An improved measure of ozone depletion in the Antarctic stratosphere

P. E. Huck,¹ S. Tilmes,² G. E. Bodeker,³ W. J. Randel,² A. J. McDonald,¹ and H. Nakajima⁴

Received 31 July 2006; revised 9 February 2007; accepted 28 February 2007; published 5 June 2007.

[1] Ozone mass deficit is a commonly used index to quantify Antarctic ozone depletion. However, as currently defined, this measure is not robust with respect to reflecting chemical ozone loss within the Antarctic vortex. Therefore, in this study, a new definition of ozone mass deficit (OMD) is developed. The 220 Dobson Unit based value currently used as the threshold for ozone depletion has been replaced with a new ozone background representative of pre-ozone-hole conditions. Second, the new OMD measure is based on ozone measurements within the dynamical vortex. A simpler method is also proposed whereby calculation of the vortex edge is avoided by using the average latitude of the vortex edge (62° S) as the spatial limiting contour. An indication of the errors in OMD introduced when using this simpler approach is provided. By comparing vortex average total ozone loss (defined using the new background and limiting contour) with partial column accumulated chemical ozone loss calculated with the tracer-tracer correlation method for 1992-2004 and in more detail for 1996 and 2003, it is shown that the new OMD measure is representative of chemical ozone loss within the vortex. In addition the new criteria have been applied to the calculation of ozone hole area. The sensitivity of the new measures to uncertainties in the background have been quantified. The new ozone loss measures underestimate chemical ozone loss in highly dynamically disturbed years (2002 and 2004), and criteria for identifying these years are presented. The new measures should aid chemistry-climate model intercomparisons since ozone biases in the models are avoided.

Citation: Huck, P. E., S. Tilmes, G. E. Bodeker, W. J. Randel, A. J. McDonald, and H. Nakajima (2007), An improved measure of ozone depletion in the Antarctic stratosphere, J. Geophys. Res., 112, D11104, doi:10.1029/2006JD007860.

1. Introduction

[2] Ozone destruction over Antarctica has been of scientific interest since the discovery of the ozone hole in 1985 [Farman et al., 1985]. Initially the severity of Antarctic ozone depletion was quantified using minimum total column ozone values over Antarctica. However, later studies [Stolarski et al., 1990] showed that in some years (e.g., 1987 and 1989) while minimum ozone values were similar, the development of the ozone hole area was quite different. The ozone hole area, introduced by Stolarski et al. [1990], and defined as the area within the 220 Dobson Unit (DU) contour, has become another standard measure of the severity of Antarctic ozone depletion. More recently, a third measure, ozone mass deficit, that combines the size and depth of the ozone hole has been used [Uchino et al., 1999].

This metric is calculated by integrating air masses depleted in ozone over latitudes poleward of 40°S. In this case air masses depleted in ozone are identified as those with less than 220 DU of total column ozone. This threshold has been generally acknowledged because it is lower than all preozone-hole values (before 1980) and the 220 DU contour lies in the region of steep ozone gradients surrounding the ozone hole. All three measures mentioned above (minimum total column ozone, ozone hole area and ozone mass deficit) have been used to describe and study interannual and intraannual variations in Antarctic ozone depletion [World Meteorological Organization (WMO), 2003].

[3] The annual minimum value in total column ozone over Antarctica is largely insensitive to reversible dynamical disturbances of the vortex edge and has shown little change through the 1990s during the annual Antarctic vortex period (18 July to 30 November) [WMO, 2003]. The minimum appears usually in late September or early October and has remained essentially unchanged at about 100 DU since 1993 after the considerable decrease during the 1980s and early 1990s. Equivalent effective Antarctic stratospheric chlorine (EEASC), a measure of the concentration in ozone depleting substances in the Antarctic stratosphere, peaks in 2001 [Newman et al., 2006]. The

¹Department of Physics and Astronomy, University of Canterbury, Christchurch, New Zealand.

²National Center of Atmospheric Research, Boulder, Colorado, USA. ³National Institute of Water and Atmospheric Research, Lauder, Central Otago, New Zealand. ⁴National Institute for Environmental Studies, Tsukuba, Japan.

Copyright 2007 by the American Geophysical Union. 0148-0227/07/2006JD007860



Figure 1. Pre-ozone-hole mean annual cycle within the Antarctic vortex (thick grey line) and its three σ uncertainty (thin black lines) displayed together with the 1979–1981 (dotted lines) and 2003 vortex mean ozone (thick black line) from the NIWA combined total column ozone database (see text). The 220 DU threshold, commonly used in calculations of OMD and OHA, is shown with a dashed line. The light grey shaded area indicates ozone loss that would not be incorporated in an ozone depletion index based on a 220 DU threshold.

insensitivity of annual minimum ozone values to changing EEASC suggests that its use in detecting a response of Antarctic ozone depletion to reductions in ozone depleting substances is limited. Similar saturation effects are observed in ozone hole area. Ozone hole area is more sensitive to changes in the vortex collar region than minimum total ozone. The springtime ozone hole area increased rapidly during the 1980s and more gradually during the 1990s [WMO, 2003] while EEASC steadily increased. From 1995 to 2005 the ozone hole area also shows a lack of response to EEASC [Newman et al., 2006]. Unlike minimum total column ozone and ozone hole area averaged over the Antarctic vortex period, the ozone mass deficit averaged over the same time period shows significant growth during the 1990s, as discussed in Bodeker et al. [2005], and greater sensitivity to recent changes in EEASC.

[4] The long-term trend in ozone depletion is mainly controlled by chemical processes and the interannual variability can be explained by variability in temperatures and planetary wave activity [Huck et al., 2005]. However, when expanding these studies to intra-annual variability, more detailed processes need to be considered which can be neglected when averaging over the Antarctic vortex period. The start date and the rate of ozone destruction depend on temperatures and on sunlight reaching areas with polar stratospheric clouds. Furthermore, ozone depletion is controlled indirectly by planetary wave activity displacing air masses out of the polar darkness and therefore causing earlier ozone depletion in some regions [Solomon et al., 1993; Austin and Butchart, 2003]. By transporting heat, waves directly affect polar temperatures and therefore the occurrence of polar stratos heric clouds (PSCs) which control chlorine activation and therefore ozone destruction [*Schoeberl and Hartmann*, 1991]. To quantify intra-annual variability in ozone depletion a measure sensitive to all of these processes is necessary.

[5] If the ozone mass deficit and ozone hole area definitions are based on a 220 DU threshold, as was done by Huck et al. [2005] and earlier studies, the identified onset of ozone depletion occurs well after sunlight returns to the polar regions, which is inconsistent with accepted theory of polar ozone depletion chemistry (i.e., that ozone depletion commences with the return of sunlight to regions containing PSCs) and with observations of chemical ozone loss within the vortex. The 220 DU based measures do not capture the full deviation from pre-ozone-hole conditions (see Figure 1; figure details presented in section 2.1). This suggests that the first step toward improving the ozone mass deficit and ozone hole area metrics is to define a better background value that describes the column ozone for pre-ozone-hole conditions. Since the 220 DU contour has previously provided the unperturbed background ozone as well as the limiting contour for the Antarctic vortex, calculation of a new contour for defining the region within which ozone depletion is considered becomes necessary when using a pre-ozone-hole background.

[6] To test whether these new parameters for ozone mass deficit and ozone hole area calculation are likely to lead to more representative measures of Antarctic ozone depletion, vortex average total column ozone loss is derived using the pre-ozone-hole background. This measure is compared with the accumulated chemical loss in partial column ozone derived using tracer-tracer correlations for Halogen Occultation Experiment (HALOE) and Improved Limb Atmospheric Spectrometer (ILAS) data. While the HALOE comparisons show the utility of the new measures in tracking springtime ozone loss, the comparisons with ILAS show how the new measures are able to track the wintertime ozone depletion.

[7] With the tracer-tracer correlation method the impact of dynamical changes on ozone within the analyzed layer are separated from the chemical ozone loss [*Proffitt et al.*, 1990; *Müller et al.*, 2001]. It can be shown that the magnitude of ozone loss in the new measure, especially in winter, is due to chemical ozone loss.

[8] The approach used to calculate the pre-ozone-hole background ozone and the two spatial integration limits considered are introduced in section 2.1. In section 2.2 the application of the pre-ozone-hole background and the new spatial integration limits to ozone mass deficit and ozone hole area is described. In section 2.3 the new method is tested by investigating whether these changes result in better tracking of Antarctic chemical ozone loss calculated with more complex and sophisticated techniques such as the tracer-tracer correlation method. The results are presented in section 3, in particular ozone mass deficit in section 3.1 and the chemical ozone loss in section 3.2. The results are summarized and conclusions are drawn in section 4.

2. Methods

2.1. Derivation of Pre-Ozone-Hole Background

[9] To estimate the pre-ozone-hole mean annual cycle in total column ozone over Antarctica, coefficients from a

 Table 1. Regression Model Coefficients to Calculate the Pre-Ozone-Hole Background

Coefficient	Value
А	302.2
В	-9.522
С	2.753
D	-8.708
Е	17.80
F	-11.07
G	-4.121
Н	1.518
Ι	-2.250

regression model applied to ozone in an equivalent latitude coordinate [*Bodeker et al.*, 2001] were used to construct the stationary part of the annual cycle within the Antarctic polar vortex. This mean annual cycle is compared with vortex average daily total column ozone from the NIWA combined total column ozone database for 1979, 1980 and 1981 in Figure 1, which shows the consistency of this background with pre-ozone-hole conditions in observations. In addition, using data for 2003 as an example, the grey shaded area in Figure 1 shows the ozone deficit below the pre-ozone-hole background that would be excluded from a 220 DU based ozone hole metric. The winter/spring decrease in the pre-ozone-hole background results from the cold polar trough region south of 70°S where planetary wave activity is weak [*Chandra and McPeters*, 1986]. A function of the form:

$$O_3 = A + B \cdot \sin\left(2\pi \frac{d}{366}\right) + C \cdot \cos\left(2\pi \frac{d}{366}\right) + \ldots + H$$
$$\cdot \sin\left(8\pi \frac{d}{366}\right) + I \cdot \cos\left(8\pi \frac{d}{366}\right)$$

was fitted to this mean annual cycle, where d is the day number of the year. The coefficients A to I are listed in Table 1 and can be used to reconstruct the grey curve shown in Figure 1. If this method is used for calculations based on chemistry-climate model output, the method described by *Eyring et al.* [2006, Appendix 1] can be used to construct the pre-ozone-hole background.

[10] To test the sensitivity of the new measures to the preozone-hole background, a three σ envelope around the grey curve in Figure 1 was calculated from 100,000 Monte Carlo calculations of the background using the parameter uncertainties provided by the regression model [*Bodeker et al.*, 2001]. The upper and lower limits for the pre-ozone-hole background are displayed in Figure 1. The resultant uncertainties in the ozone mass deficit and ozone hole area measures are indicated in the figures below.

[11] Ideally the Antarctic ozone depletion index should be sensitive only to ozone depletion that occurs within the Antarctic vortex. Therefore our primary approach is to use the inner vortex edge, derived using the *Nash et al.* [1996] method, to define the region over which the ozone loss is calculated (the spatial integration limit). NCEP/NCAR reanalyses potential vorticity (PV) values were transformed into equivalent latitude coordinates. The vortex edge is located at the steepest PV gradient multiplied by the total wind speed. The inner and outer edge of the polar vortex are

defined by the maximum and minimum in the second derivative of PV with respect to equivalent latitude. The inner vortex edge is used in these studies to avoid counting any mixed air in the vortex boundary region which would affect the value of ozone depletion. However, this also means that any ozone depletion occurring in the vortex collar region will not be accounted for and therefore a slight underestimation of depleted ozone mass and ozone hole area would occur in this case. The vortex edge has been calculated for an altitude range between approximately 17-25 km at potential temperature levels of 450, 550 and 650 K. An ozone measurement is flagged as being inside the vortex if the equivalent latitude at that location is greater than the equivalent latitude of the inner vortex edge at any of the 3 levels. It should be noted that the vortex edge might be shifted or twisted at altitudes above 650 K and therefore does not match the derived vortex edge between 450-650 K. This effect could possibly occur at the end of winter or during a disturbed vortex (e.g., in 2002 [Newman and Nash, 2005]). The entire ozone column would be affected and it could result in an underestimation of ozone mass deficit and ozone hole area.

[12] Given the additional complexity associated with calculating the vortex edge, and to ensure the utility of this method, an additional spatial integration limit of 62° S, the average latitude of the vortex edge [*Bodeker et al.*, 2002], is considered. Hereinafter, quantities calculated on the basis of the vortex edge are denoted with a *vortex* subscript while those based on the 62° S limit are denoted with a *62S* subscript. In Figure 2 the limiting contours, including the 220 DU contour, are displayed on top of a latitude-time ozone distribution for 2003. It is obvious that when using the 220 DU contour as the spatial integration limit instead of the actual inner vortex edge (thick black line and crosses in Figure 2) the area of ozone depletion is restricted to within the polar vortex. However, it also omits much of the vortex region, particularly early in the season.

2.2. Ozone Mass Deficit and Ozone Hole Area

[13] In theory, ozone depletion begins as soon as sunlight reaches areas of polar stratospheric clouds, activates chlorine, and initiates heterogenous ozone depleting reactions. These processes continue until solar radiation and dynamics warm the polar region to temperatures above the threshold for PSC existence. Activated chlorine then returns to inactive reservoir species. The warming of the polar regions also causes a weakening of the polar vortex until the final warming event breaks the structure apart. From theory it is therefore expected that the start of the ozone hole should coincide with the day when sunlight first reaches PSC areas. This date has been calculated and compared to the date when the ozone mass deficit (OMD) and ozone hole area (OHA) first become nonzero, as discussed in more detail in section 3.1.

[14] In addition to calculating the daily OMD using the method described by *Huck et al.* [2005], OMD_{vortex} and OMD_{62S} have been calculated by replacing the 220 DU threshold with the pre-ozone-hole background. Daily values of these indices were calculated from the NIWA combined total column ozone database [*Bodeker et al.*, 2005] from 1979 to 2004. Through the period when a vortex edge can be defined, ozone measurements within the vortex (section 2.1)



Figure 2. Zonal mean total column ozone over the Southern Hemisphere in 2003 shown using shades of grey indicated by the scale on the right, together with the different spatial integration limits used in this and previous studies, i.e., the inner vortex edge (black crosses), the mean location of the vortex edge at 62° S (thin black line) and the 220 DU contour (thick black line).

that fall below the background add to the OMD_{vortex} total while measurements above the background are subtracted from the OMD_{vortex} total. The same approach is used when calculating OMD_{62S} using values poleward of 62°S. The NIWA combined database has also been used to calculate daily OHAvortex and OHA62S by adding the area of each cell with an ozone value below the pre-ozone-hole background to a daily total if it is within the spatial integration limit and subtracting the area of each cell with an ozone value above the background. OMD and OHA values have only been calculated during the Antarctic vortex season and are considered to be zero outside that period. Because the date when sunlight first illuminates PSC regions occurs after the date when the dynamical vortex is first defined, this restriction does not affect the start date of OMD and OHA. Shortly after the vortex forms, in the absence of other drivers of ozone variability, the OMD and OHA values should be zero. However, the derived pre-ozone-hole background does not account for year-to-year ozone fluctuations caused by various transport processes, e.g., the quasibiennial oscillation, solar cycle or volcanic eruptions. This results in OMD and OHA values that are occasionally nonzero during this period. To correct for this artifact, the average May value for OMD and OHA for each year has been subtracted from the data. Averaged over 26 years (1979–2004) the OMD is corrected by 1.5% of the annual maximum value and the OHA by 5.6%.

2.3. Comparison With Tracer-Tracer Correlation Derived Chemical Ozone Loss

[15] To test whether the newly defined OMD measure better tracks chemical ozone loss within the Antarctic vortex, spatially averaged ozone loss over the two regions defined above, was compared with ozone loss within the vortex calculated with a tracer-tracer correlation method. These two methods for calculating Antarctic ozone depletion are completely ind ent. Daily vortex average total column ozone loss was calculated for 1992-2004 by subtracting the measured ozone column from the preozone-hole background. Chemical ozone loss was derived with the tracer-tracer correlation method for 1992-2004 [Tilmes et al., 2006a]. This method requires measurements of an ozone profile and simultaneous measurements of a passive tracer which can be used to track the diabatic descent of air parcels within the vortex. HALOE measurements were used for 1992-2004 while ILAS V6.0 and ILAS-II V1.4 measurements were used for more detailed comparisons in 1996 and 2003, respectively. For the accumulated ozone loss derived from HALOE data, CH₄ and/or HF were used as the passive tracer while for the accumulated ozone loss derived from ILAS-II, N2O was used as the passive tracer. HALOE provided discontinuous measurements of O₃, CH₄ and HF inside the polar vortex [Russell et al., 1993] which have been averaged over periods between 1 and 20 days and result in one to four data points per year. ILAS provided daily measurements of O₃ and N₂O inside the polar vortex from 30 October to 13 December 1996 [Suzuki et al., 1995; Sasano et al., 1999] and ILAS-II from 2 April to 24 October 2003 [Nakajima et al., 2006; Nakajima, 2006]. While HALOE provides more years for comparison of the tracer-tracer method with the total column ozone loss derived here (1992-2004), ILAS and ILAS-II provide better temporal coverage during the years for which data are available (1996 and 2003).

[16] For the tracer-tracer correlation method an early winter reference function is derived for chemically unperturbed conditions in an established vortex. Deviations from this reference function are used to identify chemical ozone loss. This reference function was calculated using O₃ and N₂O measurements in June from ILAS and ILAS-II. The relationship between ozone and N₂O does not change within the confined borders of the polar vortex unless ozone reacts chemically. Accumulated chemical ozone loss was calculated by integrating the tracer-tracer correlation derived ozone loss profiles between 350 K and 550 K for HALOE and between 350 K and 600 K for ILAS and ILAS-II. These altitude ranges have been identified as the region where the halogen catalyzed polar ozone loss takes place [Tilmes et al., 2006b]. Therefore the integrated values represent the total amount of chemically destroyed ozone in the polar vortex. Mixing of air from outside to inside the vortex could potentially result in an underestimation of chemical ozone loss [Müller et al., 2005].

3. Results

3.1. Ozone Mass Deficit and Ozone Hole Area

[17] The OMD_{vortex} and OMD_{62S} measures, calculated as described in section 2.2, are compared with each other, and with the OMD based on the 220 DU threshold, for 2003 in Figure 3. While the 220 DU based OMD starts about two months later than both new measures, the seasonal start date of the new OMDs is now very close to the date when sunlight first illuminates PSC areas (i.e., 21 May 2003) which is consistent with observations and theory. As defined, OMD_{vortex} terminates at the same time as the vortex breaks up (the time when no edge can be found using the criterion of *Nash et al.* [1996]) while OMD_{62S} remains positive to the end of the year. Both new measures for ozone



Figure 3. Daily values of OMD_{vortex} (thin black line), OMD_{62S} (thin grey line) and OMD_{220DU} (thick black line) for 2003. The arrow early in the period shows the first day of sunlight on PSC areas in 2003. The two error bars on the OMD_{vortex} and OMD_{62S} curves indicate the uncertainties on these measures, resulting from uncertainty in the pre-ozone-hole background (see Figure 1), which are similar throughout the season.

mass deficit track each other closely until shortly after the seasonal maximum after which OMD_{62S} shows higher values compared to OMD_{vortex} . Overall, considering the years 1979–2004 (not all shown), more depleted ozone mass is accounted for in the new OMD measures compared to the 220 DU based OMD. This confirms that it is likely that the 220 DU based OMD omits some of the depleted ozone mass, as has been suggested by Figure 1.

[18] A similar comparison of the ozone hole area measures conducted for 2003 (Figure 4), indicates similar results as for OMD. Comparing the 220 DU based OHA with OHA_{vortex} and OHA_{62S}, the seasonal evolution of the two new measures shows an earlier start compared to the original OHA but all three measures considered match at similar values after the maximum of the 220 DU based OHA is reached. Because the seasonal start date for the new ozone hole area measures is similar to that for when sunlight first reaches PSC areas (and earlier than if the 220 DU contour is used), the new measures are considered to be a better measure of the ozone hole area than the method using the 220 DU contour.

[19] Use of 62°S as the spatial integration limit rather than the vortex edge introduces errors in OMD and OHA. Calendar month averages of these errors over 26 years are summarized in Table 2.

3.2. Vortex Average Chemical Ozone Loss

[20] Two measures for vortex average chemical ozone loss have been derived as described in section 2. The two measures are (1) the total column ozone loss derived by subtracting a pre-ozone-hole background from column ozone measurements within the vortex and (2) the accumulated chemical ozone loss derived with the tracer-tracer correlation method. Unlike the tracer-tracer correlation method which is specif chemical ozone loss, the total column ozone loss method considers the combination of chemical ozone loss and dynamical resupply. The two methods are compared, from 1992 to 2004, in Figure 5. To reduce uncertainties in the accumulated chemical ozone loss, up to 20 profiles inside the vortex have been averaged for one data point. Because of the orbit of the HALOE instrument this results in 1 to 4 data points per year. Except for 2002 and 2004 (unusually dynamically perturbed years, see below) the two methods show excellent agreement, well within the 1σ uncertainties on the HALOE based ozone loss and the uncertainty resulting from the pre-ozone-hole background envelopes. In dynamically perturbed years, the vortex average total column ozone loss (and by inference the new OMD metric) is reduced as a result of dynamical resupply of ozone to the vortex; that is, it does not accurately reflect the pure chemical depletion of ozone. Dynamically perturbed years can be identified using the following indicators: (1) when the September average temperatures at 65°S and 50 hPa exceed 210 K; and/or (2) when the standard deviation on September zonal mean zonal winds at 60°S and 50 hPa exceeds 10 m s⁻¹; and/or (3) when the standard deviation on September average total wave power at 60°S and 20 hPa, derived from geopotential height fields, exceeds 300 m.

[21] The two methods (subtraction of the pre-ozone-hole background from vortex average total column ozone and the tracer-tracer correlation method) provide very similar measures of Antarctic ozone loss in spite of the fact that they cover different altitude ranges (suggesting that ozone loss above or below the region diagnosed with the tracer-tracer correlation method is negligible) and that the tracer-tracer correlation measurement samples only a few locations within the vortex (suggesting that the air within the core of the vortex is well mixed). The large discrepancy in 2002 is



Figure 4. Daily values of OHA_{vortex} (thin black line), OHA_{62S} (thin grey line) and OHA_{220DU} (thick black line) for 2003. The arrow early in the period shows the first day of sunlight on PSC areas in 2003. The error bars on the OHA_{vortex} and OHA_{62S} curves indicate the uncertainties on these measures, resulting from uncertainty in the pre-ozone-hole background (see Figure 1), which decrease throughout the season: by June the error is more than $1 \times 10^{13} \text{ m}^2$.

Table 2. Percentage Error in OMD and OHA When Calculating These Quantities Poleward of 62°S Rather Than Poleward of the Inner Vortex Edge

Period	OMD _{62S} /OMD _{vortex} 100%	OHA62S/OHAvortex 100%
June	162.9	117.9
July	110.6	107.8
August	95.3	95.8
September	72.9	83.9
October	62.6	61.9
November	70.1	67.0

not surprising since in that year a midwinter major warming [*Hoppel et al.*, 2003; *Sinnhuber et al.*, 2003] caused a significant influx of ozone rich air into the vortex. The underestimation of the ozone loss in 2002 and 2004 using the total column ozone loss method suggests that dynamical resupply of ozone to the Antarctic vortex may have become more prevalent in recent years. While no major stratospheric warming was observed in 2004, vortex minimum temperatures were higher than in the previous decade, and winter-

time PSC frequencies were significantly lower than in the previous decade [*Hoppel et al.*, 2005]. Furthermore, our analysis of wave powers in geopotential height fields from 1979 to 2004, showed that the standard deviation on September wave powers of wave number 5 were anomalously high in 2004.

[22] In Figure 6 the relationship between the two different vortex ozone depletion measures is displayed as a scatter diagram. Except for 2002 and 2004, the two ozone loss measures are correlated with a correlation coefficient of r = 0.6 at 99.9% significance (when 2002 and 2004 are excluded), with the total column ozone loss method showing smaller ozone loss as expected since it considers both the effects of chemical loss and dynamical resupply.

[23] A day-by-day comparison of total column ozone loss and accumulated chemical ozone loss for 30 October to 13 December 1996 and 1 June to 20 October 2003 is displayed in Figures 7 and 8, respectively. The accumulated chemical ozone loss for these periods is based on ILAS and ILAS-II observations [e.g., *Tilmes et al.*, 2006b]. In this



Figure 5. HALOE chemical ozone loss (black stars) compared to vortex average total column ozone loss (thick grey line) for 1992–2004. Uncertainties in calculating the accumulated chemical ozone loss (error bars on black stars) result from the estimation of the reference function as well as averaging over the profiles inside the vortex, whereas the uncertainties in calculating the vortex average ozone loss have been estimate the uncertainties on the pre-ozone-hole background (thin grey lines).



Figure 6. Comparison of vortex average chemical ozone losses derived with two different methods for 1992–2004. Different symbols stand for different years.

comparison, two different total column ozone loss time series, resulting from integration over the two regions described in section 2.1 are considered. In 1996 ILAS data are only available for late in the season. Good agreement between the accumulated chemical ozone loss and the total column ozone loss are observed when averaging the total column ozone loss within the vortex boundaries (Figure 7). From 6 December 1996 onward larger deviations occur. As was the case for OMD_{vortex} and OMD_{62S} (Figure 3), the ozone loss integrated within the vortex is greater than that integrated poleward of 62° S from October onward.

[24] In 2003 ILAS-II data are available for the whole Antarctic vortex period. High correlations between accumulated chemical ozone loss and vortex average total column ozone loss are observed from the start of the ozone hole until the minimum is reached (Figure 8). The day-today variability of accumulated chemical ozone loss and total column ozone loss (both within the vortex and poleward of 62°S) are similar. Comparing the accumulated chemical ozone loss and the vortex average total column ozone loss, some deviation occurs after the minimum value is reached whereas the average poleward of 62°S starts deviating at the beginning of September and shows greater deviations. Therefore, if ozone loss only until early September is considered, ozone loss poleward of 62°S (the simpler diagnostic) provides the same accuracy as ozone loss within the vortex. Vortex average total column ozone loss reaches its minimum at about 20 DU higher than the accumulated chemical ozone loss. The date when the minimum is reached is in both cases around 9 October 2003. The differences could be due to different altitude ranges considered for the two measures, filaments from lower latitudes intruding into the vortex and/or mixing of ozone rich air from the top of the vortex. Since the tracer-tracer correlation requires an impenetrable vortex edge, filaments from lower latitudes and mixing of ozone rich air from the top of the vortex are not accounted for. In Figure 2 it can be seen that there is a slight increase in total ozone in the vortex core after about 4 October 2 his increase is most probably



Figure 7. Chemical ozone loss derived with tracer-tracer correlations and total column ozone loss derived with the new method displayed for October to December 1996 (thick black line indicates ILAS ozone loss, dotted lines indicate uncertainties on ILAS ozone loss, thin black line indicates total column ozone loss (inside vortex), and grey line indicates total column ozone loss (poleward of 62°S)). Two single error bars on the vortex average total column ozone loss lines indicate the uncertainties on these measures.

due to mixing of polar air masses with ozone rich air from lower latitudes at high altitudes.

4. Discussion and Conclusions

[25] New definitions for the Antarctic ozone mass deficit (OMD) and Antarctic ozone hole area (OHA) have been



Figure 8. Chemical ozone loss derived with tracer-tracer correlations and total column ozone loss for 2003 (thick black line indicates ILAS-II ozone loss, dotted lines indicate uncertainties on ILAS-II ozone loss, thin black line indicates total column ozone loss (inside vortex), and grey line indicates total column ozone loss (poleward of 62°S)). Two single error bars on the vortex average total column ozone loss lines indicate the uncertainties on these measures.



Figure 9. Comparison of the new (thin black line) and 220 DU based (thin grey line) ozone mass deficits averaged over the Antarctic vortex period (18 July to 30 November) for each year from 1979 to 2004. EEASC (thick black line) is plotted against the right ordinate. Both ozone mass deficit measures are plotted on the same scale, and the EEASC scale has been selected so that the EEASC overlays best with the new OMD.

presented and discussed above. The algorithm used to calculate these indices retains some of the simplicity of earlier methods (e.g., those that use 220 DU as both the unperturbed background and spatial integration limit) but has the advantage of better representing the chemical ozone loss within the vortex. This was demonstrated by comparing total column ozone loss, calculated using the pre-ozone-hole background and spatial integration limits used for the new OMD and OHA indices, with accumulated chemical ozone loss from a more complex and sophisticated tracer-tracer correlation technique. Except for unusually dynamically perturbed years the two methods showed good agreement. Diagnostics for identifying such years are presented. As noted above, because the total column ozone loss method (which uses the same assumptions as for the OHA and OMD calculations) does not distinguish between chemical and dynamical induced changes in ozone, the newly proposed Antarctic ozone depletion indices will only be representative of chemical depletion of ozone in years when dynamical contributions to ozone variability within the vortex are small. However, even in unusually dynamically perturbed years, the new measures provide improved metrics (compared to 220 DU based metrics) of the severity of Antarctic ozone depletion and for impact studies of ozone depletion on the biosphere, since the 220 DU based metrics omit large parts of ozone depletion (Figure 1).

[26] The new methods for calculating OMD and OHA require total column ozone fields and, only for the option that uses the vortex edge, potential vorticity fields. Total column ozone fields are more readily available than the ozone and passive tracer profiles required for the tracer-tracer correlation method, and therefore accurate measures of Antarctic ozone depletion can be obtained over longer time periods. Thus the recommendation is that the former method for calculating OMD should be replaced with the new method that integrates depleted ozone mass poleward

of the inner vortex edge from a pre-ozone-hole background derived with the regression coefficients listed in Table 1. We acknowledge that this method is somewhat more complicated than the former method in that it requires calculation of the dynamical vortex edge. If the additional complication of calculating the dynamical vortex edge is avoided and the OMD is integrated poleward of 62° S (the average latitude of the vortex edge), the resultant errors in the ozone mass deficit (and OHA) are listed in Table 2. Figure 9 is an updated version of *Huck et al.* [2005, Figure 1] showing the interannual variability and long-term trend in OMD_{vortex} compared to that based on the 220 DU threshold. While the long-term trend follows the chlorine and bromine loading of the stratosphere, strong interannual variability is seen in recent years.

[27] The new OHA defined over regions within the vortex and where ozone is below the pre-ozone-hole background has also shown an improvement when compared with the OHA defined within the 220 DU contour. In particular, the new version of OHA (and the new version of OMD) shows closer correspondence with the timing of the return of sunlight to the vortex and the onset of ozone depletion.

[28] A further advantage of using the pre-ozone-hole background is that the effect of the annually repeating cycle in dynamics on ozone is better captured than when a constant background is used. This leads to better separation of the effects of chemistry and dynamics in a specific year from ozone changes driven by the long-term background meridional transport that has affected Antarctic ozone over climatological time periods. However, interannual variability due to the quasi-biennial oscillation, solar cycle or volcanic eruptions is not currently included in the preozone-hole background. To correct for these effects, an offset, derived in May when the vortex has formed but before ozone depletion has started, is subtracted from the Antarctic ozone loss measures.

[29] These new definitions of OHA and OMD should also aid chemistry-climate model intercomparisons. The new measures will be less sensitive to ozone bias since the pre-ozone-hole background from which the OMD and OHA are calculated, will be specific to each model and will result in some cancelation of any ozone bias.

References

- Austin, J., and N. Austin (2003), Coupled chemistry-climate model simulations for the period 1980–2020: Ozone depletion and the start of the ozone recovery, Q. J. R. Meteorol. Soc., 129, 3225–3249.
- Bodeker, G. E., J. C. Scott, K. Kreher, and R. L. McKenzie (2001), Global ozone trends in potential vorticity coordinates using TOMS and GOME

^[30] Acknowledgments. P. Huck is supported through funding by the DAAD (Doktorandenstipendium). NCEP Reanalysis data were provided by the NOAA-CIRES Climate Diagnostics Center, Boulder, Colorado, USA, from their web site at http://www.cdc.noaa.gov/. We gratefully acknowledge all members of the HALOE team at NASA Langley and all members of the science team of the Improved Limb Atmospheric Spectrometer (ILAS and ILAS-II), in particular Y. Sasano, for processing ILAS (V6.0) and ILAS-II (V1.4) data. ILAS and ILAS-II were developed by the Ministry of the Environment, Japan (MOE), and were aboard the ADEOS and ADEOS-II satellite launched by the Japan Aerospace Exploration Agency (JAXA). ILAS and ILAS-II data were processed at the ILAS and ILAS-II Data Handling Facility, National Institute for Environmental Studies (NIES). We would also like to thank P. Newman for providing the EEASC data. We would also like to thank S. George for reading the manuscript and giving helpful suggestions.

intercompared against the Dobson network: 1978-1998, J. Geophys. Res., 106(D19), 23,029-23,042.

- Bodeker, G. E., H. Struthers, and B. J. Connor (2002), Dynamical containment of Antarctic ozone depletion, Geophys. Res. Lett., 29(7), 1098, doi:10.1029/2001GL014206.
- Bodeker, G. E., H. Shiona, and H. Eskes (2005), Indicators of Antarctic ozone depletion, Atmos. Chem. Phys., 5, 2603–2615.
- Chandra, S., and R. D. McPeters (1986), Some observations on the role of planetary waves in determining the spring time ozone distribution in the Antarctic, Geophys. Res. Lett., 13(12), 1224-1227.
- Eyring, V., et al. (2006), Assessment of temperature, trace species and ozone in chemistry-climate model simulations of the recent past, J. Geophys. Res., 111, D22308, doi:10.1029/2006JD007327.
- Farman, J. C., R. J. Murgatroyd, A. M. Silnickas, and B. A. Thrush (1985), Ozone photochemistry in the Antarctic stratosphere in summer, Q. J. R. Meteorol. Soc., 111, 1013-1028.
- Hoppel, K., R. Bavilacqua, D. R. Allen, G. E. Nedoluha, and C. Randall (2003), POAM III observations of the anomalous 2002 Antarctic ozone hole, Geophys. Res. Lett., 30(7), 1394, doi:10.1029/2003GL016899.
- Hoppel, K., G. Nedoluha, M. Fromm, D. Allen, R. Bevilacqua, J. Alfred, B. Johnson, and G. König-Langlo (2005), Reduced ozone loss at the upper edge of the Antarctic Ozone Hole during 2001-2004, Geophys. Res. Lett., 32, L20816, doi:10.1029/2005GL023968.
- Huck, P. E., A. J. McDonald, G. E. Bodeker, and H. Struthers (2005), Interannual variability in Antarctic ozone depletion controlled by planetary waves and polar temperature, Geophys. Res. Lett., 32(13), L13819, doi:10.1029/2005GL022943.
- Müller, R., U. Schmidt, A. Engel, D. S. McKenna, and M. H. Proffitt (2001), The O₃/N₂O relation from balloon-borne observations as a measure of Arctic ozone loss in 1991/92, Q. J. R. Meteorol. Soc., Part B, 127(574), 1389-1412.
- Müller, R., S. Tilmes, P. Konopka, J. U. Grooß, and H. J. Jost (2005), Impact of mixing and chemical change on ozone-tracer relations in the polar vortex, Atmos. Chem. Phys., 5, 3139-3151.
- Nakajima, H. (2006), Preface to special section on ILAS-II: The Improved Limb Atmospheric Spectrometer-II, J. Geophys. Res., 111, D20S90, doi:10.1029/2006JD007412.
- Nakajima, H., et al. (2006), Characteristics and performance of the Improved Limb Atmospheric Spectrometer-II (ILAS-II) onboard the ADEOS-II satellite, J. Geophys. Res., 111, D11S01, doi:10.1029/ 2005JD006334
- Nash, E. R., P. A. Newman, J. E. Rosenfield, and M. R. Schoeberl (1996), An objective determination of the polar vortex using Ertel's potential vorticity, J. Geophys. Res., 101(D5), 9471–9478. Newman, P. A., and E. R. Nash (2005), The unusual Southern Hemisphere
- stratosphere winter of 2002, J. Atmos. Sci., 62(3), 614-628.
- Newman, P. A., E. R. Nash, S. R. Kawa, S. A. Montzka, and S. M. Schauffler (2006), When will the Antarctic ozone hole recover?, Geophys. Res. Lett., 33, L12814, doi:10.1029/2005GL025232.
- Proffitt, M. H., J. J. Margitan, K. K. Kelly, M. Loewenstein, J. R. Podolske, and K. R. Chan (1990), Ozone loss in the Arctic polar vortex inferred from high-altitude aircraft measurements, Nature, 347(6288), 31-36.

- Russell, J. M., III, L. Gordley, J. Park, S. R. Drayson, A. F. Tuck, J. Harries, R. J. Cicerone, P. J. Crutzen, and J. E. Frederick (1993), The Halogen Occultation Experiment, J. Geophys. Res., 98, 10,777-10,797
- Sasano, Y., M. Suzuki, T. Yokota, and H. Kanzawa (1999), Improved limb atmospheric spectrometer (ILAS) for stratospheric ozone layer measurements by solar occultation technique, J. Geophys. Res., 26, 197-200.
- Schoeberl, M. R., and D. L. Hartmann (1991), The dynamics of the stratospheric polar vortex and is relation to springtime ozone depletions, Science, 251, 46-52.
- Sinnhuber, B.-M., M. Weber, A. Amankwah, and J. Burrows (2003), Total ozone during the unusual Antarctic winter of 2002, Geophys. Res. Lett., 30(11), 1580, doi:10.1029/2002GL016798.
- Solomon, S., J. P. Smith, R. W. Sanders, L. Perliski, H. L. Miller, G. H. Mount, J. G. Keys, and A. L. Schmeltekopf (1993), Visible and nearultraviolet spectroscopy at McMurdo Station, Antarctica 8. Observations of nighttime NO2 and NO3 from April to October, 1991, J. Geophys. Res., 98(D1), 993-1000.
- Stolarski, R., M. Schoeberl, P. Newman, R. McPeters, and A. Krueger (1990), The 1989 Antarctic ozone hole as observed by TOMS, Geophys. Res. Lett., 17(9), 1267-1270.
- Suzuki, M. A., A. Matsuzaki, T. Ishigaki, N. Kimura, N. Araki, T. Yokota, and Y. Sasano (1995), ILAS, the Improved Limb Atmospheric Spectrometer, on the advanced Earth Observing Satellite, IEICE TRANS Commun, E78-B12, 1560-1570.
- Tilmes, S., R. Mller, J.-U. Groo, R. Spang, T. Sugita, H. Nakajima, and Y. Sasano (2006a), Chemical ozone loss and related processes in the Antarctic winter 2003 based on Improved Limb Atmospheric Spectrometer (ILAS)-II observations, J. Geophys. Res., 111, D11S12, doi:10.1029/2005JD006260.
- Tilmes, S., R. Müller, J.-U. Grooß, R. Spand, T. Sugita, H. Nakajima, and Y. Sasano (2006b), Chemical ozone loss and related processes in the Antarctic winter 2003 based on ILAS-II observations, J. Geophys. Res., 111, D11S12, doi:10.1029/2005JD006260.
- Uchino, O., R. D. Bojkov, D. S. Balis, K. Akagi, M. Hayashi, and R. Kajihara (1999), Essential charactersistics of the Antarctic-spring ozone decline: Update to 1998, Geophys. Res. Lett., 26(10), 1377-1380.
- World Meteorological Organization (2003), Scientific Assessment of Ozone Depletion: 2002, Global Ozone Res. and Monit. Proj. Rep. 47, Geneva, Switzerland.

H. Nakajima, National Institute for Environmental Studies, Tsukuba 305-8506, Japan. (hide@nies.go.jp)

W. J. Randel and S. Tilmes, National Center of Atmospheric Research, Boulder, CO 80307, USA. (randel@ucar.edu; tilmes@ucar.edu)

G. E. Bodeker, National Institute of Water and Atmospheric Research, Lauder, Private Bag 50061, Central Otago 9182, New Zealand. (g.bodeker@niwa.co.nz)

P. E. Huck and A. J. McDonald, Department of Physics and Astronomy, University of Canterbury, Private Bag 4800, Christchurch 8020, New Zealand. (peh26@student.canterbury.ac.nz; adrian.mcdonald@canterbury. ac.nz)