South Pole Station ozonesondes: variability and trends in the springtime Antarctic ozone hole 1986-2021

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Abstract. Balloon-borne ozonesondes launched weekly from South Pole station (1986-2021) measure high vertical resolution profiles of ozone and temperature from the surface to 30-35 km altitude. The launch frequency is increased in late winter before the onset of rapid stratospheric ozone loss in September. Ozone hole metrics show the yearly total column ozone and 14-21 km partial column ozone minimum values and September loss rate trends are improving (less severe) since 2001. However, the data series also illustrate interannual variability, especially in the last three years (2019-2021). Here we show additional details of these three years by comparing annual minimum profiles observed on the date when the lowest integrated total column ozone occurs. We also compare the July-December time series of the 14-21 km partial column ozone values to the 36-year median with percentile intervals. The 2019 anomalous vortex breakdown showed stratospheric temperatures began warming in early September followed by reduced ozone loss. The minimum total column ozone of 180 Dobson Units (DU) was observed on 24 September. This was followed by two stable and cold polar vortex years during 2020 and 2021 with total column ozone minimums at 104 DU (01 October) and 102 DU (07 October), respectively. These years also showed broad near-zero ozone (loss saturation) regions within the 14-21 km layer by the end of September which persisted into October.

Validation of the ozonesonde observations is conducted through the ongoing comparison of total column ozone (TCO) measurements with the South Pole ground-based Dobson spectrophotometer. The ozonesondes show a positive bias of $2 \pm 3\%$ (higher) than the Dobson following a thorough evaluation/homogenization of the long-term ozonesonde record completed in 2018.

1 Introduction

In 1986, NOAA began launching weekly balloon-borne ozonesondes at Amundsen-Scott South Pole Station (90°S) measuring high-resolution vertical profiles of ozone and temperature. This same year numerous field projects were deployed to Antarctica (Anderson et al., 1989; Tuck et al., 1989) to investigate the discovery of the springtime Antarctic ozone hole by Farman et al. (1985). Subsequent studies confirmed that the chlorine-catalytic destruction of ozone was enhanced over Antarctica in the presence of winter-time polar stratospheric clouds (PSCs) (Solomon et al., 1986; 1999, McElroy et al., 1986). The following decade of balloon-borne profiles, satellite, and ground-based measurements showed a broad and deepening ozone hole that stabilized in its

expansion by the early 2000s (Hofmann et al., 2009). More recently, several analyses of the ongoing ground-based and satellite measurements indicate that the ozone hole has been slowly recovering since 2000 (for list of studies see Langematz & Tully et al., WMO-report Chapter 4 (2018). The current recovery stage and upward trend in springtime ozone has been linked to the decline in the concentration of man-made ozone depleting substances (ODSs) due to the successful implementation of the Montreal Protocol international guidelines phasing out the production of ODSs. In 2020, the ODS abundance over Antarctica was 25% below the 2001 peak (Montzka et al., 2021). Full recovery is predicted to occur by around 2056-2070 when ODS levels return to the 1980-benchmark levels (Newman 2006; Dhomse et al., 2018; Amos et al., 2020). However, while long-lived ODS concentrations are steadily declining, the extent of chemical ozone loss may be quite different from year-to-year due to meteorological conditions (Newman et al., 2006; Keeble et al., 2014; de Laat et al., 2017; Tully et al., 2019; Stone et al., 2021).

After polar sunset, the strengthening of circumpolar winds and the development of a potential vorticity gradient forms the polar vortex boundary region that isolates stratospheric air over Antarctica (Nash et al., 1996). Near the center of the vortex, over the South Pole, ozonesondes measure stratospheric temperatures steadily decreasing during the polar night and remaining well below the -78° C threshold for PSC formation and growth. PSCs provide the surface reaction sites for activating stable chlorine species into radicals that rapidly destroy ozone after sunlight returns in September (WMO, 2018). However, planetary wave disturbances in late winter may weaken or completely break apart the cold and stable Antarctic polar vortex, ending the optimum conditions for rapid ozone loss in September (Schoeberl et al., 1989; Newman et al, 2004; Hassler et al., 2011a; Salby et al., 2012; Strahan & Douglas, 2016; de Laat et al., 2017; Strahan et al., 2019; Milinevsky et al, 2020). These conditions were observed in the warmer and weaker polar vortex conditions in 1986 and 1988 (Stolarski et al., (1990). The most extreme disruptions in the polar vortex occurred during the stratospheric warming events in 2002 and 2019 (Hoppel et al., 2003; Safieddine et al., 2020; Wargan et al., 2020).

The South Pole ozonesondes play a key role in monitoring ozone and temperature during all phases of the ozone hole. These unique measurements are critical after Antarctic sunset when several months of darkness limits the ground-based Dobson spectrophotometer and solar ultraviolet satellite optical measurements. Several indices and indicators have been presented in past analyses of the Antarctic ozonesonde records by Hofmann et al, (1997, 2009); Solomon et al., (2005, 2016); and Hassler et al., (2011a).

This paper is a review of the South Pole ozonesonde observations beginning with an overview in Sect. 2 of the electrochemical concentration (ECC) ozonesonde and recent homogenization of the South Pole data record by Sterling et al. (2018). Section 3 shows a comparison of ozone and temperature profiles during the last 3 years when the early polar vortex breakup and weak ozone hole in September 2019 was followed by severe ozone loss in 2020 and 2021 when cold vortex conditions persisted into early December (Kramarova et al., 2020; 2021). Section 4 shows the updated 36-year homogenized ozone time series and ozone hole metrics focusing on the 14-21 km layer column ozone minimums and linear ozone loss rates during September. In addition, we update ozone mixing ratio loss rates at selected pressure levels from the analysis by Hassler et al., (2011a) that showed maximum September loss rates occur in the 33-48 hPa region while the 89 hectopascal (hPa) was found to be the optimum layer for observing early detection of reduced ozone loss rates as ODSs decline. Section 5 illustrates the extent of ozone loss saturation observed each year during the annual minimum ozone period from 26 September to 15 October. The near-zero ozone layers were variable and narrowing after 2008, but were near maximum extent again in 2020 and 2021. The summary is given in Sect. 6.

2 ECC ozonesonde overview

The basic design of the electrochemical concentration cell (ECC) ozonesonde has remained relatively unchanged during the 36-year South Pole record (Komhyr, 1967). A Teflon piston pump bubbles ambient air into a sensor cell chamber with a platinum gauze electrode submerged in 3 ml of dilute, buffered potassium iodide (KI) solution. The ozone/iodide reaction in the ECC cell generates an electrical signal proportional to the ozone concentration.

Since about the mid-1990s, the ECC sonde manufacturers have improved the sensor cell design and the purity of the platinum electrodes, thus reducing the sensor current background to approximately 0.02 microamps when sampling no-ozone filtered air (Vömel & Diaz, 2010; Smit & Thompson, GAW Report #268, 2021). The limit of detection (3 x sensor background) converts to an ozone partial pressure of 0.10 millipascals or a mixing ratio of 0.02 ppmv at 50 hPa ambient pressure.

A Styrofoam box houses and insulates the ozonesonde pump and sensor. The weather radiosonde, attached to the outside of the box, measures and transmits meteorological and ozone data to the ground-based receiving equipment during ascent to the balloon-burst altitude of about 34 km. Consistent burst altitudes are maintained during the dark, cold months at South Pole by switching from standard rubber weather balloons to 18,000 standard cubic feet volume polyethylene film balloons during the first week of April then returning to rubber balloons by mid-October.

2.1 Data Homogenization

Each ECC ozonesonde profile represents a new instrument, used only once, thus, ozonesonde trends may show an offset or sudden bias rather than a slow drift in the data record when a new ozonesonde design or standard operating procedure change occurs (Johnson, et al., 2002; Smit et al., 2007; Tarasick et al., 2016; Van Malderen et al., 2016; Witte et al., 2017). The ozonesonde model and standard operating procedures (SOPs) at South Pole have not changed since 2006. However, prior to 2006, several dual and triple ozonesondes were flown to compare new ozonesonde models or adjustments made in the SOP in order to determine ad hoc corrections to account for these changes.

A thorough review and homogenization of the ozonesonde record was completed by Sterling et al. (2018) following homogenization methods that were formulated from the Assessments of Standard Operating Procedures (ASOPOS) workshops (Smit and ASOPOS, 2012; Deshler et al., 2017). The ozonesonde guidelines, presented in the ASOPOS GAW/WMO report # 268 (2021), are based on the Jülich World Ozonesonde Calibration Center Ozone Intercomparison Experiments (JOSIE). The JOSIE environmental simulation chamber experiments are the global reference for evaluation of new ozonesonde designs and SOPs and the foundation for improving long-term vertical ozone trends determined by ozonesondes with a goal to reduce uncertainty to ± 5% throughout the profile (Smit et al., 2007; Thompson et al., 2019).

Additional verification of the ozonesonde record at South Pole station is conducted through the ongoing comparison of total column ozone (TCO) with the NOAA ground-based Dobson spectrophotometer direct sun (DS) AD wavelength pair measurements over South Pole station from 20 October to 20 February (Komhyr et al., 1997). Globally, the Dobson network is an important long-term stable reference for ozonesonde sites and useful for identifying drifts in satellite platforms (McPeters & Komhyr, 1991; Bodeker et al., 2005; Thompson et al., 2017). Figure 1 shows the homogenized ozonesonde TCO record is a constant 2 ± 3% offset compared to the Dobson observations. The Dobson DS/AD observations are accurate to within ±1% (Köhler, et al., 2018). The ozonesonde TCO includes a residual value to account for estimated ozone above the balloon burst altitude by extrapolating a constant mixing ratio (CMR) from the balloon burst pressure (occurring between 20 to 7 hPa) to zero pressure. While the ozone residual lookup values from the satellite SBUV global climatological residual table from McPeters et al. (1997, 2012) is the

recommended procedure for determining ozonesonde residuals (Smit & Thompson, GAW Report #268, 2021), we have found that the CMR extrapolation is more consistent when comparing with the Dobson spectrophotometer TCO at South Pole Station.

2.2 Temperature Profile Validation

Sterling et al. (2018) discusses the details in the transition from three different radiosonde models at South Pole from VIZ (1986-1991) to Vaisala RS-80 (1991-2014) and the current GPS-enabled InterMet (Imet) radiosondes (2015-2021). The Imet measurements added GPS computed winds and geometric altitude to the South Pole profile data. For homogenization of the non-GPS Vaisala RS-80 data, the NOAA SkySonde software (A. Jordan author - see acknowledgements) was updated to retrieve nearby weather service radiosonde data. This provided a data source to identify and flag temperature outliers and to adjust the radiosonde pressure when offsets were > 2 hPa near burst altitude (Sterling et al., 2018).

The ozonesonde radiosonde temperatures were not adjusted in the homogenization of the data record. However, temperature accuracy for each flight was validated by comparing with an additional radiosonde flown by the Antarctic Meteorological Research Center (AMRC) at South Pole. For nearly a decade, the AMRC/South Pole Meteorology Office radiosondes (using Vaisala RS92 and Vaisala RS-41 GPS models) were "piggy-backed" on board the NOAA ozonesonde package. This is an important collaboration during the winter months when NOAA switches to the cold-resistant polyethylene 18K balloons to maintain burst altitudes of 30-34 km. This results in two independent temperature profiles to monitor the coldest temperatures in the polar stratosphere where PSCs begin to form. The typical radiosonde rubber balloon profile will fail/burst at 14-15 km over South Pole under these extreme cold and dark conditions. The comparisons between the Vaisala radiosonde models (RS80 and RS92) during 2012-2013 flights show almost no difference in stratospheric temperature measurement during the summer months and only a slight 0.2°C difference in winter. The Imet radiosonde measures slightly higher temperatures, by about 0.5°C, than the Vaisala RS41, primarily during the summer months.

3 South Pole Ozonesonde Profiles: 2019-2021

Balloon-borne ozonesondes provide a unique overview of the yearly ozone hole over South Pole by comparing vertical profiles measured during winter (before depletion begins) with the annual minimum ozone profile typically observed between 26 September to 15 October. Figure 2 shows the 2019-2021 ozone (upper panel) and temperature (lower panel) profiles representing the ozone holes of these three years. The winter profiles (blue) are an average of the 6-8 profiles measured from 15 June to 15 August, when the stratospheric temperatures range between -85 to -95 °C and total column ozone (TCO) averages 263 ±17 DU.

The wintertime ozone profiles are similar to the long-term climatology each year and typically don't provide any insight into how the polar vortex conditions and the severity of ozone depletion will unfold when rapid depletion begins by 01 September at South Pole. However, during the last two weeks of August the first signs of ozone loss are occasionally observed above 21 km, likely from transported air parcels originating near the polar vortex boundaries where sunrise, Cl2 photolysis, and chemical ozone destruction begins (Schoeberl et al., 1992; Lee et al., 2001; Hassler, et al, 2011a; Strahan et al., 2019).

The springtime minimum TCO profiles (red) average 114 ±15 Dobson Units (DU) which represents a 55-65% ozone loss when compared to the wintertime profile. The 2019 minimum TCO of 180 DU was observed on 24 September, the 2nd highest minimum in the 36-year South Pole record. This was the 3rd season when the polar vortex was dramatically disrupted leading to an early September warming and meridional mixing of ozone-rich air into the polar region from the mid-latitude stratosphere

(Wargan et al., 2020; Safieddine, et al., 2020). Table 1 lists the other two years (1988 and 2002) when similar events occurred. The table also includes the date intervals when data were excluded in our long-term median calculations since these extremes in ozone and temperatures were not representative of typical chemical ozone hole losses.

150   Table 1. Years when an early disruption of the polar vortex was observed over South Pole and the corresponding period when the profile data were excluded from the ozonesonde median and percentile climatology.

Year	Dates excluded	Event
1988	11 Aug – 01 Dec	Early vortex weakening in August
2002	22 Sep – 15 Dec	Sudden stratospheric warming / split vortex (Allen et al., 2003)
2019	05 Sep – 20 Dec	Sudden stratospheric warming /vortex shift (Safieddine et al., 2020)

The stable and cold polar vortex conditions in 2020 and 2021 led to severe ozone holes over South Pole with minimum total
155   column ozone measurements of 104 DU on 01 October and 102 DU on 07 October, respectively. The profiles in Fig. 2 show near complete destruction of ozone within the 14-21 km vertical layer during those two years. The record low total column ozone profile in 2006 (92 DU) on 09 October, shown as the dotted line in Fig. 2, had a 7 km vertical extent of near-zero ozone (ozone loss saturation) from 14 to 21 km. Thereafter, the 14-21 km layer became the baseline region for tracking ozone loss metrics and the severity of the annual ozone hole over South Pole (Hofmann et al., 2009). The term "near-zero ozone" from hereon will represent
160   stratospheric ozone partial pressure measurements that fall below 0.2 millipascals (two times the LOD). The next section shows all of the observations of the 14-21 km column ozone during 2019-2021 from July-December to illustrate the temporal evolution and variability of the ozone hole.

3.1 South Pole Ozonesondes 14-21 km time series: 2019-2021

Figure 3 shows the July-December time series of 14-21 km column ozone and temperature in 2019, 2020, and 2021 compared to
165   the 1986-2021 climatological median with 30-70th /10-90th percentiles in gray shading. The median and percentile values were calculated using a sliding time series bin that is gradually reduced from ±14-days in July to a ± 3-day bin during the month of September when more frequent ozonesonde launches track the rapidly decreasing ozone column. The slope of the median 14-21 km column ozone (black line) decreases linearly during September at a rate of -3.5 ± 0.3 DU/day. This metric is computed for each individual year and presented in Section 4 to show the ozone loss rate trend. After 01 November the ozone and temperature
170   percentiles broaden significantly due to variable dates when the Antarctic polar vortex fully dissipates (Karpetchko et al., 2005; Bodeker et al., 2005).

The stratospheric warming event in 2019 included a large-scale shift of the polar vortex towards the tip of South America (Safieddine, et al., 2020), away from the typical position centered near the South Pole. Figure 3 shows the anomalous high ozone and temperature breakout in early to mid-September 2019 over South Pole. The 08 September temperature profile showed the first
175   sign of this weakening vortex with an abrupt increase of more than 10° C in the 14-21 km layer. However, the column ozone within the 14-21 km layer remained close to the median line until 20 September when the strong depletion period ended and ozone values leveled off in the 45-50 DU range. Then on 10 October, dropped to the minimum for the year at 44 DU when the polar vortex briefly centered back over the Antarctic continent and South Pole station.

The opposite polar vortex conditions were observed in 2020 and 2021 when cold temperatures and column ozone tracked
180   well below the median near the lower edge of the 10-90th percentile line from September through December in Fig. 3. Both years
showed severe loss in the 14-21 km column with minimums of 2 DU (01 October 2020) and 3 DU (01 October 2021). The daily
Dobson TCO observations also tracked the slow return to typical seasonal values. The latest date to break above the standardized
220 DU ozone hole threshold (Stolarski et al., 1990) at South Pole was 12 December 2020 when 236 DU was measured nearly two
months after the Dobson recorded 109 DU in mid-October. The NASA satellite observations also showed the longest-lived ozone
185   hole on record in 2020 due to the very weak planetary-scale wave activity (Kramarova et al., 2021)

4 Ozonesonde Metrics: Altitude Intervals: 1986-2021

Table 2 lists the altitude layer metrics analyzed in this study related to ozone loss during September and the minimum ozone
occurring by early October. While the lowest ozone is a key metric of ozone hole severity each year, many recovery indices focus
190   on the September observations when the highest sensitivity and correlation with decreasing ODSs may be ascribed (Solomon et
al. 2016; de Latt et al., 2017; Pazmiño et al., 2018; Strahan et al., 2019). The South Pole ozonesonde metrics here focus on the 14-
21 km layer and include two additional metrics showing an update of the mixing ratio loss rate profiles at selected pressure levels
from Hassler et al. (2011a) and a metric showing the yearly vertical extent of layers with near-zero ozone.

195   Table 2. Altitude Layers and Metrics updated for 1986-2021 ozonesonde record at South Pole.

Altitude Layer	Metric – plotted data
14-21 km	Column ozone minimums (DU) & September loss rates (DU/day)
Pressure: 119-33 hPa	September mixing ratio loss rate profiles (ppmv/day)
10-24 km – curtain plot	Near-zero ozone (mPa) layers during minimum period (Sep26-Oct15)
Dobson Units (DU)	*Mixing Ratio – parts per million by volume* (ppmv)
Millipascals (mPa)	*Hectopascals (hPa)*

Figure 4a shows an overview of the 36-year time series of selected 14-21 km integrated column ozone values representing
three stages of the ozone hole over South Pole. The upper panel shows the winter average ozone observed before depletion begins
and the spring minimum ozone series. An additional series shows the 15 September values when an ozonesonde is launched each
200   year on this date to track the progress of ozone depletion (Hofmann et al., 2009).
The winter average 14-21 km column ozone (15 June - 15 August) has been relatively constant at 130 ± 10 DU. The spring
minimum ozone series bottomed out at near-zero ozone from 1993-2001 followed by an upward trend after 2001. The long-term
trends in 14-21 km column ozone are more evident in the 15 September series when the 1986-1999 period showed ozone decreasing
at a rate of -2.3 ± 0.6 DU/Yr. This was followed by an upward trend line at $+ 0.9 \pm 0.4$ DU/Yr. The simple linear regression lines
205   in Figure 4a were computed by the "least squares" method. The uncertainty is from the standard error in the slope.
Both the spring minimum and the 15 September series show year-to-year variability. However, the three anomalous polar
vortex break up years (red dots) stand out as peaks in the minimum series while the 15 September observations showed almost no
signal. This may be attributed to South Pole being near the center of the ozone hole during early September, far from the vortex
edge where there is greater dynamical influence in ozone observed (Hassler et al., 2011b). Also, the first signs of the polar vortex

 weakening typically appear as layers of higher ozone and sudden increases in temperature above 24 km altitude at South Pole Station.

The lower panel (Fig. 4b) shows the result of subtracting selected series in Fig. 4a in order to illustrate the 14-21 km layer ozone total loss each year (winter average – spring minimum) and the loss that occurred before and after 15 September. From 1991-2000, there was an increasing trend in the 14-21 km column ozone loss during the first half of September (blue line in Fig 4-b) reaching a peak of 100 DU loss in 2000. This was followed by a downward trend with significant variability, until reaching a relatively stable 60-65 DU after 2014. The loss during the second half of September depends on the amount of ozone remaining after 15 September and meteorological conditions governing the stability of the polar vortex. An early vortex weakening or break up may result in transport and mixing of high ozone air masses with the depleted ozone thus reducing or ending loss before the end of September. The year 2021 shows the second highest overall loss on record at 133 DU. This year began at a slower than average pace with only 59 DU of ozone loss by 15 September but followed with a record loss of 74 DU for the second half of September when only 3 DU remained in the 14-21 km layer on 01 October.

4.1 September column ozone loss rates: 14-21 km

The South Pole September 14-21 km column ozone loss rate is a key metric suggested by Hofmann et al. (1997, 2009) for observing changes related to potential recovery in stratospheric ozone. The metric is useful since the rapid ozone loss during the month of September follows a nearly linear decline which can be compared with the 1986-2021 median loss rate of -3.5 ± 0.3 DU/day (see Figure 3). Figure 5 shows the yearly September ozone column (14-21 km) loss rates from the ozonesondes launched every 2-4 days during late August until mid-October. The selection of the start and end day for the ~ 30-day loss period are adjusted forward or backward by ± 3 days to obtain the best linear fit to the observations (see method description in Hassler et al., 2011a). In late September, near the minimum date, the linear depletion data point selection ends when either a sharp increase in ozone is observed or, in severe depletion years, drops to near-zero or shifts to a non-linear loss rate when approaching ozone loss saturation. The selected values between the start and end points are used to determine the yearly loss rate slope and uncertainty by simple linear regression. The 36-year time series shows that the 14-21 km column ozone loss rate has increased (lower ozone loss rate) from a minimum of -3.8 DU/day during 2002-2007 to -3.0 DU/day in 2016-2021 The sudden stratospheric warming in 2002 (red dot) showed rapid ozone loss but within a shortened time period ending on September 22. This was the date when the first sign of the sudden stratospheric warming (increase in ozone and temperature) began to show at altitudes above 21 km. The following ozonesonde profile on 25 September 2002 showed substantial ozone increases throughout the 15 to 32 km layer elevating TCO to 397 DU, the highest ever observed during September and October over South Pole. In 2019, the loss rate calculation period was also shortened to just two weeks before the linear ozone decline ended on 15 September. The loss rate data point for 2019 is included in Figure 5 with high uncertainty.

4.2 September ozone mixing ratio loss rates: 119-33 hectopascals

Hassler et al. (2011a) analyzed vertical profiles of ozone mixing ratio loss rates during September in twenty layers from 200 to 10 hPa showing that peak loss rates occurred within the 48 and 33 hPa layers. The 89 hectopascal (hPa) pressure level was found to be the optimum layer for detecting significantly lower loss rates based on model estimates of future declining equivalent effective

stratospheric chlorine (EESC) and lower variability in measured ozone loss at this level. Early detection was estimated to occur sometime within the 2017-2021 period.

Figure 6 shows the September ozone mixing ratio loss rates (ppmv/day) for five selected pressure levels from 119 hPa (13.6 km) to 33 hPa (20.6 km). Following the analysis method by Hassler et al. (2011a), the ozone mixing ratios during September (~Day 235-270) are grouped in 5-year intervals to reduce the influence of prevailing phases of the quasi-biennial oscillation (QBO) and other dynamical processes affecting temperature, meridional transport, and mixing depending on polar vortex conditions. Figure 6 shows that overall loss rates peaked during 2001-2005. All the selected pressure levels showed decreases in loss rates by 2016-2020. The 89 hPa loss rate showed improvement (29% decrease) with the lowest variability as predicted in the Hassler et al. (2011a) assessment. The highest altitude layer at 33 hPa level showed a substantial 49% decline from the 2001-2005 peak loss rate.

Figure 7 shows the 1986-2021 temperature time series for the five pressure levels during the month of September and before sunrise from 15 July to 15August when stratospheric temperatures remain cold (-90 to -94 °C) and stable. These data indicate that there have been no systematic winter temperature trends at these altitudes that may affect stratospheric cloud particle surface area and heterogeneous ozone destruction chemistry. However, 2021 stands out as the coldest September observed at all pressure levels. In addition, Figure 7 shows that the stable vortex in 2021 remained very cold with almost no temperature difference between winter and September at pressure levels of 119, 89, and 67 hPa and just 2 to 3°C warmer at 48 and 33 hPa. This was similar to the record low ozone year in 2006, but the 2006 temperatures were not as cold as 2021. The highest temperatures occurred in 2002 after the sudden stratospheric warming on 22 September when the 30-50 °C increase in temperature observed from 100-20 hPa sent the September temperature average off scale in Figure 7.

5 Ozone Loss Saturation: near-zero ozone layers

The near complete destruction of stratospheric ozone (loss saturation) within the 14-21 km layer is a feature of the Antarctic ozone hole that is observed in detail by high-resolution ozonesondes. Near the end of September, as the linear decrease in ozone begins to slow, other nonlinear depletion reactions complete the pathway to near-zero ozone (Grooβ et al., 2011; Kuttippurath et al., 2018; Muller et al., 2018). A reduction in the vertical extent of the near-zero ozone layers will be an important indicator of recovery showing when decreasing equivalent effective stratospheric chlorine (EESC) is no longer the excess component in the reactions that destroy ozone (Kuttippurath et al., 2018).

The vertical extent of near-zero ozone layers observed over South Pole is shown in Figure 8 as a curtain plot of dark to light gray shaded bars showing only the lowest partial pressure minimums from 0 to 0.7 millipascals (mPa). This range of values in the shaded regions represent a 95-99% loss compared to the 14-16 mPa values observed during the winter before ozone depletion begins (see Fig. 2). The near-zero ozone values for each year are selected from the lowest partial pressure ozone out of all 7-9 profiles flown during the minimum ozone hole period (26 September – 15 October). All values greater than 0.7 mPa are not included in order to highlight the lowest ozone region. The three early vortex breakup years, listed in Table 2, are shown as light red bars when ozone minimums were > 4-5 mPa.

Figure 9 shows the average temperature curtain plot during the minimum ozone period. The PSC threshold temperature of -78° C is selected as the break point between cold (blue) and warm (red) temperature scales. Together, Figs 8 and 9 show the coincidence of years with cold temperatures and low ozone. The recurring year-to-year severe depletion from 1991-2001 also shows that the upper boundary of the near complete ozone loss layer was extending to higher altitudes each year, eventually peaking

at 21 km. Hofmann et al., (1997) and Hoppel et al., (2003) noted that a reversal of ozone loss in the upper altitudes may be an important indicator of stratospheric ozone recovery as ODSs decline.

Figure 8 shows that after the record low in 2006, the near-zero ozone vertical extent appeared to be narrowing and becoming irregular. Then in 2020 and 2021 ozonesondes observed the optimum cold polar vortex conditions in September to late October along with extensive near-zero ozone within 13.5 to 20.5 km altitude. However, the 7 km near-zero ozone layer was not observed in a single profile as it was in the record low ozone profile in 2006. For example, in 2021 the near-zero ozone layer was initially observed at 15-20.5 km on October 1, and from 13.5-17.5 km on 21 October.

Severe ozone loss extending below the 14-21 km layer is not common during the minimum ozone period over South Pole. However, the near zero ozone region may extend below 14 km following major volcanic eruptions. The volcanic plumes may eventually transport sulfate aerosol into the polar stratosphere resulting in additional ozone loss through surface reaction processes (Hofmann & Solomon, 1989). For example, there was significant depletion from 12-14 km in 1992-1994 after the major eruption of Mount Pinatubo (15.1° N; Philippines) in 1991 (Hofmann and Oltmans, 1993; Deshler et al., 1996,). The Calbuco eruption (41.2° S; Chile) (Bègue, et al., 2020) in 2015 also led to an increase in ozone loss below about 14 km (100 hPa) at South Pole (Stone et al., 2017). More recently, the 15 January 2022 Hunga Tonga - Hunga Ha´apai (20.5°S) volcanic eruption plume in the SW Pacific Ocean reached altitudes of nearly 55 km (Carr et al., 2022). This recent mass injection and transport of volcanic aerosols, SO_2, and water vapor (Millán et al., 2022; Vömel et al., 2022) may enhance future stratospheric ozone depletion within the Antarctic polar vortex. The ongoing ozonesonde observations along with the 36-year ozone climatology record over South Pole will assist in identifying any additional ozone loss layers.

6 Summary

The South Pole station ozonesondes provide essential year-round high-resolution ozone and temperature profiles tracking all phases of the Antarctic yearly ozone hole near the core of the polar vortex, monitoring both the winter development of the vortex, the conditions that lead up to rapid ozone depletion in September, and looking precisely in the region where the ozone loss is taking place. The 36-year ozonesonde record has been reviewed, homogenized, and validated by comparing total column ozone with the South Pole Dobson spectrophotometer direct-sun total column ozone measurements. Ozonesondes show a positive bias with respect to the Dobson TCO of $2 \pm 3\%$.

The 2019, 2020, and 2021 South Pole ozonesonde measurements showed the greatest year-to-year variability in ozone hole conditions ever observed in the South Pole long-term record. Table 3 shows several of the key metrics for those years compared to the long-term median and record low in 2006. The anomalous polar vortex warming in 2019 disrupted ozone depletion in early September resulting in the second weakest ozone hole on record, but the following two years saw cold and persistent vortex conditions with very low total ozone column minimums among the 10th lowest in the 36-year record. In addition, the 2020 and 2021 profiles in 1-21 October showed near-zero ozone (ozone loss saturation) within the 14-21 km altitude layer under the persistent cold and stable vortex conditions.

Table 3. The South Pole yearly ozone hole minimum total column and minimum14-21 km partial column ozone values during the long-term record (1986-2021), the record low in 2006, and the most recent 3 years (2019-2021).

Year	Total Column Minimum DU (Date observed)	14-21 km Column Minimum DU (Date observed)	14-21 km Column September Loss Rates DU/Day
1986-2021	114 ± 15	8±5	-3.5 ± 0.3
2006 (record low)	92 (09 Oct)	1 (09 Oct)	-3.9
2019	180 (24 Sep)	44 (10 Oct)	Depletion ended 15-Sep
2020	104 (01 Oct)	2 (01 Oct)	-3.0
2021	102 (07 Oct)	3 (01 Oct)	-3.0

320   The time series (1986-2021) of 14-21 km column ozone during the winter months (15 June - 15 August) shows no trend averaging 130 ± 10 DU. However, rapid ozone loss during 01-30 September at rates of -3.5 ± 0.3 DU/day results in 95-99% loss of ozone in the 14-21 km layer. Minimums of near-zero ozone (~ 1-2 DU) were observed every year from 1993-2001. This was followed by an irregular upward trend from 2002-2021 with the minimum 14-21 column ozone values ranging from (1 – 9 DU). The near-zero ozone minimum years after 2001 include: 2003, 2005, 2006, 2011, 2015, 2020 and 2021.

325   The 15 September 14-21 km column ozone time series indicates a turnaround year in 2000/2001. The simple linear regression lines show decreasing ozone from 1986-1999 at -2.3 ± 0.6 DU/Yr followed by a reversal to a slight positive trend at + 0.9 ± 0.4 DU/Yr in 2000-2021. This pattern is consistent with several Antarctic ozone hole studies focused on detection of ozone recovery due to decreasing ODSs (Langematz & Tully et al., WMO-report Chapter 4, 2018; SPARC/IO3C/GAW Report No. 241, 2019).

The September mixing ratio loss rates at selected pressure levels averaged within 5-year blocks all showed improvements by 330 2016-2020 compared to the peak loss period in 2001-2005. The ozone loss at 33 hPa showed the greatest improvement with a 49% reduction in loss rate. The optimum pressure level (89 hPa) for detecting significantly lower loss rates showed a 26% reduction with the lowest variability as predicted by Hassler et al. (2011a).

The long uninterrupted 36-year South Pole ozonesonde record and future balloon-borne measurements provides unique and vital data for ozone hole analyses. The continuing year-round ozonesonde observations at South Pole Station will be beneficial for 335 observing anomalies in the ozone layer driven by meteorological events disrupting the polar vortex and for identifying layers where volcanic aerosols influence ozone depletion.

Data availability Datasets in this study can be obtained through NOAA ozone archive https://gml.noaa.gov/aftp/data/ozwv/ .
340 NDACC from   https://www.ndaccdemo.org/stations/south-pole-antarctica

Competing interests: The authors declare that they have no conflict of interest

345 **Special issue statement:**

This article is part of the special issue "Atmospheric ozone and related species in the early 2020s: latest results and trends (ACP/AMT inter-journal SI)". It is a result of the 2021 Quadrennial Ozone Symposium (QOS) held online on 3–9 October 2021.

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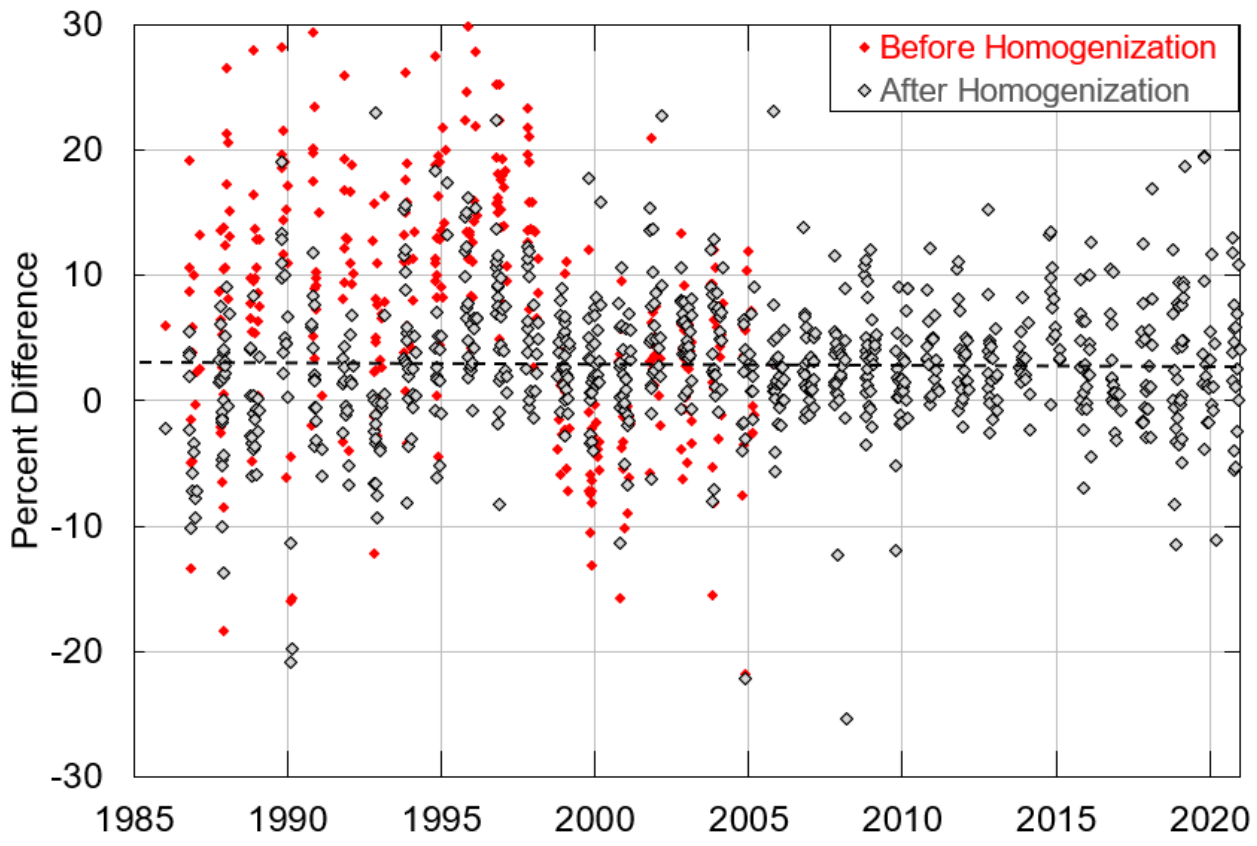
Figure 1. Total column ozone comparison of ozonesondes and the Dobson spectrophotometer direct sun measurements. The red diamonds represent the percent differences before the homogenization of 1986-2006 ozonesonde data.
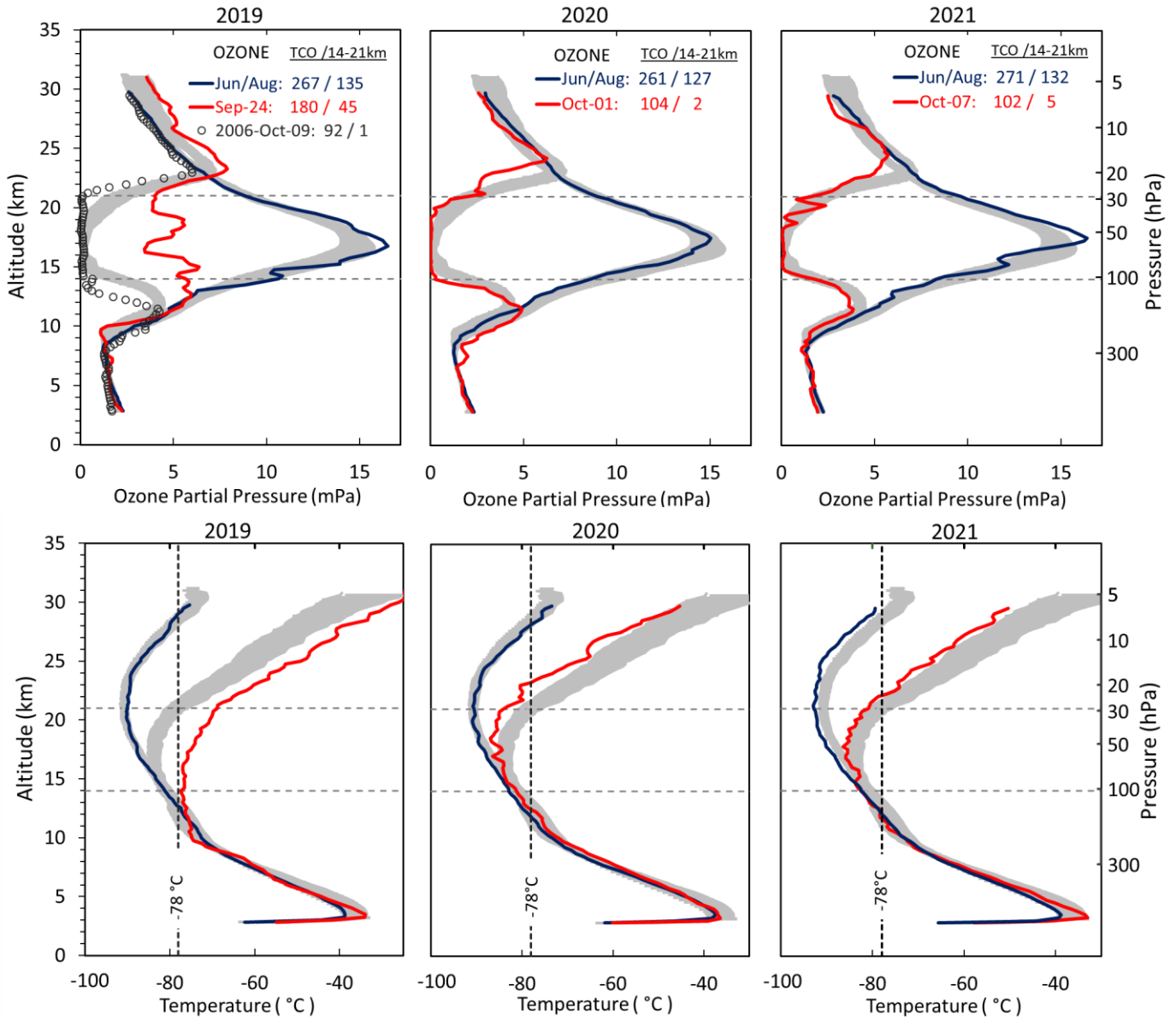
Figure 2. Selected ozonesonde profiles from 2019-2021 representing the ozone hole severity over South Pole by comparing the average winter profile (before depletion begins/blue line) to the minimum ozone profile (red). The minimum total column ozone (TCO) and 14-21 km (horizontal dashed line region) partial column ozone values are given in Dobson units (DU). The gray-shaded region represents the 1986-2018 median 30-70th percentiles for winter and minimum periods. The record low of 92 DU total column ozone measured in 2006 is shown as a dotted line in the 2019 graph. The temperature graphs show the -78 °C PSC threshold as a vertical dashed line.
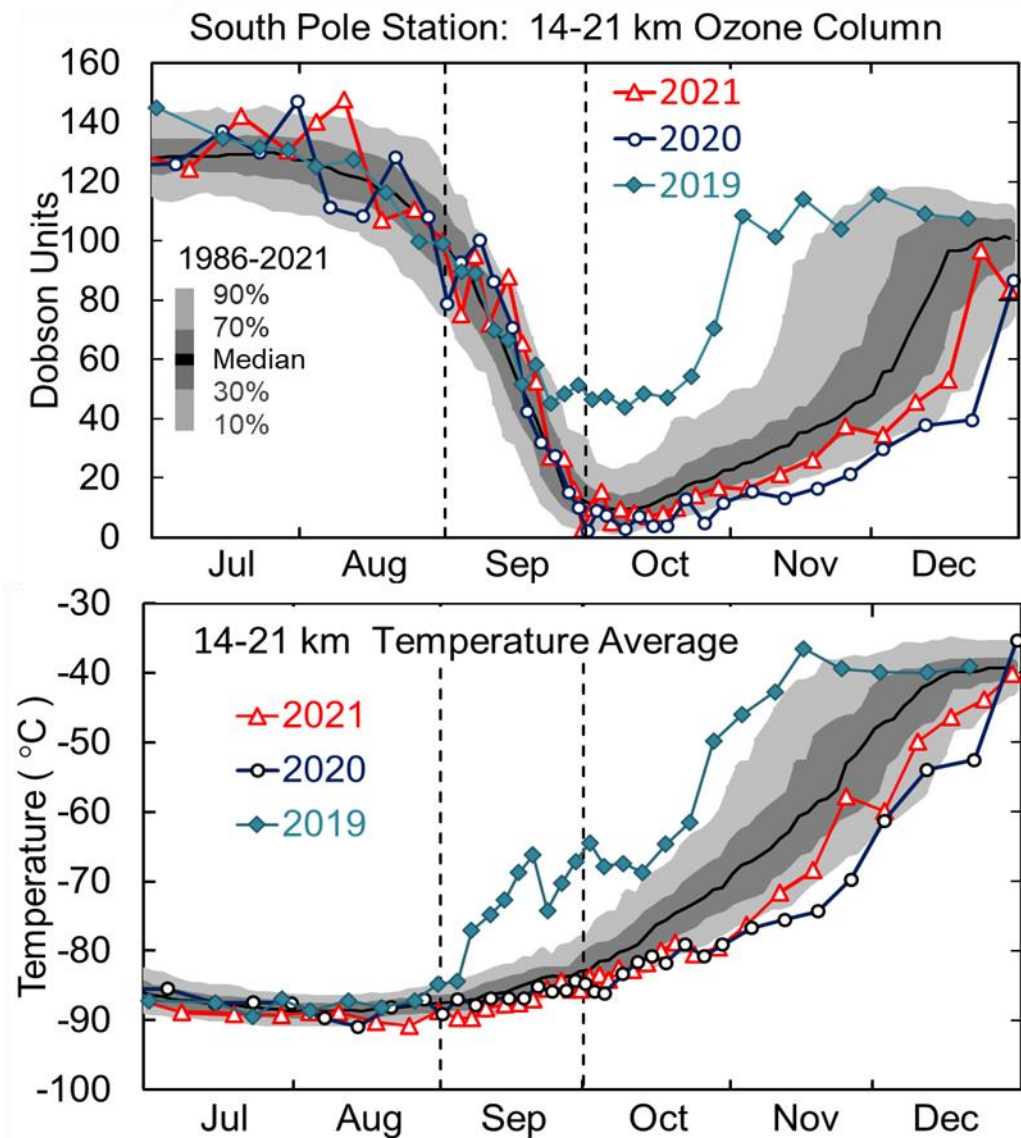
Figure 3. Ozonesonde July-December time series in 2019-2021 showing the 14-21 km column ozone in Dobson Units (DU) in the upper panel and average temperature (lower panel) compared to the 36-year median (black line) with gray percentile envelopes. The extreme variability due to the unstable polar vortex conditions in 2019 versus stable conditions in 2020 and 2021 shows in the last three months when the measurements eventually converge to normal ozone values by the end of December.
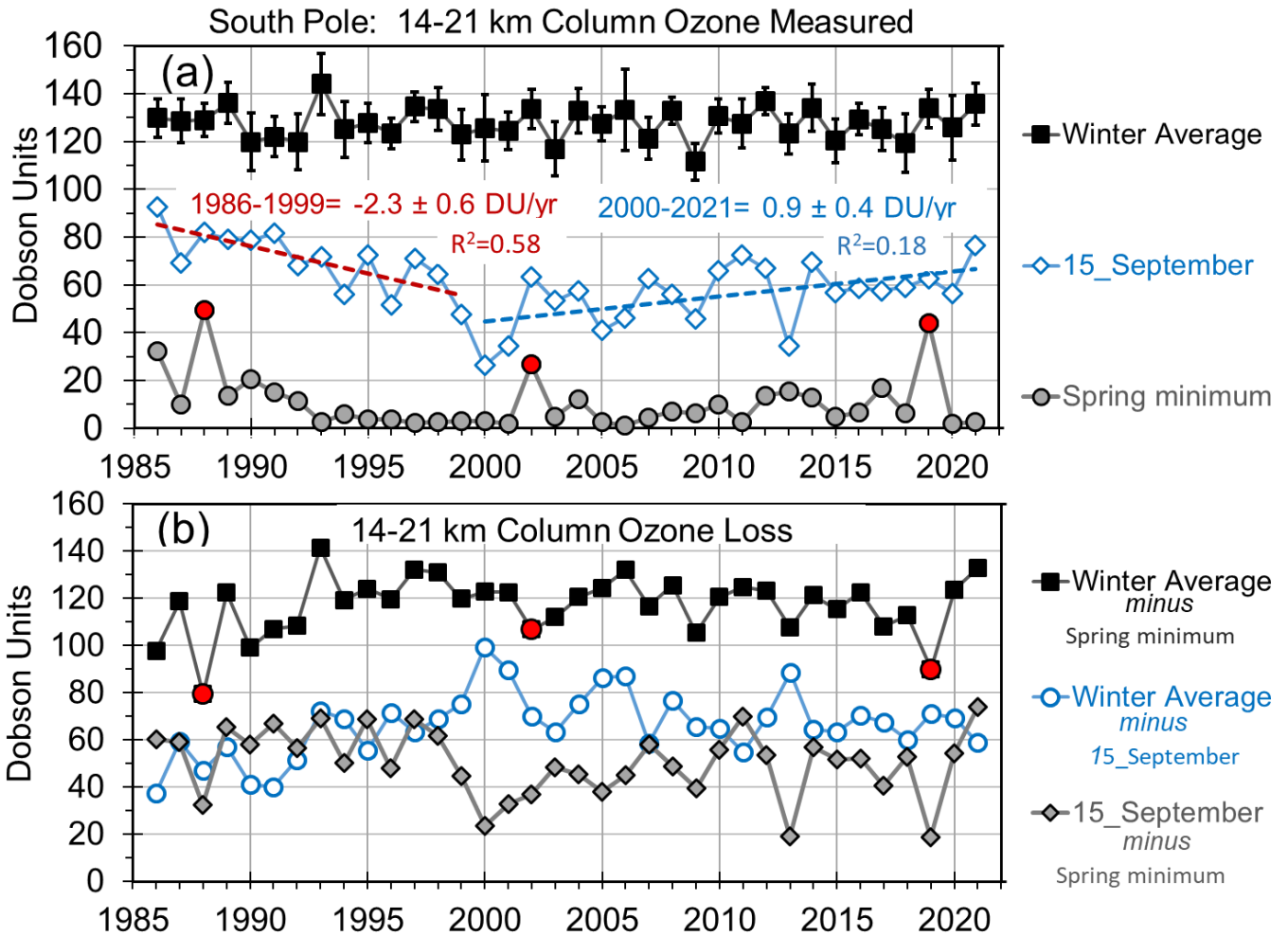
Figure 4. (a) The 1986-2021 measurements of 14-21 km column ozone over South Pole in Dobson Units (DU) during winter (15 June - 15 August average), 15 September, and the spring minimum. The two dashed lines show the simple regression linear trends and R^2 values for all 15 September measurements before and after 2000. (b) The lower panel shows the difference between the three time series in the upper panel to illustrate maximum column ozone loss, the loss during the first half of September (blue open circles line), and second half (gray diamonds line) of the depletion period during September.

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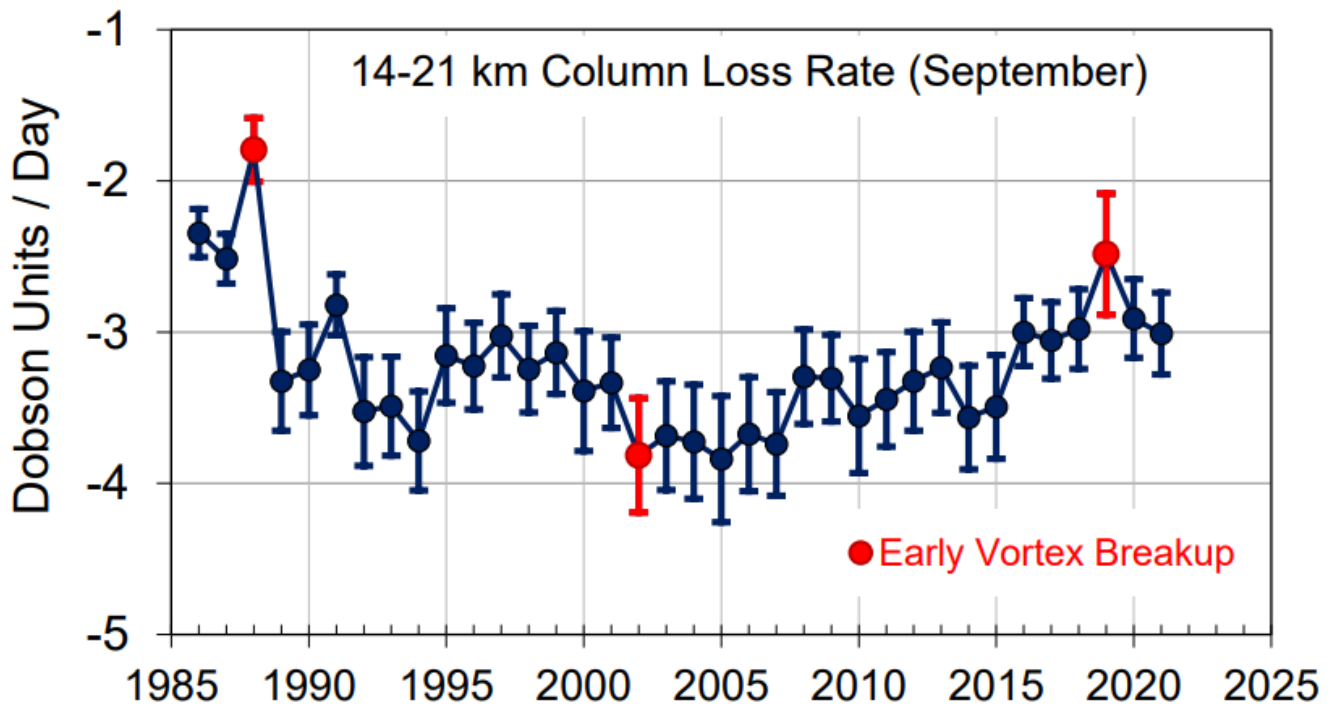
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Figure 5. Column ozone 14-21 km loss rates [DU/day] during September. The three anomalous vortex years are shown as red dots. The yearly data include 1-σ uncertainty bars.

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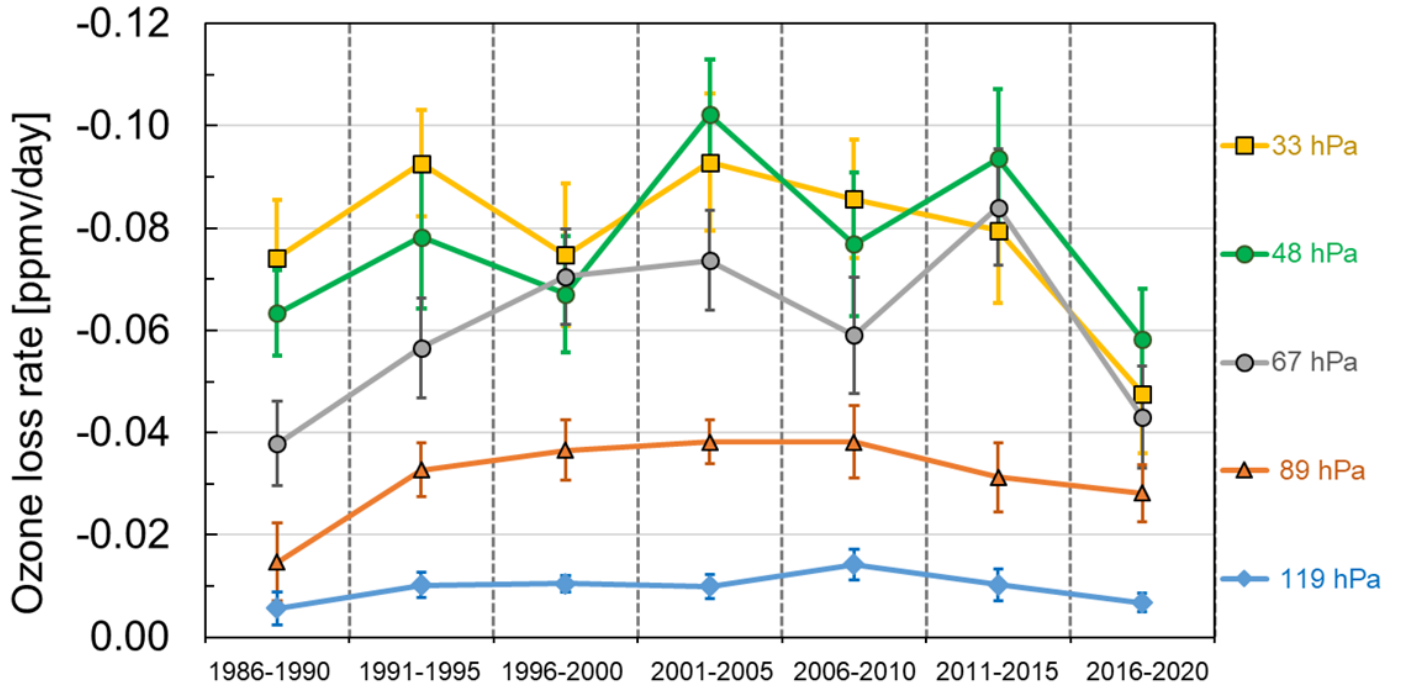
Figure 6. September loss rates [ppmv/day] calculated within 5-year blocks at five selected pressure levels within the primary depletion layer from 119 hPa (13.6 km) to 33 hPa (20.6 km) at South Pole. The linear fit to the data include 1-σ uncertainty bars.

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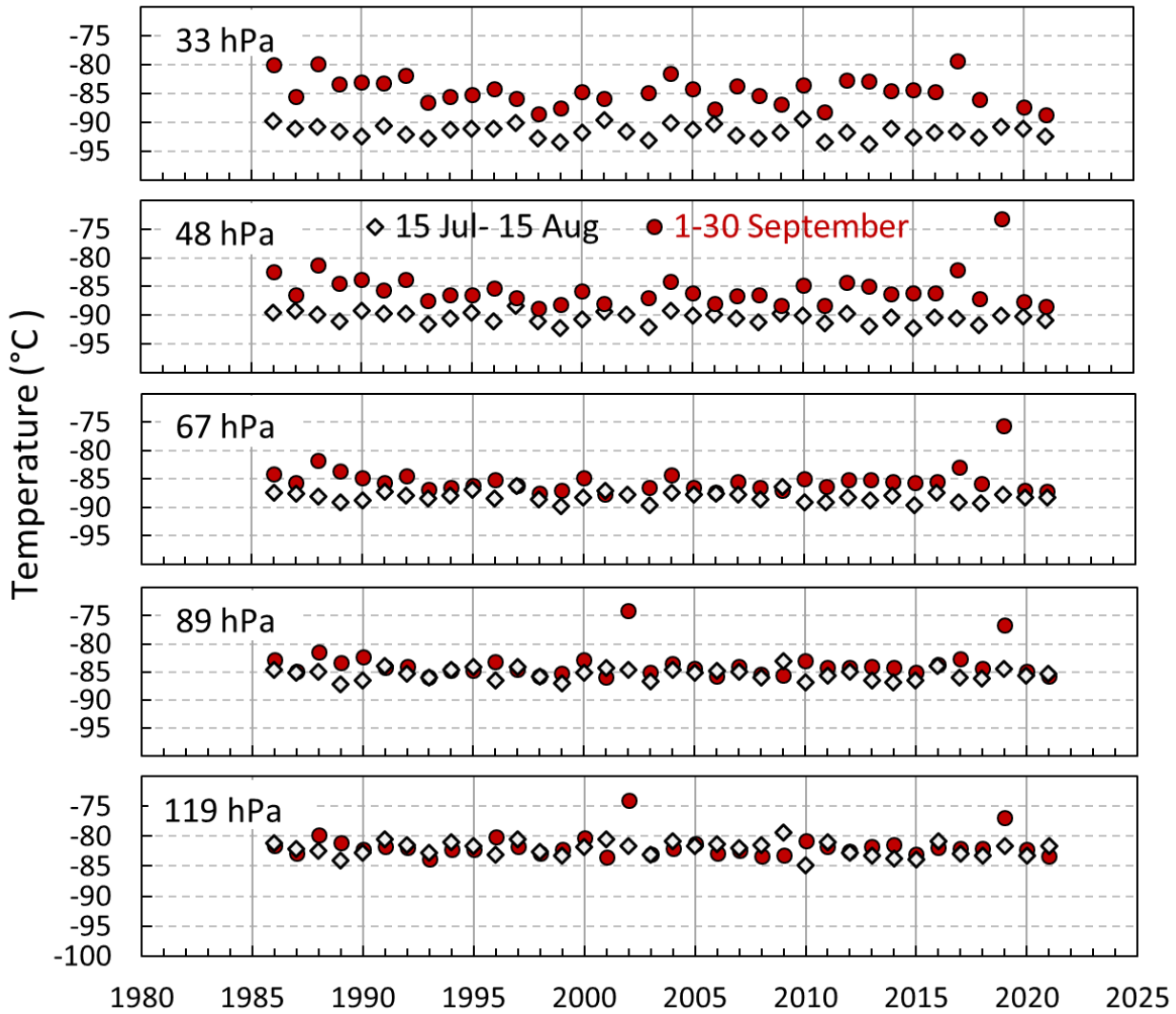
680 **Figure 7.** Average 30-day temperatures during winter from July 15 to August 15 (black diamonds) and September 1-30 (red circles) from 1986 to 202. The pressure levels are the same as in Figure 6, within the primary ozone depletion layer from 119 hPa (13.6 km) to 33 hPa (20.6 km).
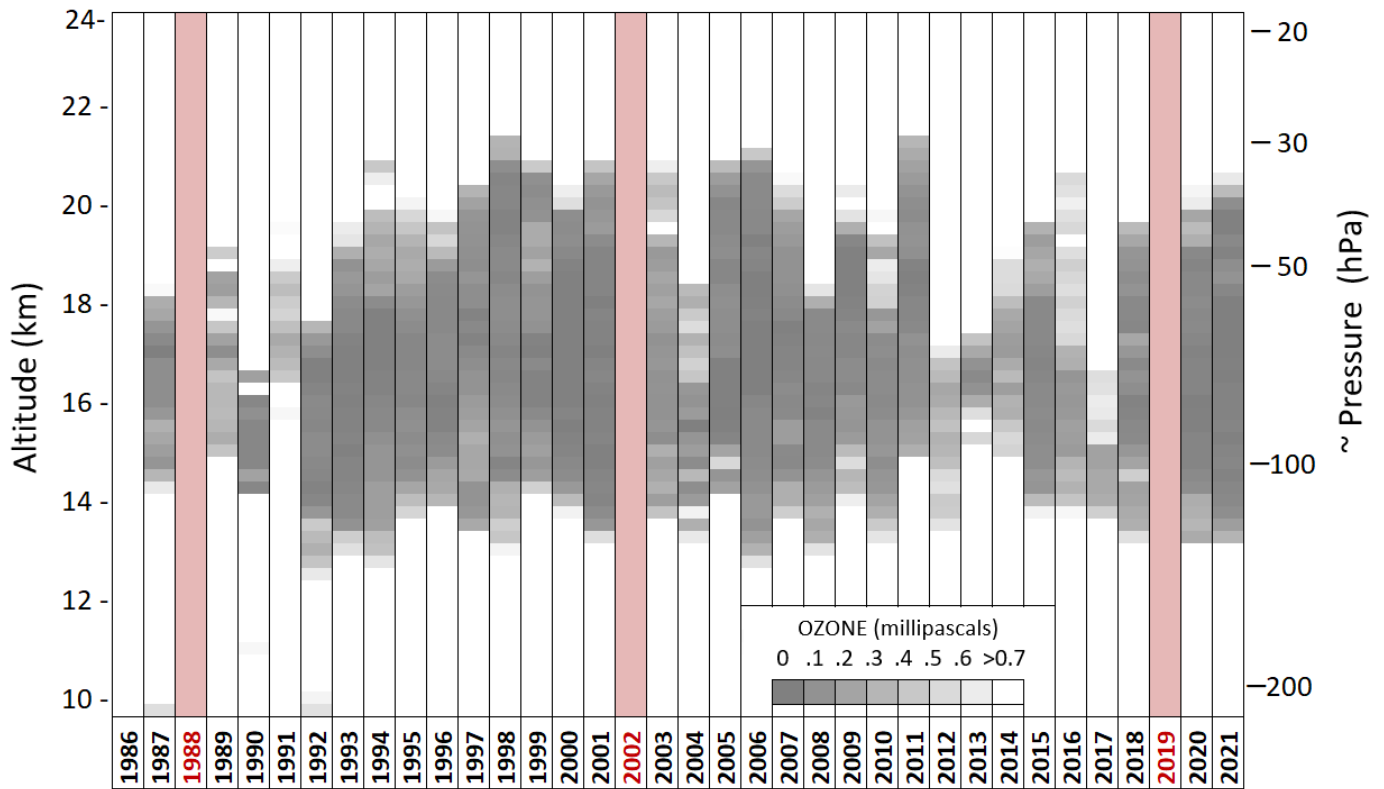
Figure 8. South Pole 10-24 km curtain plots highlighting the lowest observed ozone partial pressure 0 to 0.6 millipascals (mPa) represented by the dark to light gray shading during the yearly ozone hole minimum period from 24 September to 15 October. All values greater than 0.7 mPa are white.
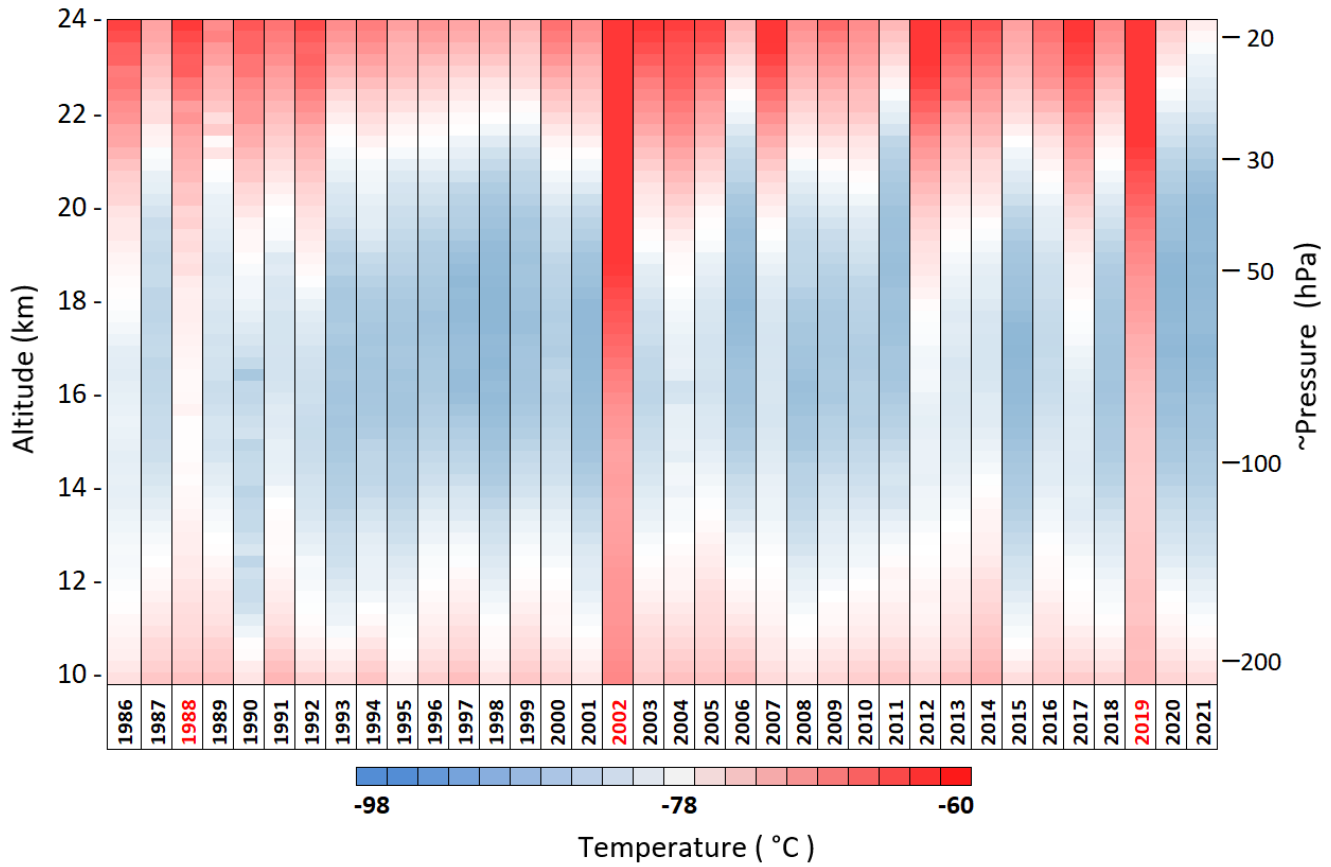
Figure 9. The average temperature during 24 September to 15 October (ozone hole minimum period over South Pole Station). The blue to red transition temperature falls at -78 ºC, the polar stratospheric cloud formation threshold temperature.