Response to Reviewer #1

General

I think this manuscript could make a contribution eventually, but it needs work. The reader cannot see clearly what the main message of the paper is (see also below). What is new? My understanding is the following: the first purpose is to introduce the new DOAS total ozone measurements at Ny-Ålesund at Yellow River Station. Then these measurements are used to investigate Arctic ozone loss in 2020. If the authors agree, then this point should come across much clearer in the manuscript. And the new DOAS instrument needs to be better described in the manuscript. Given the fact that so much has been published on the Arctic winter 2019/2020 already (see also below), it might be more appropriate to present this work in ACP as a Measurement Report. Further, the authors need to understand the background of the science they are reporting better. Some examples in detail and suggestions for improvement are given below. But as an obvious example: the authors report (on some occasions) the NAT temperature as −195 K – there are no negative values if temperature in measured in K. Overall, I think that the manuscript contains publishable material but I am afraid that restructuring and rewriting large parts of the manuscript are necessary.

Author's Response:

We would like to thank the reviewer #1 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.

We agreed to present this work as a Measurement Report, in which the measurements are reported and the consistency with other studies and measurements are shown. We focused on introducing the new DOAS total ozone measurements at Ny-Ålesund at Yellow River Station and then used these measurements to study the Arctic ozone loss in 2020. In addition, the new DOAS instrument was further described in the revised manuscript. Please see P6 lines 135–145. The temperature threshold for the existence of NAT as 195K has been revised.

"The ZSL-DOAS instrument mainly includes the prism, telescope, computer, filter, motor, and CCD spectrometer. The motor controlled the telescope that can change the angle of elevation between the horizon and the zenith. As the angle of elevation changes, the telescope can acquire scattered sunlight at different angles (2°, 3°, 4°, 6°, 8°, 10°, 15°, 30°, and 90°). The quartz fibre can transform the incident light and its numerical aperture is 0.22. The light is received by the spectrometer (Ocean Optics MAYA pro) and measured by a 2048 pixels CCD. This spectrometer was designed for wavelengths between 290 and 429 nm, and had the spectral resolution (FWHM) of 0.5 nm. The integration time varied between 100 and 2000 ms due to the light intensity. The detector operates normally at approximately 20°C with a thermal controller. The mercury lamp spectra, offsets and dark currents were calibrated ahead of the experiments. The ZSL-DOAS instrument can detect O_3, NO_2, OClO, BrO, and O_4. The ozone slant column density (SCD) was retrieved, with the raw data obtained in the zenith direction (90°). The ZSL-DOAS instrument was placed at the Yellow River Station (78.92° N, 11.93° E)

in the Arctic. Figure 1 shows the ZSL-DOAS instrument and experimental location, in Ny-Ålesund, Svalbard, Norway."

Comments

A) What are the main messages of the paper?

First: the paper states that ozone VCD from a ground-based instrument, the GOME-2 satellite, and the Brewer and SAOZ instruments agree rather well. However, this is not a very new conclusion and had been discussed in many (mostly more technically oriented) papers before (e.g., Léon-Luis et al., 2018; Fioletov et al., 2002; Fioletov, 2002; Fioletov et al., 2005; Weber et al., 2005, and references therein). Second, the paper reports that substantial ozone depletion occurred in the Arctic vortex until mid-April 2020, consistent with changes in simulated HNO3. Again this is today not very new information; there is a special issue in JGR/GRL (and some of the papers on the Arctic winter 2020 in this special issue are cited/discussed in this manuscript) but there are a few more papers on Arctic ozone in 2020 in the meantime (e.g., von der Gathen et al., 2021; Kuttippurath et al., 2021; Ardra et al., 2022). Third, ozone and temperature profiles were simulated by SD-WACCM, with these simulations corresponding well with ozonesonde measurements (but how well? – see below). The study used SD-WACCM with meteorological parameters driven by Modern Era Retrospective-Analysis for Research and Applications version 2 data; thus the simulation of temperature profiles by SD-WACCM is expected – isn't it? The fact that the ozone sonde measurements can be reproduced by the model is good but should be stated more clearly and in particular more quantitatively. Finally the paper closes with the statement that "observations of ozone VCDs over Ny-Ålesund will continue in order to monitor future ozone changes over the area." This is very good of course but not a conclusion from this paper.

Author's Response:

Thanks for the reviewer's advices. Ozone VCDs from a ground-based instrument, the GOME-2 satellite, and the Brewer and SAOZ instruments agree rather well and substantial ozone depletion occurred in the Arctic vortex until mid-April 2020, consistent with changes in simulated HNO_3. The reviewer is correctly saying that these are not very new information today. Thus, 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.

The simulation of temperature profiles by SD-WACCM indeed corresponded well with ozonesonde measurements, and this can be used to validate the simulation. The temporal resolution of the sounding data from March 25 to April 13, 2020, is once per day, whereas the others are normally once per 3 d during the spring and once per week during the other seasons. The other missing days were obtained by interpolation, so we did not show a plot of the differences (observations minus model).

The sentence has been revised. Please see P17 lines 408–414.

"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."

B) WACCM

B1) Some results of the paper rely on the model WACCM. But it is not clear how these results are obtained. I presume (although this is not stated in the paper) that openly available WACCM results have been used. If this is the case it should be clearly stated. If not, the WACCM runs conducted by the authors should be described (see also details) and then the WACCM version used should be clear.

Author's Response:

Thanks for the reviewer's suggestion. We have rewritten the description of model setting in section 2.2 of the revised manuscript. Please see P8 and P9, lines 186–222.

"The physical parameterizations employed in the Community Atmosphere Model Version 4 (CAM4) were applied to the WACCM (Neale et al., 2013). At present, the WACCM model is incorporated into a component set of the Community Earth System Model, whose source code is available online (https://svn-ccsm-release.cgd.ucar.edu/model versions/). The Model for Ozone and Related Chemical Tracers, version 3 (MOZART-3) provided the chemical parameters for the WACCM (Kinnison et al., 2007). This mechanism contains 52 neutral species, one invariant (N_2), 127 neutral gas-phase reactions, 48 neutral photolytic reactions, and 17 heterogeneous reactions [see Tables 5.1-5.5 in Neale et al. (2013)]. 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 (T_{sat}), 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). The sedimentation of HNO_3 in NAT aerosol follows the approach in Considine et al. (2000). The flux (F) of HNO_3 can be derived as follows:

$$F = V \cdot C \cdot \exp(8\ln^2\sigma). \quad (4)$$

here V represents the terminal velocity of NAT aerosol, C denotes the condensed-phase concentration of HNO_3, σ=1.6 (Dye et al., 1992) represents the width of the lognormal size distribution for NAT.

We used the SD-WACCM with meteorological parameters driven by Modern Era Retrospective-Analysis for Research and Applications version 2 (MERRA-2) data (Gelaro et al., 2017). The SD-WACCM had the horizontal resolution of 1.9° × 2.5° (lat × lon). The model was divided vertically into 88 layers, covering an altitude of ~140 km from the ground to the bottom of the lower thermosphere layer. 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). In this study, the simulation is initiated between November 1, 2019, and July 1, 2020."

B2) Also the way how the WACCM source code can be obtained should then be documented. Further, section 2.3 cites Kunz et al. (2011) – this is a good paper, but the paper does not deal with MERRA 2, so this sentence is confusing.

Author's Response:

The WACCM is a component set of CESM. And the CESM code is available online (https://svn-ccsm-release.cgd.ucar.edu/model_versions/). Similar sentences have been mentioned in P8 lines 187–188 of the revised manuscript.

We have rewritten the description of the nudging method used in SD-WACCM. Please see P9 lines 215–219.

" 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."

B3) Further, which chemical scheme has been used in these simulations? I assume the most recent JPL recommendation (Burkholder et al., 2019).

Author's Response:

The basic chemistry mechanism in the WACCM is taken from the MOZART-3. Please see P8 lines 188–190.

"The Model for Ozone and Related Chemical Tracers, version 3 (MOZART-3) provided the chemical parameters for the WACCM (Kinnison et al., 2007)."

B4) Müller et al. (1994, cited) emphasize the importance of CH_3O_2 + ClO for Arctic ozone loss – is this reaction taken into account in the WACCM simulation?

Author's Response:

This reaction is not included in the WACCM model. Müller et al. (1994) found that the CH_3O_2+ClO reaction is also important but is not included in the current chemical mechanism of the model and could be taken into account in future models to improve simulation performance. Please see P14, lines 325–327.

"On the other hand, Müller et al. (1994) found that the CH_3O_2+ClO reaction is also important but is not included in the current chemical mechanism of the model and could be taken into account in future models to improve simulation performance."

B5) More importantly, in which reference is the list of reactions described that is employed in the described chemical simulation? This information should be given in the paper.

Author's Response:

Thank for your suggestion. Please see P8, lines 190–191.

"This mechanism contains 52 neutral species, one invariant (N_2), 127 neutral gas-phase reactions, 48 neutral photolytic reactions, and 17 heterogeneous reactions [see Tables 5.1-5.5 in Neale et al. (2013)]."

B6) I also note that 'atmospheric simulations' are not mentioned in the author contribution. In general, it should be clear from the paper how the WACCM results were obtained.

Author's Response:

The WACCM simulation was conducted by Chen Pan. We have added statements in the author contribution. We also rewritten the description of the model settings. Please see section 2.2 in the revised manuscript.

C) PSCs

C1) Clearly PSCs are important to polar ozone loss. However, first, one has to discriminate between PSC 'formation' and 'existence'. For crystalline particles (NAT and ice) this is not the same thing. (see e.g. Tritscher et al 2021). Also the temperature threshold for the onset of heterogeneous chemistry is not the same thing as NAT existence (Drdla and Müller, 2012, see also Tritscher 2021;Solomon1999, cited in the paper).

Author's Response:

Thanks for the reviewer's suggestions. 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)."

C2) Further, denitrification by sedimenting NAT particles is touched upon in the paper. It is not straightforward implementing sedimentation in a model and explain the observations of large NAT particles in the atmosphere (e.g., Grooß et al., 2005; Molleker et al., 2014; Fahey et al., 2001; Tritscher et al., 2019). As simulated removal of HNO3 in the paper is mentioned, the paper should give some information how NAT sedimentation is implemented in WACCM.

Author's Response:

Thanks for the reviewer's suggestions. The sedimentation of HNO_3 in NAT aerosol follows the approach in Considine et al. (2000). The flux (F) of HNO_3 can be derived as follows:

$$F = V \cdot C \cdot \exp(8\ln^2\sigma).$$

here V represents the terminal velocity of NAT aerosol, C denotes the condensed-phase concentration of HNO_3, σ=1.6 (Dye et al., 1992) represents the width of the lognormal size distribution for NAT. Similar sentences have been mentioned in P9 lines 206–210 of the revised manuscript.

D) Ozone from sondes and simulation

D1) Figure 9 (top) shows an important comparison, namely ozone sonde measurements against simulated ozone. However I suggest not showing the region below about 10 km, which is not of interest here (it also shows basically a blue area). But I think it is important to also show a plot of the differences (observations minus model) which would reveal that the model does not very well simulate to observed ozone depletion in March between 15 and 20 km. Further questions: what is the meaning of negative ozone mixing ratios (top)?

Author's Response:

Thanks for the reviewer's advices. The region below about 10 km has been deleted. The temporal resolution of the sounding data from March 25 to April 13, 2020, is once per day, whereas the others are normally once per 3 d during the spring and once per week during the other seasons. The other missing days were obtained by interpolation, so we did not show a plot of the differences (observations minus model). The negative ozone mixing ratio occurs

because of the range of colour scale settings and it has been revised.

D2) WACCM seems to overestimate temperatures at about 25 km – is this a real effect?

Author's Response:

The SD-WACCM simulated temperatures were generally 0.6–3 K higher than the MLS temperatures between 100 and 1 hPa. For SD-WACCM, because heterogeneous chemistry is temperature-dependent, the model generally overestimated HCl and underestimated ClO in the lower stratosphere during winter, implying insufficient chlorine activation. Similar conclusions have also been reported by previous studies (Brakebusch et al., 2013; Solomon et al., 2015; Pan et al., 2018).

E) Formation of HCl

The presented WACCM results suggest that the deactivation in the Arctic in 2021 is partly caused by formation of HCl, This is the classic deactivation pathway in the Antarctic, but not in the Arctic (e.g. Crutzen et al., 1992; Douglass et al., 1995; Müller et al., 2018). The authors might want to comment on this point.

Author's Response:

Thanks for the reviewer's advices. We indeed want to comment this point. Please see P14, lines 348–352.

"Formation of HCl is considered to be the main chlorine deactivation mechanism in Antarctica (Müller et al., 2018), but not in the Arctic. HCl increases more rapidly in the Antarctic vortex in spring than in the Arctic vortex (Douglass et al., 1995). In early March 2020, in the Arctic, chlorine was deactivated as HCl and $ClONO_2$, and the PSC that permitted chlorine activation remained. Furthermore, activated chlorine compounds were mainly deactivated as $ClONO_2$ by the ClO + NO_2 reaction (Müller et al., 1994; Douglass et al., 1995)."

F) References

Several references have been cited in this review; hopefully they are helpful. The point is not that the authors should feel obliged to cite these references. However, the paper cites WMO (2014); I suggest that a more recent ozone assessment should be used in the paper (WMO, 2018). The most recent (2022) assessment has just been released (https://ozone.unep.org/science/assessment/sap) and might be helpful when revising this paper.

Author's Response:

Thanks for the reviewer's suggestion. More recent references have been cited in the revised manuscript.

G) Data availability

The data availability statement in this paper is not good. I suggest making the DOAS observations at Ny-Ålesund available for download on a server that issues a doi and where the data are permanently archived. Such links are reported for (e.g.) SAOZ but not for the DOAS

measurements presented in the paper. Further, the WACCM data need to be better described (see above). Making data available through e-mail request is no longer recommended.

Author's Response:

Thanks for the reviewer's advice. The DOAS observations at Ny-Ålesund available from https://doi.org/10.17632/jx7nkspkg7.1 and where the data are permanently archived. SAOZ data from http://saoz.obs.uvsq.fr/. The WACCM data have been further described in section 2.2 of the revised manuscript.

Details

• Title: I suggest avoiding "Research on" in the title; isn't this obvious? The title should rather reflect the fact that DOAS measurements from Ny- Ålesund are reported here.

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".

• p. 1, l. 16: why this period? (I think this is the period when measurements are available, but this should be clear from the paper).

Author's Response:

The light intensities were strong enough during this period and the measurements were available.

• p 1, l. 21: what is a "normal year" in the Arctic?

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 19–20.

"which was about 64.7±0.1% of that in the other years (2017, 2018, 2019, and 2021)"

• p. 1, l. 21: 44.3 % → here and elsewhere in the paper: add an error estimate for the ozone loss.

Author's Response:

Compared with the other years, the 2020 daily peak relative ozone difference was −44.3±0.1%. Error estimates have been added elsewhere in the revised manuscript. Please see P11, lines 268–270.

"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."

• p. 1, l. 23: here ans elsewhere: PV and ozone depletion: is this only a complicated way of

saying that there is no ozone loss outside the vortex? I think that Ny-Ålesund was located outside the vortex at about April 16 (see also Fig. 8.

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. In addition, we reviewed the literature and found that a large and strong Arctic vortex lasted from early December anomalously into the final week of April (Kuttippurath et al., 2021). As can be seen from the figure below, Ny-Ålesund was located inside the vortex on April 17.

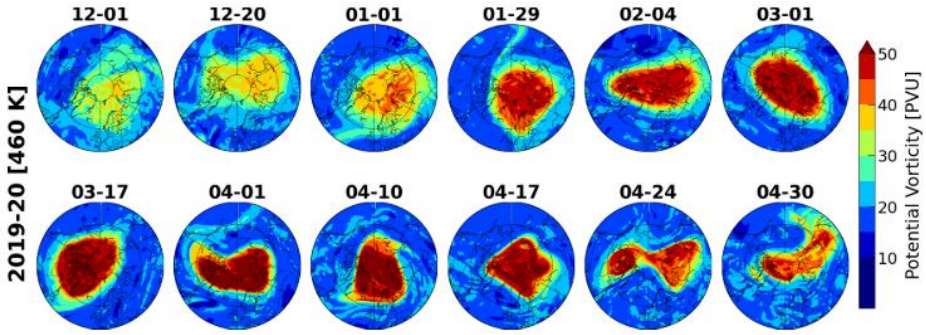

Figure 2. Polar vortex evolution in the Arctic winter/spring of 2019/2020. The evolution of polar vortex in the Arctic winter of 2020. The vortex situation in the lower-stratospheric altitude of about 460 K (~ 17 km) is illustrated. The vortex edge is calculated with respect to the Nash et al. (1996) criterion at each altitude.

Figure cited from Kuttippurath et al. (2021).

"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."

• p. 1, l. 26: how new is the peak in ClO (chlorine activation)? Compare the papers in the JGR/GRL special issue?

Author's Response:

The peak in ClO (chlorine activation) has been discussed in the JGR/GRL special issue. However, in addition to emphasizing the reliability of our observations, we also analyzed the influence of halogen chemistry processes, particularly bromine chemistry.

• p. 2, l. 38: this is not a good description of halogen induced polar ozone loss (e.g., Müller et al., 2018, and Solomon 1999, Tritscher 2021, cited in the paper).

Author's Response:

The description has been revised. Please see P2, lines 48–53.

"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).

• p 2., l. 42: 'recovery' is an important issue, it is different in the polar regions and in midlatitudes (WMO, 2018). See also further papers on the recovery of both the Antarctic ozone hole and global ozone levels (e.g., Kuttippurath and Nair, 2017; Strahan and Douglass, 2018; WMO, 2018; Bodeker and Kremser, 2021; Stone et al., 2021; Weber et al., 2022).

Author's Response:
Thanks for the reviewer's advice. The sentence has been revised. Please see P3, lines 53–56.

"As anthropogenic emissions of ozone-depleting substances have decreased 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; Dhomse et al., 2018; Kuttippurath and Nair, 2017; Strahan and Douglass, 2018)".

• p. 2, l. 49: there should be more citations here than just Hu 2020.

Author's Response:
More references have been cited. Please see P3, lines 62–64.

"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)".

• p. 3, l. 75: Simpson is on boundary layer issues: this reference needs to be changed. There are several alternative citations, already cited in the paper and there are further modelling papers cited in this review.

Author's Response:
This reference has been changed. Please see P4, lines 101–104.

"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).".

• p. 3, l. 75: These citations focus on one particular model (CLaMS), which is okay. But I think you should have citations to other models here as well (e.g., Chipperfield, 1999; Khosrawi et al., 2009; Bekki et al., 2013; Chipperfield et al., 1994; Kinnison et al., 2007; Wohltmann and

Rex, 2009; Wohltmann et al., 2010).

Author's Response:
Thanks for the reviewer's suggestion. More references have been cited. Please see P5, lines 104–107.

"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)".

• p. 4, l. 99: You cannot start the Methods section with "the DOAS instrument". Which instrument? I think it is a new instrument that is described below – correct? This should be much clearer from the paper and the instrument needs to be described first before it can be "placed" somewhere. Further, given the fact that the DOAS technique is so prominent here (or should be) a bit more background on DOAS and citations (see perhaps, Huneker et al., 2017) might be appropriate.

Author's Response:
Thanks for the reviewer's advice. The ZSL-DOAS instrument has been further described in the revised manuscript. Please see P6 lines 135–145. We have also added a bit more background on DOAS technique. Please see P4 lines 95–97.

"The ZSL-DOAS instrument mainly includes the prism, telescope, computer, filter, motor, and CCD spectrometer. The motor controlled the telescope that can change the angle of elevation between the horizon and the zenith. As the angle of elevation changes, the telescope can acquire scattered sunlight at different angles (2°, 3°, 4°, 6°, 8°, 10°, 15°, 30°, and 90°). The quartz fibre can transform the incident light and its numerical aperture is 0.22. The light is received by the spectrometer (Ocean Optics MAYA pro) and measured by a 2048 pixels CCD. This spectrometer was designed for wavelengths between 290 and 429 nm, and had the spectral resolution (FWHM) of 0.5 nm. The integration time varied between 100 and 2000 ms due to the light intensity. The detector operates normally at approximately 20°C with a thermal controller. The mercury lamp spectra, offsets and dark currents were calibrated ahead of the experiments. The ZSL-DOAS instrument can detect O_3, NO_2, OClO, BrO, and O_4. The ozone slant column density (SCD) was retrieved, with the raw data obtained in the zenith direction (90°). The ZSL-DOAS instrument was placed at the Yellow River Station (78.92° N, 11.93° E) in the Arctic. Figure 1 shows the ZSL-DOAS instrument and experimental location, in Ny-Ålesund, Svalbard, Norway."

"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)."

• p. 6, p. 140: this sentence starts with ‘parameters’ but the paper should state what was actually done regarding WACCM.

Author’s Response:
We have rewritten section 2.2. Please see response to **Comments B1**.

• p. 7, l. 166: ERA5 has 137 layers – is there a typo here?

Author’s Response:
There is not a typo here. The ERA5 data had the spatial resolution of 0.25° × 0.25° and were divided into 37 layers vertically, from 1000 hPa to 1 hPa.

• p. 7, l. 167: where have these measurements been done?

Author’s Response:
These measurements were carried out at Ny-Ålesund.

• p. 8, l. 173: what is a ‘normal year’?

Author’s Response:
This response is similar to **Details p 1, l. 21**.

• p. 9, l. 199: what is the ‘threshold temperature’? This is an important point that should be discussed in the paper.

Author’s Response:
Please see response to **Comments C1**.

• p. 9, l. 201: by definition the PV in the southern hemisphere is negative and positive in the northern hemisphere. This simple fact should be taken into account when making such statements.

Author’s Response:
Thanks for the reviewer’s advice. The sentence has been deleted.

• p. 10., l. 235: apparent → obvious?

Author’s Response:
It has been revised.

• p. 10, l. 243: ‘recover’ is problematic here, it is not the right word to use when taking about chlorine deactivation putting a halt to ozone loss.

Author’s Response:

It has been revised. Please see P15 lines 353–354.

"In mid-April 2020, $ClONO_2$ stopped increasing and ClO was almost depleted when the ozone concentration started to increase."

• p. 10., l. 237: it is not only the reaction HCl + $ClONO_2$

Author's Response:

The heterogeneous reactions HCl + $ClONO_2$ and HOCl + HCl and the gas-phase reaction CH_3O_2 + ClO contributed to the conversion of HCl to active chlorine (Müller et al., 1994; Müller et al., 2018). And the sentence has been revised. Please see P14 lines 345–346.

"Chlorine was dominantly activated by $ClONO_2$ + HCl and this reaction improved up to 10 times when the temperature reduced by 2.3 K (Wegner et al., 2012)."

• Figure 5: cold the errors of the individual measurements be used for weighting the data when calculating regression etc.?

Author's Response:

Thanks for the reviewer's suggestion. We added the errors of the individual measurements be used for weighting the data when calculating regression etc. No errors were provided from the GOME-2 dataset, so we did not consider measurement errors of GOME-2. Please see P35.

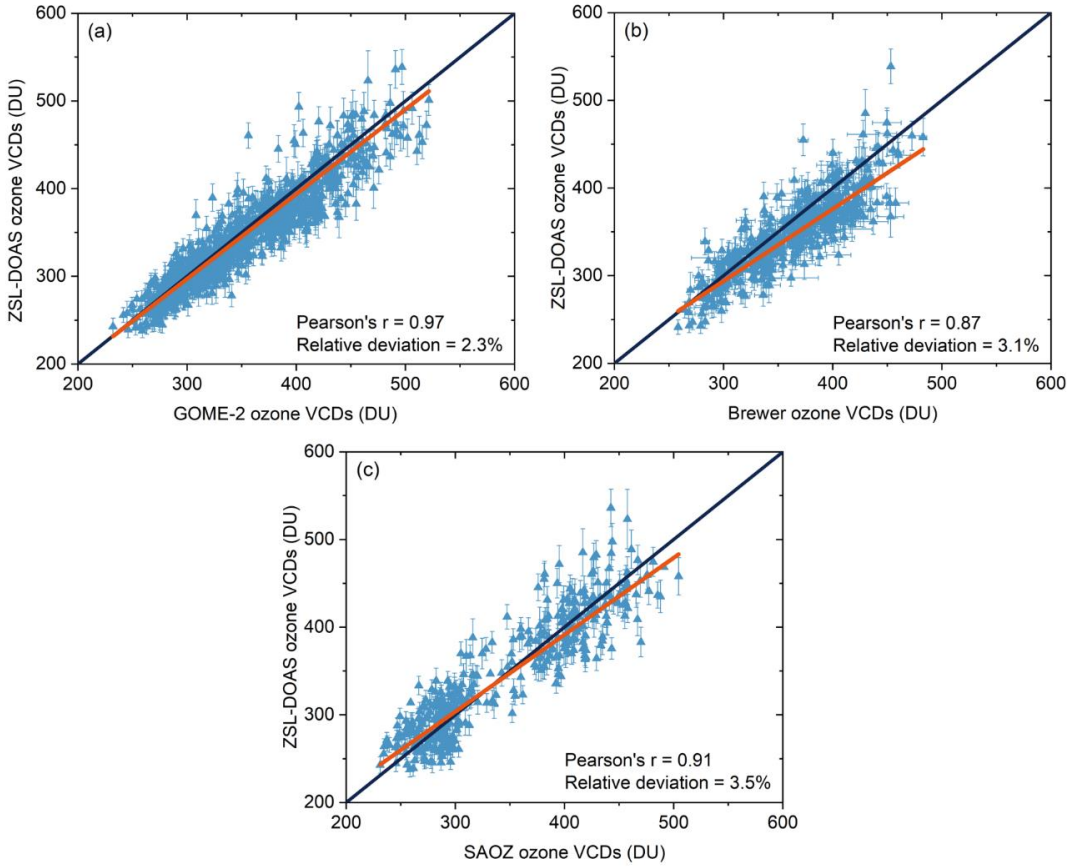

Figure 6. Scatter plots and linear fits of retrieved ozone VCDs with (a) GOME-2, (b) Brewer, and (c) SAOZ.

• Figure 6: Show error bars?

Author's Response:
The figure has been revised. Please see P34.

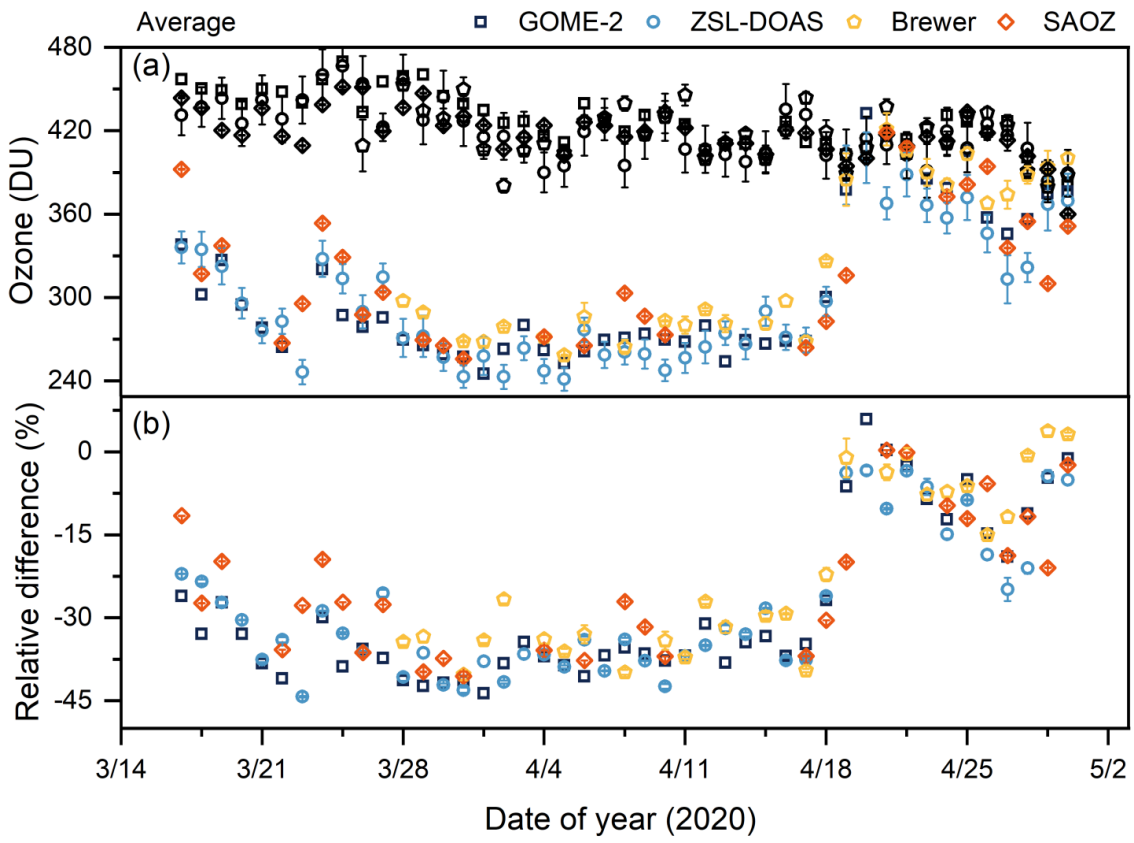

Figure 5. (a) Ozone data for 2020 and the average ozone data (black) of 2017, 2018, 2019, and 2021. (b) relative ozone difference for 2020.

• Figure 7: the blue line shows 195 K, which is an approximation for the onset temperature for heterogeneous chemistry.

Author's Response:
Thanks for the reviewer's suggestions. Please see response to **Comments C1**.

• Figure 8: show error bars?

Author's Response:
The figure has been deleted. Please see response to **Details p 1, l. 23**.