May 10, 2025

Author Comments (egusphere-2024-3719)

Manuscript title: Hemispheric differences in ozone across the stratosphere-troposphere exchange region

Referee #1

The manuscript compares ozone mixing ratios of stratospheric ozone in the UTLS between the northern and southern hemisphere mid-latitudes, and between high and low ozone depletion years. The analysis is mainly based on in-situ measurements (aircraft and ozonesondes), but is complemented with CAMSRA model output for a better spatiotemporal representation.

I first want to thank the authors for following my suggestion to extend the time range of their original study period and to incorporate additional SH ozonesonde sites (Lauder and Macquarie Island). The manuscript is now also better organized and most of my questions for clarifications have been answered.

General comments:

However, I'm still missing somewhat the purpose and focus of the paper. The purpose of the paper and the method followed by achieving this should be mentioned more clearly in the beginning of the manuscript. You describe some different elements of the puzzle in each paragraph of the introduction (STE, available data, SSW), but you do not lay the puzzle yourself by e.g. explicitly linking these phenomena (STE, SSW) with their possible impact on hemispheric ozone differences, why concentrating on mid-latitudes in this study, why it is important to discriminate between high and low ozone depleting years in this study. You assume that the reader will see the puzzle during the course of the paper. Also in the introduction, I'm missing some insight on what the manuscript wants to add to the current knowledge?

Therefore, 2 suggestions, for the introduction:

- add what is already known about UTLS ozone mixing ratio differences between the NH and SH, and what is known already about the impact of high/low ozone depletion in the stratosphere on UTLS ozone.
- Add a true roadmap for your study and give a short explanation for each step (instead of the paragraph from lines 85 to 96), for instance:
  - We want to study hemispheric ozone differences in the UTLS at mid-latitudes during spring. Why in the UTLS? Why at mid-latitudes? Why during springtime? Why are such possible hemispheric ozone differences important?
  - We will only look at the UTLS ozone of stratospheric origin. Therefore, in our analysis, we make a distinction between high and low stratospheric ozone depleting events/years.

<u>**Answer**</u>: We appreciate the suggestions, and we modified the last paragraph of the introduction accordingly. The text: *"Given the multiple dynamical and chemical processes that influence ozone in the UTLS (Millán et al., 2024; Bourgeois et al., 2020; Neu et al., 2014; Riese et al., 2012) and the interhemispheric differences in processes such as the magnitude of the stratospheric ozone depletion and frequency of SSW events, this study, investigates hemispheric differences in UTLS ozone in air masses of*

stratospheric origin, with focus on mid-latitudes, where STE plays a dominant role in determining ozone levels. We also leverage the increased ozone abundance under low-depletion conditions derived from SSW events to determine the intrahemispheric UTLS ozone differences. Through this comparative analysis, we aim to quantify the influence of two processes, i.e., stratospheric ozone depletion and SSW, on the ozone UTLS, where this species is an important radiative forcer. Our analysis utilized stratospheric and tropospheric chemical tracers measured during the SouthTRAC mission, by IAGOS commercial aircraft and by ozonesondes. Spatial coverage was further enhanced using the Copernicus Atmosphere Monitoring Service reanalysis (CAMSRA), which we compared against in situ measurements..."

Regarding the point suggesting adding what is known about hemispheric differences to the best of our knowledge, we have included relevant references to build our case. However, we apologize in advance if we are missing some key papers or review papers.

Specific comments:

- After 2 Data and before 2.1 SouthTRAC data, write a short introductory paragraph in the style of "The UTLS ozone measurements used in this study are available from (research + commercial) aircraft and ozonesondes and are complemented with chemical reanalysis vertical ozoneprofiles. To determine the stratospheric or tropospheric origin of the ozone data, we used water vapour (or humidity) measurements from the aircraft and radiosondes coupled to the ozonesondes and from the chemical reanalysis, and additionally CO, HNO3, HCl measurement from some of the aircraft data. In the next section, we give more details on these datasets.

Answer: We appreciate the suggestion and accordingly we added in the main text: *"The UTLS ozone measurements used in this study are available from (research & commercial) aircraft and ozonesondes. These in situ measurements were complemented with vertical ozone profiles obtained from chemical reanalysis. To determine the stratospheric or tropospheric origin of the ozone data, we used water vapor (or humidity) measurements from the aircraft and radiosondes coupled to the ozonesondes and from the chemical reanalysis in addition to carbon monoxide (CO), nitric acid (HNO_3) and hydrogen chloride (HCl) measurements from some of the aircraft data. In the next sections, we provide more details on these datasets".*

- Caption Fig. 1: mention explicitly for which time period the number of observations is shown here.

Answer: We added: "*SouthTRAC shows data from 4 Sep to 20 Nov 2019 and IAGOS between 4 Mar and 20 May from 2002 to 2022."*

- Again, after 3 Method and before 3.1 Study period & UTLS definition, it is important to provide some guidance to the reader. Therefore, write a short introductory paragraph in the style of "With the data available and described in the previous section, we will now describe how we will analyze springtime UTLS ozone differences at the mid-latitudes between both hemispheres. We first describe how our study period and the used UTLS definition in 3.1, mention how we distinguish between high and low stratospheric ozone depletion years in 3.2, and we show how we ascertain the stratospheric origin of the analyzed UTLS ozone concentrations in 3.3."

Answer: We added: "*In this section we describe how we used the data available to analyze springtime UTLS ozone differences at the mid-latitudes between both hemispheres. We first describe our study period and the used UTLS definition (section 3.1), then we mention how we distinguish between high and low stratospheric ozone depletion years (section 3.2) and show how we ascertain the stratospheric origin of the analyzed UTLS ozone mixing ratios (section 3.3).*"

- Section 3.1 misses a real focus. The first two lines (176-177) belong to the SouthTRAC data description. Define the UTLS (300-200 hPa) and the free troposphere (700-300 hPa) clearly. Mention that your study focus on the springtime only, and already define this periods (4 Mar –20 May, NH & 4 Sep – 20 Nov, SH) here.

Answer: We modified the beginning of 3.1 as follows: *In this study, we denominate the UTLS as the segment between 200 (~12 km) and 300 (~9 km) hPa, while the free troposphere is the segment between 700 to 300 hPa. We focus on the period between late-winter and mid-spring in both hemispheres: 4 Sep–20 Nov (SH) and 4 Mar–20 May (NH).*

- Section 3.2 and Table 2: Here, some major clarifications are needed. Low ozone depletion years are defined by the presence of a SSW event (last column of Table 2). However, for most NH years, the SSW central date lies well ahead of the study period (Mar – May), so it is not clear of the SSW event still occurs during the study period. If this is not the case, it should be mentioned what the expected impact of a SSW event earlier that year would be on the UTLS springtime ozone concentrations. To me, a more direct distinction between high and low ozone depletion years could be made by simply looking at the stratospheric (or total) ozone amounts, averaged over the 45-60° latitude bands, for the 4 Mar – 20 May (NH) or 4 Sep – 20 Nov (SH) period.

Answer: In the first revision, we were asked to include an objective criterion for the selection of high and low ozone depletion years, so we decided to choose the most commonly used metric to detect SSWs. In this metric, the central (onset) date is the first day on which the daily mean zonal mean zonal wind at 60°N and 10 hPa is easterly. Despite the metric to detect SSWs, in the NH, SSWs lead to higher ozone in springtime. See for, example, Figure 7 from *Bouillon, M., Safieddine, S., & Clerbaux, C. (2023). Sudden stratospheric warmings in the Northern Hemisphere observed with IASI. Journal of Geophysical Research: Atmospheres, 128, e2023JD038692. https://doi.org/10.1029/2023JD038692).*

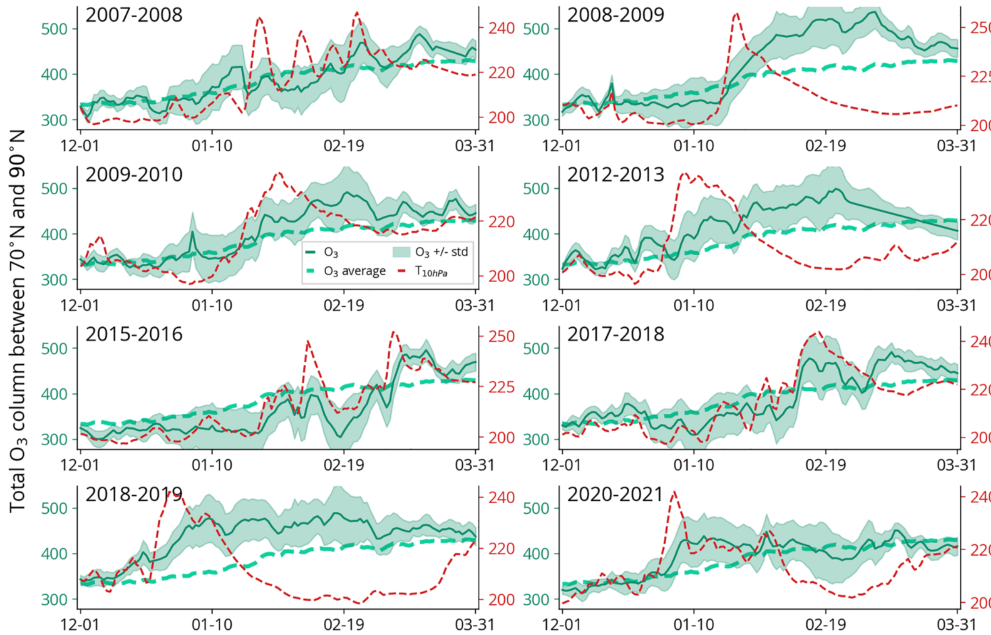


Figure 7. Daily average total ozone column during the eight major SSW winters (dark green, with the standard deviation shown around it) compared to average of all winters from 2007–2008 to 2020–2021 (dashed light green). The daily average temperature at 10 hPa is shown in red.

- Table 2: add a column with NH and SH to the left, add either "IAGOS" or "SouthTRAC" before flights and add that these latter flights only occurred in 2019. Also, I don't understand why the total number of IAGOS flights in the NH for the two periods (later winter-early spring & midspring), resp. 456 and 835 flights, is not equal to the number of IAGOS flights in Table 1 (6315).

Answer: We added the corresponding database to the number of flights. We also corrected the number of IAGOS flights and now the total is 6315 flights, as indicated in Table 1. Regarding adding new columns, we prefer not to do that since the information is sufficiently clear. We also believe it is redundant to indicate in the table that SouthTRAC occurred in 2019 as this has been clearly stated in the methodology and other sections.

- To me, it makes more sense to incorporate parts of section 4.1 in section 3.2. In its current form, section 4.1 gives the impression of being a rather standalone section, and does not entirely seem to fit within the logical flow of the paper. You could solve this by transferring parts to section 3.2. Basically, in section 3.2, you want to isolate the air of stratospheric origin in the UTLS to look at its ozone concentration properties. So section 4.1 should make a link to this section, but now using tracer correlations to look at the origin of the UTLS air masses.

Answer: These are good suggestions, and we continue to develop them in the following two points. We moved part of the first text (in section 4.1) to section 3.3 (methodology) where better fits and rephrased: "*Stratospheric and tropospheric tracer scatterplots provide a magnitude of bidirectional exchange across the tropopause by the identification of stratospheric and tropospheric branches and mixed air regions according to their tracer abundances (Gettelman et al., 2011). Hence, in this study, our first analysis used tracer correlations to compare the northern (IAGOS) and southern hemisphere (SouthTRAC) mid-latitudes, and between high and low ozone depletion years (NH).*"

- Also the choice of the 4.1 section title could be better. The sentences at the end of page 8 might be replaced by a better guidance to the analyses done in sections 4.1 and 4.2, and referring more directly to these sections (rather than describing in rather general terms, as is done now).

Answer: We changed the section title to "*Tropospheric and stratospheric tracer correlations*". We also reworded the beginning of the section: "*In the following section, we used tracer correlations to characterize the tropospheric or stratospheric origin of the air masses in the UTLS and the free troposphere of both hemispheres. In section 4.2, we applied filters mainly based on the relative humidity to assign the stratospheric origin in the UTLS and then determine the inter- and intrahemispheric ozone differences. Finally, in section 4.3, we compare the results based on situ ozone measurements with CAMSRA outputs".*

- Figure 2: in addition to my previous comment: argue why these O3-H2O tracer correlations are important, and what you learn from them, and if there is difference between the LD and HD correlations. Also, specify in the caption which periods (2002-2022?, 4 Mar – 20 May NH?, 4 Sep– 20 Nov SH?) have been used for the correlation plots.

Answer: We appreciate this comment and the intention to improve our work. Therefore, we added the following lines to ensure a better transition between sections 4.2 and 4.2: *"Overall, in this section, we found that the NH does not show differences between high and low depletion years. Therefore, in the following section, we test different filters to isolate the air of stratospheric origin in the UTLS and thus investigate its ozone mixing ratios during high and low depletion years in both hemispheres. In this section, we also showed that the NH has higher carbon monoxide levels…"*

We also added the exact periods in the caption of Figures 2 and 3.

- Figure 3: specify in the caption which periods (2002-2022?, 4 Mar – 20 May NH?, 4 Sep – 20 Nov SH?) have been used for the correlation plots.

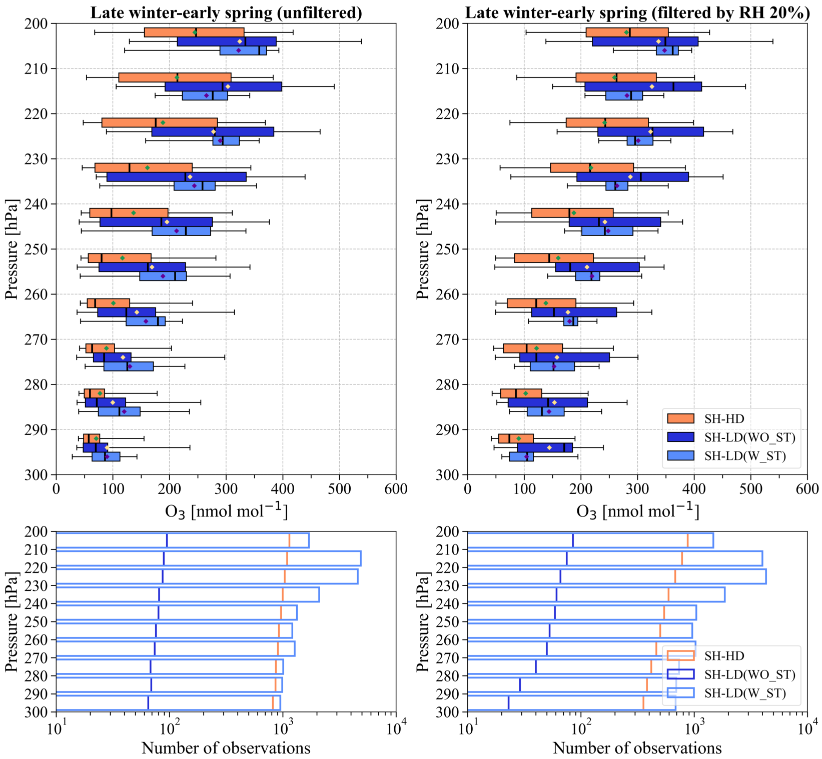
Answer: We added: "…*The left panels show scatter plots for the period between 4 Mar and 20 May from 2002 to 2022 for the NH between 300-200 and 700-300 hPa (IAGOS data set described in Table 1). The right panels show the same for the SH between 4 Sep and 20 Nov 2019 (SouthTRAC campaign data described in Table 1)...*"

- Figure 4: specify in the caption that these are SouthTRAC flights only!

Answer: We added: *"…These scatter plots are based exclusively on SouthTRAC data...*"

- Line 269: what does the comparison between Fig. 5 (stratospheric origin) and A1 (mixture of tropospheric and tropospheric origin) learn us? For instance, for late winter – early spring the SH LD ozone amounts are much larger than the SH HD ozone amounts in A1 (no RH filter), compared to the same comparison in Fig. 5 (RH filter). Any clue for this?

Answer: The filter's main function is to eliminate levels with more tropospheric (or mixed) properties. This can be clearly seen in the figure below, where the lowest ozone levels tend to be eliminated after the filter. We've added the following line to strengthen the message: "*The filter's applicability can be clearly verified for the subperiod late winter-early spring in the SH, where differences between high and low ozone depletion years can be noticed without any filter (Figure A1). However, we can also note that the 20% RH filter is very effective at removing the lowest ozone mixing ratios with tropospheric origin (or mixed air).*"



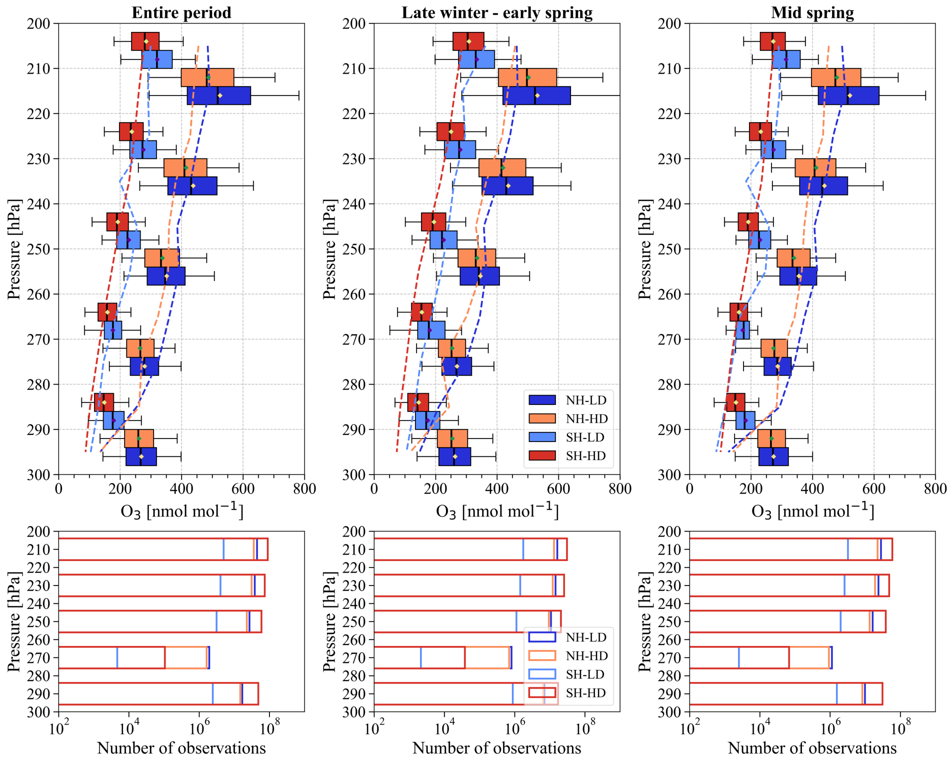
- On page 12, line 280, you define differences between high and low-depletion years as "interannual variability". I don't think that "interannual variability" is a good term for it; it is such a general term. Also you should define how you calculate the ozone difference between high and low-depletion years. It is simply the difference between the mean of the overall springtime ozone between 300-200 hPa for both the LD and HD years, or do you somewhat average out the mean values for every pressure level, as shown in Fig. 5? How are then the values mentioned in lines 280-282 calculated? Please specify! From Fig. 5, I would assume that the ozone differences between LD and HD years are nowhere significant, so I don't understand quite well how the values, and their uncertainties, in lines 280-282 have been obtained.

Answer: Instead of interannual variability, we now explicitly use *"difference between high and low ozone depletion years"*.

We specified in the text how we calculate the differences: "*To provide a quantitative magnitude of these differences, we averaged the medians between 300-200 hPa (shown in Figure 5) and then calculated the ozone difference (and standard deviation) between low and high depletion years.*"

- In Fig. 6: can't the comparisons (vertical dashed lines) with the ozone medians in the NH obtained from IAGOS added to this figure, similarly as has been done for the SH?

Answer: Yes, we did it in a new figure (see below).



- Related to previous comment: the analysis of this figure 6 is only used for comparing the SouthTRAC measurements (2019 only) with the CAMS reanalysis output (the low-depletion years 2002 and 2019?). Is this the most important message from this Figure? Shouldn't the (pattern) agreement between Fig. 5 (observations) and Fig. 6 (model) be discussed first? E.g. the higher NH ozone values, the slightly (but not significant) higher amounts for LD years compared to HD years? And how do the model values relate with the measurement values (for all measurement types, and in NH and SH)?

Answer: We made some changes to the text to address the points mentioned: "***Figure 6*** *shows that the reanalysis reproduces the ozone interhemispheric differences, i.e., higher ozone in the NH and differences between high and low ozone depletion years. We also compared the pattern agreement between the ozone boxplots (CAMSRA) and in situ measurements.* ***Figure 6*** *clearly illustrates the similarity between the ozone medians obtained from the CAMS reanalysis and in situ measurements (SouthTRAC and ozonesondes) particularly in the period with the highest number of flights, i.e., late winter-early spring, for pressures lower than 270 hPa. The lower agreement between the ozone medians of CAMS and in situ measurements in mid-spring period was determined by the lower flight frequency in this period, i.e., six flights during the mid-spring period versus ten in the late winter-early spring period (also discussed in section 4.2).*

- Lines 314-325: those findings are really not obvious at all from the figure. Are you saying that the agreement between CAMS and SouthTRAC (SH LD, light blue) is better during late winter –early spring compared to mid spring? On which ground? For pressures lower than 270 hPa (instead of below 270 hPa)? What do you mean with higher "fluctuations" in the medians for the mid-spring period? In the vertical or looking at the percentiles of the medians (I don't see this for the latter)? Are fluctuations in the vertical relevant given the coarser vertical pressure grid for CAMS?

Answer: Yes, that is our interpretation based on the ozone medians. Note that we replaced the term "below" with "lower than". Regarding the term "fluctuation," we modified the text: "*The lower agreement between the ozone medians of CAMS and in situ* *measurements in mid-spring period was determined by the lower flight frequency in this period, i.e., six flights during the mid-spring period versus ten in the late winter-early spring period (also discussed in section 4.2).*

- Figure 7: in the discussion of Fig. 7, it might be informative to give the overall mean UTLS ozone value for each considered case (NH LD, NH HD, SH LD, SH HD).

Answer: We added: "*Mean UTLS ozone during high and low ozone depletion years was 399 and 425 nmol mol^{-1} in the NH, and 228 and 265 nmol mol^{-1} in the SH.*"

- Conclusions: if you put so much weight on the analysis in section 4.1 (two paragraphs devoted to it in the conclusions), you should stress much more the importance of this analysis in the main text as well. As written before, for me, it really hampers the logical flow of the paper a bit in its present form.

Answer: We have taken the suggestions (mentioned in the above points) relating to section 4.1 in terms of logical flow and reinforcing key messages, as well as the transitional role of that section.

Technical corrections

- Line 39: remove the dot before OH

Answer: We would like to retain the dot that indicates it is a radical species

- Line 86: remove the brackets before and after Neu et al., 2014

Answer: We removed them.

- Line 86: remove "diminished"

Answer: It does not apply in the current version.

- Line 90: remove "from"

Answer: We removed it.

- Line 93: in situ measurementS. Futher: replace "air masses exhibiting stratospheric character" with "air masses of stratospheric origin"

Answer: We replaced them.

- Line 135: replace so-called initiative with the Tropospheric Ozone Assessment Report – Phase II (TOAR-II) Focus Working Group HEGIFTOM (Harmonization and Evaluation of Ground-Based Instruments for Free-Tropospheric Ozone Measurements).

Answer: We replaced it

- Line 161: an identical instead of the identical

Answer: We modified it.

- Line 184: proposed by Charlton and Polvani (2007)

Answer: We modified as suggested.

- Line 207: Section 4.1 instead of Section 3.2

Answer: We replaced 3.2 by 4.2.

- Line 237: give the pressure level instead of "above 9 km".

Answer: We made the change suggested.

- Line 266: and instead of y

Answer: We changed it.

- Line 305: add ozone between percentage and difference

Answer: We made the change suggested.

- Line 358: replace "characterized by" with

Answer: We made the change suggested.

Referee #2

I want to thank the authors for incorporating my (and other) reviewer suggestions, increasing the amount of data analyzed, and adding confidence to the results contained in this manuscript. It is a much improved final product and I have only a few final technical comments:

- Line 90: This should be "tracers", correct?

Answer: We replaced "traces" by "tracers"

- Line 193: "reverse the zonal wind at 60ºS"

Answer: We made the change

- Line 266: "300 and 280 hPa"

Answer: We made the change

- Figure 7 Caption: change "lower less" to "less than"

Answer: We made the change

- Bottom of Page 16: Please add "iii." and "iv." to the final two paragraphs/conclusions

Answer: We added just the "iii" since the last sentence is more general.