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A Strategy for Process-Oriented Validation of Coupled Chemistry–Climate Models

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Evaluating CCMs with the presented framework will increase our confidence in predictions of stratospheric ozone change.

The decreasing levels of halogens in the stratosphere should lead to a gradual recovery from the chemical ozone depletion that has occurred over the past decades (WMO 2003). However, climate change resulting from increases in greenhouse gas concentrations will influence the stratosphere through a range of radiative, dynamical, and chemical mechanisms. A schematic diagram showing the

principal regions and processes in the stratosphere is displayed in Fig. 1. An improved understanding of these processes and, more generally, of the interaction between chemistry and climate is needed if credible predictions of the future levels of stratospheric ozone, and its impact on climate and surface UV radiation, are to be made. Such predictions are required for the WMO/UNEP and IPCC assessments as part of

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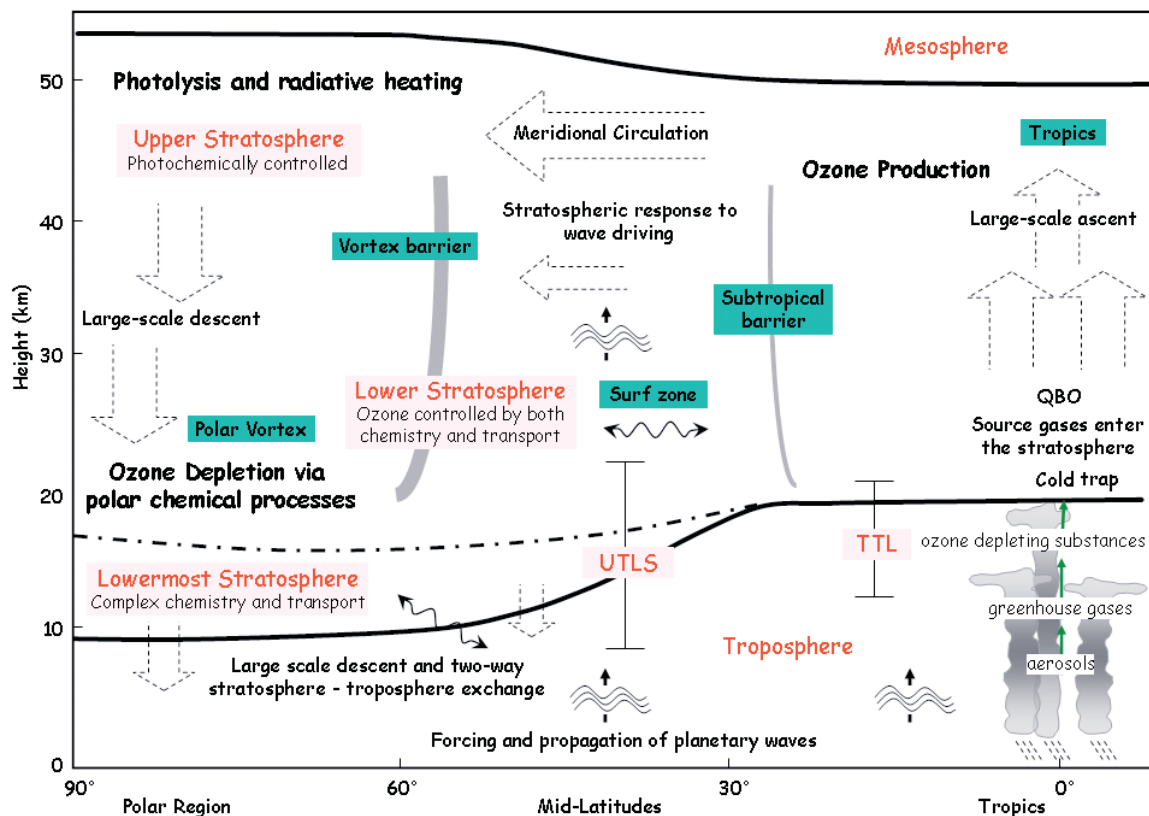


FIG. 1. Schematic diagram of the principal regions and processes in the upper troposphere and lower stratosphere. Broad arrows denote diabatic circulation and wavy arrows denote transport along isentropic surfaces. The average position of the tropopause is shown by the lower thick-black line, the average position of the stratopause by the upper thick-black line, and the 380-K isentropic surface by the thick-black dot-dashed line. The vertical bars denote the range of the UTLS and TTL.

the policy formulation processes associated with the Montreal Protocol and the Kyoto Protocol on Climate Change.

A number of CCMs with detailed descriptions of the stratosphere have been developed over the last 5–10 yr in order to provide these predictions. However, the predictions of current CCMs produce a wide range of results concerning the timing and extent of ozone-layer recovery, both in the Arctic and Antarctic winters (WMO 2003). The main features of current CCMs are summarized in Table 1. Figure 2 shows as an example the modeled minimum Antarctic total ozone for the time period 1960–2060 (Austin et al. 2003). In contrast to CTMs, which specify the meteorological conditions, CCMs specify the chemical and dynamical forcings and predict the resulting change in the chemistry–climate system. They simulate a climate that bears a statistical relationship to the real atmosphere, and so a comparison of model results with measurements must be performed in a statistical manner. This is problematic, because it appears to take many decades of observations to define a robust

stratospheric climatology, especially in the Arctic winter. While tropospheric climate models can be validated, in part, by their ability to reproduce the climate record over the twentieth century, the paucity of stratospheric climate data prior to the satellite era (post-1979) limits such possibilities for model validation of stratospheric change.

For these reasons, the validation of CCMs requires a process-oriented basis to complement the standard comparison of models with climatologies of observations. By focusing on processes, models can be more directly compared with measurements. In this case, natural variability becomes an aid because it allows dependencies between model fields to be examined in a larger variable space and, thereby, makes identifying cause-and-effect relationships within a model more reliable. An important example is the physically based relationship between planetary wave drag and polar temperatures (see the section labeled “Stratospheric response to wave drag”), which can be quantified by producing a scatterplot of the two quantities with each point representing a different year. In the context

TABLE 1. Main features of current CCMs. CCMs are listed alphabetically. The horizontal resolution is given in either degrees latitude x degrees longitude (grid point models), or as T21, T30, etc., which are the resolutions in spectral models corresponding to triangular truncation of the spectral domain with 21, 30, etc., wavenumbers, respectively. All CCMs have a comprehensive range of chemical reactions except that in the UMUCAM model the chemistry is parameterized. The coupling between chemistry and dynamics is represented in all models, but to a different degree. All models include O-GWD schemes, most models additionally include NonO-GWD.

Model	Horizontal resolution	No. vertical levels/upper boundary	Coupling chemistry/dynamics	GWD	Group and location	Reference
AMTRAC	2° x 2.5°	48/0.0017 hPa	O ₃ , H ₂ O	O-GWD + NonO-GWD	GFDL, USA	Anderson et al. (2004), Austin (2002)
CCSR/NIES	T21	30/0.06 hPa	O ₃ , H ₂ O, CH ₄ , N ₂ O, CFCs	O-GWD + NonO-GWD	NIES, Tsukuba, Japan	Nagashima et al. (2002), Takigawa et al. (1999)
CMAM	T32 or T47	65/0.0006 hPa	O ₃ , H ₂ O	O-GWD + NonO-GWD	MSC, University of Toronto, and York University, Canada	Beagley et al. (1997), de Grandpré et al. (2000)
E39/C	T30	39/10 hPa	O ₃ , H ₂ O, CH ₄ , N ₂ O, CFCs	O-GWD	DLR Oberpfaffenhofen, Germany	Dameris et al. (2005)
ECHAM5/MESSy	T42	90/0.01 hPa	O ₃ , H ₂ O, CH ₄ , N ₂ O, CFCs, NO ₂ , aerosols	O-GWD + NonO-GWD	MPI Mainz, MPI Hamburg, DLR Oberpfaffenhofen, Germany	Jöckel et al. (2004), Roeckner et al. (2003), Sander et al. (2004)
FUB-CMAM-CHEM	T21	34/0.0068 hPa	O ₃ , H ₂ O, CH ₄ , N ₂ O, CFCs	O-GWD + NonO-GWD	FU Berlin, MPI Mainz, Germany	Langematz et al. (2005)
GCCM	T42	18/2.5 hPa	O ₃	O-GWD	University of Oslo, Norway; SUNY Albany, USA	Wong et al. (2004)
GEOS CCM	2° x 2.5°	55/80 km	O ₃ , H ₂ O, CFCs, CH ₄ , N ₂ O	O-GWD + NonO-GWD	NASA GSFC, USA	S. Pawson, and P. A. Newman 2005, personal communication
GISS	4° x 5°	23/0.002 hPa	O ₃ , H ₂ O, N ₂ O, CH ₄ , CFCs	O-GWD + NonO-GWD	NASA GISS, USA	Schmidt et al. (2005a, manuscript submitted to <i>J. Climate</i>)
HAMMONIA	T31	67/2.10 ⁻⁷ hPa	O, O ₂ , O ₃ , H ₂ O, N ₂ O, CO ₂ , CH ₄	O-GWD + NonO-GWD	MPI Hamburg	Schmidt et al. (2005b, manuscript submitted to <i>J. Climate</i>)
LMDREPRO	2.5° x 3.75°	50/0.07 hPa	O ₃ , H ₂ O, N ₂ O, CH ₄ , CFCs	O-GWD + NonO-GWD	IPSL, France	S. Bekki and D. Hauglustaine 2005, personal communication
MRI	T42	68/0.01 hPa	O ₃	O-GWD + NonO-GWD	MRI, Japan	Shibata and Deushi (2005); Shibata et al. (2005)
MAECHAM4/CHEM	T30	39/0.01 hPa	O ₃ , H ₂ O, CH ₄ , N ₂ O, CFCs	O-GWD + NonO-GWD	MPI Mainz, MPI Hamburg, Germany	Manzini et al. (2003), Steil et al. (2003)
SOCOL	T30	39/0.01 hPa	O ₃ , H ₂ O	O-GWD + NonO-GWD	PMOD/WRC and ETHZ, Switzerland	Egorova et al. (2005)
ULAQ	10° x 20°	26/0.04 hPa	O ₃ , H ₂ O, CH ₄ , N ₂ O, CFCs, NO ₂ , aerosols	Rayleigh frict. + vert. diffusion	University of L'Aquila, Italy	Pitari et al. (2002)
UMETRAC	2.5° x 3.75°	64/0.01 hPa	O ₃	O-GWD + NonO-GWD	Met Office, UK	Austin (2002), Austin and Butchart (2003)
UMSLIMCAT	2.5° x 3.75°	64/0.01 hPa	O ₃ , N ₂ O, CH ₄ , H ₂ O	O-GWD + NonO-GWD	University of Leeds, UK	Tian and Chipperfield (2005)
UMUCAM	2.5° x 3.75°	58/0.01 hPa	O ₃	O-GWD, Rayleigh friction	University of Cambridge, UK	Braesicke and Pyle (2003 and 2004)
WACCM3	2° x 2.5° 4° x 5°	66/140 km	O ₃ , H ₂ O, N ₂ O, CH ₄ , CFCs	O-GWD + NonO-GWD	NCAR, USA	Sassi et al. (2005)

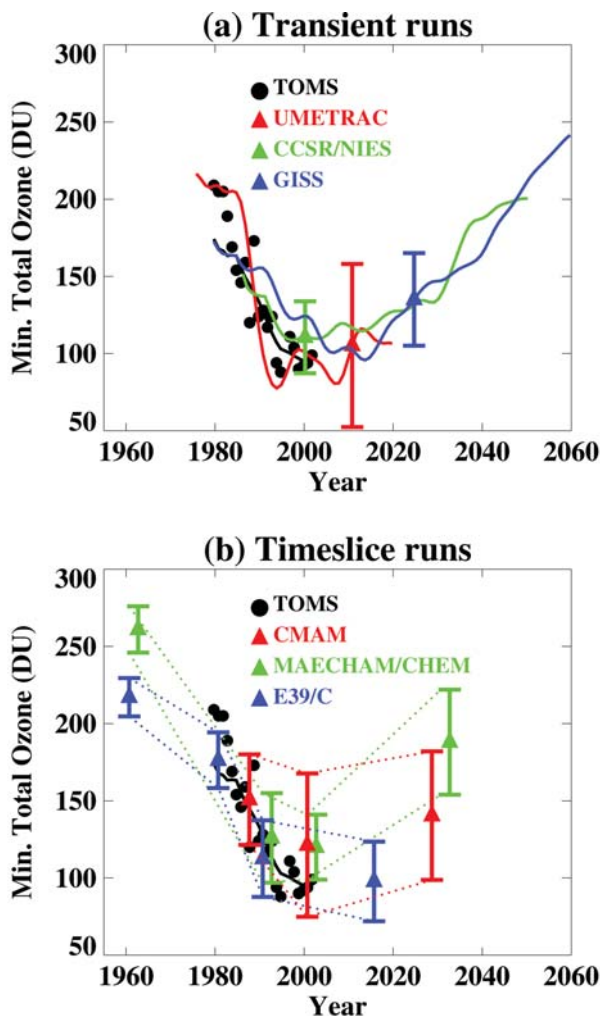


FIG. 2. Modeled and measured values of minimum total column amounts of ozone in the Antarctic (Sep–Nov). Results are shown for the period 1960–2060 derived from (a) transient runs and (b) time-slice runs of different CCM model experiments in comparison with TOMS satellite data (for the period 1980–2001). The main features of the CCMs identified in the legend are summarized in Table 1. The solid lines in (a) show the results of a Gaussian smoother applied to the results of individual years with vertical bars denoting twice the standard deviation. For the time-slice experiments, the dotted lines are drawn to assist in estimating trends. Transient as well as time-slice experiments show reasonable agreement with TOMS observations. The uncertainty in both experiment types and the differences between CCMs increases significantly for future years. Specifically, the start dates of ozone recovery, defined by when the decadal averaged minimum ozone first begins to increase, vary significantly. Similar CCM experiments for ozone depletion in Arctic winters show poorer agreement with the data and between models (WMO 2003). (Figure from Austin et al. 2003.)

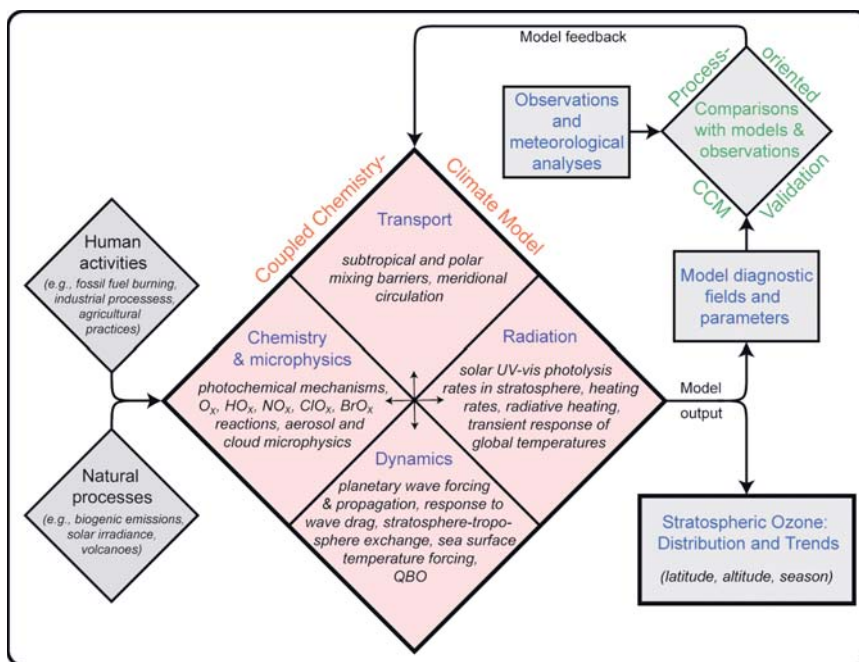
of stratospheric GCMs (i.e., those without chemistry), process-oriented validation represents the level-II tasks within GRIPS (Pawson et al. 2000). A first attempt at process-oriented validation of stratospheric CCMs is summarized in Park et al. (1999), WMO (2003), and Austin et al. (2003).

Until very recently, the components of the Earth's system (ocean dynamics, marine biogeochemistry, tropospheric and stratospheric chemistry, atmospheric dynamics and physics, terrestrial ecosystems, ecology, etc.) have been investigated separately by different disciplines. As we are moving toward more complex models that include different components of the Earth's system, the strategy of setting up benchmarks and criteria for model validation presented in this paper is also important for other modeling communities in order to consolidate their results and conclusions. Similar efforts are needed for the other components of an Earth System Model to advance our understanding of the various processes and to ensure that employing such complex models would be beneficial.

LONG-TERM APPROACH TO CCM VALIDATION. In this work we present a strategy for a more long-term comprehensive approach to CCM validation centered on four main categories: transport, dynamics, radiation, and stratospheric chemistry and microphysics. For each process, Table 2 presents the associated model diagnostics, variables relevant for validation, and sources of observational or other data that can be used for validation. The accompanying text discusses the importance of the selected processes to CCM validation and the utility of the selected diagnostics in a validation study. The relevant time scale for the diagnostic depends on the process and must be borne in mind when comparing models and measurements.

A schematic diagram of the approach to CCM validation is shown in Fig. 3. The strategy resulted from discussions at the workshop on process-oriented validation of CCMs held at Grainau, Germany, in November 2003 (Eyring et al. 2004). Members of the CCM and CTM communities came together with members of the measurement and data analysis communities to develop ideas on this issue. The role of the latter communities was crucial in understanding both the opportunities and the limitations presented by the available data. The size of the task involved with a complete validation exercise quickly became apparent and so the approach taken was to develop a range of diagnostics that can be worked through as time and interest allow. Although the focus of the

FIG. 3. Schematic diagram of the presented approach to CCM validation. The centerpiece is a CCM comprised of four basic process categories: transport, dynamics, radiation, and stratospheric chemistry & microphysics. The four categories are fundamentally interdependent and interactive and require as inputs, knowledge of human activities and natural processes. These inputs help quantitatively define processes in the atmosphere and expectations for future changes. Trends in atmospheric constituents and parameters associated with climate forcing are examples of important inputs. The CCM output includes a wide array of parameters and diagnostics associated with the four different categories. The distribution of stratospheric ozone is highlighted separately here because of the strong contemporary interest in halogen-based ozone depletion and the recovery of the ozone loss that has developed over recent decades. The comparisons of model diagnostics and other outputs with atmospheric observations and meteorological analyses are the key to process-oriented CCM validation. In the accompanying Table 2 and discussions, we define the components of these comparisons. Finally, the results of the comparisons can be used to provide feedback to the representation of processes in CCMs in order to improve subsequent CCM validation comparisons. In this way, the uncertainties in future trends in stratospheric ozone and other key model outputs can be reduced.



present discussion is on defining a methodology for the validation of CCMs, we recognize that observational uncertainties are a potentially important component of CCM evaluation. Observational uncertainties can influence the outcome of model–data consistency tests (see, e.g., Santer et al. 2003a, 2003b) and should be explicitly accounted for in any CCM validation strategy.

Stratospheric transport. Transport in the stratosphere involves both meridional overturning (the residual circulation) and mixing, which together represent the Brewer–Dobson circulation. The most important aspects are the vertical (diabatic) mean motion and the horizontal mixing. Horizontal mixing is highly inhomogeneous, with transport barriers in the subtropics and at the edge of the wintertime polar vortex; mixing is most intense in the wintertime “surf zone”—that is, the region surrounding the polar vortex—and is comparatively weak in the summertime extratropics. Accurate representation of this structure in CCMs is important for the ozone distribution itself, as well as for the distribution of chemical families and species

that affect ozone chemistry (NO_y , Cl_y , H_2O , CH_4 ; for explanations of chemical formulas used throughout, cf. appendix B). Within both the Tropics and the polar vortex, the key physical quantities to be represented are the degree of isolation and the diabatic ascent or descent, respectively. The impact of diabatic ascent or descent on the actual vertical motion of chemical species depends on the degree of isolation.

SUBTROPICAL AND VORTEX-MIXING BARRIERS. Useful information can be obtained from instantaneous snapshots of tracer fields, which makes the model–measurement comparison straightforward. For this purpose there is a wealth of high-quality observational data available. A simple check on the degree of isolation is provided by the sharpness of latitudinal gradients of long-lived species (CH_4 , N_2O , CFC-11), while a more detailed diagnosis is obtained from the structure of chemical correlations and from PDFs of such species. Just above the tropical tropopause, where the tropical mixing barrier appears to be fairly leaky, transport into mid-latitudes can be quantified by the propagation of the annual cycle in CO_2 and H_2O , which has been well

TABLE 2. List of core processes to validate CCMs with a focus on their ability to model future stratospheric ozone.				
Process	Diagnostic ^a	Variables	Data	References ^b
Stratospheric transport				
Subtropical and polar mixing barriers	PDFs of long-lived tracers	N ₂ O, CH ₄ , CFC-II, etc.; potential vorticity (PV)	Satellite and in situ (aircraft, balloons) chemical measurements and meteorological analyses ^c	Strahan and Douglass (2004)
	Latitudinal gradients of long-lived tracers			Sankey and Shepherd (2003)
	Correlations of long-lived tracers			Sankey and Shepherd (2003)
	Phase and amplitude of tropical CO ₂ or H ₂ O annual cycle in lower stratosphere (tape recorder)	CO ₂ , H ₂ O or idealized annually repeating tracer	Satellite and in situ measurements	Hall et al. (1999), Mote et al. (1996)
	Annual cycle of streamer frequency	Daily PV (maybe long-lived tracers)	Meteorological analyses ^c satellite measurements	Eyring et al. (2003), Waugh (1996), Waugh et al. (1997)
Meridional circulation	Mean age	Conserved tracer with linearly increasing concentration, SF ₆ or CO ₂	In situ measurements	Hall et al. (1999), Waugh and Hall (2002)
	Correlation of interannual anomalies of total ozone and Planetary wave flux	Total ozone and heat flux at 100 hPa, zonal and monthly means	Satellite measurements, meteorological analyses ^c	Randel et al. (2002), Weber et al. (2003)
	Vertical propagation of tracer isopleths	H ₂ O or CO ₂ or idealized annually repeating tracer (tropics), CH ₄ or N ₂ O (polar)	In situ and ground-based (polar only) and satellite data	Hall et al. (1999), Kawamoto and Shiotani (2000)
	Diabatic velocity, TEM stream-function	Diabatic velocity, residual velocities	Diabatic velocity inferred from radiative calculation	Eluszkiewicz et al. (1996), Rosenlof (1995)
UTLS transport	Vertical gradients of, and correlations between, chemical species in the extratropical UTLS	CO ₂ , SF ₆ , H ₂ O, CO, O ₃ , HCl	Balloon, aircraft	Hoor et al. (2002), Marcy et al. (2004)
	Relation between meteorological indices (e.g., tropopause height) and total ozone	Daily winds, temperature, geopotential height, total O ₃	Meteorological analyses ^c satellite measurements, ozonesondes	Santer et al. (2003a)
	Diabatic velocity, vertical O ₃ profiles in TTL	Diabatic velocity, vertical O ₃ profiles	Diabatic velocity inferred from radiative calculation, ozonesondes	Thompson et al. (2003)
Dynamics				
Forcing and propagation of planetary waves	WFA, PW spectrum (variances and covariances)	Temperature, geopotential height, horizontal winds, High-frequency (daily) data	Meteorological analyses ^c	Hayashi (1982)
	Hemispheric ozone variability indices	Total column ozone over several years	Satellite measurements of total ozone (e.g., TOMS, GOME, or SCIAMACHY)	Erbertseder et al. (2005, manuscript submitted to <i>Atmos. Chem. Phys. Discuss.</i>)

^a In addition to traditional model validation (climatological means, interannual variations etc.).

^b Listed references only provide examples.

^c Due to uncertainties use several analyses, not one.

^d Intercomparison currently not possible because process not included in most CCMs.

observed in aircraft measurements. The ascent rate of tracer isopleths in the Tropics is visible in the “tape recorder” phenomenon seen in altitude-versus-time cross sections of H₂O mixing ratios.

MERIDIONAL CIRCULATION. Both horizontal mixing and the residual circulation are largely driven by the momentum deposition (wave drag) from planetary waves propagating from the troposphere

TABLE 2. Continued.				
Process	Diagnostic ^a	Variables	Data	References ^b
Dynamics (continued)				
Stratospheric response to wave drag	Annual cycle of temperatures in Tropics and extratropics	Zonal monthly mean temperature, residual streamfunction	Meteorological analyses ^c in situ and space-based observations, profile data	Pawson et al. (2000)
	Planetary wave flux vs polar temperature, lagged in time	Heat flux ($v'T$) at 100 hPa (Jan/Feb), temperature at 50 hPa (Mar), zonal monthly means		Austin et al. (2003), Newman et al. (2001)
	Vortex definition, structure and occurrence of sudden/final warmings	PV, horizontal winds, temperature, area colder than PSC temperature, Vortex area/equivalent latitude; warming statistics; high-frequency (daily) 3D fields		Limpasuvan et al. (2004), Manney et al. (2005), Nash et al. (1996), Waugh and Randel (1999)
	Downward control integral, also scatterplot of planetary wave drag versus gravity wave drag	w^* from model PWD, GWD, other drag zonal and monthly means	Meteorological analyses ^c total drag inferred from diabatic heating calculation	Beagley et al. (1997)
	Persistence (e.g., leading empirical orthogonal functions), including Holton-Tan	Geopotential height, temperature, multiyear time series (means, frequency spectra)	Meteorological analyses ^c	Waugh et al. (1999), Zhou et al. (2000)
Stratosphere–troposphere exchange	Daily mass estimates of the lower-most stratosphere	Daily 380-K isentropic pressure and tropopause pressure	Meteorological analyses ^c	Olsen et al. (2002)
QBO^d	Horizontal winds and temperature	Horizontal winds and temperature, zonal and monthly means	Meteorological analyses ^c	Butchart et al. (2003), Giorgetta and Bengtsson (1999)
Radiation				
Solar UV–visible photolysis in stratosphere	Radiative transfer of 260–800-nm solar flux, photolysis rates comparison up to 95° solar zenith angle including clouds	Actinic flux (direct and scatter), photolysis rates of O ₃ and NO ₂ at local noon pressure, ozone, stratospheric aerosols, tropospheric clouds, aerosols and ozone	Direct flux measurements (balloon, aircraft), inferred photolysis rates (aircraft)	Bais et al. (2003), Hofzumahaus et al. (2004), Kylling et al. (2003)
Heating rates	Comparison of thermal and solar heating rates in offline runs employing column version of CCM radiation codes	Heating rates and irradiances from CCM radiation code, with a prescribed and standardized set of input atmospheric profiles	Use sophisticated reference radiation models for comparison (line by line) NLTE, discrete-ordinate scattering, etc.	Forster et al. (2001), Oinas et al. (2001)
Radiative heating	Global average of temperature profile	Annually averaged global trace gas and clouds fields, temperature	Assimilated fields derived from satellite and sonde data, meteorological analyses ^c	Pawson et al. (2000)
	Long-term globally averaged transient temperature changes	Changes in ozone, water vapor and high clouds, greenhouse gases, hydrofluorocarbons, aerosols, etc.	SSU/MSU satellite time series	Shine et al. (2003)

into the stratosphere, with more wave drag leading to a stronger Brewer–Dobson circulation. The relationship between the wave flux and the residual circulation is quantified, through temperature, in the dynamics diagnostics. With regard to chemical

transport, the seasonal cycle of O₃ in the extratropics exhibits a marked build-up during the winter/spring period due to the Brewer–Dobson circulation. Years with greater planetary wave flux have a greater ozone build-up, a relationship that is well established from

TABLE 2. Continued.				
Process	Diagnostic ^a	Variables	Data	References ^b
Stratospheric chemistry and microphysics				
Photochemical mechanisms and short-time-scale chemical processes	Offline box model comparisons of fast chemistry (of order 1 week or less)	Full chemical constituents (O_3 loss due to O_x , HO_x , NO_x , ClO_x , BrO_x , J values)	HO_x : balloon, shuttle, aircraft; NO_x : satellite, shuttle, balloon, aircraft; ClO_x : satellite, shuttle, balloon, aircraft; BrO_x : aircraft	Gao et al. (2001), Salawitch et al. (1994)
Long-time-scale chemical processes	Comparison of abundance of reservoirs and radical precursors	Instantaneous output of all chemical constituents and temperature (one per month)	Satellite measurements of reservoirs and precursors	Millard et al. (2002), Salawitch et al. (2002), Sen et al. (1999)
	Tracer–tracer relations	O_3 , NO_y , CH_4 , H_2O , N_2O		Chang et al. (1996), Fahey et al. (1996), Müller et al. (1996)
Polar processes in winter/spring	Partitioning of species within the families	Species from families (ClO_x , NO_x , HO_x , BrO_x , Cl_y , NO_y , Br_y) temperature, PV from wind fields	Satellite and aircraft measurements	Park et al. (1999), Pierson et al. (2000)
	Chemical ozone loss vs PSC activity	O_3 , passive O_3 tracer, O_3 production/loss rate, PV from wind fields, temperature	Chemical ozone loss diagnosed from frequent ozone profiles in the vortex over several years, meteorological analyses ^c	Chipperfield et al. (2005), Rex et al. (2004)
Summer processes	Ozone changes in polar regions	Total ozone, full chemical constituents, temperature	Satellite measurements of total ozone	Fahey and Ravishankara (1999)
	Ozone changes in midlatitude regions			Koch et al. (2003)
Denitrification and dehydration	NO_y vs tracer	NO_y , HNO_3 , N_2O , CH_4 , etc.	Satellite measurements of HNO_3 , H_2O , CH_4 ; aircraft observations of NO_y , H_2O , CH_4 , N_2O ; PSC size distributions	Gao et al. (2001), Popp et al. (2001), Santee et al. (2002)
	$H_2O + 2 CH_4$	H_2O particle-flux rates added to daily polar chemistry, instantaneous output, CH_4		Nedoluha et al. (2000), Park et al. (2004)
Aerosols and cloud microphysics	Cirrus cloud frequency of occurrence; H_2O distribution	Ice water content, water vapor, temperature, aerosol size distribution	Aircraft and satellite data; process/cloud-resolving models	Clark et al. (2003), Read et al. (2004), Thomas et al. (2002), Wang et al. (1996)
Stratospheric aerosol processes	Sulfuric acid size distribution, aerosol optical extinction	Sulfuric acid mass, particle number concentration, water vapor, temperature	Satellite and in situ measurements of aerosols; aerosol climatologies	Thomason and Peter (2005; submitted SPARC report)
	Temperature response in the lower stratosphere, chlorine and nitrogen partitioning after major volcanic eruptions	All species from chlorine and nitrogen families, temperature	Satellite and aircraft measurements for temperature response, e.g., MSU data	Dessler et al. (1997), Fahey et al. (1993), Labitzke and McCormick (1992)

^a In addition to traditional model validation (climatological means, interannual variations etc.).

^b Listed references only provide examples.

^c Due to uncertainties use several analyses, not one.

^d Intercomparison currently not possible because process not included in most CCMs.

observations and provides a good diagnostic for CCM validation. The Brewer–Dobson circulation also determines the mean age of air, which can be validated from measurements of long-lived species

that have increasing concentrations with time (e.g., SF_6 , CO_2). Mean age of air has been found to be a very powerful diagnostic for identifying model deficiencies (Park et al. 1999).

UTLS TRANSPORT. Transport in the UTLS region is complex. The extratropical tropopause “break” is a barrier to quasi-horizontal mixing, causing significant contrasts in chemical species between the extratropical lowermost stratosphere and the tropical upper troposphere. The degree of isolation can be assessed by the sharpness of horizontal or isentropic gradients at the tropopause (because tropopause height changes with latitude), and with chemical correlations (e.g., O_3 versus CO). There is a robust relationship between variations in total O_3 and in tropopause height, which provides a potentially important diagnostic for CCM validation. The TTL is marked by changes in the vertical stability and in chemical species beginning below the tropical tropopause. Processes in this layer are important for setting chemical boundary conditions for the stratosphere. In addition, convective processes and microphysics affect water vapor and the chemistry of ozone and other minor species. These radiatively active gases can have large impacts on the climate of the UTLS.

Dynamics. The basic dynamical state of the stratosphere which controls transport is defined by a number of physical processes. These include the forcing mechanisms and propagation of planetary-scale Rossby and gravity waves, wave–mean flow interaction, and the diabatic circulation. Correct reproduction of the climatological mean state of the stratosphere by CCMs, including interhemispheric differences, and interannual and intraseasonal variability, is important but not sufficient: the basic dynamical mechanisms must be well represented in the underlying GCMs on which the CCMs are based if future changes are to be modeled credibly.

FORCING AND PROPAGATION OF PLANETARY WAVES. The properties of planetary waves (such as their generation, propagation through the stratosphere, and role in the momentum budget of the stratosphere [i.e., the stratospheric response to planetary wave drag (PWD)], can be determined by analyzing planetary wave patterns at different altitudes between the free troposphere and the upper model layers. A WFA can help to resolve transient waves at distinct wavenumbers into standing and eastward- or westward-traveling waves at different frequencies. The amplitudes and phases of the zonal quasi-stationary planetary waves in the lower stratosphere can be found by analyzing total ozone fields using spectral statistical methods. Here, the total ozone column is considered as a conservative tracer to illuminate the variability of wave structures in the lower

stratosphere. Spectral harmonic analysis can be applied to derive the wave parameters from the ozone distribution. The spectral properties can further be used to calculate hemispheric ozone variability indices, which are defined as the hemispheric means of the zonal amplitude of the planetary wavenumbers 1 and 2.

STRATOSPHERIC RESPONSE TO WAVE DRAG. Planetary waves can only propagate into the stratosphere when the winds are relatively weak westerlies, and so the Brewer–Dobson circulation is stronger in the winter hemisphere. The wave drag can be quantified from the net planetary wave flux into the stratosphere, normally taken to be $v'T'$ (heat flux) at 100 hPa. Correlations of Eliassen–Palm fluxes (whose meridional and vertical components are respectively proportional to the meridional eddy momentum and eddy heat fluxes) with dynamical fields (e.g., temperature, wind speed) and parameters (e.g., size and persistence of the polar vortex, PSC potential) are necessary to investigate the stratospheric response to wave drag and its consequences for chemical and physical processes in CCMs. Moreover, the ability of CCMs to reproduce correctly the seasonality of the Brewer–Dobson circulation can be checked by comparing the calculated cross sections of the residual circulation mass streamfunction (latitude versus height) with those based on reanalyses (e.g., NCEP, ERA-40). The drag from dissipating gravity waves also plays a significant role in the stratospheric circulation, especially in the Antarctic winter. Here direct observations are not available, but the role of gravity wave drag in different models, and its response to changes in planetary wave drag, can be compared with appropriate diagnostics.

STRATOSPHERE–TROPOSPHERE EXCHANGE. The lowermost stratosphere is the region where isentropic surfaces intersect both the troposphere and stratosphere. The lowermost stratosphere is roughly bounded by the tropopause at the bottom and the 380-K isentropic surface at the top. The month-to-month variation of the mass of this atmospheric layer is sensitive to a number of transport and dynamical processes. Meteorological observations can be used to test this relationship in models.

QBO. CCMs are now just beginning to simulate the QBO, usually through the inclusion of enhanced GWD. It will be important to confirm that the models are obtaining a QBO for the right reasons, and that the extratropics respond in the correct manner. Meteorological reanalyses and radiosonde records can be used for this.

Radiation. Radiative calculations are used in CCMs to derive photolysis rates and heating rates. Photolysis rates in the stratosphere control the abundance of many chemical constituents that, in turn, control chemically active constituents, such as ozone. At the same time these trace gases feedback on temperature and, thus, circulation through the radiative heating rates. At present, most models calculate radiative heating rates and photolysis rates in an inconsistent manner. For example, the spherical geometry of the Earth might be included in the photolysis rate calculation, but not in the heating rate calculation. Also, different radiation schemes are usually employed for the two calculations. Such inconsistencies should be avoided. There are currently not enough high-quality measurements that can be used to validate the important radiative processes in global models. Presently, the best radiative models (currently not included in CCMs) provide an important complement to available measurements for CCM validation. Accordingly the approach taken here (unlike the other three categories) is to perform detailed model comparisons between the best radiative models and the radiation modules actually used in CCMs. We evaluate the photolysis and radiative heating rate calculations separately.

SOLAR UV–VISIBLE PHOTOLYSIS IN THE STRATOSPHERE. A photolysis rate generally requires knowledge of the actinic fluxes at solar and UV–visible wavelengths (190–800 nm) as a function of altitude and solar zenith angle. Accurate calculations of these fluxes require accurate representation of scattering, albedo, and refraction. Particular concerns in photolysis rate calculations for the lower stratosphere are the effect of tropospheric cloudiness, which can significantly increase the rates for certain gases and photolysis at solar zenith angles greater than 90°. Diagnostic parameters for photolysis rates in CCM comparisons include the radiative transfer of UV–visible wavelengths and calculated rates for individual gases. The distributions of pressure, ozone, stratospheric aerosols, and tropospheric clouds are important variables in such model comparisons. As a minimum test, the photolysis rates of O₃ and NO₂ should be stored as three-dimensional fields and compared to observations. In addition, actinic fluxes at the ground in different wavelength intervals should be compared.

RADIATIVE HEATING RATES. Radiative heating is the fundamental link between ozone and climate. As its calculation plays the central part in CCM feedbacks,

it is extremely difficult to separate cause and effect in a fully coupled model. Radiative heating rate calculations can only be truly evaluated in an offline comparison of radiation schemes. Currently, the lack of this comparison is one of the most important limitations in understanding CCM differences. A set of standardized background atmospheres and radiation scheme inputs should be compiled, along with a reference set of calculations from several state-of-the-art line-by-line and scattering models. Differences in radiative heating rates and trace gas fields can then be used to evaluate differences between the globally averaged climatological temperature of CCMs and their temperature response to changes in greenhouse gases loadings and other perturbations.

RADIATIVE HEATING WITHIN AN ONLINE FRAMEWORK. To evaluate radiative heating within an online framework, the long-term global-mean temperature climatology of CCMs can be compared to observations. An online framework allows a combined test of the model's background atmosphere and radiative heating profile. Also, the globally averaged transient temperature changes over both a single year and the past ~25 yr can be compared to SSU and MSU satellite observations. This tests both the evolution of forcing agents, as well as the radiative heating and the radiative relaxation time in the model.

Stratospheric chemistry and microphysics. One of the ways in which chemistry and dynamics are coupled is the temperature dependence of many chemical reaction rates. The importance of local control of ozone by chemistry relative to transport varies substantially between various times and places. In the upper stratosphere transport plays a role by controlling the concentrations of long-lived tracers such as inorganic chlorine, but photochemical time scales are so short that transport has a minimal direct impact on ozone. However, in the lower stratosphere, the photochemical time scales are rather longer (typically of the order of months) and interactions with dynamics are complex and more challenging to model accurately. Aerosols have an important role in chemistry in the lower stratosphere, since reactions can take place within or on the particles. Consequently, even though the photochemical lifetime of ozone is typically many months in the lower stratosphere, rapid chemical loss of ozone occurs in the Antarctic lower stratosphere, following exposure of air to polar stratospheric clouds.

PHOTOCHEMICAL MECHANISMS AND SHORT-TIME-SCALE CHEMICAL PROCESSES. Validation of chemical processes on time

scales of a couple of days can be accomplished by examining the full field of chemical constituents as well as reaction and photolysis rates found by each CCM. The comparisons should focus on times and places where the ozone loss efficiency by each of the catalytic families, as well as ozone production, can be defined from observations of the radical species. Full chemical constituent output from the CCMs (including diurnal variations, if available) would be requested for a handful of times and places of a long-term run, designed to coincide with the availability of atmospheric observations. The offline models would be constrained by abundances of long-lived radical precursors from the CCMs, to provide a meaningful test of the rapid chemistry within each CCM. The offline simulations should include Lagrangian calculations to fully understand the impact of air mass history on radical concentrations for the selected cases. Also, measurements exist for evaluation of CCM photolysis rates.

LONG-TIME-SCALE CHEMICAL PROCESSES. In contrast, the investigation of long-time-scale photochemical processes needs to be done within the CCM itself, as transport has a significant impact. All the model 3D chemical fields need to be output, as well as the appropriate dynamical variables (e.g., temperature). One instantaneous “snapshot” per month should be sufficient for the purpose of comparing the abundances of model reservoirs and radical precursors. The interrelations between long-lived tracers also need to be compared in detail with observations.

POLAR CHEMISTRY IN WINTER/SPRING. The largest chemical O_3 losses have occurred in winter/spring, when low temperatures lead to the formation of condensed matter and heterogeneous chemistry becomes important. Some aspects of heterogeneous chemistry can be investigated in box model tests, but because of the possible importance of denitrification and dehydration, as well as transport, 3D simulations are required for a complete analysis. Validating polar processes requires an extensive set of model chemical and particle fields with daily frequency. Measurements from a number of balloon and aircraft campaigns can be used to test the model chemical (and microphysical) schemes. The accumulated winter/spring polar O_3 loss is an important contributor to midlatitude trends. A validation of this modeled quantity, including its sensitivity to interannual temperature changes, is crucial for one of the main goals of CCM calculations—the prediction of polar O_3 recovery. An empirical relation between chemical O_3 loss and temperature can be used for this validation.

SUMMER PROCESSES. In summer, atmospheric chemistry in polar regions is a special case because of the continuous, or near-continuous, daylight. These conditions have revealed some possible discrepancies in NO_x chemistry. This has an impact on ozone amounts directly in the polar regions and also in midlatitudes via transport from the polar regions.

DENITRIFICATION AND DEHYDRATION. These important processes occur in the cold winters of both hemispheres and enhance O_3 loss. However, their current representation in CCMs is crude, contributing to uncertainties in polar O_3 . This is further complicated by a) an incomplete understanding of the mechanism for denitrification and b) CCM polar temperature biases. The CCM representation of denitrification can be investigated by analyzing the key nitrogen containing species, NO_y and HNO_3 , as a function of well-conserved tracers (e.g., N_2O). Similarly, the sum $H_2O + 2 CH_4$ is approximately conserved in the stratosphere except in the presence of dehydration.

AEROSOLS AND CLOUD MICROPHYSICS. Aerosol and cloud-related processes affect the whole UTLS region through changes in the radiative balance and heterogeneous reactions. Microphysical processes and gas-particle interactions are important to understand dehydration and denitrification in the polar region and the regulation of the overall stratospheric water vapor budget. The required model variables are particle number, mass densities, and relative humidity.

STRATOSPHERIC AEROSOL PROCESSES. Reactions involving sulfate aerosol are known to affect the amount of stratospheric O_3 . Only a few CCMs currently calculate the sulfur cycle and aerosol processes explicitly. Other CCMs include observed aerosol loadings and prescribed heating rates from major volcanic eruptions. In order to study the effects of these eruptions on stratospheric circulation and chemistry, the temperature response as well as changes in chlorine and nitrogen partitioning have to be examined.

SUMMARY AND THE WAY AHEAD. A table of core processes, diagnostics, and datasets for CCM validation has been developed, with a focus on the models’ ability to predict future stratospheric ozone amounts and distribution. Of the comprehensive suite of diagnostics for stratospheric CCMs listed in Table 2, several have been applied before to a range of models (Park et al. 1999; Pawson et al. 2000; Austin

et al. 2003), but most have not. Some models need further development before the diagnostics can be applied. Thus, while clearly desirable, it is a major task to perform all these diagnoses given the complexity of the CCMs. A step-wise approach is required in the use of the table. In practice, modeling groups need to develop their own priorities among these diagnostics. The choices will depend on the known strengths and weaknesses of each model, the processes and constituents already included, and the existing output from runs already performed. The choices will also depend on the scientific focus of each modeling group and the issue being addressed. For example, predictions of polar ozone loss will have more credibility if a model has been shown to compare well with diagnostics such as ozone loss versus PSC volume, heat flux, and ClO_x , NO_y , etc. Over time each model will gradually increase the number of tests applied and overall confidence will increase.

The lasting impact and the full benefit will come from concerted validation activities based on the table of processes. In order for these activities to succeed over the next several years, broad support is needed from the atmospheric sciences community and its managers. It is important that the validation procedures and goals defined for these activities are accepted at the start and valued by all participants in this joint exercise.

A new CCM Validation Activity for SPARC (CCMVal) has been established, based on experiences within GRIPS (Pawson et al. 2000) and on the concept that was developed in the workshop on process-oriented CCM validation (Eyring et al. 2004), so that real progress can be expected in the next couple of years in time for the next WMO/UNEP and IPCC assessments.

To facilitate this process-oriented validation of CCMs, we intend to provide all interested scientists with access to diagnostic software packages. These routines will be archived in a central location. The goal in supplying such software is to simplify such activities as quality control of model output, calculation of more complex model diagnostics, statistical evaluation of model/data differences and graphical display of results. Use of this software is not mandatory. Rather, the intent is to make it easier for groups to compute a broad range of calculations in a reasonably consistent way. Centralized software

repositories have been of great benefit in other MIPs, such as the AMIP and CMIP. These have freely supplied software for quality control of model output, data visualization, and interpolation of boundary condition datasets to a specific model grid. The CCM community can benefit from the experiences gained during previous model intercomparison exercises, particularly in terms of experimental design, definition of standard model output, and statistical aspects of model–data comparisons. Software developed in the course of previous MIPs, such as “performance portraits” and Taylor diagrams, provide useful means of summarizing many different aspects of climate model performance. In collaboration with groups such as the PCMDI, we intend to modify these diagnostic tools in order to suit the specific needs of the CCM community.

This suite of processes and diagnostics should become a benchmark for validation. Confidence in the performance of CCMs will increase as more model attributes become validated against the whole suite of diagnostics. Further, new models can be evaluated against an acknowledged, benchmark set of diagnostics as the models are developed. At the same time, the diagnostics themselves should develop as experience is gained and as new measurements become available allowing more processes to be diagnosed. It is hoped that this work has laid the groundwork for a more comprehensive approach to CCM validation, which will be further developed by all scientists who become involved. Updated information is available online at www.pa.op.dlr.de/CCMVal/, together with the names of people coordinating the various activities. All scientists interested in participating should contact the coordinating scientists.

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APPENDIX A: ACRONYMS AND ABBREVIATIONS

AMIP	Atmospheric Model Intercomparison Project	OCLI	Ozone Climate Interactions
AMTRAC	Atmospheric Model with Transport and Chemistry	O-GWD	Orographic gravity wave drag
CCM	Coupled chemistry–climate model	PCMDI	Program for Climate Model Diagnosis and Intercomparison
CCMVal	CCM Validation Activity for SPARC	PDF	Probability distribution function
CCSR	Center for Climate System Research	PSC	Polar stratospheric cloud
CFC	Chlorofluorocarbon	PV	Potential vorticity
CMAM	Canadian Middle Atmosphere Model	PW	Planetary wave
CMIP	Coupled Model Intercomparison Project	PWD	Planetary wave drag
CTM	Chemical transport model	QBO	Quasi-biennial oscillation
DLR	Deutsches Zentrum für Luft- und Raumfahrt (Germany)	SOCOL	Solar Climate Ozone Links
E39/C	ECHAM4.L39(DLR)/CHEM	SCIAMACHY	Scanning Imaging Absorption Spectrometer for Atmospheric Cartography
ECHAM	European Centre Hamburg Model	SSU	Stratospheric sounding unit
ECMWF	European Centre for Medium-Range Weather Forecasts (United Kingdom)	SPARC	Stratospheric Processes and their Role in Climate
ERA	ECMWF Re-Analysis	3D	Three-dimensional
EU	European Union	TEM	Transformed Eulerian mean
FUBCMAM	Freie Universität Berlin Climate Middle Atmosphere Model	TOMS	Total Ozone Mapping Spectrometer
GCCM	Global Tropospheric Climate–Chemistry Model	TTL	Tropical tropopause layer
GCM	General circulation model	ULAQ	University of L'Aquila
GEOS	Goddard Earth Observing System	UMETRAC	Unified Model with Eulerian Transport and Chemistry
GFDL	Geophysical Fluid Dynamics Laboratory (NOAA)	UMSLIMCAT	Unified Model SLIMCAT
GISS	Goddard Institute for Space Studies (NASA)	UMUCAM	Unified Model University of Cambridge
GOME	Global Ozone Monitoring Experiment	UNEP	United Nations Environment Programme
GRIPS	GCM-Reality Intercomparison Project for SPARC	UTLS	Upper troposphere/lower stratosphere
GSFC	Goddard Space Flight Center Model	UV	Ultraviolet
GWD	Gravity wave drag	$\nu T'$	Heat flux
HAMMONIA	Hamburg Model of the Neutral and Ionized Atmosphere	WACCM	Whole Atmosphere Community Climate Model
IPCC	Intergovernmental Panel on Climate Change	WCRP	World Climate Research Programme
J	Photolysis rate	WMO	World Meteorological Organization
K	Kelvin	WFA	Wavenumber–frequency analysis
LMDREPRO	Modele du Laboratoire de Meteorologie Dynamique-Chimie		
MAECHAM/	Middle Atmosphere European		
CHEM	Centre Hamburg Model with Chemistry		
MESSy	Modular Earth Submodel System		
MIP	Model Intercomparison Program		
MRI	Meteorological Research Institute		
MSU	Microwave sounding unit		
NASA	National Aeronautics and Space Administration (United States)		
NCAR	National Center for Atmospheric Research (United States)		
NCEP	National Centers for Environmental Prediction (NOAA)		
NIES	National Institute for Environmental Studies		
NLTE	Nonlocal thermodynamical equilibrium		
NonO-GWD	Nonorographic gravity wave drag		
NOAA	National Oceanic and Atmospheric Administration (United States)		

APPENDIX B: CHEMICAL FORMULAS

BrO_x	Bromine radicals
Br_y	Inorganic bromine
CFCs	Chlorofluorocarbons
CFC-11	CCl_3F
CH_4	Methane
ClO_x	Chlorine radicals
Cl_y	Inorganic chlorine
CO_2	Carbon dioxide
H	Atomic hydrogen
HCl	Hydrogen chloride (hydrochloric acid)
HNO_3	Nitric acid
HO_2	Hydroperoxyl radical
HO_x	Odd hydrogen (H , OH , HO_2 , H_2O_2)
H_2O	Water vapor
OH	Hydroxyl radical
NO	Nitric oxide
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides (NO + NO_2)
NO_y	Total reactive nitrogen (usually NO_x , NO_3 , N_2O_5 , ClONO_2 , HNO_4 , HNO_3)
N_2O	Nitrous oxide
O_x	Odd oxygen (O , $\text{O}(^1\text{D})$, O_3) or oxidant (O_3 + NO_2)
O_3	Ozone
SF_6	Sulfur hexafluoride

REFERENCES

- Anderson, J. L., and Coauthors, 2004: The New GFDL Global Atmosphere and Land Model AM2-LM2: Evaluation with prescribed SST simulations. *J. Climate*, **17**, 4641–4673.
- Austin, J., 2002: A three-dimensional coupled chemistry-climate model simulation of past stratospheric trends. *J. Atmos. Sci.*, **59**, 218–232.
- , and N. Butchart, 2003: Coupled chemistry-climate model simulation for the period 1980 to 2020: Ozone depletion and the start of ozone recovery. *Quart. J. Roy. Meteor. Soc.*, **129**, 3225–3249.
- , and Coauthors, 2003: Uncertainties and assessments of chemistry-climate models of the stratosphere. *Atmos. Chem. Phys.*, **3**, 1–27.
- Bais, A. F., and Coauthors, 2003: International Photolysis Frequency Measurement and Model Intercomparison (IPMMI): Spectral actinic solar flux measurements and modeling. *J. Geophys. Res.*, **108**, 8543, doi:10.1029/2002JD002891.
- Beagley, S. R., J. de Grandpré, J. N. Koshyk, N. A. McFarlane, and T. G. Shepherd, 1997: Radiative-dynamical climatology of the first-generation Canadian Middle Atmosphere Model. *Atmos.–Ocean*, **35**, 293–331.
- Braesicke, P., and J. A. Pyle, 2003: Changing ozone and changing circulation: Possible feedbacks? *Geophys. Res. Lett.*, **30**, 1059, doi:10.1029/2002GL015973.
- , and —, 2004: Sensitivity of dynamics and ozone to different representations of SSTs in the Unified Model. *Quart. J. Roy. Meteor. Soc.*, **130**, 2033–2046.
- Butchart, N., A. A. Scaife, J. Austin, S. H. E. Hare, and J. R. Knight, 2003: Quasi-biennial oscillation in ozone in a coupled chemistry-climate model. *J. Geophys. Res.*, **108**, 4486, doi:10.1029/2002JD003004.
- Chang, M. E., D. E. Hartley, C. Cardelino, and W.-L. Chang, 1996: Inverse modeling of biogenic isoprene emissions. *Geophys. Res. Lett.*, **23**, 3007–3010, doi:10.1029/96GL02370.
- Chipperfield, M. P., W. Feng, and M. Rex, 2005: Arctic ozone loss and climate sensitivity: Updated three-dimensional model study. *Geophys. Res. Lett.*, **32**, L11813, doi:10.1029/2005GL022674.
- Clark, H. L., A. Billingham, R. S. Harwood, and H. C. Pumphrey, 2003: Cirrus and water vapor in the tropical tropopause layer observed by Upper Atmosphere Research Satellite (UARS). *J. Geophys. Res.*, **108**, 4751, doi:10.1029/2003JD003748.
- Dameris, M., and Coauthors, 2005: Long-term changes and variability in a transient simulation with a chemistry-climate model employing realistic forcings. *Atmos. Chem. Phys. Discuss.*, **5**, 2297–2353.
- de Grandpré, J., S. R. Beagley, V. I. Fomichev, E. Griffioen, J. C. McConnell, A. S. Medvedev, and T. G. Shepherd, 2000: Ozone climatology using interactive chemistry: Results from the Canadian Middle Atmosphere Model. *J. Geophys. Res.*, **105**, 26 475–26 491.
- Dessler, A., D. Considine, J. Rosenfield, S. Kawa, A. Douglass, and J. Russell III, 1997: Lower stratospheric chlorine partitioning during the decay of the Mt. Pinatubo aerosol cloud. *Geophys. Res. Lett.*, **24**, 1623–1626.
- Egorova, T., E. Rozanov, V. Zubov, E. Manzini, W. Schmutz, and T. Peter, 2005: Chemistry-climate model SOCOL: A validation of the present-day climatology. *Atmos. Chem. Phys. Discuss.*, **5**, 509–555.
- Eluszkiewicz, J., and Coauthors, 1996: Residual circulation in the stratosphere and lower mesosphere as diagnosed from microwave limb sounder data. *J. Atmos. Sci.*, **53**, 217–240.
- Eyring, V., M. Dameris, V. Grewe, I. Langbein, and W. Kouker, 2003: Climatologies of subtropical mixing derived from 3D models. *Atmos. Chem. Phys.*, **3**, 1007–1021.
- , and Coauthors, 2004: Comprehensive summary on the Workshop on Process-Oriented Validation of Coupled Chemistry-Climate Models. *SPARC Newslett.*, **23**, 5–11.
- Fahey, D. W., and A. R. Ravishankara, 1999: Summer in the stratosphere. *Science*, **285**, 208–210.
- , and Coauthors, 1993: In situ measurements constraining the role of sulphate aerosols in mid-latitude ozone depletion. *Nature*, **363**, 509–514.
- , and Coauthors, 1996: In situ observations of NO_y, O₃, and the NO_y/O₃ ratio in the lower stratosphere. *Geophys. Res. Lett.*, **23**, 1653–1656.
- Forster, P. M. de F., M. Ponater, and W. Y. Zong, 2001: Testing broadband radiation schemes for their ability to calculate the radiative forcing and temperature response to stratospheric water vapour and ozone changes. *Meteor. Z.*, **10**, 387–393.
- Gao, R. S., and Coauthors, 2001: Observational evidence for the role of denitrification in Arctic stratospheric ozone loss. *Geophys. Res. Lett.*, **28**, 2879–2882.
- Giorgetta, M. A., and L. Bengtsson, 1999: Potential role of the quasi-biennial oscillation in the stratosphere-troposphere exchange as found in water vapor in general circulation model experiments. *J. Geophys. Res.*, **104**, 6003–6019.
- Hall, T. M., D. W. Waugh, K. A. Boering, and R. A. Plumb, 1999: Evaluation of transport in stratospheric models. *J. Geophys. Res.*, **104**, 18 815–18 839.
- Hayashi, Y., 1982: Space-time spectral analysis and its applications to atmospheric waves. *J. Meteor. Soc. Japan*, **60**, 156–171.

- Hofzumahaus, A., and Coauthors, 2004: Photolysis frequency of O₃ to O(¹D): Measurements and modeling during the International Photolysis Frequency Measurement and Modeling Intercomparison (IPMMI). *J. Geophys. Res.*, **109**, D08S90, doi:10.1029/2003JD004333.
- Hoor, P., H. Fischer, L. Lange, J. Lelieveld, and D. Brunner, 2002: Seasonal variations of a mixing layer in the lowermost stratosphere as identified by the CO-O₃ correlation from in situ measurements. *J. Geophys. Res.*, **107**, 4004, doi:10.1029/2000JD000289.
- Jöckel, P., R. Sander, and J. Lelieveld, 2004: Technical note: The Modular Earth Submodel System (MESSy)—A new approach towards Earth System Modeling. *Atmos. Chem. Phys.*, **5**, 433–444.
- Kawamoto, N., and M. Shiotani, 2000: Interannual variability of the vertical descent rate in the Antarctic polar vortex. *J. Geophys. Res.*, **105**, 11 935–11 946.
- Koch, G., H. Wernli, J. Staehelin, and T. Peter, 2003: A Lagrangian analysis of stratospheric ozone variability and long term trends above Payerne (Switzerland) during 1970–2001. *J. Geophys. Res.*, **108**, 4675, doi:10.1029/2003JD003911.
- Kylling, A., and Coauthors, 2003: Actinic flux determination from measurements of irradiance. *J. Geophys. Res.*, **108**, 4506, doi:10.1029/2002JD003236.
- Labitzke, K., and M. P. McCormick, 1992: Stratospheric temperature increases due to Pinatubo aerosols. *Geophys. Res. Lett.*, **19**, 207–210.
- Langematz, U., J. L. Grenfell, K. Matthes, P. Mieth, M. Kunze, B. Steil, and C. Brühl, 2005: Chemical effects in 11-year solar cycle simulations with the Freie Universität Berlin Climate Middle Atmosphere Model with online chemistry (FUB-CMAM-CHEM). *Geophys. Res. Lett.*, **32**, L13803, doi:10.1029/2005GL022686.
- Limpasuvan, V., D. W. J. Thompson, and D. L. Hartmann, 2004: On the life cycle of Northern Hemisphere stratospheric sudden warming. *J. Climate*, **17**, 2584–2596.
- Manney, G. L., K. Krueger, J. L. Sabutis, S. A. Sena, and S. Pawson, 2005: The remarkable 2003–2004 winter and other recent warm winters in the Arctic stratosphere since the late 1990s. *J. Geophys. Res.*, **110**, D04107, doi:10.1029/2004JD005367.
- Manzini, E., B. Steil, C. Brühl, M. Giorgetta, and K. Krüger, 2003: A new interactive chemistry climate model. 2: Sensitivity of the middle atmosphere to ozone depletion and increase in greenhouse gases: Implications for recent stratospheric cooling. *J. Geophys. Res.*, **108**, 4429, doi:10.1029/2002JD002977.
- Marcy, T. P., and Coauthors, 2004: Quantifying stratospheric ozone in the upper troposphere with in situ measurements of HCl. *Science*, **304**, doi: 10.1126/science.1093418.
- Millard, G. A., A. M. Lee, and J. A. Pyle, 2002: A model study of the connection between polar and midlatitude ozone loss in the Northern Hemisphere lower stratosphere. *J. Geophys. Res.*, **107**, 8323, doi:10.1029/2001JD000899.
- Mote, P. W., and Coauthors, 1996: An atmospheric tape recorder: The imprint of tropical tropopause temperatures on stratospheric water vapor. *J. Geophys. Res.*, **101**, doi:10.1029/95JD03422.
- Müller, R., P. J. Crutzen, J.-U. Groöf, C. Bruehl, J. M. Russell III, and A. F. Tuck, 1996: Chlorine activation and ozone depletion in the Arctic vortex: Observations by the Halogen Occultation Experiment on the Upper Atmosphere Research Satellite. *J. Geophys. Res.*, **101**, 12 531–12 554.
- Nagashima, T., M. Takahashi, M. Takigawa, and H. Akiyoshi, 2002: Future development of the ozone layer calculated by a general circulation model with fully interactive chemistry. *Geophys. Res. Lett.*, **29**, 1162, doi:10.1029/2001GL014026.
- Nash, E. R., P. A. Newman, J. E. Rosenfield, and M. R. Schoeberl, 1996: An objective determination of the polar vortex using Ertel's potential vorticity. *J. Geophys. Res.*, **101**, 9471–9478.
- Nedoluha, G., R. Bevilacqua, K. Hoppel, M. Daehler, E. Shettle, J. Hornstein, M. Fromm, J. Lumpe, and J. E. Rosenfield, 2000: POAM III measurements of dehydration in the Antarctic lower stratosphere. *Geophys. Res. Lett.*, **27**, 1683–1686.
- Newman, P. A., E. R. Nash, and J. E. Rosenfield, 2001: What controls the temperature of the Arctic stratosphere during spring? *J. Geophys. Res.*, **106**, 19 999–20 010.
- Oinas, V., A. Lacis, D. Rind, D. Shindell, and J. Hansen, 2001: Radiative cooling by stratospheric water vapor: Big differences in GCM results. *Geophys. Res. Lett.*, **28**, 2791–2794.
- Olsen, M. A., A. R. Douglass, and M. R. Schoeberl, 2002: Estimating downward cross-tropopause ozone flux using column ozone and potential vorticity. *J. Geophys. Res.*, **107**, 4636, doi:10.1029/2001JD002041.
- Park, J. H., M. K. W. Ko, C. H. Jackman, R. A. Plumb, J. A. Kaye, and K. H. Sage, 1999: Models and Measurements Intercomparison II. NASA Rep. NASA/TM-1999-209554, 502 pp.
- Park, M., W. J. Randel, D. E. Kinnison, R. R. Garcia, and W. Choi, 2004: Seasonal variation of methane, water vapor, and nitrogen oxides near the tropopause: Satellite observations and model simulations. *J. Geophys. Res.*, **109**, D03302, doi:10.1029/2003JD003706.

- Pawson, S., and Coauthors, 2000: The GCM-Reality Intercomparison Project for SPARC: Scientific issues and initial results. *Bull. Amer. Meteor. Soc.*, **81**, 781–796.
- Pierson, J. M., S. R. Kawa, R. J. Salawitch, T. F. Hanisco, E. J. Lanzendorf, K. K. Perkins, and R. S. Gao, 2000: Influence of air mass histories on radical species during the Photochemistry of Ozone Loss in the Arctic Region in Summer (POLARIS) mission. *J. Geophys. Res.*, **105**, 15 185–15 199.
- Pitari, G., E. Mancini, and D. Shindell, 2002: Feedback of future climate and sulfur emission changes on stratospheric aerosols and ozone. *J. Atmos. Sci.*, **59**, 414–440.
- Popp, P. J., and Coauthors, 2001: Severe and extensive denitrification in the 1999–2000 Arctic winter stratosphere. *Geophys. Res. Lett.*, **28**, 2875–2878.
- Randel, W. J., F. Wu, and R. Stolarski, 2002: Changes in column ozone correlated with the stratospheric EP flux. *J. Meteor. Soc. Japan*, **80**, 849–862.
- Read, W. G., D. L. Wu, J. W. Waters, and H. C. Pumphrey, 2004: Dehydration in the tropical tropopause layer: Implications from the UARS Microwave Limb Sounder. *J. Geophys. Res.*, **109**, D06110, doi:10.1029/2003JD004056.
- Rex, M., R. J. Salawitch, P. von der Gathen, N. R. P. Harris, M. Chipperfield, and B. Naujokat, 2004: Arctic ozone loss and climate change. *Geophys. Res. Lett.*, **31**, L04116, doi:10.1029/2003GL018844.
- Roeckner, E., and Coauthors, 2003: The atmospheric general circulation model ECHAM5, Part 1. MPI Rep. 349, 127 pp.
- Rosenlof, K. H., 1995: Seasonal cycle of the residual mean circulation in the stratosphere. *J. Geophys. Res.*, **100**, 5173–5191.
- Salawitch, R. J., and Coauthors, 1994: The distribution of hydrogen, nitrogen, and chlorine radicals in the lower stratosphere: Implications for changes in O_3 due to emission of NO_y from supersonic aircraft. *Geophys. Res. Lett.*, **21**, 2543–2546.
- , P. O. Wennberg, G. C. Toon, B. Sen, and J.-F. Blavier, 2002: Near IR photolysis of HO_2NO_2 : Implications for HO_x . *Geophys. Res. Lett.*, **29**, 1762, doi:10.1029/2002GL015006.
- Sander, R., A. Kerkweg, P. Jöckel, and J. Lelieveld, 2004: Technical Note: The new comprehensive atmospheric chemistry module MECCA. *Atmos. Chem. Phys.*, **5**, 445–450.
- Sankey, D., and T. G. Shepherd, 2003: Correlations of long-lived chemical species in a middle atmosphere general circulation model. *J. Geophys. Res.*, **108**, 4498, 10.1029/2002JD002799.
- Santee, M. L., A. Tabazadeh, G. L. Manney, M. D. Fromm, R. M. Bevilacqua, J. W. Waters, and E. J. Jensen, 2002: A Lagrangian approach to studying Arctic polar stratospheric clouds using UARS MLS HNO_3 and POAM II aerosol extinction measurements. *J. Geophys. Res.*, **107**, 4098, doi:10.1029/2000JD000227.
- Santer, B. D., and Coauthors, 2003a: Contributions of anthropogenic and natural forcing to recent tropopause height changes. *Science*, **301**, 479–483.
- , and Coauthors, 2003b: Influence of satellite data uncertainties on the detection of externally forced climate change. *Science*, **300**, 1280–1284.
- Sassi, F., B. A. Boville, D. Kinnison, and R. R. Garcia, 2005: The effects of interactive ozone chemistry on simulations of the middle atmosphere. *Geophys. Res. Lett.*, **32**, L07811, doi: 10.1029/2004GL022131.
- Sen, B., and Coauthors, 1999: The budget and partitioning of stratospheric chlorine during the 1997 Arctic summer, 1999. *J. Geophys. Res.*, **104**, 26 653–26 666.
- Shibata, K., and M. Deushi, 2005: Partitioning between resolved wave forcing and unresolved gravity wave forcing to the quasi-biennial oscillation as revealed with a coupled chemistry-climate model. *Geophys. Res. Lett.*, in press.
- , and Coauthors, 2005: Development of an MRI chemical transport model for the study of stratospheric chemistry. *Pap. Geophys. Meteor.*, **55**, 75–118.
- Shine, K. P., and Coauthors, 2003: A comparison of model-simulated trends in stratospheric temperatures. *Quart. J. Roy. Meteor. Soc.*, **129**, 1565–1588.
- Steil, B., C. Brühl, E. Manzini, P. J. Crutzen, J. Lelieveld, P. J. Rasch, E. Roeckner, and K. Krüger, 2003: A new interactive chemistry climate model. 1: Present day climatology and interannual variability of the middle atmosphere using the model and 9 years of HALOE/UARS data. *J. Geophys. Res.*, **108**, 4290, doi:10.1029/2002JD002971.
- Strahan, S. E., and A. R. Douglass, 2004: Evaluating the credibility of transport processes in simulations of ozone recovery using the Global Modeling Initiative three-dimensional model. *J. Geophys. Res.*, **109**, D05110, doi:10.1029/2003JD004238.
- Takigawa, M., M. Takahashi, and H. Akiyoshi, 1999: Simulation of ozone and other chemical species using a Center for Climate Systems Research/National Institute for Environmental Studies atmospheric GCM with coupled stratospheric chemistry. *J. Geophys. Res.*, **104**, 14 003–14 018.
- Thomas, A., and Coauthors, 2002: In situ measurements of background aerosol and subvisible cirrus in the tropical tropopause region. *J. Geophys. Res.*, **107**, 4763, doi:10.1029/2001JD001385.

- Thompson, A. M., and Coauthors, 2003: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology, Part 1: Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements. *J. Geophys. Res.*, **108**, 8238, doi:10.1029/2001JD000967.
- Tian, W., and M. P. Chipperfield, 2005: A new coupled chemistry–climate model for the stratosphere: The importance of coupling for future 03-climate predictions. *Quart. J. Roy. Meteor. Soc.*, **131**, 281–304.
- Wang, P.-H., P. Minnis, M. P. McCormick, G. S. Kent, and K. M. Skeens, 1996: A 6-year climatology of cloud occurrence frequency from Stratospheric Aerosol and Gas Experiment II observations (1985–1990). *J. Geophys. Res.*, **101**, 29 407–29 429.
- Waugh, D. W., 1996: Seasonal variation of isentropic transport out of the tropical stratosphere. *J. Geophys. Res.*, **101**, 4007–4023.
- , and W. J. Randel, 1999: Climatology of Arctic and Antarctic polar vortices using elliptical diagnostics. *J. Atmos. Sci.*, **56**, 1594–1613.
- , and T. M. Hall, 2002: Age of stratospheric air: Theory, observations, and models. *Rev. Geophys.*, **40**, 1010, doi:10.1029/2000RG000101.
- , and Coauthors, 1997: Mixing of polar vortex air into middle latitudes as revealed by tracer-tracer scatterplots. *J. Geophys. Res.*, **102**, 13 119–13 134.
- , W. J. Randel, S. Pawson, P. A. Newman, and E. R. Nash, 1999: Persistence of the lower stratospheric polar vortices. *J. Geophys. Res.*, **104**, 27 191–27 202, doi:10.1029/1999JD900795.
- Weber, M., S. Dhomse, F. Wittrock, A. Richter, B.-M. Sinnhuber, and J. P. Burrows, 2003: Dynamical control of NH and SH winter/spring total ozone from GOME observations in 1995–2002. *Geophys. Res. Lett.*, **30**, 1583, doi:10.1029/2002GL016799.
- WMO, 2003: Scientific assessment of ozone depletion: 2002. Global Ozone Research and Monitoring Project Rep. 47, 498 pp.
- Wong, S., W.-C. Wang, I. S. A. Isaksen, T. K. Berntsen, and J. K. Sundet, 2004: A global climate-chemistry model study of present-day tropospheric chemistry and radiative forcing from changes in tropospheric O₃ since the preindustrial period. *J. Geophys. Res.*, **109**, D11309, doi:10.1029/2003JD003998.
- Zhou, S. T., M. E. Gelman, A. J. Miller, and J. P. McCormack, 2000: An inter-hemispheric comparison of the persistent stratospheric polar vortex. *Geophys. Res. Lett.*, **27**, 1123–1126.