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Arctic on the verge of an ozone hole?

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Abstract. Severe vortex-wide ozone loss in the Arctic would expose nearly 650 million people and ecosystem to unhealthy ultra-violet radiation levels. Adding to these worries, and extreme events as the harbingers of climate change, clear signature of an ozone hole (ozone column values below 220 DU) appeared over the Arctic in March and April 2020. Sporadic occurrences of ozone hole values at different regions of vortex for almost three weeks were found for the first time in the observed history in the Arctic. Furthermore, a record-breaking ozone loss of about 2.0–3.4 ppmv triggered by an unprecedented chlorine activation (1.5–2.2 ppbv) matching to the levels of Antarctic ozone hole conditions was also observed. The polar processing situation led to the first-ever appearance of loss saturation in the Arctic. Apart from these, there were also ozone-mini holes in December 2019 and January 2020 driven by atmospheric dynamics. The large loss in ozone in the colder Arctic winters is intriguing and that demands rigorous monitoring of the region. Our study suggests that the very colder Arctic winters in near future would also very likely to experience even more ozone loss and encounter ozone hole situations, provided the stratospheric chlorine levels still stay high there.

1 Introduction

Apart from its significance of shielding the harmful ultra-violet (UV) radiation reaching the surface of earth, stratospheric ozone is a key component in regulating the climate (e.g. Riese, et al., 2012). Changes in stratospheric ozone are always a big concern for both public health and climate (WMO, 2018; Bais et al., 2019). Due to unbridled emissions of Ozone Depleting Substances (ODS) to the atmosphere since the 1930s stratospheric chlorine peaked in the polar stratosphere in the early 2000s (Newman et al., 2007; Engel et al., 2018; WMO, 2018). Therefore, the first signatures of polar ozone loss appeared over Antarctica by the late 1970s (Chubachi et al., 1984; Farman et al., 1985) and it peaked to saturation levels in late 1980s (Kuttippurath et al., 2018). Recent studies have demonstrated the effectiveness of Montreal Protocol in reducing halogen gases, with a corresponding positive trend in ozone in Antarctica (Salby et al., 2011; Kuttippurath et al., 2013; Solomon et

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al., 2016; Chipperfield et al., 2017) and in northern mid-latitudes (Steinbrecht et al., 2014; Nair et al., 2015; Weber et al., 2018). However, a positive trend in the Arctic ozone is not reported yet because of the large dynamically driven inter-annual variability of ozone there (Kivi et al., 2013; WMO, 2018).

Antarctic winters are very cold and the ozone hole is a common feature of these winters since the late 1970s. There were winters with very low temperatures with stronger vortex that showed relatively larger loss in ozone, such as the winters of 1996, 2000, 2003, 2006 and 2015 (Bodeker et al., 2005; Chipperfield et al., 2017). There were also winters with high temperatures and smaller ozone losses as in the case of 1998, 2002, 2012 and 2019 (Müller et al., 2008; de Laat et al., 2010; Kuttippurath et al., 2015). Yet, the inter-annual variability of ozone loss in the Antarctic is very small in the recent decades. On the other hand, colder winters with large losses of ozone (e.g. > 1.5 ppmv of loss) are rare in the Arctic (Rex et al., 2015). For instance, the Arctic winters 1995, 1996, 2000, 2005 and 2011 were very cold with large loss of ozone, up to 25–30%, but are still significantly smaller than the 40–55% ozone loss occurrence in the Antarctic (Kuttippurath et al., 2013; Pommereau et al., 2018).

The Arctic vortex is short-lived and frequently disturbed by planetary wave activity. The vortex normally strengthens by mid-December or early January and dissipates by mid-March. Major and minor warmings are common features of Arctic winters. However, the vortex dissipates and ozone loss terminates when the major warmings occur there. In an earlier study, Kuttippurarth et al. (2012) observed an increasing trend in major warmings, and ozone loss is found to be proportional to the timing of the major warmings, as early winter warmings produce smaller loss in the Arctic winters. Since 1979, during the satellite era, there were two extreme winters with large loss of ozone in the Arctic; 2005 and 2011 (Coy et al., 1997; Feng et al., 2007; Horowitz et al., 2011). The occurrence of extreme events is a signature of climate change and so are the extreme cold winters with large loss in ozone (e.g. IPCC, 2007). Previous studies have postulated that the cold winters will get even colder with large loss in ozone (Sinnhuber et al., 2000; Rex et al., 2004; Chipperfield et al., 2005; Rider et al., 2013). Analyses of the past colder Arctic winters indicate that it is likely that the colder winters may experience large loss in ozone, as in the case of 2005, 2016 and 2011. Here, we show that the Arctic winter in 2020 was very cold with the largest ozone loss in the observational record and met the condition for an ozone hole for the first time, by using different data, methods and other assessment parameters.

2 Methods

We have used two ozone profile datasets from the satellites. The level 2 data from the

(i) Microwave Limb Sounder (MLS) v4.2 and

(ii) Ozone Mapping and Profiler Suite (OMPS) v2.5 (ozone).

(iii) We have also used the ozonesonde measurements from the Arctic stations at Alert (62.34° N, 82.49° W) and Eureka (79.99° N, 85.90° W). These measurements have an uncertainty of 5–10%.

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Three satellite-based total column ozone (TCO) data are also employed (level 3) to analyses the ozone hole:

(iv) Ozone Monitoring Instrument (OMI, DOAS v003),

(v) OMPS (v2.1),

(vi) Global Ozone Monitoring Experiment (GOME) (GDP4.8),

(vii) Modern-Era Retrospective analysis for Research and Applications (MERRA)-2 and

(viii) Brewer spectrometers from Alert and Eureka.

These total column measurements have an uncertainty of 2–5%. The ozone and other trace gas profiles are provided in pressure co-ordinates, which are converted to isentropic coordinates using the temperature data from the same satellite. We use the European Centre for Medium-Range Weather Forecasts (ECMWF) Reanalyses ERA5 potential vorticity (PV) on a 1°x1° grid to determine the vortex edge. The PV data are also converted to isentropic coordinates using the ER5 temperature data. We computed the equivalent latitude at each isentropic level at 5 K intervals from 350 to 800 K, and are then used to compute the vortex edge using the Nash et al. (1996) criteria. We use measurements inside the polar vortex for ozone loss analysis. The missing values in satellite measurements were filled with linear interpolation (poison_grid_fill).

We have taken ozone, ClO, HNO_3 and N_2O from the Aura MLS measurements. The ozone measurements at 240 GHz have a vertical resolution of 2–3 km, vertical range of 261–0.02 hPa and an accuracy of 0.1–0.4 ppmv. The vertical range of HNO_3 measurements is 215–1.5 hPa, vertical resolution is 2–4 km, with an accuracy of 0.1–2.4 ppbv, depending on altitude. The N_2O measurements are available for the 68–0.46 hPa vertical range, and 68 hPa roughly equivalent to 400 K isentropic level. The data were extrapolated up to 350 K by performing exponential fitting to N_2O vertical distribution at 400–600 K by considering the exponential change of N_2O with altitude. The accuracy of retrievals at 190 GHz is about 2–55 ppbv at this altitude range and the vertical resolution is about 2.5–3 km. The vertical resolution of ClO measurements at 640 GHz is about 3–3.5 km over 147–1 hPa, and the accuracy of measurements is about 0.2–0.4 ppbv. The measurements also have latitude-dependent bias of about 0.2–0.4 ppbv, depending on altitude (Livesey et al., 2013; Santee et al., 2008; Froidevaux et al., 2008).

The OMPS consists of three sensors that measure scattered solar radiances in overlapping spectral ranges and scan the same air masses within 10 min. The nadir measurements are used to retrieve ozone total column and vertical profiles (NP). The Limb Profiler (LP) measures profiles with high vertical resolution (∼ 2–3 km) and the LP retrievals are in good agreement with other satellite measurements and the differences are mostly within 10% (Kramarova et al., 2018). The OMPS TCO shows 0.6–1.0% differences with Brewer and Dobson ground-based TCO measurements across the latitudes, and are also biased

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+2% when the TCO is above 220 DU (Bais et al., 2014). The Global Ozone Monitoring Instrument (GOME-2) was flown on MetOp-A satellite in 2006. The GOME-2 ozone column has a positive bias in the northern high latitudes of about 0.5–3.5% (Layola et al., 2011). The Ozone Monitoring Instrument (OMI) TCO measurements have an accuracy of about 5% in the polar regions (Kroon et al., 2008; Kuttippurath et al., 2018). The Brewer spectrometers operates in the UV region and their ozone observations have an accuracy of about 5%.

The ozone loss is estimated using two different methods and four different data sets to make sure the analyses are correct and robust. The first method used is the widely used profile descent method, wherein the N_2O data are used for the calculations of air mass descent in the polar vortex. The reference profile of N_2O was taken from the month of December, and therefore, the loss calculations are presented from December (May for Antarctic) onwards. The second method used for the calculation of ozone is the passive tracer method, for which a passive odd-oxygen tracer is simulated using a CTM (Chemical Transport Model) and is subtracted from the measured ozone to find the ozone loss, as the changes in tracer is modulated only by the dynamics (Feng et al., 2005). We have used the SLIMCAT model for the tracer calculations (Chipperfield, 2006).

3. Results and discussion

3.1 The exceptional meteorology of the Arctic winter 2020

Figure 1 shows the times series of stratospheric meteorology in the Arctic winter 2020 compared to that of the longest winters 1997 and 2011. Time series of the meteorological parameters for all Arctic winters since 1979 are also shown (grey coloured curves) for comparison. In general, the temperatures are between 210 and 195 K. In 2020, the temperatures were about 195 K in December, 190–195 K in January–March and 195–205 K in April. However, the minimum temperature in late winter 2020 is generally lower than 195 K, about 115 days from December through early April. The temperatures are lower than those in the 2011 winter, and the temperatures in late March and April are lowest on the observational record. The temperatures are below 200 K in late December and early January, which is important for chlorine activation. The lower temperatures in late December through mid-March are key to large ozone loss and are a common phenomenon in winters with large loss of ozone (e.g. 1995, 2000, 2005 and 2011). Therefore, the higher temperatures were also the reason for the relatively small ozone loss in 1997; although it was a winter with a strong vortex up to the end of April. Since minor warmings (mWs) are very common in the Arctic polar winters, we also examined the occurrence of mW events with temperature at 90° (North Pole) and 60° N at 10 hPa and zonal winds at 60° N at 10 hPa. The analyses show a small increase in temperature on 5 February 2020 (i.e. a minor warming) and a corresponding change in zonal winds.

The temperatures were consistently lower than the nitric acid trihydrate (NAT) threshold of 195 K and therefore, large areas of Polar Stratospheric Clouds (PSCs) are observed from December to mid-February. The PSC area (APSC) was about 4 million km^2 in December at 460 K, but it doubled in January through mid-March. The APSC from mid-February to late

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March is also largest on the observational record (Figure 1). The low temperatures (i.e. lower than 188 K) also produced very high amount of ice PSCs at the end of January and early February (up to 4 million km^2) when the lowest temperatures in 40 years were recorded in the Arctic. This is the largest ice PSC ever observed in terms of its area, volume and number of days of appearance (i.e. frequency) in the Arctic and the area is twice that of the winter 2011. The PSC area shrunk to half of its area in late January and February, as the lower stratospheric temperature increased during the period. This was the only occasion that the temperature increased and PSC areas limited to below 4 million km^2 in the winter 2020. Note that the PSC area and volume were largest in 2016, not in 2020 (Figure S1) (Kirner et al., 2015).

The potential vorticity (PV) analyses at ~17 km (about 460 K potential temperature level) show that the polar vortex was very strong in the lower stratosphere in 2020. The PV values were consistently higher than the previous cold (i.e. 1995, 2000, 2005 and 2011) and long-lasting (e.g. 1997 and 2011) winters in March and April. This indicates that the winter 2020 had the strongest vortex in the recent history, as demonstrated by the PV time series of different Arctic winters (Figure 1, second panel, left). However, the zonal winds were strongest in 1997 during the March-April period. The Arctic vortex in any winter would be frequently disturbed by planetary waves that emanate from the troposphere. In general, planetary wave numbers 1, 2 and 3 are mostly responsible for the momentum transfer to the stratosphere. This dynamical activity would increase the temperature in the lower stratosphere and trigger stratospheric warmings. The warmings can be minor or major, depending on the strength of wave activity, increasing the polar temperature and eventually disturbing the polar vortex. The vortex can be distorted, displaced, elongated and even split in two in accordance with the potency of momentum imparted by the waves. When the polar vortex is disturbed, the ozone loss will be smaller and the final warming can be as early as in late February or early March, as for most Arctic winters (e.g. Manney et al., 2003; Kuttippurath et al., 2012; Goutail et al., 2015). The diagnosis with heat flux and the eddy heat flux associated with waves demonstrates that the momentum transported from the troposphere to stratosphere was very weak in the winter 2020 (in the range of -20 to 30 Km s^{-1}), and the heat flux values are zero or negative (e.g. -10 Km s^{-1} in February) during most part of the winter. These analyses are also in agreement with the eddy heat flux computed for the waves, as they also show smaller wave momentum to the stratosphere. In short, the eddy heat flux and wave heat flux show smaller values in January-April; indicating the reason for the less disturbed long-lasting vortex in 2020.

The potential vorticity analyses show a strong and large vortex in early December (Figure 2). The vortex began to grow and occupied the entire polar region by early January. The lowest temperatures of the past 40 years was recorded by the end of January and the vortex was exceptionally strong and large. The mW distorted and elongated the vortex in early February, but the vortex was still strong and continued to be intact until the last week of April 2020. The extraordinary persistence of a strong and undisturbed Arctic vortex in March and April is evident in the PV maps.

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3. 2 Strong air mass descent and associated ozone distribution

Figure 3 shows the distribution of ozone, ClO, N_2O, HNO_3 and the ozone loss estimated for the winter 2020 using satellite observations. We use the measurements from MLS on the Aura satellite (Livesey et al., 2015). The MLS measurements are one of the best currently available data for polar ozone loss analyses, as the instrument provides measurements of some key ozone-related chemistry trace gases such as ClO, N_2O and HNO_3 to delineate the features of chlorine activation, vortex descent and denitrification, respectively (Manney et al., 2020). The ozone distributions in the vortex show < 1.0 ppmv in December, slightly higher values of about 1.5 ppmv in February and smaller than 1.0 ppmv from Mid-March to the end of April at 400 K. The measurements show exceptionally small values of ozone, about 0.5 ppmv or below, during the period mid-March through to the end of April at 350–450 K. The ozone values show < 2.5 ppmv from December to mid-January, < 2 ppmv January and February and < 1.0 ppmv in March-April at 350–450 K, and about 2–4 ppmv above 500 K; suggesting an unusual reduction in ozone in December and late January. The ozone values are about 3–4 ppm above 550 K throughout the winter; implying little reduction in ozone there. The unusual feature here is the extremely small ozone of 1.0 ppmv in early December and in March–April below 450 K (about 16 km). This reveals huge loss of ozone in the lower stratosphere and therefore, we have quantified the ozone loss for the winter. We use the profile descent method using the trace of air motions N_2O and is a widely used method for ozone loss estimation (Bremer et al., 2002; Rex et al., 2002; Jin et al., 2006).

For instance, the MLS measurements show N_2O values were 250 ppbv at 400 K, 150 ppbv at 500 K and 50 ppbv at 600 K in December. The N_2O observations show strong air mass descent with values down to 100 ppbv at 400 K and about 25–50 ppbv above 500 K in early February. Again, N_2O values exhibit below 50 ppbv in late March at 400 K. The N_2O distributions show below 50 ppbv at all altitudes from early February onwards; suggesting substantial dynamic descent in the stratosphere. When a particular altitude is considered, e.g. the 450 K potential temperature level, the N_2O values show 160 ppbv in early December, 100 ppbv in early January, 50 ppbv in early February and smaller than 50 ppbv thereafter. On the other hand, the N_2O distributions show 50 ppbv in early December and below that value afterwards at 500 K. The severe air mass descent in this winter is further depicted in Figure S2, where monthly correlation between ozone an N_2O are presented.

3.3 Ozone loss and mini-holes in December and January

There were vortex-wide PSC occurrences in the first week of December, about 2-4 million km^2 in area (APSC) and about 70 million km^3 in volume (VPSC). The APSC and VPSC dropped significantly afterwards and then gradually increased again by mid-December to 10 million km^2 and 120 million km^3, respectively. An unusual increase in activated chlorine is observed during the first week of December in conjunction with the appearance of PSCs. The temperatures began to decrease from 198 K in mid-December to 187 K by the end of January. The chlorine activation peaked and showed record levels of ClO, about 1.5–2.0 ppbv at 400–600 K, during this period. The chemical ozone loss began in early January with about 0.5 ppmv

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and increased to 1.5 ppmv by the end of January below 500 K. The loss above that altitude is always lower than 0.5 ppmv, which shows that the ozone loss is restricted to the altitudes below 21 km (i.e. 550 K).

In general, the ozone loss starts in December in the middle stratosphere and then gradually progresses towards the lower stratosphere by January. The loss would be below 0.5 ppmv in December and about 0.5–1.0 ppmv in January in the lower stratosphere in cold Arctic winters. However, in the Arctic winter 2020, the ClO and ozone loss show unusually high values of about 1.5–2.0 ppbv and 1.5–2.0 ppmv, respectively. Since chlorine activation of this scale requires sunlight, and there is no sunlight in the Arctic vortex in early winter, the appearance of huge amounts of ClO during this period is surprising. The only possibility to have such high-levels of chlorine activation is the displacement of vortex to the sunlit latitudes. The analyses of vortex position in early December and late January (Figure 2) reveal that the vortex was at 55°–60° N. Therefore, a strong polar vortex, very low temperatures, large volumes of PSCs and shift of vortex to the sun light part of mid-latitudes caused the unprecedented chlorine activation and ozone loss in the first week of December and late January.

In addition to the ozone loss inside the vortex, there is another interesting phenomenon in December and January. The analyses of TCO show that there were ozone holes of about 300–700 km^2 size during the first week of December (1–6 December 2019) and on 26 January 2020 (Figure 4). The lowest TCO measured of the winter was also on the latter date. However, the ozone holes were not inside the vortex, but in the mid-latitudes. A detailed analysis with TCO, PV, temperature and ClO reveal that those ozone holes were dynamically driven, as there was rapid air mass transport to the southern Arctic in early December and late January. These ozone hole occurrences due to rapid changes in weather patterns are generally called “ozone mini holes” and they return to normal levels of ozone in few days (e.g. Rieder et al., 2013). We used the HYSPLIT trajectory model to find the air mass transport at three different altitudes (17, 18 and 19 km) in the lower stratosphere, where the mini-holes are found (Figure 4, right panels). The air mass exported from mid- and low latitudes has very low PV values, lower temperature and high ClO. It suggests that the TCO transported from mid-latitudes triggered the ozone “holes” (ozone values < 220 DU). To further examine the lower ozone loss values to the levels of an ozone hole outside the vortex, we selected two ozonesonde measurements in the region (Alert: 62.34° N, 82.49° W and Eureka: 79.99° N, 85.90° W), and are shown in bottom panels for selected dates in December and January. These measurements show significant reduction in ozone Coy et al., 1997; Feng et al., 2007; Horowitz et al., 2011between 12 and 18 km; confirming the findings from satellite total column measurements.

It should be mentioned that there was already large chemical loss of ozone inside the Arctic vortex in early December and late January owing to the conventional polar ozone loss chemistry (as shown in Figure 3). However, the ozone mini-holes that appeared outside the vortex were primarily triggered by the dynamics. We cross-checked TCO from OMI (Bias et al., 2014), OMPS (Flynn et al., 2014), GOME-2 (Layola et al., 2011) and MERRA-2 (Gelaro et al., 2017), and found that the ozone mini-holes were present all these TCO datasets.

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3.4 Prolonged chlorine activation and chemical ozone loss

When the Arctic winters are very cold, chlorine activation occurs in the lower stratosphere at 400–500 K in January and February there. In 2011, the chlorine activation was observed up to the end of February and was intermittent with a peak value of about 1.6 ppbv, and was mostly at 400–500 K (e.g. Manney et al., 2011; Kuttippurath et al., 2012; Livesey et al., 2015; Griffin et al., 2018). Conversely, in the Arctic winter 2020, there was continuous and sustained chlorine activation from December to early April, except during the mW periods of mid-December and early February. The ClO values are also 0.5 ppbv larger than those observed in the winter 2011. Therefore, strong chlorine activation was observed in March-April with ClO values of about 1.0–1.6 ppbv at 400–550 K and the peak ClO value is about 2.1 ppbv.

The minor warming (Figure 1) made a break in chlorine activation (Figure 3 for ClO) in early March. Nevertheless, the temperature decreased shortly thereafter, which produced continued chlorine activation until early April at 400–550 K. The ozone loss deepened in March and peaked by the end of March, and showed the maximum of about 1.5–3.4 pmv at a broader altitude range, up to 500 K. The ozone loss above that altitude (i.e. 550 K) was about 0.5–1 ppmv, which is still larger than that of any other Arctic winter. In fact, the loss of 1.0 ppmv is the peak loss observed in normal or moderately cold winters of the Arctic (Kuttippurath et al., 2013); suggesting the severity of ozone loss even at the higher altitudes in this winter. The maximum loss was recorded at the end of March to the end of April, about 2–3.4 ppmv at 400–500 K and about 0.5–1.5 ppmv at 500–600 K. Furthermore, when compared to the early winter values, the late winter low HNO_3 values suggest very severe denitrification, about 2–4 ppbv in the same period at 350–450 K (e.g. Manney t al., 2020). The HNO_3 values in the lower stratosphere in March–April are about 60–80% lower than that of the December–February values at the same altitude levels (Pommereau et al., 2018; Lindenmaier et al., 2012). The gravest denitrification was in December, with values of about 0–2 ppbv below 400 K and 4–6 ppbv at 400–450 K. Therefore, high chlorine activation and strong denitrification (as deduced from the HNO_3 analyses shown in Figure 3) provided an unprecedented situation for large ozone loss of about 2–3.4 ppmv in the lower stratosphere in March–April.

Since the ozone loss in 2020 is exceptionally large, we have employed another set of measurements to estimate ozone loss to reconfirm that the derived results are robust. The loss estimated from OMPS measurements together with other analyses are shown in Figure 5 (left). The maximum ozone loss profile extracted from the OMPS data show very good agreement with that from the MLS measurements for the Arctic winter 2020. The peak ozone loss values show about 2–2.8 ppmv in the lower stratosphere below 550 K. Since the maximum ozone loss profiles are averaged for few days, the loss values are slightly lower than that of MLS measurements. The lower stratosphere shows similar ozone loss values, but the loss above 500 K shows slightly smaller values (0.1–0.5 ppmv) due to the low bias of OMPS measurements at these altitudes as compared to the MLS measurements (Kramarova et al., 2018). The comparison with OMPS confirms that the method adopted for ozone loss is robust. Our estimates are in good agreement with that of Manney et al. (2020) and Wohltmann et al. (2020), as they also derive a loss of about 2.3–2.8 ppmv below 450 K from the MLS measurements inside the vortex.

3. 5 The Arctic ozone loss in the context of other Arctic winters

Arctic winters are normally warmer and occurrences of PSCs are sparse and infrequent. Therefore, chlorine activation and ozone loss is limited to the winters with very low temperatures in December–February (Tilmes et al., 2014; Goutail et al., 2015; WMO, 2018; Newman et al., 2008; Kuttippurath et al., 2012). The ozone loss observed in warm winters (e.g. 2006 and 2009) is about 0.5–0.7 ppmv, moderately cold winters (e.g. 2008 and 2010) is about 1.0–1.2 ppmv and very cold winters (e.g. 2005) is 1.4–1.6 ppmv (e.g. WMO, 2018). However, the ozone loss in the winter 2011 was about 1.0 ppmv (or 30–40 DU), which is higher than that of other Arctic winters (about 2.1–2.3 ppmv or 100–100 DU) and was similar to the loss found in the warm Antarctic winters (e.g. 1988 and 2002) (Manney et al., 2011; Kuttippurath et al., 2012; Feng et al., 2015; Pommereau et al., 2018). We applied the same loss estimation method to the measurements for the Arctic winter 2011 to compare with that of the Arctic winter 2020. This would also test veracity of the loss estimation procedure and the results are shown in Fig. 5.

The peak ozone loss in the Arctic winter 2011 shows about 2.1 ppmv, which is in very good agreement with all other available analyses for that winter (WMO, 2014, 2018; Griffin et al., 2018; Livesey et al., 2015). However, the ozone loss in the Arctic winter 2020 is about 0.7 ppmv higher than that in 2011, about 2.8 ppmv. The difference in ozone loss between the winters is negligible above 480 K. Therefore, it is evident that the ozone loss in the Arctic winter 2020 is undoubtedly the largest on the record and is significantly higher than that of any previous Arctic winter.

Furthermore, we applied another loss estimation method to test robustness of the extreme ozone loss values; the passive method that uses a passive tracer (i.e. no chemistry) simulation. We have used the well-known and widely used TOMCAT/SLIMCAT model simulations for the tracer calculations (Chipperfield et al., 1999; Dhomse et al., 2019). The ozone loss computed with the passive method shows the peak value of about 2.3–2.5 ppmv at about 450 K in the Arctic winter 2020 (Figure 5, Second panel from the left). This ozone loss is slightly higher than that of the Arctic winter 2011, about 0.2 ppmv. It is also observed that the ozone loss in 2020 is higher than that of 2011 below 475 K, but the loss estimated in latter winter exceeds about 0.3–0.5 ppmv above 475 K up to 700 K (e.g. Manney et al.,2020; Wholtmann et al., 2020). However, these ozone loss estimates are lower than that estimated with the descent method, about 0.5–0.7 ppmv depending on altitude. The analysis with ozone and N_2O from the model indicates that modelled ozone is higher than (about 1–1.5 ppmv) the measurements at these altitudes, which could be due to the slower dynamical descent in the model.

It is clear that the ozone loss in 2020 is the largest among Arctic winters. Therefore, we also examined the evolution of chlorine activation in terms of the amount of ClO in each Arctic winter, as the total chlorine is decreasing in the stratosphere

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due to the effect of Montreal Protocol (e.g. Strahan et al., 2017; WMO, 2018; Dhomse et al., 2019) and we expect a corresponding response in ozone loss in the polar winters. Figure 6 shows the MLS ClO observations in each winter since 2005. The analyses show that the chlorine activation was very severe and continuous for about four months in 2020. However, the highest ClO was observed in winter of 2016, in February. Many colder winters had the ClO values around 2 ppbv as found in 2020, but the sustained chlorine activation that observed in 2020 was unique. Although the high ClO values in March were also observed in 2011, the chlorine activation was not as severe as in 2020 in early winter (December – January). On the other hand, the unprecedented chlorine activation observed in 2016 was more episodic, such as in mid-December, mid-January to early February and late February. Therefore, the continuous and severe chlorine activation from December through March was the key for the record-breaking ozone loss in 2020. Figure 6(b) and (c) further illustrate that the peak ClO profiles or the time series of average ClO for the entire winter will not reveal the depth of chlorine activation.

3.6 The Arctic ozone loss equals the levels of the Antarctic ozone loss

The peak ozone loss in the Antarctic happens at around 500 K and the loss is severe from 400 to 600 K for five months continuously from August to November (Tilmes et al., 2006; Huck et al., 2005; Sonkaew et al., 2013; Kuttippurath et al., 2015). In contrast, the Arctic winters are normally short and maximum ozone loss occurs at around 425–475 K for a period of about two months, from mid-January to mid-March (e.g. Kuttippurath et al., 2010; Manney et al., 2004). The ozone loss in the Arctic is restricted to the altitudes below 500 K. The ozone loss in the Arctic winter 2020 is very high and therefore, we compare the ozone loss with that in the Antarctic winters 2015 and 2019. The Antarctic winter 2015 was one of the coldest and 2019 was one of the warmest, and therefore, the assessment would give an upper and lower bound of ozone loss estimate for the Arctic winter 2020.

The peak ozone loss estimated using the vortex descent method is about 2.8 ppmv at 480 K in the Antarctic winter 2015 and about 2.3 ppmv at 490 K in 2019 (Figure 5). The ozone loss in Antarctic 2015 shows consistently higher values (about 0.1–0.5ppmv) than that of 2019 up to 550 K, and the loss is similar above that altitude in both winters. The ozone loss is about 1.0 ppmv at 370 K, and 2.6 ppmv at 460 K, 1.5 ppmv at 550 K, 0.5 ppmv at 650 K and it terminates at 700 K in the Antarctic winter 2015. In the Arctic winter 2020, the ozone loss shows about 0.3 ppmv at 370 K, 2.0 ppmv at 430 K and 480 K, 1.5 ppmv at 550 K and loss terminates above that altitude. The peak ozone loss is about 2.3 ppmv and is at 460–470 K. On the other hand, the loss in the Antarctic winters above 470 K is very large and reaching up to 700 K. The peak ozone loss in the Arctic winter 2020 is about 2.8 (2.3) ppmv and is at 460–470 K. This is also the main difference between the Arctic and Antarctic ozone loss, as the broader and larger ozone loss above the 470 K in the Antarctic. The difference is almost 1.0 ppmv above the peak ozone loss altitude. Therefore, the ozone loss in the Arctic winter 2020 is either equal or larger than that of the Antarctic winter 2019 below 470 K, but the loss is smaller than that of the Antarctic winters above 525 K.

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We applied the passive method also to further examine the estimated loss in the Arctic and Antarctic winters (Figure 5, second panel from the left). The ozone loss estimated with the passive method exhibits smaller values in the lower stratosphere in comparison with that derived from the descent method. The loss is about 0.2 ppmv at 350 K, 1.6 ppmv at 400 K and 2.3 ppmv at 450 K in the Arctic winter 2020. The peak loss is recorded at 450–460 K and the loss decreases with altitude, about 1.5 ppmv at 500 K and 0.1 ppmv at 530 K. In the Antarctic winter 2019, the ozone loss shows similar values as that of the Arctic winter 2020 at 370–420 K, but slightly smaller than that of the Arctic winter at 420–470 K. The maximum ozone loss in Antarctic winter 2019 is estimated at 470 K, about 2.3 ppmv, and about 0.5–1.5 ppmv above that altitude, which is higher than that of the Arctic winter 2020. Furthermore, the Arctic ozone loss halts at about 550 K, whereas the Antarctic ozone loss at this altitude is as high as 1.5 ppmv. In the Antarctic winter 2015, the ozone loss is about 1.0 ppmv at 370 K, 2.0 ppmv at 400 K and the peak loss of about 2.8 ppmv at 475 K. The loss gradually decreases with altitude, such as 2.1 ppmv at 500 K, 1.5 ppmv at 550 K, 1.0 ppmv at 600 K and 0.5 ppmv at 650 K. The ozone loss in the Antarctic winter 2015 is thus, higher than that of the Antarctic winter 2019 and the Arctic winter 2020, about 0.5–1.5 ppmv, depending on the altitude. The assessment further gives strong evidence that the peak ozone loss in the Arctic winter 2020 equals to that of the warm winters of Antarctic (e.g. 2019). The loss estimation method can have uncertainty in the range of 3–5%, depending on the winter months. For instance, the monthly mean ozone loss and its standard deviation for each winter month of 2020 are shown in Figure S3. A complete error analyses of the ozone loss method is already presented in Kuttippurath et al. (2010).

3.7 The first appearance of ozone loss saturation in the Arctic

Ozone loss saturation is a common feature of Antarctic winters since 1987 (Kuttippurath et al., 2018; Jin et al., 1996). However, as compared to the Antarctic, the Arctic winters are relatively short (December to March), stratospheric temperatures are about 10 K higher, occurrence of PSCs are infrequent, denitrification is modest and thus, ozone loss is moderate. Therefore, the Arctic never encountered the ozone loss saturation (i.e. the complete loss of ozone at some altitudes in the lower stratosphere between 400 and 550 K) there. Apart from these, the ozone loss normally happens only up to 25–30% in the Arctic winters and henceforth, the loss saturation was unexpected for the Arctic conditions. Figure 5 (right) shows the ozone profile measurements by ozonesondes at two Arctic stations, Alert (82.50° N, 62.33° W) and Eureka (80.05 N, 86.42 W), on selected days. The ozone profiles measured at selected Antarctic stations are also shown for comparisons. In general, the ozone loss saturation in Antarctica occurs at the altitude between 400 and 500 K (e.g. Davis: 68.6°S, 78.0°E and Marambio: 64° S, 56° W), and the altitude range would go up to 550 K for the stations that are always inside the vortex, as shown for Syowa. The ozone loss observed at Davis and Marambio is always smaller than that at Neumayer, South Pole and Syowa. Therefore, ozone loss saturation is also comparatively infrequent and occurs at limited altitudes in the Antarctic. Here, the ozonesonde measurements at Alert (on 08 April 2020) show loss saturation at the altitudes 420–475 K (e.g. Wilka et al., 2021). The measurements at Eureka (on 10 April 2020) show loss saturation with about 99% ozone loss at altitudes between 420 and 460 K. The time series of ozone measurements, as analysed from the available measurements, show that the ozone loss saturation occurred at these station in early April (Figure S4). The vertical shading in the figure for 0.2 ppmv shows the ozone loss saturation criterion with respect to the ozone volume mixing ratios (Smit et al., 2007), and the ozonesonde measurements have an uncertainty of 5–10%. Yet, the ozone measurements at Alert and Eureka are in the saturation limit and thus also provide first evidence for the occurrence of ozone loss saturation in the Arctic. The loss saturation suggests that the Arctic has entered an exigent climate change scenario and our analyses are consistent with the analyses of Wohltmann et al. (2020), who also report about 90–93% loss of ozone in the 450–475 K altitude range in this winter.

3.8 The first-ever ozone hole in the Arctic?

Since the ozone hole is defined with respect to TCO measurements (i.e. below 220 DU), we analysed the TCO measurements, which are shown in Figure 7. It shows the lowest TCO measurements made in the Arctic polar region in the winter 2020 by three different satellite instruments, OMI, OMPS and GOME. As shown in the figure, the OMI measurements show TCO below 300 DU for almost all winter months inside the vortex, as defined by the Nash et al. (1996). The measurements show around 230 DU in early December, about 260 DU in January, about 218–260 DU in February, around 220 DU in March and around 240 DU in April. There are ozone values lower than or equal to 220 DU in early (01–05) December, late (25–26) January, some days (05, 12, 17–22) in March and few days in early (06–07) April. The occurrences of these low ozone values in December and January are associated with ozone mini-holes triggered by dynamics. However, the appearances of ozone hole values in March and April are driven by chemistry and is our topic of discussion. The ozone holes measured by OMI corresponding to the dates are also shown in the ozone maps in the top panel and the exact dates of ozone hole occurrences based on OMPS and MERRA-2 data are given in Table S1. The OMPS total column agrees well with that of the OMI measurements throughout the period, where the differences are mostly 2–3 DU and are within the uncertainty of both instruments (i.e. about 5–10%). The OMPS measurements have captured all features of OMI measurements throughout the winter. The GOME measurements are very close to the OMI and OMPS measurements too, but are slightly higher in January and February due to the limited coverage of northern polar region by GOME in winter months. As the winter progresses, its coverage improves and therefore, the March and April measurements are in excellent agreement with other satellite observations. The TCO measurements at Alert also manifest the ozone hole values of about 200 DU in two days of April; corroborating the satellite observations (Figure 7).

We also estimated the partial column ozone loss from the ozone profiles of OMPS and MLS satellites (Figure 7, bottom panel). The ozone loss is calculated with respect to the passive method (Feng et al., 2005). The Arctic winters usually show total column ozone (TCO) loss of about 70–80 DU in cold winters, about 45–50 DU in warm winters, and about 90–120 DU in exceptionally cold winters such as in 2005 and 2011 (Goutail et al., 2015; Kuttippurath et al., 2012b; Rex et al., 2005; Manney et al., 2003). The largest column ozone loss ever measured was in the Arctic winter 2011, and was about 100 DU as assessed from all available studies (Griffin et al., 2018; Kuttippurath et al., 2012; Manney et al., 2011). On the other hand,

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the Antarctic ozone column loss is about twice that of the Arctic, about 150–160 DU, but slightly lower about 100–120 DU in very warm winters (1988 and 2002) and in early years (e.g. 1979–1985) of ozone loss there (Huck et al., 2005; Tilmes et al., 2006; Kuttippurath et al., 2015). The analyses clearly suggest that even the partial column ozone loss in the Arctic winter 2020 is about 115 DU at 350–550 K, which is higher than that of the Arctic winter 2011 and equal to that of the loss found in the Antarctic winters 1979–1985, 2002 and 2019.

Since the ozone loss in the Arctic winter 2020 is up to the levels of that found in Antarctic winters, we examined the occurrence of ozone holes using TCO data from OMPS and MERRA-2 and the results are presented in Figure 8 for selected ozone hole days. The first appearance of ozone holes in the Antarctic winters are also shown for comparison. There are clear and identifiable ozone holes in March and April 2020 and were hundreds of kilometres wide to demarcate the regions below 220 DU. The ozone maps show that the holes in March and April 2020 were larger than that of the Antarctic ozone holes in October 1979 and 1980. Therefore, ozone loss in the Arctic winter 2020 is roughly comparable to the Antarctic ozone loss and the appearance of ozone hole for several weeks demonstrate that the Arctic winters enter a new era of ozone depletion events, and signal significant changes in the climate of the region. However, as the ozone holes were not very large and were not present for continuously for months as they occur over the Antarctic, the situation could termed only as the appearance of signatures of ozone hole in Arctic winter 2020.

4. Conclusions

The Antarctic ozone hole is present for the past forty years, and the impact of ozone hole on public health is mostly restricted to the southern high and mid-latitudes. The ozone hole has also influenced the climate of the southern hemisphere by changing the winds, temperature and precipitations in different regions. On the other hand, the biggest concern about the polar ozone loss in the stratosphere has always been an Arctic ozone hole, because such an ozone hole can occur anywhere beyond 45° N in the densely populated northern mid and high latitudes. The changes in associated UV radiation incidence would also affect the flora and fauna of the region. If such a situation arose, that would trigger ecosystem damage and impose serious threat to public health (e.g. Newman et al., 2009). Nevertheless, it is believed that an ozone hole over the Arctic would be unlikely due to relatively higher temperature and shorter wintertime ozone loss period there. Furthermore, the winters are always prone to several minor and frequent major warmings (almost a major warming per winter), which would restrict the lifetime of the polar vortex, PSC occurrence and chlorine activation to limit the extent and severity of ozone loss. However, the Arctic winter 2020 was exceptional as it was characterised by strong vortex from December through the end of April, large and widespread PSCs, and unprecedented and prolonged chlorine activation with peak ClO values of about 2.0 ppbv. The high chlorine activation in early December and early January produced larger loss in ozone (e.g. 1–1.5 ppmv below 430 K in early January) in the Arctic that has never occurred before. The continued high chlorine activation from January to mid-April caused a record-breaking ozone loss of about 2.5–3.4 ppmv at 400–600 K, and triggered the first-ever ozone hole of Arctic in March and April 2020. The unprecedented chlorine activation (e.g. January through March, above 0.7 ppbv) and severe denitrification (60–80%) also set up an atmosphere to have the first ever occurrence of ozone loss saturation in the Arctic.

Another interesting aspect of this winter was the dynamically driven, but chemically modified ozone mini-holes in December and January. These mini- holes were larger than the Antarctic ozone holes of 1979 and early 1980s.

The analyses presented use multiple data sets, different ozone loss estimation methods, and several parameters to make a robust statistics and balanced assessment on the polar processing ozone loss in the Arctic winter 2020. Yet, all the methods, data, and parameters converge to provide an undeniable fact of the first-ever ozone hole opened over the Arctic with the first appearance of ozone loss saturation. Henceforth, the unique Arctic winter surfaces the first sign of climate change over the region, and sends warning signals across the latitudes about the impact of changes in climate. Yet, the ozone loss in the Arctic cannot not be called as "an ozone hole", as it was in an extreme winter, and the ozone hole (or sustained large loss) and loss saturation situations were not continuous for months as in the Antarctic ozone hole. Extreme weather events are harbingers of climate change and therefore, the ozone loss in the Arctic winter 2020 can also be considered as a signature of climate change in the region.

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Data availability

The MLS data are available on https://disc.gsfc.nasa.gov/. The MODIS datasets were acquired from the Level-1 and Atmosphere Archive & Distribution System (LAADS) Distributed Active Archive Center (DAAC), located in the Goddard Space Flight Center in Greenbelt, Maryland (https://ladsweb.nascom.nasa.gov/). The ozonesonde data are available from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC, https://woudc.org/). The OMPS ozone data are available on https://earthdata.nasa.gov/earth-observation-data/near-real-time/download-nrt-data/omps-nrt. The meteorological analyses: temperature, winds, heat flux, PSC and wave heat flux data are taken from https://ozonewatch.gsfc.nasa.gov/. The OMI data

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are available on https://disc.gsfc.nasa.gov/datasets/. The GOME data are downloaded from the https://atmosphere.copernicus.eu/data.

Author Contributions

JK conceived the idea and wrote the original manuscript. The manuscript was subsequently revised with inputs from RM and WF. JK, PK, SR, RR and GK analysed the data and produced the figures. WF designed the model runs and carried out the model simulations. All authors participated in the discussions and made suggestions, which were considered for the final draft.

Additional information

Supplementary Information accompanies the paper on this journal website.

Competing Interests

The authors declare no competing and conflict of interests

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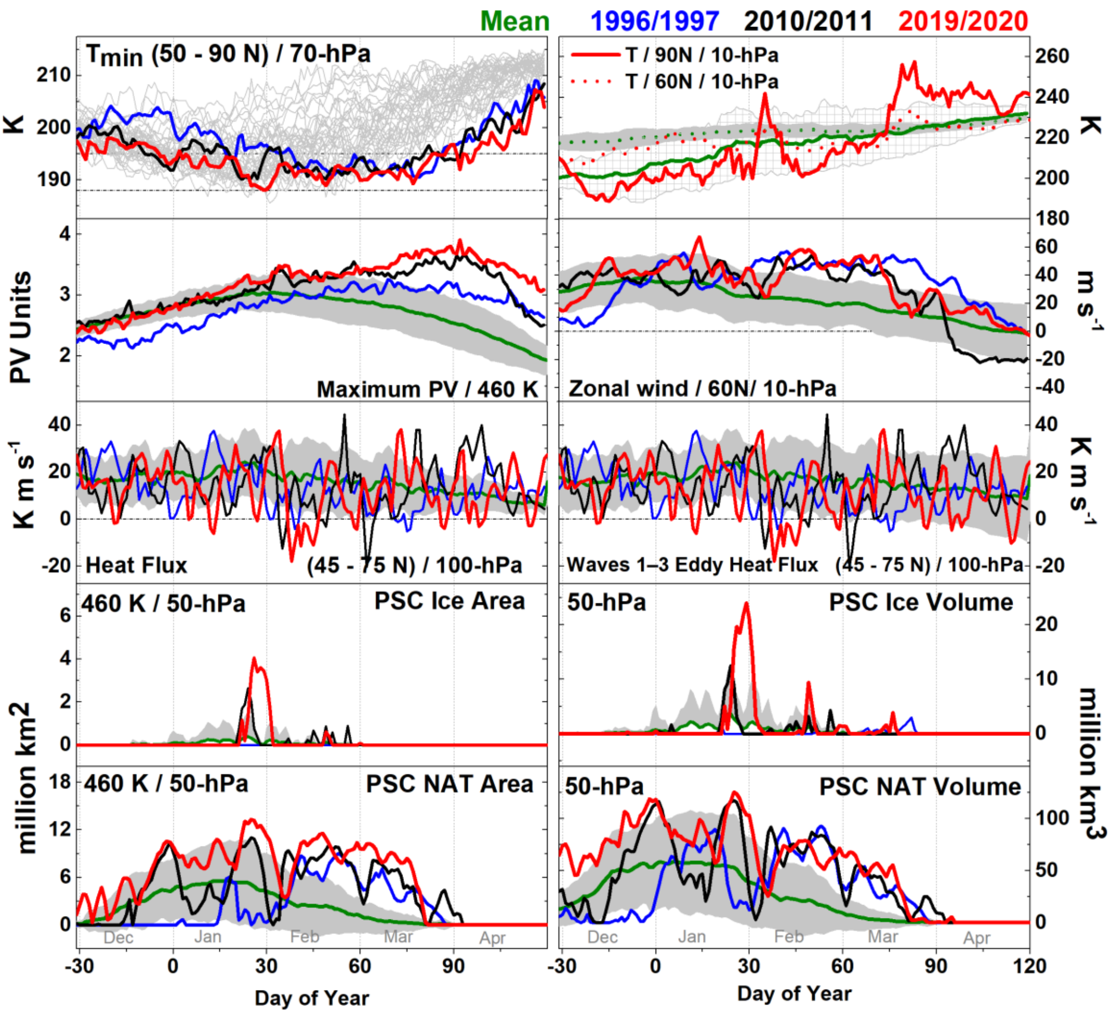

Figure 1: Meteorology of the Arctic winter 2020. The temperature, zonal winds, potential vorticity (PV), heat flux, wave eddy heat flux, and area and volume of polar stratospheric clouds (PSC) for the Arctic winter 2020 as compared to previous Arctic winters. The shaded area shows the standard deviation from the mean.

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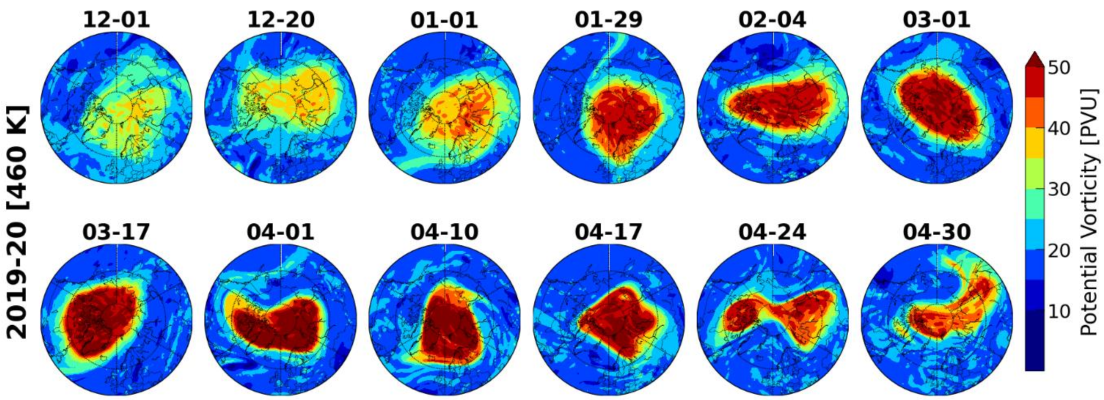

Figure 2: Polar vortex evolution in the Arctic winter 2020. The evolution of polar vortex in the Arctic winter 2020. The vortex situation in the lower stratospheric altitude of about 460 K (~17 km) is illustrated. The vortex edge is calculated with respect to the Nash et al. (1996) criterion at each altitude.

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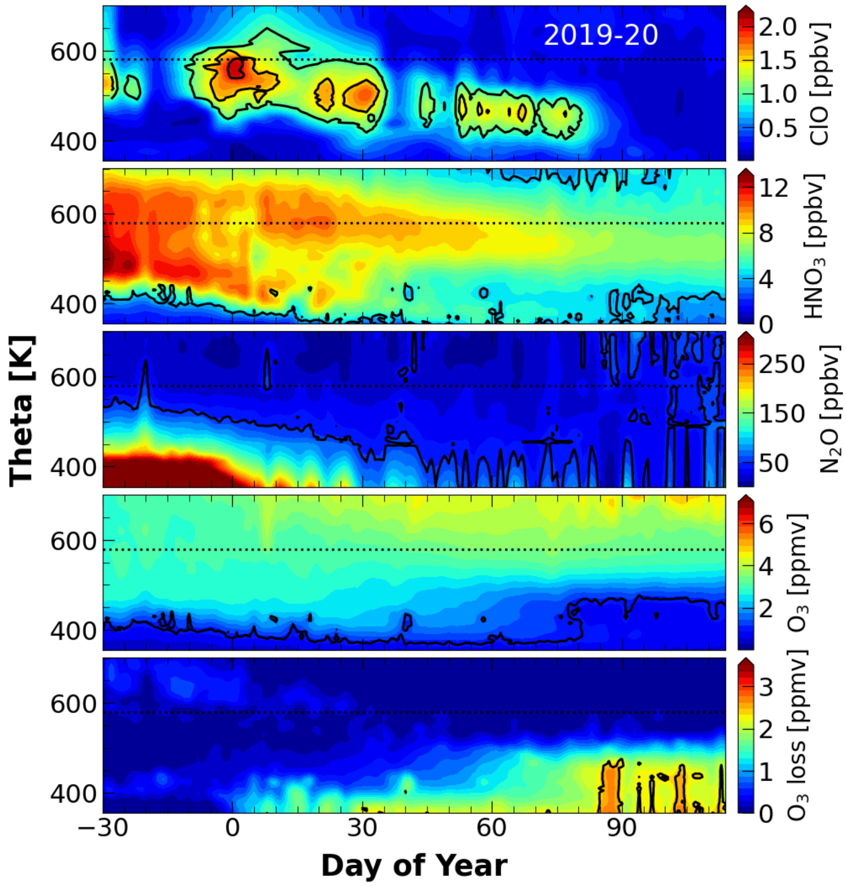

Figure 3: Ozone loss in the Arctic polar vortex in 2020. The distribution of ClO, HNO_3, N_2O and ozone (top to bottom) as measured by the Microwave Limb Sounder (MLS) for the Arctic winter 2020. The bottom panel shows the ozone loss estimated using the MLS ozone by applying the tracer descent method (see Methods and supplementary file). The vortex edge is computed in accordance with Nash et al. (1996) criterion. The vortex-sampled data are then averaged over each day and are shown.

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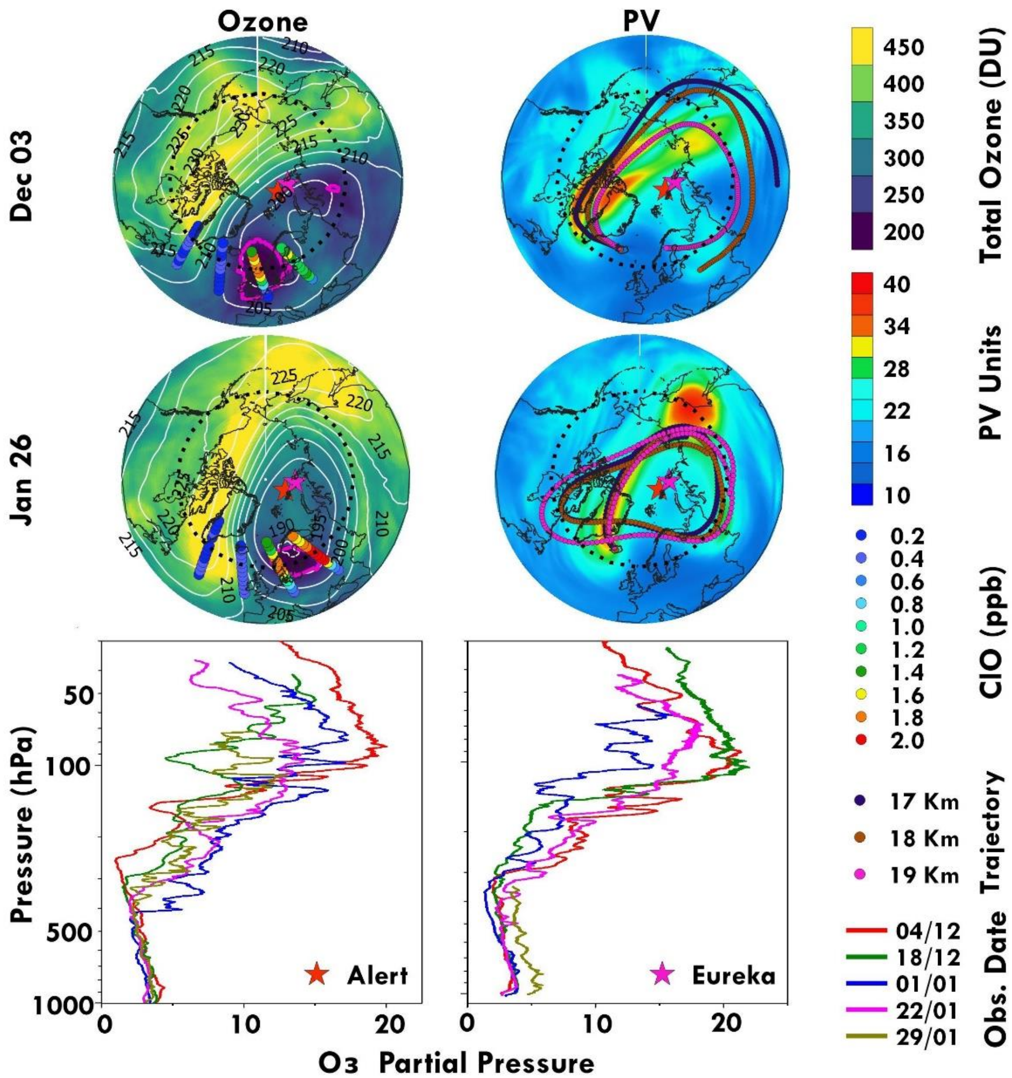

Figure 4: The ozone mini-holes in December 2019 and January 2020. The total ozone observations by Ozone Monitoring Instrument (OMI) on 03 December 2019 and 26 January 2020. The potential vorticity (PV) maps for the corresponding dates are shown on the right. The air mass trajectories computed using the HYSPLIT model at 17, 18 and 19 km are also illustrated in the PV maps. The ozonesonde measurements in December and January at Alert (62.34° N, 82.49° W) and Eureka (79.99° N, 85.90° W) are shown in the bottom panel and are also shown in the maps as red and magenta stars, respectively.

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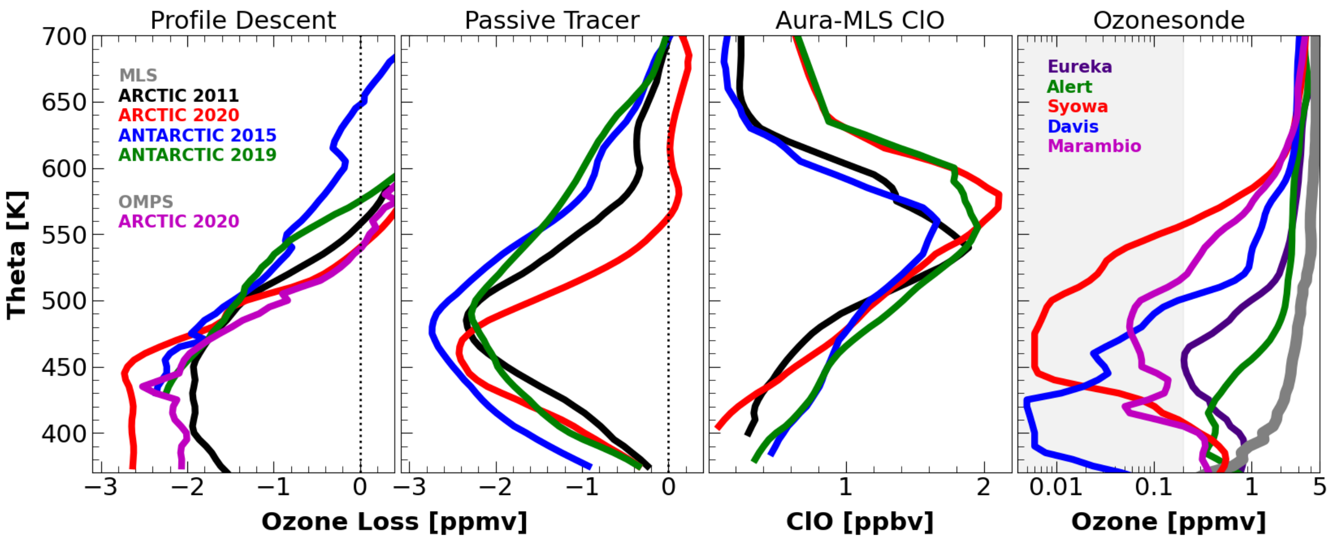

Figure 5: The Arctic and Antarctic Ozone Loss Saturation and Chlorine activation. Left. The ozone loss estimated using the Microwave Limb Sounder (MLS) measurements by applying the vortex descent method for the Arctic winter 2019–2020 compared to the Arctic winter 2011, and the Antarctic winters 2015 and 2019. The ozone loss estimated with Ozone Mapping and Profiler Suite (OMPS) measurements is also shown. **Second from the left**: The ozone loss estimated using the passive tracer method for the Arctic winter 2020, and the Antarctic winters 2015 and 2019. **Second from the right**: The activated profiles ClO measured by MLS for the Arctic winters 2011 and 2020, and the Antarctic winters 2015 and 2019. The profiles of peak ClO values are shown from each winter. **Right:** Ozonesonde measurements from selected Antarctic and Arctic stations. The Antarctic ozonesonde measurements (Davis, Marambio and Syowa) from past winters and the Arctic measurements (Alert and Eureka) from the Arctic winter 2020. The grey color represents an ozone profile without ozone depletion in Arctic and Antarctic. The grey-shaded region represents the ozone loss saturation threshold.

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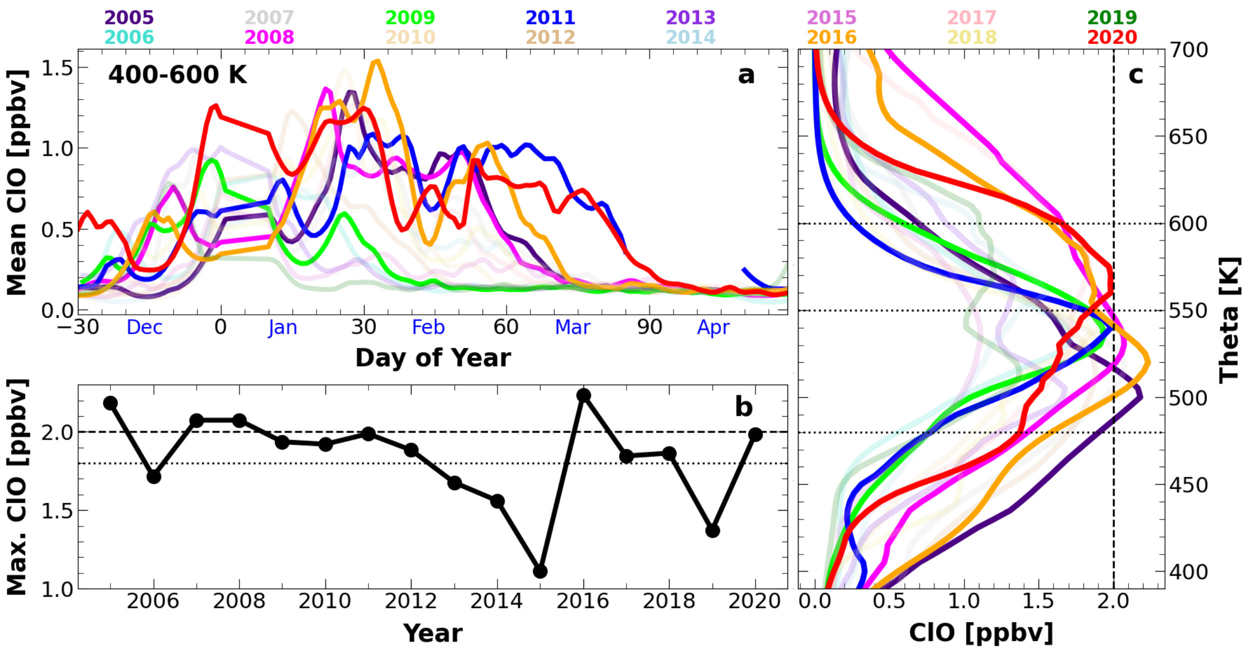

Figure 6: Chlorine activation the Arctic winters 2005–2020. (a) The temporal evolution of ClO in the Arctic winters as measured by the Microwave Limb Sounder (MLS) inside the vortex. **(b)** The maximum ClO measured inside the vortex in each winter from 2005 to 2020. **(c)** The maximum ClO profiles measured inside the vortex for Arctic winters since 2005. The high chlorine activation with high ClO value are shown in bright colours and others are faded in (a) and (c). Since the chlorine activation timing is different in different winters, the peak ClO observed between December and April/March are shown.

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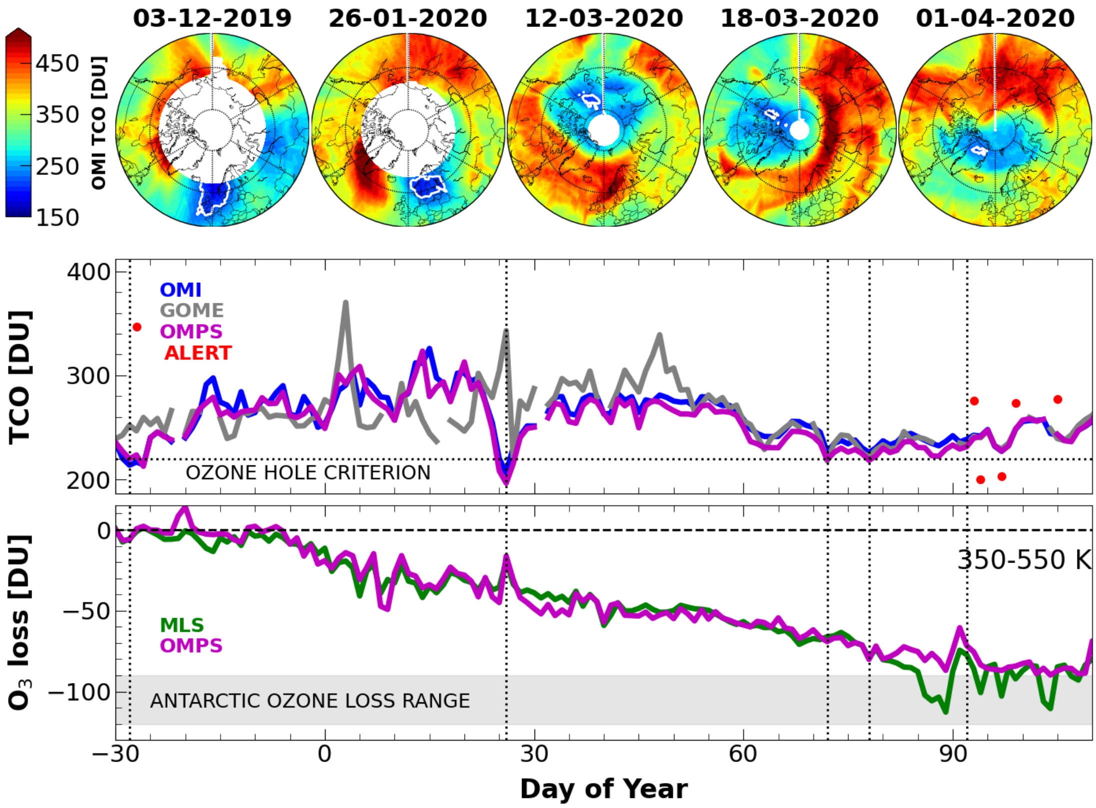

Figure 7: Arctic ozone hole in the total column and partial column ozone. Top: The maps of total column ozone from the OMI satellite measurements in the Arctic for selected ozone hole days for the winter 2020. **Middle panel**: The lowest (5%) TCO measured inside the vortex from three different satellite measurements (OMI, GOME and OMPS). The difference in total column measurements are due to the difference in coverage of the measurements in the Arctic region. The ozone hole criterion of 220 DU is indicated by the dotted line. The total column ozone (TCO) measurements at Alert station are also shown (red solid circles). **Bottom**. The partial column ozone loss computed at the altitude range 350–550 K from the MLS and OMPS measurements. The ozone loss estimated in the Antarctic winters at the same altitude range is shown as the grey-colored area.

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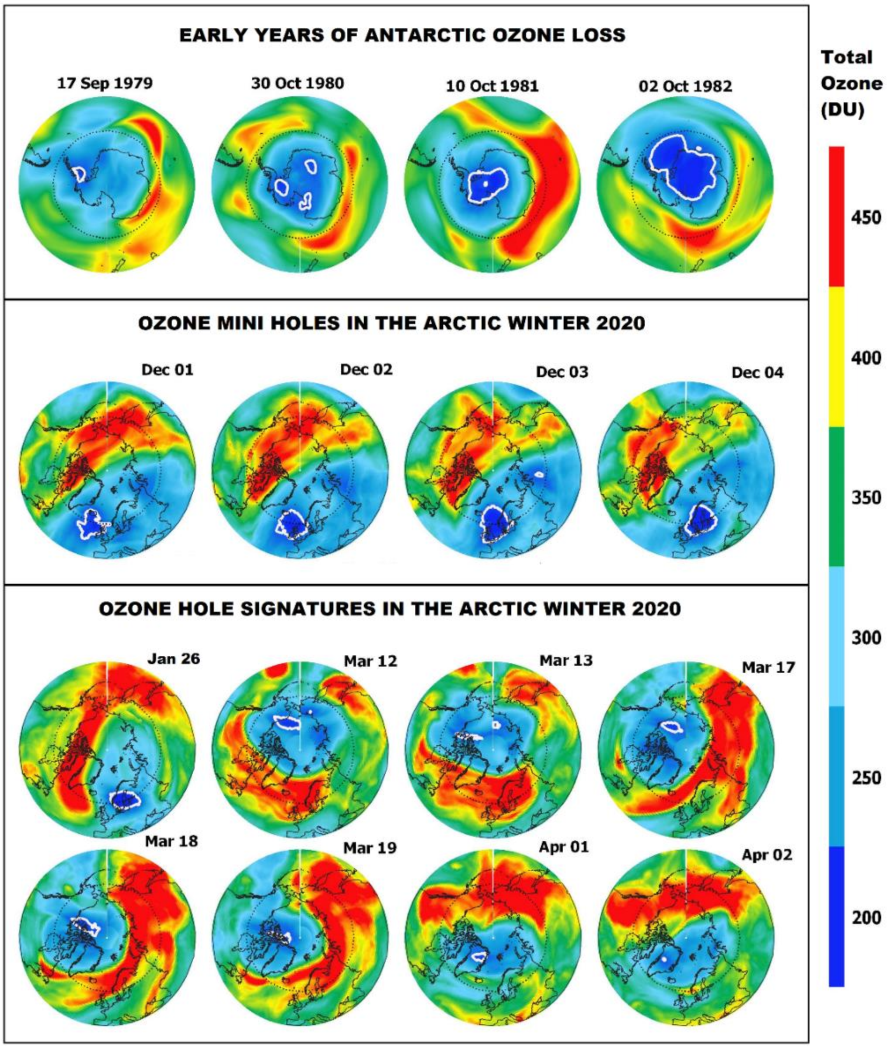

Figure 8: The first-ever ozone hole in the Arctic? The maps of total column ozone from MERRA-2 and OMPS satellite measurements for selected days. Ozone hole is defined as the area below 220 DU of ozone, as demarcated by the white contour. The top panel shows the early years of Antarctic ozone hole, the middle panel shows the ozone-mini holes driven by dynamics, and the bottom panel shows the ozone holes observed in the Arctic winter 2020.