Response to reviews on "Stratospheric impact on the Northern Hemisphere winter and spring ozone interannual variability in the troposphere"

by Junhua Liu et al.

We thank the three reviewers for their helpful comments and Ryan Williams for his interactive comment. We have addressed all comments in detail below and have clarified the text in the relevant sections.

In the following, we address the concerns raised by all the reviewers. Reviewers' comments are italicized.

Anonymous Referee #1

Overview: This paper uses modeled and observed ozone to examine the interannual variation of the impact of stratospheric ozone on tropospheric concentrations and is restricted to mid to high latitudes in the NH during winter and spring. The authors conclude that the model well reproduces the interannual variations in tropospheric ozone, except over North America following the eruption of Mt. Pinatubo. They infer that the STE was too strong over NA after the Pinatubo eruption. The paper will be suitable

for publication, but I recommend revision prior to acceptance, after the authors have considered the questions noted below.

Question 1: The authors state that the stronger and deeper stratospheric contributions in the tropospheric O_3 variability shown by the model is related to the ozonesondes being closer to the polar vortex in winter over NA than over Europe. This doesn't make sense to me. Does it mean that you're effectively comparing apples and oranges, in that you're looking at different meteorological regimes when looking at your NA data vs your European data? The text makes it sound like the ozonesondes are somehow controlling what the model does.

Thanks a lot for the comments by the first reviewer. The text has been modified to avoid the confusion. Figure 9 and 10 in revised manuscript show that there are strong longitudinal variations in NH (averaged between 30°N to 80°N) meteorology (tropopause pressure, geopotential heights), which results in the longitudinal variations of stratospheric O_3 contribution between N. America and Europe. Please see below for the modified text:

Our analysis of the MERRA2 assimilated fields shows strong longitudinal variations in meteorology over northern hemisphere (NH) mid-high latitudes, with lower tropopause height and lower geopotential height over North America than Europe. These variations associated with the relevant variations in the location of tropospheric jet flows are responsible for the longitudinal change in the stratospheric O_3 influence and result in a deeper and greater stratospheric O_3 influence on the tropospheric O_3 over North America than that over Europe.

Question 2: The Orbe 2017 paper referenced talks about multiple version of a replay simulation, and discusses various deficiencies in the large-scale transport depending on how the simulation was done. Which one of the runs discussed in the Orbe paper is this study using? Or, because it seems this is a higher horizontal resolution run than discussed in Orbe et al, 2017, is it something completely different? My concern is that the Orbe paper talks about potential issues (i.e., regarding age of air in particular) regarding the replay simulations, so have you picked a version of the model that would best represent overall transport?

We are referring the Orbe et al (2017) paper to explain the detailed description of the "replay" methodology. The runs discussed in the Orbe paper are performed at a coarser resolution. Neither of them is the one used in our study. The simulation used in our study has the similar setting as RAs3, which best represents overall transport. The text has been modified as below:

We use a replay simulation (http://acd-ext.gsfc.nasa.gov/Projects/GEOSCCM/MERRA2GMI) of the GEOSCCM with the Global Modeling Initiative (GMI) chemical mechanism (Strahan et al., 2007;Duncan et al., 2007) for trace gas chemistry, which includes a complete treatment of stratospheric and tropospheric chemistry, and the Goddard Chemistry Aerosol Radiation and Transport (GOCART) module (Chin et al., 2002;Colarco et al., 2010) for aerosols. The replay simulation follows the replay methodology as described in Orbe et al. (2017) and uses the RAs3 setting, which best represents overall transport. The model reads in the three-hourly time-averaged output of MERRA-2 meteorology (U, V, T, pressure) and recomputes the analysis increments, which are used as a forcing to the meteorology at every time step over the 3 h replay interval. More detailed information on replay methodology can be found in Orbe et al. (2017). The replay simulation is run at a MERRA-2 native resolution of ~50 km in the horizontal dimension and 72 vertical levels. This replay simulation is referred to as the 'MERRA2-GMI" simulation.

Question 3, discussion of figure 4 tropospheric comparison. The authors states that the phase is in agreement but the magnitude is underestimated by the model for the observed anomalies. (and, do you calculate the anomalies from the individual stations and then average, or from the averaged ensemble of 17 stations? This should be stated before the figure is presented.) I think really you mean sign is in agreement rather than phase. I also don't see that in general that the absolute value is underestimated by the model. At 700 mb, the model and obs don't agree on the sign for the period from 2012- 2015. At 400 mb, they don't agree on the sign for 1990-end of 1991. At 400 mb, there is an underestimate sometimes, and an overestimate from 1997-2001. I also don't understand the statement that both obs and simulations show the largest interannual variations in winter and spring. Am I supposed to be able to discern that from Figure 4? Perhaps that statement shouldn't be made until you've presented figure 5. And, in the caption of figure 4, please say what the red and black numbers are supposed to mean.

We agree with the reviewer 2 that Figure 4 did not provide more useful information by comparing observations from all stations with the model simulation. We therefore removed Figure 4 and section 4.1.

Question 4, discussion of figure 5. The authors state that, for 200 mb, the IAV is larger over NA than Europe, and larger in spring than winter. These appear to be qualitative statements. Do you have a way to calculate a value for IAV (i.e., perhaps the standard deviation of your anomalies)?

It would then be possible to apply some sort of statistical test to assess whether there really is a regional or seasonal difference.

Thanks a lot for the reviewer's suggestion on statistical analysis. We calculated the standard deviations of the anomalies to support our arguments of IAV. We also performed several statistical F-test to assess the equality of variance (standard deviation) for the selected anomalies. The significance of F-test is a value in the interval [0.0, 1.0]; a small value (< 0.2) indicates that the selected two datasets have significantly different variances. Below are two tables to assess whether there is a significant difference in the IAVs 1) between North America and Europe, 2) between DJF and MAM. The objective of our paper is quantifying the stratospheric O_3 influence on the tropospheric O_3 IAV, the seasonal or regional difference of O_3 IAV is not the focus of our paper. We therefore add those tables into supplementary materials. Corresponding discussions are added into text.

		DJF	**MAM**
200 hPa	Std_{na} (Std_{eu})	44 (44)	57 (54)
	F-test	0.99	0.82
400 hPa	Std_{na} (Std_{eu})	**3.08 (2.34)**	**4.94 (2.54)**
	F-test	**0.17**	**0.001**
700 hPa	Std_{na} (Std_{eu})	**2.94 (1.59)**	**2.56 (1.73)**
	F-test	**0.002**	**0.05**

Table R1: Standard deviations and F-test statistics of the observed O_3 anomalies over N. American sites (Std_{na}) and European sites (Std_{eu}), to assess whether there is significant regional difference in the amplitude of IAVs between North American and European sites.

At 200 hPa, there is not significant regional difference in the magnitude of O_3 IAV between North America and Europe in both seasons. At 400 hPa and 700 hPa, ozonesonde observations show significantly greater IAV over North America than Europe in both seasons.

		North America	**Europe**
200 hPa	Std_{djf} (Std_{mam})	**44 (57)**	**44 (54)**
	F-test	**0.19**	**0.28**
400 hPa	Std_{djf} (Std_{mam})	**3.08 (4.94)**	2.34 (2.54)
	F-test	**0.02**	0.69
700 hPa	Std_{djf} (Std_{mam})	2.94 (2.56)	1.59 (1.73)
	F-test	0.5	0.66

Table R2: Standard deviations and F-test statistics of the O_3 anomalies in DJF (Std_{djf}) and MAM (Std_{mam}), to assess whether there is significant seasonal difference in the IAVs.

At 200 hPa, ozonesonde observations show significantly greater IAV in MAM than DJF over both regions. At 400 hPa and 700 hPa, there is not significant seasonal difference in the magnitude of O_3 IAV between MAM and DJF, except for over North America at 400 hPa, where observed O_3 IAV is greater in MAM than DJF.

Question 5, The author's state that the correlation between polar winter 150 mb temps and 200 mb ozone anomalies being lower in spring is "consistent with our understanding of the impact of

temperature variations on the formation of polar stratospheric clouds and polar vortex isolation with reduced transport of o3 from the tropics at low temperatures....". I personally don't follow this at all. Are you trying to explain why there is a correlation, or why the correlation is different between winter and spring?

We deleted our discussion about the relationship between O_3 IAV and temperature at 150 hPa averaged over latitude north of 60°N. The averaged temperature is a good measure of the overall temperature in the polar vortex (https://ozonewatch.gsfc.nasa.gov/facts/vortex_NH.html). Although data show the high correlations between polar vortex temperature and O_3 IAV over selected sondes station in the lower latitudes, we cannot derive the directly causality without more detailed examinations.

Question 6, I think you need a quantifiable definition of what you mean by IAV in order to compare where it is larger or smaller in different seasons or in different regions. The paper is written as though IAV is the same as the deviation (anomaly) from the seasonal mean. One then has to determine the interannual variations from looking at wiggles in anomaly plots.

Please see our response to Question 4.

The definitions of IAV amplitude has been added in text. Text has been modified based on the statistical comparisons of standard deviations.

Question 7: Discussion of Table 3, Please explain how, from looking at the correlation coefficients in Table 3, that one concludes that 27% of the NA interannual variation is related to 200 mb changes in winter.

We calculate the percentage of variance explained (r^2) through the correlation. The correlation between O_3 anomalies at 200 hPa and 400 hPa in winter is 0.52 means 0.52^2x100 = 27% of the variance in 400 hPa is "explained" or related to 200 hPa O_3 anomalies.

To avoid confusion, we replaced r with r^2 in Table 3 and modified corresponding discussions in the text. We also add the definition of explained variance in the revised manuscript.

Question 8: Discussion of Figure 6, Mt Pinatubo erupted in June 1991. Your 700 mb DJF NA plot shows a large difference between the red, black and green lines for 1990. What are you defining as the "Pinatubo period" and do you keep 1990 in your re-calculations of strato3-o3 correlation when you say you omit the Pinatubo period?

We define the Pinatubo period as year 1991-1995. No, the re-calculation is from 1996 to 2016. The text has been modified to avoid the confusion.

Question 9: around line 260-265 it states that anomalies in strato3 diverge from simulated o3 near the end of the period, and looking at figure 5, that seems to be around 2012. Do precursors really become significantly important only in the past decade?

No. Precursors are important through the whole time period in the lower troposphere, especially over Europe, where there are less stratospheric intrusions. That is why we see small correlations between $StratO_3$ and O_3 at 700 hPa. Below figure shows the $StratO_3/O_3$ averaged over Europe sites

at 700 hPa and 900 hPa in boreal winter season (DJF) from 1990 to 2016. We can see that the $StratO_3/O_3$ ratio is less than 0.5 and decreases sharply at 900 hPa after 2014.

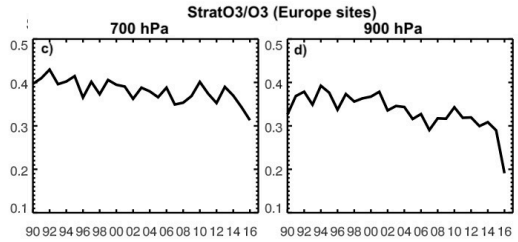

Figure R1: Time series of $StratO_3/O_3$ averaged over North America sites (top) and Europe sites (bottom) at 700 hPa and 900 hPa in boreal winter season (DJF) from 1990 to 2016.

We modified the text as below:

There is no significant relationship between $StratO_3$ and simulated O_3 at 700 hPa. This is expected since the impact of stratospheric ozone decreases, and the impact of ozone production from its precursors becomes more important at lower altitudes.

Question 10: If you separate the analysis more finely than simply Europe vs NA, and compared comparable latitudes, do you come to the same conclusions? How different are Madrid and Wallops? Your NA comparison includes more high latitude stations than your European one does. Is it longitude you're finding differences between, or latitude?

Regarding to reviewer's comments about N. American sondes, we have analyzed the latitudinal difference of N. America ozonesonde by separating ozonesonde stations into 3 groups (> 70N, 70N-50N, and <50N). We do find that O_3 IAV over N. America varies with latitudes, but the longitudinal difference of $StratO_3$ influence to the troposphere between N. America and Europe is persistent over most NH mid-high latitudes (Figure R2).

We identified that the stronger and deeper stratospheric O_3 influence over N. America than Europe through the comparisons sampled at sonde stations. In section 5.2, we extend our analysis from O_3 sampled at stations to the latitudinal average between 30°N and 80°N. As shown in Figure 9 and 10 in revised manuscript, the stronger and deeper stratospheric O_3 influence over N. America than Europe is a large-scale phenomenon, and not artificially caused by the locations of sondes stations. Below figure (Figure R2, also Figure S3) shows the climatology map of $StratO_3/O_3$ at 400 hPa in DJF and MAM averaged from 1990 to 2016. Red thick line is the location of strongest winds, which indicates the approximation of the jet climatology locations. Due to large latitudinal temperature and strong westerly upper level winds, the westerly jet breaks down into large-scale eddies, which are called the baroclinic eddies. The baroclinic eddies push warm air poleward and cold air southward, cooling the subtropics and warming the polar latitudes, in wavelike pattern. As we can see from the figure below, the jet meanders to the south over central and eastern N. America and bringing cold polar air with more stratospheric subsidence. The jet moves to the north over Europe and brings in warm air with less stratospheric O_3 influence. The longitudinal difference is persistent between N. America and Europe over most mid-high latitudes.

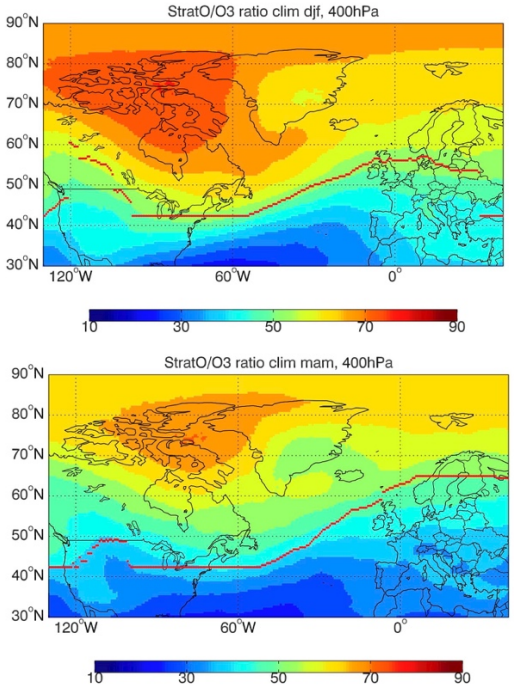

Figure R2: Spatial maps of simulated $Strat O_3/O_3$ ratio climatology at 400 hPa in DJF (top) and MAM (bottom) averaged from 1990 to 2016. Red thick lines indicate the approximated climatological jet locations, where the strongest winds are.

Question 11: On line 306, replace "changes" with "relationship" Your plot shows snapshots of winter and spring 1993, not differences (or changes).

The text has been modified as suggested.

Question 12: Final paragraph, the implication here is that the underlying meteorology was deficient over NA in the early period, but perhaps not over Europe. What would be the reason for that? And, can you look at any other fields in the model/sonde comparisons to assess whether this is the issue (maybe tropopause pressure, or the temperature from the radiosonde that flew with the ozone sonde?

One possible reason is related to the spatial representativeness of meteorological measurements over these two regions. As we can see from Figure 7a in revised manuscript, the westerly subtropical jets show a southward shift over N. America and moves northward over Europe. Most stations over Europe are located south of the subtropical jet, with less dynamic perturbation. Over N. America, the excessive STE are inferred over two stations between 50N and 70N (Figure S2), which are on the edge of the subtropical jet, a region with complex metrological regimes and strong O_3 gradient. Considering the much coarser and low-resolution observations in the underlying meteorology in earlier period, problems tends to occurs over a region with more dynamical perturbations (e.g. N. America) than a meteorologically stable region (e.g. Europe).

Evaluating the regional accuracy of underlying meteorology in the early period is beyond the scope of this paper.

specific comment: please change "amplitude" on line 194 to "magnitude".

The text has been modified as suggested.

Anonymous Referee #2

Liu et al. use model simulations of ozone and a stratospheric ozone tracer together with observations from ozonesondes to investigate the interannual variation of ozone and the vertical extent of the impact of stratospheric ozone on tropospheric ozone. Before the simulations are used for the analyses their quality is checked by first comparing the simulations to measurements. I am confident that the study itself is important and deserves to be published, however, I am not happy with how the result from these studies are presented. The manuscript in its present form is confusing and needs thorough structuring and a clear line. From the current manuscript is not clear what the major focus of this study is: Do you want to evaluate the model or do you want to investigate the stratospheric impact on the NH winter and spring interannual variability in the troposphere as it is stated in the title.

The manuscript in its current form has a stronger focus on the evaluation of the model than on the analyses of the interannual variability. Further, a lot of information is packed into the figures and thus makes it quite hard to follow and get the major results through. I would suggest major revisions before the manuscript can be published.

We acknowledge the comments by the second reviewer. But we disagree with the reviewer's comments "The manuscript in its current form has a stronger focus on the evaluation of the model than on the analyses of the interannual variability." The purpose of the paper is not just model validation, but primarily to use observations, model and StratO_3 to answer the question of how the stratospheric O_3 impacts the troposphere O_3 IAV.

Considering that there is no publication on evaluation of the tropospheric O_3 simulation from the MERRA2-GMI run, we think it is very important to do the model evaluation before using the model to explain the cause of tropospheric O3 variations.

Specific comments:

P1, general: Why is it important to look at the interannual variation? What are the unanswered questions? The motivation for this study is not clear. In the introduction (P2, 58-59) a motivation is given. Something like this could be repeated in the abstract.

Using the interannual variation is a good way to evaluate stratospheric impact, since we are looking at the response of tropospheric ozone to stratospheric forcing.

We have a brief discussion of motivation in the 1st paragraph of the introduction, which lead to the main objective of our study in the 2nd paragraph of the introduction. In the 3rd and 4th paragraph we give a more detailed description of background and unanswered questions of this topic and our approach to achieve the goal. We think the motivation are sufficiently described in the introduction and we don't think it is necessary to include it in the abstract.

P1, L1: How long is the model run? That should be mentioned here.

The run period has been added in section 2.2. The analysis period is added in the abstract.

P1, L29-30: Why should ozone sondes be closer to the polar vortex? This sentence is somewhat weird and misleading and thus should be rephrased.

Discussion has been modified to avoid the confusion.

Please see our response to the Question 1 from the reviewer 1.

P2, L44: What exactly are these "replay" simulations? This should be explained. What atmospheric conditions or initial conditions have been assumed for this simulation?

Please see our response to Question 2 from the reviewer 2.

P2, L48: Which parameters exactly? Can you give some examples?

We replace 'parameters' into 'system'. The parameters we used in our study include air mass flux, tropopause pressure and geopotential heights.

P3, L75ff: Here you give a better description of the aim of this study. Something like this should be also added in the abstract, so that it also there becomes more clear why it is important to investigate these processes.

Please see our response above. We discuss the objective of this study in 2nd paragraph of the introduction.

P3, Section: A comparison for each station would also be quite useful to understand local differences and which stations/locations maybe mess up the mean.

Our examination on individual stations shows that the underestimate of tropospheric O_3 depletion during the DJF and MAM of the Pinatubo period exist over most N. American stations. Simulations at Wallops Island did a better job among all the N. America stations. Over Wallops Island, model reproduce the O_3 variation at 400 hPa, but still underestimate the decreased O_3 at 700 hPa.

P5, L135ff: The comparison to the satellite data has not been mentioned in the abstract or introduction. Why? If it is a part of this study it should be mentioned there. Why do you this comparison in the first place? Is this really necessary? You anyway comparethe model simulations to sonde data so. Therefore, I do not understand what additional information is gained by doing an additional comparison. Especially, if your focus is not on the evaluation of the model but on the investigation of the impact of stratospheric ozone on tropospheric ozone.

We think it is necessary and important to include satellite comparison. We want to know that the model performs well in the large scale before looking at sondes. But we can put these figures in the supplement if the reviewer insists.

P5, L154ff: Reference to the figure is missing.

The reference to the figure is at the end of the sentence.

P7, L205ff: I cannot follow how you derive this conclusion. Which season and time periods are you referring to? How have the numbers in percent been derived?

Please see our response to Question 7 of the reviewer 1.

To avoid confusion, we replaced r with r^2 in Table 3 and modified corresponding discussions in the text.

P8, L228: What exactly is the StratO3 tracer? What is included in the diagnostic? How is it calculated? Is this simply the stratospheric O3 flux?

More detailed discussions of the $StratO_3$ tracer setting in the model have been added in section 2.2. Please see below:

A $StratO_3$ tracer is included in the model to track the stratospheric O_3 influence on the troposphere. $StratO_3$ is set equal to simulated O_3 in the stratosphere and is removed in the troposphere based on interannually-varying monthly mean loss rates and surface deposition fluxes archived from the standard full chemistry simulation, thus diagnosing the relative importance of stratospheric ozone at all locations in the troposphere. $StratO_3$ tracer is defined relative to a dynamically varying tropopause tracer (e90) (Prather et al., 2011). The e90 tracer is set to a uniform mixing ratio (100 ppb) at the surface with a 90-day e-folding lifetime everywhere in the atmosphere. This lifetime is long enough for the tracer to be well mixed throughout the troposphere but short compared to the transport time scales in the stratosphere, resulting in sharp e90 gradients across the tropopause. In our simulations, the e90 tropopause value is set to 90 ppb. The e90 tracer has been used in many studies of STE as an accurate tropopause definition and an ideal transport tracer in UTLS (e.g., Hsu and Prather, 2014;Liu et al., 2016;Pan et al., 2016;Randel et al., 2016;Liu et al., 2017).

P8, L234: Where exactly do we see this in Figure 6?

Figure 5 e, f in the revised manuscript. The reference to figure has been added in the text.

P9, L266ff: Also here it is not clear how the numbers in percent have been derived.

Please see our above response. The numbers are square of correlation coefficients.

P9, L267-267: Here an important result is given, but it gets somehow lost in the discussion of the differences between the model simulations and observations.

We include this result in the abstract.

P9, L269: Reference? Has this relation seen before? Has this relation already been discussed somewhere else?

This sentence provides a hypothesis to explain the difference in the stratospheric O_3 influence between North America and Europe as shown in Figure 5 in the revised manuscript. We move this sentence to the next paragraph to lead the discussion of Figure 6 in the revised manuscript.

P10, L298-299: This sentence is too complicated and should be rephrased. Maybe it would be better to split this sentence also into two sentences.

The text has been modified as suggested:

In the equatorward breaking, tongues of high PV and stratospheric air extend equatorward associated with frequent STE processes. In the poleward breaking, tongues of low PV and upper tropospheric air extend poleward.

P10, L308: It would be worth to more clearly state that because of the different tropopause heights different pressure levels are shown in the figures.

The text has been modified as suggested.

P10, L315: How it the air mass flux derived/calculated?

The air mass flux is air mass flow rate, which is calculated by multiplying omega (the volume flow rate which depends on the pressure difference) with the density of air.

The text has been added into revised manuscript.

P10, L320: not shown? Or is this shown? Can this be seen when comparing 1993 to 1998?

This can be seen when compare 1993 and 1998. You can see the difference of longitudinal variations of subtropical jets between Figure 7 and 8 in revised manuscript.

P10, general: In the introductory part of this section StratO3/O3 distinction based on PV is mentioned, but in the analyses the air mass flux is used.

PV is mentioned in Thorncroft's paper to characterize these wave-breaking events, which also closely associated with STE process. In our analysis, we rely on air mass flux to infer the strength of STE process.

P11, L327: Here four panels are given, but only 2 panels show the 400 hPa level.

The labels have been corrected in the text.

P11, L330: Why is there less dynamic perturbation?

We infer this from the maps of winds (Figure 7a) and air mass flux (Figure 7c) in the revised manuscript. Both horizontal and vertical transport is smaller over north of 70°N than regions between 50°N and 70°N.

P12, L363: Why are these three parameters used? What is the connection between these? This is not really discussed. Wouldn't it then be easier to just show StratO3/O3?

Figure 9 and 10 in the revised manuscript examine whether the longitudinal variations of $StratO_3$ influence on tropospheric O_3 inferred from observations and simulations over North America and Europe sonde stations is a large-scale phenomenon and related to the large-scale circulation patterns. The geopotential heights and tropopause pressure are good representors of large-scale circulation patterns. We therefore use these two parameters to represent the dynamic system.

P12, L383: maximum? Shouldn't it read minimum? Generally, I have the feeling that in this paragraph the description does not agree with the figure shown.

We change 'correlation' into 'anticorrelation". In this way, it is correct to say the anticorrelation reaches maximum at the surface.

P13, L396: This is not clear. How does the Pinatubo eruption deplete ozone? Do you mean in the troposphere or the stratosphere and by which process?

There are many studies examine the stratospheric and tropospheric O_3 depletion after the Pinatubo eruption through dynamics and chemistry processes. Please see discussion in section 4.1.

P13, L410-411: This does not become comprehensible from what is shown in the manuscript.

This is a conjecture based on our analysis. The observations show that tropospheric O_3 decreases after the Pinatubo, reflecting the decreased O_3 as seen in the stratosphere. Model reproduced the observed stratospheric O_3 decrease, but did fully reproduce the observed tropospheric O_3 decrease. Our model analysis shows that there is an increase of $StratO_3$ in the troposphere after the Pinatubo eruption. $StratO_3$ changes in the troposphere are due to two factors: ozone concentrations in the stratosphere, and the mass flux from stratosphere. We therefore speculate that model may overestimate the downward flux at this period and the effect of decreased stratospheric ozone to the tropospheric O_3 could thus masked by this overestimation in the model analysis.

Figure 2 and 3: Are these figures really useful? Especially, since later anyway the simulations are compared to ozone sonde data. This part of the study could (if required) be provided in the supplement.

We think that all these figures lend credibility to the model. We think it is necessary and important to include satellite comparison. We want to know that the model performs well in the large scale before looking at sondes.

Figure 4: What does the reader gain from this Figure? Is there any more information gained when comparing observations from all stations with the model simulation?

We remove the Figure 4 and its discussion as suggested by the reviewer.

Figure 5, 6, and 7: I would suggest to split these by North America and Europe and discuss the regions separately. As you do it now, you compare different pressure levels, seasons and regions and it gets really hard to follow since you also above all that additionally discuss the differences between model simulation and observations.

We keep these figures unchanged, since they show direct regional and seasonal comparison.

But we modify the text to discuss the regions separately and a summary of the difference between N. America and Europe.

Figure 8, 9: Again too many panels and too many things discussed at the same time. I would suggest to solely show the anomalies in the figure and to provide the airmass flux in the supplement.

There are four situations in flux change: 1) increase of downward flux, 2) decrease of upward flux, 3) decrease of downward flux 4) increase of upward flux around the tropopause. We cannot distinguish these four situations based only on the anomalies of airmass flux. We have to combine the maps of air mass flux and its anomalies to determine how flux changes.

Technical comments: P2, L18: add "of O3" after input and maybe use a different wording

for "input", e.g. entrainment.

The text has been modified as suggested.

P2, L47: "in so doing" ! "in doing so"?

The text has been modified as suggested.

P4, L99: present = 2019? It would be better to clearly state the year here.

The text has been modified as suggested.

P4, Section 4 header: remove colon.

The text has been modified as suggested.

P4, Section 4.1 header: remove full stop after title.

The text has been modified as suggested.

P7, L219: space between "correlation" and reference of "Terao" missing.

We have the reference of "Terao et al 2008"

P7, Section 4.3 header: Remove colon.

The text has been modified as suggested.

P12, L360: "impact on tropospheric O3 from the upper to lower troposphere" ! not clear. Please rephrase the sentence.

We rephase the sentence into: the significant impact of the StratO_3 IAV on tropospheric O_3 reach to the lower troposphere.

P13, Section 6 header: remove colon.

The text has been modified as suggested.

Figure 8 and 9: Panel labelling with a,b,c: : :.. is missing.

Labels have been added in the figures.

Figure 8: Adjust both columns so that they are next to each other at the same height. At the moment there is a shift between the columns.

Figure 8 has been reproduced as suggested.

Figure 10 and 11: 180 W on the right side of the x-axes should read 180 E.

The label has been corrected.

Figure 10 and 11: To use white dashed lines instead of black dashed lines would increase the readability.

We tried the white lines. The effect is not good. We therefore keep the black lines.

Figure 12: Also here North America and Europe should be marked.

The figure has been modified as suggested.

Anonymous Referee #3

The paper compares the 1990-2015 ozonesonde observations at 8 North American and 9 European sites with CCM output of tropospheric ozone levels to study the stratospheric impact on the observed tropospheric ozone concentration time series. The (total + tropospheric) ozone output of the model is first validated by comparison with satellite ozone retrievals. Making use of a model stratospheric ozone tracer, the impact of STE on tropospheric ozone is assessed, together with the analysis of model wind patterns and airmass fluxes.

GENERAL COMMENTS

The study is scientifically sound and takes into account all relevant literature. The analysis is detailed and all relevant aspects are considered. The presentation is clear, although somewhat verbose at some locations, and follows a very logical structure. It therefore deserves publication in ACP, if some remaining issues can be described better or clarified. These are summed up here below.

Thanks a lot for the comments by the third reviewer.

SPECIFIC COMMENTS

** From the text (page 4, lines 114-120), it is not clear how the stratospheric ozone tracer (StratO3) is defined. Please be more specific on this important variable of your analysis.*

More detailed discussions of the $StratO_3$ tracer setting in the model have been added in section 2.2.

** On page 5, lines 147-150: please, be more quantitative when comparing the magnitude, IAV and trend of the tropospheric ozone satellite retrieval and model replay simulation.*

More in general, I agree with reviewer 1 that, throughout the entire manuscript, you should quantify the comparison of "IAV" between two datasets.

Please see the response to Question 4 of the reviewer 1.

The revised manuscript included two statistical tables as supplementary materials. Those tables include 1) standard deviations of each time series (representing of IAV), as well as F-test statistics to assess whether there is a significant difference in the IAVs 1) between North America and Europe, 2) between DJF and MAM. The corresponding discussion are added into text along the lines we discussed about seasonal and regional IAV.

**On Page 5, lines 154-156, please describe more clearly how the ozone anomalies are calculated. For instance, for every ozonesonde site, you first calculate the monthly anomalies, and then you calculate the monthly mean of those monthly anomalies for all sites together? What does the 95% confidence interval represents ? The site to site variability with or without the variability within one month at a given site?*

Please see the response to Question 2 of the reviewer 1.

**Coming back to the previous point: quantify the statements on page 6, lines 168-169: "Both observations and simulations show the largest interannual variations in the winter and spring, when the strongest IAVs occur" and on page 6, lines 176-177: "The IAV of ozone is larger over North America then over Europe, and larger in spring than in winter".*

Discussions on IAV comparison have been revised based on statistical analysis.

** In sect 4.1, in which you describe Fig. 4, it should be mentioned that the comparison between ozonesonde data and model simulation decrease with increasing pressure and why this is the case.*

We remove the Figure 4 and corresponding discussion.

** Page 6, lines 184-188: I do not understand the link between the winter polar 150 hPaAVERAGED temperature – 200 hPa O3 IAV correlation and PSC formation, which only happens at very low stratospheric temperatures (< -80_C).*

Please see our response to Question 5 of the reviewer 1.

We deleted our discussion about the relationship between O_3 IAV and temperature at 150 hPa average over latitude north of 60°N. Although they show high correlations, we cannot derive the directly causality without more detailed examinations.

** Page 7, lines 206-209: where do these explained variances come from (in Table 3, only correlations are shown)? Please explain. Same comment for the percentages for the explained variations, mentioned on Page 8, line 234, and page 9, lines 265-267.*

Please see our response to Question 7 of the reviewer 1.

To avoid confusion, we replaced r with r^2 in Table 3 and modified corresponding discussions in the text. We also add the definition of explained variance in the revised manuscript.

** Page 9: why are you using the alternative definition of tropopause pressure by Browell et al. (1996)? Is this tropopause identical to the ozonopause? What is the effect of this choice for the tropopause (compared to the thermal tropopause, as defined by the WMO) on the mentioned correlations with the IAV of O3 and stratO3?*

Bethan et al. (1996) has compare the calculated tropopauses using WMO temperature lapse rate criteria with that defined by the ozone gradient. They demonstrated that it is feasible to define the tropopause in terms of ozone concentration, by identifying the sharp gradient in concentration that occurs at the base of the stratosphere. They also argued that for high latitude in winter, by nearly isothermal profile that could lead to indefinite thermal tropopauses. Another reason is that for ozonesonde data, we did not process its co-measured temperature profile. We therefore used ozone tropopause here.

** Page 12, lines 372-388: the analysis of the correlations between AO and ozone is not very convincing. First of all, please mention the months for which Fig. 12 is constructed (DJF and/or MAM?). Secondly, on which ground do you classify the correlation profiles (with low correlation coefficients after all) in Fig. 12 as significantly different between North America and Europe? And similar between sonde and model data in Figure S2?*

Figure R3 (Figure 11 in the revised manuscript) shows the correlation during the winter season. The caption has been modified. We are arguing the deeper and stronger AO-O_3 coupling over N. America than over Europe. We add dashed lines to indicate regions with statistically significant correlation (df=25, $p<0.05$). Please see below figure.

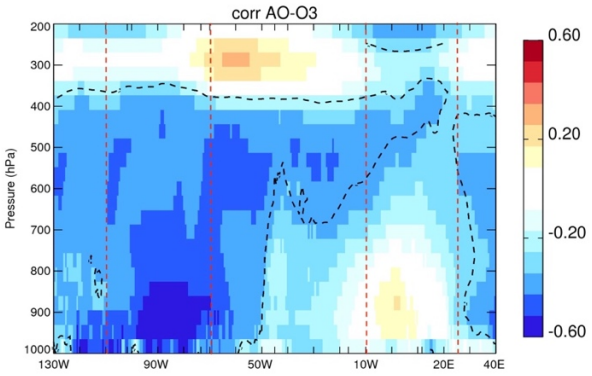

Figure R3: Longitudinal variations of correlation profiles (r) between AO index and simulated O_3 averaged over 30°N and 80°N in DJF from 1000 hPa to 200 hPa. Correlations inside black dashed lines are statistically significant (df=25, $p<0.05$). Red dashed lines indicate the longitudinal range for the North American region (120°W-60°W) and the European region (10°W-26°E).

TECHNICAL CORRECTIONS

** Pag 1, line 29: remove the ‘ after ozonesondes*

The text have been modified as suggested.

** Page 2, line 46-47: replace “In so doing” with “In doing so”.*

The text have been modified as suggested.

** Page 5, before Section 4: Here, you can add that some features in tropospheric ozone are well reproduced (e.g. 2015), while others not (e.g. 2013) and that those differences will be analyzed further in the paper.*

The text has been modified as suggested.

** Page 9, after line 269: please mention here that the longitudinal difference in dynamics between North America and Europe will be further analyzed in Sect. 5.2.*

We move this sentence to the next paragraph to lead the discussion of Figure 6 in the revised manuscript.

** Page 10, line 310: replace “asterisks” by “lines” (referring to Fig. 8)*

The text has been modified as suggested.

** Page 13, line401: replace “resulting” with “result”.*

The text has been modified as suggested.

** Please remove the : in the section titles (e.g. 6: Conclusions and discussion)*

We removed all the : in the section titles.

** Please acknowledge the data repositories properly for the ozone data used (ozonesondes: WOUDC, SBUV, OMI, etc.).*

Below texts are added in the acknowledge:

We thank the World Ozone Data Centre and the SHADOZ program for making the routine sonde data accessible. We gratefully acknowledge Dr. Jerry R Ziemke from NASA for providing the OMI/MLS TCO data and Dr. Stacey M. Frith from NASA for providing SBUV total ozone column data. We thank the reviewers for their helpful comments and suggestions to improve this paper.

Interactive comment by Ryan Williams

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This is an interesting new article on the role of the of the stratosphere on tropospheric ozone interannual variability during Northern Hemisphere winter and spring (when the STE flux is at a maximum). We however feel that our most recent study that looks at the stratospheric influence on tropospheric ozone should additionally be cited within the introduction:

"Characterising the seasonal and geographical variability in tropospheric ozone, stratospheric influence and recent changes" by Ryan S. Williams et al. (2019) (https://www.atmos-chem-phys.net/19/3589/2019/)

We would suggest adding a citation to this paper either on P2, L38: "Stratosphere-troposphere exchange (STE) has been shown to impact the tropospheric ozone distribution (e.g., Terao et al., 2008; Hess et al., 2015; Holton et al., 1995)."

Or alternatively on P2, L50: "STE has been widely studied for several decades (Danielsen, 1968; Holton et al.,1995; Olsen et al., 2002; 2003; 2013; Stohl et al., 2003a; 2003b; Sprenger and Wernli, 2003; Thompson et al., 2007; Lefohn et al., 2011; Skerlak et al., 2014)".

Since our study does not look at STE explicitly (only implicitly using tagged stratospheric ozone tracers from the EMAC and CMAM CCMs), a citation on L38 would be more applicable in our view.

Thanks a lot for the short comments on the references. The reference has been added in the revised manuscript as suggested.

Furthermore, we feel that a mention to nudged, specified-dynamics CCM simulations should be later included in the introduction, in addition to free-running CCM simulations and CTMs (P2-3, L61-72), as a useful tool for assessment of the stratospheric influence on tropospheric ozone (using stratospheric tagged ozone tracers). Compared with free-running CCMs, "the influence on composition of dynamical biases and differences in variability between the reanalysis and the models can be assessed" – Morgenstern et al. (2017), P648 (https://www.geosci-model-dev.net/10/639/2017/). This point could also be made in highlighting the role of constraining the dynamics on influencing the distribution of model composition fields.

Below text is added in the introduction:

Williams et al (2019) used nudged CCM simulations by the ERA-Interim reanalysis dataset and a stratospheric tagged O_3 tracer to assess the role of stratospheric ozone in influencing both regional and seasonal variations of tropospheric O_3. Their study shows that stratosphere has a much larger influence than previously thought, although some differences result from the definition of stratospheric tracer.

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Stratospheric impact on the Northern Hemisphere winter and spring ozone interannual variability in the troposphere

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Abstract. In this study we use O_3 and stratospheric O_3 tracer simulations from the high-resolution (0.5°x0.5°) Goddard Earth Observing System, Version 5 (GEOS-5) Replay run ~~(MERRA-2 GMI at 0.5° model resolution ~ 50 km)~~ and observations from ozonesondes to investigate the interannual variation and vertical extent of the stratospheric ozone impact on tropospheric ozone. Our work focuses on the winter and spring seasons from 1990 to 2016 over North America and Europe. The model reproduces the observed interannual variation of tropospheric O_3, except for the post-Pinatubo period ~~from 1991 to 1995~~ over the region of North America. Ozonesonde data show a negative ozone anomaly in 1992-1994 following the Pinatubo eruption, with recovery thereafter. The simulated anomaly is only half the magnitude of that observed. Our analysis suggests that the simulated Stratosphere-troposphere exchange (STE) flux deduced from the analysis might be too strong over the North American (50°N-70°N) region after the Mt. Pinatubo eruption in the early 1990s, masking the impact of lower stratospheric O_3 concentration on tropospheric O_3. European ozonesonde measurements show a similar but weaker O_3 depletion after the Mt. Pinatubo eruption, which is fully reproduced by the model. Analysis based on the~~a~~ stratospheric O_3 tracer ($StratO_3$) identifies differences in strength and vertical extent of stratospheric ozone impact~~influence~~ on the tropospheric ozone interannual variation (IAV) between North America and Europe. Over North America stations, the $StratO_3$ IAV has a significant impact on tropospheric O_3 from the upper to lower troposphere and explains about 60% and 66% of simulated O_3 IAV at 400 hPa, ~11% and 34% at 700 hPa in winter and spring respectively. Over Europe stations, the influence is limited to the middle to upper troposphere, and becomes much smaller at 700 hPa. The MERRA2 assimilated fields exhibit strong longitudinal variations in meteorology over northern hemisphere (NH) mid-high latitudes, with lower tropopause height and lower geopotential height over North America than Europe. These variations associated with the relevant variations in the location of tropospheric jet flows are responsible for the longitudinal change in the stratospheric O_3 influence and result in a stronger stratospheric O_3 impact on the tropospheric O_3 over North America than over Europe. ~~The stronger and deeper stratospheric contributions in the tropospheric O_3 IAV over North America shown by the model is likely related to ozonesondes' being closer to the polar vortex in winter with lower geopotential height, lower tropopause height, and stronger coupling to the Arctic Oscillation in the lower troposphere (LT) than over Europe.~~

1 Introduction

Tropospheric ozone plays an important role in the oxidative capacity~~chemistry~~ of the troposphere. It is the third most important climate forcing gas after carbon dioxide and methane, and affects the radiative balance of the atmosphere (Forster et al., 2007). Unlike the well-mixed greenhouse gases, tropospheric ozone and its radiative forcing are spatially and temporally inhomogeneous (Lacis et al., 1990; Forster and Shine, 1997; Joiner et al., 2009; Worden et al., 2008; 2011;

Bowman et al., 2013). Stratosphere-troposphere exchange (STE) has been shown to impact the tropospheric ozone distribution (e.g., Holton et al., 1995; Terao et al., 2008; Hess et al., 2015; Williams et al., 2019). Liu et al. (2017) showed that stratospheric ozone input plays a dominant role in driving the interannual variation (IAV) of upper tropospheric ozone over the southern hemisphere ocean, where its radiative impact is largest. Considering the observed and expected net global decrease in emissions of ozone precursors and the predicted increase in ozone STE (e.g., Collins et al., 2003; Sudo et al., 2003; Hardiman et al., 2014; Banerjee et al., 2016), it is important to quantify the role of STE compared to that of precursor emissions in determining tropospheric ozone variations.

In this study we use a long-term ~~Goddard Earth Observing System (GEOS) – chemistry climate model (CCM)~~ replay simulation of O_3 and 'stratospheric ozone tracer diagnostic' ($StratO_3$) by the Goddard Earth Observing System (GEOS) - chemistry climate model (CCM), as well as the ~~model's~~analyzed meteorological fields, to interpret the tropospheric O_3 IAV obtained from the ozonesonde records in the northern hemisphere ~~troposphere at~~ mid-high latitudes. In doing so~~so doing~~, we examine the vertical and longitudinal distribution of the stratospheric O_3 impact on the IAV of tropospheric O_3 and their linkage to dynamical systems~~parameters~~.

STE has been widely studied for several decades (Danielsen, 1968; Holton et al., 1995; Olsen et al., 2002, 2003; 2013; Sprenger and Wernli, 2003; Stohl et al., 2003a; 2003b; Thompson et al., 2007; Lefohn et al., 2011; Skerlak et al., 2014; Williams et al., 2019). It contributes significantly to ozone in the upper troposphere, where ozone has a strong radiative effect. Observations, assimilations and simulations from high resolution models show that deep STE events occasionally reach ground level, adversely affecting the air quality near the surface (e.g., Haagenson et al., 1981; Davies and Schuepbach, 1994; Lefohn et al., 2001; Lin et al., 2012;2014; Ott et al., 2016; Knowland et al., 2017; Akritidis et al., 2018). In addition, various chemistry climate models project increased STE leading to a higher contribution of stratospheric ozone to tropospheric ozone (Sudo et al., 2003; Collins et al., 2003; Zeng et al., 2010; SPARC-CCMVal, 2010). Limitations in the representation of subscale processes lead to large uncertainties in the calculated stratospheric contribution to concentrations and variations of tropospheric ozone. These limitations also lead to uncertainty in their relative magnitudes compared to the effects of increased or decreased emissions of ozone precursors. The uncertainties in stratospheric contribution to tropospheric ozone variations lead to similar uncertainties in resulting ozone radiative forcing, a key area of focus in climate change studies.

Various studies have used tropospheric chemistry transport models (CTMs) to examine the response of tropospheric ozone to changes in stratospheric input and in surface emissions; these models used a simple treatment of stratospheric-tropospheric flux, adopting either the SYNOZ (synthetic ozone) approximation developed by McLinden et al. (2000) to specify the stratosphere-to-troposphere flux (e.g., the GEOS-Chem model in Hess and Zbinden, 2013;Fusco and Logan, 2003), or specifying ozone in the lower stratosphere (LS) (the GISS model in Fusco and Logan, 2003; Karlsdottir et al., 2000). Hess et al. (2015) analyzed the effects of stratospheric input to tropospheric ozone variations over the northern hemisphere mid-latitudes with four ensemble simulations of the free running Whole Atmosphere Community Climate Model (WACCM) for 1953 to 2005. Their model used a standard stratospheric chemical mechanism and simple CH_4-NO_X chemistry in the troposphere with constant surface emissions of ozone precursors. The study reproduced well the observed tropospheric O_3 IAV, suggesting that natural variability in transport and stratospheric ozone plays a significant role in the tropospheric ozone IAV over the northern hemisphere. Williams et al (2019) used a nudged CCM simulations by the ERA-Interim reanalysis dataset and a stratospheric tagged O_3 tracer to assess the role of stratospheric ozone in influencing both regional and seasonal variations of tropospheric O_3. Their study shows that stratosphere has a much larger influence than previously thought, although some differences result from the definition of stratospheric tracer.

In this study, we use a long-term full chemistry ~~O_3~~ simulation with a ~~and an online~~ stratospheric O_3 tracer ~~simulation~~ with a horizontal resolution of 0.5°, suggested to be the minimum model resolution needed to resolve the structure of deep STE events (Ott et al., 2016). We focus on 1990 - 2016, a period of considerable IAV in STE (James et al., 2003), varied trends in

emissions of ozone precursors, and greater availability of reliable ozone observations than in prior periods. We examine the vertical extents of STE impact on tropospheric ozone using model simulations and ozonesonde measurements sampled over North America and Europe. We rely on the $StratO_3$ tracer simulation to quantify the contribution of stratospheric O_3 to tropospheric O_3 at different levels, as well as its contribution to the IAV.

2 Data and Model

2.1 Ozonesondes

We select 17 ozonesonde sites including eight from North America and nine from Europe, all of which have a record of at least 3 profiles every month between 1990 and 2016 (Figure 1 and Tables 1 & 2). The data are obtained from the World Ozone and Ultraviolet Data Center (WOUDC, http://www.woudc.org). Observations over most stations were obtained using electrochemical concentration cell (ECC) ozonesondes, which rely on the oxidation reaction of ozone with potassium iodide in solution (Komhyr et al., 1995). At Hohenpeissenberg, Germany, observations were obtained using the Brewer/Mast ozonesonde. The sondes profiles have a vertical resolution of ~150 m for ozone, with an accuracy about ±5% in the troposphere (WMO, 2014).

2.2 MERRA2-GMI

We use a replay simulation (http://acd-ext.gsfc.nasa.gov/Projects/GEOSCCM/MERRA2GMI) ~~(Orbe et al., 2017)~~ of the GEOSCCM with the Global Modeling Initiative (GMI) chemical mechanism (Duncan et al., 2007; Strahan et al., 2007) for trace gas chemistry, which includes a complete treatment of stratospheric and tropospheric chemistry, and the Goddard Chemistry Aerosol Radiation and Transport (GOCART) module (Chin et al., 2002; Colarco et al., 2010) for aerosols. The replay simulation follows the replay methodology as described in Orbe et al. (2017) and uses the RAs3 setting, which best represents overall transport. The model reads ~~ingests~~ in the three-hourly time-averaged ~~essential~~ output of MERRA-2 meteorology (U, V, T, pressure)~~ every 3 h, and~~ and recomputes the analysis increments, which are used as a forcing to the meteorology at every time step over the 3 h replay interval. ~~then utilizes the model physics and chemistry to calculate the evolution of meteorology and chemical composition from the ingested analysis and the species concentrations at the end of the previous replay interval at every model time step~~More detailed information on replay methodology can be found in Orbe et al.~~,~~ (2017). The replay simulation is run at a MERRA-2 native resolution of ~50 km in the horizontal dimension and 72 vertical levels. ~~(http://acd-ext.gsfc.nasa.gov/Projects/GEOSCCM/MERRA2GMI).~~ This replay simulation is referred to as the 'MERRA2-GMI" simulation.

The MERRA2-GMI simulation was run from 1980 to 2018~~present~~. The emissions in this run include anthropogenic, biofuel, biomass burning, and biogenic emissions. The values for fossil fuel and biofuel emissions are from the MACCity inventory (Granier et al., 2011) until 2010, and then derived by following the Representative Concentration Pathway (RCP) 8.5 scenario after 2010. The MACCity anthropogenic emissions are derived by interpolating the Atmospheric Chemistry and Climate - Model Intercomparison Project (ACCMIP) emissions (Lamarque et al., 2010) on a yearly basis between the base years 1990, 2000, 2005 and 2010. For the years 2005 and 2010, the interpolation follows the RCP 8.5 emission scenario. Biomass burning emissions are from the Global Fire Emissions Dataset (GFED) version 4s (Giglio et al., 2013) after 1997. Prior to 1997, biomass burning emissions are based on a GFED4s climatology with year-to-year variability imposed using regional scale factors derived from the Total Ozone Mapping Spectrometer (TOMS) aerosol index (Duncan et al., 2003). The simulation used the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006) to simulate biogenic emissions, including isoprene, within the model. The lightning parameterization in the model (2010) is constrained by the MERRA-2 detrended cumulative mass flux, with seasonal constraints from the Lightning Imaging Sensor (LIS) / Optical Transient Detector (OTD) v2.3 climatology (Cecil et al., 2014). Methane is specified using latitude and time

dependent surface observations from the NOAA Earth System Research Laboratory (ESRL) Global Monitoring Division (GMD) network (Dlugokencky et al., 2011).

A StratO$_3$ tracer is included in the model to track the stratospheric O$_3$ influence on the troposphere. ~~simulate O$_3$ of stratospheric origin in the troposphere at all locations and times.~~ StratO$_3$ is set equal to simulated O$_3$ in the stratosphere and is removed in the troposphere based on interannually-varying monthly mean loss rates and surface deposition fluxes archived from the standard full chemistry simulation, thus diagnosing the relative importance of stratospheric ozone at all locations in the troposphere. StratO$_3$ tracer is defined relative to a dynamically varying tropopause tracer (e90) (Prather et al., 2011). ~~The StratO$_3$ tracer is defined relative to a dynamically varying tropopause tracer (e90) .~~ The e90 tracer is set to a uniform mixing ratio (100 ppb) at the surface with a 90-day e-folding lifetime everywhere in the atmosphere. This lifetime is long enough for the tracer to be well mixed throughout the troposphere but short compared to the transport time scales in the stratosphere, resulting in sharp e90 gradients across the tropopause. In our simulations, the e90 tropopause value is set to 90 ppb. The e90 tracer has been used in many studies of STE as an accurate tropopause definition and an ideal transport tracer in UTLS (e.g., Hsu and Prather, 2014; Liu et al., 2016; 2017; Pan et al., 2016; Randel et al., 2016). ~~StratO$_3$ is set equal to O$_3$ in the stratosphere and is removed in the troposphere using chemical loss and deposition output from the standard full chemistry simulation.~~ The MERRA2-GMI simulation has hourly output for ozone and three-hourly output for StratO$_3$ tracer at each model level. When comparing to the ozonesonde measurements, the model outputs are sampled at the nearest grid point and launch time for each sonde.

3 Model evaluation with satellite observations

We first evaluate the overall model performance by comparing the simulated total column O$_3$ with the version 8.6 merged total ozone datasets from the Solar Backscatter Ultraviolet (SBUV) (McPeters et al., 2013; Frith et al., 2014). These evaluations support the suitability of the model to then examine the IAV impact of stratospheric ozone. The SBUV observing system measures concentrations at different levels from the ground to the top of the atmosphere, with a vertical resolution changing from 6 km resolution in the middle and upper stratosphere to about 15 km in the troposphere. The SBUV v8.6 merged total ozone dataset are monthly-mean zonal and gridded average products from 1970 to 2017 constructed by combining individual data sets of ozone from a series of SBUV instruments – the Nimbus 4 BUV, the Nimbus 7 SBUV, and SBUV/2 instruments (https://acd-ext.gsfc.nasa.gov/Data_services/merged/). Figure 2 compares the zonal mean of the simulated and observed total ozone columns (TOZ) averaged over 30°N-60°N and 60°N-75°N as well as their anomalies from 1991 to 2017. The anomalies are calculated by removing the monthly mean averaged from 1991 to 2017. The model reproduces the magnitude and seasonal cycle of the observed total column ozone over the mid-high latitudes of the northern hemisphere. Although the model has a low bias compared to the observations, the discrepancy is not statistically different from zero. The model reproduces the observed IAV well, showing more positive ozone anomalies in 2000s and negative ozone anomalies in early 1990s.

We then compare the tropospheric O$_3$ column between model and values derived from a combination of measurements from the Aura Ozone Monitoring Instrument (OMI) and Microwave Limb Sounder (MLS) for January 2005 to December 2016 (Ziemke et al., 2019). The OMI/MLS tropospheric ozone column (TCO) is determined by subtracting the MLS stratospheric ozone column (SCO) from OMI total column ozone each day at each grid point from 60°S to 60°N. The tropopause pressure is defined using the World Meteorological Organization (WMO) 2K-km^{-1} lapse-rate definition from the National Centers for Environmental Prediction (NCEP) re-analyses. The data set has included a +2 DU offset correction and a -0.5 DU/decade drift correction following evaluation with ozonesondes, cloud slicing measurements, and the OMI row anomaly. More detailed description of this dataset is given in Ziemke et al. (2019). We select the same definition of the tropopause pressure for the model simulation to calculate tropospheric column ozone in the model.

Figure 3 shows the tropospheric ozone columns and their anomalies from the MERRA2-GMI replay simulation, together with the OMI/MLS TCO averaged between 30°N and 60°N. The anomalies are calculated relative to their respective 2005 to 2017 monthly mean. The MERRA2-GMI simulation reproduces well the phase of observed seasonal cycles, but underestimates slightly the observed summer maxima (r = 0.93, Figure 3a). The correlation between the OMI/MLS TCO and the MERRA2-GMI TCO decreases ~~decreases from 0.93 for the ozone concentrations~~ to 0.67 for the anomalies. Model did not reproduce the observed anomalies in year 2013 and 2016, but in general model and observations show similar magnitude, IAV and trend, with more negative anomalies before 2009, followed by a continuous increase. Thus, the MERRA2-GMI replay simulation results are in good agreement with the seasonality and IAV of the total and tropospheric column ozone from satellite observations.

4~~.~~ Results

~~4.1 O_3 IAV for all sondes and seasons.~~

~~The monthly means of observed (black) and simulated (red) O_3 anomalies averaged over the selected seventeen ozonesonde sites at 200 hPa, 400 hPa and 700 hPa are shown in Figure 4. Anomalies are smoothed with a 6-month running mean to highlight the interannual variations.The shaded area represents the 95% confidence interval (CI) of the observed mean. The simulated anomalies are calculated from the output at the grid point nearest the ozonesonde site on the date of the measurement. Anomalies are smoothed with a 6-month running mean to highlight the interannual variations.~~

~~In the lower stratosphere (200 hPa), both the phase and magnitude of the simulated anomalies agree very well with that derived from observations (r = 0.93). For example, both observation and simulation show a strong negative O_3 anomaly of about 166 ppb with ~ 39% relative change in February 1993 after the Pinatubo eruption. Both show strong positive anomalies in the winter-spring seasons of 1991, 2013, and some years after strong El Ninos, e.g., 1998-1999, 2001-2004, 2009-2010, with an increased stratospheric circulation (e.g., Neu et al., 2014).~~

~~In the troposphere, the model is in reasonable agreement with the phase of observed anomalies, but tends to underestimate the magnitude. For example, the model underestimates the observed O_3 depletion after the Mt. Pinatubo eruption at 400 hPa (relative anomalies of -14% in measurements vs. -8% in model) and 700 hPa (-13% vs. -5%), as well as the observed maximum O_3 increase in February 2013 at 400 hPa, and in December 2003 at 700 hPa. Still, most of model results fall within the 95% CI of observations.~~

~~(e.g., Holton et al., 1995;Stohl et al., 2000;Skerlak et al., 2014;2015;e.g., Lin et al., 2012;2015)~~

~~Both observations and simulations show the largest interannual variations in the winter and spring, when the strongest IAVs occur. In the next section, we examine time series for these two individual seasons over North America and Europe and investigate their controlling factors.~~

4.12 Winter and spring O_3 IAV in the lower stratosphere and troposphere over North American and European sites

Previous studies have shown that the impact of STE is greatest in the free troposphere during winter (e.g., Holton et al., 1995; Stohl et al., 2000; Skerlak et al., 2014; 2015) and at the surface during spring (e.g., Lin et al., 2012; 2015). In summer, the impact of stratospheric ozone to the troposphere is relatively weaker due to the increased chemical ozone production during that season. Here, we focus on the winter (DJF) and spring (MAM) seasons to examine the interannual variations of the strength and vertical extent of stratospheric ozone impact on the tropospheric ozone.

Figure 45 compares the anomalies of modeled and observed ozone at 200 hPa, 400 hPa and 700 hPa in the winter and spring seasons from 1990 to 2016 averaged over sites from North America and Europe. Anomalies at each site are calculated by removing the respective seasonal mean climatology from 1995 to 2016, and then averaged over all sites for each region. The shaded area represents the 95% CI of the calculated mean from daily observations over all the selected stations. To quantify

the magnitude of IAVs, we calculate the standard deviations of these O_3 anomalies. We also perform the statistical F-test to assess the regional and seasonal variations of O_3 IAVs. The calculated standard deviations and F-test statistics are shown in Table S1 and S2.

At 200 hPa, the model reproduces very well the observed IAV in both seasons over both regions ($r \geq 0.91$, Figure 4 a-d). There is not significant regional difference in the magnitude of O_3 IAV between North America and Europe (Table S1). But over both regions, the O_3 IAV show significant seasonal difference with greater magnitude in spring than in winter (Table S2). ~~The IAV of O_3 is larger over North America than over Europe, and larger in spring than in winter.~~ Negative O_3 anomalies occur in the early 1990s and at the end of the record from 2014 to 2016, while positive anomalies are obtained for most years between 1998 and 2013. The negative ozone anomalies during the period of 1992-1996 are consistent with the chemical and dynamical perturbations following the June 15th 1991 eruption of Mt. Pinatubo (Hadjinicolaou et al., 1997; Stenchikov et al., 2002;Rozanov et al., 2002). The negative ozone anomaly in 2015-2016 is associated with stratospheric circulation changes caused by the unusually warm ENSO event aligned with a disrupted Quasi-Biennial Oscillation (QBO) during that period (Tweedy et al., 2017; Diallo et al., 2018)

~~Both observed and simulated O_3 IAV at 200 hPa are correlated with the temperature at 150 hPa averaged over latitudes north of 60°N ($r > 0.7$ over North America and $r \geq 0.55$ over Europe, Figure S1) in the winter. The correlation is weaker in the spring. These results are consistent with our understanding of the impact of temperature variations on the formation of polar stratospheric clouds and polar vortex isolation with reduced transport of O_3 from the tropics at low temperatures (e.g., Schoeberl and Hartmann, 1991), all of which modulate ozone concentrations in the lower stratosphere during winter/spring. The IAV at 400 hPa over North American sites is similar to those at 200 hPa (Figure 5 e-f).~~ At 400 hPa, ~~The~~the model reproduces the overall variations as inferred from observations, showing negative anomalies in early 1990s, with mostly positive anomalies thereafter. The observed O_3 depletion after the Mt. Pinatubo eruption reaches its maximum amplitude of 7 ppb (-13% relative anomaly) in the winter 1992-1994. The model underestimates the observed peak depletion in winter of 1992, with the simulations falling outside the 95% CI of the observations from 1992 to 1994 (~~Figure 5~~Figure 4 e). ~~I~~In spring, the model reproduces well the timing of observed O_3 depletion, but again underestimates its amplitude (~~Figure 5~~Figure 4 f). At 700 hPa, the observations from the North American sites show a similar negative ozone anomaly in 1992-1994 to that obtained at 200 hPa and 400 hPa, with prevailing positive anomalies thereafter. The model results for the sign of the interannual variations are in relatively good agreement with observations, but again underestimate the magnitudes of the negative anomalies in the early 1990s after the Mt. Pinatubo eruption.

Over European sites, the observed O_3 IAV, excluding year 1990-1991, exhibits a similar pattern to the one at 200 hPa after 1991, although the minima after the Mt. Pinatubo eruption are not as pronounced as over North America (Figure 4 g-h). The maximum positive anomaly, observed in 1990-1991, is not reproduced by the model. The model-observation correlation coefficients increase significantly if we omit these two years (from 0.18 to 0.58 in the winter and from 0.43 to 0.58 in the spring). At 700 hPa, ~~Over the European sites, unlike that over North America,~~ the model reproduces the amplitude of observed O_3 depletion after the Mt. Pinatubo eruption and shows similar magnitude of observed variations. Unlike at 200 hPa, the regional difference of the magnitude of O_3 IAV is significant at 400 hPa and 700 hPa, with smaller O_3 IAV over European sites than over North American sites in both seasons (Table S1). While the seasonal difference between DJF and MAM in magnitude of O_3 IAV is small, only become significant over North America at 400 hPa (Table S2).

We use explained variances (r^2) to assess the strength of the relationship between O_3 IAV in the lower stratosphere and in the troposphere. Table 3 shows the ~~correlation coefficients~~explained variances of the winter and spring ozone IAVs between 200 hPa and 400 hPa, 200 hPa and 700 hPa for the observations and simulations averaged over the North American and European stations. Both the model and observations suggest that about 27% of North American interannual ozone variability at 400 hPa is accounted for by~~related to~~ changes at 200 hPa in the winter. The explained variance is higher in the spring with 40% and 46% respectively in the observation and simulation. Over Europe, the 200-400 hPa O_3 relationship~~correlation~~ in the

observation is relatively low (r^2 = 0.131 in DJF and 0.0213 in MAM), due to the phase shift of these two-time series during the first two years, where observed O_3 anomalies are negative at 200 hPa, but reach a maximum at 400 hPa. The explained variance correlation increases to 0.4567 after removing these two years in the winter, but not that much in the spring (r^2 = 0 = 0.0523). High correlations of the O_3 IAV between 200 hPa and 400 hPa is seen in the model in both seasons. The highest relationshipcorrelation coefficient between 200 hPa and 700 hPa is found over the North American sites in the spring with r^2 = 0.2146 & 0.1741 respectively in observations and simulations, which is consistent with the previous findings of the deep STE hot spots along western U.S. in the spring season (Skerlak et al., 2014).

The correlations analysis between the stratosphere and troposphere IAV in both observations and model simulations suggest a potential impact of stratospheric O_3 on tropospheric O_3 variations. Previous studies have found high correlations between ozone in the lower stratosphere with that in the middle and lower troposphere with the largest effects in late winter and spring. Correlation does not necessarily mean causality, and to date model investigations of this correlation (Terao et al., 2008; Hess and Zbinden, 2013) did not use a model with both stratospheric and tropospheric chemistry, and realistic stratospheric circulation. The MERRA2-GMI simulation has both of these attributes, detailed dynamic diagnostics, and the StratO_3 tracer as described in Section 2.2. In the next section, we use the StratO_3 tracer to examine the contribution of stratospheric ozone to the IAV of tropospheric ozone, as a function of altitude, season, and location. We will also use diagnostics from the model to explore the influence of dynamics on the stratospheric O_3 contribution to the tropospheric O_3 and its IAV.

4.23: Impact of stratospheric O_3 on tropospheric O_3 IAV

Figure 6Figure 5 shows the same comparison between the observed (black lines) and simulated ozone (red lines) anomalies as in Figure 5Figure 4, but adding the anomalies of simulated StratO_3 tracer (green lines). As expected, the StratO_3 tracer anomalies at 200 hPa are almost identical to the simulated O_3 anomalies, since most measurements are in the stratosphere at this level.

The IAV of StratO_3 tracer in the troposphere reflects a combined effect of the changes in the lower stratospheric O_3 concentrations and in the strength of stratosphere-to- troposphere (STE) mass flux. These two effects may either cancel or reinforce each other, depending on their relative phases. At 400 hPa, over the North American stations, the minimum and maximum of StratO_3 tracer is highly correlated with the minimum and maximum of simulated O_3. The IAV of StratO_3 explains more than 60% of simulated O_3 variations (Figure 5 e, f, r = 0.77 in DJF and 0.81 in MAM), suggesting that the changes of stratospheric O_3 input strongly impact provides a significant influence on the simulated O_3 IAV in the upper troposphere. The correlation between StratO_3 and observed O_3 is slightly lower (0.44) than that with simulated O_3 in DJF over North America. The decreased correlation is mainly due to the model-observation discrepancy between 1992-1994. The sondes at 400 hPa show a similar O_3 depletion through 1992-1994 as seen at 200 hPa after the Mt. Pinatubo eruption, while the model shows an O_3 increase after 1992 through 1994, which is driven by changes in the stratospheric O_3 contribution to the modeled O_3 (Figure 6Figure 5e). This suggests that the impact of the negative anomalies of stratospheric ozone (200 hPa) may be counterbalanced by an increase in downward mass flux from the stratosphere, thus leading to the model underestimation of the negative anomaly in observations at 400 hPa. In MAM, the StratO_3-O_3 correlation to the observed O_3 stays high (0.74) over North America. Over European sites, a similar correlation is observed between simulated O_3 and StratO_3 at 400 hPa in the winter (r = 0.78), with a slightly smaller value in the spring (r=0.61). The correlation decreases when comparing StratO_3 to the observed O_3, mainly because of the model-observation discrepancy during the first two years. Omitting the first two year gives a fair correlation between StratO_3 and observed O_3 (0.66 in DJF and 0.34 in MAM). The fair to good correlations between StratO_3 and observed O_3 give credence to the reality of the impact of stratospheric ozone on the troposphere. In general, the good agreement between ozone IAV with that of StratO_3 at 400 hPa indicates that

changes in the stratospheric ozone contribution play an important role in the simulated upper tropospheric O_3 IAV in winter-spring over North America and Europe.

The bottom panel of ~~Figure 6~~Figure 5 compares the simulated StratO_3 tracer anomalies to the observed and simulated O_3 anomalies at 700 hPa over North American and European ozonesonde sites in the winter and spring seasons. Over North America, the observed O_3 anomalies stay low in the early 1990s, and increase thereafter in both seasons, which is underestimated in the model. In the winter, StratO_3 anomalies decrease slightly in contrast to increases in both observed and simulated O_3 anomalies. The winter StratO_3-O_3 correlation is ~ 0. In spring, sonde and model exhibit similar IAV of O_3 and are similar to the phase of the IAV of the StratO_3 after the Pinatubo period (1991-1995). The StratO_3-O_3 correlation increases from 0.07 to 0.33 in winter and from 0.36 to 0.58 in spring from 1996 to 2016~~after omitting the Pinatubo period~~.

Over North America, our model results are in good agreement with the observed IAV at all levels except right after the Mt. Pinatubo eruption. The model only reproduces about half of the observed tropospheric depletion over North America. As discussed above, this ~~is likely~~could be due to an excessive mass flux from the stratosphere in the MERRA-2 analysis during this period. Model results are in better agreement with the magnitude of observed O_3 depletion after the Mt. Pinatubo eruption in the middle and lower troposphere over Europe. There is no significant relationship between StratO_3 and simulated O_3 at 700 hPa. This is expected since the impact of stratospheric ozone decreases, and the impact of ozone production from its precursors becomes more important at lower altitudes. ~~We also note that the anomalies in StratO_3 diverge from those of simulated O_3 towards the end of our analysis period, particularly at 700 hPa. This is to be expected since the impact of stratospheric ozone decreases at lower altitudes, and the impact of ozone production from its precursors becomes more important.~~ In summary, our model analysis identifies differences in the strength and vertical extent of stratospheric ozone impact on the tropospheric ozone IAV between North America and Europe. Over North America, the StratO_3 IAV has a significant impact on the tropospheric O_3 IAV from the upper to lower troposphere and explains about 60% and 66% of the simulated O_3 IAV at 400 hPa, ~11% and 34% at 700 hPa in winter and spring respectively, after 1995. Over Europe, the influence is limited to the middle to upper troposphere, and becomes much less at 700 hPa.

The difference in the stratospheric O_3 influence between North America and Europe is likely due to ~~the~~ longitudinal difference in dynamics. Previous studies have suggested that the IAV of the STE mass flux is likely correlated to changes in the tropopause height (e.g., Gettelman et al., 2011). The top panel of ~~Figure 7~~Figure 6 shows the comparison of the observed O_3 mixing ratio anomalies at 400 hPa and the tropopause pressures derived from the observed O_3 profiles following the criteria in vertical gradient and O_3 mixing ratio given by Browell et al. (Browell et al., 1996). The tropopause pressure was estimated to be at the pressure where a linear regression line passing through the lower stratospheric O_3 profile (150 ppb - 400 ppb, lower than 100 hPa) intersects with the 100 ppb O_3 level. The bottom panel of ~~Figure 7~~Figure 6 compares the simulated O_3 and StratO_3 anomalies at 400 hPa with the tropopause pressures derived from simulated O_3 profiles following the same criteria as for the observations. As expected, the IAV of O_3 and StratO_3 positively correlates with that of the derived tropopause pressure (anticorrelates with the tropopause height) in both model and observation. In general, during years with a lower tropopause, stratospheric O_3 influence at 400 hPa increases and results in a positive O_3 anomaly. During years with a higher tropopause, decreased stratospheric O_3 influence leads to a negative O_3 anomaly at 400 hPa.

The above high correlations between the IAV of tropopause pressure and StratO_3 raise the question of what dynamical conditions control the higher/lower tropopause pressures, STE mass fluxes, and the subsequent impact of stratospheric ozone on tropospheric ozone. These questions are particularly important if these dynamical conditions may exhibit future changes as a result of climate change. In the next section, we rely on the model's 3-d dynamical diagnostics, including air mass flux and horizontal wind patterns, to examine both the vertical and horizontal transport influence of the stratospheric O_3 contribution on the tropospheric O_3 and its IAV. We also examine the longitudinal difference in the model's dynamics to explain the identified longitudinal difference in stratospheric O_3 influence in the troposphere between North America and Europe.

5 Influence of dynamics

5.1 Case study of 3-d dynamic characteristics

~~The p~~Planetary-scale Rossby waves, including quasi-stationary Rossby waves and Rossby wave-breaking events, superimposed on the mean westerly zonal flow are the dominant dynamical variability over northern midlatitudes in winter and spring. Homeyer and Bowman (2013) have shown that Rossby wave-breaking occurring in the upper troposphere can affect the flow at all tropospheric levels and plays an important role in the meridional transport of both tropical and subtropical air masses. Ozone transport associated with these wave disturbances are responsible for a large fraction of ozone temporal and spatial variability in winter and spring (e.g., Kinnersley and Tung, 1998; McCormack et al., 1998; Lozitsky et al., 2011; Zhang et al., 2015). Thorncroft et al. (1993) classified Rossby wave-breaking events as either "equatorward breaking" or "poleward breaking". In the equatorward breaking, tongues of high PV and stratospheric air extend equatorward associated with frequent STE processes. In the poleward breaking, tongues of low PV and upper tropospheric air extend poleward.~~In the equatorward breaking, tongues of high PV, stratospheric air extend equatorward associated with frequent STE processes, while, in the latter case, tongues of low PV, upper tropospheric air extend poleward.~~

In this section, we rely on the model's 3-d dynamic diagnostics, including air mass flux and horizontal wind patterns, to examine both vertical and horizontal transport influence of the stratospheric O_3 contribution to the tropospheric O_3 and its IAV. By doing that, we examine the linkages of the dynamical structures at the lower stratosphere to the stratospheric O_3 contributions in the upper and middle troposphere and how they vary with the changes in wave disturbances year by year. Our analysis first focuses on the year 1993, when there is a major discrepancy with the observations at 400 hPa as shown in Figure 5.

~~Figure 8~~Figure 7 illustrates the relationship ~~corresponding changes~~ of $Strat O_3/O_3$ ratio at 400 hPa to ~~changes in~~ dynamics including horizontal winds at 400 hPa, and vertical airmass flux near the seasonal mean tropopause pressure in the year 1993. Because of the different tropopause heights different pressure levels are shown in the figures. The tropopause pressure in the model averaged from 30°N to 80°N is around 250 hPa in winter and around 300 hPa in spring. The vertical airmass flux is calculated by multiplying ω (the volume flow rate, which depends on the pressure difference) with the density of air. The top row of Figure 7 shows maps of simulated $Strat O_3/O_3$ ratio in the winter and spring of 1993; prevailing wind patterns at 400 hPa are superimposed on this ratio. The jet locations, approximated by the strongest winds, are indicated by red thick lines~~asterisks~~ in the top row of Figure 7. The second row of Figure 7 shows the respective anomalies of simulated $Strat O_3/O_3$. The third row shows the airmass flux around the tropopause pressure with 250 hPa in winter and 300 hPa in spring (Blue color represents the downward motion and red color represents the upward motion near the tropopause) and the fourth row shows the anomalies of airmass flux at the same pressure (Blue color represents an increase of downward flux or a decrease of upward flux. Red color represents a decrease of downward flux or an increase of upward flux around the tropopause).

$Strat O_3/O_3$ ratio represents the impact of stratospheric air on tropospheric ozone at this level. Regions with the maximum $Strat O_3/O_3$ ratio at 400 hPa in general show a similar longitudinal distribution in the winter and spring seasons with a southward shift over eastern North America and a poleward shift over western North America and Europe. However, there are year by year variations in this longitudinal distribution of the $Strat O_3/O_3$ ratios, associated with the IAV of wave disturbances in the westerlies.

In the winter of 1993, strong northwesterly winds prevailed north of 50°N and the westerlies dominated between 30°N and 50°N over western North America. The winds converged around 45°N over eastern North America and moved northwestward into the North Atlantic and Europe. The winds changed direction to northwesterly over Europe, bringing higher stratospheric O_3 air into eastern Europe (Figure 7a). The maps of the airmass flux and its anomalies (Figure 7e, g) suggest that North America between 50°N and 70°N was dominated by more vigorous downward mass fluxes of

stratospheric air. Meanwhile, the southeasterly winds brought ozone rich air from high latitudes, resulting in a positive anomaly of stratospheric ozone influence at 400 hPa (Figure 7c). Our results suggest that although the stratospheric O_3 depletion modulated this process, the enhanced STE in the model counteracted the depletion and reduced the negative anomalies expected at 400 hPa over North American between 50°N and 70°N (Figure S12). Over the high latitudes (> 70°N), where there is less dynamic perturbation (including both vertical and horizontal transport), the stratospheric O_3 contribution at 400 hPa was largely driven by the depletion of the O_3 concentration in the lower stratosphere and showed a strong negative anomaly in 1993. Although most of the European region was covered by the increased downward airmass flux at 200 hPa in the winter of 1993, a negative anomaly of the StratO_3 contribution at 400 hPa was seen over western Europe. It is likely that the combined negative effects of the O_3 depletion in the lower stratosphere and the downwind of the low stratO_3 air from the subtropical North Atlantic Ocean exceeded the positive effect of the increased downward airmass flux over this region.

In the spring of 1993, southwesterly wind prevailed south of 65°N over western North America, bringing in low StratO_3 oceanic air from the subtropics. The wind was deflected to the south around 120°W and 65°N and flowed to Hudson Bay around 60°N, then transported to the east until reaching the west coast of Europe, where the winds bifurcated into two branches: one passed by the northern side of Europe and the other flowed around the southern side of Europe. In North America south of 50°N, there were three cells with increased upward airmass fluxes ranging from 110°W to 50°W, resulting in a decreased StratO_3 contribution in the downwind regions (Figure 7d, f, h). Most regions of western North America north of 50°N showed a decreased StratO_3 contribution, likely contributed jointly by the lower stratospheric O_3 and the decreased STE flux. Decreased stratospheric O_3 contribution occurred over most of Europe, especially the northwest coast, which was downwind of the westerly flows with low stratospheric O_3.

Figure 8 shows the similar analysis as Figure 7, except for 1998, when stratospheric O_3 levels have recovered from the Mt. Pinatubo eruption and reached a regional maximum (Figure 4). In the winter of 1998, a poleward shift of the jet occurred over most of North America. The jet location as well as the regions with the maximum StratO_3/O_3 ratio moved to the north by about 7° compared to the winter of 1993. Strong southwesterly winds combined with increased ascending air dominated over western North America between 45°N and 70°N and brought in tropical oceanic low O_3 air. Over middle and eastern North America, weakened descending air resulted in a minimum of stratospheric O_3 influence over these regions. Therefore, although there was an increase in the stratospheric O_3 concentrations, the weaker STE flux associated with the northward movement of the jet system over North America produced only a small O_3 variation at 400 hPa.

Our analysis suggests that the IAV of wave disturbances in the westerlies likely affect the IAV of the regional distributions of prevailing wind patterns as well as the strength of STE flux. The IAV of stratospheric O_3 influence in the troposphere reflects a combined effect of the changes in the lower stratospheric O_3 concentration and in the 3-d dynamics, which may either cancel or reinforce each other.

5.2 Longitudinal difference of stratospheric O_3 influence

Our ~~model~~ analysis based on data and model sampled at sonde sites, identified differences in the strength and vertical extent of stratospheric ozone impact on tropospheric ozone IAV between North America and Europe. Over North American sites, the significant impact of the StratO_3 IAV ~~has a significant impact~~ on tropospheric O_3 reach to the~~from the upper to~~ lower troposphere. Over European sites, the influence is limited to the middle to upper troposphere. In this section, we examine whether the longitudinal difference as seen in ozonesonde sites is a large-scale phenomenon and relevant to longitudinal dynamical variations, by extending our analysis to a broader spatial domain. ~~The difference of stratospheric O_3 impact between North America and Europe is caused by the longitudinal differences in dynamics.~~

Figure 9 and 10 show the latitudinal average (30°N to 80°N) of tropopause pressure, geopotential height at 400 hPa, and the StratO_3/O_3 ratio at 400 hPa at each longitude from 180°W to 180°E from 1990 to 2016 in winter and spring. The

geopotential heights and tropopause pressure are good representors of large-scale circulation patterns. All of them show strong longitudinal difference between North America (120°W-60°W) and Europe (10°W-26°E), with lower geopotential height, higher tropopause pressure (lower tropopause height), and greater stratospheric O_3 contribution over North America than Europe. The longitudinal gradients between North America and Europe are slightly weaker in spring than in winter. The spatial map of $Strat O_3/O_3$ climatology at 400 hPa averaged from 1990 to 2016 suggests that the longitudinal difference is persistent over most mid-high latitudes (Figure S2), which is closely related to the wavelike pattern in jets. The climatology of jet meanders to the south over central and eastern N. America and brings in cold polar air with more stratospheric subsidence. The jet moves to the north over Europe and brings in warm air with less stratospheric O_3 influence. Skerlak et al. (2014) identified the deep STE hot spots along the western North America using the ERA-Interim reanalysis data set from the European Centre for Medium-Range Weather Forecasts (ECMWF) from 1979 to 2011. Therefore, over North America, the stratospheric subsidence inside the polar vortex as well as deep stratospheric intrusion events results in a deeper and greater stratospheric O_3 influence on the tropospheric O_3 than ~~that~~ over Europe.

A modulating factor in the IAV is ~~In terms of IAV, we refer to~~ the Artic Oscillation (AO) – the primary mode of IAV in the troposphere during winter. Several studies have examined the mechanism for downward transport from the stratosphere to the troposphere and attributed changes in the strength of lower-stratospheric polar vortex to AO anomalies at the surface, with a positive AO phase linked to a more isolated and stronger polar vortex (Ambaum and Hoskins, 2002;Perlwitz and Harnik, 2003) and lower tropopause heights. Lamarque and Hess (2004) found that the AO explains up to 50% of the IAV in tropospheric ozone over North America in January-March, but did not find any significant correlation in European sonde data, with similar results from the Model for OZone And Related chemical Tracers (MOZART) model. They argued that the correlation may be caused by the influence of the AO on its modulation of STE as well as transport of O_3 and its precursors. Kivi et al. (2007) found that changes in the AO explained most of the tropospheric ozone trends in January–April, based on analysis of Arctic ozonesonde data. Figure 11 shows the longitudinal variations of simulated O_3 and AO correlation profiles averaged over 30°N and 80°N from 1000 hPa to 200 hPa. Over North America (120°W to 60°W), the correlation between simulated O_3 and the AO index is negative and stays low above 400 hPa. ~~,~~ The anticorrelation increases with increased pressure and reaches its maximum near surface around 90°W. The anticorrelation averaged over Europe (10°W-26°E) stays low above 400 hPa, increases slightly from 400 hPa to 700 hPa, then decreases sharply near the surface. This is similar to the correlations obtained from the ozonesonde profiles (Figure S3). The similarity of correlation patterns over sonde sites and their surrounding broader regions indicates that the AO-related stratospheric subsidence is a large-scale phenomenon and also show a similar longitudinal variation between North America and Europe.

6~~.~~ Conclusions and discussion

In this study we used O_3 and stratospheric O_3 tracer simulations from MERRA-2 GMI and observations from ozonesondes to investigate the interannual variations and vertical extents of stratospheric ozone influence on tropospheric ozone. Our work focuses on the winter and spring seasons over North America and Europe.

The model reproduces the observed interannual variations of tropospheric O_3 in the troposphere over North America except for the Pinatubo period from 1991 to 1995. The ozonesonde data show a negative ozone anomaly in 1992-1994 following the Pinatubo eruption, with recovery thereafter. However, the simulated anomaly is about half the magnitude of the observed tropospheric ozone depletion. Over European regions, ozonesondes show a similar but weaker O_3 depletion, which was fully reproduced by the model. We use a stratospheric ozone tracer to gauge the impact of stratospheric ozone variations in different regions of the troposphere. Our results based on the stratospheric O_3 tracer suggest that the influence of the stratospheric IAV is significant in the middle to lower troposphere over North America, while over Europe, the stratospheric influence is limited to the middle to upper troposphere. Our analysis of the MERRA2 assimilated fields shows strong

longitudinal variations in meteorology over northern hemisphere (NH) mid-high latitudes, with lower tropopause height and lower geopotential height over North America than Europe. These variations associated with the relevant variations in the location of tropospheric jet flows are responsible for the longitudinal change in the stratospheric O_3 influence. The increase in frequency in stratospheric folds near the jets, and the strong winter subsidence inside the polar vortex lead to stronger stratospheric impact over North America than over Europe.

~~Over North America, the stratospheric subsidence insides the polar vortex as well as deep stratospheric intrusion events resulting in a deeper and greater stratospheric O_3 influence on the tropospheric O_3 than over Europe.~~

We examine the linkages of horizontal and vertical dynamical structures in the lower stratosphere to the contributions of stratospheric O_3 in the upper and middle troposphere. Our analysis suggests that the IAV of wave disturbances of the westerlies likely affect the IAV of the regional distributions of prevailing wind patterns as well as the strength of STE flux. The IAV of stratospheric O_3 influence in the troposphere reflects a combined effect of the changes in the lower stratospheric O_3 concentration and in the 3-d dynamics, which may either cancel or reinforce each other, depending on their relative phases.

Our analysis provides an in-depth understanding of how dynamics influences the O_3 redistribution in the troposphere, and reveals deficiencies in the transport produced by the input meteorological fields. The observed O_3 at 400 hPa over the North American sites show a similar O_3 depletion as that at 200 hPa, while in the model, the effect of lower stratospheric O_3 concentration seems masked by increased stratospheric-tropospheric flux, indicated by increased tropopause pressure accompanied by a stronger downward airmass flux in the model, especially between 50°N and 70°N. Therefore, the model underestimation of the observed O_3 depletion after the Mt. Pinatubo eruption over North America in the lower troposphere could be due to the STE flux being too strong in the model for this region during that period. The assimilated MERRA-2 meteorological fields are significantly improved after the year 1998 when many higher-resolution meteorological observations are included in the assimilation (Bosilovich et al., 2015; Stauffer et al., 2019).

Author contributions

JL performed the data and model analysis and wrote the paper, JL and JMR conceived and planned the project and participated in the numerous scientific discussions. LDO performed and provided the MERRA2-GMI simulation. ARD provided insights on interpolation of model and data comparison. LDO and MAO helped on dynamical analysis of model simulations and interpolation of the findings. LH prepared ozonesondes data. All authors provided critical feedback and helped shape the research, analysis and manuscript.

Code/Data availability

All data used for this article can be obtained by contacting J. Liu (email:Junhua.liu@nasa.gov).

Competing interests

The authors declare that they have no conflict of interest.

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Tables

Sonde station	(lat, lon)	Time	Freq (n/mon)
Alert	82.50°N, 62.33°W	1990-2017	4.1
Eureka	79.99°N, 85.94°W	1993-2015	5.5
Resolute	74.72°N, 94.98°W	1980-2017	3.1
Churchill	58.75°N, 94.07°W	1980-2014	3.2
Edmonton	53.55°N, 114.10°W	1980-2017	3.4
GooseBay	53.32°N, 60.30°W	1980-2017	3.8
Boulder	40.00°N, 105.25°W	1980-2017	3.0
Wallops	37.93°N, 75.47°W	1985-2017	3.4

Table 1: The longitude, latitude, measurement time period and mean sampling frequency of the selected north American ozonesonde sites.

Sonde station	(lat, lon)	Time	Freq (n/mon)
Ny-Aleasund	78.93°N, 11.95°E	1991-2013	7.1
Sodankyla	67.39°N, 26.65°E	1989-2007	5.4
Legionowo	52.40°N, 20.97°E	1980-2015	4.1
Lindenberg	52.21°N, 14.12°E	1980-2014	5.0
DeBilt	52.10°N, 5.18°E	1992-2014	4.3
Uccle	50.80°N, 4.35°E	1980-2014	10.8
Hohenpeissenberg	47.80°N, 11°E	1980-2017	10.0
Payerne	46.49°N, 6.57°E	1980-2014	11.2
Madrid	40.47°N, 3.58°W	1995-2015	3.6

Table 2: The longitude, latitude, measurement time period and mean sampling frequency of the selected European ozonesonde sites.

	North American stations (1990-2016)		European stations (1990-2016)		(1992-2015)	
	DJF	MAM	DJF	MAM	DJF	MAM
r^2 (200 hPa - 400 hPa)	0.27 (0.27)	0.41 (0.46)	0.1 (0.5)	0.02 (0.37)	0.45(0.62)	0.05 (0.41)

r^2 (200 hPa - 700 hPa)	0.06 (0.002)	0.21 (0.17)	0.1 (0.01)	0.07 (0.03)	0.18(0.12)	0.15(0.08)

Table 3: Variance explained (r^2) ~~correlation coefficients~~ of ozone between 200 hPa and 400 hPa, 200 hPa and 700 hPa in observations and simulations. The numbers in parentheses are variance explained ~~correlation coefficients~~ for simulations.

Figures

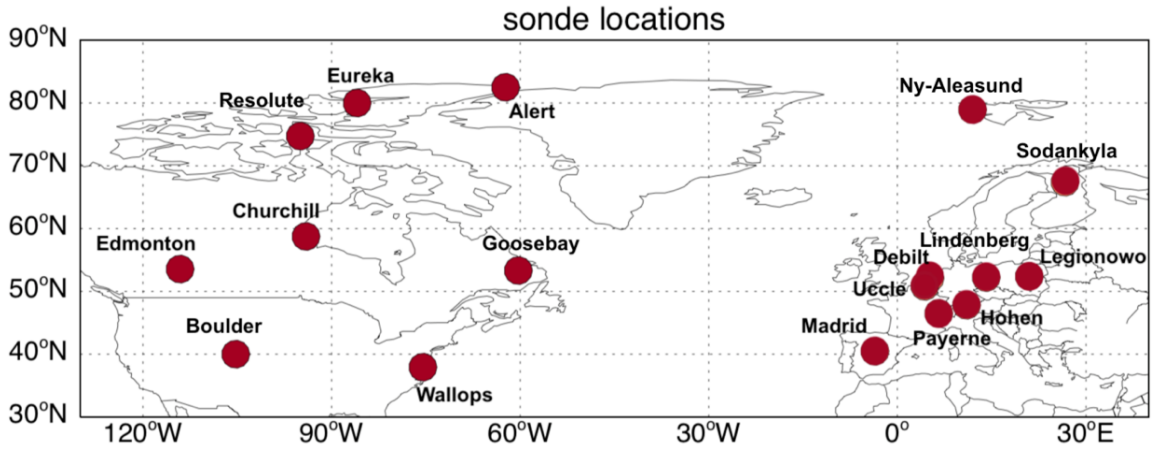


Figure 1: Map of ozonesonde sites selected in this study.

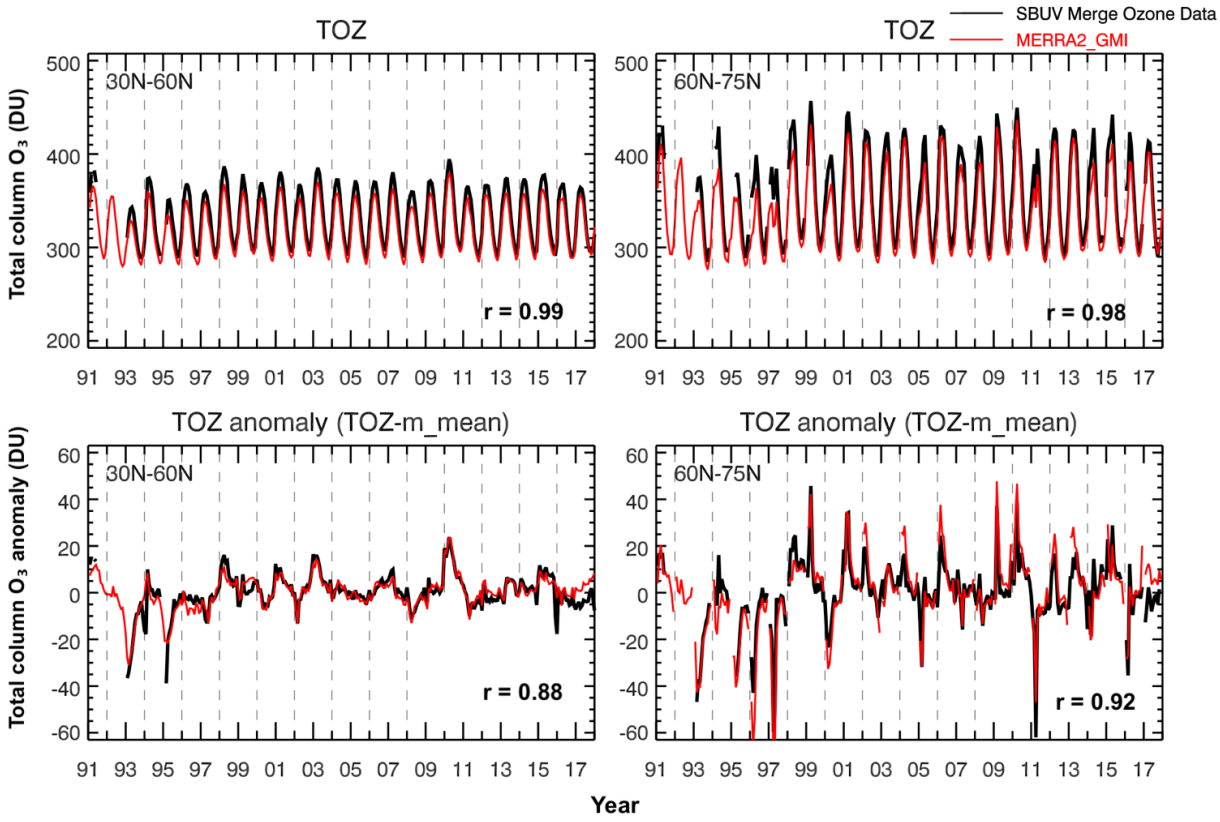


Figure 2: Monthly zonal mean of total column ozone (top) and its anomalies (bottom) averaged over (left) 30°N-60°N and (right) 60°N-75°N from the observations of the SBUV version 8.6 merged total ozone datasets (black lines) and the MERRA2-GMI simulations (red lines) from 1991 to 2016. The anomalies are calculated by removing the monthly mean averaged from 1991 to 2016.

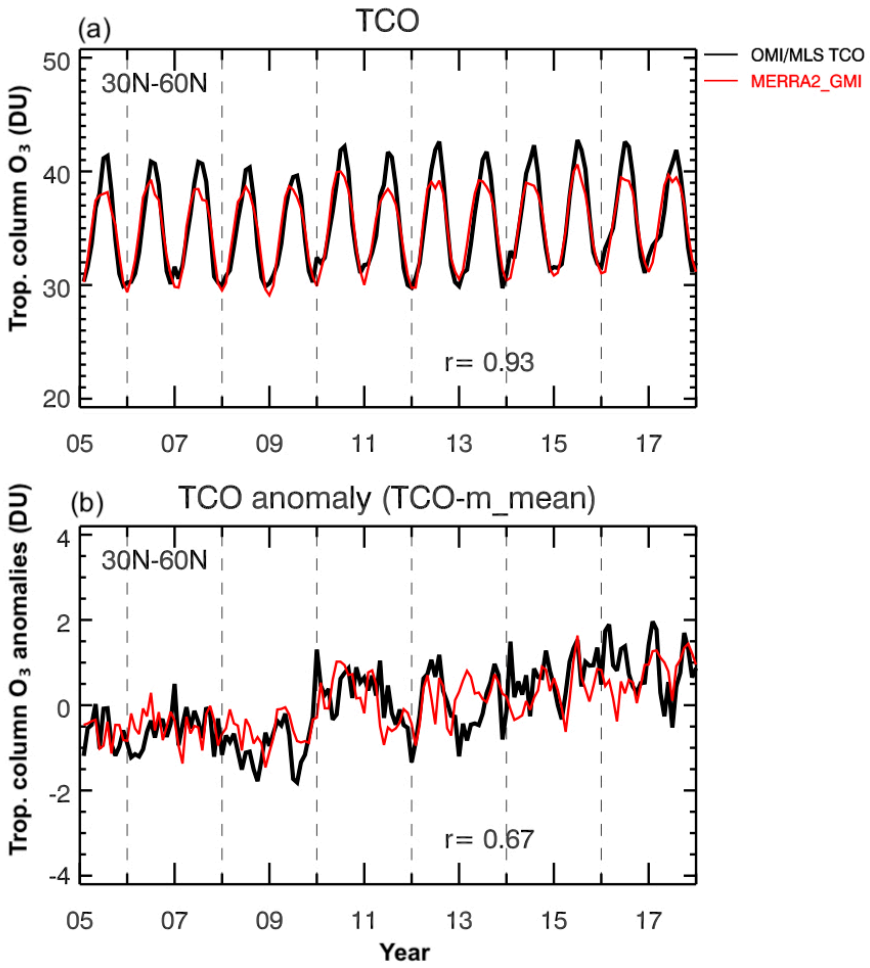
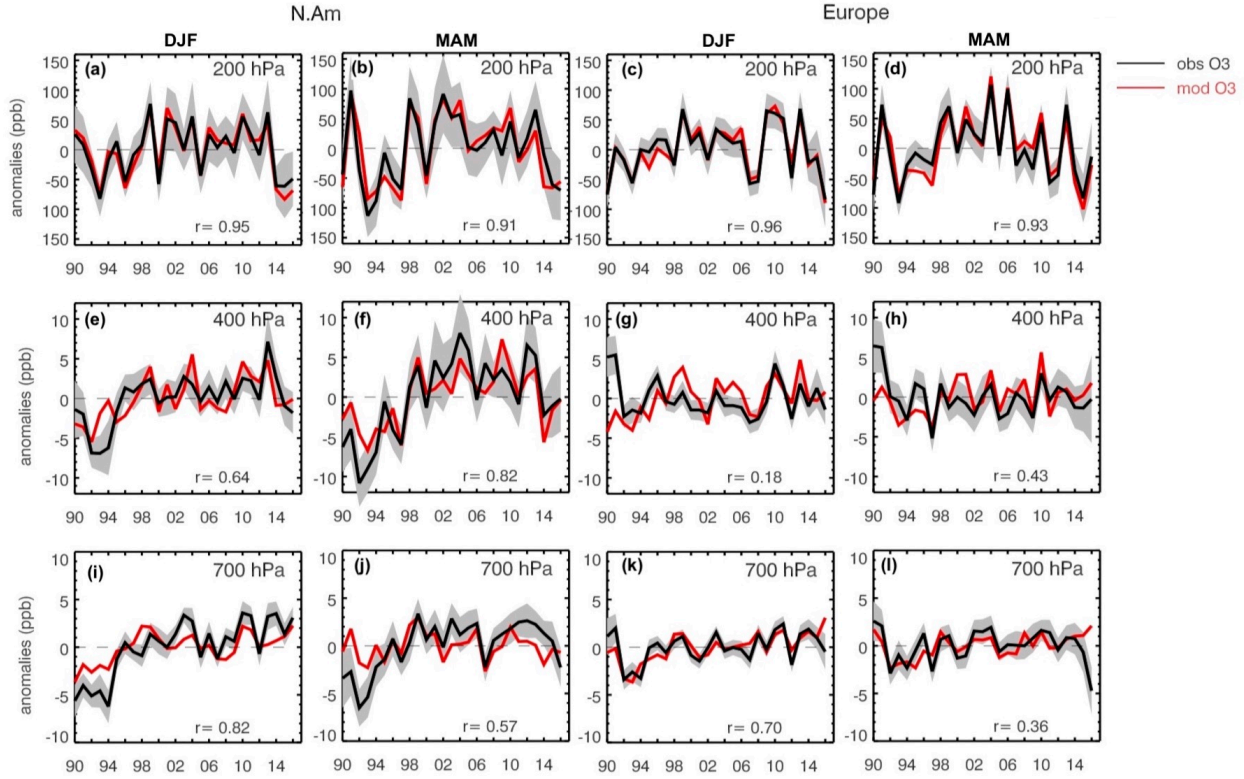
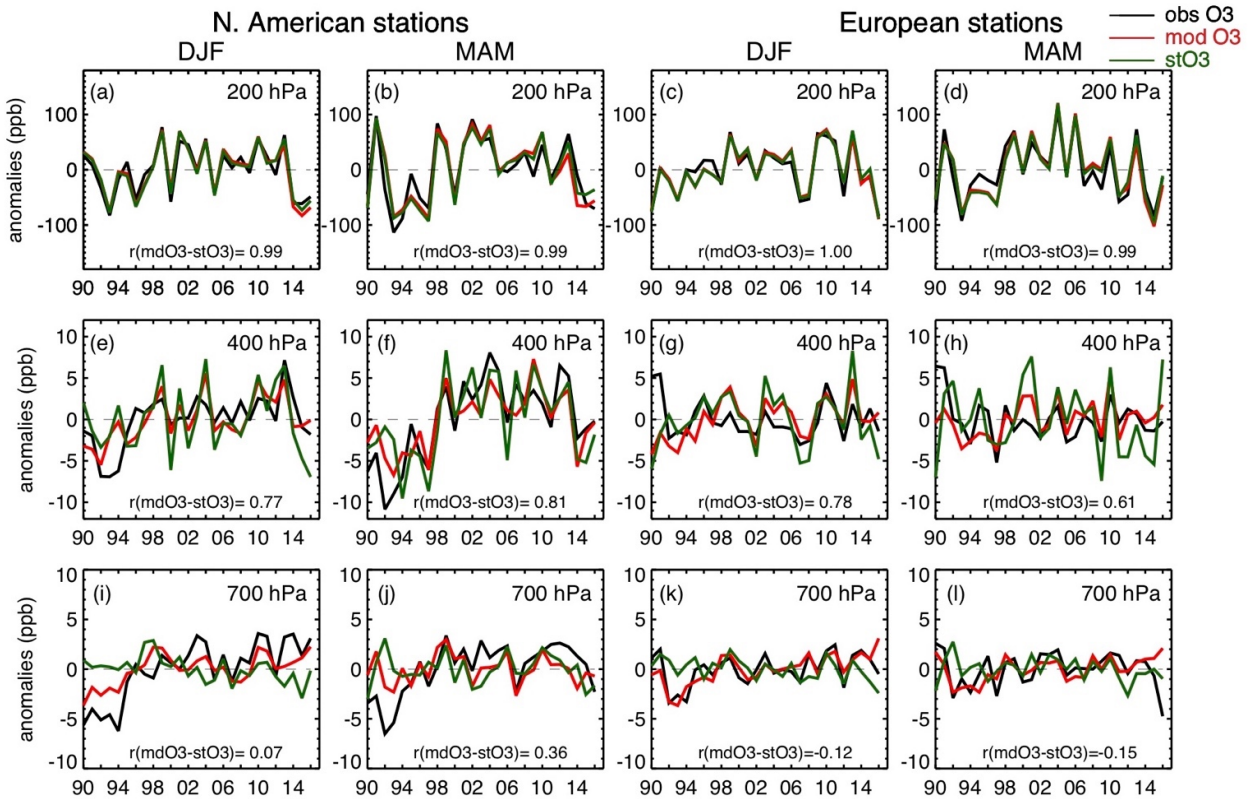


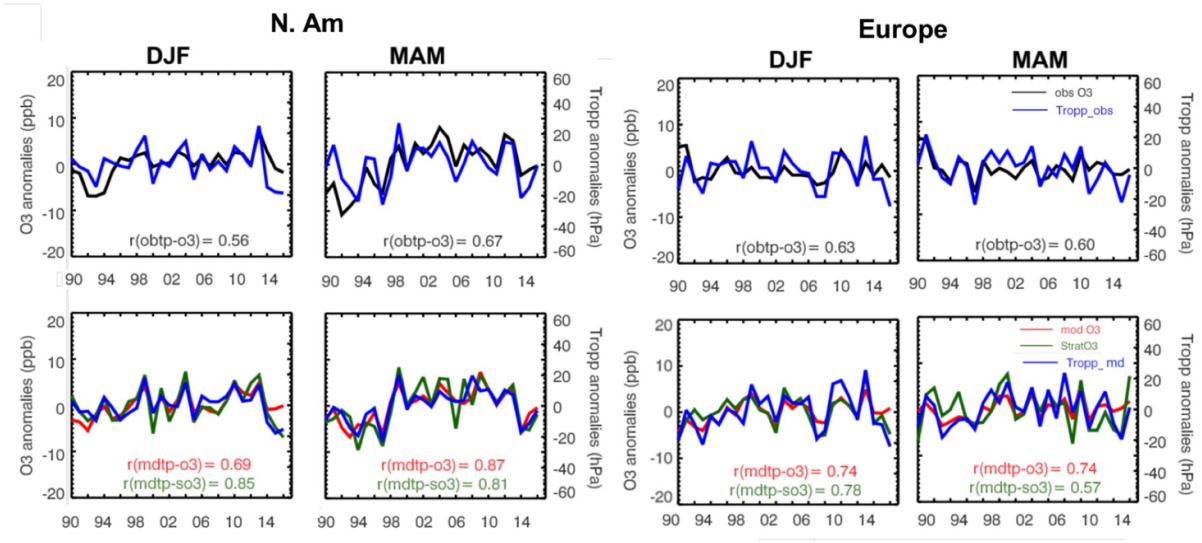
Figure 3: Monthly zonal mean of total column ozone (top) and its anomalies (bottom) averaged over 30°N-60°N from the observations derived from OMI/MLS residual analysis (black lines) and the MERRA2-GMI simulations (red lines) from 2005 to 2017. The anomalies are calculated by removing respective monthly mean averaged from 1991 to 2017.



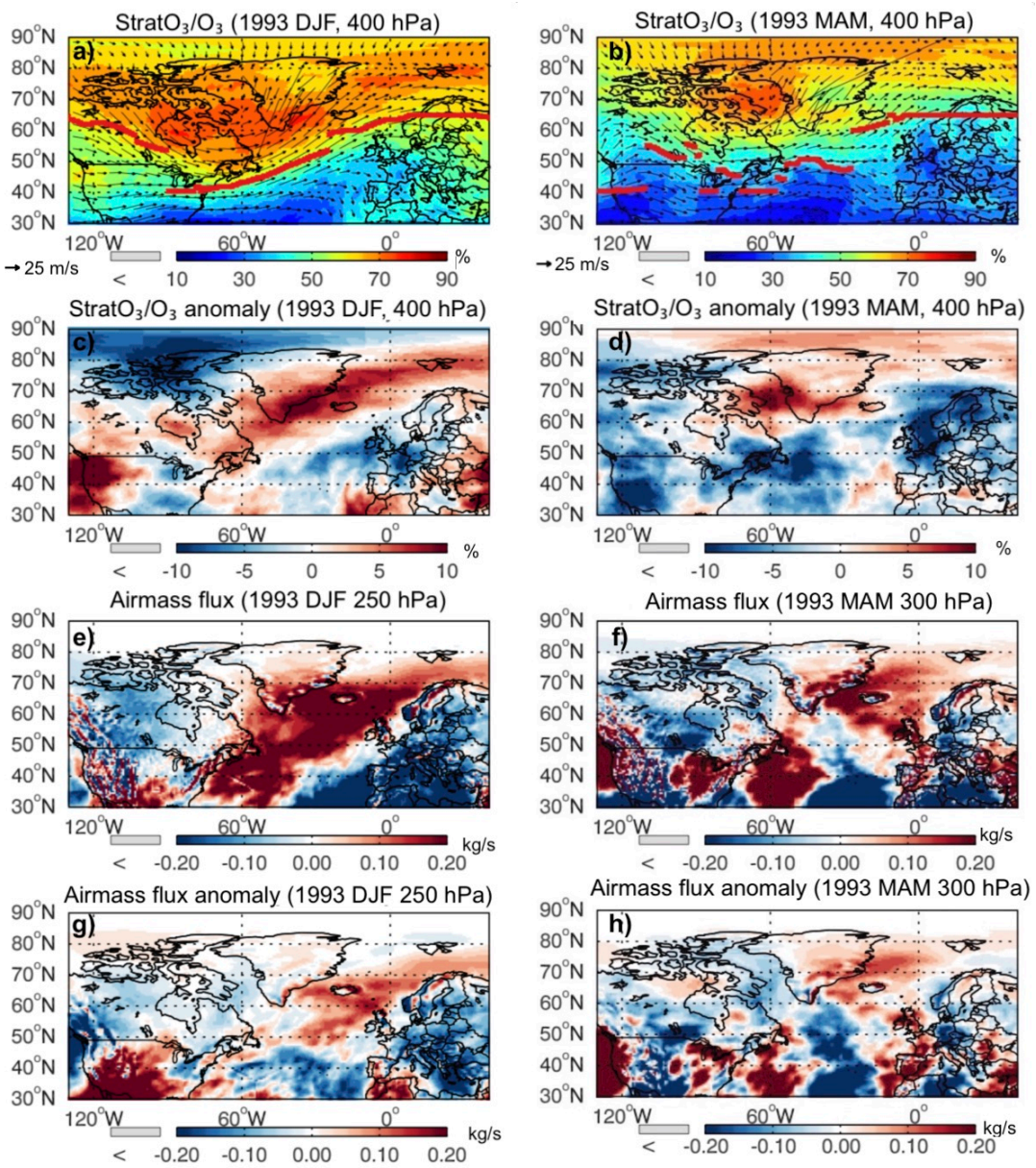
~~Figure 5~~Figure 4: Time series plots of observed (black) and simulated (red) ozone anomalies (unit: ppb) at 200 hPa (top), 400hPa (middle) and 700 hPa (bottom) averaged from selected ozonesonde sites over North America and Europe in winter and spring seasons from 1990 to 2016. The anomalies are calculated by removing the seasonal mean averaged from 1990 to 2016. The shaded area represents the 95% confidence interval (CI) of observed mean, which is calculated by multiplying the standard error of observations by 1.96.



~~Figure 6~~Figure 5: Similar to ~~Figure 5~~Figure 4, but adding the simulated $StratO_3$ anomalies (green). The correlation coefficients between simulated O_3 and $StratO_3$ are shown in text.



~~Figure 7~~Figure 6: **(top) Time series of the observed O_3 mixing ratio anomalies at 400 hPa and the tropopause pressures derived from observed O_3 profiles averaged over the North American and European sites in winter and spring. Their correlation coefficients are shown in black text. (bottom) Time series of the simulated O_3 and StratO_3 anomalies at 400 hPa with the tropopause pressures derived from simulated O_3 profiles, with the respective correlation coefficients shown in red and green text.**



~~Figure 8~~Figure 7: : **Spatial maps of simulated StratO_3/O_3 ratio (1st row) and its anomaly (2nd row) at 400 hPa, Airmass flux (3rd row) and its anomaly (4th row) at 250 hPa in winter (left) and at 300 hPa in spring (right) of 1993.250 hPa and 300 hPa are the closest model pressure levels to the area averaged tropopause pressure between 30°N and 80°N. Black thin arrows in 1st row represents the prevailing wind pattern at 400 hPa. Red thick lines indicate the approximated jet locations, where the strongest winds are.**

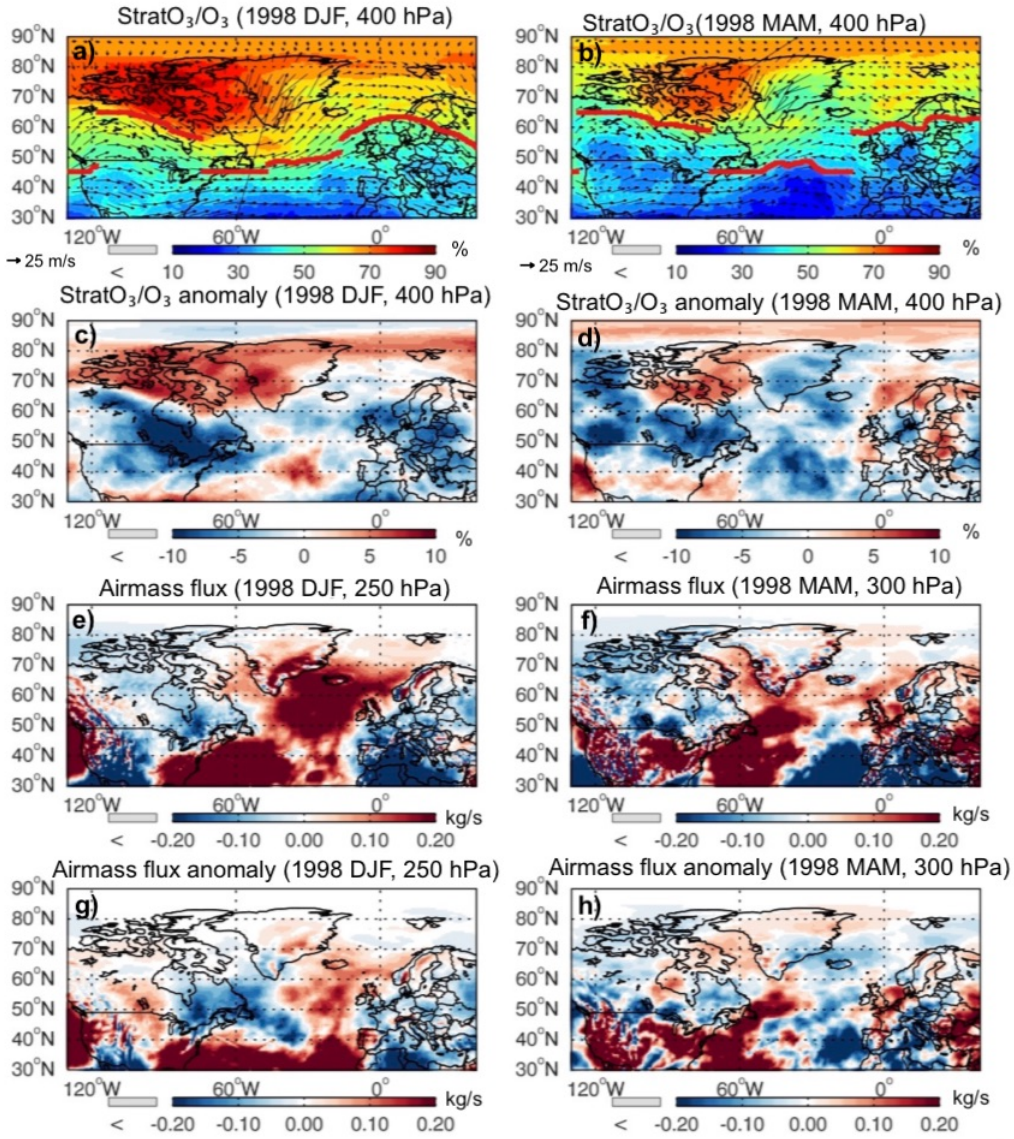
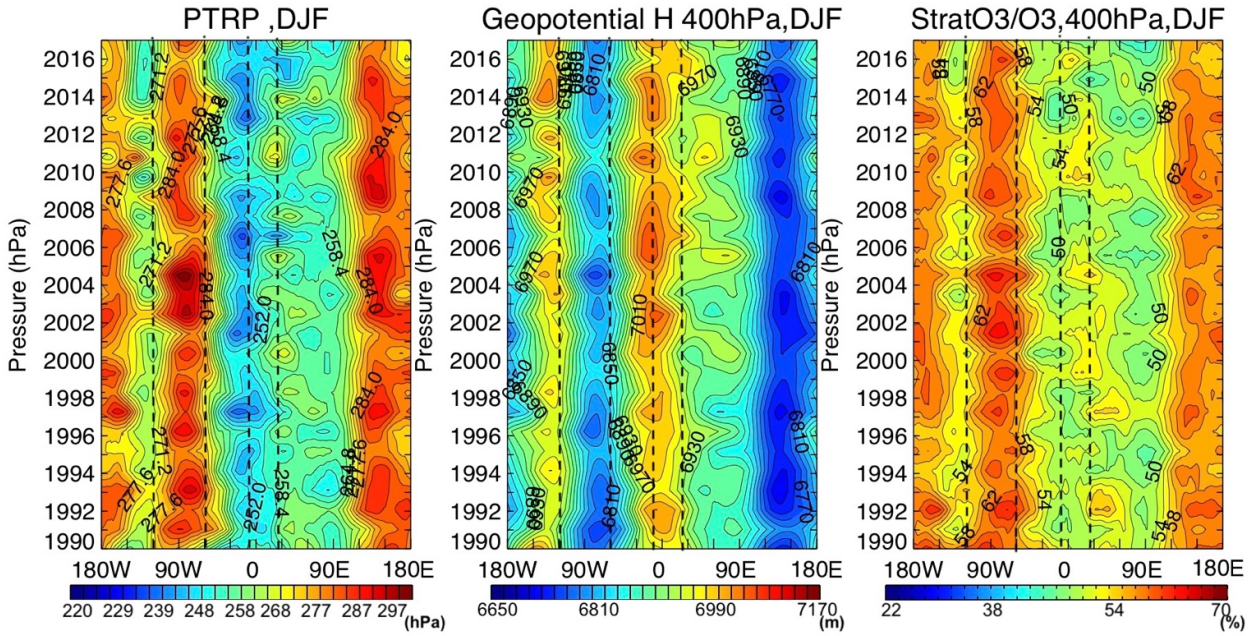
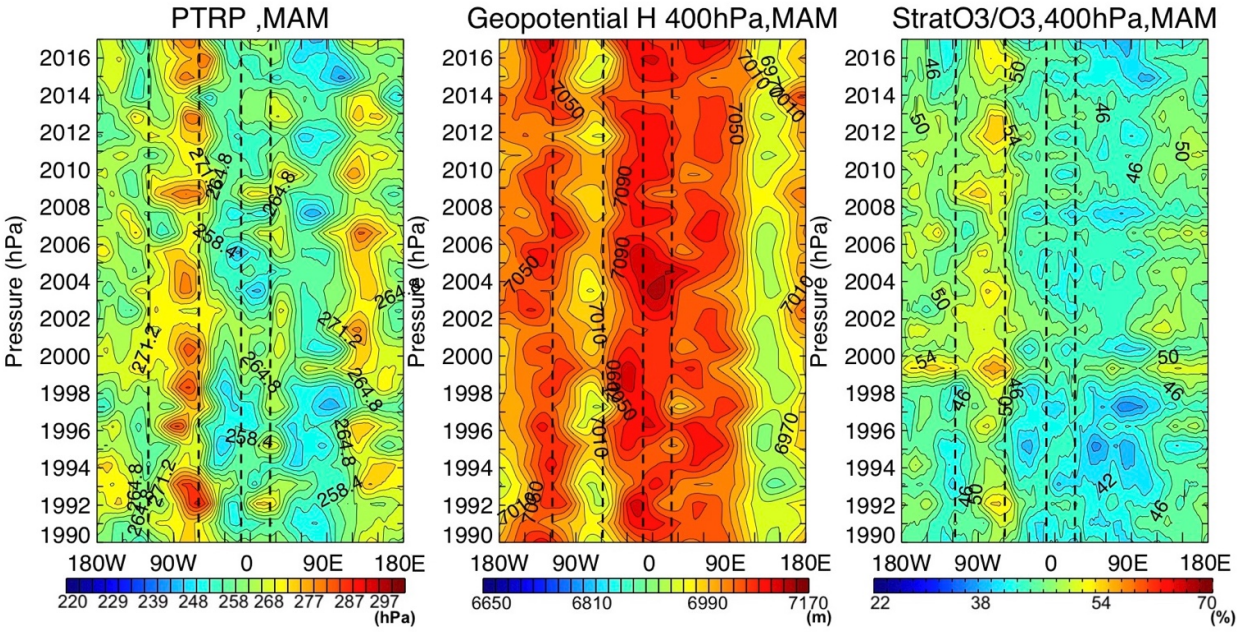


Figure 8: Similar to Figure 7 but for year 1998.



765 **Figure 9: Latitudinal average between 30°N to 80°N of (left) the tropopause pressure; (middle) the geopotential height at 400 hPa; (right) the $Strat O_3/O_3$ ratio at 400 hPa along each longitude from 180°W to 180°E from 1990 to 2016 in winter (DJF). Dashed lines indicate the longitudinal range for the North American region (120°W-60°W) and the European region (10°W-26°E).**



770 **Figure 10: Latitudinal average between 30°N to 80°N of (left) the tropopause pressure; (middle) the geopotential height at 400 hPa; (right) the $Strat O_3/O_3$ ratio at 400 hPa along each longitude from 180°W to 180°E from 1990 to 2016 in spring (MAM). Dashed lines indicate the longitudinal range for the North American region (120°W-60°W) and the European region (10°W-26°E).**

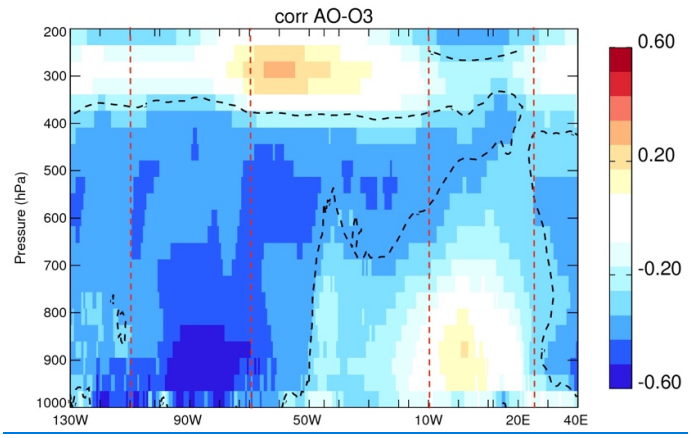


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Figure 11: Longitudinal variations of ~~simulated O_3 and AO~~ correlation profiles (r) between AO index and simulated O_3 averaged over 30°N and 80°N in DJF from 1000 hPa to 200 hPa. Correlations inside black dashed lines are statistically significant (df=25, p<0.05). Red dashed lines indicate the longitudinal range for the North American region (120°W-60°W) and the European region (10°W-26°E).

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