

Simulation of the record Arctic stratospheric ozone depletion in 2020

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Abstract

In Arctic winter and spring 2020, the stratospheric temperatures were exceptionally low for a long time period and the polar vortex was very stable. As a consequence, significant ozone depletion occurred in Northern polar regions in spring 2020. Here, we present simulations by the Chemical Lagrangian Model of the Stratosphere (CLaMS) that addresses the development of chlorine compounds and ozone in the polar stratosphere in 2020. The simulation is able to reproduce relevant observations which is shown by comparisons with MLS, ACE-FTS and OMI data. Although the concentration of chlorine and bromine compounds in the polar stratosphere has decreased by more than 10% compared to the peak values around the year 2000, the meteorological conditions in winter and spring 2020 caused an unprecedented ozone depletion. The simulated lowest ozone mixing ratio was around 0.05 ppmv and the ozone depletion in the vortex core in the lower stratosphere reached 133 Dobson Units, which is more than the loss in the years 2011 and 2016 that had the largest Arctic ozone depletion so far.

Abstract

In Arctic winter and spring 2020, the stratospheric temperatures were exceptionally low for a long time period and the polar vortex was very stable. As a consequence, significant ozone depletion occurred in Northern polar regions in spring 2020. Here, we present simulations by the Chemical Lagrangian Model of the Stratosphere (CLaMS) that addresses the development of chlorine compounds and ozone in the polar stratosphere in 2020. The simulation is able to reproduce relevant observations which is shown by comparisons with MLS, ACE-FTS and OMI data. Although the concentration of chlorine and bromine compounds in the polar stratosphere has decreased by more than 10% compared to the peak values around the year 2000, the meteorological conditions in winter and spring 2020 caused an unprecedented ozone depletion. The simulated lowest ozone mixing ratio was around 0.05 ppmv and the ozone depletion in the vortex core in the lower stratosphere reached 133 Dobson Units, which is more than the loss in the years 2011 and 2016 that had the largest Arctic ozone depletion so far.

1 Introduction

It is well established that the Antarctic ozone hole (Farman et al., 1985; Jones & Shanklin, 1995) is caused by chemical ozone depletion in spring through catalytic cycles driven by chlorine and bromine compounds (e.g., Canty et al., 2016; WMO, 2019). For these cycles to run efficiently, chlorine needs to be activated from the so called reservoir compounds HCl and ClONO₂ by heterogeneous reactions. These heterogeneous reactions take place only at low temperatures present typically in polar winter and spring; these reactions occur on the surfaces of Polar Stratospheric Clouds (PSCs) and on cold sulfate aerosol (Solomon, 1999; Drdla & Müller, 2012).

The reason for an enhanced stratospheric chlorine and bromine loading, commonly referred to as Equivalent Effective Stratospheric Chlorine (EESC) is chlorine and bromine released from anthropogenically emitted CFCs and halons (Newman et al., 2007; Engel et al., 2018). Because of the regulations of the Montreal protocol and its amendments and adjustments, the stratospheric halogen loading peaked around the year 2000. Due to the long atmospheric lifetime of the CFCs and halons, the EESC (polar winter conditions) is currently reduced by about 11-12% of the peak value in the year 2002 (Newman et al., 2007; Engel et al., 2018; WMO, 2019). This level corresponds to the EESC of about the year 1992. In addition to the long-term decline of EESC in the atmosphere, there is also an inter-annual variability of inorganic chlorine (Cl_y , i.e. the sum of the destruction products of anthropogenically emitted CFCs) in the polar stratosphere, which is caused by the inter-annual variability of descent in the polar vortex (Strahan et al., 2014). In Antarctica, the expected decline of Cl_y is about 20 pptv/y, whereas the year-to-year variability ranges from -200 to $+150$ pptv (Strahan et al., 2014); this effect will be present in the Arctic as well and points to the importance of an accurate representation of diabatic descent in studies of polar ozone loss.

Strong chemical polar ozone loss in the stratosphere has been identified in cold Arctic polar winters and springs exhibiting a persistent polar vortex into late winter and early spring e.g., in March 1997, 2011, and 2016 (Newman et al., 1997; Müller, Crutzen, et al., 1997; Müller, Grooß, et al., 1997; Tilmes et al., 2004; Weber et al., 2011; G. L. Manney et al., 2011; Grooß et al., 2014; Pommereau et al., 2018). However, stratospheric temperatures in the Arctic in winter and spring are generally much higher than in the Antarctic. However, there are reported cases, where chlorine activation occurs in association with the formation of PSCs over only a small portion of the vortex, but this small cold portion nonetheless resulted in a substantial activation vortex wide (Wegner et al., 2016). Because of a higher dynamic activity in the Arctic, which results in a lower stability and in higher temperatures of the Arctic polar vortex, Arctic ozone depletion is typically much

60 less pronounced and much more variable than in the Antarctic (WMO, 2019; Bernhard
61 et al., 2020).

62 Despite decreasing halogen levels in the stratosphere (Newman et al., 2007; Engel
63 et al., 2018), the lower and more variable stratospheric temperatures may cause larger
64 areas to be perceptible for heterogeneous chemistry and chlorine activation, resulting in
65 ozone depletion, especially in years with a stable polar vortex extending into spring. Rex
66 et al. (2006) put forward the hypothesis that cold Arctic stratospheric winters tend to
67 get colder, resulting in an increasingly strong ozone loss. There is a debate on the ac-
68 curacy of such projections based on studies using extreme value statistics (Rieder & Polvani,
69 2013) or sunlit vortex volumes (Pommereau et al., 2013). Detailed studies of re-analyses
70 and homogenized radiosonde data (Bohlinger et al., 2014) confirm a cooling of -0.41 K/decade
71 over the past 3 decades in Arctic spring that fosters ozone depletion, but also an increase
72 of planetary wave activity that disturbs the stability of the polar vortex, which is not
73 favourable for chemical ozone loss. However, in different models participating in the Chemistry-
74 Climate Model Initiative (CCMI), there is no statistically significant change in the fre-
75 quency of major mid-winter stratospheric sudden warmings (SSWs), the largest instance
76 of wintertime variability in the Arctic stratosphere (Ayarzagüena et al., 2018). Rieder
77 et al. (2014) show that the temperature decrease in the lower stratosphere is dominated
78 by ozone depleting substances as opposed to well-mixed greenhouse gases that dominate
79 the temperature trend in the upper stratosphere. In the Arctic, the role of dynamical
80 processes for determining springtime ozone will likely increase in the future, because of
81 the effect of declining ODSs and rising greenhouse gases. There are Chemistry Climate
82 Model (CCM) simulations that show that even after 2040, when EESC has substantially
83 declined, early springtime Arctic total column ozone in particular years can drop by about
84 50 to 100 DU below the long-term average (Langematz et al., 2014; Bednarz et al., 2016).

85 Even in cold Arctic winters, ozone mixing ratios at the end of the ozone depletion
86 period are commonly much higher than in the Antarctic, which has implications for chlo-
87 rine recovery. In the Antarctic, for very low ozone mixing ratios, chlorine recovery is largely
88 into HCl, whereas in the Arctic, for higher ozone mixing ratios, chlorine recovery is largely
89 into ClONO₂ (Crutzen et al., 1992; Müller et al., 1994; Douglass et al., 1995; Groöb et
90 al., 1997). For the chlorine recovery into HCl in the Antarctic, the occurrence of extremely
91 low ozone mixing ratios in the vortex is essential (Groöb et al., 2011; Müller et al., 2018).

92 Moreover, a strong Arctic polar vortex leads to a reduced poleward transport of
93 ozone, contributing further to low total column ozone values in spring. There have been
94 warm Arctic years in which no chlorine-catalyzed ozone depletion was detected and cold
95 years with a stable vortex that experienced significant ozone depletion (e.g., Tilmes et
96 al., 2006; Müller et al., 2008; Bernhard et al., 2020).

97 Although the main aspects of polar chlorine activation and the resulting catalytic
98 ozone loss are established (WMO, 2019), some scientific questions in this area remain
99 that have not yet been resolved. In particular, there is observed HCl depletion in the dark
100 polar vortex which cannot be reproduced given the processes currently implemented in
101 models (Groöb et al., 2018).

102 Here we report on the Arctic ozone depletion in the year 2020 that is unprecedented
103 (Bernhard et al., 2020; G. Manney et al., 2020). The chemical composition observed by
104 satellite instruments is compared with the simulations by the Chemical Lagrangian Model
105 of the Stratosphere (CLaMS). The major causes of the reported severe ozone depletion
106 in the Arctic in 2020 are the low stratospheric temperatures and the exceptionally sta-
107 ble polar vortex extending into spring (Lawrence et al., 2020).

2 Data description

2.1 AURA-MLS

The observations by the Microwave Limb Sounder (MLS) on board the AURA satellite are the main data set used in this study. Further details of the MLS measurements in the Arctic winter 2019/2020 are reported by G. Manney et al. (2020). MLS observes in limb viewing geometry on the so-called A-train orbit, circling the earth 15 times daily covering latitudes from 82°S to 82°N. We use the MLS version 4.2 data (Livesey et al., 2017). Here observations of O₃, N₂O, HCl, H₂O, and HNO₃ are used for model initialisation, boundary conditions and for comparison with the model results.

2.2 ACE-FTS

The experiment ACE-FTS on the satellite SCISAT is a Fourier Transform Spectrometer (FTS) with high spectral resolution (0.02 cm⁻¹) operating from 2.2 to 13.3 μm wavelength employing a Michelson interferometer (Bernath et al., 2005). Since 2004, it observes around 30 profiles per day in solar occultation geometry with the majority of the measurements being in high latitudes. Here we use version 3.6 data of O₃, N₂O, as well as the five main components of total inorganic nitrogen NO_y (NO, NO₂, N₂O₅, HNO₃, ClONO₂).

2.3 AURA-OMI

The Ozone Monitoring Instrument (OMI) onboard the the satellite AURA is a limb viewing spectrometer that measures the solar radiation back-scattered by the Earth's atmosphere and surface over the entire UV/VIS wavelength range from 270 to 500 nm (Levelt et al., 2006). It is in the succession of previous Total Ozone Monitoring Spectrometers (TOMS). Since 2004, OMI provided measurements in global coverage of the distribution of atmospheric ozone column on a daily basis.

3 Model description

3.1 Model setup

The simulations described here have been performed with the Chemical Lagrangian Model of the Stratosphere (CLaMS). Unlike in previous publications, the chemical transport model CLaMS is driven here by operational analyses by the European Centre of Medium Range Weather Forecasts (ECMWF), as neither re-analyses ERA-Interim nor ERA-5 were available when the simulations were performed. Thus the vertical velocities were derived from heating rates calculated by an offline radiation module based on the Morcrette scheme as in an earlier model setup (Morcrette, 1991; Zhong & Haigh, 1995; Konopka et al., 2004). The vertical model coordinate is the hybrid potential temperature ζ (Konopka et al., 2004; Pommrich et al., 2014) that is identical to potential temperature above 100 hPa and corresponds to pressure levels below with a smooth transition. The model simulation described here starts on 1 November 2019 and runs until mid-April. The model domain consists of the Northern hemisphere with a horizontal resolution of 100 km. The vertical range is from the surface to 900 K potential temperature divided into 32 levels with a variable resolution in the stratosphere ranging from 0.7 km at 9-12 km to 1 km at the top layer yielding about 820.000 air parcels in total.

The chemical scheme used here is the same as described earlier (McKenna et al., 2002; Groöf et al., 2011, 2018). In particular, a comprehensive stratospheric chemistry scheme is employed which includes a full set of heterogeneous reactions. The vertical redistribution of NO_y and H₂O due to sedimenting NAT and ice particles is determined by Lagrangian NAT and ice particle tracking (Groöf et al., 2018; Tritscher et al., 2019).

154 This algorithm allows denitrification to be represented in the model and also dehydra-
 155 tion, albeit to a smaller extent in the shown simulation.

156 3.2 Initialisation and boundary conditions

157 Both the chemical composition at the begin of the simulation and the boundary
 158 conditions at the surface and the top model layer at 900 K potential temperature are de-
 159 rived from a variety of data and model results. This is similar to previous work (Grooß
 160 et al., 2014). Data from the Microwave Limb Sounder (MLS) within ± 2.5 days of the
 161 initial time for O_3 , HNO_3 , H_2O , HCl and N_2O have been used. The observation loca-
 162 tions have been transferred to the initial time using CLaMS trajectories and averaged
 163 to a 2° latitude \times 6° longitude grid. In the troposphere below 350 K potential temper-
 164 ature the initialisation of these compounds is taken from a multi-annual CLaMS sim-
 165 ulation with simplified chemistry setup (Pommrich et al., 2014). For total inorganic ni-
 166 trogen (NO_y), chlorine (Cl_y) and bromine (Br_y), tracer correlations with N_2O have been
 167 used. NO_y was derived from ACE-FTS from the sum of the observations of HNO_3 , NO ,
 168 NO_2 , N_2O_5 and $ClONO_2$ on 1 October to 21 November 2019 in different latitude bands
 169 that correlate well with N_2O . Inorganic chlorine (Cl_y) and bromine compounds (Br_y)
 170 were initialized using correlations with N_2O derived from balloon observations (Grooß
 171 et al., 2018). The partitioning within these chemical families has been taken from a 2-
 172 D reference model (Grooß, 1996) with updated boundary conditions (WMO, 2019).

173 The chemical composition at the upper boundary at the 900 K potential temper-
 174 ature level was calculated using the same data sources. Twice per month, the data as
 175 described above at this level are mapped and averaged into bins of equivalent latitude
 176 (Lary et al., 1995). From that, the chemical composition at the upper boundary is de-
 177 termined. Simulations with this setup had been performed for Arctic winters since 2010.

178 4 Results

179 The CLaMS simulation aims to reproduce the processes involved in the Arctic strato-
 180 spheric ozone depletion over the winter 2019/2020. In the simulation, the formation of
 181 NAT particles starts already on 16 November 2019. We analyze the observations and the
 182 model results in the polar vortex using the concept of equivalent latitude Φ_e (Lary et
 183 al., 1995). The resulting chlorine activation is exceptionally long. (ClO_x/Cl_y) averaged
 184 over the polar vortex core ($\Phi_e > 75^\circ N$) increases above 50% rather early on 14 Decem-
 185 ber between about 490 K and 540 K potential temperature. Therefore some ozone de-
 186 pletion occurred already in December. Chlorine activation in 2020 lasts until 22 March,
 187 and the polar vortex remains still stable into the month of April (Lawrence et al., 2020).

188 4.1 Descent in the polar vortex

189 For validation of the simulation results, it is essential to demonstrate the ability
 190 of the model to reproduce atmospheric observations. One important aspect of the sim-
 191 ulation is whether the transport and dynamics is represented correctly. Especially the
 192 diabatic descent of air inside the polar vortex should be realistic. Diabatic descent in a
 193 particular winter determines the ozone mixing ratios at a given potential temperature
 194 level in spring, but also the available Cl_y (Strahan et al., 2014). Thus we investigated
 195 whether the descent of vortex air is simulated properly. Figure 1 shows a comparison of
 196 N_2O profiles in the vortex core observed by ACE-FTS between 23 and 29 March in the
 197 vortex ($\Phi_e > 70^\circ N$) and the corresponding CLaMS data evaluated at the observation lo-
 198 cations. The solid lines show the mean mixing ratio profile and the dotted lines show the
 199 standard deviation. The CLaMS simulation shows a slightly lower descent than observed
 200 between early November and late March, for 200 ppbv N_2O corresponding to a change
 201 of 54 K instead of 64 K in potential temperature. For 100 ppbv N_2O the simulated de-

202 scent is 68 K instead of the observed 90 K. This difference of vertical displacements is im-
 203 portant, but is in the order of the vertical model resolution.

204 4.2 Ozone depletion

205 The focus of this study is on the simulation of ozone depletion in the Arctic in 2020.
 206 First, the overall development of ozone is compared with MLS observations. As MLS mea-
 207 sures about 3500 profiles per day, the comparison is done in a way that similar air-masses
 208 are averaged. For that, both the data and the simulations are averaged daily into equiv-
 209 alent latitude and potential temperature bins. The width of the equivalent latitude bins
 210 is chosen such that they correspond to equal geographical areas. Figure 2 displays dif-
 211 ferent illustrations of these averages. Panel a shows the time development of the vortex
 212 core average profiles for equivalent latitudes $\Phi_e > 75^\circ\text{N}$, panel b shows the time develop-
 213 ment of the mixing ratio at the 450 K potential temperature level as a function of equiv-
 214 alent latitude. Panels c and d show this average for two chosen dates, 31 January and
 215 25 March respectively. Figure 3 shows the corresponding development of ozone derived
 216 from the CLaMS simulation. Although there are some small differences, the ozone de-
 217 pletion over winter and spring is well reproduced by the simulation. The strongest ef-
 218 fect of the ozone depletion is seen between 400 K and 500 K potential temperature to-
 219 wards the end of March. The gradients of ozone at the edge of the polar vortex are re-
 220 produced well. On 27 March, the mean MLS ozone mixing ratio (\pm standard deviation)
 221 on the 450 K level for equivalent latitudes above 75°N is 0.21 ± 0.25 ppmv. The correspond-
 222 ing ozone mixing ratio from the CLaMS simulation is 0.29 ± 0.26 ppmv.

223 Figure 4 shows the chemical ozone change that is calculated as the difference be-
 224 tween the simulated ozone and a passive ozone tracer that has identical initialisation and
 225 boundary conditions and transport, but no chemical change. The maximum ozone de-
 226 pletion in the vortex core average ($\Phi_e > 75^\circ\text{N}$) is 2.74 ppmv reached on 30 March on the
 227 470 K potential temperature level. After that day this value remained constant until mid-
 228 April indicating no additional ozone loss but also no significant mixing with mid-latitude
 229 ozone-rich air.

230 Also visible is an ozone depletion in mid-latitudes in the middle stratosphere be-
 231 tween 600 K and 700 K. This ozone depletion is dominated by NO_x -induced catalytic cy-
 232 cles, but this topic is not further considered here.

233 With respect to the variability of surface UV radiation, the total ozone column is
 234 important. To calculate the model total ozone column, the ozone column above 900 K
 235 potential temperature needed to be added to the column over vertical model domain.
 236 The column above 900 K amounts to about 30 DU as determined from a climatology de-
 237 pending on equivalent latitude and time of year (Groß & Russell, 2005). The geograph-
 238 ical distribution of ozone columns is also well reproduced by the simulation. Figure 5
 239 shows the comparison of the geographical distribution of ozone columns from AURA-
 240 OMI and CLaMS for March 29. Note that the OMI data are observed and accumulated
 241 over a 24-h period (resulting in a discontinuity on the date line) while the model data
 242 are displayed for the synoptic time 12:00 UTC. This comparison shows the ability of the
 243 CLaMS simulation to reproduce the gradients in the ozone distribution accurately.

244 Figure 6 shows the average development of the simulated ozone column depletion
 245 within the vortex ($\Phi_e > 70^\circ\text{N}$) between 350 and 600 K potential temperature. For this vol-
 246 ume the maximum ozone depletion of 133 DU has been reached on 25 March. For com-
 247 parison, we also show this quantity for comparable CLaMS simulations for the years with
 248 significant Arctic ozone depletion in the previous decade including 2011 and 2016. The
 249 calculated column ozone depletion in 2020 clearly exceeds the depletion in 2011 and 2016.

250 The simulated ozone profiles were also compared with observations. Figure 7 shows
 251 the average vortex core ozone profile observed by ACE-FTS for the time between 23 and

252 29 March. The corresponding CLaMS simulations were evaluated at the ACE-FTS tan-
 253 gent point locations. Dotted lines show the standard deviation within these profiles. Around
 254 450 K potential temperature, the simulations show lower ozone mixing ratios but within
 255 the standard deviation. This could possibly be caused by the fact that the ACE-FTS oc-
 256 cultation retrieval are also influenced by larger ozone concentrations at higher levels along
 257 the viewing beam.

258 The simulated ozone mixing ratios did not quite reach the low levels reported for
 259 the Antarctic ozone hole (Solomon et al., 2005). Grooß et al. (2011) showed that in the
 260 Antarctic ozone hole, the chemical ozone depletion can continue until very low ozone mix-
 261 ing ratios of the order of 10 ppbv have been reached. Before the minimum ozone mix-
 262 ing ratios are reached, chlorine activation is maintained even though there is a consid-
 263 erable speed up of the reaction of Cl with CH₄ (Douglass et al., 1995; Grooß et al., 2011;
 264 Müller et al., 2018) caused by decreasing ozone mixing ratios themselves. However, this
 265 chlorine deactivation reaction is first balanced by immediate heterogeneous chlorine ac-
 266 tivation. If this balance terminates, it is possible that practically all available inorganic
 267 chlorine species are converted to HCl at a time when PSCs are still available (Müller et
 268 al., 2018). For typical Arctic ozone mixing ratios, the dominant fraction of the active chlo-
 269 rine species undergo deactivation into ClONO₂ by the reaction ClO+NO₂ (Müller et al.,
 270 1994; Douglass et al., 1995).

271 The development of individual air masses with very low ozone mixing ratios can-
 272 not be well simulated, if mixing or diffusion is over-estimated in a numerical model. Here
 273 we employ Lagrangian simulations which have an advantage as they follow individual
 274 air masses for some time without mixing. Also, Lagrangian simulations did reflect the
 275 low Antarctic ozone mixing ratios (Grooß et al., 2011). In the current simulation, the
 276 lowest simulated ozone mixing ratio in the polar lower stratosphere of 48.5 ppbv was reached
 277 on March 24 (84°N, 131°E, $\theta=439$ K). This individual air parcel was not affected by the
 278 mixing algorithm for 50 days. Figure 8 shows the temporal development of ozone and
 279 chlorine compounds of this air parcel. For an extended period, from about mid Febru-
 280 ary to early March ClO_x levels remain strongly elevated in this air parcel, while the mix-
 281 ing ratios of HCl, ClONO₂, and HOCl remain very low. The NAT equilibrium temper-
 282 ature T_{NAT} is shown in the top panel calculated using simulated HNO₃ and H₂O. Changes
 283 in T_{NAT} are due to local denitrification and nitrification by the sedimenting NAT par-
 284 ticles. Also shown is the threshold temperature for chlorine activation on liquid aerosols
 285 T_{ACL} as defined by Drdla and Müller (2012) that does not depend on HNO₃. There is
 286 no deactivation occurring during until early March, neither into ClONO₂, nor into HCl.
 287 Starting in early March, similar as in the Antarctic (e.g., Grooß et al., 1997), the air par-
 288 cel experiences a complete deactivation into HCl with temperatures partly below T_{NAT} ,
 289 albeit on a longer time scale of about 2 weeks. The exact location of the simulated low-
 290 est ozone mixing ratio is not relevant and it is very sensitive on the ozone initialisation
 291 cannot be done with an accuracy of a few ppbv. Also with respect to denitrification and
 292 T_{NAT} , the model results are unlikely to represent the precise day-to-day variability at
 293 a specific location. But on a larger vortex scale, they should be representative. The vor-
 294 tex core average HCl and ClONO₂ ($\Phi_e > 70^\circ\text{N}$) at 500 K potential temperature are shown
 295 in Fig. 9a together with the corresponding MLS HCl data. The rate of increase of HCl
 296 mixing ratios in late March is similar suggesting also similar low ozone mixing ratios.

297 Figure 9 indicates a discrepancy in HCl during the onset of chlorine activation. Grooß
 298 et al. (2018) showed that there is a yet unexplained discrepancy in HCl between simu-
 299 lations and observations. In the cold dark polar vortex core in early winter stratosphere
 300 the observed depletion of HCl is faster than simulated. This effect is most pronounced
 301 in the Southern hemisphere, but has been also seen in the 2016 Arctic winter. It is also
 302 seen here for the year 2020, but to a lesser extent than in the simulation for the winter
 303 2015/2016 (Grooß et al., 2018). This is probably due to the earlier onset of chlorine ac-
 304 tivation in 2019/2020 when still more sunlight was available.

5 Conclusions

The Arctic winter 2019/2020 was exceptionally cold in the polar stratosphere and the polar vortex was stable for a long time with the consequence of significant ozone depletion. This has been shown by CLaMS simulations that correspond well to the observations. Even though the stratospheric halogen loading (EESC) has decreased since its peak in 2000 by about 11-12%, these meteorological conditions cause an Arctic ozone depletion that has been unprecedented. The calculated vortex average ozone column depletion between 350 and 600 K potential temperature was 133 DU in late March. On the 470 K level in the vortex core the ozone mixing ratio was depleted by 2.74 ppmv on 30 March. The lowest simulated ozone mixing ratio in the lower stratosphere was about 50 ppbv. If the temperature trend in the lower polar stratosphere continues, more Arctic winters with similar ozone depletion are possible even though the chlorine loading is decreasing.

Acknowledgments

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The satellite data used in this study are publicly available. MLS level 2 data (version 4) were obtained from https://acdisc.gesdisc.eosdis.nasa.gov/data/Aura_MLS_Level2, OMI level 3 data from <https://ozonewatch.gsfc.nasa.gov/data/omi> and ACE-FTS data (version 3.6) from https://databace.scisat.ca/level2/ace_v3.5_v3.6. CLaMS model results in the equivalent latitude/potential temperature averages are available from https://datapub.fz-juelich.de/slcs/clams/ozoneloss_2020.

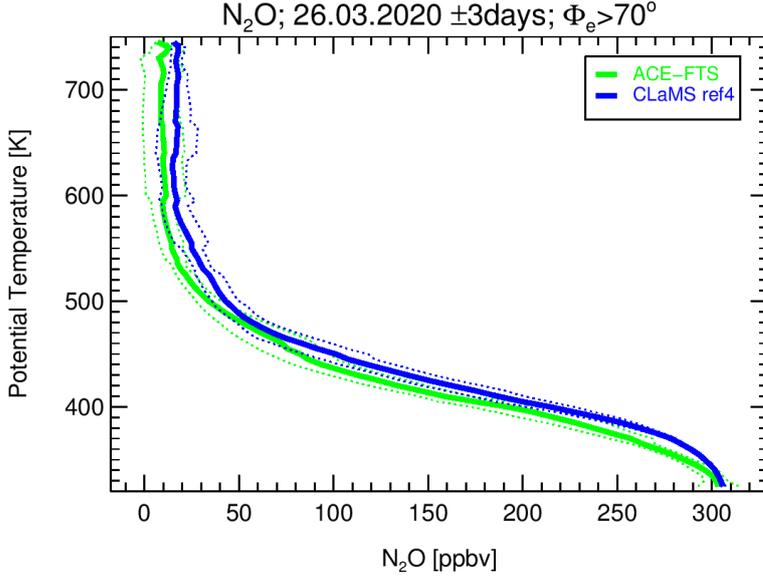


Figure 1. ACE-FTS comparison: vortex ($\Phi_e > 70^\circ\text{N}$) N_2O mixing ratios in the time frame 26 March ± 3 days. The green line shows the average N_2O mixing ratio profile. Dotted lines correspond to the standard deviation of the measurements ($\pm 1\sigma$). Blue lines are from the CLaMS simulation interpolated to ACE-FTS profile locations evaluated correspondingly.

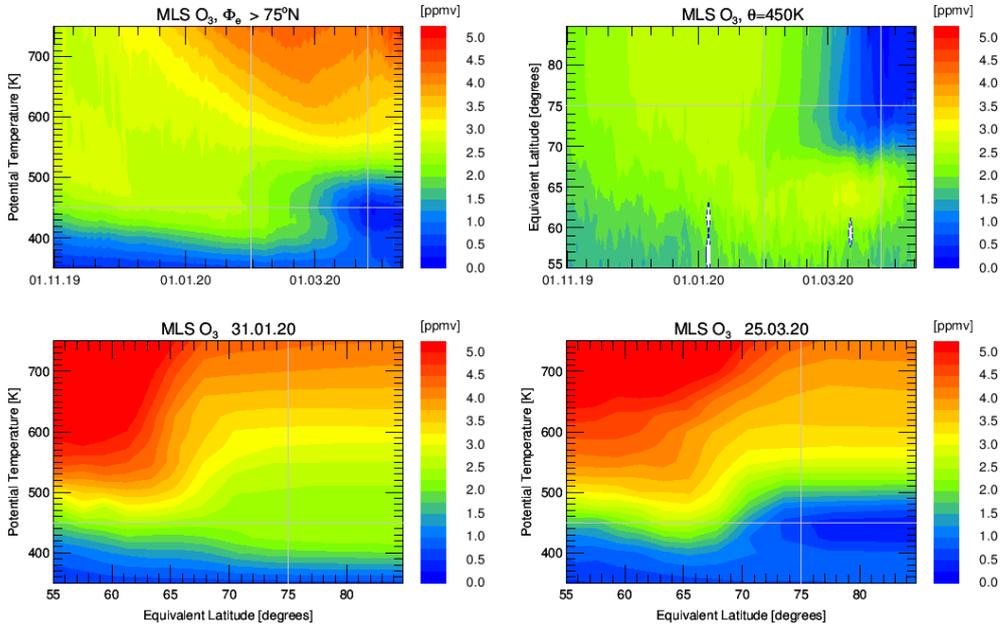


Figure 2. MLS ozone observations averaged in equivalent latitude/potential temperature space. The top left panel shows the vortex ($\Phi_e > 70^\circ\text{N}$) average as a function of potential temperature and time. The top right panel shows the observations on the 450 K potential temperature levels as function of equivalent latitude and time and the lower panels show these data for two chosen days, 31 January and 29 March.

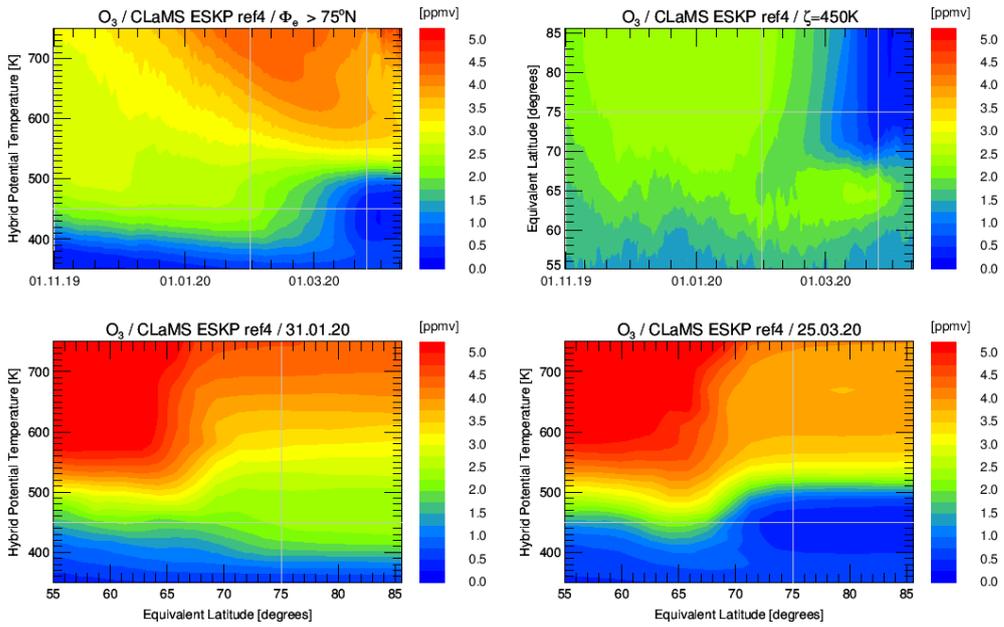


Figure 3. CLaMS simulation of ozone plotted corresponding to Figure 2.

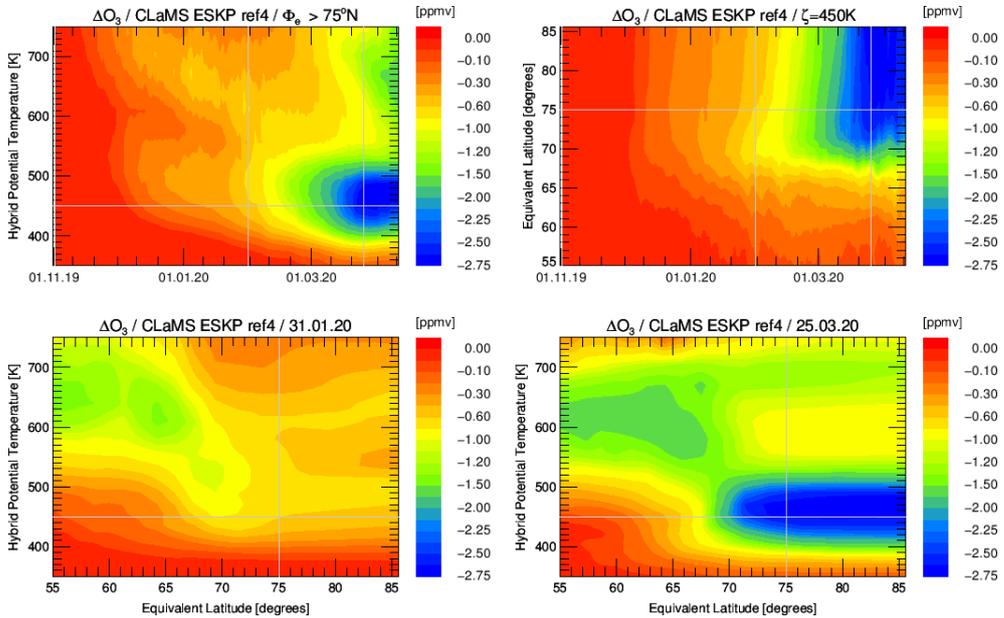


Figure 4. Simulated chemical ozone depletion determined by the difference of simulated ozone and the passive ozone tracer corresponding to Figures 2 and 3.

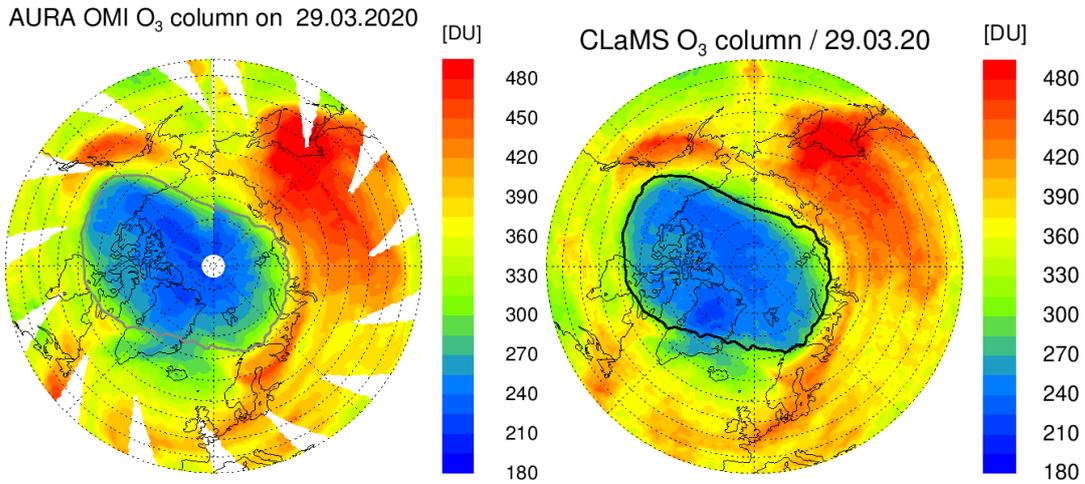


Figure 5. Ozone columns for 29 March (a) from AURA-OMI and (b) from the CLaMS simulation. Ozone column above 900 K in CLaMS plot is taken from the HALOE-climatology (Groß & Russell, 2005). The solid line marks the vortex edge after Nash et al. (1996) on the 450 K potential temperature level.

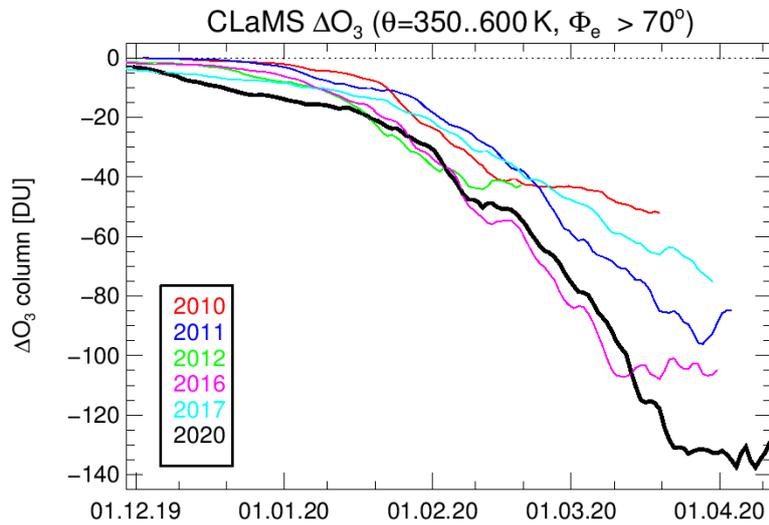


Figure 6. Development of simulated ozone column depletion between 350 and 600 K potential temperature. For comparison, also comparable simulations for other years of significant Arctic ozone depletion including 2011 and 2016 are shown. The maximum column ozone depletion on 2020 is 133 DU at 25 March.

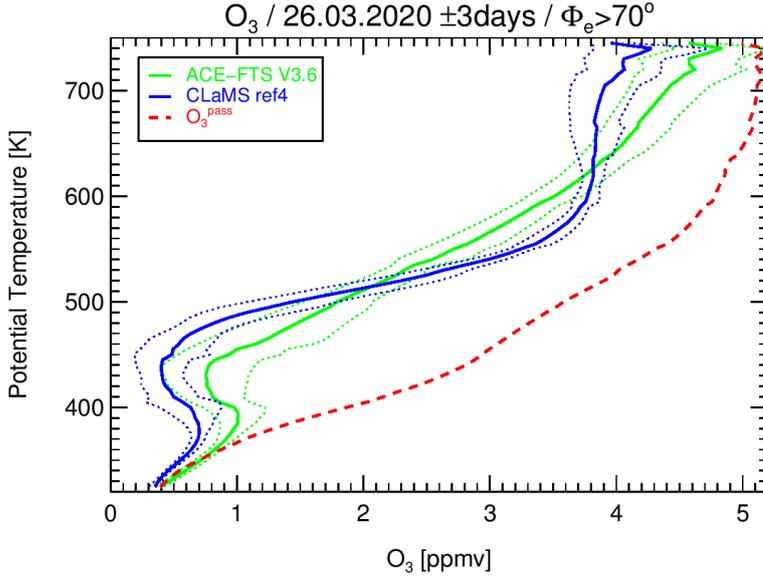


Figure 7. ACE-FTS comparison: vortex ($\Phi_e > 70^\circ\text{N}$) ozone mixing ratios in the time frame 26 March ± 3 days. The green line shows the average ACE-FTS ozone mixing ratio profile. Dotted lines correspond to the standard deviation of the measurements ($\pm 1\sigma$). Blue lines are from the CLaMS simulation interpolated to ACE-FTS profile locations evaluated correspondingly. The red dashed line marks the mixing ratio of the passive ozone tracer.

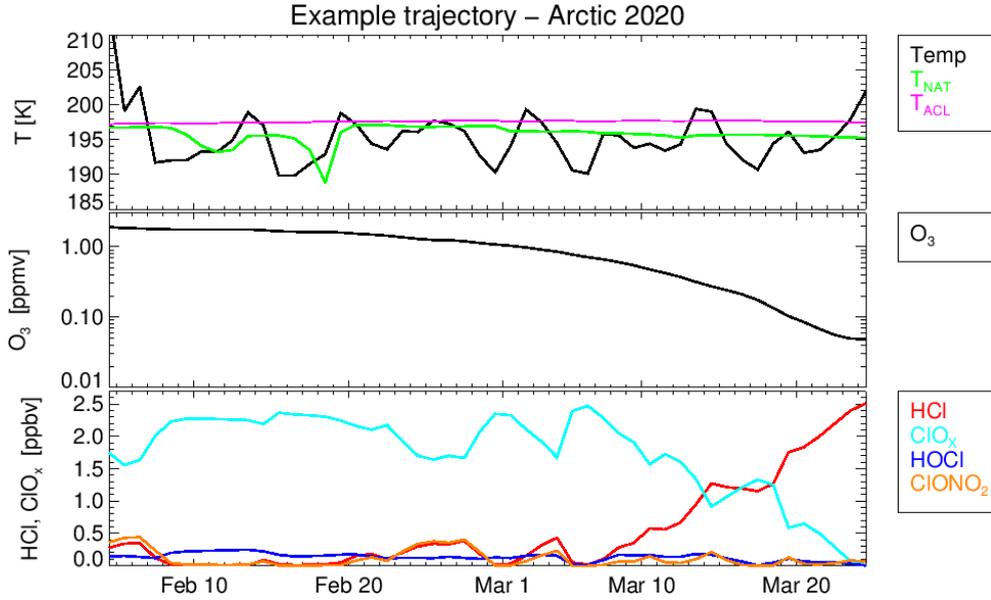


Figure 8. 50-day development of one example air parcel trajectory from the CLaMS simulation that is not affected by mixing over the shown time period. The top panel shows the temperature, the NAT equilibrium temperature T_{NAT} as well as the threshold temperature for chlorine activation on liquid aerosols T_{ACL} after Drdla and Müller (2012). The mixing ratio of ozone (middle, logarithmic ordinate) and chlorine compounds HCl, ClONO₂, ClO_x (=ClO+2×Cl₂O₂, 2×Cl₂) and HOCl (bottom) are shown. The ozone mixing ratio on 24 March (84°N, 131°E, $\theta=439$ K) is 48.5 ppbv.

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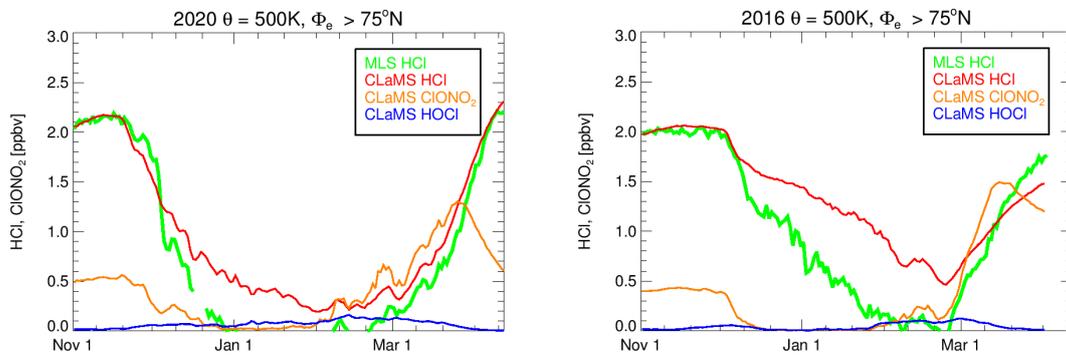


Figure 9. Vortex core average HCl, ClONO₂ and HOCl mixing ratios on the 500 K potential temperature level simulated by CLaMS and observed HCl by MLS. (a) Current simulation for 2019/2020 and (b) simulation for 2015/2016 by Groöß et al. (2018).