same constraints (solar zenith angle, overhead ozone, and local concentrations of the long-lived species) when used as a box model. Since the 2-D models transport zonal mean concentrations, the zonal mean production and loss rates are needed in the mass continuity equations for the long-lived species in these models. Different techniques are used to obtain the zonal mean production and loss rates. These include integrating the diurnally varying concentrations of the radicals obtained by explicit time marching to obtain the zonal mean rates; and using diurnally-averaged radical concentrations calculated from average solar zenith angles corrected by pre-calculated correction factors. The AER model employs the former method, while most of the other 2-D models use some variation on the latter method. The different approaches are summarized in Table 1.6.

UNIVAQ uses 24 hour average photolysis rates to calculate 24 hour average radical concentrations and the production and loss terms for long-lived species. CSIRO and LLNL use similar techniques to obtain zonal mean production and loss rates, but they employ pre-calculated factors to correct for diurnal effects. LLNL calculates the diurnal factors for a variety of chlorine loadings and sulfate surface area densities. THINAIR calculates daytime average radical concentrations. Production and loss rates for the long-lived species are an average of the daytime rates, obtained using daytime radical concentrations, and nighttime rates, most of which are assumed to be zero. Non-zero nighttime rates mainly come from washout removal or reactions involving species which have little diurnal variation. However, the $NO_2+NO_3 \rightarrow N_2O_5$ reaction is assumed to occur only at night. The GSFC model calculates daytime average concentrations of most radical species, but performs explicit time-marching for night species, including HNO_3, NO_2, NO_3, N_2O_5, HOCl, HCl, ClO, $ClNO_3$, BrO, HOBr, and $BrNO_3$.

All models except LLNL employ family groupings to minimize the number of transported species. The transported species vary somewhat, especially for the nitrogen and chlorine species. Some models transport HNO_3, N_2O_5, and HNO_4 separately from the NO_y family, while some consider these species part of NO_y. Likewise, some models transport $ClNO_3$ and HCl separately from the Cl_y family, while others do not. Transported species for each model are listed in Table 1.6. Transport is performed with a variety of advection schemes and different time steps. The frequency with which production and loss rates for the slow species are updated also varies. This information is provided in Table 1.6 also.

1.3 Sources and Sinks for Some Species

Aircraft emissions include H_2O, oxides of nitrogen, and oxidation products from sulfur in the fuel. We will discuss how these are treated in the models.

Most 2-D models assigned tropospheric concentrations of H_2O based on relative humidity and temperature. The exception is GSFC, which uses a first order removal time constant to relax the calculated

Table 1.6: Chemistry Solvers Used in the Models

Model	solve continuity equations for:	radicals and P & L for long-lived species	time step
2-D Models			
AER	source gases, O_x, HNO_3, NO_y, Cl_y, Br_y, solid HNO_3, solid H_2O	do explicit time-marching of radical species for 24 hours; use diurnally varying radical concentrations to compute 24-hour averaged P & L for long-lived species.	no operator splitting; 6 hour time step; Smolarkiewicz scheme for advection; P & L updated daily
CSIRO	source gases, O_x, NO_y, Cl_y, Br_y	24-hour averaged zonal-mean P & L calculated using correction factors obtained from a 24-hour integration of all species;uUpdated every 10 days	use variable order Bott scheme with 6 hour time step; keep same P & L for 10 days
GSFC	source gases, O_x, NO_z, HNO_3, N_2O_5, Cl_z, $ClNO_3$, Br_y, solid HNO_3, solid H_2O	use time marching for night-species (HNO_3, NO_2, NO_3, N_2O_5, HOCl, HCl, ClO, $ClNO_3$, BrO, HOBr and $BrNO_3$); use daytime averaged J-rates and dawn values of night-species to get day-averaged values of all species; diurnal average P & L updated daily	use split operator; Lin and Rood scheme; 12 hour for advection, 3 hour for vertical diffision, 24 hour for horizontal diffusion, 24 hour for chemistry
LLNL	explicit, non-family approach	use 24-hour averaged sun and diurnal factors to time march the species with 15 minute time step; diurnal factors are calculated for each chlorine loading and sulfate surface area density	use split operator; use Smolarkiewicz with 2 hour time step to do transport for 2 days, then time march chemical species for 2 days.
THINAIR	source gases, O_x, NO_y, HNO_3, N_2O_5, HNO_4, HO_x, Cl_y, Br_y	use daytime average J-rate to calculate daytime mixing ratios; 24-hour average P & L is weighted sum of day and night rates; most night rates zero except for NO_2+NO_3 $\rightarrow$ N_2O_5	Adams-Bashforth scheme with 4 hour time step
UNIVAQ	source gases, O_x, NO_y, HNO_3, $ClNO_3$, Cl_y, Br_y	use 24-hour averaged sun to calculate 24-hour averaged radical concentrations to get P & L for long-lived species; updated every 2 hours	explicit time marching with 2 hour time step
3-D Models			
LARC	source gases, O_x, NO_y, HNO_3, N_2O_5, Cl_y, HCl, $ClONO_2$, H_2O_2, solid HNO_3	use instantaneous J-rates to calculate photochemical equilibrium concentrations of the radicals every 30 minutes	forward Euler with 30 minute time step
SCTM1	source gases, O_x, NO_x, NO_y, Cl_x, Cl_y, Br_y, HO_x	QSSA method; family approach	10 minute integration step; rate coefficients updated every 40 min
SLIMCAT	source gases, O_x, NO_x, HNO_3, HNO_4, N_2O_5, NO_3, $ClNO_3$, HCl, HOCl, OClO, ClO_x, BrO_x, HBr, HOBr, BrCl, $BrNO_3$, H_2O_2	semi-implicit symmetric method (Ramaroson et al., 1992); explicit calculation of rates every time step with no diurnal averaging	operator split; 20 min for chemical time step, 30 min for dynamical time step

Table 1.7: Parmeterization of Lightning and Rainout

Model	Lightning source, source region	species removed by washout	washout time constant
2-D Models			
AER	2 MT/yr, 4-14 km	H_2O_2, CH_3OOH, CH_3CO_3H, CH_2O, HNO_3, HCl, HBr, F_x, CF_2O	5 days at the surface, 40 days at 10 km
CSIRO	2 MT/yr, 4-14 km	NO_y, Cl_y, Br_y, H_2O_2, CH_2O, CH_3COOH	3 days at the surface, 28 days at 10 km
GSFC	2 MT/yr, 4-14 km	H_2O_2, CH_3COOH, HNO_3, HO_2NO_2, HCl, HBr, HF, CClFO, and COF_2	25 days to 100 days
LLNL	5 MT/yr, from ISCCP cloud database	N_2O_5, HNO_3, HNO_4, H_2O_2, CH_2O, ClO, HCl, HOCl, $ClONO_2$, $BrONO_2$	3 days below 4 km, 50 days near tropopause
THINAIR	none	HNO_3, HNO_4, HCl, HBr, H_2O_2, CH_2O, CH_3OOH	7 days
UNIVAQ	5 MT/yr, from ECHAM3 GCM output	HNO_3, HCl, HBr, H_2O_2, CH_3COOH, CH_2O, H_2SO_4, SO_2, aerosols	5 days at the surface, 50 days at 10 km
3-D Models			
LARC	2 MT/yr, 4-14 km	H_2O_2, HNO_4, HNO_3, HCl, HBr	Mahlman and Moxim (1978)
SCTM1	7 MT/yr, from GCM output	HNO_3, individual members of Br_y, Cl_y, and NO_x families	Isaksen and Rodhe (1978)
SLIMCAT	none due to lower boundary location (at 335 K)	HNO_3, HCl, HBr, HNO_4, H_2O_2	20 days

concentrations to a pre-assigned relative humidity in the troposphere. The concentration in the stratosphere is computed using appropriate sources (in situ oxidation from methane, transport from the troposphere, and aircraft emission along flight corridors) and sinks (removal by transport out of the stratosphere and photochemical removal) in the mass continuity equation using an assigned boundary condition at the tropopause. The LARC model uses a fixed background H_2O determined from climatology. The distribution of water emissions from aircraft is solved as a separate species and added to the background in the calculations.

To simulate the concentration of NO_y in the lower stratosphere, all models include in situ oxidation of N_2O, transport of NO_x produced by lightning in the tropical upper troposphere, and NO_x emission from aircraft along the flight corridors. To simulate the effect of lightning, the models assigned a production rate of NO in the tropical upper troposphere. The exact location and the total amount differ slightly from model to model. The AER, CSIRO, GSFC and LARC models use a total of 2 MT(N)/yr uniformly (constant in molecules/cm^3/sec) distributed between 4 and 14 km in the tropics. The LLNL and UNIVAQ models use a total of 5 MT(N)/yr. The distribution in LLNL is derived from the ISCCP cloud database. The distribution in UNIVAQ is from the ECHAM GCM output. The THINAIR model does not use a lightning source of NO. Table 1.7 summarizes the lightning treatment in the models.

Most models use first order removal rate constants to simulate washout rates for soluble species, with time constants of 3-5 days at the ground, increasing to 30-50 days at 10 km. The washout rate in GSFC is longer, ranging from 25 days to 100 days. The CSIRO model allows no washout within 3 km of the tropopause. Washout in the LARC model follows the treatment of Mahlman and Moxim (1978). The SCTM1 model follows Isaksen and Rodhe (1978). SLIMCAT uses a constant 20 day washout lifetime. It should be noted that different models pick different species for washout. Some models remove NO_y, some only HNO_3 while others include N_2O_5, HNO_4, and $ClNO_3$. These are itemized by model in Table 1.7. With efficient washout, surface sources of NO_x have little impact on stratospheric NO_y.

There are significant differences in the H_2O and NO_y distributions (both in the background and the perturbed atmosphere) simulated by the models. No observational databases have been identified as the benchmarks for the climatological mean for these species in the present day atmosphere. It remains unclear as to whether the differences among the models are due to transport differences or differences in treatment of the sources. The quadratic loss of NO_y by photolysis of NO followed by the N+NO reaction may be a significant source of model NO_y differences.

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