Ozone loss in the Arctic polar vortex inferred from high-altitude aircraft measurements

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The Arctic polar vortex in winter is known to be chemically primed for ozone depletion, yet it does not exhibit the large seasonal ozone decrease that characterizes its southern counterpart. This difference may be due in part to a net flux of ozone-rich air through the Arctic vortex, which can mask ozone loss. But by using a chemically conserved tracer as a reference, significant ozone loss can be identified. This loss is found to be correlated with high levels of chlorine monoxide, suggesting that much of the decrease in ozone is caused by anthropogenic emissions of chlorofluorocarbons.

IN 1985 Farman *et al.*[1] reported that spring values of total column ozone over Halley Bay, Antarctica, had decreased considerably during the previous decade, and they suggested that certain chlorine-containing compounds could be causing the observed ozone depletion. Further studies have supported these findings[2-5] and refined understanding of high-latitude ozone loss.

During the 1987 Airborne Antarctic Ozone Experiment (AAOE)[5] *in situ* measurements of many chemical species and meteorological parameters were made from a NASA high-altitude aircraft (ER-2) on twelve flights out of Punta Arenas, Chile (53° S), at cruise altitudes of ~17–19 km. *In situ* measurements of ozone (O_3)[6], nitrous oxide (N_2O—a chemically conserved tracer)[7], chlorine monoxide (ClO)[8], temperature, pressure and wind speed[9] were made.

Potential temperature (Θ) is a measure of entropy and is calculated from measured pressure and temperature. In the absence of diabatic processes, parcels tend to move on constant Θ surfaces and mix with other parcels on that surface. Because Θ increases with altitude, and air movement is stratified somewhat by Θ, we use it here in place of altitude as a vertical coordinate. Using Θ, diabatic processes (net heating or cooling) can be more easily identified. The aircraft flights were usually to 72° S on surfaces of constant Θ (isentropic). AAOE lasted from 17 August to 22 September, covering late winter and early spring in the Southern Hemisphere. It was found that the O_3 mixing ratio at 72° S decreased by 61% at $\Theta = 425$ K (17 km) over a 30-day period[6] and was precisely collocated with very high ClO mixing ratios[10]. Calculations indicated that the observed concentration of ClO was high enough to link the O_3 decrease to catalytic destruction by chlorine[11-13]. These calculations, however, included two important assumptions: that diabatic cooling was negligible and that the vortex air was not being replenished by O_3-rich air from outside the polar jet. This view of the polar jet as a 'containment vessel' is supported in the analyses of some authors[1,14-16] but not by others[17-22].

Possible Arctic ozone loss has also been reported recently. Balloon data taken during 1989 suggest that ozone depletion occurred late in January at an altitude of 22–26 km over Kiruna, Sweden (68° N)[23], and that there was a depleted ozone layer from 18 to 24 km from 24 January to 22 February over Alert, Canada (82° N)[24]. Another indication of possible ozone loss in the Arctic winter was found by the Ozone Trends Panel[25], where a comparison of total column ozone data from 1979–80 with 1986–87 shows a decrease of 4–10% between 65° and 80° N during the period from November to February.

An aircraft campaign, similar to AAOE, the Airborne Arctic Stratospheric Expedition (AASE) was based at Stavanger, Norway (59° N) from 3 January to 10 February 1989, to study ozone destruction mechanisms during the Arctic mid-winter[26]. The instrumentation used on the 14 ER-2 flights was virtually identical to that used in the Antarctic, but the aircraft flew nearer the pole in the Arctic, usually to 79° N. Although the Antarctic vortex persists well into spring, the less stable Arctic vortex usually dissipates in late winter and so the Arctic measurement campaign had to be conducted earlier in the season. Large photochemical ozone loss was not expected during this period of minimum solar radiation and, as anticipated, no large isentropic temporal decrease in ozone was found in the Arctic.

The analysis we present is based on a moving coordinate system defined vertically by Θ and horizontally by latitude referenced to the vortex boundary, which was defined as the latitude of peak wind speed as measured from the aircraft. The average latitude of the boundary is 68° N with a standard deviation of 3°. For the analysis that follows, the data will be considered in three regions: the vortex exterior (from aircraft take-off at 59° N to the boundary), the outer vortex (the first 8° of latitude inside the vortex boundary, comprising about 60% of the vortex volume) and the inner vortex (the remainder of the vortex, usually including the pole). The outer vortex receives more solar radiation than the inner vortex, and so photochemical ozone loss is more likely to occur there.

An analysis similar to that used to show that there is ozone loss outside the Antarctic ozone hole[27] will be used to show that there is also significant ozone loss in the Arctic vortex. Fundamental to the arguments presented is that standard gas-phase photochemistry identifies the primary source region for O_3 and the loss region for N_2O as the tropical middle stratosphere and also predicts long photochemical lifetimes for N_2O and O_3 (more than a year) for high-latitude winter[28-31]. From this, we deduce that air parcels at high latitudes with the same N_2O mixing ratios should have very similar O_3 mixing ratios. Deviations from this behaviour thus indicate chemical ozone loss occurring by processes not included in standard gas-phase photochemistry models[32].

In the following section, O_3 and N_2O data are presented and their relative behaviour during the mission characterized. In the section that follows, we argue that the vortex behaves as a 'flowing processor' and these data are evaluated in this context, and an apparent O_3 loss is identified within the vortex. The origin of the loss is shown to be chemical rather than dynamical, to be enhanced in the outer vortex, and to have occurred both during and before the mission. The O_3 and N_2O data taken outside the vortex where O_3 loss is expected to be small are compared with values found within the vortex where an O_3 loss

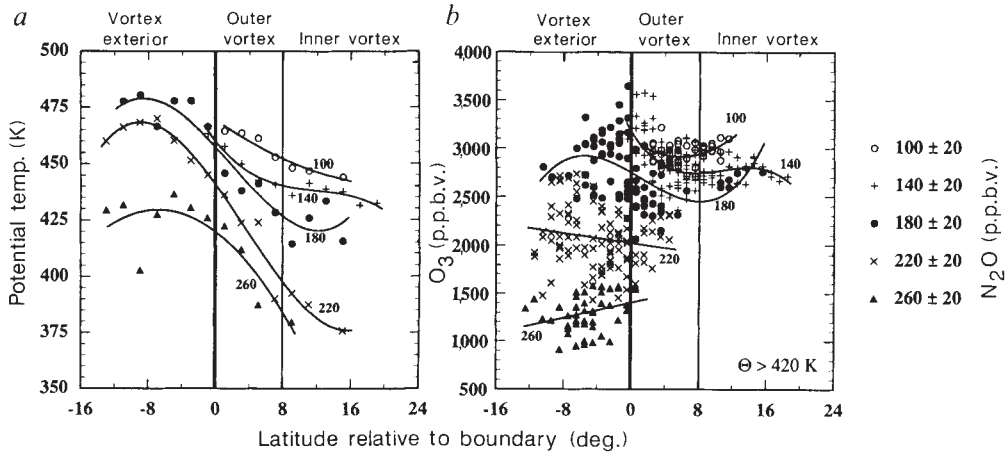

FIG. 2 *a*, N_2O isopleths on a coordinate system of Θ plotted against latitude relative to the boundary of the vortex. Data from the entire mission are averaged over 1° of latitude relative to the vortex boundary and plotted along with polynomial fits for each of the N_2O bins. *b*, All O_3 data with Θ > 420 K are plotted against latitude relative to the boundary and binned by N_2O. Data are averages over 100-s intervals (about 10′ of latitude). A fit is provided for each bin.

fit is shown for the 410 K bin with quadratic fits for the three middle bins. The highest bin (490 K) has too few data to justify a fit. Figure 1*d* shows data from the inner vortex. Comparing Fig. 1*b–d*, the fits for the 410 K bin are all very similar, but there is little similarity in the fits to the other Θ bins. The uniformity below 420 K indicates that air is free to move from inside to outside the vortex at this level, suggesting that the 'bottom of the vortex' is where Θ = 410 ± 10 K.

Another important criterion for comparing Fig. 1*b–d*, and indicative of chemical loss of O_3, is the deviation of O_3 mixing ratios relative to N_2O from the mid-latitude values shown in Fig. 1*a*. It is clear that for a constant value of N_2O, O_3 progressively decreases from the mid-latitudes, to the vortex exterior, to the outer vortex. In Fig. 1*b*, *c*, O_3 also decreases with decreasing Θ. The characteristic of decreasing O_3 with decreasing Θ for a constant value of N_2O will be referred to as 'O_3–Θ dependence'. There is a small O_3–Θ dependence in the vortex exterior and a larger one in the outer vortex, but none is evident in the mid-latitude data, nor in the inner vortex. The O_3–Θ dependence in Fig. 1*b*, *c* is not a result of downward mixing of air from above the O_3 maximum because O_3 and Θ are negatively correlated above the maximum, whereas the O_3–Θ dependence in Fig. 1*b*, *c* is positive. This implies that the O_3–Θ dependence is not a result of dynamics, but of chemistry.

This O_3–Θ dependence was also clearly seen in the Antarctic exterior (Fig. 1*e*) and seems much like the Arctic outer vortex (Fig. 1*c*). Also given in Fig. 1*e* is the Antarctic reference that was used for assessing O_3 loss[27]. That reference was based on a more restricted data set than is available for the Arctic, and may underestimate O_3 loss because it was chosen very conservatively.

Isopleths for N_2O (lines of constant N_2O) within our Θ-latitude coordinate system are shown in Fig. 2*a*. Each point is an average over 1° of latitude and over all flights of the mission. Polynomial fits are provided for each of the N_2O bins and are good approximations to the N_2O isopleths. The poleward decrease in Θ along the isopleths indicates diabatic cooling within the vortex. Diabatic cooling rates during the mission have been approximated theoretically[34] and from AASE N_2O data[35]. Although these analyses do not necessarily represent average vortex conditions, they do imply that cooling of at least 0.9 K per day in Θ (0.4 K per day in temperature) occurred during the mission both inside and outside the vortex.

Figure 2*b* confirms the O_3 loss seen in Fig. 1*a–d* but in a spatial context, by plotting O_3 against latitude relative to the vortex boundary for the same five N_2O bins of Fig. 2*a*. Only data above 420 K are plotted, to exclude data that may be affected by the relatively unrestricted horizontal exchange of air at the bottom of the vortex. Again polynomial fits are included for each of the N_2O bins and terminated where the data end. In the outer vortex, O_3 decreases towards the pole, and where there are data within the inner vortex, the fits there either increase or flatten out indicating less ozone loss. The outer vortex receives more solar radiation, and so the observed spatially enhanced decrease is consistent with O_3 being photochemically destroyed.

O_3 loss in a flowing processor

Early research indicated that tropospheric air enters the lower stratosphere in the tropics and exits at higher latitudes[36,37] and this is still generally accepted[30]. That is, the dominant atmospheric circulation is upward at low latitudes where heating lifts air into the region of the stratosphere where ozone is produced, then poleward and at high latitudes downward, due to diabatic cooling. As previously mentioned, some authors believe that the Antarctic jet effectively terminates the poleward component, but only the question of Arctic containment will be discussed here.

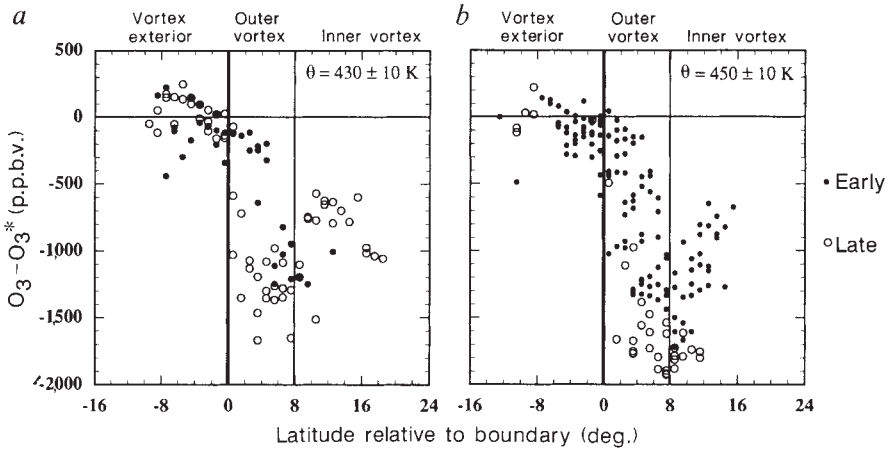

FIG. 3 $O_3 - O_3^*$ plotted against latitude relative to the vortex boundary for the early data (before 30 January) and the late data (the remainder of the mission), each point representing, from a flight leg, the average over 1° of latitude relative to the boundary. Negative values indicate loss of O_3 relative to the vortex exterior. *a*, Includes only those points with Θ = 430 ± 10 K. *b*, Includes only those points with Θ = 450 ± 10 K.

discussed earlier[41] shows a column increase of 12–20% per month from December to February, but also shows that the column decrease reaches a maximum in May at 12% per month, decreasing almost linearly to no change in October. Assuming the column loss in summer is representative of chemical O_3 loss, we conclude that a summer loss of O_3, terminating months before AASE, is not likely to contribute substantially to the loss reported here. But to quantify its possible effect requires very accurate O_3 and N_2O data measured simultaneously throughout the polar lower stratosphere in autumn and winter, and no such data are available.

Returning to the O_3–Θ dependence, a plausible explanation for the low O_3 and its dependence on Θ can be constructed in the context of a 'flowing processor'. First, recall that air is diabatically cooling both inside and outside the vortex, that N_2O is chemically inert in the high latitude winter, and that mixing ratios do not change due to expansion and contraction. Therefore, if mixing of air parcels of differing N_2O content is ignored, the movement of a parcel that is diabatically cooling must be restricted to a surface described by the N_2O isopleth on which that parcel resides. This can be visualized with or without containment of the lower vortex. Consider an air parcel initially a few degrees of latitude outside the vortex that is diabatically cooling. The parcel remains on a surface of constant N_2O, so as Θ decreases, the parcel moves poleward and downward into the vortex (see Fig. 2*a*) thus describing a 'flow' into the vortex in the lower stratosphere. Alternatively, if the lower vortex is assumed to be contained by the polar jet, the cooling would be characterized as a change in N_2O on a surface of constant potential temperature, Θ (ref. 35). In this case the flow is vertical and downward as the isopleths sink. But in both cases, there is flow through the surfaces of constant Θ within the vortex in the lower stratosphere.

Let us hypothesize that by some chemical 'process', as an air parcel diabatically cools, O_3 is lost, and see if the resulting 'flowing processor' would produce the observed data. Figure 2*b* shows O_3 decreasing poleward along the N_2O isopleths, in the outer vortex, but not in the inner vortex. That is, the O_3 loss seems to be less in the inner vortex. Therefore our cooling/O_3-loss hypothesis, together with an enhanced loss in the outer vortex (where there is greater solar radiation), is consistent with Fig. 2, with or without mass flux into the lower vortex.

Similar reasoning can also be applied to Fig. 1*b*–*d* to again test our expanded hypothesis, this time assuming that there is mass flux into the lower vortex. Consider an air parcel with, for example, $N_2O = 175$ p.p.b.v. originally outside the boundary at $\Theta = 470$ K and $O_3 = 3{,}050$ p.p.b.v. (Fig. 1*b*). Later that parcel contains only 2,400 p.p.b.v. of O_3 when $\Theta = 430$ K inside the boundary (Fig. 1*c*), representing a 20% O_3 loss. But the inner vortex O_3 at 430 K is 2,700 p.p.b.v. and, as previously noted, shows no O_3–Θ dependence (Fig. 1*d*). Again, these data indicate that increased exposure to solar radiation in the outer vortex enhances the O_3 loss. The lack of O_3–Θ dependence in the inner vortex could be a result of enhanced isentropic mixing in that region and the low O_3 may be due to transport from the outer vortex where the loss is primarily occurring. The summertime O_3 loss discussed earlier could also explain part of the inner vortex loss.

As discussed above, air parcels at high latitudes with the same N_2O mixing ratios would be expected to have very similar wintertime O_3 mixing ratios. This was deduced without requiring the somewhat controversial assumption of significant mass flux into the vortex in the lower stratosphere. How accurately we can quantify O_3 loss from N_2O depends on how well the O_3 and N_2O reference relationship is known, and this is related to the altitude of mass flux. That is, a significant flux across the boundary at or near flight altitudes implies that air sampled inside the boundary recently entered the vortex, carrying with it the measured characteristics of the exterior. A path higher in the stratosphere requires the assumption that the O_3–N_2O reference relationship is preserved at altitudes above the region sampled by the aircraft, and therefore implies greater uncertainty in the amount of O_3 loss. Figure 1*a* supports this assumption.

To quantify O_3 loss within the vortex, a linear least-squares fit to all the exterior data (Fig. 1*b*) is calculated, excluding those points at the bottom of the vortex where $\Theta < 420$ K. This fit is shown in Fig. 1*c*, *d* and is given by

$$O_3^*(\text{p.p.b.v.}) = 7{,}019 - 22.42 \times N_2O\ (\text{p.p.b.v.})$$

where O_3^* approximates the O_3 mixing ratio in the vortex exterior from the mixing ratio of N_2O. For any sampled parcel inside the vortex, O_3^* can be calculated from its N_2O content with a negative value for $O_3 - O_3^*$ representing loss relative to the vortex exterior. The mid-latitude O_3–N_2O fit (Fig. 1*a*) is not used as a reference, instead we use the data in the vortex exterior for the comparison. For the reasons discussed in this section, $O_3 - O_3^*$ may overestimate O_3 loss within the vortex.

Figure 3 shows $O_3 - O_3^*$ plotted against latitude relative to the vortex boundary for $\Theta = 430 \pm 10$ K and 450 ± 10 K. Loss in excess of 1,500 p.p.b.v. is found in the outer vortex with about two-thirds of that loss in the inner vortex. The data are also binned into early (before 30 January) and late data (the remainder). There are small temporal changes seen in the outer vortex, although Fig. 3*a* includes virtually no data from the first half of January because early flight legs were above $\Theta = 440$ K. Temporal changes are better represented in Fig. 3*b*, showing a decrease of ~500 p.p.b.v. This isentropic decrease in O_3 is a result of the temporal changes in N_2O, not absolute O_3 changes. Therefore, isentropic temporal analysis of O_3 alone will not reveal the true temporal decrease.

An O_3 decrease of 13 ± 8 p.p.b.v. per day has been reported by others[35] also using AASE data. The analysis was restricted to a few vertical profiles within the vortex and required a diabatic cooling rate estimated from N_2O and an assumption of strictly vertical descent. If the same cooling rates (0.9 K per day in Θ) are applied to the fits in Fig. 1*c* and no account is made for O_3 flux into the vortex, loss rates of from 9 p.p.b.v. per day where $N_2O = 210$ p.p.b.v. to 24 p.p.b.v. per day where $N_2O = 130$ p.p.b.v. are obtained.

As O_3 is low within the vortex early in the mission, a result of O_3 flux from above or from the exterior of the vortex without concurrent O_3 loss would be increased O_3 relative to N_2O; that is, the interior O_3–N_2O relationship would appear increasingly similar to the exterior relationship. This was not observed, again implying, as found in the discussion on summertime loss, that O_3 was chemically destroyed during the mission. From Fig. 3, O_3 loss had occurred by January. Further analysis of individual flights reveals that, on the first flight, 3 January, ~1,000 p.p.b.v. of O_3 had been lost at 465 K; that is, substantial O_3 loss had occurred before the mission.

Our analysis of the flowing processor still leaves an important question unanswered: 'What is the cause of the O_3 loss?'. As previously mentioned, the cause of the Antarctic O_3 loss has been attributed to catalytic destruction by chlorine, and, to a lesser degree, bromine. Model calculations for the Arctic based on measured ClO and BrO suggest that O_3 loss approached 12% (about 12 p.p.b.v. per day) during the Arctic campaign[46]. Coupled photochemical–microphysical lagrangian model calculations indicate that an O_3 loss of more than 20 p.p.b.v. per day could have been sustained throughout much of February[47]. Solar radiation and measured ClO increased substantially during the mission[48], so losses should be much less in early January. Therefore, the loss indicated in our analysis for early January is too large to match these model studies. But meteorological data show that temperatures were low enough to form polar stratospheric clouds at 30 mbar by late November (A. F. Tuck, personal communication), and the eccentricity of the Arctic vortex is sufficient to frequently carry outer vortex air into sunlit regions. These episodic conditions may be sufficient to produce a significant O_3 loss at 30 mbar during late November and December.

Diabatic cooling could bring this somewhat depleted air to ER-2 flight altitudes by early January. Summertime O_3 loss might also have some residual effect.

One of the striking results in the Antarctic campaign was the observed rapid decrease in O_3 and simultaneous increase in ClO as the aircraft crossed into the ozone hole along an isentropic flight leg[10]. On 9 February 1989, a similar but less dramatic isentropic O_3 decrease was observed poleward of the Arctic boundary[49] that was coincident with very high ClO and is shown in Fig. 4*a*, *b*. Figure 4*a* shows decreasing O_3 poleward of the vortex boundary that is collocated with decreasing N_2O. This positive correlation between O_3 and N_2O inside is contrasted by the usual negative correlation seen outside the boundary, implying O_3 loss within the vortex. Figure 4*b* shows no O_3 loss outside the boundary, substantial loss inside (implied by negative values for $O_3 - O_3^*$), and that the loss occurs precisely where ClO increases dramatically. Figure 4*c* demonstrates that this correlation between O_3 loss and ClO in the outer vortex persists throughout the mission and is evident even with constant N_2O. In this plot of all outer vortex data, ClO is the abscissa and $O_3 - O_3^*$ the ordinate with Θ restricted to 435 ± 15 K. The data are tightly binned by N_2O value and each bin is provided with a linear fit. It is evident that O_3 loss and ClO are positively correlated over a wide range of N_2O, and the linear fits are surprisingly similar, although their slopes increase at the higher N_2O values.

Summary

Evaluating O_3 loss in the Arctic winter is complex because of our limited understanding of the effects of polar dynamics. Although there seems to be a consensus on significant diabatic cooling at high latitudes, and poleward flux into the Arctic vortex, there are uncertainties in the cooling rates and differing opinions on the altitude at which air is supplied to the vortex. Descent and poleward flux into the vortex (regardless of the altitude of the flux), along with concurrent O_3 loss together form a 'flowing processor', accepting air rich in O_3 from lower latitudes, and liberating air somewhat depleted in O_3 through the bottom of the vortex. The relative rates of O_3 loss and its resupply determine whether a signature of that loss will be evident in isentropic and column analyses. To avoid these problems we have used a chemically conserved tracer, N_2O, to infer Arctic O_3 loss, a method similar to that used to infer loss in the exterior of the Antarctic ozone hole[27].

The analysis reveals an *in situ* O_3 loss of 500–1,500 p.p.b.v. (12–35%) throughout the vortex relative to its exterior. This corresponds to a decrease in column O_3 of about 6% over the 4 km depth covered in this analysis. If this loss is of anthropogenic origin emerging during the past decade, the decrease we report is consistent with the 4–10% long-term seasonal column change found by the Ozone Trends Panel[25]. The rate of O_3 loss has been approximated by assuming vertical descent and a conservative cooling rate. Losses of 9–24 p.p.b.v. per day are obtained for parcels with N_2O from 210 p.p.b.v. to 130 p.p.b.v., respectively. These rates are comparable to but slightly higher than model calculations. Larger cooling rates and deviations from vertical descent will both increase this difference.

An O_3 loss was observed in early January that is too large to match model studies. The Arctic vortex polar asymmetry contrasts with the more symmetric Antarctic vortex, and the asymmetry could enhance early O_3 loss in the Arctic relative to the seasonally comparable period in the Antarctic. This would be due to episodic exposure to sunlight during the low-latitude excursions of the polar jet. The low values of O_3 found on 3 January indicate substantial O_3 loss before the mission and may result from this intermittent exposure.

We have found that the O_3 loss is correlated with elevated ClO in the outer vortex, along with enhancement of O_3 loss where there is maximum exposure of the vortex to sunlight. This suggests, but does not prove, that chlorine is an important factor in the ozone decrease. The agreement between this analysis and model studies is not perfect, particularly in early January. Perhaps alternative explanations of the data will emerge that do not require such large wintertime O_3 loss. In any case, it is clear that accurate evaluation of Arctic O_3 loss must account for the effects of polar dynamics. □

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