Severe Arctic ozone loss in the winter 2004/2005: observations from ACE-FTS


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The severe Arctic ozone reduction in the winter 2004/2005 is analyzed using ACE-FTS observations and four different analysis techniques: correlations between ozone and long-lived tracers (adjusted to account for mixing), an artificial tracer correlation method, a profile-descent technique, and the empirical relationship between ozone loss and potential PSC volume. The average maximum ozone loss was about 2.1 ppmv at 475 K–500 K (18 km–20 km). Over 60% of the ozone between 425 K–475 K (16 km–18 km) was destroyed. The average total column ozone loss was 119 DU, ~20–30 DU larger than the largest previously observed Arctic ozone loss in the winter 1999/2000. Citation: Jin, J. J., et al. (2006), Severe Arctic ozone loss in the winter 2004/2005: observations from ACE-FTS, Geophys. Res. Lett., 33, L15801, doi:10.1029/2006GL026752.

1. Introduction

ACE-FTS was a Canadian satellite mission (SCISAT-1) whose primary science objective is to study processes related to Arctic stratospheric ozone (O₃) loss [Bernath et al., 2005]. The primary instrument is a high spectral resolution (0.02 cm⁻¹) Fourier Transform Spectrometer (ACE-FTS) operating from 2.3 to 13.3 μm (750–4400 cm⁻¹) that measures temperature and many species involved in ozone-related chemistry. The retrieval approach for temperature, pressure, and volume mixing ratios (VMRs) is described by Boone et al. [2005]. Validation studies of ACE-FTS version 1.0 and 2.1 data show that stratospheric ozone is in good agreement with the observations from instruments including GOMOS [Fussen et al., 2005], HALOE [McHugh et al., 2005], POAM III, SAGE III, ozonesondes [Walker et al., 2005], Odin/OSIRIS [Petelina et al., 2005], and EOS MLS [Froidevaux et al., 2006]. In this study, the new version 2.2 ACE-FTS data with updated O₃ retrievals, which give more consistent results in the lower stratosphere, are employed to analyze the Arctic O₃ loss during the winter 2004/2005.

2. O₃ Reduction Analyses from Correlations between O₃ and Long-Lived Tracers

The SCISAT-1 orbit allowed ACE-FTS to observe the Arctic (50°N–80°N) between 1 January and 26 March 2005, and the data at potential temperatures 350 K–1700 K (~12 km–45 km) are used here. We consider measurements north of 50°N with sPV (scaled potential vorticity [Manney et al., 1994]) >1.8 × 10⁻⁴ s⁻¹ above 450 K and sPV > 1.4 × 10⁻⁴ s⁻¹ between 350 K–450 K to be inside the polar vortex. Measurements with sPV < 1.0 × 10⁻⁴ s⁻¹ are categorized as outside the polar vortex. We estimate the O₃ loss between 1–7 January 2005 and 8–15 March 2005,
after which the polar vortex broke up [MG2006]. sPV is calculated from UK Met Office analyses [Swinbank et al., 2002].

[5] Figure 1 shows correlations between nitrous oxide (N\textsubscript{2}O) and methane (CH\textsubscript{4}) for the Arctic. For N\textsubscript{2}O values below 200 ppbv (above \textlesssim15 km), the correlations inside the vortex are different from those outside. From early January to March 2005, the vortex correlation curve shifts to its concave side (aside from a few observations shown by the blue dots). This shift suggests that mixing within the vortex or across the vortex edge occurred. Moreover, EOS MLS observations show that the mixing occurred mainly from the inner vortex edge to the vortex core [MG2006].

[6] The blue dots (observations for 8–15 March) in Figure 1 and Figure 2 have relatively large CH\textsubscript{4} values and indicate mixing of extra-vortex air which is not surprising as it was near the time of vortex break-up. These points are excluded in all the following O\textsubscript{3} loss estimates to reduce the uncertainty caused by the mixing.

[7] However, when using the O\textsubscript{3}/CH\textsubscript{4} correlation to quantify the chemical O\textsubscript{3} loss, the mixing effect still needs to be removed. Rex et al. [1999] constructed mixing lines to correct for effects on the denitrification estimates using the change in the N\textsubscript{2}O/CH\textsubscript{4} correlation. Using that method and the vortex O\textsubscript{3}/CH\textsubscript{4} and N\textsubscript{2}O/CH\textsubscript{4} correlations, the effect of mixing within the vortex can be roughly removed. Still, since the O\textsubscript{3}/CH\textsubscript{4} correlations are different inside and outside the vortex even without chemical O\textsubscript{3} loss and both of the end members of the mixing line are set on the vortex correlations, this method cannot correct for the mixing across the vortex edge.

[8] Polynomial fits of the 1–7 January 2005 O\textsubscript{3}/CH\textsubscript{4} correlations are shown as the black and red lines in Figure 3. The black line part is obtained using CH\textsubscript{4} VMRs smaller than 0.30 ppmv but only retains the part below 0.22 ppmv. The red line part is obtained using the entire data set but retains only the part with CH\textsubscript{4} VMRs larger than 0.22 ppmv. In the calculation for the mixing lines, vortex observations for 8–15 March 2005 are binned onto isentropic surfaces in 20 K intervals. For more details about this technique, see Rex et al. [1999].

[9] The adjusted correlation points and their polynomial representation are shown as the grey triangles and line in Figure 3. The modifications mainly impact the middle and lower stratospheric vortex with CH\textsubscript{4} mixing ratios larger than 0.5 ppmv. This suggests that the impact of mixing above \textlesssim35 km during the intervening two months was negligible for the air below.

[10] Employing the adjusted correlation function to the CH\textsubscript{4} measurements for 8–15 March 2005, we can estimate the O\textsubscript{3} inside the vortex, as shown in Figure 4. O\textsubscript{3} reduction occurred between 375 K–800 K (\textsim14 km–30 km) with a maximum loss of 1.8 ppmv between 475 K–500 K (\textsim18 km–20 km). For individual data points, O\textsubscript{3} loss was as large as 2.4 ppmv at these levels. The O\textsubscript{3} loss exceeded 45% between 400 K–500 K (\textsim15 km–20 km) and reached over 60% between 425 K–450 K.

[11] Using vortex averaged profiles of temperature and pressure for the periods 1–7 January and 8–15 March 2005, the total column O\textsubscript{3} loss between 375 K and 800 K for 8–15 March 2005 is estimated to be \textsim114 DU from the O\textsubscript{3}/CH\textsubscript{4} correlations. This value is \textsim25 DU smaller than the estimate of 139 DU obtained using the correlation function without the mixing correction. Based on the adjusted O\textsubscript{3}/N\textsubscript{2}O and O\textsubscript{3}/CFC-12 (dichlodifluoromethane, CCl\textsubscript{2}F\textsubscript{2}) correlations, the maximum O\textsubscript{3} loss were 2.0 ppmv and 2.1 ppmv, and the total column O\textsubscript{3} loss estimates were 125 DU and 130 DU, respectively (Table 1). For the three tracers, the averages are 2.0 ppmv and 123 DU for maximum O\textsubscript{3} loss and total column O\textsubscript{3} loss, respectively.
Figure 4. Vertical vortex O$_3$ and O$_3$ loss profiles. Dash-dot red line, averaged O$_3$ observations for 1–7 January; blue line, averaged O$_3$ observations for 8–15 March. Green circles and purple triangles are calculated O$_3$ and O$_3$ loss values for 8–15 March using the modified O$_3$/CH$_4$ correlations (the grey line in Figure 3), while the green solid and dashed lines are their averages on isentropic surfaces. The black solid and dashed lines show the calculated O$_3$ and O$_3$ loss values for 8–15 March using CH$_4$ and the profile-descent technique, while the grey solid and dashed lines, and the yellow solid and dashed lines show the same result but using N$_2$O and CFC-12, respectively.

3. O$_3$ Loss from Artificial Tracer, Profile-Descent, and Potential PSC Volume Methods

[12] Using the artificial tracer technique proposed by Esler and Waugh [2002] we compose an artificial tracer from simultaneous CH$_4$, N$_2$O, CFC-11 (trichlorofluoromethane, CCl$_3$f) and OCS (carbonyl sulphide) observations from 350 K to 800 K within the vortex for 1–7 January 2005 with the coefficients determined by a linear regression.

\[
\text{Artificial tracer} = 3.262 \times 10^{-3} \text{ CH}_4(\text{ppbv}) - 1.678 \times 10^{-2} \text{ N}_2\text{O}(\text{ppbv}) + 6.903 \times 10^{-3} \text{ CFC-11}(\text{pptyv}) - 1.251 \times 10^{-2} \text{ OCS}(\text{pptyv}) + 3.622
\]  

[13] Using equation (1) we can obtain the artificial tracers inside and outside the vortex for 1–7 January and 8–15 March and their correlations with O$_3$ as shown in Figure 5. The decrease of O$_3$ with respect to this artificial long-lived tracer can be regarded as the chemical O$_3$ loss. However, because the correlations inside and outside the vortex are different, this method cannot correct for the mixing across the vortex edge. This kind of mixing can only increase O$_3$ for an artificial tracer value, which suggests that neglecting mixing across the edge gives a conservative O$_3$ loss estimate for this method. However, because of the early vortex linear correlation, which cannot be changed by inner vortex mixing, this method still can reduce the uncertainty due to the mixing within in the vortex. Figure 6 shows the vertical distribution of O$_3$ loss estimated by this method. The loss maximized at around 450 K with an average reduction of ~2.1 ppmv, slightly larger than the estimate from the modified correlations. The reduction extended up to about 650 K (~25 km) and the column O$_3$ loss between this level and 375 K was 116 DU.

[14] Next, we estimate O$_3$ loss using the profile-descent technique [MG2006]. Using the vortex averaged long-lived tracer profiles (e.g., CH$_4$ in Figure 2) for 1–7 January and 8–15 March 2005, vortex descent rates can be deduced. The descent rates are then applied to the O$_3$ profile for 1–7 January (dash-dot red line, in Figure 4 and Figure 6) to derive new O$_3$ profiles for mid-March. The difference between the O$_3$ measurements and the derived O$_3$ values for 8–15 March is considered as O$_3$ loss. Estimates based on these inferred descent rates from the profiles of CH$_4$, N$_2$O and CFC-12 are shown in Figure 4 and Table 1. The maximum O$_3$ loss averaged for the three species occurred at 500 K and was ~2.3 ppmv, which is slightly larger than the estimates from the artificial tracer method and the adjusted correlations. The average total column loss using the CH$_4$, N$_2$O and CFC-12 profiles was 128 DU, close to the results from the adjusted correlation analysis but 10% larger than the estimate from the artificial tracer method.

[15] Finally, using National Center for Environmental Prediction/National Center for Atmospheric Research prediction/National Center for Atmospheric Research (NCEP/NCAR) temperature data [Kistler et al., 2001], and typical HNO$_3$ and H$_2$O mixing ratios from ACF-FTS measurements from January to March 2005 we estimate the potential PSC volume (V$_{PSC}$) to be $41 \times 10^6$ km$^3$. According to the empirical relation of Rex et al. [2004], this would imply column O$_3$ loss of 108±15 DU. The uncertainty range is due to uncertainties in the HNO$_3$ and H$_2$O values used for the potential PSC volume calculation. This

Figure 5. Correlations between O$_3$ and an artificial tracer composed of CH$_4$, N$_2$O, CFC-11, and OCS.

Figure 6. Vertical vortex O$_3$ and O$_3$ loss profiles. Lines and symbols are the same as in Figure 4 except that the O$_3$ and O$_3$ loss values are derived from the correlations between O$_3$ and the artificial tracer.
estimate is about 12%, 7%, and 15% smaller than the direct estimates using ACE-FTS data from the adjusted correlation analysis, the artificial tracer method and the profile-descent technique, respectively. A possible reason for relatively smallness of this estimate is that the NCEP/NCAR temperature is biased high in the lower stratosphere compared to other data sets [Manney et al., 2005, and references therein].

On average, ACE-FTS maximum O₃ loss by mixing ratio is estimated to be ~2.1 ppmv between 475 K–500 K. This value is ~0.8 ppmv larger than estimates from EOS MLS and POAM III using vortex-averaged descent from a radiation calculation [MG2006]. Between 450 K–500 K, the ACE-FTS estimate is ~2.0 ppmv from the profile-descent method, which is close to the loss in the outer vortex and ~0.5 ppmv larger than the vortex average from EOS MLS using the same method.

Before the winter 2004/2005 the largest Arctic O₃ loss on record occurred in the winter 1999/2000 [WMO, 2003]. The above average maximum O₃ loss is ~0.8 ppmv larger than the maximum loss estimated from POAM III using the profile-descent technique and modeling in the Arctic for the winter 1999/2000 [Hoppel et al., 2002]. However, the loss in 2004/2005 was about 0.5 ppmv smaller than the maximum loss at 450 K at the end of March 2000 from the MATCH analysis [Rex et al., 2002]. Nevertheless, since loss extended farther down where ozone VMRs are smaller, the maximum fractional loss reached 60% at 425 K – 475 K (~16 km – 18 km), close to the estimate using various methods and data sets during SOLVE/THESOO 2000 [Newman et al., 2002]. The average of the total column loss estimates from the four methods is 119 DU. This is ~20–30 DU larger than estimates at the end of March 2000 [WMO, 2003].

4. Summary

The Arctic winter 2004/2005 was particularly cold [MG2006] with substantial chlorine activation [Dufour et al., 2006]. Table 1 shows the estimates of the maximum O₃ mixing ratio loss and total column O₃ loss from various methods. The maximum mixing ratio loss over the entire winter was ~1.8 ppmv based on the adjusted O₃/CH₄ correlations and ~2.0 ppmv of average from the adjust O₃/CH₄, O₃/N₂O and O₃/CFC-12 correlations, ~2.1 ppmv from the correlation between O₃ and an artificial tracer, and ~2.3 ppmv from the profile-descent method. These estimates are ~0.5 ppmv larger than those from EOS MLS [MG2006]. Total column O₃ loss was ~123 DU based on the adjusted O₃/long-lived tracer correlations and 116 DU based on the artificial tracer method. Only the effect of mixing within the vortex is corrected for in the tracer correlation methods used here. Hence, the results are likely to be conservative. However, exact mixing effects cannot be assessed due to the unusual and complex morphology of O₃ in this year [MG2006]. A smaller estimate of ~108 DU is obtained from the empirical relationship of Rex et al. [2004], while a larger value of ~127 DU is obtained from the profile-descent technique. The average from the four methods is 119 DU, which is the largest Arctic column O₃ loss observed. Further investigation using modeling and other observations is necessary to better understand the O₃ depletion during this winter.

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References


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Table 1. Total column O₃ loss in DU and maximum VMRs loss in ppmv (in brackets)


