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Interactive comment on "Persistance of upper stratospheric winter time tracer variability into the Arctic spring and summer" *by* David E. Siskind et al.

Anonymous Referee #2

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This paper contains an analysis of measurements and model simulations of methane (CH4) and chlorine monoxide (ClO) in the upper stratosphere at northern high latitudes from late winter through the early fall seasons. It is shown that interannual variability in winter/spring dynamics leads to large changes in the mean CH4, which persists into the summer, and impacts the level of summertime ClO and ozone (O3). Overall, this is a very significant result as it shows that high latitude winter/spring dynamics in the upper stratosphere and mesosphere has an important effect on summertime O3 in the upper stratosphere through chlorine chemistry, which is distinct from the well-established link to ozone through the descent of nitrogen compounds.

The manuscript is fairly clear overall; the organization of topics, the text, and figures are all in good shape for publication. However, there are a few critical issues that should be addressed by the authors in order to make this story complete. I recommend that the paper should be published in ACP, provided that the two major issues below can be adequately addressed.

Major

1. Possible effects from sampling biases of SOFIE observations: The manuscript makes it clear that the sampling latitude for SOFIE varies with season between 65oN and 82oN. This is highly appropriate to point out to the reader, but it leads to a number of possible issues that may impact the analysis. Perhaps these issues are addressed in detail by previous publications. If so, then a summary discussion should be included here (with references). If not, then this paper should explore the following impacts in greater detail:

(i) Interpretation of "zonal means" in time-pressure coordinates (Figs 1 and 2) is clouded by the mixing of time and latitude dimensions. Inferring the amount of descent from Fig 1 (discussion lines 115-148) and the impact of photochemistry on the summertime decrease in CH4 (lines 136-139) can only be done if latitudinal sampling effects can be ignored. If the observation latitude is changing by ∼6deg every month, then a month-to-month variation is likely induced through the sampling of background latitudinal gradients in CH4. For example, it is possible that a large fraction of the CH4 decrease seen in every year from day 172 to day 264 in Fig 2 is a result of the latitude decreasing from 56 to 82deg latitude (and similarly for the abrupt increase in CH4 after day 264).

(ii) The number of longitude samples per zonal mean, and their distribution in longitude for the daily (Fig 1) and five-day (Fig 2) means should be reported, in order to address the question of whether these data truly represent zonal means.

(iii) The possible use of equivalent latitude coordinates should be explored. It has been well established that for long-lived tracers such as CH4, time variations in zonal means can be altered by non-zonal circulation patterns. It is a particularly pronounced effect for zonal means taken near the winter/spring vortex edge (this impacts up to day ∼100 in Figs 1, 2, and Fig 5 for O3). Equivalent latitude transformation is probably not possible with SOFIE measurements, but it can and should be done here for MLS ClO observations and also for the WACCM model results. It should be clearly demonstrated that either (i) the impact of combining different dynamical regimes (e.g. inside/outside vortex) has little impact on the CH4, ClO, and O3 variability in zonal means, or (ii) even a large impact in winter/spring does not affect the main conclusion for summertime ozone.

2. A more complete picture of the chemistry, including other compounds, is needed. One of the main conclusions of paper draws on photochemical links, from CH4 to ClO, and from ClO to O3. The description of this chemistry is lacking in detail and leaves important questions unanswered. At a minimum, some interested readers will not be experts in stratospheric/mesospheric chemistry so that a discussion of the issues below is warranted.

(i) The reaction Cl+CH4 -> HCl+CH3 is identified on line 160 as being the reason that ClO varies inversely with CH4 as in Figures 3 and 4. The changes are not 1:1 (hundreds of ppbv for CH4, tenths of ppbv for ClO), but is this expected? Is it a result of other loss processes for CH4? Are the observed slopes of ClO vs CH4 consistent with expectations from chemistry? For example, if there is a true inverse relationship (y=1/x, and not y=1/x2 or y=1/sqrt(x)), then we might expect dy/y=-dx/x. Is this the case here? If a causal link is being argued, then it is important to build the foundation by considering and discussing the important chemical processes impacting both CH4 and ClO, such as major production and loss mechanisms.

(ii) Connected to the above questions, it is puzzling why there are no MLS HCl measurements included in the analysis, considering that it is a product of the Cl+CH4 reaction. Are the effects from CH4 variability seen in HCl, and if not, why? A positive results would clear bolster the discussion related to (i) above.

(iii) Diurnal variations. I believe that there may be significant diurnal variability in both ClO and O3 in the upper stratosphere and stratopause regions. Is this an issue for sampling MLS at the SOFIE latitudes? Since MLS is sun-synchronous, then the local time of the sampled MLS measurements will vary with season.

(iv) A clearer picture of the changes in ozone attributed to ClOx chemistry is needed. Although Fig 6 from WACCM/NOGAPS is useful, interpretation of model results is sometimes just as complicated as observations unless there is an accompanying, simplified analytic/theoretical basis. In particular, on lines 208-220 it is stated that ozone loss from chlorine is about 20% larger in 2009 than in 2008. Yet, the net effect considering ClOx, NOx, and HOx loss is only 2% larger in 2009 vs 2008. Is this just an additive sum of the loss rates? Why is NOx lower in 2009? Large descent in that year should have led to higher NOx. How does the WACCM NOx for 2008/2009 compare with measurements, i.e., can we really believe the NOx behavior? (Important - as it offsets most of the ClOx effect.) It also seems odd that the HOx cycle is relatively unchanged between 2008 and 2009; changes in descent likely changed upper stratospheric water vapor (measured by MLS), which should impact HOx. Although it's true that NOx and HOx are not the main foci of this paper, any conclusions involving ClOx - O3 links based on observations should carefully consider the impacts of these other chemical families. This would include both their influence on O3, and possible interrelationships between HOx, NOx, and ClOx.

Minor things to consider:

1. Lines 54-60 and Table 1: Two things to consider here. Since their are 4 combinations to two quasi-binary states, is there any coupling between the two states, i.e., are winter descent and spring PW independent? Also, there is no clear statement of the criteria used to gauge both descent and PW activity. This would include both the quantity used in each case (e.g., slope of tracer contours for descent, or heat fluxes for PW) and the corresponding thresholds for distinguishing "high" from "low".

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Discussion paper

2. Line 104: should probably define "MERRA"

3. Figure 1: As noted on lines 113-117, this figure illustrates variations in upper stratospheric CH4 that can be driven by variations in descent from the mesosphere . In looking a cross-stratopause transport, it would be useful to show the stratospause as dashed curves in Fig 1. Since SOFIE measures T, this should be a straightforward addition.

4. Lines 146-148: Contrary to the statement that "once the relative abundance of CH4 was established by May, it remained mostly unchanged until October", it appears from Figure 2 that in 2011, 2014, and 2013, CH4 decreased by around 60%. It is harder to judge some of the other years, but they appear to be around 50%. On the other hand, is this statement mean to convey that all of the years seem to decrease by the same fraction?

5. Figures 5 and 6: The choice of using 80N for looking at ozone and ozone loss rates is not well justified. On the one hand, Figure 4 states that the mean latitude of SOFIE for August is 78deg (and 78deg is used also used in Fig 8 for ozone). On the other hand, if one is trying to quantify the impact on ozone from ClO variations observed over the months of May-August (and sampled at the SOFIE latitudes as in Figure 3), then the mean latitude might be closer to 70deg .

6. Lines 219-220: In Figure 6, at 4 hPa ozone loss is about 10% less in 2009 compared with 2008, whereas at 1.47 hPa it is about 2% larger in 2009 compared with 2008. This is stated as the reason why, in Figure 7, 2009 minus 2008 ozone is positive at 4 hPa and negative at 1.47 hPa. Yet, the 4 hPa change in loss is 5 times larger than at 1.47 hPa whereas the ozone response is smaller at 4hPa than at 1.47 hPa. This is not explained. Is the ozone response nonlinear?

7. Figure 1 caption: "This latitude varies has some variation..."

8. Figure 7: "monthly and daily averaged" is unclear.

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