

Response to the Reviewer 1

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We deeply thank the reviewers for their works and constructive comments on our article. Please find below our general response followed by the responses to each comment.

We found that both reviewers pointed out the weakness in the comparison methodology of the original submitted manuscript. To improve the quality of the paper, we applied the following major updates on the manuscript:

- Ground-based measurements are newly added for the comparison. We would like to include B. J. Connor, who is responsible for the analysis of the ground-based CIO measurements, as a coauthor of this paper. In order to make the paper title consistent with the updated content, we would like to change it to “**Comparison of SMILES CIO profiles with satellite, balloon-borne, and ground-based measurements**”.
- Comparison of the SMILES and Envisat MIPAS CIO profiles has been updated using coincidences of the two datasets while the zonally-averaged data comparison has been removed. With this change, all the comparisons described in this paper are now done with the same comparison methodology, which makes the discussion more straightforward to understand.

We believe these major changes will improve the quality of the article, and they make the paper more useful for readers.

The responses to the individual comments are described in detail below. The original comments from the reviewers are cited with an *italic* style.

#1 Specific comments :

First, there are two competing algorithms employed for the analysis, only one of which is addressed here. Some of the differences between the algorithms (for example, spectroscopic parameters) should be resolved internally, or at least assessed in detail. It should not be left to the data user to assess how much error is due to which choice.

The SMILES level-2 (L2) processing in NICT (hereafter SMILES NICT L2 data) is

being developed not in a competitive stance against that of JAXA, but rather as an independent research activity to investigate different retrieval strategies. The motivation for having several developments is to try to exploit the full potentiality of the SMILES instrument. For the validation of these L2 products, the SMILES mission team made a policy that the validation studies should be performed independently for each L2 processing since they are separate activities. The target of this paper is to evaluate the quality of ClO profiles from the SMILES NICT L2 processing (as mentioned in the manuscript) and we do not intent to discuss which of the NICT or JAXA L2 product would be better-recommended for data users. In this version, unlike the JAXA product, the NICT L2 processing mostly targets the altitudes above ~25 km. Altitudes below have been less prioritized because of some calibration issues that have a strong impact in the lower altitudes. This statement is added in the manuscript at Section 2.2.

We included the comparison between NICT and JAXA's ClO profiles in this manuscript as a first-step check of the general consistency between these two datasets. A very good agreement in the stratosphere and mesosphere is found. The evaluation of the differences between the two processing is stated in the paper (Sect. 4) as one of the main target of this study. Hence, unlike stated by the reviewer, we believe that the paper does really help the users for understanding the difference between the retrieval, and the impacts of these differences are clearly described in term of bias, standard deviation, and vertical resolution (Sect. 4, and Fig. 5). The discussion also includes the spectroscopic parameters.

Second, the use of a single a priori profile always and everywhere is clearly causing a significant, entirely unnecessary, error in some circumstances, as the authors themselves freely acknowledge.

For the current version of the SMILES NICT L2 processing, a single a priori state was used because we wanted to avoid variability induced by ClO a priori. The information about a priori contamination is provided with the data, and the error analysis presented in this paper shows that errors due to the ClO apriori is negligible where the a priori contamination is less than 10% at most of the interested altitudes (about 20–80 km and 20–70 km for the daytime and nighttime mid-latitude conditions, respectively). Activated ClO in the polar vortex is an exceptional case (a priori contamination is estimated as ~35%), but for such a case data users can take into account this error.

Third, the narrow spectral region considered also causes errors which could be avoided or minimized. Both my second and third points will be addressed in the next version of the data set, according to the authors, at which time I would think a new validation paper would be submitted. Thus I question the need for this one.

The use of narrow spectral region is adopted in the NICT L2 v2.1.5 processing in order to reduce the errors due to the contamination of other opacity sources when retrieving the ClO at the stratosphere and mesosphere. We are aware that such a narrow bandwidth brings disadvantages for some specific conditions (e.g., altitudes below about 20 km and when ClO is enhanced at the polar lower stratosphere, as we described in the manuscript). But since the original motivation of the NICT L2 v2.1.5 processing was to analyze the SMILES data at the stratosphere and mesosphere, we adopted this retrieval strategy. In the revised manuscript, we added some more explanation about the sensitivity decrease due to the use of the limited spectral bandwidth (Appendix A).

The future versions of the NICT L2 processing is planned to include the lower stratosphere for the target of the retrieval analysis by using the spectra over the full bandwidth (1.2 GHz). The good agreement of the current NICT L2 data with the JAXA product indicates that the quality of retrievals in the stratosphere and mesosphere will not be improved so drastically compared to the current version (v2.1.5). Therefore we believe that publishing the validation paper at this timing is reasonable. In addition, there are already several scientific papers which include the current SMILES NICT ClO data for their researches (e.g., Khosravi et al. (2013); Kuribayashi et al. (2013); and Sugita et al. (2013)). Having a reference paper about the data quality is considered to be useful for the science community.

#2 Comments on the comparison methodology :

First, the use of averaging kernels to compare different data sets is justified and reasonably well described, but it does not completely eliminate error due to the measurement's vertical resolution and use of different a priori profiles. There seems to be no attempt made to assess the remaining uncertainty. Further, the use of Eq (1) is justified if one measurement's resolution is \gg than the other's. But Fig 5 suggests the difference is no more than a factor of 2. Again, what error

remains in the comparison? Finally, the use of triangular smoothing functions rather than averaging kernels in some comparisons (p 626 l. 13) seems arbitrary, and its impact is also not quantified.

For the comparison with other microwave/submm limb sounders (MLS and SMR), the difference in the averaging kernel and a priori assumption are not considered. This is because the a priori contamination is considered very small. The average good agreement shown in these comparisons indicates that the a priori error is not a significant issue.

The comparison with the Envisat MIPAS is now updated to comparing the coincidence profiles. For this, we considered the differences in the vertical resolution and the a priori assumptions for both datasets (the triangular smoothing function is no longer used). The difference is within the expected systematic errors due to the instrument functions and spectroscopic parameters.

#3 Comment about using ground-based observations as comparison dataset :

The authors say they do not compare to ground-based measurements because of the “large difference in sensitivity”. But CIO measurements by the ground-based radiometer at Mauna Kea, Hawaii have accuracies very similar in the mid-stratosphere to those claimed here for SMILES, namely 22–40 pptv in 20–40 km (Solomon et al, 2006). Those measurements have been made continually over 30 years, and near continuously for the last 20. Further SMILES emphasizes its diurnal variation measurements. The first such were made in the stratosphere from Mauna Kea 30 years ago, and are arguably as good as any such measurements available at present for a single location (Solomon et al, 1984). The data from the Mauna Kea instrument and its sibling in Antarctica have been used extensively for validation by the Aura and UARS MLS teams (Nedoluha et al, 2011, Santee et al, 2008, Connor et al, 2007). Thus I think there is a compelling case for their use in a study such as this, which otherwise seems to include an exhaustive list of CIO instruments.

We fully agree with this comment that the ground-based CIO observations are very important in the atmospheric researches, particularly in studying long-scale temporal trends of CIO. Following this comment, now we included the ground-based data into the analysis. Also we included the reference papers of the ground-based observations in the introduction section.

About the diurnal variation observation of SMILES, what we intended to mention in the original manuscript is that SMILES has the capability of detecting the “global” distribution of ClO diurnal variations at a “wider” altitude range including the mesosphere where, to our knowledge, the microwave ground-based instruments do not have good sensitivity and the possibility to get independent points at different heights.

#4 Other comment :

P 618, 17: 'latitude range shifted' -- from what to what?

It is from 65°S to 38°N. Manuscript is updated.

References:

Khosravi, M. et al. (2013), “Diurnal variation of stratospheric HOCl, ClO and HO₂ at the equator: comparison of 1-D model calculations with measurements of satellite instruments”, *accepted to Atmos. Chem. Phys.* (the discussion paper was published as *Atmos. Chem. Phys. Discuss.*, **12**, 21065-21104, 2012)

Kuribayashi et al. (2013), “Direct estimation of the rate constant of the reaction ClO + HO₂ → HOCl + O₂ from SMILES atmospheric observations”, *Atmos. Chem. Phys. Discuss.*, **13**, 12797-12823.

Sugita et al. (2013), “HCl and ClO profiles inside the Antarctic vortex as observed by SMILES in November 2009: Comparisons with MLS and ACE-FTS instruments”, *Atmos. Meas. Tech. Discuss.*, **6**, 6729-6765.