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

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[1] 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

[2] The Atmospheric Chemistry Experiment (ACE) is a Canadian satellite mission (SCISAT-1) whose primary science objective is to study processes related to Arctic stratospheric polar ozone (O_3) 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 O_3 retrievals, which give more consistent results in the lower stratosphere, are employed to analyze the Arctic O_3 loss during the winter 2004/2005.

[3] To quantify chemical O_3 loss throughout the winter, and to isolate it from dynamical effects, a tracer correlation method has been used [e.g., *Proffitt et al.*, 1990; *Müller et al.*, 1997; *Salawitch et al.*, 2002]. This method assumes that the correlation between O_3 and a long-lived tracer is preserved inside the polar vortex if there is no chemical O_3 reduction, and thus any change of the correlation relationship yields information on chemical O_3 loss. However, mixing inside the polar vortex and across the vortex edge can also change the correlations. *Rex et al.* [2002] and *Müller et al.* [2005] argued that neglecting mixing across the polar vortex edge could only result in an underestimation of the chemical O_3 loss. For the winter 1999/2000, when the largest ever Arctic O_3 loss was reported [*World Meteorological Organization (WMO)*, 2003], mixing across the vortex edge was negligible [*Ray et al.*, 2002; *Richard et al.*, 2001; *Salawitch et al.*, 2002]. *Ray et al.* [2002] also showed that mixing within the vortex can be responsible for most long-lived tracer correlation changes. In a study on denitrification, *Rex et al.* [1999] proposed a mixing-line technique to correct for the effects of mixing across the vortex edge when using the correlation method. Later, *Esler and Waugh* [2002] composed an artificial tracer that has a linear relation with the nitrogen family (NO_y), so mixing does not change the correlations, to get a robust estimate of denitrification. Here, the correlation method with the mixing correction and the artificial tracer method are both used to quantify the O_3 loss. In addition, a profile-descent method [*Manney et al.*, 2006] (hereinafter referred to as *MG2006*) is used. Finally, *Rex et al.* [2004] showed that there exists a quantitative empirical linear relation between the Arctic total column chemical O_3 loss and the potential PSC volume from mid-December to March, which offers another tool for chemical O_3 loss estimation.

2. O_3 Reduction Analyses from Correlations between O_3 and Long-Lived Tracers

[4] 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]) $>1.8 \times 10^{-4}$ s^{-1} above 450 K and sPV $> 1.4 \times 10^{-4}$ s^{-1} between 350 K–450 K to be inside the polar vortex. Measurements with sPV $< 1.0 \times 10^{-4}$ s^{-1} are categorized as outside the polar vortex. We estimate the O_3 loss between 1–7 January 2005 and 8–15 March 2005,

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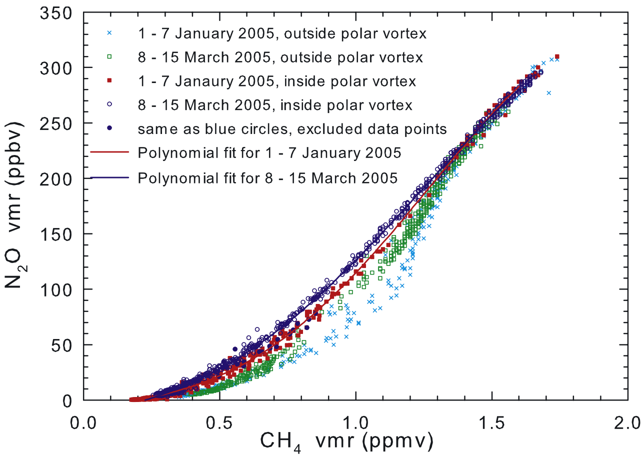

Figure 1. Correlations of N_2O and CH_4 between 350 K–1700 K.

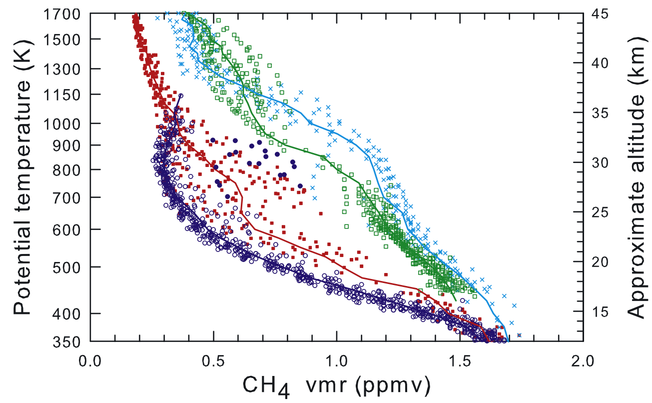

Figure 2. Vertical distribution of CH_4. The symbols are the same as in Figure 1 and the lines are the averages.

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_2O) and methane (CH_4) for the Arctic. For N_2O values below 200 ppbv (above ~15 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_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_3 loss estimates to reduce the uncertainty caused by the mixing.

[7] However, when using the O_3/CH_4 correlation to quantify the chemical O_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_2O/CH_4 correlation. Using that method and the *vortex* O_3/CH_4 and N_2O/CH_4 correlations, the effect of mixing within the vortex can be roughly removed. Still, since the O_3/CH_4 correlations are different inside and outside the vortex even without chemical O_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_3/CH_4 correlations are shown as the black and red lines in Figure 3. The black line part is obtained using CH_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_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_4 mixing ratios larger than 0.5 ppmv. This suggests that the impact of mixing above ~35 km during the intervening two months was negligible for the air below.

[10] Employing the adjusted correlation function to the CH_4 measurements for 8–15 March 2005, we can estimate the O_3 inside the vortex, as shown in Figure 4. O_3 reduction occurred between 375 K–800 K (~14 km–30 km) with a maximum loss of 1.8 ppmv between 475 K–500 K (~18 km–20 km). For individual data points, O_3 loss was as large as 2.4 ppmv at these levels. The O_3 loss exceeded 45% between 400 K–500 K (~15 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_3 loss between 375 K and 800 K for 8–15 March 2005 is estimated to be ~114 DU from the O_3/CH_4 correlations. This value is ~25 DU smaller than the estimate of 139 DU obtained using the correlation function without the mixing correction. Based on the adjusted O_3/N_2O and O_3/CFC-12 (dichlodifluoromethane, CCl_2f_2) correlations, the maximum O_3 loss were 2.0 ppmv and 2.1 ppmv, and the total column O_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_3 loss and total column O_3 loss, respectively.

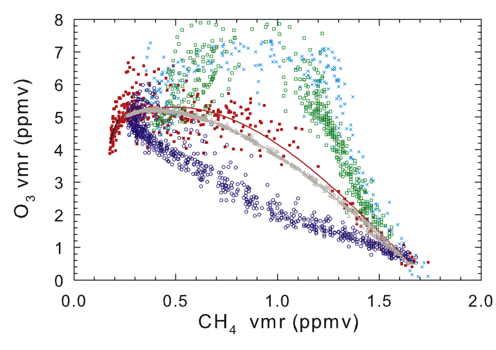

Figure 3. Correlations of O_3 and CH_4 between 350 K–1700 K. The color symbols are the same as in Figure 1. See text for details of the lines and grey triangles.

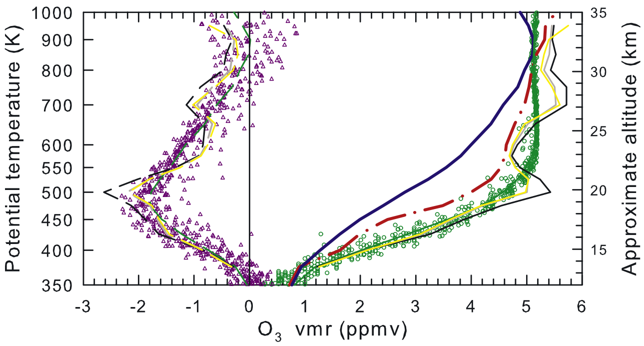

Figure 4. Vertical vortex O_3 and O_3 loss profiles. Dash-dot red line, averaged O_3 observations for 1–7 January; blue line, averaged O_3 observations for 8–15 March. Green circles and purple triangles are calculated O_3 and O_3 loss values for 8–15 March using the modified O_3/CH_4 correlations (the 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_2O 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_2O, CFC-11 (trichlorofluoromethane, CCl_3f) 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.

$$\begin{aligned} \text{Artificial tracer} = {} & 3.262 \times 10^{-3}\ CH_4(\text{ppbv}) \\ & -1.678 \times 10^{-2}\ N_2O(\text{ppbv}) \\ & +6.903 \times 10^{-3}\ \text{CFC-11}(\text{pptv}) \\ & -1.251 \times 10^{-2}\ \text{OCS}(\text{pptv}) + 3.622 \end{aligned} \quad (1)$$

[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.

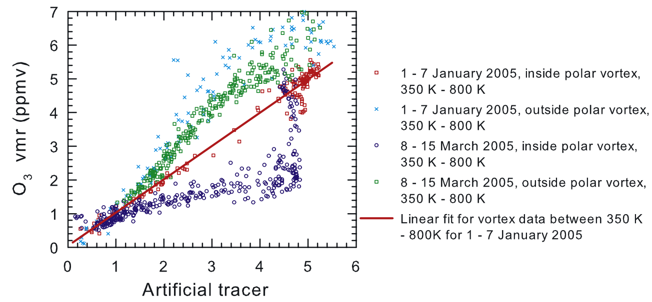

Figure 5. Correlations between O_3 and an artificial tracer composed of CH_4, N_2O, CFC-11, and OCS.

[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_2O 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_2O 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 Predication/National Center for Atmospheric Research Reanalysis (NCEP/NCAR) temperature data [*Kistler et al.*, 2001], and typical HNO_3 and H_2O mixing ratios from ACE-FTS measurements from January to March 2005 we estimate the potential PSC volume (V_{PSC}) to be 41×10^6 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_2O values used for the potential PSC volume calculation. This

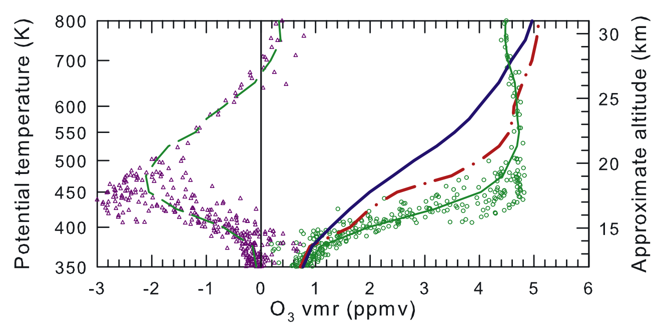

Figure 6. Vertical vortex O_3 and O_3 loss profiles. Lines and symbols are the same as in Figure 4 except that the O_3 and O_3 loss values are derived from the correlations between O_3 and the artificial tracer.

Table 1. Total column O_3 loss in DU and maximum VMRs loss in ppmv (in brackets)

Method	CH_4	N_2O	CFC-12	Average
Original correlation	139 (2.1)	131 (2.1)	131 (2.1)	134 (2.1)
Adjusted correlation	114 (1.8)	125 (2.0)	130 (2.1)	123 (2.0)
Profile-descent technique	137 (2.6)	125 (2.2)	123 (2.2)	128 (2.3)
Artificial tracer method				116 (2.1)
V_{PSC} method				108

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].

[16] On average, ACE-FTS maximum O_3 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.

[17] Before the winter 2004/2005 the largest Arctic O_3 loss on record occurred in the winter 1999/2000 [*WMO*, 2003]. The above average maximum O_3 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 between 425 K–475 K (~16 km–18 km), close to the estimate using various methods and data sets during SOLVE/THESEO 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

[18] 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 O_3 mixing ratio loss and total column O_3 loss from various methods. The maximum mixing ratio loss over the entire winter was ~1.8 ppmv based on the adjusted O_3/CH_4 correlations and ~2.0 ppmv of average from the adjust O_3/CH_4, O_3/N_2O and O_3/CFC-12 correlations, ~2.1 ppmv from the correlation between O_3 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 O_3 loss was ~123 DU based on the adjusted O_3/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 O_3 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 O_3 loss observed. Further investigation using modeling and other observations is necessary to better understand the O_3 depletion during this winter.

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