

Fundamental differences between Arctic and Antarctic ozone depletion

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Antarctic ozone depletion is associated with enhanced chlorine from anthropogenic chlorofluorocarbons and heterogeneous chemistry under cold conditions. The deep Antarctic “hole” contrasts with the generally weaker depletions observed in the warmer Arctic. An unusually cold Arctic stratospheric season occurred in 2011, raising the question of how the Arctic ozone chemistry in that year compares with others. We show that the averaged depletions near 20 km across the cold part of each pole are deeper in Antarctica than in the Arctic for all years, although 2011 Arctic values do rival those seen in less-depleted years in Antarctica. We focus not only on averages but also on extremes, to address whether or not Arctic ozone depletion can be as extreme as that observed in the Arctic. This information provides unique insights into the contrasts between Arctic and Antarctic ozone chemistry. We show that extreme Antarctic ozone minima fall to or below 0.1 parts per million by volume (ppmv) at 18 and 20 km (about 70 and 50 mbar) whereas the lowest Arctic ozone values are about 0.5 ppmv at these altitudes. At a higher altitude of 24 km (30-mbar level), no Arctic data below about 2 ppmv have been observed, including in 2011, in contrast to values more than an order of magnitude lower in Antarctica. The data show that the lowest ozone values are associated with temperatures below -80°C to -85°C depending upon altitude, and are closely associated with reduced gaseous nitric acid concentrations due to uptake and/or sedimentation in polar stratospheric cloud particles.

stratosphere | atmospheric chemistry

The extensive springtime depletion of Antarctic ozone has attracted both public and scientific interest since its discovery (1) and explanation in the 1980s. The ozone hole has been linked to the coupling of human-made chlorofluorocarbons with surface chemistry on and in polar stratospheric clouds (PSCs) that form during extreme cold conditions (2). Polar stratospheric clouds are composed of nitric acid hydrates, liquid solutions of sulfuric acid, water, and nitric acid, and (under very cold conditions) water ice (e.g., ref. 3 and citations therein). Some of the key reactions are photochemical, so that the ozone hole does not form during midwinter when the polar cap is dark, but rather in late winter/spring as sunlight returns, provided that temperatures remain low. Although the same basic chemical mechanisms operate in both hemispheres, the Arctic winter stratosphere is generally warmer than the Antarctic, and it warms up earlier in the spring. These two factors taken together explain why ozone depletion in the Arctic is generally much smaller than in the Antarctic. A particularly cold Arctic stratospheric winter and spring in 2010/2011 displayed much larger ozone depletion than typical years, as highlighted by Manney et al. (4). This noteworthy geophysical event has intrigued scientists and raised several important questions: Could this be the first Arctic ozone hole? Are Arctic ozone losses ever observed to be as extreme as those in the Antarctic? Some authors have variously characterized Arctic ozone loss in 2011 as unprecedented, an echo of the Antarctic, or on the brink of an Antarctic ozone hole (e.g., refs. 4 and 5). The unusual meteorology of this year has been explored by several studies (6–8), and a rich suite of observations of stratospheric chemical composition has been presented from

both ground-based and satellite methods (e.g., refs. 4, 7, and 9–11). Understanding how the ozone losses of the two polar regions compare is important not only to ensure a clear understanding of ozone depletion chemistry but also to accurately communicate the state of the science to the public.

Solomon et al. (12) presented ozonesonde and total ozone column data up to 2006 from stations in the Arctic and Antarctic. Here we update and expand the comparison of ozone and related chemistry over the two polar regions. Our goal is to present the observations in a manner that readily shows similarities and differences, and provides insights into chemical processes, particularly the role of polar stratospheric cloud chemistry. The data presentation should also be useful for future studies testing the ability of numerical models to fully simulate ozone depletion. Both average changes and the range of extreme values are presented, because these each provide important and distinct tests for physical and chemical understanding (just as in, for example, climate change studies).

We first examine in situ ozone observations obtained by balloonsondes at ground stations. Although limited to a few sites in each hemisphere, these are the only data that extend from the 1960s onward, before the satellite era. We next present microwave limb sounder (MLS) satellite observations (available from 2004 to present), to probe the consistency between the limited spatial sampling of the balloons from a few surface sites to the extensive coverage of the satellite and to examine how data from the MLS platform compare with the most extreme local depletions observed in situ. We will present information on the range of extreme ozone observations in individual air parcels as observed by MLS, as well as averages of the measurements over the cold polar regions. There are important limitations of such simple comparisons, and these are noted where appropriate. Stratospheric temperatures, gaseous nitric acid concentrations, and their relationship to ozone losses in the two hemispheres are also discussed to provide insights to chemical processes. We include ozonesonde data for the region near 20 km altitude (50-mbar

Significance

Fundamental differences in observed ozone depletion between the Arctic and the Antarctic are shown, clarifying distinctions between both average and extreme ozone decreases in the two hemispheres. Balloon-borne and satellite measurements in the heart of the ozone layer near 18–24 km altitude show that extreme ozone decreases often observed in the Antarctic ozone hole region have not yet been measured in the Arctic in any year, including the unusually cold Arctic spring of 2011. The data provide direct evidence that heavily depleted air contains reduced nitric acid abundances, and better quantify the roles of polar stratospheric cloud chemistry and temperatures below -80°C to -85°C in ozone destruction.

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pressure), where the largest local Arctic losses are typically found, but also present results for other pressure levels. Antarctic depletion is typically somewhat greater at lower altitudes near 15–18 km (higher pressures) but the 50-mbar level provides the best chance for the Arctic to mirror Antarctica. We also present MLS data near 18 km (70 mbar) and 24 km (30 mbar), and show that comparison of different levels aids in understanding differences in chemical processes in the two hemispheres.

Ozone Observations in the Lower Stratosphere

Stratospheric air flows largely in the zonal (east to west) direction, particularly in winter when the strong thermal gradient between the polar regions and lower latitudes leads to a circumpolar vortex. Disturbances due to atmospheric waves linked to the underlying surface topography (e.g., mountains, oceans) displace and alter the shape of the vortex, so that stations in polar regions generally sample air that is both inside and outside the vortex over the course of the winter. Therefore, long station records with frequent temporal sampling display a range from heavily depleted air, when they are located inside the vortex, to far less depleted, when they happen to be on the edge or outside the vortex.

Fig. 1 compares Antarctic ozonesonde data for the month of September to corresponding Arctic data for March since the mid-1960s. Here we have included all available historical data, including that taken in the early part of the record by less precise measurement techniques (Regener and Brewer ozonesondes) for comparison. At least for the Antarctic where ozone depletion is so large, the uncertainties in these early data do not substantially affect their usefulness in showing the marked differences in current ozone abundances compared with the preozone hole era. Results are plotted on a logarithmic scale to reveal the deepest depletions that indicate how effective the depletion chemistry can be.

Fig. 1 shows that ozone in the Antarctic decreases to extremes that are profoundly different from the Arctic, which must reflect differences in the effectiveness of chemical loss processes. Antarctic local ozone concentrations can drop to minimum values as low as about 0.02 parts per million by volume (ppmv) and even less, i.e., more than 99% decrease in some air parcels. In contrast, the figure demonstrates that the lowest ozonesonde data

obtained in the Arctic in March 2011 were about 1 ppmv at this altitude. Somewhat lower local values of about 0.5 ppmv were detected in a few observations using the more complete sampling available from satellites (see Fig. 4). The observed minima in March 2011 Arctic ozone data at 50 mbar in Fig. 1 are close to the low values obtained in several prior years (1996, 1997, 2000, 2005, and 2007). Therefore, on the basis of local ozonesonde minima covering many decades, the 2011 ozone values are low but not unprecedented (see refs. 4–6).

It is important to emphasize that transport by stratospheric winds can increase or decrease ozone along with chemistry, so that dynamical terms must be quantified if the amount of chemical ozone depletion is to be identified distinct from transport-related decreases (see, e.g., ref. 13). Recent studies by several groups suggest that unusual meteorological conditions (and associated transport of ozone) accounted for some of the apparent 2011 decreases in Arctic ozone at this level and in the total ozone column compared with typical Arctic years. One study suggested that chemical loss accounted for perhaps half of the apparent reduction in the Arctic total ozone column observed in 2011 (8), whereas another deduced only a 23% contribution from chemistry (14). The differences between these studies show that there are large uncertainties in the meteorological parameters needed to quantify chemical losses distinct from dynamical effects in the Arctic. Here we present ozone observations but do not attempt to evaluate dynamical contributions or perform Lagrangian calculations.

Fig. 2 compares the seasonal cycles of ozone and temperature data from the Arctic to the conjugate Antarctic conditions, with a shift of 180 d applied to the data to align solar illumination conditions for representative years and stations. The comparison of data in recent years to that obtained in the 1960s and 1970s illustrates the timing and impact of the remarkable seasonal ozone loss. Antarctic ozone typically begins to decline around the end of August (day 240), and reaches its minimum by late September or early October (and this season is shaded gray in the figure; see further below).

Arctic ozone began to decline in 2011 at a conjugate northern hemisphere point in the season (i.e., similar solar illumination conditions), around day 60, but the decreases are smaller and occur over a more limited period than in typical Antarctic years,

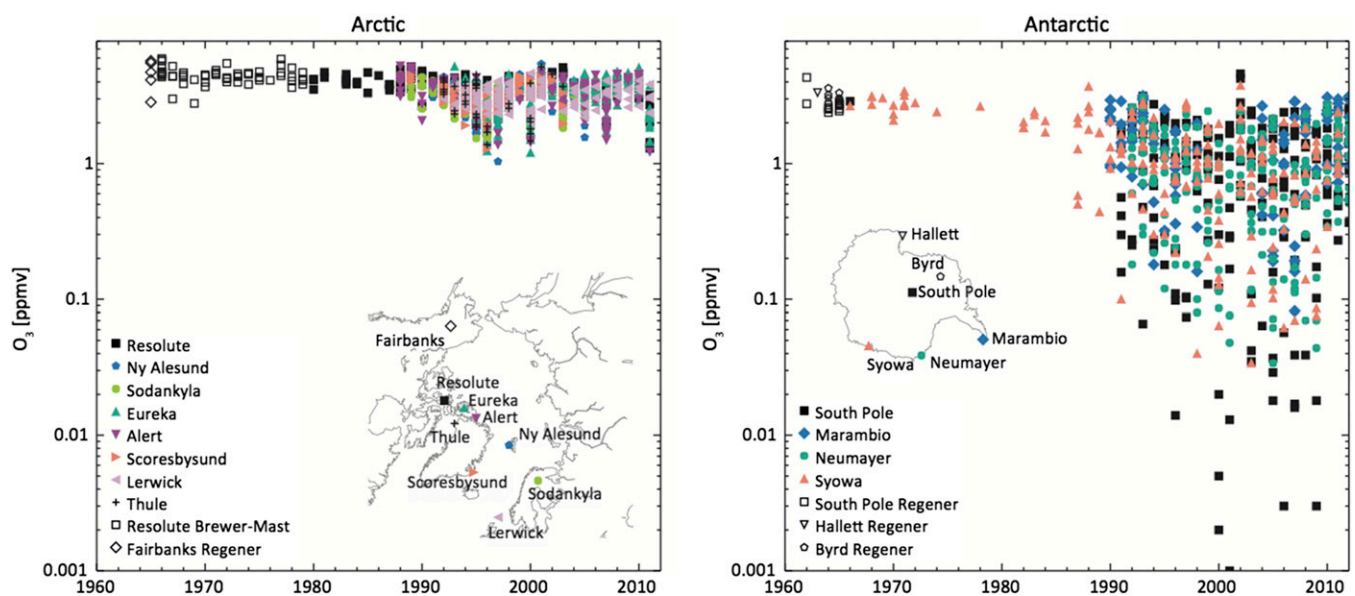


Fig. 1. Observations of the local ozone abundance at 50 mbar near the heart of the ozone layer in the Arctic in March (Left) and Antarctic in September (Right) at ozonesonde stations, in ppmv. Historical observations obtained using less precise Regener and Brewer–Mast approaches are shown using open symbols.

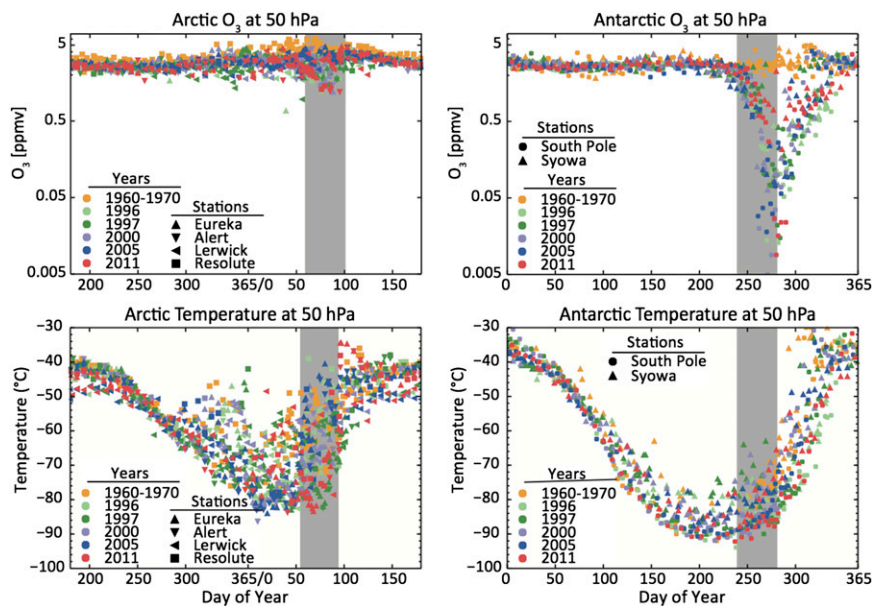


Fig. 2. (Upper) Seasonal cycles of ozone at 50 mbar from ozonesondes for representative stations and years in the Arctic (Left) and Antarctic (Right). The Arctic has been displaced by 180 d to align analogous illumination conditions in the two hemispheres. (Lower) Concurrent temperature measurements on the same balloon flights. The gray shaded regions delineate the time of year when ozone declines.

suggesting a pronounced difference in photochemistry. Some of the observed Arctic local minima occur at earlier times in the season, when they were probably due in part to transport processes (via transient synoptic disturbances, also sometimes referred to as “miniholes”; see refs. 4 and 8).

Fig. 2 (Lower) compares the associated seasonal cycles of 50-mbar temperatures in the two hemispheres as reported on the same balloon flights as the ozonesonde observations. The figure shows that the lowest temperatures in the Antarctic sometimes reach values below -90 °C, considerably colder than in the Arctic. Further, in the Arctic, there has not yet been a year in which lower stratospheric temperatures remained below about -80 °C much beyond day 80, whereas temperatures below -80 °C have

frequently been measured in the Antarctic as late as day 290, rendering the ozone loss season a full month longer than in the Arctic (see ref. 4 and references therein). The colder temperatures and longer duration of overlap between cold temperatures and sunlight allow photochemistry to attack ozone for many weeks, and lead to the extreme depletions obtained in Antarctica.

Fig. 3 shows the seasonal cycle of satellite local ozone observations from the MLS platform (see instrument validation in refs. 15 and 16), for days 1–180 in the Arctic near 70 mbar (poleward of $75^\circ N$) in two different years, compared with days 180–360 for Antarctic data (poleward of $75^\circ S$). At this pressure level, both the precision and accuracy of MLS data have been

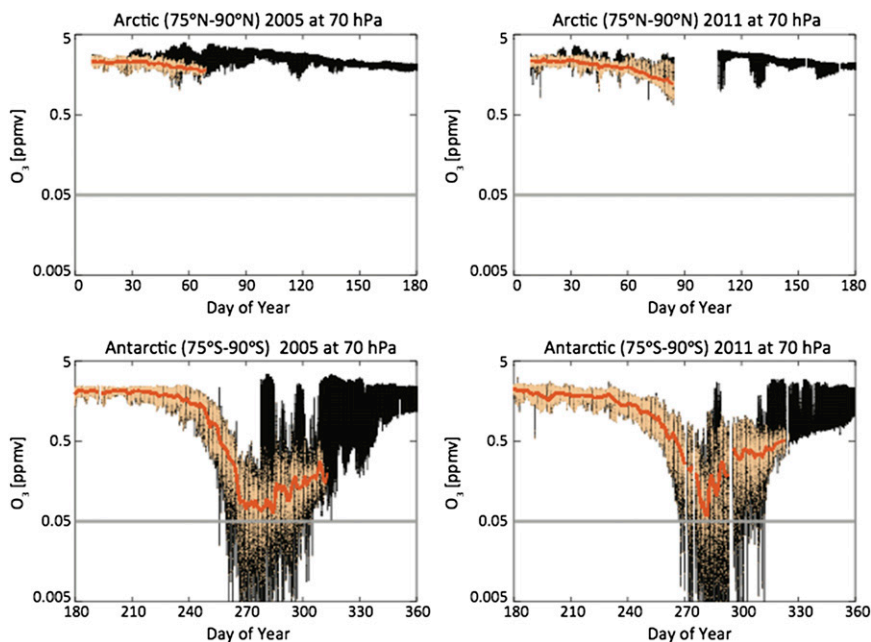


Fig. 3. Comparisons of the seasonal cycles of ozone from MLS satellite data taken poleward of 75° in each hemisphere near 70 mbar. Orange points denote data taken where local temperatures were below -75 °C, and orange lines represent the averages of the orange points. The horizontal lines indicate the nominal estimated accuracy and precision of MLS data (about 0.05 ppmv at this level).

estimated to be typically about ± 0.05 ppmv (16), indicated by the line on the figure; data below this level are subject to large uncertainty relative to their values, but are included because MLS data flags indicate useable observations. We use 70-mbar (18 km) MLS data here instead of 50 mbar (20 km) because estimated uncertainties are smaller at 70 mbar (15, 16). Data are shown for 2005 and 2011, two years that display differences in ozone values for each hemisphere. The dark shaded region in Fig. 3 represents individual satellite observations, whereas the orange dots and lines show individual observations as well as averages of data taken where the simultaneously measured local temperature was at or below -75°C (thus confining attention to the cold part of the polar vortex with a potential for rapid chemical ozone depletion). The results obtained here for the Arctic in 2011 are very close to those obtained in Manney et al. (4) using a dynamical definition of the vortex edge. The MLS instrument yields a range of values that are broadly similar to those seen from the electrochemical balloon soundings shown in Fig. 2. The average of the satellite data obtained in the cold vortex (orange line) declines to below 1 ppmv in the Arctic spring of 2011 (as shown in ref. 4, supplemental figure 4) before an instrument failure interrupted spring MLS data in that year. Fig. 3 indicates that although the average of the ozone values in the Antarctic in 2011 do rival those of the Arctic up to that point, the extreme low ozone values in the Antarctic are far lower,

suggesting substantial differences in the chemistry that are examined in *Lower Stratospheric Chemistry*.

Lower Stratospheric Chemistry

We next probe the thermal and chemical conditions under which severe ozone losses occur in the Arctic and Antarctic. It has been clear since the discovery of the Antarctic ozone hole that heterogeneous reactions involving particulate matter at cold temperatures are essential to its formation. Surface reactions involving hydrochloric acid (HCl), chlorine nitrate (ClONO₂), and hypochlorous acid (HOCl) serve to "activate" chlorine, converting relatively inert molecules into much more active forms (such as Cl₂) that photolyze readily, thereby producing chlorine free radicals (Cl, ClO) and catalytic destruction of ozone; several of the activation reactions proceed more rapidly as temperatures fall (see the review in ref. 3, and references therein). The relative roles and temperature dependencies of surface reactions on and in water ice, nitric acid trihydrate ice, and supercooled liquid sulfuric acid–water–nitric acid and binary sulfuric acid–water particles at background levels have been research topics since the late 1980s, and some important questions have recently reemerged. Some recent studies have suggested that heterogeneous chemistry taking place on background particles of sulfuric acid and water is sufficient to explain nearly all of the chlorine activation (17–19) and ozone losses in both the

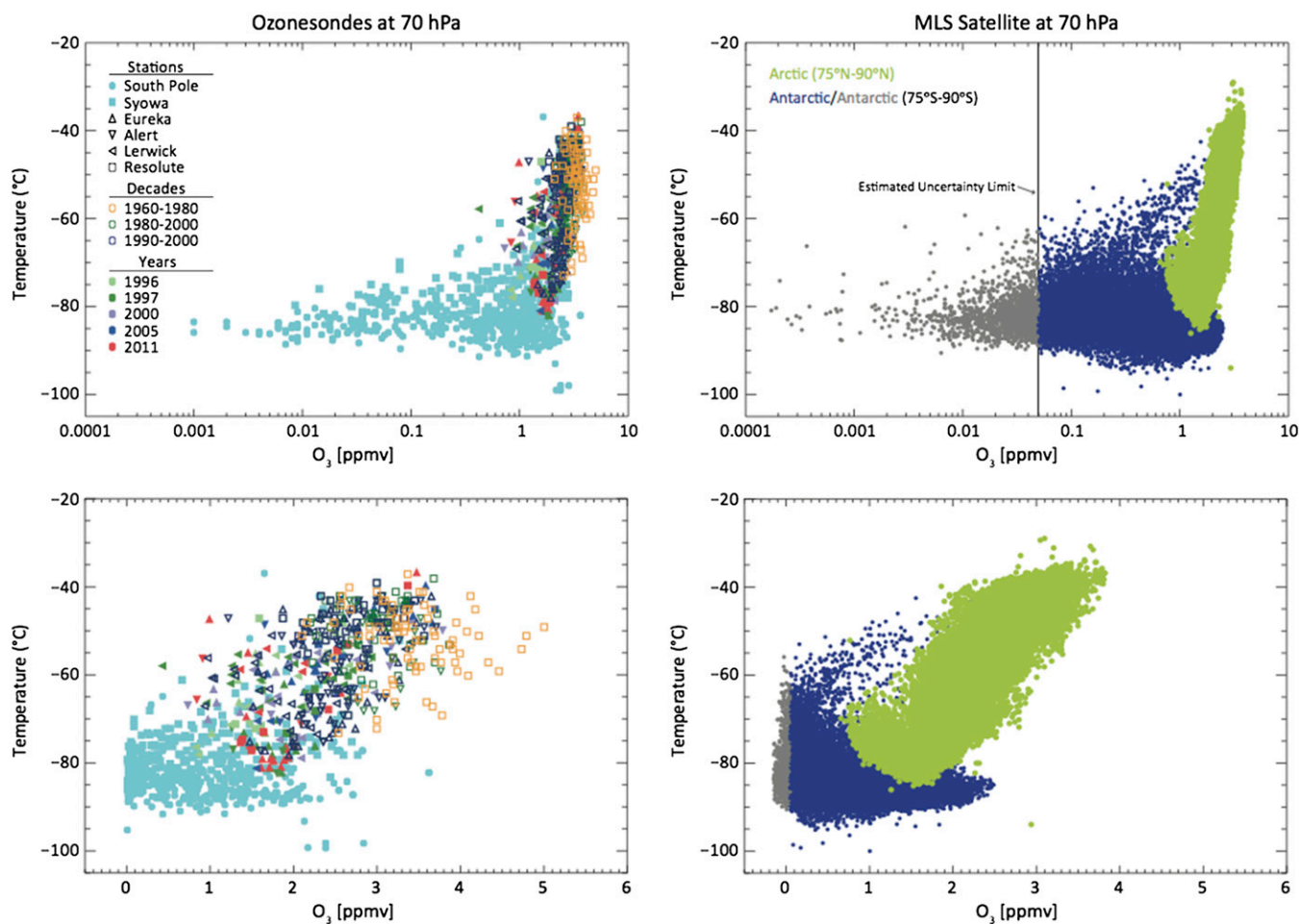


Fig. 4. Observations of ozone at 70 mbar versus temperature for days 60–100 in the Arctic and days 240–280 in the Antarctic (gray shaded region in Fig. 2) using (Upper) logarithmic and (Lower) linear scales. (Left) Data from ozonesondes in the Arctic and Antarctic for representative stations and years. (Right) Data from the MLS satellite instrument taken poleward of 75° in each hemisphere. The nominal estimated accuracy of the MLS data near 70 mbar is indicated by the change in color for Antarctic data from blue to gray and the vertical line in Upper Right.

Arctic and Antarctic (17) without any need for temperatures below -78°C or formation of polar stratospheric clouds, which would represent a major change in understanding. The potential for liquid binary aerosols to play some role in ozone loss under cold conditions was first identified decades ago (20, 21). Hanson et al. (21) showed that reactions of binary sulfuric acid–water aerosols can deplete some polar ozone for background aerosol at a particular range of temperatures cold enough to drive relatively rapid reactions but warmer than those typically associated with polar stratospheric clouds. The key question is not whether or not such surfaces can deplete some ozone but rather whether or not they actually do contribute substantially to ozone destruction compared with chemistry driven on PSCs that will certainly form as temperatures fall.

Activated chlorine needs to remain so for several weeks if extensive ozone loss is to occur. Activation is not a one-way process, and competition with deactivation (mainly reforming HCl and ClONO₂) increases the rate at which surface reactions must proceed if they are to keep pace with deactivation. In sunlit air, the rate of deactivation dramatically increases, for example, because any nitric acid present will photolyze, producing NO₂ and consequently converting ClO to the ClONO₂ reservoir. The rate of formation of HCl increases as well (22). In the late 1980s, it was emphasized that polar stratospheric cloud formation could reduce gas-phase nitric acid concentrations at temperatures below about -80°C , either temporarily by incorporation into particles or permanently via particle sedimentation (23), a phenomenon called “denitrification.” Reductions of the gas-phase nitric acid retard deactivation by incorporation of nitric acid into PSC particles and because of sedimentation of large particles, thereby acting to enhance polar ozone losses (e.g., refs. 4, 23, and 24). Photochemical ozone loss in polar spring is therefore strongly linked to the factors that affect both activation and deactivation, notably, cold temperatures that affect the rates of surface chemical processes, aerosol surface area, illumination, and denitrification.

Fig. 4 presents local observations of ozone versus temperature for balloonsondes and MLS satellite data in the Arctic and Antarctic near 70 mbar for the time of year when ozone decreases (gray shaded regions in Fig. 2, days 60–100 for the Arctic and 240–280 for Antarctica). Some very cold air parcels may display high ozone values if the air has not been cold enough long enough to become depleted, or because they are in the dark. Fig. 4 displays data using both logarithmic and linear scales, illustrating instrument precision, and the estimated MLS uncertainty is indicated. The logarithmic graphs in Fig. 4 provide an indication of the lower temperature limits required for ozone to drop, although it is important to note that the air sampled could have been colder before the time of the measurement (as will be shown in Fig. 5) and the temperature observations are subject to uncertainties of the order of a few degrees (25). Further, air parcels are subject to mixing, which acts to homogenize both ozone losses and temperatures; indeed, the triangular slopes of the data in Fig. 4 are suggestive of mixing.

The logarithmic graphs in Fig. 4 show that the deepest ozone losses are sharply clustered at temperatures near -80°C (193 K) in the Antarctic MLS data and as low as -85°C (188 K) in South Pole sondings, suggesting a key role for polar stratospheric cloud chemistry. To further probe the ozone depletion chemistry, Fig. 5 provides additional information obtained from the MLS satellite data. Here we again show ozone versus temperature in the times of year when ozone decreases (as in Fig. 4), but for individual years and with the gas-phase nitric acid content of the air indicated by the color scale.

Fig. 5 shows that a marked change in ozone occurred in the Arctic in 2011 near 70 mbar associated with air that was denitrified, in contrast to earlier years; the results in Fig. 5 are complementary

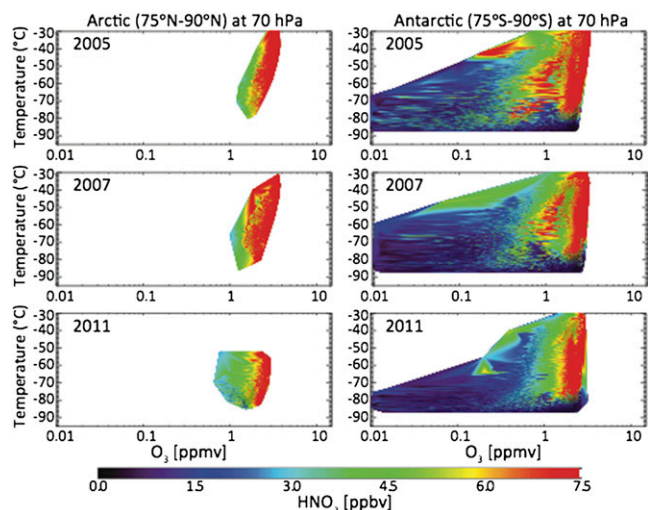


Fig. 5. Observations of ozone versus temperature poleward of 75° latitude near 70 mbar in the Arctic (Left) and Antarctic (Right) for particular years as indicated for days 60–100 in the Arctic and days 240–280 in the Antarctic (gray shaded region in Fig. 2), colored according to the simultaneously measured nitric acid abundances.

to the findings in ref. 4 regarding average values but also show local extremes. Although there is likely some ozone loss in the warmer and nondenitrified air that is present in other Arctic years at this pressure, its chemical character is different from 2011. Nitric acid decreases in the Arctic in 2011 in association with temperatures between -80°C and -85°C , and Arctic extremes are about 0.5 ppmv at this level. It is notable that Fig. 5 shows smaller ozone decreases in the Arctic in 2005 and 2007, when temperatures did not fall below -80°C and reductions in nitric acid appear to be more limited. This indicates that a temperature threshold of -78°C , for example, may produce some ozone decrease but does not yield extreme ozone losses. There is some Arctic air observed at warmer temperatures in 2011 showing low ozone, but it also displays significant denitrification, demonstrating that it must have been

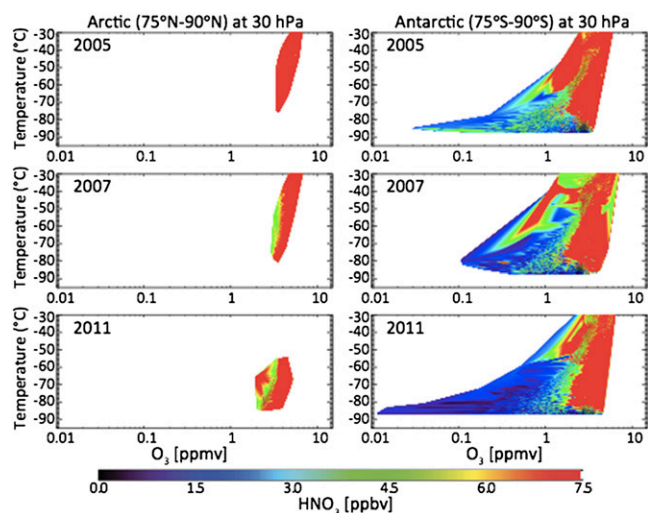


Fig. 6. Observations of ozone versus temperature poleward of 75° latitude at 30 mbar in the Arctic (Left) and Antarctic (Right) for particular years as indicated for days 60–100 in the Arctic and days 240–280 in the Antarctic (gray shaded region in Fig. 2), colored according to the simultaneously measured nitric acid abundances.

exposed to polar stratospheric clouds and colder temperatures at a time before the observation.

The Antarctic data in Fig. 5 show much lower ozone minima than in the Arctic in all years shown, and are associated with far greater denitrification. The differences between the Arctic and the Antarctic as seen in Fig. 5 suggest that temperatures colder than -80°C at 70 mbar are probably key to the differences in ozone destruction between the hemispheres. Analysis of the data in Fig. 5 shows that more than 90% of the Antarctic air parcels with ozone mixing ratios below 1.5 ppmv near 70 mbar contain less than 5 ppbv of nitric acid, compared with typical unperturbed nitric acid values of 10–20 ppbv. Denitrification is a distinctive marker for the chemical effects of PSCs, and Fig. 5 thus demonstrates that the bulk of the Antarctic ozone loss, and essentially all of the deepest losses driving ozone below 0.1 ppmv, has occurred in air exposed to PSCs.

Fig. 6 supports and expands upon these findings by a similar depiction of observations for 30 mbar (24 km). At this level, the Arctic in 2011 is again a year displaying relatively cold temperatures (minima of -85°C in Fig. 6). However, Arctic ozone at 30 mbar in 2011 does not suggest much depletion, with ozone minima of about 2 ppmv at 30 mbar, and little denitrification. In sharp contrast, far lower ozone of a few tenths of a part per million by volume or less is observed at 30 mbar in the Antarctic, and is associated with extensive denitrification and colder temperatures from -85°C to -95°C , providing additional evidence of the key role of extreme cold temperatures, PSC chemistry, and denitrification.

Although Figs. 5 and 6 establish that cold temperatures below -80°C to -85°C and PSCs are required to drive ozone abundances below 1 ppmv, we cannot determine whether the cold temperature (which implies faster rates of surface chemical activation) or the associated denitrification (implying slower deactivation) is more important for the ozone extrema. In other words, the association of denitrification with extreme ozone loss does not prove a causal mechanism, because both are driven by cold temperatures.

Conclusions

In situ and satellite measurements have been used here to study both averages and extremes in the depth of local Arctic and Antarctic ozone losses near 18, 20, and 24 km. The average local ozone decreases in the coldest part of the stratosphere above Antarctica at 20 km exceed those obtained in the Arctic, although the average values in the Arctic in 2011 at this level do rival those of some Antarctic years. However, Antarctic ozone also displays remarkable extremes in which local ozone frequently drops to or below 0.1 ppmv at 18, 20, and 24 km, in marked contrast to the Arctic (where, even in 2011, the minima did not fall below about 0.5 ppmv at 18 km and 2 ppmv at 24 km); undepleted ozone values at these altitudes are around 2–4 ppmv. The deepest levels of depletion in each hemisphere are clustered at temperatures below about -80°C at 18 km and below -85°C at 24 km, and occur in air parcels with reduced gaseous nitric acid concentrations. These results show how examination of the range of ozone depletion and the chemical composition associated with low ozone values provide important insights into the role of PSCs and the temperatures required to drive extreme ozone losses, and should prove useful for future evaluation of numerical models of ozone depletion. The observations demonstrate that the local extremes of ozone loss observed in the Antarctic have not yet been observed in the Arctic, and show the importance of temperatures below -80°C and polar stratospheric cloud chemistry in driving extensive ozone depletion.

Materials and Methods

In situ ozone observations by balloonsonde presented in this paper primarily use electrochemical detection (26) and have been checked for errors in background signals that can lead to spurious points. Historical balloon data using the less precise Regener and Brewer–Mast methods are also presented for comparison. The balloon data are available at the World Ozone and UV Data Center, except for the South Pole measurements, which are available from the National Oceanic and Atmospheric Administration. Satellite observations of ozone and nitric acid using microwave limb sounding methods (version 3.3) are also presented, and those data are available from the National Aeronautics and Space Administration's Aura MLS site at <http://mls.jpl.nasa.gov/>.

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