Popular Summary of
The Sensitivity of Arctic Ozone Loss to Polar Stratospheric Cloud Volume and Chlorine and Bromine Loading in a Chemistry and Transport Model

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Scientists use computer models to predict how the atmosphere will change over the next few decades. One of the things scientists worry about is how to find out if their predictions are believable. Scientists have been observing the atmosphere for many years with ground-based and satellite instruments, and studying the data to find patterns in the way the atmosphere behaves. If we can show that find the same behavior patterns in our models, we have confidence in the predictions from that model.

In the Arctic winter, thin clouds form when the lower stratosphere is very cold. The cloud particle surfaces speed up chemical reactions involving chlorine and bromine that destroy ozone. A group of researchers showed that the amount of ozone lost each year depends on the amount of clouds. More polar stratospheric clouds mean more ozone loss and fewer clouds mean less ozone loss. The amount of clouds varies from year-to-year. The chemical reactions that lead to ozone loss only happen if there are clouds, so this observation that relates the amount of ozone loss to the amount of clouds makes sense. The amount of ozone loss plotted versus the total volume of clouds forms a straight line, and our model produces a straight line with the same slope compared with data.

We know that the amount of clouds is not the only thing that can affect the amount of ozone loss in the model. For example, the amount of ozone loss calculated for a set polar stratospheric cloud volume will be too small if the model mixes middle latitude air into the polar vortex. We used new data from the MLS instrument on the Aura satellite to find patterns in another gas that tells us about mixing. Nitrous oxide (N_2O) has a long life-time and so the N_2O patterns tell us about the patterns of transport and mixing in the stratosphere. Very low concentrations of N_2O are measured inside the polar vortex, while much higher concentrations are measured outside. There is a sharp change that shows us the edge and tells us that the air does not mix between one side and the other. Our model

produces this pattern in N_2O, showing that our model keeps the air inside the polar vortex separated from the air outside.

Together these two comparisons tell us that this important part of our model behaves like the atmosphere. This increases confidence in the predictions of the future behavior of ozone in the Arctic stratosphere.

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Abstract

The sensitivity of Arctic ozone loss to polar stratospheric cloud volume (V_{PSC}) and chlorine and bromine loading is explored using chemistry and transport models (CTMs). A simulation using multi-decadal output from a general circulation model (GCM) in the Goddard Space Flight Center (GSFC) CTM complements one recycling a single year's GCM output in the Global Modeling Initiative (GMI) CTM. Winter polar ozone loss in the GSFC CTM depends on equivalent effective stratospheric chlorine (EESC) and polar vortex characteristics (temperatures, descent, isolation, polar stratospheric cloud amount). Polar ozone loss in the GMI CTM depends only on changes in EESC as the dynamics repeat annually. The GSFC CTM simulation reproduces a linear relationship between ozone loss and V_{PSC} derived from observations for 1992 – 2003 which holds for EESC within ~85% of its maximum (~1990 – 2020). The GMI simulation shows that ozone loss varies linearly with EESC for constant, high V_{PSC}.

1. Introduction

Chemistry climate models (CCMs) combine a general circulation model (GCM) with a model of physical and photochemical processes that affect atmospheric constituents. CCMs are self-consistent in that dynamics and constituents feedback through radiative processes and are used to predict future atmospheric climate and distributions of trace gases. *Eyring et al.* [2005] describe a process-oriented approach to CCM evaluation as key to obtaining believable predictions. *Rex et al.* [2004, hereinafter R2004] report a linear relationship between winter-spring loss of Arctic ozone and the volume of polar stratospheric clouds (PSCs). R2004 used data for 10 winters between 1992 and 2003, a period when inorganic chlorine in the upper stratosphere was close to its maximum. R2004 suggest this relationship as an element of CCM evaluation and speculate that

additional stratospheric cooling would lead to more PSCs and additional polar ozone loss. Chemistry and transport models (CTMs) are driven by input meteorological fields and ignore feedback processes, but should reproduce this relationship. R2004 show that the sensitivity of polar ozone loss to the volume of PSCs (V_{PSC}) in a version of the SLIMCAT CTM, driven by meteorological fields from the United Kingdom Meteorological Office UKMO, is less than that derived from observations. Recently, *Chipperfield et al.* [2005] show that a modified version of SLIMCAT, driven by meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF), reproduces the observed relationship.

Here we focus on simulations driven by meteorological output from a GCM. *Stolarski et al.* [2006] show that simulated mean total ozone for 60°S-60°N reproduces many aspects of TOMS observations. We show the realism of the simulated polar votex by comparing N_2O and its horizontal gradients with N_2O observed by the Microwave Limb Sounder (MLS) on NASA's Aura satellite [*Waters et al.*, 2006]. We show that the sensitivity of simulated winter chemical loss of ozone to V_{PSC} follows the R2004 relationship for 1990 – 2020, years when the equivalent effective stratospheric chlorine (EESC), i.e., chlorine and bromine available in the stratosphere to destroy ozone, is within 85% of its maximum. This result does not include modifications to the photochemical input and boundary conditions made to SLIMCAT. We also investigate the dependence of polar ozone loss on EESC for fixed V_{PSC}.

Simulations use the GSFC CTM and the Global Modeling Initiative (GMI) CTM [*Douglass et al.*, 2004], described in Section 2. Section 3 shows comparisons with N_2O to support the realism of the simulated polar vortex and verifies the method used to account for the ozone increase due to transport. Results are presented in section 4 followed by discussion and conclusions.

2. Models

The GSFC CTM

Stolarski et al. [2006] describe the GSFC CTM and the primary simulation used here; aspects important to this analysis follow. Rate constant data and cross sections are taken from Jet Propulsion Laboratory (JPL) Evaluation 14 [*JPL*, 2003]. The polar stratospheric cloud parameterization follows [*Considine et al.*, 2000] and accounts for denitrification through PSC sedimentation. Solar radiation for 1973-2003 is specified measurements from Solar Ultraviolet Spectral Irradiance Monitor (SUSIM) [*Brueckner et al.*, 1993] for 1992-2003 and ground-based measurements of 10.7 cm radio flux at Ottawa (F10.7). Later years repeat the solar cycle for 1992-2003. Aerosols are specified from a monthly mean climatology based on satellite data [*Jackman et al.*, 1996] and updated for *WMO* [2003]. Surface boundary conditions for source gases including CFCs, halons, methane and nitrous oxide are specified from Scenario A2 of the Scientific Assessment of Ozone Depletion: 2002 [*WMO*, 2003]. A simulation, identical except that boundary conditions for chlorine and bromine source gases are held fixed at background levels (~1960), is used to quantify the transport contribution to winter polar ozone change. The *Lin and Rood* [1996] scheme is used for constituent transport. The horizontal grid is 2.5°

longitude and 2° latitude. The 28 vertical levels between the surface and 0.4 hPa use a terrain following coordinate in the troposphere and pressure above the interface at 247 hPa. Vertical spacing is about 1 km near the tropopause and increases to 4 km near the upper boundary.

Meteorological fields are taken from a 50-year integration of the Goddard Earth Observing System (GEOS-4) GCM. GEOS-4 GCM uses a flux-form semi-Lagrangian transport code with a quasi-Lagrangian vertical coordinate [*Lin*, 2004; *Lin and Rood*, 1997]. Resolution is 2.5° longitude by 2° latitude with 55 layers between the surface and 0.01hPa. Physical tendencies are calculated with parameterizations from of *Kiehl et al.* [1998]. Drag resulting from the dissipation of a coarse spectrum of gravity waves with non-zero phase speeds is included, to improve the simulation of the upper stratosphere and mesosphere, using the method of *Garcia and Boville* [1994]. The lower-boundary sea-surface temperatures and sea-ice distributions were imposed from *Rayner et al.* [2003]. *Olsen et al.* [2004] show that exchange of mass and ozone between stratosphere and troposphere is realistic compared with observational estimates.

The GMI CTM

The GMI CTM uses essentially the same chemical mechanism, the same rate constant and cross section data [*JPL,* 2003], the same PSC parameterization, the same numerical transport and horizontal grid resolution as the GSFC CTM and is described elsewhere [*Rotman et al.* 2001; *Douglass et al.,* 2004]. There are 5 additional vertical levels with an upper boundary at 0.01 hPa. The GMI CTM uses SMVGEAR to solve the photochemical part of constituent continuity equations [*Jacobson,* 1995].

This simulation uses a single year's meteorological fields from the GCM described above. A year with a cold Arctic vortex was chosen to estimate the maximum possible impact of polar ozone loss. The GMI CTM produces realistic stratospheric transport and age-of-air using GEOS-4 GCM meteorological fields [*Strahan and Polansky,* 2005]. The distribution of polar stratospheric clouds is nearly identical each year for recycled meteorology, varying slightly due to small changes in nitric acid.

3. Analysis

The Arctic Polar Vortex

Simulation of winter polar ozone loss requires realistic simulation of temperatures, size and isolation of the Arctic winter vortex. V_{PSC} as used by R2004 is a proxy for temperature, thus the realistic range for simulated V_{PSC} compared with that derived from meteorological analyses shown in the next section implies realistic temperatures. We compare simulated N_2O with that observed by Aura MLS [*Froidevaux et al.,* 2006] to show credibility of vortex size and mixing barriers.

MLS Feb. 2005 N_2O, binned and averaged by equivalent latitude on potential temperature surfaces (Fig. 1a), is compared with the same average for simulated year 1983 (a year with a cold stable vortex) (Fig. 1b) and with the Feb. mean of 17 simulated years between 1990 and 2020 with cold stable vortices (Fig. 1c). All distributions are normalized by N_2O at 450K in the tropics. The simulation reproduces low values of high

latitude N_2O observed by MLS. Comparisons of tracer gradients are more revealing [*Sankey and Shepherd,* 2003]. The location and strength of MLS horizontal N_2O gradients for Jan. and Feb. (Figs. 1d and 1g) compare well for simulated year 1983 for Jan. and Feb. (Figs. 1e and 1h) and for the cold winter mean (Figs. 1f and 1i), showing that the vortex size and mixing barrier are credible. As observed, simulated February gradient maxima are stronger and equatorward of January maxima. The gradient of the mean distribution is weaker than the MLS gradient, due to interannual variability in vortex size. These comparisons show that, as observed, simulated N_2O descends from the upper stratosphere without significant horizontal mixing. Descent is necessary to obtain high values of Cly and Bry in the lower polar vortex. A strong barrier to mixing is necessary because mixing inhibits ozone loss both by decreasing Cly and Bry inside the vortex and by speeding conversion of chlorine radicals to reservoir species.

Ozone increase due to transport

High northern latitude ozone generally increases due to descent during winter. This seasonal change in ozone must be accounted for in determining the vortex-average polar ozone chemical loss between 380K and 600K. We quantify this contribution using the temporal changes in the vortex average profiles of N_2O (a conserved tracer) to quantify descent. We verify the results by comparing the ozone increase estimated from the change in N_2O with the increase obtained from the background chlorine simulation. The differences between the vortex averaged lower stratospheric ozone columns at 5-day intervals and columns on January 1 are plotted versus the estimated differences for each simulated year (Fig. 2). Scatter is similar for different time intervals, and errors do not accumulate over the winter. We estimate the transport contribution using N_2O for the remainder of this work.

Polar stratospheric cloud volume

R2004 estimate polar stratospheric cloud volume (V_{PSC}) using temperatures from ECMWF meteorological analyses and assumed profiles for HNO_3 and H_2O following *Hansen and Mauersberger* [1988]. In the CTM we use a similar procedure except with simulated profiles of H_2O and HNO_3. Averaged over all winters the CTM V_{PSC} exceeds that estimated by applying the method of R2004 to CTM temperatures by 30%. Without global observations of PSCs for winters analyzed in R2004 we cannot confirm the accuracy of the R2004 estimate. The difference between V_{PSC} from the CTM and V_{PSC} using CTM temperatures and fixed profiles of HNO_3 and H_2O following R2004 is an estimate of the uncertainty in the R2004 procedure. A few simulated winters are warm and no PSCs are formed; these years are not included when calculating the sensitivity of ozone loss to V_{PSC}.

4. Results

Ozone loss is shown as a function of V_{PSC} in Fig. 3. Center solid lines are the best fit for V_{PSC} between 5 and 40 x 10^6 km^3. The sensitivity of the ozone loss to V_{PSC} is 2.2±0.5 DU/$10^6$$km^3$ for V_{PSC} from the CTM (Fig. 3a) and 2.4±0.6 DU/$10^6$$km^3$ for V_{PSC} from CTM temperatures following R2004 (Fig. 3b). In both panels the fit from R2004 (dashed line) is nearly the same as the CTM best fit.

Figure 4 shows ozone loss from both CTMs vs. EESC; points are colored according to V_{PSC}. Squares are from the GSFC CTM and asterisks are from the GMI CTM. For EESC between 1.8 and ~ 3.6, and moderate values of V_{PSC}, the ozone loss varies linearly with EESC. The solid line in the stippled area (pre-1990 EESC) is the best fit between EESC and GSFC CTM. For warm winters, V_{PSC} ~ 0 (blue), the simulated winter ozone change is small or positive.

The GMI CTM sensitivity to EESC (dashed line) nearly parallels that of the GSFC CTM. The orange points (same values of V_{PSC}) from both simulations fall on or near the dashed line. EESC reaches 85% of its maximum in ~1990 and remains in that range until ~2020. For this EESC range ozone loss is sensitive to V_{PSC}. Winters with low V_{PSC} (blue) have little ozone loss, those with moderate V_{PSC} (green) have moderate ozone loss, and those with high V_{PSC} (red) have substantial ozone loss. The dashed line fit to GMI results shows that ozone loss varies linearly with EESC for the entire EESC range for fixed V_{PSC}.

5. Discussion and Conclusions

The GSFC simulation reproduces the slope of the empirical relationship between polar ozone loss and V_{PSC} described by R2004. *Chipperfield et al.* [2005] show that a modified version of SLIMCAT reproduces the observed empirical relationship. Modifications include improving the denitrification scheme, an updated calculation of diabatic heating, and addition of 100 pptv of chlorine and 6 pptv bromine to represent effects of very short-lived halocarbons. *Burkholder et al.* [1990] cross sections for photolysis of Cl_2O_2 are extrapolated to long wavelengths as suggested by *Stimpfle et al.* [2004], leading to more ozone destruction than using cross sections recommended by the JPL Evaluation 14 [*JPL,* 2003].

If the latter two modifications were implemented in the GSFC CTM, its sensitivity of winter ozone loss to V_{PSC} would exceed that derived from observations. The obvious question is why the SLIMCAT CTM and the GSFC CTM differ when the input photochemical data and boundary conditions are the same. The most important difference is that SLIMCAT uses meteorological fields from an assimilation system and the GSFC and GMI CTMs use meteorological fields from a GCM. Analysis has shown that GSFC CTM driven by meteorological fields from various versions of the GEOS Data Assimilation System exhibits excess horizontal mixing [e.g., *Considine et al.*, 2003]. *Schoeberl et al.* [2004] use trajectory calculations to show that use of the diabatic heating for vertical motion does not improve the horizontal transport using meteorological fields from GEOS DAS or from UKMO. Simulated winter polar ozone will be less sensitive to V_{PSC} if there is excess mixing of middle latitude air into the polar vortex, as such mixing reduces the active chlorine and bromine and also speeds re-formation of chlorine reservoirs. We suggest that the main difference between the GSFC CTM and SLIMCAT is isolation of the polar vortex produced by the input meteorological fields. Physical interpretation of agreement (or disagreement) between simulation and observations is

more possible if the R2004 diagnostic is applied and interpreted with diagnostic of transport barriers such as discussed by *Sankey and Shepherd* [2003]

Sensitivity of ozone loss to V_{PSC} depends on several factors, including descent, temperature, vortex isolation and photochemistry. A simulation may produce the wrong sensitivity if any of these are in error, or may produce the correct sensitivity in the case of compensating errors. Comparisons of simulations with observations provide insight into the realism of the photochemical mechanism, but do not support changes to photochemical input data unless aspects of simulation performance such as vortex isolation are also evaluated. Although the increase to the *JPL* [2003] recommendation for Cl_2O_2 cross sections used by *Chipperfield et al.* [2005] is supported by the work of *Stimpfle et al.* [2004], *Pope et al.* [2005] report measurements of the Cl_2O_2 cross-sections that are significantly smaller than the current JPL evaluation. It is prudent that controversy concerning recommended cross sections for atmospheric photolysis of Cl_2O_2 be resolved through analysis of laboratory results.

Finally, this analysis may lead to an improved statistical model for quantifying ozone trends. Statistical analysis derives ozone sensitivity to chlorine by simultaneously accounting for other factors (seasonal, solar, quasi-biennial oscillation, volcanic aerosols) known to contribute to observed ozone variability [*Stolarski et al.,* 2006]. R2004 and this analysis show that winter polar vortex temperature modulate the sensitivity of high northern latitude ozone to chlorine for high EESC. This effect is not accounted for in present statistical models.

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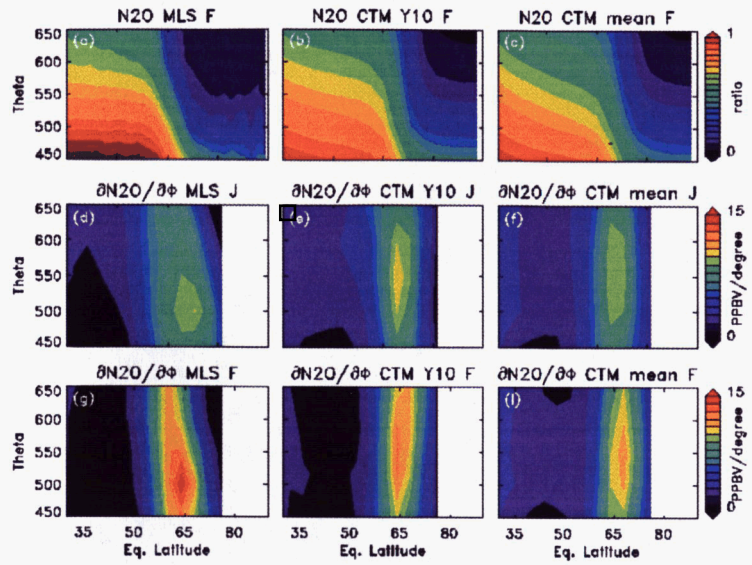

Figure 1 (Top) Feb. mean N_2O: (a) MLS (b) GSFC CTM 1983) (c) GSFC CTM multiyear mean. (Middle) Jan. horizontal N_2O gradients (d) MLS; (e) GSFC CTM 1983 (f) CTM multiyear mean. (Bottom) Feb. horizontal N_2O gradients (g) MLS; (h) GSFC CTM 1983; (i) GSFC CTM multiyear mean.

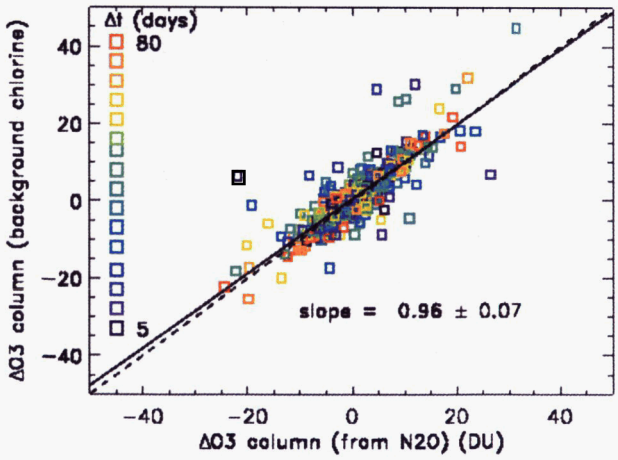

Figure 2 The increase in ozone column due to descent from N_2O vs. that from the background chlorine simulation.

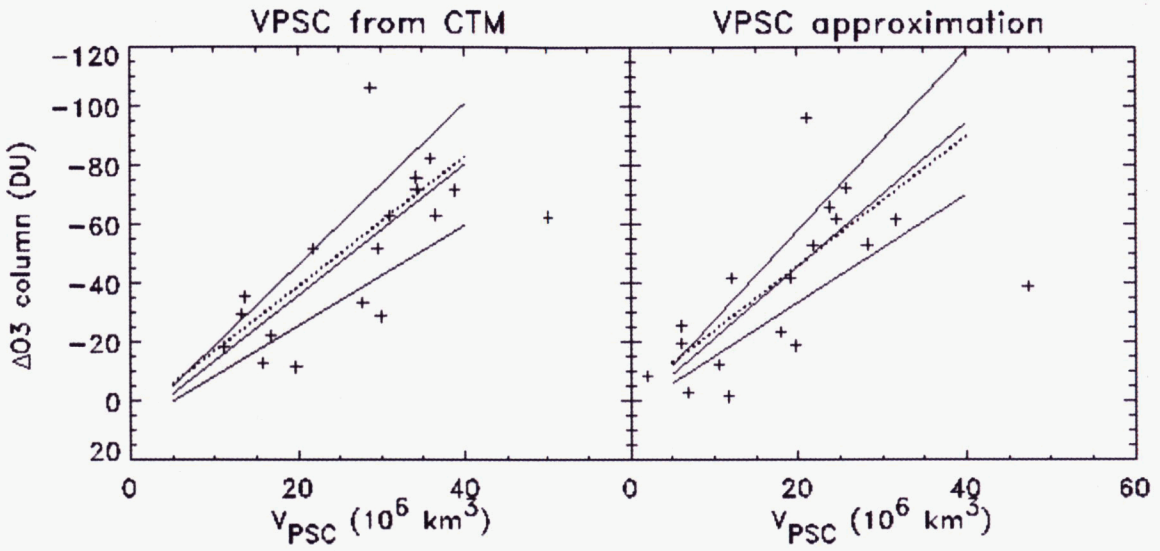

Figure 3 a) CTM vortex averaged chemical ozone loss vs CTM V_{PSC}; b) same as (a) except V_{PSC} calculated following R2004. The central lines are best fit for V_{PSC} greater than 5; the outer lines include 1-sigma slope errors.

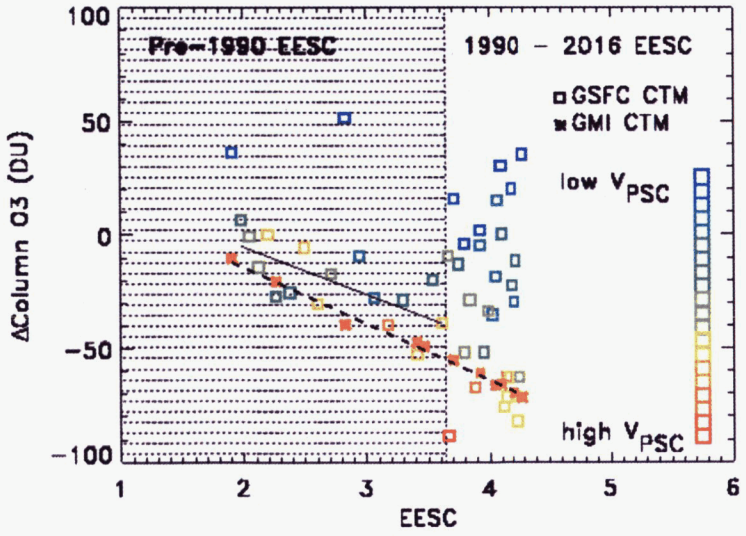

Figure 4 Ozone loss vs. EESC for the GSFC CTM (squares) and for the GMI CTM (asterisks). Colors show V_{PSC} (V_{PSC} ~ 0 blue ; VPSC ~ 60 red). The solid line is the best fit for GSFC CTM results for pre-1990 EESC. The dashed line is the best fit for the GMI CTM results.