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Interactive Comment

Interactive comment on "Evidence for heterogeneous chlorine activation in the tropical UTLS" *by* M. von Hobe et al.

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We thank the reviewer for helpful comments and a constructive review. We will address the points raised below, and most of them will be reflected in the revised manuscript.

With the type of measurements this study is based upon, data artifacts can never be ruled out with 100 % certainty. However, the data have been most carefully analyzed and quality checked and if we had any substantial evidence for a data artifact we would not publish the data. As a matter of fact, some peaks in the ClO data (including some in the vicinity of cirrus clouds that would have gone nicely with the story presented in this paper) have been removed during the data analysis due to instrumental conditions for which we could not rule out data artifacts to the best of our knowledge.

Furthermore, although we cannot explain everything we observed (e.g. we are unable to quantitatively simulate the high ClO mixing ratios seen in the case studies using the known trace gas concentrations and process parameterizations), we believe that the evidence that tropical cirrus clouds can cause high ClO is compelling.

On the notion of high ClO normally being present only in aged stratospheric air mentioned in the abstract and summary, a sentence will be added in the discussion of Figure 3, and a reference to the low ClO/Cl_y ratios in Figure 4 will also be made. A notion that observed O_3 abundances are not reproduced in the standard simulations will be added in the abstract.

For our "definition" of tropospheric air masses, we chose a cut-off value of 100 ppb ozone, as will be described in the revised manuscript. More details, including the exact numbers, will also be given for the offset between observed and ECMWF temperatures. The reference for the "typical ice particle radius of 10 μm" is Krämer et al., 2009.

The 10 ppm H_2O as initialization for the CLaMS_BT runs were chosen so that the observed total water measured by FISH at the end of the simulation is reproduced. 10 ppm does not seem so unrealistic taking into account the tropospheric character of the air masses and the presence of ice particles expected to lead to considerable dehydration over the course of the simulation. The latter is supported by the CLaMS_CTM simulations.

For the case studies, we clearly focus on the incidences with highest ClO mixing ratios, because they are "significant beyond doubt". The 5 – 20 ppt points are shown in Figures 4 and 5 (although as ClO/Cl_y ratios rather than ClO mixing ratios), and many of them do show evidence for possible heterogeneous activation. The significance of individual points – at least statistical – can be assessed by the error bars in Figure 3 that do combine accuracy and precision. A notion on this will be added to the caption of Figure 3.

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We do not state that "most enhanced ClO events occur at Ts below 195 K" but that "most events of enhanced ClO are linked to either temperatures < 195 K and/or the occurrence of cirrus clouds during or preceding the flight". The dark red and orange points in Figure 4 at temperatures significantly above 195 K are one of the reasons for the more detailed investigation that led to Figure 5, where we look at temperature histories and indicators for the presence of ice particles other than just temperature. We will reword and rearrange the relevant paragraph to hopefully make this clearer.

In our discussion on the possibility of transport of inorganic chlorine to the TTL directly from the boundary layer, we did adopt the suggested rewording. In addition, we expanded this entire paragraph as suggested in the other reviews.

Concerning our description of the observations in Figure 6, the reviewer is correct that the peaks in the FISH and FSSP data are not always very big, and that there are points in these plots with higher ClO than the background and no indication for ice. We will attempt to be more exact with this discussion. We will also expand the figure caption to better explain the colours and lines in the lower two panels.

The observation of elevated night-time ClO and the corresponding CLaMS_BT results will be discussed in more detail and quantitatively (cf. response to Ross Salawitch's review). In Figures 7 and 8, the observations will be added to facilitate comparison between the simulation results and the actual data. Concerning the statements on the chemical conversions and the relative importance of individual reaction chains found in the simulations, they are the result of a detailed analysis of the model output of chemical reaction rates. We will add a corresponding statement in Section 4.

We agree with the reviewer that we do indeed have to go through some "gymnastics" in order to reach agreement between our CLaMS_BT simulations and the observations. However, we do comment on each individual assumption made in Section 4 as to how realistic it is. In our conclusions we clearly state the difficulties with reaching this agreement and the discrepancies with some of the observations, coming to the conclusion that "the understanding and, consequently, model implementation of the relevant processes in the TTL is yet incomplete". Given the well known uncertainties that do exist for some trace gas concentrations and reaction rates altered in our simulations, this statement does not seem unwarranted.

On the technical side, all the corrections suggested by the reviewer will be implemented in the revised manuscript. The font size in the figures will be increased and a general reference to ACE-FTS will be given.

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