Reply to Anonymous Referee #1

Thank you very much for the valuable comments and questions. We deeply appreciate that. We have improved the manuscript according to your comments, and also underwent careful spell-check and proof-reading by the native speakers. Furthermore we plan to have a final English correction by another native speaker after revision of all the scientific comments by all the reviewers.

General comments

[Question and comment 1] ----------
(a) I think the discussion of the diurnal cycle of ClO can be better presented. Especially the discussion of the mesospheric diurnal cycle seems incomplete – why is it reversed? Where does the maximum at 70 km come from?

[Our answer]
The diurnal cycle of mesospheric ClO is strongly correlated with that of mesospheric ozone. In the mesosphere, ozone is photo-dissociated and atomic O radical is generated in daytime. Therefore the amount of ClO is decreased by the reaction R1 (ClO + O -> Cl + O2). In nighttime, the amount of ozone is increased and that of ClO is also increased by the reaction R2 (Cl + O3 -> ClO + O2). We added the description for the ClO enhancement in our manuscript.

[Modification in the manuscript]
p. 4689 l. 28 " The “ClO mesospheric enhancement” seems to start in October–November 2009 and fade in March–April 2010." -> "The "ClO mesospheric enhancement" that is located close to the tertiary O3 maximum (Marsh and Smith, 2001) seems to start in October-November 2009 and fades in March-April 2010. ClO is enhanced through the reaction (R2) because of the O3 enhancement."

References "Marsh, D. and Smith, A.: The existence of a tertiary ozone maximum in the high-latitude middle mesosphere, Geophys. Res. Lett., 28, 4531-4534, 2001." was added.

[Question and comment 2] ----------
(b) For the observed ClO enhancements, a test of significance should be carried out, and stated

precisely.

[Our answer]
We observed the ClO enhancement at 70 km with an amplitude of more than 100 pptv. We calculated the total error at 70 km to be less than 20-30 pptv when more than 100 profiles are averaged (please see Fig. 9). The numbers of averaged profiles in an SZA bin of 10 deg where the ClO enhancement occurs are larger than 100. The enhancement amplitude is three times larger than the estimated total error at 70 km. Therefore we conclude that the observed ClO enhancement is significant.

[Modification in the manuscript]
p. 4690 l. 1 " The amplitude of ClO enhancement is about 100 pptv, which is larger than the total error of 20–30 pptv at 70 km averaging 100 profiles." -> "The amplitude of the ClO enhancement is about 100 pptv and it is calculated with averaging more than 100 profiles in an SZA bins of 10 deg. The total error at 70-80 km is estimated to be 20-30 pptv by the error analysis, which is three times smaller than the amplitude of the ClO enhancement. Therefore the observed ClO enhancement at 70-80 km is significant."

[Question and comment 3] ----------
(c) I also found quite a large number of typos and/or language errors. All three points are discussed in more detail in the list attached below, which also includes a couple of minor comments.

[Our answer] and [Modification in the manuscript]
Thank you very much for your careful English corrections. We would like to answer your comments individually as follows.

Comments on context

[Question and comment 4] ----------
Page 4670, Line 24: has the diurnal variation of mesospheric ClO been observed by ground-based or airborne instruments yet?

[Our answer]
In the case of ozone, the diurnal variation has been observed by ground-based instruments (e.g., Zommerfelds et al., 1989) up to 74 km. But not for the mesospheric ClO. UARS/MLS produced the ClO diurnal variation at 53 km (Ricaud et al., 2000) although it was noisy. For an example of the ground-based observations, there is a very recent work made by Kuwahara et al., (2012). It measured ClO diurnal variation up to 60 km. We believe that our measurements are the first observations of ClO diurnal variations up to 70-80 km. We changed the statement as follows.

Kuwahara, T., Nagahama, T., Maezawa, H., Kojima, Y., Yamamoto, H., Okuda, T., Mizuno, N., Nakane, H., Fukui, Y., and Mizuno, A.: Ground-based millimeter-wave observation of stratospheric ClO over Atacama, Chile in the midlatitude Southern Hemisphere, Atmos. Meas. Tech. Discussion, 5, 1907–1945, doi:10.5194/amtd-5-1907-2012, 2012

Ricaud, P., Chipperfield, M. P., Waters, J. W., Russell III, J. M., and Roche, A. E.: Temporal evolution of chlorine monoxide in the middle stratosphere, J. Geophys. Res., 105, 4459–4470, doi:10.1029/1999JD900995, 2000. 4688, 4709

Zommerfelds, W. C., K. F. Kunzi, M. E. Summers, R. M. Bevilacqua, D. F. Strobel, M. Allen, and W. J. Sawchuck (1989),Diurnal Variations of Mesospheric Ozone Obtained by Ground-Based Microwave Radiometry, *J. Geophys. Res.*, *94*(D10), 12,819–12,832, doi:10.1029/JD094iD10p12819.

[Modification in the manuscript]
p. 4670 l. 24 "We observed the global diurnal variation of mesospheric ClO for the first time." -> "The diurnal variation of mesospheric ClO up to 80 km is observed by SMILES for the first time from space."

[Question and comment 5] ----------
Page 4680, discussion of m in Figure 3: can you discuss what it means that m (the sum of the averaging kernel elements at one altitude) is larger than one over nearly the whole altitude range?

[Our answer]
In our definition, the measurement response is computed as the summation of absolute values of each element in the averaging kernel (See Eq. (19)). As there is no constraint on the area of the

averaging kernel (unlike the Backus-Gilbert method, see Rodgers, 2000), a measurement response can be larger than one. In our experience it is found that measurement responses can be slightly larger than one (the value is smaller than 1.2) when the wings of averaging kernel are oscillating around zero. There is no physical meaning in the measurement response being larger than one, but it is mathematical artifact. Oscillations in the wings of averaging kernels are found when the width of the averaging kernel (i.e. vertical resolution of the retrieved profile) is larger than the retrieval altitude grid spacing. Note that for limb sounding we are taking into account only averaging kernel close to the Dirac peak and errors related with the small a priori contamination are taken into account in the analysis.

Rodgers, C. D.: Inverse Methods for Atmospheric Sounding: Theory and Practice, Series on tmospheric, Oceanic and Planetary Physics, vol. 2, World Scientific, Singapore, 3605–3609, 2000. 4678

[Modification in the manuscript]
Nothing was changed.

[Question and comment 6] ----------
Page 4687, Line 20: As far as I see, the vertical resolution of SMILES is comparable to MLS and Odin at 10 hPa, better than MLS and Odin at 2 hPa.

[Our answer]
Your suggestion is right. We changed the statement as follows.

[Modification in the manuscript]
p. 4687 ll. 19-20 "The vertical resolution of SMILES is larger than those of Aura/MLS and Odin/SMR." -> "The vertical resolution of SMILES is comparable to those of Aura/MLS and Odin/SMR."

[Question and comment 7] ----------
Page 4688, Line 17 ff, line 22: “qualitatively reliable” is a term that does not appear to have a clearly defined meaning. What you do show is that the observation of stratospheric diurnal variation is consistent with the observations of MLS / UARS within error bars; what you also should show is whether the observation is significant at a given confidence limit. Same goes for

the mesospheric diurnal variation.

[Our answer]
We changed the statements as follows.

[Modification in the manuscript]
p. 4688 ll. 19-22 "Moreover, there is good agreement between the behaviors of the diurnal variations in the ClO VMR for the stratosphere deduced from SMILES observations and UARS/MLS observations as shown in Fig. 3. We conclude that the ClO diurnal variations observed by SMILES are qualitatively reliable." -> "Moreover, the behaviors of diurnal variations in the ClO VMR for the stratosphere deduced from SMILES and UARS/MLS observations gave a good agreement within their 1-sigma standard deviations as plotted in Fig. 10, although the artificial bias was added to the UARS/MLS data."

[Question and comment 8] ----------
Page 4689, Line 1: To show whether the variation is realistic, you should show whether it is significant (at the 90%, 95%, 99%? confidence limit at least).

[Our answer]
The interval of color contour levels of Fig. 11 is set to be 25 pptv, which corresponds to the value of total error estimated in our error analysis when 100 profiles are averaged. The contour plot of Fig. 11 was made with averaging more than 100 profiles in each SZA bin of 10 deg except for the area where there are few observations because of the orbit of SMILES. Please note that the numbers of averaged profiles in the SZA bin are shown at the top of each panel. Therefore the difference of colors (discretized colors) in Fig. 11 is significant. But as you point out, the current manuscript was not clear for that. We changed the statements as follows.

[Modification in the manuscript]
p. 4688 l. 27 - p. 4689 l. 1 "The interval of color counter levels is representative of a VMR of 25 pptv, which is the total error value estimated in this study for averaging 100 profiles. Thus, the variations apparent in Fig. 11 are considered to be realistic." -> "The contour intervals in Fig. 11 are 25 pptv, which is the total error estimated for an average of 100 profiles. In Fig. 11, each SZA bin of 10 deg was calculated by averaging more than 100 profiles except for the bins where there were fewer observations because of the orbit of SMILES. The ClO features observed by SMILES shown in Fig. 11 are significant."

[Question and comment 9] ----------

Page 4689, Line 20, 21: what about photolysis of ClO? Does that play a role?

[Our answer]

As you mentioned, ClO is photo-dissociated in the middle atmosphere. But the reaction R1 (ClO + O -> Cl + O2) is faster than the ClO photolysis.

[Modification in the manuscript]

Nothing was changed.

[Question and comment 10] ----------

Page 4689, Line 24, 25: What is the reason of the reversed diurnal cycle in the mesosphere? If it was predicted by models, the reason should be known, and should be stated here.

[Our answer] and [Modification in the manuscript]

Please see [Question and comment 1]

[Question and comment 11] ----------

Page 4689, Line 26 ff: This ClO maximum at _70 km seems to appear right at the place where the third ozone maximum is observed (see, e.g., Marsh et al., GRL, 2001), so the reason might be that reaction R2 works faster in the presence of enhanced O3 values.

[Our answer] and [Modification in the manuscript]

Please see [Question and comment 1]

[Question and comment 12] ----------

Page 4690, Line 1-3: again, how significant is the observed ClO enhancement?

[Our answer] and [Modification in the manuscript]

Please see [Question and comment 2]

[Question and comment 13] ----------
Page 4690, Line 17: “... was reasonably explained ...” is not a very precise formulation.
Better would be “... is consistent with ...”

[Our answer]
We changed the statement according to your suggestion as follows.

[Modification in the manuscript]
p. 4690 l. 17 "The behavior of the diurnal variation in ClO was reasonably explained by known diurnal chemistry." -> "The behavior of the diurnal variation in ClO is consistent with known diurnal chemistry."

[Question and comment 14] ----------
Page 4690, Line 18: “... from the stratopause to the mesosphere ...” could be a really small step. What you mean is “... from the stratopause well into the mesosphere, to altitudes of more than 70 km”.

[Our answer]
We changed the statement according to your suggestion as follows.

[Modification in the manuscript]
p. 4690 ll. 18-19 "The global diurnal variation of ClO from the stratopause to the mesosphere were obtained for the first time by the SMILES observations." -> "The global diurnal variations of ClO from the stratopause well into the mesosphere, to altitudes of more than 70 km, were obtained for the first time by using SMILES observations."

[Question and comment 15] ----------
Page 4690, Line 22: Again, you should show whether these observations are significant, and then state the significance here instead of the reliability (which is not a well-defined term).

[Our answer]
We changed the manuscript according to your suggestion as follows.

[Modification in the manuscript]

p. 4690 ll. 21-22 "The quantitative error analysis shows that these ClO features are reliable." -> " The quantitative error analysis provided here indicated that these ClO features were atmospheric in nature."

Language

[Question and comment 16] ----------

Abstract: the abstract might be more concise if the order was different: (1) line 1-6; (2) line 13-23, (3) line 7-12.

Abstract, Line 7/8: "... stratospheric ClO, with an enhancement during daytime..."

Abstract, Line 9: " ... a different ..." –> "... the opposite ..."

Abstract, Line 20: "... with averaging ..." –> " ... when averaging ..."

Page 4669, Line 2: "... ClO activation on PSC surfaces ..."

Page 4669, Line 20: "... a sensitive observation of the diurnal variation of short-lived ..."

Page 4670, Line 13: " ... of the antenna ..."

Page 4670, Line 20: "... sturdy ... " –> "... study ... "

Page 4670, Line 22: "Stratospheric diurnal variations observed by SMILES are compared to those observed by UARS / MLS. The diurnal variation of mesospheric ClO is observed by SMILES for the first time from space."

Page 4671, Line 22: "... depends on the temperature ..."

Page 4673, Line 3: "An error in the ClO VMR ..."

Page 4673, Line 23: "... brightness temperature, Ochiai et al., 2012b)."

Page 4674, Line 17: "... of the antenna beam pattern."

Page 4674, Line 19: "... of the SBS is ..."

Page 4675, Line 21 – 25: this sentence is very long – maybe make a full stop after the first "respectively".

Page 4677, Line 7: "...is the transmission coefficient of SWM, ..."

Page 4677, Line 11: "... have identical surfaces with -.."

Page 4679, Line 2: "We use measurements whose tangent heights ..."

Page 4681, Line 8: "... sturdy ..." –> "... study ..."

Page 4681, Line 14: "... the reference spectrum ..."

Equation 29: try to use same size brackets for both terms

Page 4683, Line 18: "... for the random error Erandom and the systematic ..."

Equation 32: it would look better if the '2's were on the 'E's, not on the brackets

Page 4684, Line 24: “The error from the dry-air ...”
Page 4685, Line 19: “Comparing these three instrument functions, ...” (?)
Page 4685, Line 20: “..., the error from the SBS has only a small contribution.”
Page 4688, Line 1: “Two months of data from SMILES were ...”
Page 4688, Line 11: “The night-time ClO VMR values are near zero ...”
Page 4689, Line 18: “The stratospheric ClO amount is controlled by the following reactions ...”
Page 4690, Line 6: “.. the error in the ClO L2r ...”

[Our answer] and [Modification in the manuscript]
We changed the manuscript according to your comments. Please see the modified manuscript.

[Question and comment 16] ----------
Figure 4: the cyan symbols are difficult to see, maybe use larger symbols or a different color.

[Our answer] and [Modification in the manuscript]
Fig. 4 We changed the symbol size larger and used solid line. The same goes to figures 5, 7 and 8. We are also pointed out the color in figure by L. Froidevaux as "Figure 9: The color you refer to as “yellow” is not that close to yellow… (and yellow is not a good color choice anyway)." We changed the colors for the total systematic and random errors in figures 4, 8 and 9.

[Question and comment 17] ----------
Figure 10: is the offset added to UARS or to SMILES?

[Our answer]
The offset is added to UARS/MLS. We would change the sentence in the caption of Fig. 10 as follows.

[Modification in the manuscript]
Caption of Fig. 10 "We add arbitrary offsets of 100, 200, 400, 200 and 100 pptv at 0.46, 1, 2.1, 4.6 and 10 hPa, respectively, since UARS/MLS data have a negative bias." -> "We respectively added arbitrary offsets to UARS/MLS data of 100, 200, 400, 200 and 100 pptv at 0.46, 1, 2.1, 4.6 and 10 hPa, since UARS/MLS data have a negative bias."

[Question and comment 18] ----------

Figure 11: this figure is too small.

[Our answer]

Fig. 11 may be small to see as you mentioned. But we strongly would like to show all the panels in one figure. Please see the attached pdf file of the revised manuscript. We guess Fig. 11 will be printed in this size in the final print. We would like to keep this style.

We found a mistake in Fig. 11 that an unexpected number "39" was printed in the panel of 20S-20N from Jan.-Feb. (2010).

[Modification in our manuscript]

Fig. 11: An unexpected number "39" in the panel of 20S-20N from Jan.-Feb. (2010) was removed.

References

We updated the references as follows.

Manabe, T., Nishibori, T., Mizukoshi, K., Otsubo, F., Ochiai, S., and Ohmine, H.: Measurement of the offset-Cassegrain antenna of JEM/SMILES using a near-field phase-retrieval method in the 640 GHz Band, IEEE T. Antenn. Propag., 60, doi:10.1109/TAP.2012.2201080, accepted, 2012.

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Manabe, T., Nishibori, T., Mizukoshi, K., Otsubo, F., Ochiai, S., and Ohmine, H.: Measurement of the offset-Cassegrain antenna of JEM/SMILES using a near-field phase-retrieval method in the 640 GHz Band, IEEE T. Antenn. Propag., 60, 3971-3975, doi:10.1109/TAP.2012.2201080, 2012.

Marsh, D. and Smith, A.: The existence of a tertiary ozone maximum in the high-latitude middle mesosphere, Geophys. Res. Lett., 28, 4531-4534, 2001.

was added.

Ochiai, S., Kikuchi, K., Nishibori, T., and Manabe, T.: Gain nonlinearity calibration of submillimeter radiometer for JEM/SMILES, J. Sel. Topics Appl. Earth Obs. Remote Sens., 5, 962–969, 2012a.

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Ochiai, S., Kikuchi, K., Nishibori, T., and Manabe, T.: Gain nonlinearity calibration of submillimeter radiometer for JEM/SMILES, IEEE J. Sel. Top. Appl., 5, 962–969, 2012a.

Ochiai, S., Nishibori, T., Kikuchi, K., Mizobuchi, S., Manabe, T., Mitsuda, C., Baron, P., and Ueno, S.: Tangent height accuracy of Superconducting Submillimeter-wave Limb-emission Sounder (SMILES) on International Space Station (ISS), Proc. IEEE Int. Geosci. and Remote Sens. Symp., 1290--1293, 2012b.
was added.

Ochiai, S., Kikuchi, K., Nishibori, T., Manabe, T., Ozeki, H., Mizobuchi, S., and Irimajiri, Y.: Receiver performance of Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) on the International Space Station, IEEE T. Geosci. Remote, submitted, 2012b.
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Ochiai, S., Kikuchi, K., Nishibori, T., Manabe, T., Ozeki, H., Mizobuchi, S., and Irimajiri, Y.: Receiver performance of Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) on the International Space Station, IEEE T. Geosci. Remote, submitted, 2012c.