Author Comment to Referee #2

ACP Discussions doi:10.5194/acp-2020-747 (Editor - Farahnaz Khosrawi) 'Potential of future stratospheric ozone loss in the mid-latitudes under climate change and sulfate geoengineering'

We thank referee #2 for specific guidance on how to revise our paper. We performed a sensitivity study assuming 5 K less temperatures than simulated in GLENS to show the impact of lower temperatures for this ozone loss process more clearly. Further, we discussed the missing of convective overshooting events shortly and do not claim our results "upper boundary" any more. Our reply to the reviewer comments is listed in detail below. Questions and comments of the referee are shown in italics. Passages from the revised version of the manuscript are shown in blue.

This is a well-designed study that seeks to examine the potential for and impacts of heterogeneous chlorine activation in the lower stratosphere on ozone in current and future climates. In particular, using global climate model projections of the future with and without geoengineering assumptions, the likelihood of chlorine activation is assessed over time and evaluated to assess future impacts on lower stratosphere ozone. Overall, I found the paper to be a valuable contribution and worthy of publication. My only significant criticism would be in the weight given to the results and their interpretation in the text throughout, as the narrative broadly glances over the limitations/caveats of the GLENS model that are relevant to the subject matter. This is not to say that important elements are ignored or simply not acknowledged. Rather, they are largely dismissed when discussing the significance of the results. Either more evidence needs to be given to favor or increase confidence the GLENS results or the limitations of the model should be more routinely stated in context of the results.

Major points

One of the more concerning limitations of GLENS in my assessment is the apparent warm bias of the model in the UTLS (which is common in most models given their relatively coarse vertical resolution near the tropopause). In the paper, this is assessed using airborne observations from the SEAC4RS campaign, which are very good but ultimately too limited for comprehensive validation of the model. I would strongly recommend that the authors consider using high-resolution radiosonde observations to characterize the true temperature bias in the UTLS (by comparing tropopause-relative T) as it may be as high as 5 K based on the data shown and is a major source of sensitivity to the chlorine activation results. The authors do show what an assumed warm bias of 2 K would lead to, but even this number appears to be conservative in my opinion. Rather than the messaging throughout stating that the assessments in the paper are an "upper bound" to chlorine activation, I would argue that in many ways they are a lower bound. Better assessments of model biases will help to focus the messaging more on the expected likelihood and impacts of this important process.

We agree with the referee that the warm bias of the model could be as high as 5 K from the comparison with SEAC^{4}RS measurements. Determining the temperature bias more extensively (e.g. using radiosonde observations) would be enough content of a new study. Hence, it is out of the scope of our study, which focuses on the likelihood for the ozone loss process to occur in the mid-latitude lowermost stratosphere. Because the temperature plays a key role for the likelihood of the ozone loss process, we analysed a further sensitivity case with 5 K lower temperatures in Sec. 4.5 of the revised version of the manuscript. The results of this case study are also included in the conclusions to estimate the range of uncertainty of our results better.
The occurrence of conditions which would lead to chlorine activation and thus ozone loss in the mid-latitude lowermost stratosphere is not yet clear (e.g. the occurrence frequency of convective overshooting, the temperature and the ozone mixing ratio of moist air masses, the duration of the conservation of low temperatures together with elevated water vapour). Because of this range of uncertainty, we decided to claim our results not as "upper boundary" to the impact of chlorine activation on ozone.

Sec. 4.5 was revised as follows discussing in addition a temperature shift of

−5K.

Likelihood of heterogeneous chlorine activation and its impact on ozone for low temperatures

As analysed in Sec. 3.1, the temperatures in the mixing-layer above the tropopause simulated in GLENS may be higher than the real atmospheric temperatures in this region. Therefore, a sensitivity study is performed assuming a shift in GLENS temperatures of –2 K and of −5 K to explore the impact of uncertainties in the temperatures calculated in GLENS. However, the focus of this sensitivity assumption is only on the temperature shift without considering a potential ice formation at very low temperatures. The likelihood for the occurrence of heterogeneous chlorine activation assuming lower temperatures and its impact on ozone in the lowermost stratosphere is presented in Fig. 1 (of this reply).

The likelihood that chlorine activation occurs would increase significantly assuming lower temperatures (Fig. 1, top). For case C2010, chlorine activation would occur with a likelihood of 3.7% assuming 2 K lower temperatures and of 10.9% assuming 5 K lower temperatures than in the GLENS simulation (assuming GLENS temperatures, the likelihood accounts to as 1.0%). In the global warming cases C2040 and C2090 the likelihood increases likewise assuming lower temperatures. In case C2040, assuming temperatures of 2 K less yields a likelihood of 1.4% and assuming 5 K less of 6.4% (instead of 0.1% assuming GLENS temperatures). In case C2090, for −5 K, the likelihood accounts to as 2.7%. Applying geoengineering would cause the highest likelihood for chlorine activation to occur. Assuming 2 K lower temperatures than the GLENS simulations, in case F2040 6.7% (16.9% for −5 K) and in case F2090 7.4% (19.5% for −5 K) of the air masses would yield chlorine activation.

Despite the higher likelihood of chlorine activation in the F2090 case, ozone is more affected in the F2040 case, because the ozone values in the range where ozone destruction would occur in the years 2040–2050 are higher than in the years 2090–2100 (not shown). For 2 K lower temperatures, activated chlorine would destroy up to ∼0.8% of ozone in the lowermost stratosphere in the F2040 case, but only up to 0.4% in case F2090 (Fig. 1, bottom). Assuming

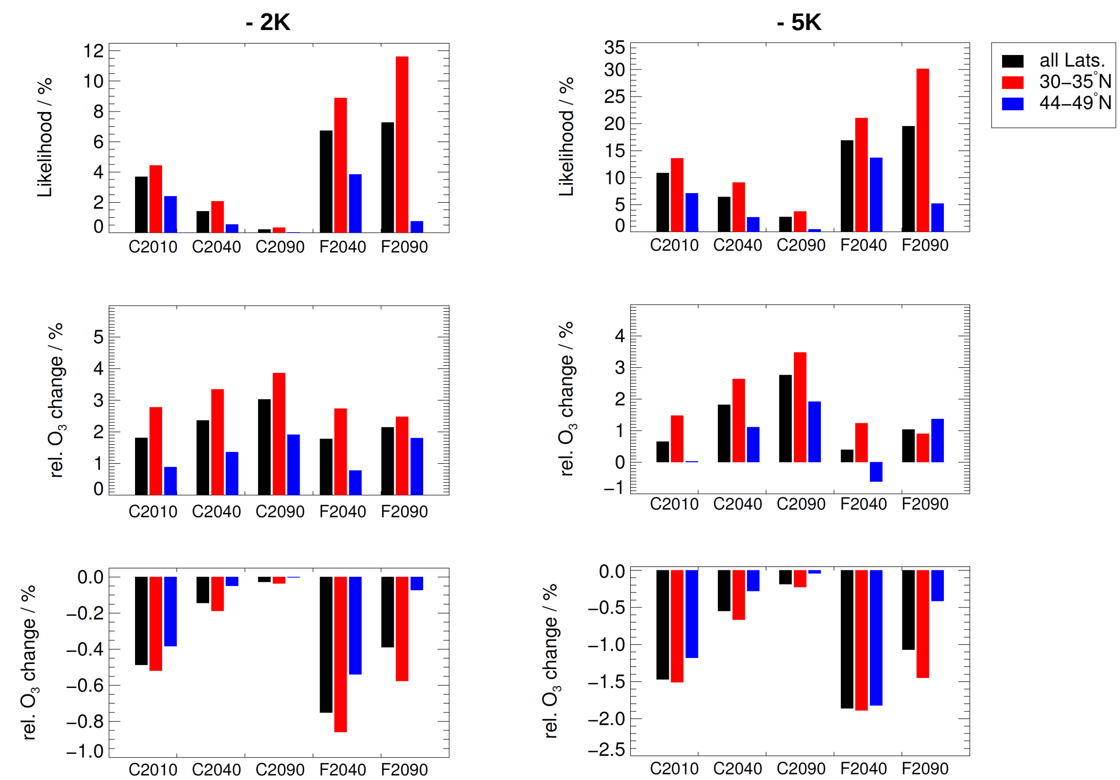

Figure 1: Likelihood (top) for the occurrence of chlorine activation as well as its impact on ozone in the lowermost stratosphere assuming 2 K (left) and 5 K (right) lower temperatures than simulated in GLENS. Further, the chemical ozone change in the mixing layer assuming 10 consecutive days without mixing of air parcels (middle) and the relative ozone change in the mixing layer caused by heterogeneous chlorine activation (bottom) is shown for the assumption with 2 K and 5 K less temperatures. Note that the scale on the y-axes differs. (See Tab. 1 for case descriptions.)

5 K less, 1.9% (F2040) and 1.1% (F2090) of ozone in the lowermost stratosphere are destroyed. In the global warming scenario, more ozone would be likewise destroyed due to heterogeneous chlorine activation.
The higher ozone destruction due to chlorine activation for lower temperatures results in a reduced net ozone formation in the mixing layer. For all cases considered (global warming and geoengineering), the relative net ozone change (Fig. 1, middle) is significantly reduced. In case F2040 in the extratropical latitude range, even a net ozone destruction occurs in the mixing layer assuming 5 K less than simulated in GLENS. However, comparing the behaviour in different latitude regions, the impact of heterogeneous chlorine activation on ozone is mostly higher in lower latitudes.
The likelihood for heterogeneous chlorine activation to occur and its impact on ozone in the mixing layer determined in this section is summarized in Tab. 3 referred to as '2 K' and '5 K' lower temperatures. Assuming less temperatures than those calculated in GLENS increases the likelihood for heterogeneous chlorine activation to occur as well as its impact on lowermost stratospheric ozone. In all cases, the relative ozone loss in the mixing layer is two to three times higher assuming 2 K lower temperatures than in the reference and six to ten times higher in the −5 K assumption. Assuming low temperatures, in all cases considered, an upper limit of 0.3 DU from a total ozone column of ∼303 DU in this region (which is ∼0.1%) has been estimated as the total ozone reduction caused by heterogeneous chlorine activation.

The results of the "−5 K"-study are added to Tab. 3,

Table 3. Overview on the likelihood for chlorine activation to occur in the mid-latitude mixing layer above the tropopause, its impact on ozone in the mixing layer and the relevance for ozone column. Further the net chemical ozone change in the mixing layer is specified. Three latitude ranges are considered here: 30–49°N, only the subtropical latitude band in 30–35°N and only the extra-tropical latitude band in 44–49°N. The considered cases today (C2010) and in the future scenarios assuming global warming (C2040, C2090) and additional geoengineering (F2040, F2090) are further described in Tab. 1. The reference refers to GLENS results for the mixing layer. In the assumption with 2 K (5 K) lower temperatures, temperatures of GLENS air masses are reduced by 2 K (5 K) to infer uncertainties in the simulated temperatures. The chemical ozone changes here are determined based on 10-day box-model simulations neglecting mixing between neighbouring air masses. Thus conditions causing chlorine activation are assumed here to be maintained for 10 consecutive days without perturbations.

			Reference				**2 K lower temperatures**				**5 K lower temperatures**			
	O_3-Column / DU	O_3-Column in the mixing layer / DU	Likelihood for chlorine activation	net ozone change	rel. O_3-loss	O_3-loss / DU	Likelihood for chlorine activation	net ozone change	rel. O_3-loss	O_3-loss / DU	Likelihood for chlorine activation	net ozone change	rel. O_3-loss	O_3-loss / DU
C2010														
all Lats.	295.8	15.7	1.0%	2.3%	0.1%	0.02	3.7%	1.8%	0.5%	0.08	10.9	0.7%	1.5%	0.23
30–35°N	289.9	14.2	1.1%	3.4%	0.2%	0.02	4.4%	2.8%	0.5%	0.07	13.6	1.5%	1.5%	0.21
44–49°N	307.8	20.3	0.9%	1.2%	0.1%	0.03	2.4%	0.9%	0.4%	0.08	7.1	0.0%	1.2%	0.24
C2040														
all Lats.	307.2	16.9	0.1%	2.5%	<0.1%	<0.01	1.4%	2.4%	0.1%	0.02	6.4%	1.8%	0.5%	0.08
30–35°N	299.0	13.9	0.1%	3.6%	<0.1%	<0.01	2.1%	3.3%	0.2%	0.03	9.1%	2.6%	0.7%	0.10
44–49°N	319.2	21.4	0.1%	1.4%	<0.1%	<0.01	0.5%	1.3%	0.1%	0.01	2.7%	1.1%	0.3%	0.06
C2090														
all Lats.	321.7	18.8	0.0%	3.0%	0.0%	0.00	0.2%	3.0%	<0.1%	<0.01	2.7%	2.8%	0.2%	0.04
30–35°N	309.6	15.5	0.0%	3.9%	0.0%	0.00	0.3%	3.9%	<0.1%	<0.01	3.7%	3.5%	0.2%	0.03
44–49°N	336.9	23.8	0.0%	1.8%	0.0%	0.00	<0.1%	1.9%	0.0%	0.00	0.4%	1.9%	<0.1%	<0.01
F2040														
all Lats.	302.7	15.2	3.3%	2.3%	0.3%	0.05	6.7%	1.8%	0.8%	0.11	16.9%	0.4%	1.9%	0.29
30–35°N	296.0	12.8	4.1%	3.4%	0.4%	0.05	8.9%	2.7%	0.9%	0.11	21.0%	1.2%	1.9%	0.24
44–49°N	313.6	19.1	1.3%	1.2%	0.2%	0.03	3.8%	0.8%	0.5%	0.10	13.7%	−0.6%	1.8%	0.34
F2090														
all Lats.	321.1	17.0	2.7%	2.6%	0.1%	0.02	7.3%	2.1%	0.4%	0.07	19.5%	1.0%	1.1%	0.19
30–35°N	310.0	14.7	4.5%	3.2%	0.2%	0.03	11.6%	2.5%	0.6%	0.09	30.1%	0.9%	1.4%	0.19
44–49°N	334.3	21.5	0.2%	1.9%	<0.1%	<0.01	0.8%	1.8%	0.1%	0.02	5.2%	1.4%	0.4%	0.09

the conclusion of our study (p. 31, l. 19–20 of the revised version of the manuscript)

Assuming 2 K lower temperatures, the likelihood increases accounting for 3.7% in case C2010, 6.7% and 7.3% in the cases F2040 and F2090, respectively, and 1.4% and 0.2% in the cases C2040 and C2090, respectively. Assuming 5 K lower temperatures, the likelihood is higher with 10.9% in case C2010, 2.7% in case C2090 (global warming) and 19.5% in case F2090 (geoengineering).

and for comparison again to the final section of the conclusion (p. 32, l. 13–21 of the revised version of the manuscript).

In summary, we show that heterogeneous chlorine activation affects ozone in the lowermost stratosphere in mid-latitudes, but the impacts are very small. Sulfate geoengineering leads to a 2–3 times higher likelihood for the occurrence of chlorine activation. However, in the geoengineering case most likely for chlorine activation, chlorine is activated with a probability of 3.3% (16.9% assuming 5 K lower temperatures) in the entire latitude region considered here. In all cases today and in future, less than 0.4% (1.9% assuming 5 K lower temperatures) of ozone in the mixing layer are destroyed caused by heterogeneous chlorine activation. This leads to a reduction in column ozone of 0.1 DU (0.3 DU if 5 K lower temperatures are assumed), which are 0.1% of column ozone. Thus according to the results of this study, the relevance of ozone destruction caused by heterogeneous chlorine activation in the mid-latitude mixing layer between stratospheric and tropospheric air is negligible with respect to the ozone column and small in the mixing layer even if sulfate geoengineering would be applied.

The second limitation that I believe needs to be better addressed and highlighted is the representation of convection in the GLENS model. Climate models are often not classified as resolving (or even representing) convection well. Rather, global coarse horizontal resolution models such as GLENS are often better used to assess changes in convective environments. Dynamically downscaled climate simulations have become increasing used to study convection since it can be better simulated (and even resolved) over regions of interest by using the large-scale environments projected by the global model *as input. Since this study relies on the global large-scale climate projection alone, the realism of UTLS water vapor and its variability due to convection is highly questionable. It is very likely a significantly underestimated reference point, which again is in contrast to the messaging throughout in the paper. I would like to see these points better highlighted and used to interpret the results. I'm not asking for additional analysis to respond to this point, but more appropriate messaging/discussion in the text and perceived importance or likelihood of chlorine activation.*

Besides the temperature, the water vapour content in the lowermost stratosphere plays a key role for the ozone loss process analysed in this study. Hence, we investigated the water vapour transport to the lowermost stratosphere in case C2010 roughly for getting a first impression on the question, how high water vapour mixing ratios reach altitudes of the lowermost stratosphere in the GLENS case C2010. One example with more than 10 ppm H_2O at a pressure level of $\sim$100 hPa (shown in Fig. 3 of this reply) was investigated. For this example, convection up to a pressure level of $\sim$165 hPa was found in the GLENS simulation over Central North America (Fig. 2 of this reply). The successive advective transport from the maximum level of convective outflow up to $\sim$100 hPa follows the anticyclonic flow of the North American summer monsoon (indicated as pathway a and b in Fig. 3 of this reply). A similar transport mechanism was also found by (Wang et al., 2020). Further, transport from the pacific region (path c) could contribute to elevated water vapour in this specific case. However, analysing the transport of water vapour into the lowermost stratosphere for more cases in GLENS would be an entirely new study and is hence out of the scope of our study.

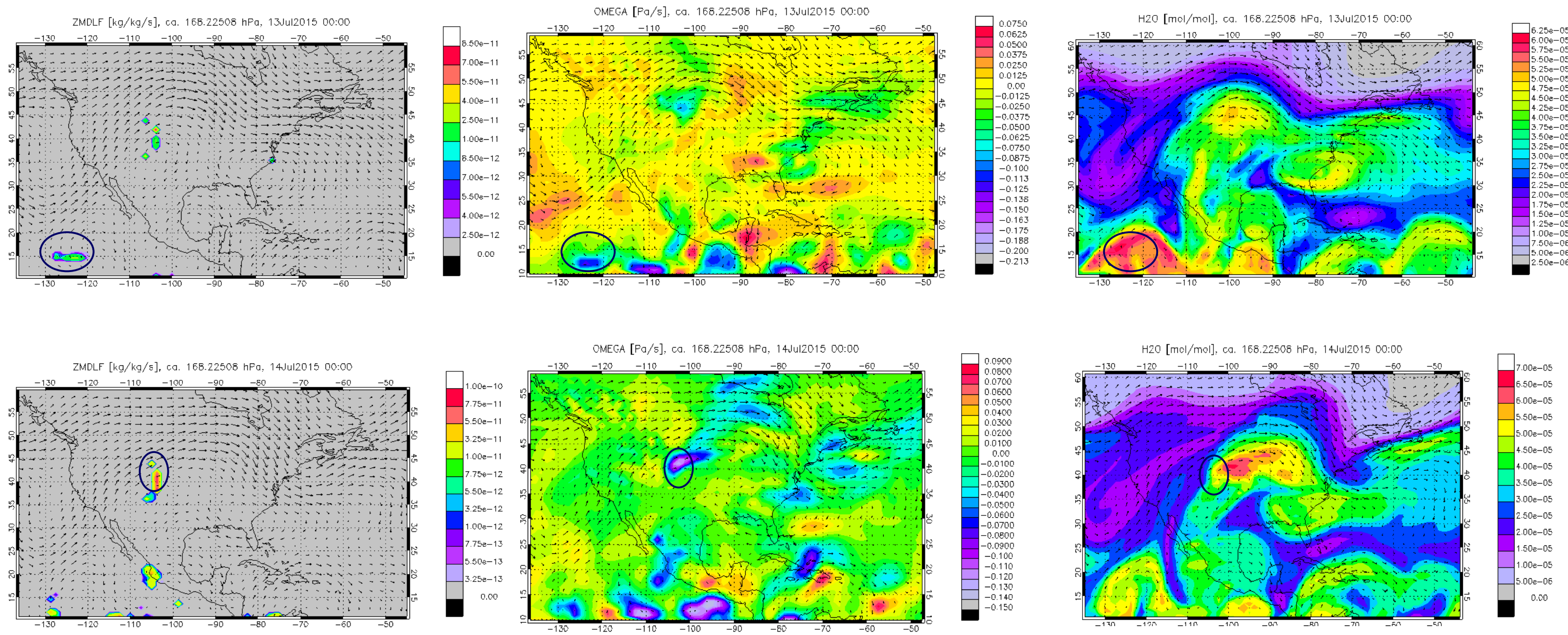

Figure 2: Measure for air masses transported by convection to the 168 hPa level in WACCM (left), upwards transport (middle) and water vapour (right) in the 168 hPa level on 13 July (upper panels) and 14 July (bottom panels) of case C2010.

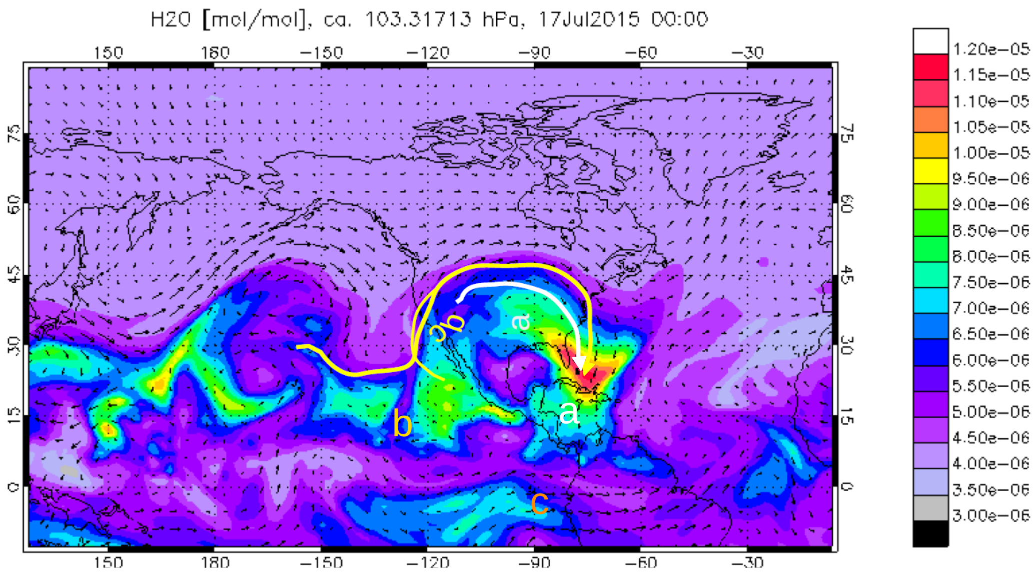

Figure 3: Example for an event with elevated water vapour mixing ratios at ∼100 hPa simulated in GLENS (case C2010). The coloured lines (a,b and c) refer to potential pathways causing the water vapour enhancement.

To mention that the water vapour transport in GLENS does not represent the reality properly we added a sentence in the discussion section of our manuscript and mention results of the study of Anderson and Clapp (2018), where higher water vapour mixing ratios are assumed, in comparison to the results in our study (p. 29, l. 12–19 of the revised version of the manuscript).

This chlorine driven ozone loss process could occur today above central North America in relation to stratospheric moistening through convective overshooting events during the North American Summer Monsoon (NAM). However, convection implemented in WACCM does not consider overshooting convection (i.e. convection up to above the local tropopause) and therefore the transport of enhanced water vapour into the lower stratosphere by convection is most likely underestimated in GLENS simulations. Anderson and Clapp (2018) performed a box-model study assuming that conditions yielding heterogeneous chlorine activation, as low temperatures and a high water vapour mixing ratio of 20 ppmv, are maintained for 14 days. With this assumption, they simulated a maximal fractional ozone loss of −2.5 to −67% (depending on the HCl mixing ratio) for the lower stratosphere between 12 km and 18 km. In our study, chlorine activation would reduce ozone in the mixing layer by 0.1% for today's conditions (case C2010; 0.7% assuming 5 K lower temperatures).

Further, we mention in the conclusion (p. 31, l. 28–29 of the revised version of the manuscript), that convective overshooting is not implemented in GLENS.

In addition it should be noted that convection used in WACCM does not consider overshooting convection (i.e. convection up to above the local tropopause) and therefore the transport of enhanced water vapour into the lower stratosphere by convection is most likely underestimated in our study.

Minor points

Page 11, line 4 - should cite Smith et al, 2017 (doi:10.1002/2017JD026831) and Herman et al, 2017 (doi:10.5194/acp-17-6113- 2017) as well since this studies more extensively evaluate delivery of water to the stratosphere by convection during SEAC⁴RS.

We thank the referee for this remark and cited the study of (Smith et al., 2017) and (Herman et al., 2017) (p.12, l.3 of the revised version of the manuscript).

Page 26, line 21 - contrary to this statement, I found very little discussion of the apparent temperature bias in GLENS in Section 3.1.

To make the temperature difference between the GLENS C2010 mixing layer and the $SEAC^4RS$ measurements more clear, we added the temperature ranges more clearly to avoid any misunderstandings (p. 12 l. 2–7 in the revised manuscript).

Measurements during $SEAC^4RS$ sampled convective injections of water vapour into the stratosphere (Toon et al., 2016; Smith et al., 2017; Herman et al., 2017) and thus provide unusual cold and moist conditions for the lowermost stratosphere, which are lower than the temperatures in the simulated C2010 case (∼195–209 K mainly prevailing in SEAC^{4}RS instead of ∼201–209 K in case C2010). To consider the impact of this temperature bias on ozone further simulations are preformed (see Sec.) assuming temperatures to be 2 K and 5 K lower than found in GLENS.

References

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