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The 1997 Arctic ozone depletion quantified from three-dimensional model simulations

Franck Lefèvre, François Figarol

Centre National de Recherches Météorologiques, Météo-France/CNRS, Toulouse, France

Kenneth S. Carslaw, Thomas Peter

Max-Planck Institut für Chemie, Mainz, Germany

Abstract. Three-dimensional simulations of total ozone are reported for the 1996-97 Arctic winter. The record low ozone values observed by satellite in late March are well reproduced by the chemistry-transport model. The comparison between the chemically integrated ozone and a passive tracer with identical initialization allows us to discriminate chemical changes from variations due to dynamical processes. In addition to a substantial total ozone chemical loss (60 to 120 Dobson Units), the simulation reveals an dynamically-induced reduction of ~ 70 DU also responsible for the ozone minimum observed in the Arctic in late March 1997.

Introduction

Observations by the Total Ozone Mapping Spectrometer (TOMS) satellite instrument in spring 1997 revealed anomalously low ozone columns over the Arctic [Newman *et al.*, 1997], with reductions of about 30-40% compared to the early 1980s climatology. This suggests that significant chemical ozone destruction occurred during the 1997 Arctic spring. However, chemical losses are difficult to quantify from total ozone data alone. In winter and spring, planetary wave activity and synoptic weather systems lead to considerable day-to-day variations of the ozone column, which usually outweigh the effects of chemistry. Interpretation is complicated further by diabatic descent which tends to increase the ozone column in the polar vortex, and this may vary substantially from year to year. We use a three-dimensional Chemistry-Transport Model (CTM) to simulate the evolution of the total ozone field during the 1996-97 winter and to map the accumulated total ozone chemical loss over the entire northern hemisphere. In recent years, CTMs constrained by meteorological analysis have successfully reproduced the ozone column variability measured by TOMS [Douglass *et al.*, 1996]. They have also proved to be powerful tools to discriminate chemical change from large variations due to dynamical processes [Chipperfield *et al.*, 1996; Goutail *et al.*, 1998a]. This approach is used here for the interpretation of the record low ozone columns measured in March 1997.

Model Experiment

The Reprobus CTM is used in this study [Lefèvre *et al.*, 1994]. The chemical package calculates the densities of 55

species by means of a comprehensive set of 102 gas phase reactions and 36 photodissociation rates [DeMore *et al.*, 1997]. Among them, 40 chemical families or individual constituents are explicitly transported by the semi-lagrangian scheme. A new heterogeneous chemistry routine was implemented for the present study. This is a "liquid only" scheme which assumes that polar stratospheric cloud particles remain in liquid phase down to the ice frost point. An analytical expression [Carslaw *et al.*, 1995] is used to calculate the equilibrium composition and volume of the $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ binary and $\text{HNO}_3\text{-H}_2\text{SO}_4\text{-H}_2\text{O}$ ternary droplets as a function of temperature and the total amounts of H_2O , HNO_3 , and H_2SO_4 . The routine also computes the aqueous phase concentrations for the soluble species HCl, HBr, HOCl, and HOBr, to calculate the rates of the most important heterogeneous reactions on stratospheric liquid aerosols. Background concentrations of H_2SO_4 were used, leading to aerosol surface area smaller than $1 \mu\text{m}^2\text{cm}^{-3}$ above 200K.

Reprobus was coupled to the temperature and wind fields analysed every 6 hours by ECMWF, and was integrated from December 18, 1996 to April 4, 1997. The model has 31 levels from the surface up to 10 hPa, and a resolution of 2 degrees latitude by 2 degrees longitude. The ozone field was initialized in the 100-10 hPa pressure range from the MLS Version 4 O_3 data [Froidevaux *et al.*, 1996; Manney *et al.*, 1996]. Ozone at lower levels and other gases were initialized from a 5-year run of a version of Reprobus coupled to Météo-France's Arpège general circulation model. For all results presented here the contribution of upper stratospheric O_3 to the total column was calculated assuming an O_3 mixing ratio identical to the one calculated at 10 hPa over a scale height of 6 km. This amount ranges typically from ~ 50 DU in the tropics to ~ 30 DU at high latitudes.

Comparisons With Observations

The model output is compared with the observations of the TOMS instrument on the NASDA (Japan) ADEOS spacecraft. Because at middle latitudes chemical processes are not expected to play a significant role in the observed ozone column variability on this timescale, the comparison at Haute-Provence (44 N, 6 E) provides a measure of the accuracy of the transport in the model (Figure 1A). The model reproduces realistically over the 3 1/2 month simulation the short timescale changes in the observations, which result from dynamical events. In contrast to Haute-Provence, the Arctic station of Ny Ålesund (79 N, 12 E) was usually located well inside the polar vortex during the 1996-97 winter. The observed behavior of the ozone column at this location

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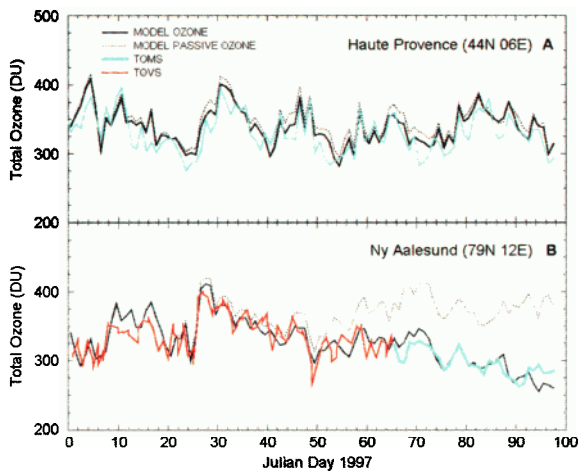


Figure 1. Time series of total ozone (Dobson Units) at (A) Haute-Provence and (B) Ny Ålesund from January 1, 1997, to April 7, 1997. Daily model calculations are compared to ADEOS-TOMS observations when available, complemented by TOVS nighttime measurements [Lefèvre *et al.*, 1991] at Ny Ålesund before day 65. Also shown is the evolution of a passive tracer with the same initialization as ozone.

is therefore the result of a combination of transport processes and rapid photochemical loss induced by enhanced active chlorine and bromine concentrations. Observations and model show an apparent decrease in ozone column from about 400 DU in late January to 280 DU in late March (Figure 1B). The good overall agreement between satellite data and Reprobus in January and early February, when the polar night prevents large O_3 loss, gives a strong indication that transport processes at all scales, including diabatic descent in the polar vortex, are realistically described by the model. A comparison of observed and calculated long-lived tracers in February 1997 [A. Engel, personal communication] also showed that diabatic descent is well reproduced by the model. The total ozone chemical loss can therefore be quantified using a passive tracer, initialized with concentrations identical to ozone, but transported without any chemical production or loss. The difference between the "passive ozone" column and the true ozone column allows to eliminate the synoptic variability as well as the effect of diabatic descent in polar regions, to give the accumulated chemical loss since the beginning of the simulation. At Ny Ålesund, the difference between the chemical and passive ozone columns diverges rapidly after day 50 in mid-February. This reflects the increase in available sunlight which drives the ozone-destroying catalytic cycles. At the end of March the passive ozone tracer indicates column amounts of about 375 DU, which would have been the total ozone content at Ny Ålesund in absence of chemistry. Compared to the ~ 275 DU obtained from the chemically integrated ozone, the accumulated ozone loss is therefore of ~ 100 DU.

Figure 2 (top and middle rows) compares a series of ADEOS-TOMS global observations to snapshots of Reprobus model calculations at 2-week intervals from February 12, 1997 to March 26, 1997. Throughout the period the synoptic features observable in the TOMS total ozone fields correspond closely with similar features in the calculated fields.

At middle latitudes maxima and minima in the observed and calculated fields, which result from dynamical events, are collocated with a one-to-one degree of correspondence. At high latitudes, the Reprobus simulation captures the observed steady decline of the total ozone content. On March 26 both the amplitude and the almost circular morphology of the simulated ozone minimum centered on the North Pole compare well with TOMS data. The minimum values given by TOMS and the model on March 26 are both lower than 230 DU. At middle latitudes, numerical diffusion of ozone-poor air through the vortex edge leads however to an underestimation of about 25 DU (or 5%) in the ozone maxima calculated by the model at the end of the simulation. Uncertainties in the estimation of the ozone column above 10 hPa in the tropics also cause an amplification of the slow increase observed by TOMS in this region from early February to late March.

Ozone Loss

We now use the difference between the chemically integrated and passive ozone columns to map out the spatial extent of the total ozone chemical loss (Figure 2, bottom row). The gradual increase in chemical loss is evident at high latitudes, while chemical production dominates in the tropics. A moderate ozone column destruction (10–25 DU) occurred by February 12. Although significant chlorine activation is calculated by the model on that day, the circular shape of the vortex and its centered position over the Pole did not favor long exposures to sunlight, limiting ozone chemical loss in early winter. More significant are the losses calculated on February 26, with values ranging from 30 to 50 DU at high latitudes. The very low temperatures observed during the second half of February led to an almost complete chlorine activation throughout the polar stratosphere in the model: inspection of model output on the 465K isentropic level (not shown) indicates ClO mixing ratios larger than 1.7 ppbv on February 26, in rather good agreement with MLS observations [Santee *et al.*, 1997]. In contrast to the February calculations, the total ozone loss maps for March show largest destructions more towards the vortex interior. Maximum losses reach 80 DU on March 12 and 120 DU on March 26. A large variability in the reduction is found in the vortex region, with a factor of two between minimum and maximum losses north of 70°N. This inhomogeneity is consistent with the findings of Müller *et al.* [1997b], but the maximum losses calculated from our analysis appear to be larger than the 60–70 DU maximum losses derived from the HALOE instrument. Two reasons partly explain this difference. First, as shown from the MLS ozone measurements [Manney *et al.*, 1997], the vertical extent of the O_3 decrease in 1997 was larger than in previous years and extended as high as 650 K, i.e. above the 350 K–550 K domain used to compute the total ozone loss from HALOE. At high latitudes our model simulation confirms that a 10–18 DU cumulative destruction occurred above 550 K. Second, due to the unusual position and morphology of the 1997 polar vortex many of the HALOE observations were not performed in the center of the vortex, where the strongest ozone loss is predicted by the model. When using the same vertical and horizontal sampling as Müller *et al.* [1997b], our simulation indicates a large variability inside the vortex, with maximum losses between 80–90 DU.

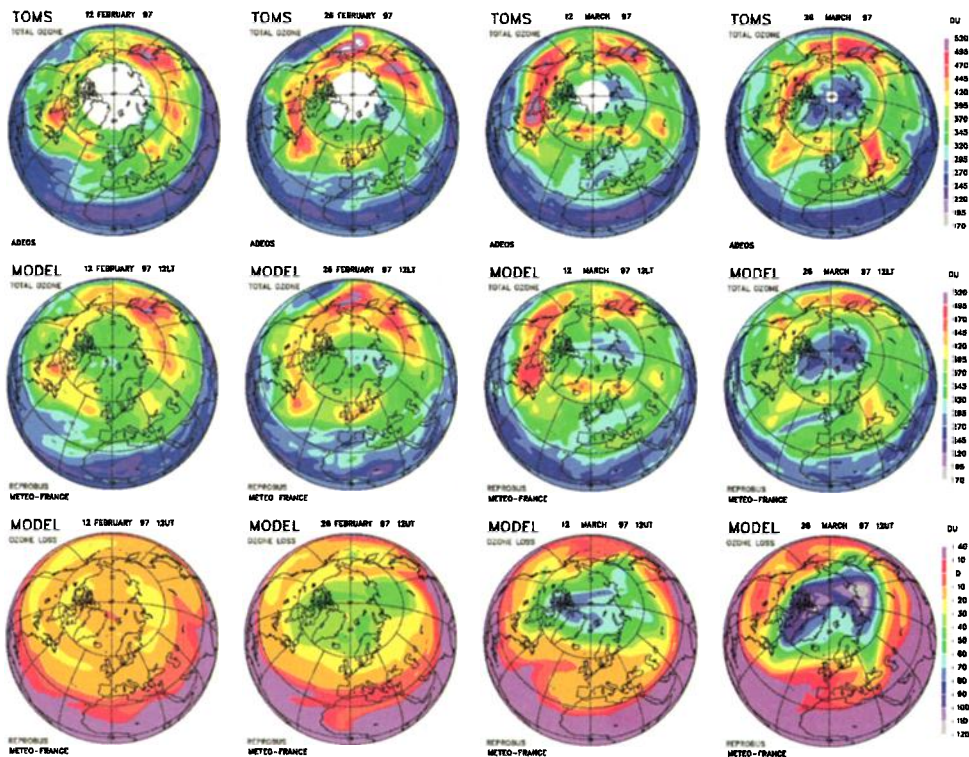


Figure 2. Total ozone distribution (Dobson units) measured by ADEOS-TOMS (top row) and calculated by the model (middle row) on February 12, February 26, March 12, and March 26, 1997. Both observed and calculated data are asynoptic fields obtained near local noon. Also shown is the accumulated total ozone chemical loss or production (bottom row) calculated by the model since the beginning of the simulation.

Discussion and Conclusion

Although considerable, the 60–120 DU chemical loss derived from the model in late March 1997 appears to be close

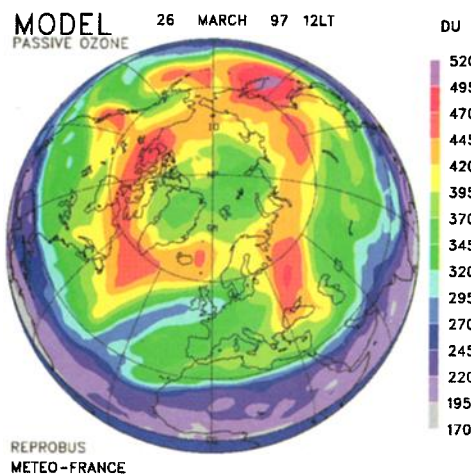


Figure 3. Asynoptic total column distribution (Dobson units) on March 26, 1997 12 00 LT of a passive tracer initialized like ozone at the beginning of the simulation. A large-scale dynamically induced minimum is obtained over the North Pole.

to what was found in 1995 and smaller than in 1996 [Müller *et al.*, 1997a; Goutail *et al.*, 1998b]. Therefore, photochemistry alone cannot explain the record low ozone columns measured by TOMS. Figure 3 shows the simulated passive ozone distribution on March 26, which reflects what the ozone field would have been in the absence of chemical processes. Interestingly, the model reveals a morphology very similar to the TOMS observations, with a distinct polar minimum of 380 DU surrounded by a belt of maxima of 450–500 DU around 60°N latitude. Dynamical processes are therefore also responsible for the unusual pattern of the TOMS total ozone data in late March 1997. Low temperatures in the lower stratosphere are frequently linked to high tropopause and favor low ozone columns [Petzoldt *et al.*, 1994]. This dynamical situation occurred during the second half of March, when an anticyclonic disturbance developed near the pole in the upper troposphere [European Met. Bull., 1997]. Compared to the March climatology of the late 1970s [Newman *et al.*, 1997], before the appearance of increased levels of anthropogenic chlorine in the stratosphere, the passive O_3 column amounts calculated north of 70°N are lower by ~70 DU. This reduction is fully dynamical in origin.

The record low ozone columns measured in the Arctic in 1997 were not due to record large chemical ozone loss. They resulted from the conjunction of a substantial photochemical loss, favored by high levels of chlorine and cold temperatures persisting until late March, and from a large-scale dynamically-induced reduction linked to unusual meteorological conditions near the pole. Combining both effects leads to a total ozone reduction of 130–190 DU in the

Arctic region in late March 1997, in good agreement with the 30-40% observed deviation from the late 1970s TOMS climatology.

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F. Figarol and F. Lefèvre, Centre National de Recherches Météorologiques, Météo-France/CNRS, F-31057 Toulouse Cedex, France. (e-mail: figarol@xdata.cnrm.meteo.fr, franck.lefevre@meteo.fr)

K. Carlsaw and T. Peter, Max-Planck Institut für Chemie, Postfach 3060, D-55020 Mainz, Germany.

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