

Dear authors,

I would recommend a major revision. This manuscript has to improve significantly before I can recommend it for publication.

I have to admit that I found it difficult to make a decision for this paper. Some parts of this paper make the impression that the authors have a good knowledge of the topic. Most of these parts are scientifically sound, and show knowledge of the literature. The structure and readability of the paper are acceptable and on an average level in these parts. Language could be improved, but I acknowledge that the authors are no native speakers (neither am I). There are also numerous small errors, omissions and inconsistencies, and this manuscript could need more care and attention.

But some other parts make the impression as if the manuscript was written by several different authors, with a different level of scientific background. These parts sometimes show a lack of understanding of the stratospheric processes and fundamental flaws in the methods. In particular, section 4 is in a bad shape.

While the topic is within the scope of the journal and of interest for the audience, another problematic issue is that large parts of the results shown in this paper have already been published elsewhere in similar or identical form. In the end, I am in favor of the authors, since I think that as a reviewer my main responsibility is to make sure that the results are scientifically sound. There is nothing wrong with confirming the results of others with different data sets or models. But some parts of this manuscript make the impression that the authors only reviewed the literature and rephrased the work of others, instead of doing original research. This is ok up to some point, but it should not be taken too far to avoid any negative connotations.

Best regards, Ingo Wohltmann

## **Major comments**

### **1. Result that the contribution of gas phase nitrogen cycles to ozone loss in the polar vortex in spring is significant cannot be correct**

The results of the noPSCaer CTM run for the contribution of gas phase nitrogen cycles to ozone loss in the polar vortex cannot be correct (section 4, e.g. your Figures 12-15 and accompanying text). It is basic textbook knowledge (if not to say the most important scientific result of research related to the ozone layer of the last decades) that chemical ozone loss in spring in the polar vortices is dominated by loss from catalytic chlorine and bromine cycles and caused by heterogeneous chemistry. Your results imply that NO<sub>x</sub> cycles contribute not only significantly to column ozone loss in the vortex in spring, but even dominate the ozone loss. If that would be true, it would mean that the Montreal Protocol and the ban of CFCs would have been unnecessary and that we should have seen large ozone loss before the rise of CFCs in the 1960s or 1970s.

Unfortunately, there is no sufficient information to figure out what is going wrong here. There is either a bug in your program code (in the CTM or in your scripts that produce the figures), or there are some fundamental flaws in your methods.

There are some candidates:

- a) Not considering that NO<sub>y</sub> and NO<sub>x</sub> are not the same in the noPSCaer run without PSCs and the full run with PSCs (because PSCs change NO<sub>y</sub> and NO<sub>x</sub>, e.g. by denitrification or changes in partitioning).
- b) A problem with the initialization of the noCHEMall run that you use as a passive ozone tracer. It may be that you initialize the passive ozone tracer too early and get too much NO<sub>x</sub> chemistry from outside of the vortex or in early winter or late autumn into your estimates.

c) A problem with the upper model boundary, which may be too low to obtain meaningful values with the passive ozone tracer.

d) Using a vertical range for column integration which exceeds the range where the passive ozone column method gives reliable results (relates e.g. to c) or the ozone lifetimes getting shorter with increasing height).

I will go into more detail in the specific comments when necessary.

I would recommend either to solve this issue or, if it is not possible to find out what is going wrong here, to remove all discussion of nitrogen cycles and the noPSCaer run, and to delete the corresponding results from the figures.

There is plenty of literature showing how the results should look like. I will give you only one example. Figure 20 of Wohltmann et al. (2017) shows that more than 95% of ozone loss at about 50 hPa in the vortex is caused by the catalytic ClO-ClO and ClO-BrO cycles (for one Arctic and one Antarctic winter). You will find more figures for different altitudes and years in the supplement of this study, showing that this is a general statement.

For specific comments of relevance to this major comment, see e.g. comments to line 24-25, 184, 194-195, 197.

## 2. Not sampling the polar vortex homogeneously when determining ozone loss

In the method to determine ozone loss that you introduce in sections 2.2 and 3.2: For no apparent reason, you start trajectories only on the 85 degrees latitude circle instead of sampling the vortex homogeneously (which would have been easy). This introduces a bias, which you could have easily avoided. Please see the specific comment to lines 332-333 for details. Please repeat the runs.

## 3. Low novelty value and low scientific significance, redundancy, many repetitions of results from other studies, and the paper needs to be shortened considerably

You are probably aware that many results which are similar or even identical to what is shown in your manuscript have already been published elsewhere (e.g. Manney et al., 2020, Lawrence et al., 2020, Wohltmann et al., 2020, Dameris et al., 2021). In my opinion, the main responsibilities of a reviewer is to make sure that the results are scientifically sound and that the paper is well structured and readable, and I will concentrate on these aspects in my review. It does not hurt when a paper confirms results of other studies. But I think that this manuscript can be considerably shortened, in particular when you only repeat results from other studies and do no original research on your own.

I find section 3.1 particularly problematic (see also specific comments). There are at least 5 Figures which have already been published in identical or very similar form in other manuscripts (where 4 figures alone are from Lawrence et al., 2020). Large parts of the text only repeat findings from other studies. Please shorten this section considerably and concentrate on your original research (that would be mainly the Plumb fluxes).

Section 4 is equally problematic. It could be shortened considerably. And it suffers from the fundamental flaws that I discuss in major comment 1.

The introduction is almost 3 pages and it is definitely too long. It is a little bit tedious to read. Try to concentrate on what is important for your study. Not all details are important enough to mention them here, e.g. the UV radiation in April in Moscow or the Indian Ocean Dipole. Your introduction would be very fine for a review paper and is a nice review on the literature for the winter 2019/2020, but this is maybe not the right place for it.

## 4. Language

This manuscript would benefit from the help of a native speaker. You have to improve on the use of the English language. Note that I am well aware that the authors are no native speakers and I won't base my judgement on this manuscript on the use of language by the authors, because this would not be fair. But it certainly will help to make the manuscript more readable and also to remove some confusion or misunderstandings. See also similar comment of reviewer #3 in the pre-review.

#### 5. Numerous small errors, omissions and inconsistencies

There are numerous small errors, omissions and inconsistencies in the manuscript. While this can happen to everyone, I think that this manuscript would have needed some more care and attention and this would not have been necessary.

E.g., a figure is missing (Figure 2b), section numbering is not correct in one place (two sections 2.3), at least one paper in the references is not cited (Madrid et al.), there seems to miss a sentence in the author contributions, and so on.

#### 6. Make sure that the title, abstract and conclusions reflect what you say in the main text:

When reading the main text, I discovered that you put quite an emphasis on the troposphere. This is totally fine, but it isn't really reflected in e.g. the title or abstract of the paper. Also, if you would like to put an emphasis on the situation in Russia, this is fine, but then it should be more clearly stated in abstract, title and introduction. Make sure that you don't disappoint the reader who has read the title and abstract and then does not really find what he expects in the main text.

### **Specific comments**

#### Line 9-10:

State in the abstract which CTM you are using (i.e. the RSHU CTM). This will be of interest for many readers.

#### Lines 21-22:

The phrasing is a little bit misleading, since people often think immediately of "chemical ozone loss" when they read "ozone loss". Suggestion: "...indicated that both dynamical and chemical processes make contributions to ozone changes inside the polar vortex"

Lines 22-23: *"In this case, dynamical processes predominate in the western hemisphere, while in the eastern hemisphere chemical processes make an almost equal contribution with dynamical factors"*

I don't think that this is correct (at least in the way that it is phrased here, it is misleading). The simple reason for this is that the polar vortex is moving. Air masses that are in the western hemisphere at a particular date will be somewhere else a few days later. You have to make sure that you phrase that carefully. Ideally, one would follow the air masses inside the vortex and make statements for the vortex as a whole. That being said, it is of course a valid approach to look at specific locations, but then, for a single location, fast dynamical changes will often dominate. This is probably a question of what you use as a reference frame when you define the chemical and dynamical change.

Lines 24-25: *"the chemical depletion of ozone is determined not only by heterogeneous processes on the surface of the polar stratospheric clouds, but by the gas-phase destruction in nitrogen catalytic cycles as well."*

This would only be correct as a very general statement. But from the context is clear that you refer to ozone loss in spring in the polar vortex here and that you think that the contribution from NO<sub>x</sub> cycles is significant. The sentence is misleading and not correct in the end. You have to be careful about the message that you convey here. There is general agreement that chemical depletion in spring in the polar vortices is dominated by heterogeneous processes, see major comment 1. Delete this sentence.

Line 28:

Would be nice if you would not only cite references for the tropospheric influence, but also for the statement *“the circulation of the Arctic stratosphere in the winter-spring season (hereinafter winter season) is characterized by strong interannual and seasonal variability”*. Suggestions: e.g. Tegtmeier et al., 2008, Solomon, 1999.

Line 30:

Pedatella et al. is a news article and not a peer-reviewed paper. Delete the reference.

Line 38-39: *“the largest decrease in the Arctic ozone was observed...”*

This statement needs some references. For the 2019/2020 winter, e.g. Manney et al., 2020, Wohltmann et al., 2020. For 2015/2016, e.g., Khosrawi et al., 2017.

Line 62: *“record low temperatures were observed in the Arctic lower stratosphere, and, as a result, a record volume of Polar Stratospheric Clouds (PSCs) was expected”*

Again, this statement needs some references, e.g. Lawrence et al., 2020 or Wohltmann et al., 2020. And you surely not mean “was expected” but “was observed”.

Lines 115-116: *“Signs of recovery in ozone levels began to be noted in the polar regions, in particular, a decrease in the depth of the ozone hole and its size in Antarctica. The 2019 ozone hole in Antarctica was one of the lowest in decades”*.

These statements need some references.

Line 138:

The canonical reference for the ERA5 dataset is Hersbach et al., 2020. Your reference is outdated and not peer-reviewed.

Line 136-139:

It makes the impression to me that some of the meteorological analyses that you mention here are never used in the paper. Please do only mention the analyses that are actually used. In addition, please make sure that you give the analysis that you use to calculate quantities or that you discuss in the text at the appropriate places in the paper. This information is missing in several places.

Line 141-142:

State the analysis you used for the temperatures.

Line 159:

Why forward and not backward trajectories? Backward trajectories would probably have the advantage that you would lose less trajectories that leave the vortex. In addition, forward trajectories the disadvantage that at the end of the run, trajectories started at the same potential temperature level will be distributed over a range of potential temperatures. Since you would like to know the ozone loss at the end of the run at a certain level, this is unfortunate.

Section 2.2:

Important information is missing or only given later in section 3.2.

What are the initial locations of the trajectories? What is the start date? These two questions are answered in section 3.2, but you should either move this from section 3.2 to section 2.2. Or vice versa (you could delete this section then)

Did you use vertical winds or heating rates to calculate the vertical motion? What happens with trajectories that leave the vortex? What is your criterion for the vortex edge (some PV value)? These questions are not answered here or in 3.2.

It would also be important to have some more information about the ERA5 ozone product, since your results crucially depend on it. E.g. which measurement data are assimilated, how well does the ERA5 ozone product compare to observations?

#### Section 2.3:

Important information is missing, that is only given later in section 3.2. Please state the individual stations. What is the time period you are looking at and the starting date you use as the reference? What is your criterion for the vortex edge?

Again, you could move this information from 3.2 to 2.3. Or vice versa (and delete 2.3)

#### Line 172:

State the name of the radiative transfer model. Just “the radiative transfer model” is no sufficient information.

#### Line 170-172:

It would be appropriate to go a little bit more into detail here (even though this is an established method). So, you probably first interpolated all the ozone sonde measurements on an isentropic surface. When you fit the linear regression line to the timeseries, you probably don't use all of the ozone data but a time window around some given date to be able to obtain a value for the rate of change for a specific date, I suppose? What is the length of the time window?

#### Line 175:

I wasn't able to find the reference Tsvetkova et al. (2004) at the home page of the journal. Please replace the reference by something easier to access. I think there are many articles describing the method.

#### Section 2.4 (misabeled as a second Section 2.3 in the manuscript):

Again, important information is missing. When did you start the model run? See also comment to lines 194-195.

#### Line 180:

Smyshlyaev et al., 2017 is only available in Russian language (at least when I access it through the doi). This is very unfortunate, because I can't read it. I don't know what the guidelines of ACP are regarding this, but possibly you have to remove this reference.

#### Line 183:

5 degrees x 4 degrees is an extremely coarse resolution and is not state-of-the-art anymore (maybe it was 15 years ago). Is it possible to run the model in a higher resolution (say 2 degrees x 2 degrees or 1 degree x 1 degree) to exclude that the coarse resolution has any negative effects on the results?

#### Line 184:

You should make sure that you don't look at air masses for your “passive ozone tracer” from the noCHEMAll run that were initially (at the start of the model run) at the upper model boundary or above, because this leads to meaningless values for the “passive ozone”. From model runs that I performed for 2019/2020 with an upper boundary at 50 km, I estimated that values for the ozone loss above 550 K are not reliable anymore with the “passive ozone tracer” method.

Line 186-187:

It is a rather unusual choice for the PSC scheme that it is based only on STS clouds. Can you elaborate a little bit on the reasons for that (not only in the reply, but also in the manuscript)? Why don't you simulate NAT and ice clouds in addition? Note that I am aware that the addition of NAT and ice clouds would probably only have a small effect on your results, since the heterogeneous reactions are usually sufficiently fast and since the temperature dependence is similar for NAT and STS clouds. But you possibly introduce some uncertainty by this, this should be discussed.

Lines 194-195:

This is not quite clear to me. Do you want to say that you initialize your "passive ozone tracer" from the noCHEMall run for estimating ozone loss on 1 November? But only north of 64 degrees? What are you doing in the following days? Calculating chemistry south of 64 degrees and then switching chemistry off when air masses are transported inside the 64 degree latitude circle?

This is of particular importance for your method to determine ozone loss. You set the reference date here for your determination of ozone loss. Ozone loss is extremely sensitive to the start date for the passive ozone tracer. If you choose a date too early, you will get a significant contribution from ozone loss from outside of the vortex caused by NO<sub>x</sub> cycles (since the air masses that are inside the vortex and the end of the model run would have been far outside the vortex at the start of the model run). Or you get loss from NO<sub>x</sub> cycles in autumn before the formation of the vortex, when there still is sunlight. And if you set the start date too late, you miss some ozone loss. In my experience, a date between 15 December and 1 January works best for 2019/2020. November 1 is probably much too early.

And please state clearly what you are doing here. I.e., setting the reference date for calculating ozone loss. This is probably not clear to the majority of readers.

If you switch off chemistry only north of 64 degrees, that makes it hard to reason about the results. This way, your passive ozone tracer will not really be "passive", but some mixture of air masses that did experience ozone loss and air masses that did not. This makes it hard to understand what is actually shown in Figs. 12-15. Certainly not just "the" ozone loss.

Maybe this method would have worked if you would have chosen a PV contour at the edge of the vortex instead of 64 degrees. But it still would be a problem that there is probably ozone loss by NO<sub>x</sub> cycles in November at high latitudes, since there is still sunlight.

But in any case, this would need much more explanation. The reasoning behind your setup is not at all obvious to the reader. I think I figured it out after some time (you want only to count ozone loss in the vortex and get rid of the loss by NO<sub>x</sub> cycles outside of the vortex by switching on chemistry south of 64 degrees), but that does not get clear at all. And probably, 64 degrees as a boundary will not work because this always includes air from outside the vortex.

Line 197: *"Comparison of the baseline scenario with these two additional scenarios makes it possible to estimate the periods when the chemical destruction of ozone is most effective after heterogeneous activation on the PSC surface, and when the gas-phase destruction of ozone in nitrogen catalytic cycles is more significant."*

You are very probably not doing this correctly or as you intended it. Switching off heterogeneous chemistry on PSCs (or not forming PSCs at all in the model) will also affect the partitioning and chemistry of NO<sub>y</sub> and NO<sub>x</sub>, and it will affect denitrification. E.g., the heterogeneous reaction N<sub>2</sub>O<sub>5</sub>+H<sub>2</sub>O will be important for the partitioning. And effectively switching off denitrification will lead to higher NO<sub>x</sub> in the "noPSCaer" run, which could lead to more ozone depletion by NO<sub>x</sub> compared to the reference run. This way, you will obtain a different result as if you have kept NO<sub>x</sub> constant.

What is not quite clear to me: Do you also switch off chemistry on the binary background aerosol at higher temperatures (which would also lead to unwanted changes)?

It is very likely that you will not obtain the result that you are hoped for: A clean separation of the amount of ozone that is depleted by chlorine and bromine cycles from the amount of ozone that is depleted by NO<sub>x</sub> cycles.

Therefore alone, I would recommend to delete all of the discussion on the contribution of the nitrogen cycles to the chemical ozone loss. I don't think that results are reliable.

A clean way to do this correctly would be to keep track of it in the chemistry module of your model, by e.g. looking at the rates of the rate-limiting steps of the different catalytic ozone destruction cycles. A study that shows how to do this correctly and that shows that heterogenous chemistry dominates ozone loss is Wohltmann et al., 2017.

Lines 207-210 *“On the other hand, in the absence of the Sun, the chemical destruction of ozone does not yet reach high values associated with the previous halogen activation on the surface of polar stratospheric clouds, therefore, it can be assumed, that there should not be extremely low values of TCO in the region of absence of observations by the OMI instrument.”*

Delete this sentence. This is not correct.

It is well known that ozone values inside the vortex are often relatively homogeneous. It is also well known that the movement of the vortex and the movement of air inside the vortex cause a homogenization. Air is often processed by the PSCs like in a “flow processor”, and air masses are transported into the sunlit regions of the vortex and move back into the dark regions again. Of course, there are exceptions, and air masses may sometimes remain in darkness for a long time. But what you are doing here is pure speculation.

And you don't even need to speculate. There are measurements from e.g. the MLS instrument which show the regions that OMI can't measure (and which already have been used for studies of this winter). Why don't you base your discussion on these measurements?

Lines 214-215: *“Again, the minimum values are detected along the border of the region of absence of observations - the zone of polar night.”*

See above. Delete this statement.

Lines 202-220:

While I won't judge your paper by relevance, I wonder whether this detailed description of the position and movement of the polar vortex is really necessary. I don't really see the scientific significance of these results. In addition, this can easily be deduced from Figure 1. Furthermore, the development of the vortex has been described elsewhere in studies that already have been published, e.g. some figures in Manney et al., 2020, and more importantly, Dameris et al., 2021. Their figure 1 has a large similarity to your Figure 1.

Line 227: *“The winter season 2019-2020 in the Arctic stratosphere was one of the coldest in the last 40 years.”*

Give references for this statement (e.g. Wohltmann et al., 2020, Lawrence et al., 2020)

Line 229:

Would be better to speak of STS and NAT clouds and not of Type I clouds for clarity.

Line 232:

Figure 2b is missing.

Lines 245-246: *“The first period (February 7 - March 7) corresponds to strongest weakening of wave activity propagation in 2020, the second: January - February, the third: January - March.”*

Sorry, but I have no clue what you want to say to me here. Is a part of the sentence missing?

Figure 3:

I would find it helpful to have some kind of colorbar for panels (a) and (c). Or at least to have the unit directly in the plots. It is given in the caption, but it is a little bit hard to bring this together.

Line 258: *“Where the absolute maximum of the average temperature in February was reached.”*

Sorry, but again, I can't follow you. Do you mean that the highest temperatures over the course of the year were reached in February in Siberia. Or does the maximum refer to location? Please clarify.

Line 260:

Do you mean “obtained” and not “retained”? Or what do you want to say?

Large part of section 3.1 (Lines 221-end and Figures 2-4):

Without going through Lawrence et al. (2020) in detail, I have the impression that almost all of your section 3.1 only repeats what has already been written in the text and shown in the figures in Lawrence et al. Only for example: Your Figure 2 is Figure 11a from Lawrence et al., your Figure 3a is similar in meaning to Figure 7f from Lawrence et al., your Figure 3c is Figure 6a from Lawrence et al., your Figure 4a is Figure 3a from Lawrence et al. While I think this was probably not intentional, I think this is problematic. You should really think about shortening this section, to delete some of the figures and to refer to Lawrence et al. where appropriate.

Section 3.1: Meteorological data:

What is the reanalysis data that you are using here for the temperatures and the EP fluxes? You only make a very general statement in section 2 that you use ERA5, NCEP, JRA and MERRA, and don't state anything in section 2.1. It makes the impression that you use a particular reanalysis here. Which one?

Line 267: *“and the negative one, on the contrary”*.

Sorry, I can't follow you here. What do you want to say? That the negative phase shows opposing changes? I don't think you need to state that. This is obvious and follows from the definition of the AO.

Line 267-270: *“With a positive AO phase, a stronger western zonal transport leads to milder winters, but with more precipitation in Southern Europe. In the negative AO phase, this transfer is weaker; as a result, cold air masses from the Arctic spread more strongly to the territory of Europe.”*

It seems to me that this needs a reference.

Line 271: *“AO is the result of interaction between the dynamics of the stratosphere and the troposphere.”*

I don't know if I would phrase it like this. I would suggest to write something like “Interaction between the dynamics of the stratosphere and the troposphere can cause changes in the AO” or that the changes in the stratosphere (polar vortex strength) and troposphere (AO) are closely correlated.

Line 273-274: *“which is facilitated by an increase in the temperature gradient between the heated by the sun and shaded parts of the atmosphere”*

I have no idea what you want to say here. Do you mean “associated” again and not “facilitated”? That would make more sense. But why do you mention the sunlit and dark parts of the atmosphere in



conjunction with the temperature gradient? Assuming that you are talking about the zonal temperature gradient in the stratosphere, of course the polar vortex is in the end caused by radiative cooling in the polar night. But I think you are talking about interannual or seasonal changes in the temperature gradient related to polar vortex strength, which are not caused by changes in solar illumination (which is the same in every year).

What do you mean by “zonal mean flux”? Flux of which quantity?

Lines 275-276:

What is cause and effect here? Less wave activity means a stronger and undisturbed vortex, and that in turn means altered conditions for wave propagation. Again, probably better to speak of correlation or association.

Line 284:

Up to here, you talk of the AO. Now, you suddenly start to use the term NAM, which is just another word for the AO. And instead of talking of an AO index value which is just a number like 4 in the paragraph before, you start giving the value as 1.5 sigma. But probably I am not wrong when I suppose that these indices measure the same quantity. And what is sigma? The standard deviation of the AO time series, I suppose?

Line 284-285:

I have no idea what you mean by “spread continuously”. Do you mean “propagate downward” or “extend into the troposphere”? Or that there is a clear signal in this time period?

Line 286:

At least in the stratosphere, the plot shows high values of the AO index up to the end of April.

Line 286:

You suddenly talk of a SSW in March which you never have mentioned before. It would be helpful to introduce the warming before you refer to it. It would also be helpful to have a reference or some more explanation. As far as I can see from e.g. PV maps, the vortex was quite stable until the end of April (although there was warming at the end of March).

**(Note: Lines 288-322 are not really my area of expertise. I hope another reviewer can say more to this. I cannot judge whether the results are scientifically sound or not)**

Line 290-292: *“It is known that the main source of wave activity propagation into the stratosphere, characterized by the maximum of the vertical component  $F_z$  of Plumb's fluxes, (e.g. Jadin 2011) is located over this region [north-Eastern Eurasia].”*

This is not really my area of expertise and this may be correct. But this is a rather bold statement, and I have never heard of this before. Unfortunately, the only reference that you give is from a predatory journal, and therefore, is no reliable source and I refuse to read it. Please give references from legitimate peer-reviewed sources or delete this statement.

In any case, remove the reference.

Lines 294-295:

Since you already stated this, you should refer to your earlier statements. E.g. “As shown above...”

Figure 4a:

The green contour mentioned in the caption is missing from the plot. The units for the colorbar are not given (neither in the plot nor in the caption).

Figure 4b and c, Figure 5a and b:

Same comment as to Figure 3. Would be helpful to have the units for the contours directly in the plots.

Lines 306-307:

You are again talking about a SSW event which you never have introduced before.

Lines 331-339:

Seems like part of the description of your method is in these lines and the other part is in section 2.2. Can you please describe the method only at a single place? Either, you have to move these lines to the description of the method in section 2.2. Or vice versa (and then delete section 2.2).

Some of the questions I had for section 2.2 are answered here, but others are not (see also following comments and comment to section 2.2)

Lines 332-333: *“For simplicity the trajectories were initiated uniformly distributed on the 85 N latitude circle, when it was completely located inside the polar vortex”*

For no apparent reason, you start trajectories only on the 85 degrees latitude circle instead of sampling the vortex homogeneously, which would have been easy. This introduces a bias which you could easily have avoided. Please repeat your calculations with trajectories that sample the vortex homogeneously.

You need to test whether a trajectory is inside the vortex in any case, so that should not introduce any additional effort. Unfortunately, you don't give any information on your criterion for testing whether a trajectory is located inside the polar vortex. Do you use a fixed PV contour (say 36 PVU), equivalent latitude or the edge as defined by Nash?

It should also be easy to sample the vortex homogeneously, e.g. by starting trajectories on more than one latitude circle and starting less trajectories on latitude circles closer to the pole to make sure that every trajectory represents the same area, or by using a random generator to distribute points evenly (the only thing you have to take care of is taking the arcsin of the random number for latitude).

Line 335: *“and mostly remained inside the vortex”*

You give no information what you do with trajectories that leave the vortex. Do you ignore that or do you sort them out? This is potentially important for the results.

Line 332, 339:

You start the trajectories only on two levels, 475 K and 550 K. I think that would have been fine if you would have used backward trajectories, but it is unfortunate with forward trajectories. The quantity that you usually would like to know is the ozone loss at some level in spring at the end of your trajectory run (i.e. in a given well defined air mass). The trajectories which you start at 475 K or 550 K will not only descend, but will also cover a range of potential temperatures (and horizontal locations) at the end of the run, which makes the ozone loss hard to interpret.

The very least you could do is to give the range of potential temperatures that is covered by the trajectories at the end of the run (and the mean value to estimate the diabatic descent).

It would however be much more straightforward to base your method on backward trajectories.

Line 338: *“average ozone value”*

I assume you mean the average over all trajectory locations at this date?

Line 355-356:

I would expect this information earlier in the description of the method.

Line 358: “overestimation”.

You don't know whether this is an overestimation (compared to “reality”) or not. I would say “leads to higher estimates for...”

Line 367-368:

Please give the exact dates. Does beginning of January mean January 1? What is end of March?

Line 369: “Significant ozone loss had been seen from the mid-January till the end of March between 400-525 K isentropic levels (~15-22 km).”

Where does the information mid-January until end of March come from? Is this your result from your method? This cannot be deduced from the figure, so you should state that more clearly. E.g. “Analysis of the ozone sonde data shows that significant ozone loss is observed from mid-January to end of March between...”

Lines 377-378:

Are these values your results or the results of Peters (2010)?

Line 382-384: “The use of temperature, wind speeds, surface pressure and air humidity from the reanalysis data made it possible to simulate the effect of atmospheric circulation on the transport of ozone and associated gases close to reality. Variability of specified dynamical parameters determines the dynamical decrease in ozone content, as well as the atmospheric temperature govern the rate of chemical reactions, polar stratospheric clouds formation and the rate of heterogeneous reactions on their surface, which determine the chemical destruction of ozone.”.

Delete these sentences. They have no information content and are phrased awkwardly. Everybody in the community knows what a CTM is good for. The information given here is much too basic.

Figure 9:

There is a strange zig-zag pattern in some of the contours in the plots. It seems that there is a bug in your plotting or interpolation functions.

Figure 9:

Would be nice again to have the units in the plots.

Figure 9:

Please indicate the vortex edge in the plots.

Line 390: “First, a basic numerical experiment was performed taking into account all factors affecting Arctic ozone”.

I think what you wanted to say here is: “First, a reference run with full chemistry was performed.” Your sentence sounds odd.

Line 390-391: “The variability of the atmospheric gas composition during the winter of 2019-2020 was calculated in the basic numerical experiment”

Again, you state the obvious. Delete the sentence.

Line 395-396: “In particular, its movement in the eastward direction and the minimum values at the beginning of March in the Northern part of the European territory of Russia are reproduced”.

Well, if you would not reproduce these very basic features, you would have a problem anyway. I don't know whether this sentence is really needed.

Line 396-397: *“In mid-March – in the western part of the Arctic, in the area of Greenland, Svalbard and Franz Josef Land in early April and North to the mainland of the ETR in mid-April.”*

This is not a complete sentence, and therefore, unintelligible. Delete. And if it would be a complete sentence, I have the feeling that the information given here would be irrelevant. You don't need to state the position of the vortex every few days and every location that it covers. This is not only easily visible in the plots, but also no relevant or scientifically interesting information in my opinion.

Line 399-400: *“The model results demonstrate that the minimum values are observed at the boundary of the polar night in the part where the Sun has already returned”*

I find this statement problematic since the vortex is constantly moving. And I don't think it is correct, see my earlier comment on line 207-210. It also hard to see in the plots because there is no line showing the area of polar night. Delete the sentence.

Lines 400-402: *“This confirms the hypothesis of the effect of the chemical destruction of ozone, which intensifies after heterogeneous activation in polar stratospheric clouds and the return of the Sun.”*

This is basic textbook knowledge that everybody who reads this paper is aware of. That would be fine for the introduction, but not for the main text. In addition, it is phrased quite awkwardly, up to the point that it is not quite correct or very hard to understand what you mean. Delete the sentence.

Lines 402-404: *“However, the results of model calculations reveal that relatively low values of the total column ozone (below 300 DU) are also observed in the polar night zone, where the chemical destruction of ozone is slowed down. This also indicates a significant influence of dynamical factors on the formation of regions of low ozone content.”*

This is again basic knowledge, misleading and scientifically not relevant. I start to wonder whether the author of this section has a basic lack of understanding of the relevant science.

The total column value is a cumulative quantity, which is not determined by the position of the vortex at a particular date. In addition, the Arctic vortex is more dynamically active than the more circular Antarctic vortex, where it is more likely that air masses stay in darkness for a long time.

I think the only part of interesting information here would be the minimum column values. I also acknowledge that the discussion of dynamical factors can be interesting, but that would include things like interannual variations in diabatic descent or ozone mini-holes. The simple fact that air inside the vortex is moving and that the vortex is moving as a whole does not belong to this. This could be interesting if the authors would have done a detailed trajectory study of the history of air masses, but stating the obvious here is not enough in my opinion.

Line 406-407 *“To better understand the relative role of dynamical and chemical processes in the formation of the Arctic ozone anomaly in spring 2020”.*

I don't understand why it leads to a better understanding of the *relative* roles of dynamics and chemistry to look at the PSC area, i.e. what has the discussion in the following paragraph to do with that?

Line 407:

“Type I cloud” is ambiguous. Please clarify whether you mean STS or NAT clouds.

Line 408-409: *“but the inertia in their melting with increasing temperature is also taken into account.”.*

Would be worth noting here that your CTM does not use an equilibrium scheme as some other CTMs and to cite Smyshlyaev et al., 2010.

Lines 414-416: “*which suggests that the relationship between the formation of PSCs and ozone destruction is not linear and confirms the theory of several stages of the formation of ozone anomalies.*”

Apart from the fact that this sentence is phrased very awkwardly, this again states textbook knowledge and the obvious. Delete the half-sentence.

Line 416-417: “*In the polar stratosphere, associated, first, with the formation of PSCs, then with halogen activation on their surface, and only then with ozone destruction after the return of the Sun after the polar night.*”

Again, this is basic knowledge. Delete the sentence. This would be fine in the introduction, but not as a scientific result in the main text. And it is phrased so awkwardly that it is almost unintelligible.

Figure 10, Figure 11:

Indicate the edge of the polar vortex. It seems to me the area covered by PSCs is much larger than the polar vortex. Is this really correct?

Figure 10, Figure 11:

Again, give the units in the plots.

Figure 11 caption:

“low stratospheric coefficient of ozone destruction” does not give enough information to find out what is shown here.

Line 425: “*the coefficient of chemical ozone destruction in the lower stratosphere*”

This is introduced as it would be a well-known quantity (known by everyone under this name), but I think in fact it will cause confusion for many readers. E.g., what does “lower stratosphere” mean here? There is no altitude range mentioned in the following. This is important information that you need to give here.

Line 425-426: “*This coefficient is a factor by which the concentration of ozone should be multiplied in order to obtain the rate of its chemical destruction.*”

This definition seems not to be consistent with the magnitude of the values shown in Figure 11. If I understand you correctly, a value of  $1/s$  would mean that all of the ozone at a particular location would be depleted by chemical processes in 1 second. But the figure shows values on the order of  $10^8$  per second. Either I have difficulties to understand your definition or there is something wrong with the magnitude of the values given in the plot.

Lines 425-426:

Just to make sure that nothing is going wrong here. You take into account that there is a fast equilibrium between O and O<sub>3</sub> and only look at the net change of O<sub>3</sub>?

Lines 425-426:

Do you show and discuss instantaneous values at a given point in time (say 12 UTC) or daily averages? The plots make the impression that the latter is the case. But you don't tell us anything about that. This is important. Please clarify.

Lines 425-426:

I have the impression there might be quantities that would be more easy to understand which you could show here, e.g. simply the rates in ppb/day or a similar unit, or the reciprocal of what you show

here (the time scale needed for complete ozone destruction). But maybe my confusion is just caused because I have difficulties to understand your text. A better explanation and definition may help here.

Lines 428-429: *“It should be noted that the rate of ozone destruction in March has its maximum values at the boundary of the polar night in the region of the newly returned Sun.”*

This is hard to see, because the boundary of the area of polar night is not shown in the plots. And looking at Figure 11, I doubt that this statement is correct (assuming that you show daily averages). Delete the statement.

Lines 430-434: *“In this case, the maximum rate of ozone destruction is a necessary, but not sufficient condition for the formation of a zone of low ozone content in the spring. Dynamical factors also play an important role, in particular, for definition of the zone where the polar vortex is located. In particular, in early March, the rate of destruction of the base is maximum over the entire circle of latitude near the boundary of the polar night, and the minimum values of the ozone content are noted only in the eastern hemisphere (Figs. 8 and 9). Also in mid-March and April, areas of high ozone depletion cover a wider zone than areas of minimum total ozone.”*

Delete all of these statements. First of all, you obviously can't deduce the cumulative chemical destruction of ozone over a longer time period (that causes the low ozone columns) from the chemical rates at a single date, because the values add up. You don't need to argue with dynamical reasons here. This is really a basic flaw in your reasoning here.

And with respect to dynamics: And again, you are stating the obvious here. The polar vortex and the air contained in the vortex are moving. It makes no sense to note that the minimum values of ozone are over the eastern hemisphere in this context. The vortex is moving, and a few days later, this may look totally different.

Lines 436-439: *“For a more detailed study of the influence of dynamical and chemical factors on the local variability of the ozone content Figures 12 - 15 present the simulated with the CTM and measured by the OMI instrument changes in the total ozone content at four stations (two in the Western Hemisphere and two in the Eastern Hemisphere) during six months from the beginning to mid-2020.”*

I have a general comment here: While it is certainly fine to perform case studies like this for single locations, it would be have been so easy here to make more general and scientifically relevant statements by looking at vortex means (which should have been easily possible). I don't really see why the situation at a particular location is so scientifically interesting, but that may be my personal opinion. I think that you wasted a chance here without necessity.

Lines 436-439:

It seems to me that the information given in Figures 12-15 is largely redundant. Figure 12 and 13 (Pechora and Tura) show almost identical results. The same is true for Figure 14 and 15 (Resolute and Eureka). You could easily do with only two figures here. I would suggest to shorten the text in lines 450-526 significantly.

Line 440:

Is there any reason why you use SBUV data here and OMI data earlier in the manuscript?

Line 442:

Earlier in the manuscript, you give a value of 64 degrees N and not 66 degrees N. Only one of these values can be correct. Please correct.

Lines 444-446: *“Comparison of the baseline scenario with these two additional scenarios allows us to estimate the periods when chemical destruction of ozone is most effective after heterogeneous*

*activation on the PSC surface, and when gas-phase destruction of ozone in nitrogen catalytic cycles is more significant”*

See major comment 1. There is something fundamentally going wrong here.

Lines 446-448: *“In addition, the comparative role of dynamical and chemical processes of ozone reduction can be assessed by comparing these scenarios with each other and to mean climatic values presented at the bottom of these figures.”*

While I agree that you can disentangle dynamical and chemical changes when looking at these plots, this sentence is easy to misunderstand. E.g. you can't assess the comparative role of dynamical and chemical processes from comparing the “PSC” and “noPSCaer” runs.

Lines 450-526:

I think this part can be shortened considerably, not only for the reason stated above (lines 436-439). It is a little bit tiring that this is basically a description of what you can see in the figures (in very much detail), without so many scientifically interesting results.

Line 461-463: *“Based on a comparison of the noPSCaer and noCHEMall scenarios, it can be concluded that in the chemical destruction of ozone at Pechora station, the heterogeneous part is about one third (~ 25 DU), and the gas-phase part is ~ 45 DU.”*

See major comment 1. This can't be correct. Unfortunately, you don't indicate in the plots when the station is located inside the polar vortex.

Figures 12-15:

Indicate when the station is in the polar vortex. This is *very* important to be able to interpret the results correctly.

Figures 12-15:

In case you don't find the problem, remove the noPSCaer run from the plots.

Figure 12-15:

In the b panels, I would have found it more intuitive when the blue line would have been the difference between “noCHEMall” and “noPSCaer”.

Lines 471-481:

This is largely redundant with the paragraph about Pechora. I won't comment in detail and suggest to delete this.

Line 495: *“It should also be noted that there are two peaks of maximum chemical destruction of ozone: in late March and mid-April.”*

This is only the observation at this location because of the movement of the vortex. If you would look at the same air mass, this would be different.

Line 496-500: *“At the same time, chemical destruction in the second half of March is superimposed on a dynamic decrease in its content, which leads to a minimum in the seasonal variation of the total ozone content, while in April, when the chemical destruction of ozone is even greater than in March, the polar vortex is already shifting towards the eastern hemisphere (Fig. 8 and 9), and the total ozone content is higher than in March.”*

I find this sentence very hard to understand and unintelligible. For example, what do you mean by minimum in seasonal variation?

Line 507-509: *“Comparison of calculations for different scenarios of accounting for the chemical destruction of ozone depicts that the destruction of ozone over heterogeneous reactions in the western hemisphere exceeds 30 DU, which is more than in the eastern hemisphere, while the gas-phase destruction of ozone in the Western hemisphere is greater than in the Eastern Hemisphere.”*

Delete these sentences. See major comment 1. There seems to be a fundamental flaw in your method.

Lines 510-513: *“It should also be noted that in the Western Hemisphere, the minimum values of the ozone content according to satellite measurements in March are lower than the values calculated using the model, while in the Eastern Hemisphere the satellite and model results are closer. This result may be due to relatively coarse model resolution to simulate fine local effects in the western hemisphere.”*

Delete these sentences. You show a fundamental lack of understanding of the processes here. Since the vortex is moving, air masses that are located in the eastern hemisphere will be located somewhere else a few days later. This has nothing to do with the hemispheres.

Line 521-524: *“Additional numerical calculations to assess the effect of various catalytic cycles of chemical ozone destruction on a decrease in its content in April-May 2020 revealed that the main increase in the gas-phase ozone destruction occurs in the nitrogen catalytic cycle, in which the chemical reaction with the participation of nitrogen dioxide and atomic oxygen plays a determining role.”*

Since there is a fundamental flaw in your method (major comment 1), these results are very likely not correct. Delete this sentence.

Lines 524-526: *“In the Arctic stratosphere, in contrast to the Antarctic stratosphere, significant denitrification does not occur, and therefore a sufficient amount of nitrogen oxides remains in it, which plays a decisive role in the destruction of stratospheric ozone.”*

This statement is not correct, and it would have been easy to see that if you would have looked into the literature (e.g. Manney et al., 2020). Note that this statement is not correct in general, and not only for the winter 2019/2020. There are many Arctic winters which show a significant amount of denitrification, this is basic knowledge. Delete the sentence.

In addition, it seems that you use it here as an (wrong) explanation for your flawed results. I wonder why you did not notice that something must be wrong here.

Lines 528-531:

This sentence is phrased so awkwardly that I have a very hard time to understand what you want to say. It is almost unintelligible. Please rephrase. I won't give a suggestion here, because this is the conclusions and I am not sure what you want to tell us.

Line 535: *“The of SSW event in the middle of March 2020”*

This part of the sentence makes no sense. What do you want to tell us?

Line 535: *“although it did not satisfy the WMO definition of Major SSW event”*

Earlier in the paper, I had some comments that you were referring to a SSW event that you never mentioned before. And now, in the last lines of the paper, you tell me that it actually was no SSW event. What does this mean? That I can forget about everything that I have read about the SSW event? This information should have been given much earlier.

I agree that the warming of the vortex in late March led to a relatively abrupt stop of chemical ozone depletion. Can you rephrase this.

Lines 537-545, 554-557:



This is not my area of expertise. I will skip this part.

Lines 558-560:

You need to be more specific here. You have only results for two potential temperature levels. You don't mention that you refer to vortex means. You don't mention the date you are referring to. And give numbers for the ozone loss.

Line 566-569: *"...reveal that both dynamical and chemical processes make significant contributions to the decrease in the ozone content inside the polar vortex. In this case, the chemical ozone depletion is determined not only by heterogeneous processes on the surface of polar stratospheric clouds, but by gas-phase destruction in nitrogen catalytic cycles as well."*

This is not correct and misleading. See major comment 1. Delete this from the conclusions.

Line 573:

It seems that there is something missing in the "Author contributions". It starts with "All other authors...", implying that a sentence is missing at the start. There is information missing who has written the main text.

## **Technical corrections (language etc.)**

Title:

"Dynamical and chemical processes contributing to ozone loss in the exceptional Arctic stratosphere winter-spring of 2020" (added "the")

Line 8:

You can delete "The features". Just start with "Dynamical processes and changes..."

Line 17:

"repeated" is probably not the perfect choice of word. Maybe "which was similar to the depletion in 2010/2011"

Line 33:

Change "the main SSW" to "a main SSW"

Line 38:

Change: "the largest decrease in the Arctic ozone was observed" to "the largest decreases in Arctic ozone were observed"

Line 49:

You certainly mean "statistically" and not "statically"

Line 52:

You probably mean something like "nevertheless" and not "in the meanwhile"

Line 132:

Change "reveal" to something like "estimate" or "determine"

Line 148:

Change “the Lagrangian approach” to “a Lagrangian approach”

Line 161:

Change “were interpolated into the points of each trajectory” to “were interpolated to the positions of each trajectory”

Line 164:

Change to “Ozone sonde data ... have been used”

Line 168:

You misspelled the name in the reference. The correct name is in line 175 (Braathen). There is also a superfluous “;”

Line 176:

Section 2.4 is mislabeled as Section 2.3

Line 182:

Split the sentence and shorten: “Meteorological fields are specified...”

Lines 184-186:

Awkward phrasing. Change to e.g. “The model includes 74 oxygen, hydrogen, nitrogen, chlorine, bromine, carbon and sulfate species. The chemistry of the species is calculated as described in Smyshlyaev et al. (1998).”

Line 191-192:

Again, phrased awkwardly. Suggestion: “For a more detailed study of the influence of dynamical and chemical factors on the local variability of the ozone content, two additional numerical experiments with the RSHU CTM were performed in addition to the reference run (termed “PSC” here).”

Line 204:

Change “at the early March” to “in early March”

Line 206:

Do you mean “north of Alaska”?

Line 207:

Maybe “which are based on solar radiation” is better English.

Line 215:

Change “north to” to “north of”

Line 217:

Change “values less than 220 DU” to “values of less than 220 DU”

Line 220:

Change “territory” to “area”

Figure 1 caption:

The text speaks of OMI and the caption speaks of AURA. Would be nice to have that consistent.

Line 229:

Split sentence. Write something like: “Temperatures were sufficiently low to allow the formation of NAT and STS clouds”.

Line 229:

Can we stick to Kelvin and not to degree Celsius?

Line 230:

Change “Figure 2a” to “Figure 2”. There is only one panel here and 2a does not exist.

Line 232:

Do you mean “Two main causes of the cold and stable Arctic polar vortex“ and not “Two main causes of so cold and stable Arctic polar vortex”? Or what were you trying to say?

Figure 2 caption:

Change “climate mean” to “climatological mean”

Figure 2 caption:

There is a Russian letter in the caption (probably means “and”)

Line 242:

Change to “Furthermore, we compare...”

Caption Figure 3:

Change “Latitudes are from 30 N” to “The map shows only latitudes north of 30 N”

Line 263-264:

“Notably that described positive temperature anomalies were observed not only near surface but at higher levels in troposphere.” This is phrased awkwardly. Suggestion: “Positive temperature anomalies were observed not only near the surface but also at higher levels in troposphere.”

Line 266:

Change “and increased in” to “and increased pressure in”

Lines 273-274:

You probably mean “between parts of the atmosphere that are heated by the sun and parts that are shaded”? But I think a native speaker probably wouldn’t phrase it like this. I would talk of the sunlit part.

Line 294:

Change “In the same time” to “In the same time period”

Line 304:

Change “till” to “until”

Line 305:

Change “This is confirmed by the diagram with...” to “This can be seen in Figure 5a showing ...”

Figure 4 caption:

Change “climate mean” to “climatological mean”

Line 316:

Change “display dominated” to “show pronounced”

Line 334:

Change “descent” to “descend”

Figure 7 caption:

Change “0 day...” to “The horizontal axis shows the number of days since December 1.”

Line 358:

Change “average vertical descending” to “average vertical descent” (this time the “t” is correct!)

Line 360:

Change “As well to estimate chemical ozone loss” to “As another method to estimate chemical ozone loss”

Line 361:

Change “Ny-Ålesund” to “Ny-Ålesund”

Line 367:

Change to “Figure 8 shows the vertical profile of the vortex-averaged cumulative ozone loss...”

Line 369:

“with largest losses”: Start a new sentence and write “These winters showed the largest ozone losses previous to the winter 2019/2020.”

Line 371:

Split into two sentences. “...than in 2010/2011. That is consistent...”

Line 380:

Awkward phrasing. Change “For a more detailed study of the degree of dynamical and chemical processes influence on the formation of ozone anomalies...” to “For a more detailed study of the dynamical and chemical processes that influence the formation of ozone anomalies...”

Line 381-382:

Awkward phrasing. Split into two sentences. Change “in which the dynamic parameters were set from the MEPRA-2 reanalysis data” to “Meteorological data were obtained from the MERRA-2 reanalysis”.

Line 382:

Note the change “MEPRA-2” to “MERRA-2” in the previous comment

Line 391-392:

Change “Figure 9 demonstrates” to “Figure 9 shows”

Line 392:

Awkward phrasing and a lot of repetition of information: Change “the results of calculations of the total column ozone for March-April 2020, performed using the CTM with the specified dynamical parameters from the MERRA-2 reanalysis” to “... shows the total ozone column for March-April 2020 from the CTM”.

Line 410:

Replace “territory” by “region” or “area”

Line 411:

Change “the area of the PSCs zone is maximum” to “the area covered by the PSCs is maximum”

Line 413:

Change “the area covered by PSCs significantly reduced” to “the area covered by PSCs are significantly reduced”

Line 425:

Change “Fig. 11 demonstrates” to “Figure 11 shows”

Line 454:

Phrased awkwardly. Change “which maximally affect the ozone depletion in April” to something like “Cumulative ozone depletion shows maximum values in April”

Line 458:

Change “two times” to “by a factor of two”

Line 460:

Change “if compare” to “when compared”

Line 492:

“the total content fluctuates” This is phrased awkwardly.

Line 531:

Change “Further” to “Furthermore”

Line 532:

Change “ozonosondes” to “ozone sondes”

Line 532:

Split into two sentences: “...observations. Finally, ...”

Line 537:

Delete “revealed”

Line 562:

I don't think that “repeat” is the best choice of word here. Maybe “rivalled”

Line 586:

Change “Ny-Aalesund” to “Ny-Ålesund”

Line 607:

Don't abbreviate "QJRMS"

Lines 644-645:

Jadin et al. is an article from a predatory journal. Delete the reference.

Line 686:

It seems to me that the reference Madrid et al. is not cited in the paper. Delete.

Line 703-704:

Pedatella et al. is a news article. Delete the reference.

Line 711:

Change "Lefe`vre" to "Lefèvre"

Line 738-740:

Smyshlyaev et al., 2017 is only available in the Russian language, so I can't read it. See specific comments to line 180.

Line 760-761:

I wasn't able to find this article on the home page of the journal.

### **References**

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