The unusual stratospheric Arctic winter 2019/20: Chemical ozone loss from satellite observations and TOMCAT chemical transport model

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Abstract

Satellite observations of relevant trace gases, together with meteorological data from ERA5, were used to describe the dynamics and chemistry of the spectacular Arctic 2019/20 winter/spring season. Exceptionally low total ozone values of slightly less than 220 DU were observed in mid March within an unusually large stratospheric polar vortex.

This was associated with very low temperatures and extensive polar stratospheric cloud formation, a prerequisite for substantial springtime ozone depletion. Very high OClO and very low NO2 column amounts observed by GOME-2A are indicative of unusually large active chlorine levels and significant denitrification, which likely contributed to large chemical ozone loss. Using results from the TOMCAT chemical transport model (CTM) and ozone observations from S5P/TROPOMI, GOME-2 (total column), SCIAMACHY and OMPS-LP (vertical profiles) chemical ozone loss was evaluated and compared with the previous record Arctic winter 2010/11. The polar-vortex-averaged total column ozone loss in 2019/20 reached 88 DU (23%) and 106⁻DU (28%) based upon observations and model, respectively, by the end of March, which was similar to that derived for 2010/11. The total column ozone loss is in agreement with OMPS-LP-derived partial column loss between 350 K and 550 K to within the uncertainty. The maximum ozone loss ($^{80\%}$) observed by OMPS-LP was near the 450 K potential temperature level (~18 km altitude). Because of the larger polar vortex area in March 2020

compared to March 2011 (about 25% at 450 K), ozone mass loss was larger in Arctic winter 2019/20.

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Special section

¹² The Exceptional Arctic Polar Vortex in 2019/2020: Causes and Consequences

13	Key Points:
14	Very large OClO and very low NO_2 slant columns were observed by GOME-2A
15	during Arctic winter 2019/20
16	Chemical total column ozone loss of 88 DU and 106 DU was derived from TROPOMI
17	satellite observations and the chemical transport model
18	Chemical ozone loss derived from OMPS-LP satellite data reached 2.1 ppmv (80%)
19	near the 450 K potential temperature level $(\sim 18 \text{ km})$

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22 Abstract

Satellite observations of relevant trace gases, together with meteorological data from ERA5, 23 were used to describe the dynamics and chemistry of the spectacular Arctic 2019/20 win-24 ter/spring season. Exceptionally low total ozone values of slightly less than 220 DU were 25 observed in mid March within an unusually large stratospheric polar vortex. This was 26 associated with very low temperatures and extensive polar stratospheric cloud forma-27 tion, a prerequisite for substantial springtime ozone depletion. Very high OCIO and very 28 low NO₂ column amounts observed by GOME-2A are indicative of unusually large ac-29 30 tive chlorine levels and significant denitrification, which likely contributed to large chemical ozone loss. Using results from the TOMCAT chemical transport model (CTM) and 31 ozone observations from S5P/TROPOMI, GOME-2 (total column), SCIAMACHY and 32 OMPS-LP (vertical profiles) chemical ozone loss was evaluated and compared with the 33 previous record Arctic winter 2010/11. The polar-vortex-averaged total column ozone 34 loss in 2019/20 reached 88 DU (23%) and 106 DU (28%) based upon observations and 35 model, respectively, by the end of March, which was similar to that derived for 2010/11. 36 The total column ozone loss is in agreement with OMPS-LP-derived partial column loss 37 between 350 K and 550 K to within the uncertainty. The maximum ozone loss ($\sim 80\%$) 38 observed by OMPS-LP was near the 450 K potential temperature level (~ 18 km alti-39 tude). Because of the larger polar vortex area in March 2020 compared to March 2011 40 (about 25% at 450 K), ozone mass loss was larger in Arctic winter 2019/20. 41

42 **1** Introduction

While large springtime polar ozone depletion has been observed above Antarctica 43 in most years since the 1980s (the "ozone hole"), such events occur only sporadically in 44 the Arctic (Langematz et al., 2018). The chemistry involved in this depletion process 45 is well understood (Solomon, 1999; Solomon et al., 2015). A prerequisite for substantial 46 polar ozone depletion during winter/spring is sufficiently low stratospheric temperatures 47 to form polar stratospheric clouds (PSCs) (e.g. Spang et al., 2018), which activate halo-48 gens, mainly chlorine, from their reservoir species. Sunlight returning to the polar re-49 gion then allows rapid catalytic reactions involving the active halogens to destroy ozone. 50

Above Antarctica temperatures in the lower stratosphere are persistently below the 51 PSC formation threshold. In contrast, above the Arctic such low temperatures are reached 52 only sporadically and rarely persist over a long enough period to sustain the ozone de-53 pletion process. The strong variability in stratospheric meteorology, associated with vari-54 ations in atmospheric dynamics, is responsible for the high variability in Arctic ozone 55 and stratospheric temperatures due to enhanced ozone transport in warm winters and 56 enhanced chemical loss in cold Arctic winters (e.g. Chipperfield & Jones, 1999; Tegtmeier 57 et al., 2008; Weber et al., 2011; Strahan et al., 2016). The very low ozone observed in 58 cold polar winters is therefore due to a combination of reduced transport and chemical 59 loss (e.g. Weber et al., 2003; Tegtmeier et al., 2008). 60

The Arctic winter/spring 2019/20, along with 2010/11 and 1996/97 exhibited low 61 stratospheric temperatures throughout February and well into March, associated with 62 a deep depression in polar ozone in March resembling the Antarctic ozone hole (Lefèvre 63 et al., 1998; Kuttippurath et al., 2012; Manney et al., 2011, 2020; Dameris et al., 2020; 64 Lawrence et al., 2020) as shown in Fig. 1. In March 2020 total ozone was up to 200 DU 65 lower than the year before. Arctic winter 2019/20, in particular, has some similarity to 66 the winter 2010/11 (Manney et al., 2011) which, until now, showed the largest estimated 67 ozone depletion. 68

In this paper we report on chemical ozone loss in Arctic winter 2019/20 derived from total ozone data from TROPOMI (TROPOspheric Monitoring Instrument) and ozone profiles from OMPS-LP (Ozone Mapping and Profiler Suite - Limb Profiler) satellite data



Figure 1. Arctic March mean total ozone (DU) from TROPOMI in (a) 2019 and (b) 2020, representative for years with average conditions and above-average-sized polar vortices, respectively. (c) Difference (DU) between mean March 2020 and 2019 total ozone. The total ozone was retrieved using WFDOAS (weighting function DOAS) V4.

in combination with results from the 3D chemical transport model (CTM) TOMCAT
(Chipperfield, 2006). A particular focus in this paper is on the comparison between 2019/20
and the previous record winter 2010/11 for which ozone column data from GOME-2A
(Global Ozone and Monitoring Experiment- Metop A) and limb data from SCIAMACHY
(SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY) are also
used.

The structure of this paper is as follows. Section 2 describes the observational data and CTM used here. Section 3 gives a brief description of the polar meteorology in the Arctic winter/spring 2019/2020, including a comparison to the record winter/spring in 2010/11. Section 4 shows results from other trace gas observations (NO₂ and OClO) along with ozone followed, in Section 5, by chemical ozone loss calculations using the combination of model and observational data. Our summary and concluding remarks are provided in Section 6.

85 2 Data

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2.1 Merged WFDOAS total ozone

The merged GOME, SCIAMACHY, GOME-2, and TROPOMI (GSG) total ozone 87 timeseries consists of total ozone data retrieved using an advanced version of the Uni-88 versity of Bremen Weighting Function DOAS (WFDOAS) algorithm (Coldewey-Egbers 89 et al., 2005; Orfanoz-Cheuquelaf et al., 2020). The merging of the various instruments 90 has been briefly described in Weber et al. (2018). A monthly mean latitude-dependent 91 bias correction, used to successively adjust SCIAMACHY (2002-2012) and GOME-2A 92 (2007-present) to the initial GOME (1995-2011) data record, has been applied here to 93 daily gridded data. Recently bias-adjusted WFDOAS data from GOME-2B (with a better global coverage than GOME-2A) starting in 2015 and TROPOMI (Veefkind et al., 95 2012) starting in 2018 have been added into the merged daily WFDOAS total ozone time-96 series, available at a spatial resolution of $1.25^{\circ} \times 1^{\circ}$ (longitude × latitude). 97

98 2.2 SCIAMACHY and OMPS-LP ozone profiles

SCIAMACHY aboard Envisat (2002-2012) and OMPS-LP aboard SUOMI-NPP 99 (2012-present) observe the atmosphere in limb geometry from a sun-synchronous orbit 100 and collect radiances in the UV-VIS spectral region. Ozone concentrations are retrieved 101 from 60 down to 10 km (or cloud top height), by using for both instruments the SCI-102 ATRAN radiative transfer model and retrieval software package (Rozanov et al., 2014). 103 The typical vertical resolution of the retrieved profiles is about 2.5 km (OMPS-LP) and 104 3.7 km (SCIAMACHY). Details of the retrieval algorithm and a validation of the ozone 105 profiles can be found in Jia et al. (2015) and Arosio et al. (2018). 106

2.3 GOME-2A OClO and NO₂ columns

Stratospheric OClO and NO₂ columns retrieved from UV/visible observations of 108 instrument such as GOME and SCIAMACHY have already been used in previous stud-109 ies (Wagner et al., 2001; Weber et al., 2003; Richter et al., 2005). Here, the data anal-110 ysis follows Richter et al. (2005) but using GOME-2A data instead of GOME observa-111 tions. Since OClO is rapidly photolyzed, substantial amounts can only be measured at 112 very large solar zenith angles (SZA). In order to remove the effect of changing illumi-113 nation during the time series, only observations at 90° SZA are used. As rapid photol-114 ysis also changes the OClO concentration along the light path, no attempt is made to 115 convert OClO slant columns into vertical columns. As the geometry of the light path re-116 mains the same for all measurements at 90° SZA, the results are still comparable from 117 day to day and between years. 118

The variations in local equator crossing times (and twilight zones at a given day of the year) of the various satellites complicate the comparison between satellites. For this reason, we limit our comparisons to results from the GOME-2A instrument (launched in 2006) that covers both cold Arctic winters studied here. As a result of the sun-synchronous orbit of GOME-2A, the latitude probed at 90° SZA varies from 65° to 85° over the winter / spring period, and does not reflect vortex or polar-cap averages as the ozone data used here.

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2.4 TOMCAT chemical transport model

TOMCAT/SLIMCAT (hereafter TOMCAT) is a 3-D chemical transport model (CTM), 127 which has been described in Dhomse et al. (2019). The model contains a detailed de-128 scription of stratospheric chemistry, including heterogeneous reactions on sulfate aerosols 129 and PSCs. Here the model was forced using European Centre for Medium-Range Weather 130 Forecasts (ECMWF) ERA5 winds and temperatures (Hersbach et al., 2020) and run with 131 a horizontal resolution of $2.8^{\circ} \times 2.8^{\circ}$ with 32 altitude levels from the surface to ~60 km. 132 The run was initialised in 1977 and forced using specified surface mixing ratios of the 133 long-lived source gases. In recent model updates the supersaturation of HNO_3 for type 134 I PSC formation was implemented according to Grooß et al. (2018) and the Cl_2O_2 ab-135 sorption cross sections are from Burkholder et al. (2015) with an assumed quantum yield 136 of 1. Solar flux variations [1980–2019] are taken from the NRLSSI2 empirical model (Coddington 137 et al., 2016) that are recommended for CMIP6 simulations as implemented in Dhomse 138 et al. (2016). For the year 2020, solar fluxes are held constant at December 2019 values. 139

2.5 ERA5 Reanalysis

¹⁴¹ Wind and temperature data from the ERA5 re-analysis (Hersbach et al., 2020) are ¹⁴² used here for determining dynamical properties (vortex and PSC volume) and driving ¹⁴³ the TOMCAT CTM. For the polar vortex diagnostics 6-hourly data at a spatial reso-¹⁴⁴ lution of $0.75^{\circ} \times 0.75^{\circ}$ were used.



Figure 2. (a) Type I and type II PSC volume ($\times 10^6$ km³) in Arctic winters 2010/11 and 2019/20 derived from ERA5 reanalysis in the potential temperature range 400 to 750 K. The grey shading indicates the maximum values of type I PSC volume from 1979 to spring 2019. All curves have been smoothed with a running [1,2,1] triangular filter. (b) Monthly mean ERA5 100 hPa eddy heat flux integrated between 45°N and 75°N.

145 **3** Meteorology

The Arctic winter/spring 2019/20 exhibited a strong polar vortex with persistent PSCs observed from mid November until April, as shown in Fig. 2. Here the PSC volume was calculated using ERA5 data at potential temperature levels in order to identify grid boxes with temperatures below the PSC formation threshold as described in Feng et al. (2007). PSC volumes were derived in steps of 25 K potential temperature from 400 K to 750 K altitudes. For the vertical extent it was assumed that 25 K roughly corresponds to 1 km altitude (Knox, 1998).

In Arctic winter 2019/20 the PSC volume was at a record high since 1979 in the 153 second half of November. It remained high throughout December and January and was 154 again at a record high in the second half of February through to nearly the end of March. 155 The early PSC formation (chlorine activation) and very high PSC volumes in March, as 156 shown in Fig. 2a, favoured strong depletion in ozone. The previous record winter 2010/11157 showed a similar evolution in the volume of type I and type II PSCs as this year, with 158 the exception that the volumes were generally smaller in 2011 and PSCs started to form 159 later (end of November). Temporary lows in PSC volume had a very similar timing in 160 both winters with local minima observed in the middle of January and a sharp short-161 term drop in early February, most likely related to minor stratospheric warming events 162 perturbing the polar vortex. Maximum PSC volumes were reached at the end of Jan-163 uary in both winters. At that time type II (ice) PSCs were also maximum in both win-164 ters. Ice PSCs can lead to strong dehydration and removal of water vapour that may re-165 sult in a delay in deactivating active chlorine into HCl in early spring (Manney et al., 166 2020). 167

Fig. 2b shows northern hemisphere (NH) monthly mean eddy heat fluxes at 100 hPa 168 for the two cold Arctic winters considered here, along with data from their preceding win-169 ters 2009/10 and 2018/19, respectively. The eddy heat flux is a measure of the plane-170 tary wave activity determining the strength of dynamical activity (ozone transport, driver 171 of the Brewer-Dobson circulation) and stratospheric meteorology. From January to March 172 the eddy heat flux was persistently below the long-term mean in both 2010/11 and 2019/20, 173 resulting in reduced ozone transport as well as lower stratospheric temperatures (Newman 174 et al., 2001; Randel et al., 2002; Weber et al., 2011). In February 2020 the 100 hPa eddy 175

heat flux reached the lowest value since 1980, which may have been responsible for set ting record high PSC volumes starting by the end of February 2020. Both the contin-

uous low dynamical activity, also linked to the positive anomaly in the Arctic Oscilla-

tion (Lawrence et al., 2020), and very low stratospheric temperatures contributed to the

very low ozone in Arctic winter 2019/20.

Further details on the stratospheric meteorology in this particular winter and comparisons to past winters can be found in this journal's special issue and other studies (Dameris et al., 2020; Inness et al., 2020; Lawrence et al., 2020; Manney et al., 2020).

¹⁸⁴ 4 Trace gas observations

The evolution of ozone, NO_2 , and OClO, above the Arctic in 2019/20 are displayed 185 in Fig. 3. Corresponding timeseries for the year 2010/11 (previous record winter) as well 186 as the years preceding both cold winters, 2009/10 and 2018/19, with more typical con-187 ditions are also shown to demonstrate the large variability from year-to-year. Panel (a) 188 shows the evolution of the polar cap mean total ozone $(50^{\circ}N-90^{\circ}N)$. Due to the Brewer-189 Dobson circulation, total ozone normally increases over the winter reaching, on average, 190 an annual maximum in March (thick grey curve). Starting in mid February, however, 191 polar ozone strongly declined in 2011 and 2020. In mid March polar-cap mean were the 192 lowest in both winters since the mid 1990s (since start of the WFDOAS merged total 193 ozone timeseries), a time when stratospheric halogens originating from man-made ozone 194 depleting substances (ODSs) were maximum (Newman et al., 2007). In 2020 the polar 195 cap mean remained very low from March until May. 196

The polar minimum total ozone evolved in a very similar way to the mean (Fig. 197 3b). A first record minimum was observed in March 2011 and this record was broken again 198 in March 2020 with total ozone being slightly below 220 DU for a brief period (Inness 199 et al., 2020; Wohltmann et al., 2020), a value which commonly defines the boundary of 200 the Antarctic ozone hole (e.g. NASA Ozone Watch, 2020). The steady decline in min-201 imum ozone in polar winter is considered a good proxy for continued polar chemical ozone 202 loss (Müller et al., 2008). The rapid declines and rises in minimum ozone observed in 203 early November and late January 2020 are, in contrast, purely dynamical in nature. They 204 are caused by subtropical streamers intruding into polar latitudes producing so-called 205 ozone mini holes by fast horizontal advection of ozone away from a region with a strongly 206 elevated tropopause (James et al., 2000; Weber et al., 2002; Dameris et al., 2020). 207

In both Arctic winters 2010/11 and 2019/20 the mean NO₂ and OClO slant columns were record minima and maxima, respectively, by late February and early March (Fig. 3c,d). In particular, OClO levels reached a new record (since 2007) in March 2020, which indicates substantial chlorine activation up to the last measurements mid of March. After that, the sun is too high for GOME-2A measurement at 90° SZA and the time series ends before the end of chlorine activation is reached.

Stratospheric NO_2 levels are generally small in the winter polar regions as most NO_2 214 is converted into its night-time reservoirs $(N_2O_5 \text{ and } HNO_3)$ during polar night (e.g. Bur-215 rows et al., 1999). As the sun returns to the Arctic, NO_2 levels usually increase through 216 mixing of polar air masses with mid-latitude air and destruction of HNO₃ by photoly-217 sis and reaction with OH. A stable vortex and denitrification by subsidence of condensed 218 HNO_3 in PSCs delay this process as well as subsequent deactivation of active chlorine 219 into their reservoir species (ClONO₂), explaining both the low NO₂ columns and the ex-220 tended chlorine activation observed in 2011 and 2020. 221



Figure 3. Time series of Arctic polar cap $(50^{\circ}N-90^{\circ}N)$ (a) mean total ozone (DU), (b) minimum total ozone (DU), (c) mean NO₂ and (d) mean OCIO. Panels (c) and (d) show mean 90° solar zenith angle (SZA) column densities (total columns) of NO₂ and 90° SZA slant column densities (SCD) of OCIO, respectively. Selected years are shown by coloured lines, the two cold Arctic winters 2010/11 and 2019/20 and the two years (2009/10 and 2018/19) preceding both cold winters. Daily minimum total ozone columns shown here are averages of the ten grid boxes $(1^{\circ} \times 1.25^{\circ})$ with the lowest total ozone. The grey shading provides the range of maximum and minimum values since 1995 (ozone) and 2007 (minor trace gases), respectively. The thick grey lines in the ozone panels show the long-term mean since 1995. All curves have been smoothed with a running [1,2,1] triangular filter.



Figure 4. Evolution of Arctic vortex-mean (a) total ozone (DU) and (b) chemical ozone loss (DU) in winter 2019/20. In panel (a) timeseries of TROPOMI (green) and TOMCAT (black) are shown. Dotted black line is TOMCAT data limited to the sunlit part of the polar vortex, while the red line displays the passive ozone from TOMCAT. The vortex area was determined here for the 450 K surface. In panel (b) the light blue line represents the ozone loss derived from TROPOMI using TOMCAT passive ozone (see main text for explanations). Dark blue line shows the TOMCAT-derived column ozone loss. For comparisons triangles show the total column ozone losses from the observations and the model on March 15 and 30 in 2011, respectively. Also shown in panel (b) are the partial column ozone losses (350 K-550 K altitude) from OMPS-LP and TOMCAT.

5 Chemical ozone loss

The chemical ozone loss in both cold Arctic winters 2010/11 and 2019/20 is esti-223 mated using the vortex-average approach (Harris et al., 2002). In this approach total ozone and ozone profiles are averaged daily within the confines of the polar vortex and the dif-225 ference to a passive ozone tracer, here from the TOMCAT CTM, is considered as the 226 accumulated ozone loss. The vortex edge is here defined by the combination of maximum 227 wind speed and potential vorticity (PV) gradient (Nash et al., 1996) and was determined 228 at the 450 K potential temperature level. The timeseries of vortex-averaged total ozone 229 from TROPOMI and TOMCAT are shown in Fig. 4 for the winter 2019/20. The same 230 quantities for the winter 2010/11 are displayed in Fig. 5 using observations from GOME-231 2A and SCIAMACHY. While panel (a) shows the various timeseries over the course of 232 the winter, corresponding polar ozone losses are displayed in panel (b). The TOMCAT 233 vortex-averaged ozone has evidently a negative bias with respect to TROPOMI, which 234 is particularly large in the middle of the winter, which may be partly due to the strong 235 descent in the model forced by the ERA5 reanalysis. 236

TROPOMI, OMPS-LP (and SCIAMACHY) measure in the optical range and thus 237 do not observe the polar night region, such that the polar vortex is usually only covered 238 by 10 to 20%, depending on its exact location, in late December and early January. The 239 agreement between observations and model improves significantly when using model av-240 erages from the sunlit part of the polar vortex only (dotted line in Fig. 4a). In the be-241 ginning of December the observations agree well with the model, but the bias increases 242 with time until the end of March (when the polar vortex is completely illuminated) in-243 dicating that modelled ozone loss is slightly larger than that observed (by ~ 15 DU). The 244 observed ozone loss is here approximated by subtracting the difference between obser-245 vations (green curve) and the sunlit part of the model ozone (dotted line) from the mod-246 elled ozone loss. Alternatively, one could also have taken the difference between the sun-247



Figure 5. Same as Fig. 4 but for winter 2010/11. Triangles show for comparison the total column ozone loss on March 15 and March 30 in 2020. Here observational data from GOME-2A (total column) and SCIAMACHY (subcolumns) are shown.

lit part of modelled passive ozone and observations. One important assumption for both
alternatives of the observed ozone loss calculation here is that the negligible difference
between passive ozone and satellite observations in the sunlit part of the polar vortex
ozone in early December also holds for the entire polar vortex. The estimated observed
chemical loss curve is thus not necessarily representative for the entire polar vortex in
early winter but approaches the full vortex value by late March.

By the end of March 2020 the TROPOMI accumulated total ozone loss amounts to 88 DU (23%) and for the TOMCAT CTM to 106 DU (28%). These losses are quite similar to the results from the previous record winter 2010/11 as indicated by the triangles in Fig. 4b (see also Fig. 5). On March 15, where observed minimum total ozone is near its lowest value (Fig. 2b) the mean total ozone loss is even slightly higher in 2011 than 2020, for both observations and CTM.

Fig. 6 shows the time series of March daily mean vortex-averaged ozone profiles 260 from SCIAMACHY (2011) and OMPS-LP (2020). Only profiles with a PV value higher 261 than 38 PVU (at 475 K) were averaged to obtain the daily Arctic vortex mean. The rapid 262 decline of ozone near 450 K potential temperature levels throughout March is clearly ev-263 ident. It appears that the largest decline in 2020 was slightly below 450 K, a bit lower 264 than in 2011 apparently in agreement with Microwave Limb Sounder (MLS) observations 265 reported in Manney et al. (2020). However, one needs to be cautious here as the verti-266 cal sampling of SCIAMACHY (3.3 km \approx 75 K) is too coarse to clearly support this. 267

Figure 7 shows a time-altitude cross-section of the accumulated ozone loss from SCIA-268 MACHY (2010/11) and OMPS-LP (2019/20). Similar to total ozone, the ozone loss here 269 is calculated from the difference of the daily mean observed ozone profiles to the pas-270 sive ozone from TOMCAT. For this purpose, only TOMCAT profiles collocated with SCIA-271 MACHY / OMPS-LP observations are considered. Passive ozone is initiated in the model 272 on December 1 each Arctic winter. A comparison between observations and passive ozone 273 in early December, a period where chemical ozone loss is still very small, revealed dif-274 ferences of about 12 DU (2010) and 21 DU (2019) in the 350-550 K column which are 275 accounted for in the ozone loss shown in Fig. 7. By the end of March 2020 a maximum 276 ozone loss of 2.1 ppmv near 450 K was observed. In 2011 the accumulated loss reached 277 a maximum of 2.2 ppmv which is comparable to the value from 2020. These values are 278 slightly smaller than, but in good agreement with, Manney et al. (2011, 2020), confirm-279 ing that the ozone losses observed in both winters were at a record low. 280



Figure 6. March sequence of daily mean Arctic vortex ozone profiles as a function of potential temperature from (a) SCIAMACHY in 2011 and (b) OMPS-LP in 2020. Colors indicate individual dates as provided in the legend. The Arctic vortex region was here determined from the 475 K altitude level.

Since most of the ozone decrease occurs between 350 and 550 K, subcolumn ozone 281 loss values were derived from the profile observations and CTM. The subcolumn (350-282 550 K) ozone loss is displayed along with the total ozone data in Figs. 4b and 5b. The 283 OMPS-LP subcolumn ozone loss is 105 DU, about 20 DU larger than the TROPOMI 284 total column ozone loss at the end of March 2020. The modelled subcolumn loss on the 285 other hand (95 DU) is closer to the observation-derived total column loss of 88 DU. In 286 2011 the observation-derived and modelled subcolumn ozone loss at the end of March 287 2011 differ slightly by 5 DU. The largest error in the estimated ozone loss comes from 288 uncertainties in establishing a proper initial Arctic vortex-mean ozone value from the 289 UV/visible observations as well as uncertainties in the CTM, e.g. vertical transport in 290 the polar vortex and uncertainties in photochemical data propagating into uncertain-291 ties in the model chemistry. The overall uncertainty in the established polar ozone loss 292 is estimated to be about 15% (about 15 DU). Nevertheless, both CTM and observations 293 agree that Arctic vortex-averaged ozone losses in both Arctic winters 2011 and 2020 were 294 very similar. 295

The product of mean column ozone loss and average vortex area provides an estimate of the total number of ozone molecules lost (which is proportional to the ozone mass loss). At 450 K the vortex area was, on average, about 20 million square km, about 4 million square km larger than in 2011 (see also Fig. 10a in Lawrence et al. (2020)). As a consequence, the mass loss in ozone was about 25% larger in 2020 compared to 2011 assuming a similar vortex-averaged column ozone loss in both winters. In terms of ozone mass loss the Arctic winter 2019/20 therefore sets a new record high.

³⁰³ 6 Summary and conclusions

The Arctic winter/spring 2019/20 is one of the coldest on record (Lawrence et al., 2020; Dameris et al., 2020; Inness et al., 2020) with temperatures sufficient for PSC occurrence from November until April. The PSC volume was at a record high (based on available observations) in November and from late February throughout March, which indicates substantial polar ozone loss. OCIO slant columns were very high and NO₂ slant



Figure 7. Time-altitude cross-section of estimated ozone loss in late Arctic winter and early spring (a) from SCIAMACHY in 2011 and (b) from OMPS-LP in 2020. The daily ozone loss was determined from the difference between Arctic vortex-mean passive ozone profile from TOMCAT and observed ozone profiles. Biases in passive ozone with respect to observations in early December have been corrected for in the loss calculations (see main text). Vortex area is derived here at the 475 K altitude level.



Figure 8. ERA5 100 hPa polar-cap (blue) and tropical temperature (red) timeseries from 1980 to 2020. Blue dots indicate mean polar temperature anomalies outside the 1σ standard deviation of the long-term mean indicated by the two blue dashed lines.

columns very low in February and March, indicating large chlorine activation and extensive denitrification, both consistent with continued chemical ozone losses well into early
spring. In February 2020 the planetary wave activity was at a record low resulting in very
low temperatures in February and March and record high PSC volume.

Using total ozone observations from TROPOMI and TOMCAT/SLIMCAT model 313 simulations, the total column ozone loss was estimated to be 88 DU and 106 DU, respec-314 tively, by the end of March (23-28% loss). From OMPS-LP observations ozone profile 315 losses were found to reach 2.1 ppmv at 450 K (\sim 18 km) in good agreement with MLS 316 observations (Manney et al., 2020). The combined uncertainty of derived polar ozone 317 loss (from model and observations) is estimated to be on the order of 15%. The vortex-318 averaged ozone loss was very similar in both Arctic winters 2010/11 and 2019/20, but 319 the ozone mass loss was significantly higher (about 25%) in 2020 than 2011 due to the 320 larger area of the Arctic vortex in March 2020. 321

An important question concerns the reason behind such a large ozone loss observed 322 in winter 2019/20, given the fact that ODSs are mostly declining as a consequence of the 323 Montreal Protocol and Amendments. While in the Antarctic first signs of ozone recov-324 ery have been detected (Solomon et al., 2016; de Laat et al., 2017; Weber et al., 2018), 325 the variability in stratospheric meteorology and ozone in the Arctic is still too large to 326 uniquely identify ozone recovery (e.g. Chipperfield et al., 2017; Dhomse et al., 2018). CTM 327 calculations, however, show that the ozone loss would have been larger in 2019/20 with 328 ODS at levels of the mid 1990s (Feng et al., 2020). 329

The Arctic winter 2019/20 exhibited record-low polar-cap temperature at 100 hPa 330 based upon ERA5 reanalysis data as shown in Fig. 8. Although most Arctic winters were 331 rather warm (above 1σ variability) after the mid 1990s, about two winters in a decade 332 were extremely cold. The hypothesis discussed for some time that cold Arctic winters 333 are getting colder in a changing climate (Rex et al., 2004, 2006; Rieder & Polvani, 2013) 334 gains new relevance after the record temperatures observed in 2019/20. Also shown in 335 Fig. 8 are tropical 100 hPa temperatures during boreal winter, displaying an anti-correlation 336 with polar-cap temperatures (Yulaeva et al., 1994). This correlation is a result of the inter-337 annual variability in the Brewer-Dobson circulation (weak circulation associated with 338

higher tropical upper troposphere - lower stratosphere (UTLS) temperatures due to weaker
 vertical ascent in the tropics).

The two unusual cold Arctic winters of the past decade fall in a period where a weak positive tropical temperature trend is apparent. This suggests that a slight weakening of the Brewer-Dobson circulation, opposite to the expected long-term trend from climate change (e.g. Aschmann et al., 2014; Garfinkel et al., 2017), may have contributed to these two recent extreme Arctic winters.

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