

N95-11019

Calculations of Arctic Ozone Chemistry Using Objectively Analyzed Data in a 3-D CTM

by

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Abstract

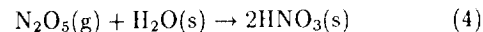
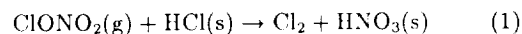
A three-dimensional chemical transport model (CTM) (Kamiński, 1992) has been used to study the evolution of the Arctic ozone during the winter of 1992. The continuity equation has been solved using a spectral method with Rhomboidal 15 (R15) truncation and leap-frog time stepping. Six-hourly meteorological fields from the Canadian Meteorological Centre global objective analysis routines run at T79 were degraded to the model resolution. In addition, they were interpolated to the model time grid and were used to drive the model from the surface to 10 mb. In the model, processing of Cl_x occurred over Arctic latitudes but some of the initial products were still present by mid-January. Also, the large amounts of ClO formed in the model in early January were converted to ClONO₂. The results suggest that the model resolution may be insufficient to resolve the details of the Arctic transport during this time period. In particular, the wind field does not move the ClO_x 'cloud' to the south over Europe as seen in the MLS measurements.

Introduction

Although the dynamical situation during the Arctic winter is less stable than that in Antarctica and the temperatures are not as cold, Polar Stratospheric Clouds (PSC) type I and type II PSCs do form (eg. GRL, 1990). Also, air remains in the vortex sufficiently long that there may be extensive processing (eg., McConnell et al., 1991) so that Cl_x may be converted to active forms of chlorine within extensive regions of the polar vortex. This has clearly been observed during the Arctic Airborne Stratospheric Expedition (AASE) during the winter of 1989/90 where enhanced levels of BrO and ClO were observed along with some denitrification (GRL, 1990). Further, the microwave limb sounder (MLS) instrument on board the UARS satellite has observed extensive 'clouds' of ClO over northern latitudes and as far south as the Mediterranean (Waters et al., 1992).

In this note we present the results of an attempt to simulate the dynamics and chemistry of Arctic stratosphere, and in particular the formation of the ClO 'cloud' observed over Northern Europe during the 1991/1992 winter (eg. Waters et al., 1992). The York Chemical Transport Model (CTM) (Kamiński, 1992) was run in an off-line mode from December 15th to January 22nd. The CTM was driven by the objectively analyzed wind and temperature data generated by the Canadian Meteorological Centre (CMC) global spectral model (Ritchie, 1991) run in the Analysis Cycle (zero hour forecast at 00Z, 06Z, 12Z, and 18Z was used). The CMC's global objective analysis (OA) stops at 10 mb. The upper level temperature fields are inverted from satellite observations (TOVS), and the wind fields are derived from temperature.

The chemistry in the model consists of 49 reactions (which includes gas phase, heterogeneous and photochemical reactions) and 21 species. The following heterogeneous reactions were included



and were activated by temperature switches. For $T < 195$ K the reactions (1) and (2) were activated, while for $T < 190$ K the reactions (3) and (4) were added. Although this does not simulate the details of the effects of processing by type I and II PSCs it should produce the correct general conversion from inactive forms of Cl_x to active forms, ClO_x. We define $\text{Cl}_x = \text{ClO}_x + \text{HCl} + \text{ClONO}_2$ and $\text{ClO}_x = \text{Cl} + 2\text{Cl}_2 + \text{ClO} + \text{HOCl} + \text{ClONO}_2 + 2\text{Cl}_2\text{O}_2$.

Seven tracers are used: O₃, Cl_x, NO₂, CH₄, N₂O, H₂O and passive-O₃. The passive tracer has the mixing ratios of the initial O₃ field and provides a measure of the effects of transport without chemistry and is thus particularly useful for comparing with the chemically active O₃ field. In addition, it provides a useful check for the mass

conservation properties of the code. For this experiment a half-hour dynamical time step (required by the CFL condition) and a 1.5 hour chemical time step are used to calculate the species concentrations. The chemical time step of 1.5 hours was selected in order to adequately simulate the diurnal behaviour of the Arctic chemistry. The chemistry solver is stable and previous experiments have indicated that much longer time steps could be used. The J values are evaluated by means of a look-up table at each chemical time step. A two-dimensional look-up table is generated using combinations of the logarithm of O_2 and O_3 optical thickness, and temperature, depending on the J value. Currently we allow the J value look-up tables to have a maximum error of about 5% as compared with those generated by a detailed wavelength integration method (cf. Henderson et al., 1987).

The CTM employs a spectral solution to the continuity equation (eg. Williamson et al., 1987) and was run with Rhomboidal 15 (R15) truncation, corresponding to 48 longitudes by 40 latitudes in grid space. A leapfrog scheme in spectral space, and a finite difference scheme in the vertical are used to integrate the continuity equation. The initial conditions, representative for mid-December, were generated by a two-dimensional zonal average model of the middle atmosphere that was supplied by Dr G. Brasseur of NCAR (Brasseur et al. 1990). The model was run from 15th December, 1991 to 22nd January, 1992. Since this is a short note and the results are extensive we have chosen to concentrate on the output for a single day and a single pressure level, viz. the 30 mb surface on the 16th January, 1992.

Analysis and Conclusions

It is quite clear from a preliminary comparison of the model O_3 data with the TOMS O_3 that the dynamical field rapidly imposes its main features in a relatively short period. A comparison between the model active and passive ozone and the TOMS ozone field is shown in Figure 1 for the 16th January, 1992. It can be seen that many of the features exhibited by the TOMS ozone are reproduced by the model. Noteworthy are the highs over eastern North America and eastern Asia and the low over the eastern Atlantic. There are several locations where the detail is not good. However, considering the limited resolution of the model and the fact that it presents a snapshot whereas the TOMS data represent the ozone field gathered over a day we consider the agreement quite reasonable. Since the effects of chemistry in the polar regions are marginal over this short time period what we are seeing in the model is largely the effects of transport. This is also seen in a comparison of the active and passive ozone fields in Figure 1. At low latitudes the difference between the passive and active ozone is due to the effect of substantial ozone generation, as expected, due to

photolysis of O_2 . The effect of ozone depletion at boreal latitudes is not immediately evident and still requires further quantification. We note that the model ozone highs are within 5% of the TOMS data.

At the beginning of the simulation the temperature is sufficiently cold to form PSC type I and the heterogeneous reactions listed above are activated. During the second week of January, 1992 the boreal temperatures are warmer than 195 K and there is no further formation of active chlorine. Figure 2 shows time series of the percent volume of air within each layer that has (a) temperatures less than 190 K, (b) temperatures less than 195 K and (c) temperatures less than 200 K for latitudes greater than 50° N. It shows quite dramatically the evolution of the processing in the model. The 200 K temperature was included because the CMC temperatures in this region of the stratosphere for this period of the forecast were probably up to 5 degrees too warm (Dr H. Ritchie, private communication, 1992; this problem with the stratospheric temperatures has now been corrected).

Although the processing has finished by the 16th of January it takes several weeks for the processed air to be reconverted back to HCl and $ClNO_3$ (eg. Henderson et al., 1990). This is indicated in Figure 3 which shows the fields for HCl, ClO_x , and $ClNO_3$ on the 30 mb level for 00Z on the 16th January, 1992. Figure 3a shows that a large region of HCl has been completely processed to active chlorine and has not been reformed. At the time of the snapshot the hole in the HCl field is over Greenland and northern Europe. Other regions with low values are due to transport effects which are immediately apparent in the Cl_x field (not shown). Furthermore, we note that there as almost no PCS type II activated in the model.

The active chlorine field, ClO_x , is shown in Figure 3b. It is similar in shape to the processed HCl hole, but somewhat smaller and this is significant as illustrated later. The peak ClO_x mixing ratio is 3.6 ppb. The ClO_x is composed mainly of $ClNO_2$, HOCl and Cl_2 with peak mixing ratios ~ 2.2 ppb (over Greenland), ~ 0.45 ppb over Scandinavia and ~ 0.6 ppb over Greenland, respectively. Cl_2O_2 and ClO are relatively small components of the ClO_x field. The fact that $ClNO_2$, HOCl and Cl_2 predominate, indicates that the air in this region has not seen the sunlight otherwise it would have been processed into ClO and Cl_2O_2 . Given that the MLS measurements of Waters et al. (1992) show several ppb of ClO, our results indicate that there may be serious problems with the dynamical field run at R15.

The missing Cl_x between the HCl hole and the ClO_x 'cloud' is found as $ClNO_3$ and is shown in Figure 3c. It appears as a collar of $ClNO_3$ around the ClO_x 'cloud' with the highest values ~ 3 ppb occurring over northern Russia and with mixing ratios ~ 1 ppb around the collar. What appears to have occurred is that HCl has been processed and the products photolyzed to form ClO. This has then reacted with the NO_2 present in the model to form $ClNO_3$.

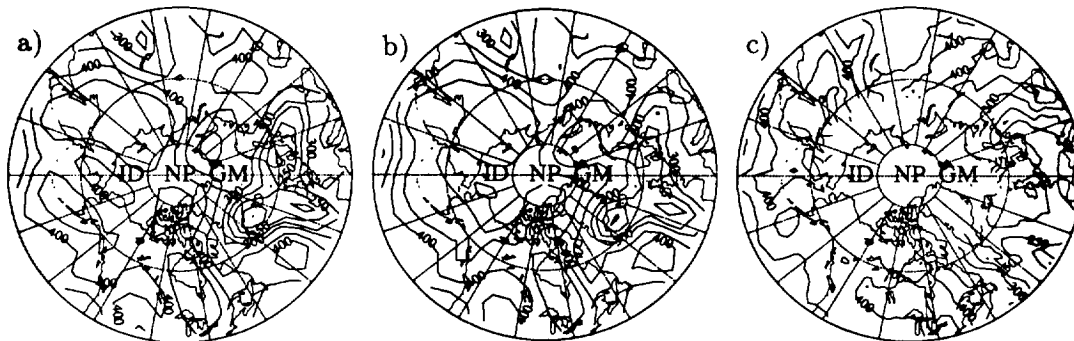


Figure 1. a) Active, b) passive and c) TOMS column ozone for the 16th January, 1992

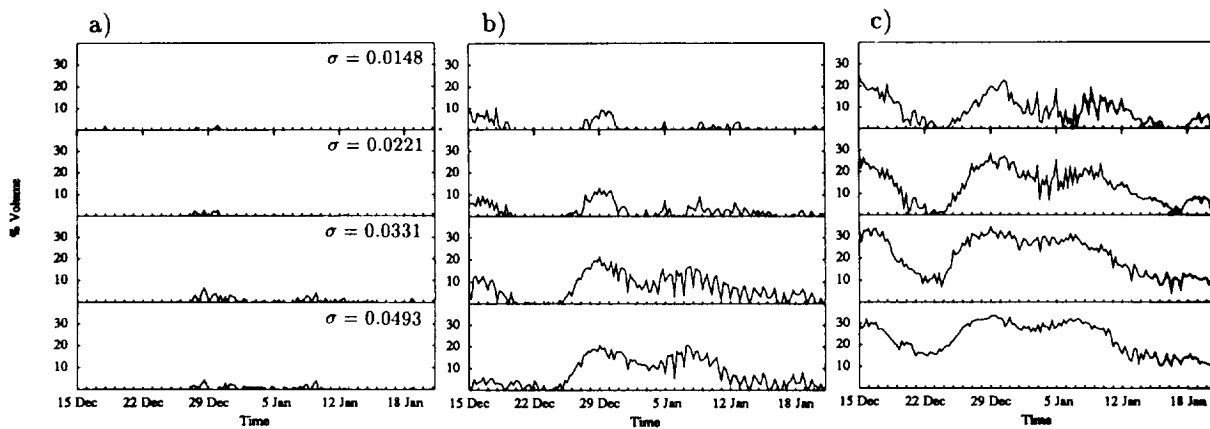


Figure 2. Time series showing the percent volume of air that has a) temperatures less than 190 K, b) temperatures less than 195 K and c) temperatures less than 200 K for latitudes greater than 50° N (time series are shown for 4 model levels).

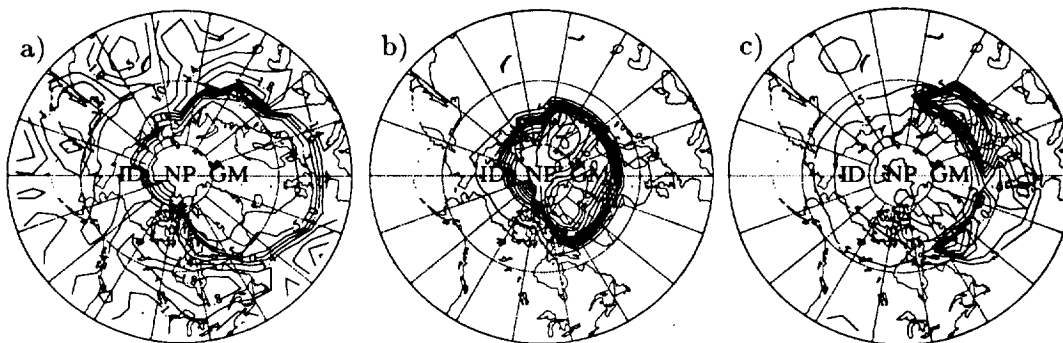


Figure 3. a) The HCl mixing ratios on the 30 mb surface for 00Z on the 16th January, 1992 using the CTM showing the region of reduced mixing ratios due to processing simulated by the model; b) ClO_x c) ClNO_3 .

We note that we have not included any processing of N_2O_5 in the model to HNO_3 by stratospheric aerosols which would occur at all temperatures. This would remove any N_2O_5 formed in the model and so the NO_x . We find that N_2O_5 mixing ratios are $\sim 1\text{--}3$ ppb in the polar region ($> 50^\circ \text{N}$) and NO_x values are ≤ 2.5 ppb. However, this would be adequate to convert any free ClO to ClNO_3 on a time scale of a day. The edge of the ClNO_3 collar reaches as far south as northern France. This is about 1 model grid point further north than measured by the MLS instrument for earlier dates. Again, we suspect that this is due to the limitations of the R15 simulation and possibly poor representation of the N_2O_5 as mentioned above.

Acknowledgments

This work was supported by the Atmospheric Environment Service of Environment Canada and by the Natural Science and Engineering Research Council of Canada. We wish to thank Dr G. Brasseur for the use of his 2-D model output used in the development of the initial conditions. Dr H. Ritchie of RPN in Dorval was very helpful in discussing the use of the CMC spectral forecast model.

References

Brasseur, G., M.H. Hitchman, S. Walters, M. Dymek, E. Falise, and M. Pirre, An interactive chemical dynamical radiative two-dimensional model of the middle atmosphere. *J. Geophys. Res.*, **95**, 5639–5655, 1990.

G.R.L., *Geophys. Res. Lett.*, **17**, 313–564, special issue, 1990.

Henderson G.S., J.C. McConnell, and W.F.J. Evans, The effects of initial active chlorine concentrations on the Antarctic ozone spring depletion, *J. Geophys. Res.*, **95**, 1899–1908, 1990.

Henderson, G.S., W.F.J. Evans, J.C. McConnell and E.M.J. Templeton, 1987, A numerical model for one dimensional simulation of stratospheric chemistry, *Atmos. Oceans*, **25**, 427–459.

Kamiński, J.W., Study of Stratospheric Chemistry Using a Three-Dimensional Spectral Chemical Transport Model, Ph D. Thesis, York University to be submitted, 1992.

McConnell, J.C., W.F.J. Evans, and E.M.J. Templeton, Model simulation of chemical depletion of Arctic ozone during the winter of 1989, *J. Geophys. Res.*, **96**, 10923–10930, 1991.

Ritchie, H., Application of the semi-Lagrangian method to a multilevel spectral primitive-equations model, *Q. J. R. Meteorol. Soc.*, **117**, 91–106, 1991.

Waters, J.W., L. Froidevaux, W.G. Read, L.S. Elson, E.F. Fishbein, T. Lungu, D.A. Flower, R.F. Jarnot, Stratospheric ClO and O_3 measured by the Upper Atmosphere Research Satellite (UARS) Microwave Limb Sounder (MLS) experiment. Quardennial Ozone Symposium, 1992.

Williamson, D.L., J.T. Kiehl, V. Ramanathan, R.E. Dickinson, J.J. Hack, Description of NCAR Community Climate Model (CCM1), NCAR/TN-285+STR, Boulder, Colorado, USA, 1987.