Response to Reviewer #2

General

In the current manuscript, Li et al has introduced the retrieval method (ZSL-DOAS) by the ground-based instrument installed at Yellow River Station in the Arctic. Then they have validated the retrieved ozone VCD with some widely used measurements including GOME2, Bremer spectrophotometers and SAOZ, the latter uses DOAS with precise measurements of stratospheric constituents during twilight. All these show the observed ozone value is much lower in late winter/early spring in 2020 comparing with the recent five years. Then the authors have investigated the daily variability in ozone changes by calculating the 4-year mean ozone value (2017-2021 excluding the extreme year 2020) and the absolute (and the relative percentage) difference with respace to the mean (?) and have tried to link it to the dynamics (for example by looking at the temperature, PV evolutions). They have also done a model simulation (SD-WACCM) to look at the model performance and the chemical species changes. However, the paper is not well written though the message is still clear for me. There are many confusions (for example, definitions like ozone loss etc. that are quite different from the community has used). It is no doubt that the current ZSL-DOAS observations give another evidence for the unusual Arctic 2020 spring ozone and the unique dataset will be of interest to the atmospheric community. There are many studies over many years show that chlorine and bromine compounds are responsible for the polar ozone depletion in winter and spring. However, the current manuscript has not provided the firm conceptual advance in our understanding of Arctic ozone depletion and there is no new insight into the underlying mechanism responsible for the Arctic ozone depletion. Therefore, the paper has to be rejected or rewritten to find something new.

Author's Response:

We would like to thank the reviewer #2 for the careful and valuable comments, which enable us to improve our study and the manuscript remarkably. Please kindly find our point-to-point response to the problems/comments below in blue and the change of the manuscript in orange.

Figure 6 shows indeed not the ozone loss, but the ozone difference between 2020 and the 4-year mean. It has been revised. Please see P34. There are many studies on the Arctic winter 2019/2020 already, so we have presented this work as a Measurement Report, in which the measurements are reported and the consistency with other studies and measurements are shown.

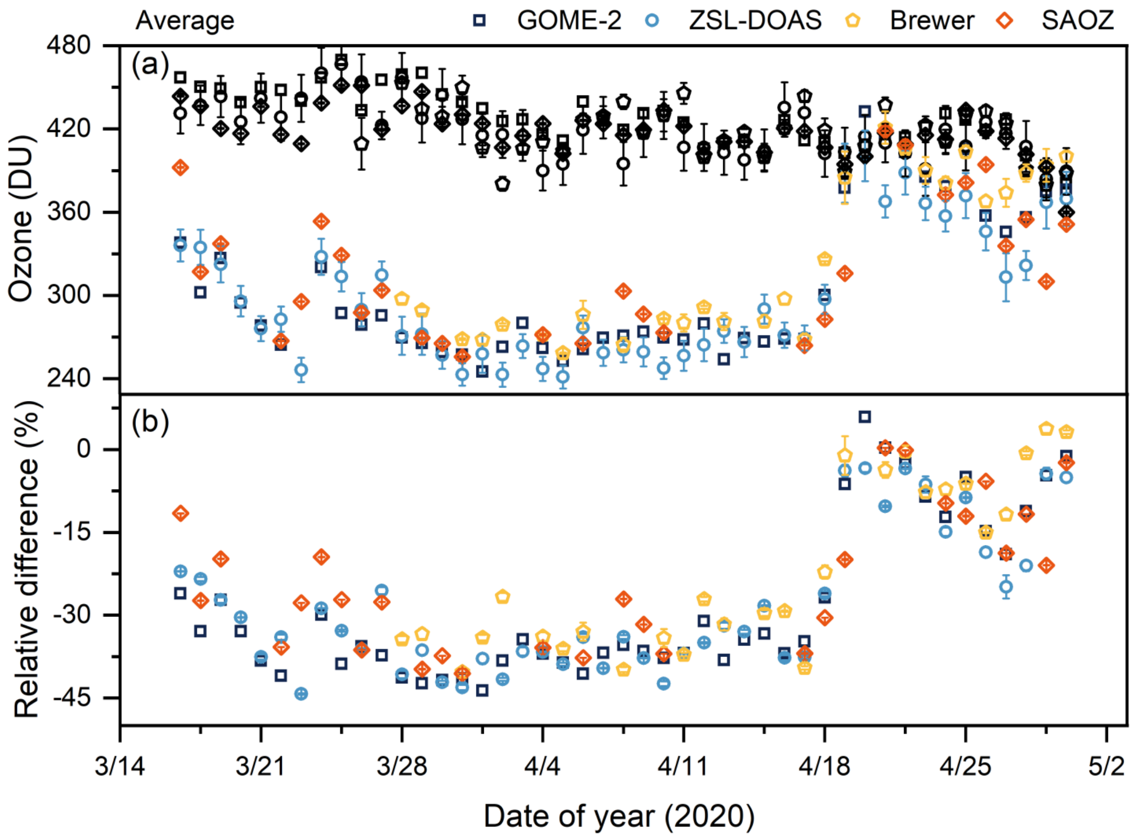

Figure 5. (a) Ozone data for 2020 and the average ozone data (black) of 2017, 2018, 2019, and 2021. (b) relative ozone difference for 2020.

Specific Comments:

1. The title is too general. "Research" is quite broad.

Author's Response:

Thanks for the reviewer's suggestion. The title has been revised as "The unusual spring 2020 Arctic stratospheric ozone depletion above Ny-Ålesund by ground-based ZSL-DOAS".

2. Line 13 in the Abstract: "Severe", the "third and most severe" is vague. There are still larger Arctic ozone loss for other years. Do you mean the long-lasting cold polar vortex years?

Author's Response:

The sentence has been revised. Please see P1 lines 15–16.

"Of the severe stratospheric ozone depletion events (ODEs) reported over the Arctic, the most severe occurred during the spring of 2020."

3. Actually, I find "event" is confusing.

Author's Response:
Many studies have reported this event about unprecedented Arctic ozone depletion in the year 2020 (Dameris et al., 2021; Feng et al., 2021; Manney et al., 2020; Wohltmann et al., 2020).

4. There are many "normal" in the whole text. What is the definition for the normal year? Need to be clear with it.

Author's Response:
In this manuscript, the data for 2020 were compared with that for the other years (2017, 2018, 2019, and 2021) in the Arctic. The sentence has been revised. Please see P1 lines 18–20.

"The average ozone VCD over Ny-Ålesund between March 18 and April 18, 2020, was approximately 274.8 Dobson units (DU), which was about 64.7±0.1% of that in the other years (2017, 2018, 2019, and 2021)"

5. Some results are obvious: "effect of the polar vortex on stratospheric ozone depletion", "Chlorine activation" and "bromine compounds"

Author's Response:
Thanks for the reviewer's suggestion. We have presented this work as a Measurement Report, in which the measurements are reported and the consistency with other studies and measurements are shown.

6. The last sentence in the abstract. What is the main point here? Is this relevant to this work?

Author's Response:
The sentence has been deleted. We have rewritten the part in the revised manuscript. Please see P2 lines 38–41.

"By ZSL-DOAS observations, we provided another evidence for unprecedented ozone depletion during the Arctic spring of 2020. The ZSL-DOAS ozone VCD observations can also provide calibration for satellite observations and model simulations, and in the future can provide the support for observations at more Chinese research stations or international local stations in the polar area."

7. Introduction is not well written. Most of them are too general and the background is well known. If you focus on Arctic winter/spring 2020, then you need to brief summary the available publications and what are the unique research questions you need to address or the methods you have applied.

Author's Response:
Thanks for the reviewer's suggestion. The introduction has been added. Please see P2–5 lines 44–126.

“Stratospheric ozone is essential for human health, surface ecosystems, and the climate in general (McKenzie et al., 2011) because it absorbs ultraviolet (UV) solar radiation and converts it into thermal energy. The characteristic absorption bands of stratospheric ozone are mainly located in the Hartley and Huggins zones of the UV region and in the Chappuis zone of the visible spectrum, thereby absorbing almost all UV-C (i.e., wavelengths < 280 nm) and some UV-B (i.e., wavelengths ranging between 280 and 315 nm) radiation. Since the late 1970s, Antarctic stratospheric ozone during the austral spring has decreased sharply, mainly because of elevated concentrations of active chlorine (Farman et al., 1985). When the weather is cold and there is sufficient sunlight, chlorofluorocarbons derived from anthropogenic emissions can be converted to produce active chlorine, and then to maintain the chlorine activation process, which causes ozone depletion (Müller et al., 2018; Solomon, 1999; Tritscher et al., 2021).. As anthropogenic emissions of ozone-depleting substances since the Montreal Protocol was enforced, the concentrations of ozone in the Antarctic stratosphere were predicted to recover to pre-1980 values in 2060 (Solomon et al., 2016; Stone et al., 2021; WMO, 2018; Kuttippurath and Nair, 2017; Strahan and Douglass, 2018).

The severe ozone depletion over the Arctic is relatively uncommon compared with that in the Antarctic. During normal Arctic winters, the polar vortex usually fractures and disperses early due to huge planetary wave activities and Brewer–Dobson circulation dynamics (Manney et al., 2003; Dameris, 2010; Harris et al., 2010). Thus, in the Arctic, the duration of the vortex is shorter and relative ozone loss is also lower (Solomon et al., 2007). However, irregular changes in Arctic ozone in recent years have attracted worldwide attention and challenged the existing model. The most severe Arctic ozone depletion lasted for nearly a month, from March to April 2020 (Dameris et al., 2021). Between mid-February and late March 2020, the persistence of anomalously faint wave activities in the Arctic led to an abnormally persistent and cold vortex, which caused significant ozone loss (Hu, 2020; Kuttippurath et al., 2021; Ardra et al., 2022). This event was the most severe reported low Arctic ozone event, following those that occurred in the springs of 1997 and 2011 (Hansen and Chipperfield, 1999; Manney et al., 2011).

The powerful and persistent vortex during the winter and spring is considered as a main cause of significant ozone depletion in the Arctic (Bognar et al., 2021). Polar stratospheric clouds (PSCs) are classified into three types: nitric acid trihydrate (NAT), ice PSCs, and supercooled ternary solution (STS), and their threshold temperatures for existence are T_{nat} (195 K), T_{ice} (188 K), and T_{sts} (195–197 K), respectively (Toohey et al., 1993; Poole and McCormick, 1988; Solomon, 1999). Extremely low air temperatures are essential to produce PSC. The PSC can be used as a surface for heterogeneous interactions, leading to the conversion of reactive halogens from the halogen reservoirs, which can cause serious ozone loss (Frieβ et al., 2005; Marsing et al., 2019). Although the PSC is not only composed of NAT (Pitts et al. 2009; Spang et al. 2018), the temperature threshold for the existence of NAT provides a good estimate on the occurrence of heterogeneous chemistry (Drdla and Müller 2012; Kirner et al. 2015; Grooß and Müller 2021; von der Gathen et al. 2021). PSC might also grow large enough to precipitate and remove HNO_3 in the stratosphere, which is the reservoir of NO_2. The resulting denitrification from the polar vortex hinders chlorine deactivation by NO_2 (Salawitch et al., 1989; Arblaster et al. 2014). Active chlorine is rapidly photolyzed because of the recovery of spring sunlight when ozone loss occurs via the self-reaction of ClO (Molina and Molina, 1987), as well as the cross-reaction of ClO and BrO (McElroy et al., 1986). It is essential that the vortex retains low temperatures and carries on as a transport impediment so that ozone can remain depleted without NO_2 to inactivate chlorine.

The observed Arctic ozone depletion is invaluable for validating stratospheric ozone simulations and for understanding the processes that cause Arctic stratospheric ozone depletion. Currently, ozone vertical column density (VCD) detection utilizes the characteristic ozone absorption in the UV and visible spectra, which provides accurate ozone identification and quantitative measurements. Ground-based observation of ozone VCD started in the first decades of the twentieth century (Dobson, 1968; Brewer, 1973; Solomon et al., 1987; Bognar et al., 2021). From the 1960s, ozonesondes began to acquire atmospheric ozone data (Logan, 1994; Thomason et al., 2011; Wohltmann et al., 2020; Grooß and Müller, 2021).Since 1978, satellite observations have provided essential data for atmospheric ozone related studies (Kuttippurath et al., 2012; Manney et al., 2020). Among these, ground-based observations are crucial to calibrate remotely sensed observations and optimizing inversion results (Lu et al., 2006). In the 1970s, differential optical absorption spectrometry (DOAS) was developed by Platt and Stutz (2008) and has been widely used to measure several trace gases of ozone, nitrogen dioxide, bromine monoxide, and sulfur dioxide (Hüneke et al., 2017).

The impacts of the aberrantly powerful and persistent vortex on ozone in the Arctic were investigated using satellite observations, ozonosonde measurements, and data from the European Centre for Medium-Range Weather Forecasts (ECMWF) (Wohltmann et al., 2020; Lawrence et al., 2020). The major stratospheric halogen species, chlorine, and bromine were investigated in this ozone depletion event (ODE) (Wohltmann et al., 2017, 2021). In addition, compared to ground-based observation, modelling provides a wider coverage and favours the investigation of ozone depletion. (Müller et al., 1994; Wohltmann et al., 2010; Griffin et al., 2019; Grooß and Müller, 2021). Recently, stratospheric chemical patterns, consisting of a group of heterogeneous reactions, have been developed in various models according to investigations and experiments conducted in the polar area (McKenna et al., 2002; Grooß et al., 2011, 2018; Chipperfield, 1999; Khosrawi et al., 2009; Bekki et al., 2013; Chipperfield et al., 1994; Kinnison et al., 2007; Wohltmann and Rex, 2009). Global and area models using different stratospheric chemical patterns have been applied to simulate ozone columns, which usually compare well with satellite observations and ozonosonde data (Pan et al., 2018; Grooß and Müller, 2021).

Accurate ground-based observations can improve the accuracy and reliability of models as well as enhancing our understanding of the reasons for ozone depletion. In this study, we have developed a ground-based DOAS system that can conduct ozone VCD observations in the Arctic. The zenith scattered light observation mode was applied to measure ozone VCD using the Langley Plot method (Frieß et al., 2005).

We analyze the reasons for this ODE in the unusual spring of 2020 above Ny-Ålesund, Norway. The methods and data are given in Sect. 2, which covers the presentation of the experimental location and DOAS instrument, the DOAS method, the specified dynamics version of the Whole Atmosphere Community Climate Model (SD-WACCM), Global Ozone Monitoring Experiment 2 (GOME-2) observations, Brewer measurements, Système d'Analyze par Observation Zénithale (SAOZ) measurements, ECMWF data, and ozonesonde data. Section 3 presents the results, where Sect. 3.1 describes the results of ozone VCDs from February 2017 to October 2021 and ozone difference in spring 2020. The zenith scattered light DOAS (ZSL-

DOAS) retrieved the daily variations in ozone VCDs, which were in comparison with GOME-2 observations, Brewer, and SAOZ measurements. A detailed characterization of this ODE is presented for establishing the basis of the subsequent analysis. The relationship between Arctic ozone depletion and meteorological conditions in terms to temperature and potential vorticity (PV) is described in Sect. 3.2. In Sect. 3.3, this ODE was analyzed using the SD-WACCM to further illustrate the ozone depletion process, and to explore the effects of chemical depletion and dynamic transport on this ODE. The influence of the halogen species is discussed in Sect. 3.4. The comprehensive summary is provided in Sect. 4."

8. I also find it is difficult to see the purpose of this work. Are you aiming to validate your ZSL-DOAS observations using other measurements? I am not an expert in the retrieval method so it is hard for me to judge your method. What is the difference in the retrieval of ozone from ZSAL-DOAS and SAOZ because SAOZ also uses DOAS method? For the Air Mass Factor (AMF) in section 2.2, the authors have mentioned the related parameters in Table 2 and AMF will be quite different for different assumed profiles (for example Lines 121-123). So I am curious what are these profiles from. It is vague just to mention SCIATRAN without any reference there.

Author's Response:

Thanks for the reviewer's advice. We have presented this work as a Measurement Report, in which the measurements are reported and the consistency with other studies and measurements are shown. Both ZSL-DOAS and SAOZ use the DOAS method, but ZSL-DOAS is an instrument self-developed by our group. A priori ozone profile is obtained from the monthly mean climatology. The sentence has been revised. Please see P7 lines 164–167.

"Here, the Air Mass Factor (AMF) can be obtained from the SCIATRAN model and is influenced by a priori ozone profile, SZA (solar zenith angle), wavelength, and surface albedo. Based on the average monthly climate, a priori ozone profile can be achieved. The SZA calculated in this research ranged between 35° and 80°, with surface albedos between 0.08 and 0.6. Table 2 lists these parameters."

9. SD-WACCM. This section needs some more details since the authors have carried out the simulation. It should come from CESM1 (but which version) but not sure what other changes have been made by the author. I assume this is a released version but ported and run on a different HPCx in China.

Author's Response:

Thank for your suggestion. We have rewritten this section and added description on the WACCM model. In this study, we replace the standard polar stratospheric cloud module with that from Wegner et al. (2013). Similar sentences have been written in P9 lines 204–206.

"The polar stratospheric cloud module used in this study followed Wegner et al. (2013) rather than the standard module of Kinnison et al. (2007), improving the capabilities of WACCM in modelling ozone and its associated components (Brakebusch et al., 2013)."

10. However, some of the emissions and other input data have not been updated/available for year 2020. One simple example is that this version uses the prescribed stratospheric sulphur aerosol density (SAD) which is important for the ozone depletion. How the SAD used in SD-WACCM4 for this? Do you use the previous year's values or fixed values? Which component you are using? It seems that the authors only mentioned MOZART3, so I am not sure if this one "pp_waccm_mozart" used or you also have MAM model in the model simulation etc..

Author's Response:
We have rewritten this section and added description on the WACCM model. Please see P8 and P9, lines 191–206.

"The chemical mechanism of WACCM4 also contains 4 aerosol types heterogeneous reactions: liquid binary sulfate (LBS), supercooled ternary solution (STS), nitric acid trihydrate (NAT), and water-ice. When model temperatures above 200K, only the LBS exists. The surface area density (SAD) of LBS is from SAGE, SAGE-II and SAMS observations (Thomason et al., 1997) and Considine update it (World Meteorological Organization, 2003). With the model atmosphere cooling, the LBS aerosol expands and absorbs both HNO_3 and H_2O to obtain the STS aerosol. Tabazadeh et al. (1994) derived the composition of STS by the Aerosol Physical Chemistry Model (ACPM). The STS aerosol median radius and SAD is derived following the approach of Considine et al. (2000). When model temperatures reach a specified supersaturation ratio of HNO_3 for NAT, HNO_3 containing aerosols are allowed to form. In WACCM4, Peter et al. (1991) set this ratio to 10. NAT median radius and SAD are derived in the same way with STS aerosol. If the derived atmospheric temperature does not exceed the saturation temperature of water vapour on ice (Tsat), then this results in the formation of water-ice aerosols. In WACCM4, the CAM's prognostic water routines gives the condensed phase H_2O, which is conveyed to the chemistry module. According to the method of Considine et al. (2000), the median radius and SAD of water-ice can be derived by this condensed phase H_2O. The polar stratospheric cloud module used in this study followed Wegner et al. (2013) rather than the standard module of Kinnison et al. (2007), improving the capabilities of WACCM in modelling ozone and its associated components (Brakebusch et al., 2013)."

11. For the SD, the author need to realize that this is not a fully nudged version, which all depends on the relaxation time etc. This needs to make clear.

Author's Response:
Thank for your suggestion. We have added some description of the calculation of the meteorological fields. Please see P9, lines 214–221.

"Meteorological fields were calculated using a nudging method in the model (Lamarque et al., 2012). Data for the horizontal winds, temperature, and surface pressure from MERRA-2 were used to drive the physical parameterization from the surface to 50 km (Kunz et al., 2011), which allowed for more accurate comparisons between the measurements of atmospheric composition and the model output (Lamarque et al., 2012). This can be employed for the study of specific weather events. Linear transitions were used in the 50–60 km altitude range and over 60 km, and online calculations were performed. In this study, the MERRA-2 dataset has the same resolution with the SD-WACCM, which can be accessed on the Earth System Grid (https://www.earthsystemgrid.org/home.html) and are obtained from the original resolution (1/2°×2/3°) by a conservative re-gridding procedure (Lamarque et al., 2012; Pan et al., 2019)."

12. I have not heard "thermogenic" layer, should be use the correct term like "lower thermosphere". It is strange to say the "parameters" in the Line 140. Please note it uses CAM4 physics.

Author's Response:
Thank for your suggestion. The word has been revised and we have rewritten the section 2.2. The WACCM4 model is based on the physical parameterizations used in the Community Atmosphere Model version 4 (CAM4) (Neale et al., 2013).

13. It is impropriate to say "the SD-WACCM with meteorological parameters driven by …", note even SD-WACCM most of the temperature etc. are still from WACCM itself (driven by SST, solar etc.…). How can you say "Data from MERRA-2 guaranteed the accuracy of simulated values"? Note that other processes play roles.

Author's Response:
we have rewritten the section 2.2. These sentences have been corrected. Please see P9, lines 215–217.

"Data for the horizontal winds, temperature, and surface pressure from MERRA-2 were used to drive the physical parameterization from the surface to 50 km (Kunz et al., 2011), which allowed for more accurate comparisons between the measurements of atmospheric composition and the model output (Lamarque et al., 2012)."

14. Auxiliary data: Can be concise and have proper references. For example ERA5. Some sentences are not necessary at all. For example. Lines 152, 165.

Author's Response:
Thanks for the reviewer's suggestion. Some sentences have been deleted.

15. It reads to me that the authors just SHOW the results itself, rather than describe the results in a correct/proper way. For example, for Figure4, we can see large daily variability that has never mentioned. We also see the differences among these measurements but have never explained. For example, why your data ZSL-DOAS is much lower than other observations? I don't think the gradient of "~0.92DU per day" is similar for all the "normal" years claimed. It is also not correct to say "Ozone VCD begins to decrease in March."

Author's Response:
Thanks for the reviewer's advice. During the observation period, the average ozone VCD from

ZSL-DOAS was 2.25% lower than that from GOME-2, 4.92% lower than that from Brewer, and 2.26% higher than that from SAOZ. These differences of ZSL-DOAS observations compared to other observations are due to systematic errors of the instrument. These descriptions have been revised. Please see P11, lines 257–261.

"In the other years (2017, 2018, 2019, and 2021), ozone VCD showed a fluctuating downward trend between March and September, with a small upward trend around March and August. In 2020, however, severe ozone depletion occurred between March 18 and April 18, after which ozone VCD gradually increased. Ozone VCD decreased further in mid-May. In around September, the ozone VCD increased obviously again, probably due to clear warming of the polar stratosphere."

16. For Figure 5, I understand the VCD and TCO can be used but the authors need to be consistent in the whole text.

Author's Response:
It has been revised. We have used VCD in the whole text.

17. The presentation for Figure 6 is not good at all. Why the authors term this "ozone loss"? How do you estimate "ozone loss"? It looks that this is just ozone difference between 2020 and the 4-year mean.

Author's Response:
Figure 6 shows indeed not the ozone loss, but the ozone difference between 2020 and the 4-year mean. It has been revised. Please see response to **General** above.

18. For Figure 7, it seems that temperature is from ERA5, why use "measured" in Line 190? It is a reanalysis product, which is from ECMWF model simulation using the data assimilation from the measurements.

Author's Response:
It has been revised. Please see P12, lines 283–284.

"Daily average temperatures of Ny-Ålesund between November 2016 and September 2021 were showed at 70 hPa in the low stratosphere, where significant ozone depletion tends to occur (Fig. 7)."

19. Figure 8, it seems that the authors just look at one time period of ozone and PV evolution, then comes the conclusion of "PV correlates negatively with ozone VCD" etc. The authors seems not have a deep understanding their figures even they made it (for example, stratospheric warming after mid-April that made polar vortex weaker and TOC higher) etc..

Author's Response:
Thanks for the reviewer's suggestion. The Fig. 8 has been deleted. We have rewritten this part.

Please see P12, lines 293–300.

"A cold and stable polar vortex is a prerequisite for ensuring that Arctic stratospheric temperatures are sufficiently low. The 2019/2020 winter was unique and the polar vortex was unusually stable, prolonged, and cold (Lawrence et al., 2020; Wohltmann et al., 2020; Rao and Garfinkel, 2020). A large and strong Arctic vortex lasted from early December anomalously into the final week of April (Kuttippurath et al., 2021). The faint planetary wave activity in the Northern Hemisphere also contributed to the formation of a cold and strong vortex (Feng et al., 2021). Unusually low temperature and strong and prolonged vortex in the 2019/2020 winter provided favourable meteorological conditions for ozone depletion in the Arctic."

20. Figure9, the authors said "unusual low". This is only for 2020, have you made O3 volume mixing ration comparison with others.

Author's Response:
I made O_3 volume mixing ration comparison with that of 2017,2018, and 2019 from the ozonesonde. Besides, I have reviewed relevant literatures that reported unprecedented Arctic ozone depletion in the year 2020 (Dameris et al., 2021; Feng et al., 2021; Manney et al., 2020; Wohltmann et al., 2020).

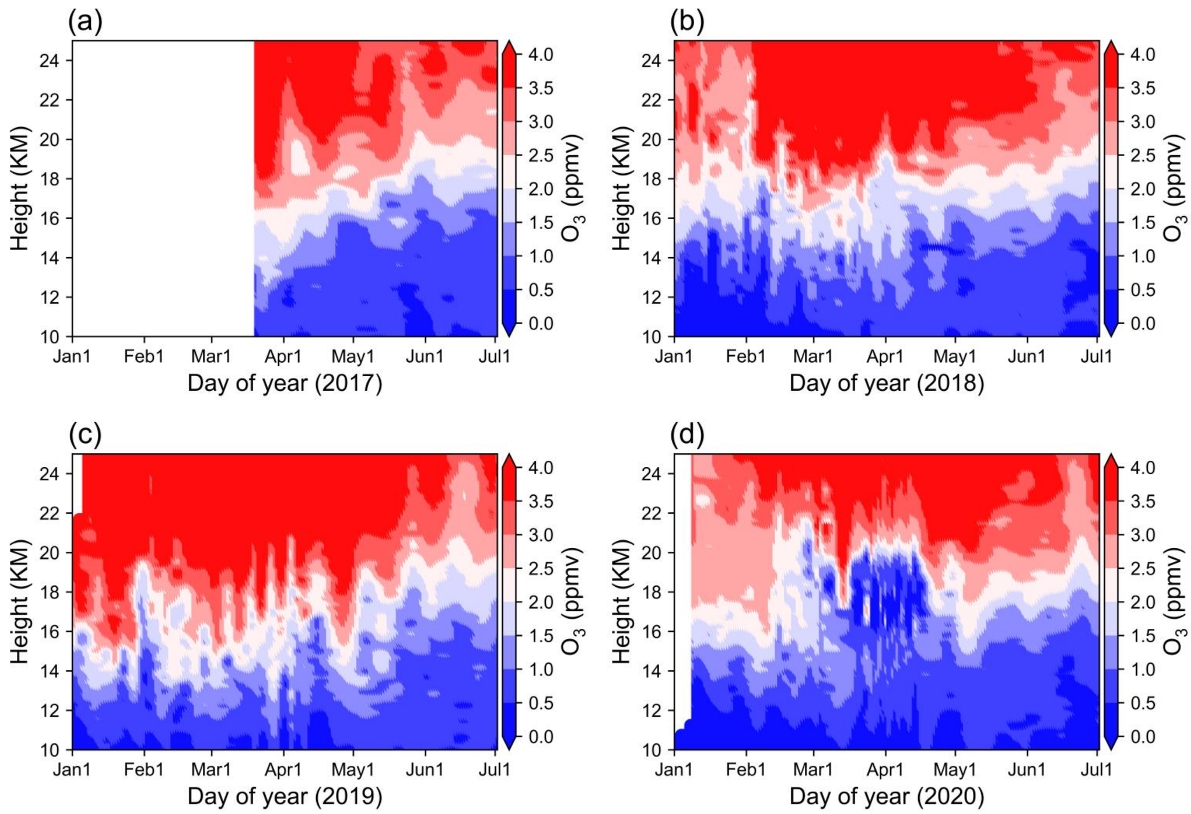

Figure A1. Between January 1 and July 1, ozone profiles of 2017 (a), 2018 (b), 2019 (c), and 2020 (d) from ozonesonde measurements.

21. Why "< 0.5ppmv suggested the ozone was nearly completed depleted. "?.

Author's Response:

By reviewing the literature, "Mixing ratios were consistently below 0.5 ppmv in a wide altitude range (with minima below 0.2 ppmv), indicating near-complete depletion of ozone (Bognar et al., 2021)".

22. For the Tnat, it also depends on H_2O, HNO_3 and H_2SO_4? What is their values used for the Tnat?

Author's Response:

Please see P3, lines 68–76.

"Polar stratospheric clouds (PSCs) are classified into three types: nitric acid trihydrate (NAT), ice PSCs, and supercooled ternary solution (STS), and their threshold temperatures for existence are T_{nat} (195 K), T_{ice} (188 K), and T_{sts} (195–197 K), respectively (Toohey et al., 1993; Poole and McCormick, 1988; Solomon, 1999). Extremely low air temperatures are essential to produce PSC. The PSC can be used as a surface for heterogeneous interactions, leading to the conversion of reactive halogens from the halogen reservoirs, which can cause serious ozone loss (Frieβ et al., 2005; Marsing et al., 2019). Although the PSC is not only composed of NAT (Pitts et al. 2009; Spang et al. 2018), the temperature threshold for the existence of NAT provides a good estimate on the occurrence of heterogeneous chemistry (Drdla and Müller 2012; Kirner et al. 2015; Grooß and Müller 2021; von der Gathen et al. 2021)."

23. Figure 10, why "HNO_3 changes abruptly from abnormally high values to normal values, which indicated the abundant PSC activities of the period" only applies for "Between late January and early February"? What caused the low value patches around 20-22km?

Author's Response:

It also applies other period and the sentence has been revised. The low value of HNO3 at 20-22 km is probably due to low temperatures (Fig. 9c–d) leading to PSC activity and severe denitrification (Ardra et al., 2022). Please see P14, lines 335–337.

"HNO_3 changed abruptly from abnormally high values to normal values, which indicated the abundant PSC activities of the period (Bognar et al., 2021). The low value of HNO_3 at 20-22 km is probably due to low temperatures (Fig. 8c–d) leading to PSC activity and severe denitrification (Ardra et al., 2022)."

24. Figure 11, I saw the model showed a complete HCl depletion on 21 Feb. How the modelled HCl etc. chemical species compared with ACE observation for example?

Author's Response:

The figure displays the simulated average diurnal mixing ratios of ozone, chlorine, and bromine compounds for heights of 17.5 km above Ny-Ålesund, but ACE observations do not have the corresponding altitude. Then, we reviewed the literature and also found that a complete HCl depletion around 20 Feb (Grooß and Müller 2021). Furthermore, the reliability of the model can be validated by comparing it with ozone sonde measurements.

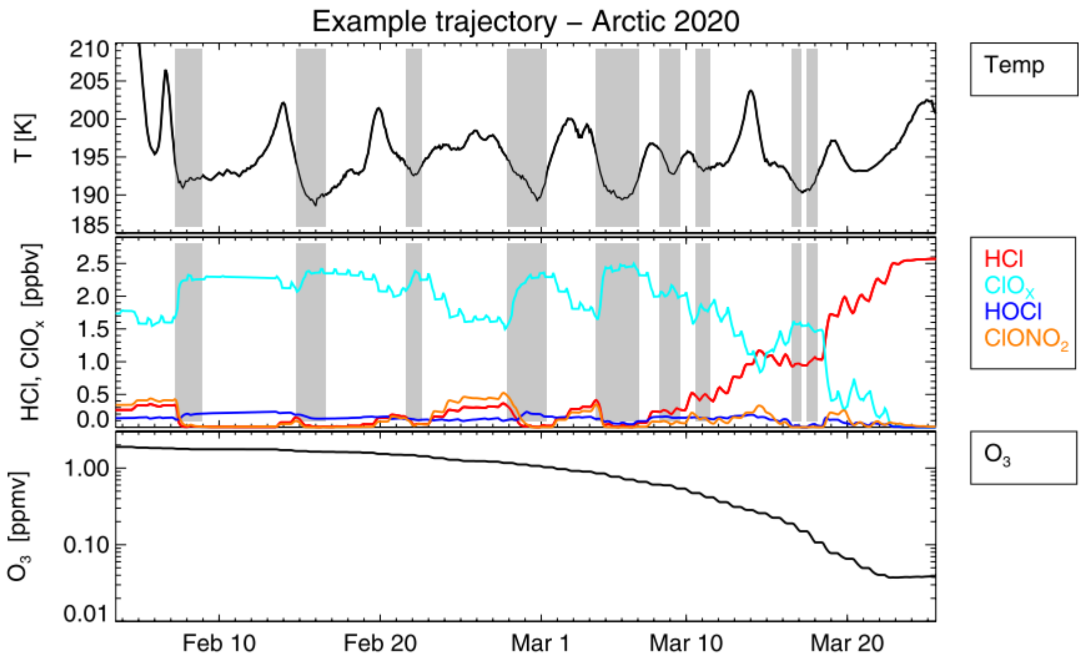

Figure 10. Fifty day development of one example air parcel trajectory from the CLaMS simulation that is not affected by mixing over the time period shown. The top panel shows the temperature along the air parcel trajectory (black). The periods when heterogeneous HCl loss rates ($k_{HCl+ClONO2}$ [$ClONO_2$] + $k_{HCl+HOCl}$ [HOCl]) are larger than 1 day^{-1} are marked as gray shaded areas. The middle panel shows the mixing ratio of the chlorine compounds HCl, $ClONO_2$, ClO_x (= ClO + 2 × Cl_2O_2 + 2 × Cl_2) and HOCl. Ozone is shown in the lowest panel on a logarithmic ordinate. The minimum ozone mixing ratio on 24 March (84°N, 131°E, θ = 439 K) is 38 ppbv.

Figure cited from Grooß and Müller. (2021).

25. This reads like a summary from the main text. I am not sure why the last sentence is matter based on this study.

Author's Response:

Thanks for the reviewer's suggestion. The conclusion part has been revised. Please see P16 and P17, lines 376–414.

"In this research, the ozone VCD was obtained from a ground-based instrument, the GOME-2 satellite, and the Brewer and SAOZ instruments and further evaluated with a correlation analysis. The Pearson correlation coefficients were 0.97, 0.87, and 0.91, and the relative deviations were 2.3%, 3.1%, and 3.5%, respectively. Therefore, we can conclude that the method of observing the VCDs of Arctic ozone using a ground-based DOAS instrument is reliable and valid. Compared to the other four years, the 2020 daily average relative differences from March 18 to April 18 from the GOME-2, ZSL-DOAS, Brewer, and SAOZ datasets were −36.5%, −35.3±0.4%, −33.1±0.7%, and −32.0±0.1%, respectively. The results indicated that all instruments recorded severe ozone depletion from March 18 to April 18, 2020.

Unusually low temperature and strong and prolonged vortex in the 2019/2020 winter provided favourable meteorological conditions for ozone depletion in the Arctic. The ozone and temperature profiles were simulated by SD-WACCM, and these simulations corresponded well with ozonesonde measurements. The model results show that ozone depletion at a height range of 16–20 km is evident from late March to early April, which corresponds to the ozone VCDs obtained from the ground-based instrument. Chlorine and bromine activation were clearly obvious during the Arctic spring of 2020, whereas the partitioning of bromine compounds was different from that of chlorine. Chlorine was predominantly present as HCl and $ClONO_2$ before activation, whereas bromine was predominantly present as HOBr and BrCl before activation. Particularly, bromine existed mainly as HOBr before chlorine activation began. When chlorine was activated, bromine existed mainly as BrCl. In addition, formation of HCl is considered to be the main chlorine deactivation mechanism in Antarctica (Müller et al., 2018). However, in the Arctic, due to HCl increased more slowly than in the Antarctic, chlorine was mainly deactivated as $ClONO_2$.

In summary, by ZSL-DOAS observations, we provided another evidence for unprecedented ozone depletion during the Arctic spring of 2020. The ZSL-DOAS ozone VCD observations can also provide calibration for satellite observations and model simulations, and in the future can provide the support for observations at more Chinese research stations or international local stations in the polar area. Additionally, although WACCM can depict the evolution of ozone during this Arctic ozone depletion event, there are some problems such as overestimation of the temperature and the CH_3O_2+ClO reaction is not considered in the current chemical mechanism of the model. This could be considered in future models to improve the simulation performance."