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Emergence of ozone recovery evidenced by reduction in the occurrence of Antarctic ozone loss saturation

J. Kuttippurath¹, P. Kumar¹, P. J. Nair¹ and P. C. Pandey¹

Industrial emissions of ozone depleting substances (ODSs) during the second half of the twentieth century have led to one of the most visible human impacts on the Earth: the Antarctic ozone hole. The ozone loss intensified in the 1980s and reached the level of saturation (i.e., complete loss of ozone) due to the high levels of ODSs in the atmosphere. Significant changes in the southern hemispheric climate have been observed in the past decades due to this unprecedented ozone loss. Although the most recent studies suggest healing in the Antarctic ozone hole, the status of ozone in the loss saturation layer (~13–21 km) has not been discussed in detail. Here, a comprehensive analysis of vertical, spatial and temporal evolution of ozone loss saturation (ozone mixing ratio ≤ 0.1 ppmv) in the Antarctic vortex using high resolution measurements for the 1979–2017 period reveals that the loss saturation began in 1987 and continued to occur in all winters thereafter, except in the major warming winters of 1988 and 2002. However, our analysis shows a clear reduction in the frequency of occurrence of ozone loss saturation over the period 2001–2017 consistently throughout various datasets (e.g., ozonesonde and satellite measurements of ozone profiles and total columns), thereby revealing the emergence of an important milestone in ozone recovery.

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INTRODUCTION

The ozone loss process in the stratosphere is relatively well understood, and much of the loss is driven by the chemical cycles involving chlorine and bromine compounds, whose abundance peaked around 2000 in the polar stratosphere.¹ With the abatement of atmospheric loading of ozone depleting substances (ODSs) due to the implementation of Montreal Protocol,² the extent of ozone loss is expected to have decreased since 2000.¹ Early signs of the rebound of Antarctic ozone have already been reported.^{3–5} However, one of the main difficulties in determining accurate trends in ozone in the southern polar stratosphere is the complete loss of ozone there. The saturation of ozone loss, i.e., the total or near-zero destruction of ozone in the lower stratospheric layers, mostly at 13–21 km, has reportedly begun in 1991,^{6–8} although there are references for the near-complete loss of ozone at some lower stratospheric altitudes in McMurdo observations in 1987.^{9,10} Nevertheless, those studies did not mention “saturation” of the ozone loss there. Studies have shown that this loss saturation may hamper determining accurate trends, but has not been investigated in much detail.^{7,8} Both chemistry-climate and chemical transport modelling studies on the ozone loss and projection of ozone changes often struggle to simulate the features of loss saturation⁹ such as the altitude range of saturation and ozone values below 0.1 ppmv.^{11,12} Henceforth, precise information on the saturation of ozone loss in the Antarctic vortex would allow a better assessment of the evolution of ozone in that region towards its recovery detection.

In addition, although recent studies indicate a significant positive change and a healing in the Antarctic ozone hole, details of ozone change in the loss saturation altitudes in the polar vortex is not clearly known yet, albeit there are studies on ozone loss for

individual or few years together.^{1,2,6–10} This situation thus warrants a careful and detailed examination of ozone at these loss saturation altitudes to make a clear statement on ozone recovery and its robustness at this sensitive vertical region. Here, we present, to our knowledge, the first detailed long-term (four decades) analysis of Antarctic ozone loss saturation in terms of its first occurrence, timing, spatial differences, vertical spread, inter-annual changes and temporal evolution using high-resolution ozonesondes and satellite measurements inside the vortex for the 1979–2017 period.

RESULTS AND DISCUSSION

Antarctic ozone loss saturation

The Antarctic ozone loss saturation is a unique phenomenon, which is more frequent and very severe at latitudes near the South Pole and hence, the balloon-borne vertical ozone profiles at the South Pole, Neumayer and Syowa research stations show near-complete ozone destruction as the values are about 1 mPa (~0.1 ppmv) or smaller, as depicted in Fig. 1. The depletion of ozone up to 0.1 ppmv indicates loss of about 95–99% of ozone in the lower stratosphere. The magnitude and spatial extension of ozone loss shown are the maximum measured at the respective stations, as analysed from their entire measurement records. It also shows the spatial differences in the vertical distribution of ozone loss in Antarctica. In general, the ozone profiles illustrate that the complete loss of ozone begins at the altitude of about 350 K (~12 km) and vertically spreads up to 550 K (~22 km) with frequent loss saturation occurrences at 400–450 K (~13–16 km), where the total loss of ozone occurs at most stations.

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Fig. 1 Ozone loss saturation in the Antarctic. The vertical profiles of ozone measured by ozonesondes on different dates at different stations in Antarctica. The profiles in red show the ozone distribution prior to the ozone loss period and those in blue show the ozone loss saturation observed in spring. The ozone loss saturation observed in 1987 from then available measurements are shown in green. The dotted horizontal lines represent altitude 360 K (~12 km) and 510 K (~20 km), where the loss saturation is very common and frequent. The vertical dashed lines represent 0 mPa

Fig. 2 shows the distribution of ozone measured at different stations at 400 K (~13 km) in October during the period 1979–2017. Only Syowa measurements are available during 1979–1984 and Neumayer in 1985, and these measurements show concentrations of approximately 1 ppmv in this period, but Neumayer measurements show values ≤ 0.1 ppmv in 1985. The ozone measurements from all stations again show below 0.1 ppmv in 1987, where the South Pole measurements register the smallest values. However, the measurements show about 0.6 ppmv in 1988 and they exhibit incredibly similar values at all stations in this winter. The ozone values show about 0.01–1 ppmv at all stations thereafter, except in 2002. The amount of ozone prior to the onset of ozone loss in spring at these altitudes is about 3–5 ppmv and ozonesonde measurements have an uncertainty of about 5% in the lower stratosphere. Therefore, we define 0.1 ppmv as the ozone loss saturation threshold, which represents about 95–99% of ozone loss in the lower stratosphere, depending on altitude. Additionally, this would represent the loss incurred due to ozone loss catalytic cycles alone. Note that the loss saturation threshold is different from the detection limit of ozonesondes. There is no consensus on the detection limit of ozonesonde measurements, as studies have considered values from 1 to 40 ppbv for this (see Methods). We have taken 10 ppbv as the detection limit of ozonesondes, as most studies have considered this value for scientific analyses.^{12,13}

The analyses with ozonesonde measurements reveal that the ozone loss saturation first occurred in October 1987 and continued to occur in all winters thereafter, except in the warm winters of 1988 and 2002.^{14,15} Note that, the first appearance of ozone loss saturation was in 1985 at Neumayer (there were no other station measurements and no references too), but there was no loss saturation in 1986. Hence, it can be argued that widespread saturation of Antarctic ozone loss began in the cold

winter of 1987, and is found in all then available measurements. The near-complete loss of ozone in the lower stratosphere in 1987 was also found in some previous analyses,^{9,10} although never mentioned about the “saturation” of loss. Therefore, to date, the early 1990s have been regarded as the beginning of loss saturation in the Antarctic stratospheric ozone.¹⁷ The role of Pinatubo volcanic aerosols is undeniable in the large loss of ozone in 1992, when all measurements show values below the saturation threshold.^{16,17} Similarly, the very cold winters 2001, 2003, 2006 and 2015 show most measurements down to 0.1–0.01 ppmv. The saturation was very severe, in terms of its vertical and spatial expansion, from 1991 through 2000, but signature of a positive change with comparatively higher values in ozone is observed for the period 2001–2017. This could be partly due to the dynamics, as there were a few warm winters in 2010s (e.g., 2010, 2012, 2013 and 2017). Yet, as mentioned earlier, the profiles having ozone depletion up to 0.1 ppmv still indicate the loss of about 95–99% ozone at these altitudes.

We also examined the satellite measurements to diagnose the signatures of loss saturation (Figure S1). Note that the comparison with satellite measurements should be taken qualitatively throughout this study, as they can hardly measure very small values of ozone below the saturation level. The satellite measurements also have an uncertainty of about 5–10% in the lower and middle stratosphere. The features extracted from satellite measurements largely depend on horizontal and vertical resolution, and time and mode (e.g., nadir or limb) of the measurements; and the resolution of satellite measurements is very coarse. As shown by the ozonesondes, the satellite measurements show symptoms of loss saturation at 400 K in 1985 and 1986 with values of about 0.15 ppmv. The measurements show loss saturation values in all winters except in 1988 and 2002. Since the frequency and coverage of Aura MLS (Aura

Fig. 2 Spatial distribution of ozone loss saturation. Ozone mixing ratios measured by ozonesondes at different Antarctic stations at the potential temperature level of 400 K (~13 km) in October during the 1979–2017 period. Only Syowa measurements are available for the years 1979–1984 and Neumeyer in 1985. The individual measurements are shown against the equivalent latitudes (EqLs) inside the polar vortex. The horizontal dashed lines represent 0.1 ppmv

Microwave Limb Sounder) observations are larger than that of SAGE-II (Stratospheric Aerosol and Gas Experiment-II), the former makes more and vortex-wide loss saturation measurements. The Aura MLS measurements suggest that the saturation occurs more near the polar latitudes, as the South Pole measurements exhibit higher number of saturation occurrences.

Fig. 3 shows the box plots of ozone values at 400 K from the ozonesonde measurements. As found in Fig. 2, the measurements show saturation values at Neumayer, South Pole and McMurdo in 1987. The median of measurements show about 0.35 ppmv in 1985, although the 1%-whiskers are at the threshold value of saturation; indicating the first apparent signature of beginning of loss saturation. The median values are below 0.15 ppmv at Neumayer and South Pole, but slightly higher at McMurdo in 1987. It also demonstrates the gravity of loss saturation at South Pole and McMurdo, as the measurements show loss saturation in all winters, except in 1988 and 2002. Although the loss saturation is evident at Neumayer, it was not very strong in 1988–1991. The very cold winters often inflict the loss saturation at Syowa, Marambio and Davis. The Mirny and Maitri stations have only few years of measurements at this altitude inside the vortex, and the loss saturation occurs more often at Maitri. It suggests that the results also depend on the frequency and timing of measurements.

VERTICAL, SPATIAL AND TEMPORAL FEATURES

The saturation of ozone loss is further examined with its temporal and spatial evolution at different altitudes. Figure 4 shows the

evolution of ozone inside the vortex ($>65^\circ$ equivalent latitude (EqL))¹⁸ at different vertical levels in the lower stratosphere between the potential temperatures 375 K (~12 km) and 575 K (~23 km) in October for the period 1979–2017. The measurements at different altitudes show similar features of time evolution of loss saturation. The ozone loss started in the early 1980s, intensified in the mid-1980s, the level of saturation first reached in 1985 and again in 1987, and it stayed around this saturation level to date at altitudes 375–550 K (~12–22 km). The loss saturation is conspicuous from 375 to 500 K (~20 km) in most winters and for most stations since 1987, but it seldom occurs above that altitude as found in 1998. Among the winters, however, there are two exceptions, the warm winters 1988 and 2002, as mentioned previously. The loss is unprecedented at altitudes 400–450 K, where all measurements show loss saturation at all stations since 1987. It is also interesting to note the loss saturation at higher altitudes at 500–575 K in 2013, and was a unique winter in this regard. In summary, the analyses at different vertical levels clearly show that the loss saturation first appeared in 1985, initiated the vortex-wide saturation in 1987 (e.g., all four stations show loss saturation values in 1987) and it continued to occur thereafter. The break in loss saturation in 1988 was due to a major warming, in which no measurement showed saturation threshold of ozone loss at any altitude. This is also evidenced by the lower stratospheric temperature measurements shown by Kuttippurath and Nair.³

The satellite measurements show loss saturation threshold of about 0.1 ppmv in 1985, and is evident at 375 K (Figure S2). The ozone values smaller than 0.1 ppmv again appeared in 1987 at 375–450 K and continued to occur in the succeeding winters too,

Fig. 3 Spatial distribution of ozone loss saturation. The box plots of ozone mixing ratios measured by ozonesondes at different Antarctic stations at the potential temperature of 400 K (~13 km) in October during the 1979–2017 period. The box represents 25–75% and whiskers represent 1 and 100%. The average values are shown in red and the median in black. The horizontal dashed lines represent 0.1 ppmv

except in 1988 and 2002. The Aura MLS measurements show the loss saturation in all winters from 2004 to 2017 at different altitudes in the lower stratosphere (375–525 K or ~12–21 km), and was very severe at 400 K, consistent with the ozonesonde measurements.^{19,20} The satellite measurements, however, show no loss saturation at 550 and 575 K, which could be due to the lower vertical resolution of the measurements at these altitudes. These results are further verified using box-whisker plots at different altitudes and are consistent with previous discussions (Figure S3), such as the loss saturation signatures (0.15 ppmv at the 1%-whisker) in 1985 at 375 K and the onset of vortex-wide loss saturation from 1987 onwards.

Ozone column characteristics

To examine the characteristics of ozone at the core of loss saturation layer, we computed the partial columns at 400–500 K from the ozonesonde measurements and compared to the total column ozone mapping spectrometer/ozone monitoring instrument (TOMS/OMI)^{21,22} overpass observations, and are displayed in Fig. 5. The comparison shows excellent agreement between ground-based and satellite measurements (Figure S4). As expected, the partial column ozone shows very small values at all stations after the first appearance of loss saturation in 1985. The estimated columns show about 25–80 Dobson Units (DU) in 1979–1984, 6–25 DU in 1985 and 15–60 DU in 1986. The

measurements demonstrate the smallest and largest of that decade in 1987 (0.8–25 DU) and 1988 (25–40 DU), respectively. However, the columns show below 10 DU thereafter, with a gradual decrease that peaked (i.e., the smallest column) in 1998 with values of about 0.2 DU at South Pole. The coldest spring of 1997, 1998, 2001, 2006 and 2011 show partial columns below 1 DU, indicating the void of ozone in the lower stratosphere. The historic stations South Pole, Syowa, McMurdo and Neumayer exhibit ozone columns less than 50 DU in all years in 1989–2017; consistent with those observed from ozonesonde mixing ratio analyses. The South Pole soundings show smaller ozone columns (<10 DU) compared to the measurements from all other stations. There are high ozone column episodes at these stations (e.g., Marambio) due to the occasional excursions of vortex to the South American Peninsula. Additionally, most years, except the major warming years and 1990, show very small values; suggesting the saturation of ozone loss.

The TCO measurements from TOMS and OMI (Fig. 4, upper panel) show about 300–450 DU in 1979–1981, 210–400 DU in 1982–1983, 250–350 DU in 1984, 225–400 DU in 1985 and 80–400 DU thereafter. The measurements show below 220 DU in 1981–1982 and in all winters since 1987, and this is the TCO criterion used for defining “the ozone hole”.¹⁶ The winter followed experienced ozone values below the ozone hole TCO criterion at Neumayer and South Pole. In 1987, an unprecedented 110 DU was

Fig. 4 Vertical structure of ozone loss saturation. Ozone mixing ratios measured by ozonesondes at different Antarctic stations at potential temperature (nine potential temperatures) levels 375–575 K (~12–23 km) in October during the 1979–2017 period. The solid horizontal line connects the mean ozone values in each year. The horizontal dashed line represents 0.1 ppmv. The vertical dashed lines represent year 1987 and 2000

measured at the South Pole, and about 150 DU at Neumayer and McMurdo. Ever since, the South Pole and Neumayer soundings continued to measure around 120 DU except for the warm winters. On average, the total columns show around 220 DU in 1982, 1983 and 1985, but about 80–150 DU in other winters. Therefore, the TCO measurements attest the fact that the loss saturation has undoubtedly begun in 1987 and its healing started in 2001.

Trends in ozone loss saturation occurrences

To find the changes in ozone loss saturation with time, the saturation threshold reached at each altitude (from 375 to 575 K, nine altitudes) for each station from 1979 to 2017 is analysed. We divided the number of saturation occurrences by total number of observations in the respective years to find the proportion (in %) of loss saturation measurements. Figure 6 shows the percentage of saturation occurrences at different vertical levels in each year during the ozone hole period, from September through November, to include all saturation episodes in spring. These analyses, therefore, show the history of ozone loss saturation in Antarctica, as they show the onset, progress, and peak of loss saturation, and emergence of ozone hole recovery. For instance, at 375 K, the loss saturation occurrences show about 5% until 1990, rose to 45% by 1992, and stayed around 15–50% in 1992–2001. The loss saturation gradually decreased to 1% by 2014. Similarly, the loss saturation frequency increased from about 2% in 1985 to 60% in

2001 at 400 K, and from about 15% in 1987 to 52% in 2000 at 425 K (~15 km). However, the loss saturation occurrences show a steep decrease, such as from 60% to 1% (~4.9%/year) at 400 K and from 52% to 2% (~4.1%/year) at 425 K from 2001 to 2017. The temporal evolution of loss saturation frequency is very similar at other altitudes and in the satellite measurements (Figure S5). At 400 K, for example, the satellite measurements show about 6% in 1987 and that increased to 25% by 2001. It shows a clear negative trend from 2001 onwards at all altitudes, such as from 20% in 2001 to 2% in 2017 at 400 K. Although the trend in loss saturation frequency is no different at other altitudes, the highest frequency was observed in 2015 in the satellite measurements.

We also examined the loss saturation occurrences at all altitudes in September–November from ozonesonde and satellite measurements (Figure S6). As shown by the ozonesonde measurements, the loss saturation was about 1% in 1985, 2–3% until 1991 and then it increased to 21% by 1992. The number of saturation measurements stayed around 15–20% until 1996, rose to 29% by 1998 and again increased to 34% by 2001. The proportion of saturation was highest (36%) in the very cold winter of 2006. It is also evident that loss saturation started to subside after 2001. The satellite measurements confirm the ozonesonde observations of loss saturation, albeit with smaller amplitude due to their relatively coarser vertical and horizontal resolution.

The box plots made from ozonesonde measurements clearly depict values below 0.1 ppmv from 1987 onwards, as illustrated in

Fig. 5 Ozone column changes in the loss saturation layer. Total column ozone (TCO) distribution inside the vortex measured by TOMS/OMI and the partial column ozone computed from the ozonesonde measurements over the ozone loss saturation layer 400–500 K (~13–20 km) in the vortex in October during the 1979–2017 period. The solid horizontal line connects the mean ozone values in each year. The horizontal dashed lines represent 220 DU (i.e. the ozone hole criterion) in the upper panel and 10 DU in the lower panel. The vertical dashed lines represent year 1987 and 2000

Fig. 6 Frequency of loss saturation occurrences. The percentage of number of saturation occurrences (the number of loss saturation occurrences divided by the total number of measurements) at different altitudes in the lower stratosphere at 375–500 K (~12–20 km) estimated from the ozonesonde measurements in spring (September–November). The vertical dashed lines represent year 1987 and 2000. The non-saturation years are not connected with the lines, to easily identify those events

Fig. 7. The 1%-whiskers show a progressive increase in the number of saturation occurrences at 400 K from 1987 onwards. The 1%-whiskers also show the largest ozone values in 1988 and 2002, and the smallest in 1992 and 1996 (very cold winter). Nevertheless, the whiskers show a gradual reduction in the number of saturation occurrences in the post-2001 period. Similarly, the satellite measurements illustrate the first appearance of loss saturation in

1987 (Figure S3, 400 K) and the healing indicated by its fewer occurrences in the 2001–2017 period. It is evident from both analyses that the loss saturation has started to decrease substantially after 2001, in agreement with the decline in ODSs.

Henceforth, this study reveals clear observational indications that the Antarctic ozone inside the loss saturation altitudes is increasing and recovering, although the saturation of ozone loss is

Fig. 7 Trends in ozone loss saturation. The box-whisker plot of ozone mixing ratios at 400 K (~13 km) as measured by the ozonesondes in the Antarctic in Spring (September–November) from 1979 to 2017. The box represents 25–75% and whiskers represent 1% and 100%. The mean is shown in red and median in black. The horizontal dashed line represents 0.1 ppmv

expected to continue to occur in very cold winters due to the still high levels of ODSs in the stratosphere, as the reduction processes of atmospheric burden of halogens are comparatively slow.^{23–25} The disappearance of near-zero ozone concentrations between 12 and 21 km is a key milestone in the recovery of Antarctic ozone and it would also mark the return of ozone to 1980 levels, as 1987 is the start year of occurrence of loss saturation.²⁶ Since there are already significant changes in the southern hemispheric climate owing to the Antarctic ozone loss,²⁷ the recovery from loss saturation is very likely to affect that. Although not discussed the policy aspect in detail, the recovery indicated in the loss saturation layer robustly suggests that the Montreal Protocol has definitely saved the ozone layer and climate of the southern hemisphere.

METHODS

In this study, ozonesonde measurements from 10 Antarctic stations are used.²⁸ The soundings include all available observations namely from the South Pole, Neumayer or Georg-Forster/Novorolaska, Syowa, McMurdo, Marambio, Davis, Maitri and Mirny, and deployment of the stations are shown in Figure S7. Since the Antarctic vortex is strong and stable for more than 6 months from June through November in most winters, the stations are always inside the polar vortex. In addition, we have selected the measurements exclusively inside the vortex using the Nash et al.¹⁸ criterion. The frequency of individual station measurements is very small in a month, yet there are sufficient measurements during the ozone hole period and have used all publicly available measurements from the region to date. Although the historic stations have measurements from 1960s, this study concentrates different aspects of ozone loss saturation since the 1980s. The ozonesonde data include both electrochemical cell and carbon-iodine type of measurements, which insignificantly differ in absolute values and hence, are suitable for our studies.²⁹ The ozonesonde measurements in general have an accuracy of about 5% in the lower stratosphere.³⁰ Studies have taken different values for the detection limit of ozonesondes, such as 0.4,¹³ 2,³¹ 10,³² and 15 ppbv,³² and we have considered 10 ppbv as the detection limit. Individual ozonesonde profiles from each station are checked manually to exclude any unexpected erroneous values or profiles. The soundings with bursting altitude below 450 K are excluded from the analysis. The measurements with corrupted metadata or bulb temperature are also omitted.

Apart from the ozonesonde measurements, we have also used the satellite measurements of SAGE-II and Aura MLS. These data have comparatively lower vertical resolution and hence, the fine scale features presented with the ozonesonde can be compared qualitatively. The period of analyses of both satellite data is 1985–2003 for SAGE-II and 2004–2017 for Aura MLS. The SAGE-II ozone retrievals have an accuracy of about 5–10% at 15–45 km, depending on altitude. The Aura MLS measurements have a vertical range of 12–73 km and their accuracy is about 5–10% between 16 km and 60 km. The vertical resolution of SAGE-II and Aura MLS ozone is about 1 km and 3 km, respectively, and both measurements have a horizontal resolution of about 200 km.^{33,34} The TOMS measurements aboard three different satellites (Earth Probe, Meteor and NIMBUS 7) from 1979 to 2004 and OMI on-board the Aura satellite from 2005 to 2017 are considered for the comparisons. The accuracy of the

TCO data is about 5–10%.^{22,35} The TOMS/OMI daily retrievals are checked for any unreasonable values and are excluded, if found.

The analyses presented are in terms of volume mixing ratios (VMRs) and the partial columns in DU. For the conversion from VMR to DU, temperature and pressure data are taken from the European Centre for Medium-Range Weather Forecasts (ECMWF) analyses and are interpolated to the station co-ordinates, if not provided with ozonesonde data files. The same meteorological data are used for conversion of the altitude levels (pressure or geometric altitude) to potential temperature (θ , K) levels in 25 K intervals between 275 K (~7 km) and 950 K (~35 km). The EQL of each profile at each altitude is determined with the ECMWF data. The profiles having EQL values above 65°S at each altitude level are considered inside the polar vortex for further analyses. The EQL is a good measure of vortex edge and a cut off of $\geq 63^\circ$ S is found to be apt for the Antarctic ozone trend or ozone loss studies [³⁵, references therein]. Therefore, we have taken $\geq 65^\circ$ EQL for our analyses to confirm the considered measurements are inside the vortex. Since the Antarctic vortex is more or less stable in most years, this criterion with respect to EQL, which is also a measure of PV, is a good criterion to segregate vortex measurements.

To test the ability of satellite measurements to diagnose the loss saturation, we interpolated the SAGE-II and Aura MLS profile measurements to the ozonesonde stations to make the overpass measurements. The October measurements at 400 K from 1985 to 2017 from SAGE-II^{36,37} and Aura MLS^{38,39} do not show loss saturation until 1990 at McMurdo (Figure S8). The next appearance of loss saturation was in 1997 at McMurdo and Syowa, the continued appearance of loss saturation values started in 1996, and the loss saturation was never attained in 2002. These suggest that the overpass measurements do not represent the region, as the satellite coverage is poor at the high polar latitudes. Nevertheless, the overpass measurements for the recent decades also show significant reduction in the occurrences in loss saturation frequency, as analysed from the measurements for all altitudes for the September–November period (Figure S9).

Code availability

The analyses codes are available on request.

DATA AVAILABILITY

The ozonesonde data are available from <http://www.ndacc.org> and <http://www.woudc.org>. The SAGE-II data were obtained from the NASA Langley Research Center Atmospheric Science Data Center, and the data are available on <https://eosweb.larc.nasa.gov/>. The TOMS data are from <http://toms.gsfc.nasa.gov>, and OMI and Aura MLS data <http://avdc.gsfc.nasa.gov/pub/data/satellite/Aura/OMI/>. The ECMWF data are available on <https://www.ecmwf.int/>. The authors are happy to share the data used in the manuscript up on request. Since the ozonesonde and satellite data used are already freely available on public domains, the analysed data can also be provided for any scientific study.

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AUTHOR CONTRIBUTIONS

J.K. and P.J.N. designed the research and performed the preliminary analysis. P.K. performed the data analysis and extended the data up to 2017. J.K. and P.K. drew the figures. J.K. wrote and revised manuscript with inputs from P.K., P.J.N. and P.C.P.

ADDITIONAL INFORMATION

Supplementary information accompanies the paper on the *npj Climate and Atmospheric Science* website (<https://doi.org/10.1038/s41612-018-0052-6>).

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