Reply to Referee #1

Dear Referee #1

We would like to thank you for your helpful comments and improvements on the manuscript. Please find our answer to your comments and the revisions according to your suggestions.

Sincerely yours,

Tomohiro Sato

Note
GC: General comments
SC: Specific comments
MC: Minor comments
AR: Our answer and revision

GC 1-1:

This short paper introduces a new algorithm for the retrieval of ozone isotopic enrichments vertical profiles from the Superconducting Submillimiter Wave Limb Emission Sounder (SMILES). First, the retrieval, forward model and errors are explained. Then a comparison is performed. Followed by a brief discussion.

Although the overall material is publishable, I believe there are a number of points which need to be address before the paper can get into AMT. In particular, the title, the introduction and some of the retrieval intricacies could be explained better.

Furthermore, one major concern is the use of English, there are many sentence that could be rewritten for a more enjoyable reading, the manuscript needs to be thoroughly re-checked, and proof read to improve its readability before the paper is re-submitted.

Please find below my specific comments.

Change title to: Vertical Profile of delta18OOO from Middle Stratosphere to Lower Mesosphere from SMILES Spectra

Introduction comments

Please start by mentioning that there is a heavy ozone anomaly (ie. the measured magnitude of the ozone enrichments are large compared to the expected values).

Perhaps give a bit of history (ie. Cicerone and McCrumb [1980]; Mauersberger [1981]) to give the reader a historical perspective.

Break equation 1 into 2 and add the 100 factor since all the results are given in percentage. (i.e. deltaˆm O = (ˆm R_obs / ˆm R_std -1) * 100. where ˆm R_obs is .... and ˆm R_std ...)

Then explain previous measurements clearly (although briefly), detailing their finding and their measurements caveats rather than quoting Mauresberg, (1981) and then giving the Krankowsky (2007) results (as in p8891 line 5 through 7).

Overall, the SMILES section of the introduction is fine, just some minor comments (that can be found in the minor comments section)

AR to GC 1-1:

Thank you for suggesting many improvements. We revised the title and manuscript following your suggestions. The title was changed to

"Vertical profile of δ^{18}OOO from the middle stratosphere to lower mesosphere from SMILES spectra".

We applied major update on the introduction section including the history of ozone isotope studies. We believe now the manuscript contains a useful introduction for readers.

With respect to the use of English, we went through all the manuscript and improved the languages. We also intend to use the AMT copy edit service before publishing the paper.

SC 1-1:

p8895 line 19: If window b0 is the same as b1, why did you retrieved O3 plus other molecules using a fix pressure and temperature profile if later you were going to retrieved temperature. Please clarify, wouldn't be better to retrieve O3 and temperature from b0 (b1) at the same time.

AR to SC 1-1:

The main target of the TOROROS retrieval is ozone isotopic ratio. VMRs of O_3 and ^{18}OOO were retrieved under the same temperature and pressure profiles. Temperature and pressure profiles were fixed to the a priori in the retrievals of O_3 and ^{18}OOO. Temperature retrieved in b0 is a supplemental information for discussion of ozone isotopic ratio. As you mentioned the explanation in our manuscript was unclear for this point, so we modified the statement as follows.

P 8895 L 19-20: "The frequency range and the retrieved parameters were the same as window b1." → "The frequency range and the retrieved parameters were the same as window b1. The temperature profile retrieved in window b0 is used for a discussion of temperature dependence of $\delta^{18}OOO$."

SC 1-2:

p8897 eq2: You don't need ny or nx to increase the contribution of the apriori you just need to decrease the apriori noise, which is essentially what you are doing with ny and nx. Please delete nx and ny and report the corresponding apriori error .

AR to SC 1-2:

Yes, thank you for this correction. We deleted n_x and n_y from Eq. (2) as follows.

P 8897 Eq. (2):

$$\chi^2 = \{y - F(x,b)\}^T S_y^{-1}\{y - F(x,b)\} + \{x - x_a\}^T S_x^{-1}\{x - x_a\}. \quad (2)$$

P8897 L 7-10: "This definition of ... and **y**, respectively." was deleted.

A priori error is described in page 8899 Eqs. (7) and (8) and corresponding texts.

SC 1-3:

p8898 line 16: include a brief explanation of eq 21 in Worden (2006) or give the full description in the explanation.

AR to SC 1-3:

We added descriptions for Eq. (21) in Appendix A1, and changed the sentence in the main text as follows.

P 8898 L 16: "…(see the explanation of Eq. (21) in Worden et al., 2006)." → "… (see Appendix A1 in detail)"

SC 1-4:

p8899 eq7 and 8: These two equations will be better after eq 4. So when eq6 refers to e_z the reader already know what it is.

For instance change line 9 in p8898 to: The weighting function Kx in the linear scale was projected onto the log scale by (no period) K_z = dy / dz ... as well as, the apriori error (e_x) by (again no period) followed by eq7 and 8. (rearranging the rest of the text appropriately)

AR to SC 1-4:

We re-arranged the statement from page 8898 line 3 to page 8899 line 13 following your comment.

SC 1-5:

p8899 eq10: Why did you need this normalization? Please explain the advantages/disadvantages of doing this.

AR to SC 1-5:

This normalization is needed for a mathematic aspect of the matrix operations (e.g., calculation of inverse matrices) in the retrieval calculation. This normalization is useful for the matrix operations because we have to handle large matrices with large ranges that include several parameters. For example, VMRs of O_3 and ^{18}OOO are simultaneously included in the retrieval state **x** and their values are different with three order, O_3 VMR is about ppm and ^{18}OOO VMR is about ppb.

SC 1-6:

p8900 eq13: please define how did you map Kz (equation 4) to K (in eta space) I understand that the a priori section of the chi2 (eq2) in eta space becomes zero but please clarify for the reader.

Also, in the last iteration the levenberg-marquadt parameter needs to become zero otherwise is introducing a regularization effect that needs to be taken into account for the computation of the averaging kernels and the error covariance matrix. For a description of this effect see Raspollini et al 2013 – Ten years of MIPAS measurements with ESA level 2 processor V6- part 1: Retrieval algorithm and diagnostics of the products.

AR to SC 1-6:

We added the equation,

$$K_\eta = \frac{\partial y}{\partial \eta} = \frac{\partial y}{\partial z}\frac{\partial z}{\partial \eta} = K_z \epsilon_z,$$

after the sentence of page 8900 line 8.

We did not force the Levenberg-Marquardt parameter Γ to be zero at the end of iteration, but actually Γ becomes close to zero at the end of retrieval iteration (approximately less than 0.001).

SC 1-7:

p8901 line3: Please explain the methodology described in Sato (2012) briefly. Line15: Why did you not include the xtrue – xref error? This error give you an approximation of the retrieval error in itself.

AR to SC 1-7:

The difference of $\mathbf{x}_{\mathbf{true}}$ and $\mathbf{x}_{\mathbf{ref}}$ is less than 0.5% in isotopic enrichment δ^{18}OOO at 28-57 km. It is much smaller than other considered errors such as γ_{air} (its error is larger than 4% in δ^{18}OOO), thus we ignored the difference between $\mathbf{x}_{\mathbf{true}}$ and $\mathbf{x}_{\mathbf{ref}}$ in the presented work.

SC 1-8:

p8902 line 12: Define the contribution matrix D. Is this the same D as in Baron et al 2011 or in Sato (2012). Please clarify.

Line17: Define A (the averaging kernel matrix) ie. A = dˆx / dx = DK

AR to SC 1-8:

We defined **D** and **A** in the manuscript by adding the following equation.

P 8902 L 12 "**D** is the contribution function matrix." → "**D** is the contribution function matrix and is given by

$$\mathrm{D} = \frac{\partial \hat{\mathrm{x}}}{\partial \mathrm{y}} = \left(K_x^T S_y^{-1} K_x + S_x^{-1}\right)^{-1} K_x^T S_y^{-1},$$

where $\hat{x}$ is a retrieval solution of **x**."

P 8902 L 17-18: The equation:

$$\mathrm{A} = \frac{\partial \hat{\mathrm{x}}}{\partial \mathrm{x}} = DK$$

is inserted after "**A** is the averaging kernel matrix."

SC 1-9:

p8903 line 3: Sato et al 2012 refers the reader to Baron et al 2002 for a detail explanation of the measurement response. Please summarize here or add the description in the appendix. line 6: what does the m = 0.9 implies? Is this just the integrated area under each kernel? If it is just say: values near unity indicate that most information was provided by the measurements while lower values indicate that the retrieval was influenced by the apriori.

AR to SC 1-9:

The measurement response vector **m** is defined as $\mathrm{m}[\mathrm{i}] = \sum_j |A[i,j]|$. Using the value of **m**, we can estimate contributions of observed spectrum and a priori state to the retrieved state. As you mentioned, the m = 0.9 roughly means 90% contribution from the observed spectrum. We defined the measurement response vector in the manuscript with brief explanation about what the measurement response is.

P 8903 L 2-6: "The measurement response ***m***, that indicates the sensitivity of the observation (see Eq. 19 in Sato et al., 2012), of the b1 O_3 was almost equal to one for all altitudes between 20 and 80 km, thus the retrieved O_3 was less dependent on the a priori VMR. On the other hand, the ***m*** of ^{18}OOO in both windows b2 and c1 was larger than 0.9 at altitudes between 28 and 62 km." → "The measurement response vector ***m*** is given by

$$\mathrm{m}[\mathrm{i}] = \sum_j |A[i,j]|\,,$$

and indicates a sensitivity of the observation to the retrieved result (see Eq. 19 in Sato et al., 2012 and reference therein), i.e., values of ***m*** near unity indicate that most information in the retrieval results is provided by observations while low values of ***m*** indicate that the retrieval results are largely influenced by the a priori state and forced to be identical to the a priori values. The ***m*** values of b1 O_3 were almost equal to unity for all altitudes between 20 and 80 km. For ^{18}OOO in the windows b2 and c1, the ***m*** values were larger than 0.9 at altitudes between 28 and 62 km."

SC 1-10:

Line25: Please explain why for 18OOO in window b2 the gamma_air error increased compared to V215, particularly around 30 km. Is this also due to the fix vertical grid that helped reduced the gamma_air error in O3. Please clarify.

AR to SC 1-10:

To investigate a vertical grid dependence of the error in b2 ^{18}OOO VMR due to the γ_{air} uncertainty, we performed the error analysis for the γ_{air} uncertainty in TOROROS retrieval with different vertical grid: i.e. the one used for the V215 processing. Figure SC 1-10 shows the result and there is no significant difference in the errors for the two vertical grid conditions: the TOROROS grid (T-grid) and the V215 grid (V-grid). This suggests that the vertical grid is not a cause of the issue that you pointed out.

One possible explanation for the different behaviors in the error from γ_{air} between TOROROS and V215 is an effect of O_3 in the ^{18}OOO retrieval. The error in ^{18}OOO VMR due to γ_{air} in TOROROS is larger than that in V215 (as you pointed out), which is only the case for Band B and not for Band C. The biggest difference of ^{18}OOO retrieval at near 30 km between Bands B and C is effects from O_3. The O_3 spectral line is overlapping on the ^{18}OOO line which makes difficult to distinguish ^{18}OOO spectral feature from a wing slope of O_3 line. Considering that γ_{air} is effective to describe the width of spectral line, errors originated from γ_{air} can be larger if spectral line width of ^{18}OOO is better distinguished from other interfering spectral features (nearby O_3 line). The ^{18}OOO retrieval in TOROROS employs information from O_3 more effectively than V215 does. This may make TOROROS's ^{18}OOO (in Band B) more sensitive to γ_{air} than V215.

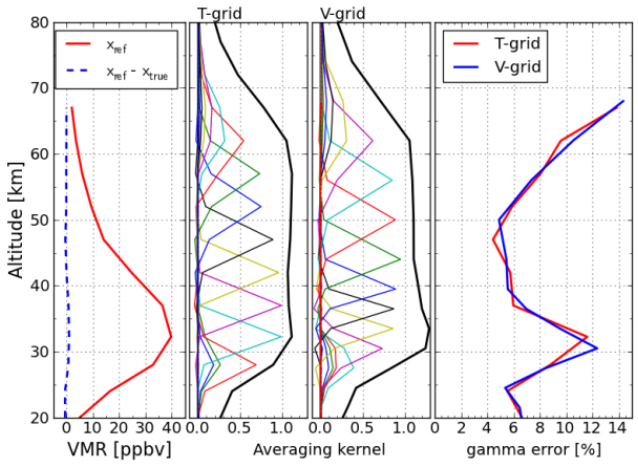

Fig. SC 1-10: Vertical grid dependence of error in b2 ^{18}OOO VMR due to uncertainty in γ_{air} of the ^{18}OOO transition. Reference profile, averaging kernels and the errors are plotted. "T-grid" and "V215" mean the grid of TOROROS and V215, respectively.

SC 1-11:

p8904 line1: The smoothing error drops from 55 to 57 and then sharply increases. Why were the values of Sx multiplied by two above 55km. Did you try not multiplying by two to verify your hypothesis.

AR to SC 1-11:

When this manuscript was submitted to AMTD, we considered that the smoothing error was dropped at 57 km and related it to the use of twice large $\mathbf{S_x}$ above 55 km. But now we regard it as a part of oscillation through altitudes from 25 to 60 km. We newly investigated the cause of such an oscillation in the smoothing error by calculating the smoothing error with and without the cross terms (CT) in $\mathbf{S_x}$. Figure SC 1-11 shows that the oscillation is vanished when CT is removed from $\mathbf{S_x}$. So the drop at 57 km in the smoothing error is not caused from the twice multiplied $\mathbf{S_x}$ above 55 km. We changed the manuscript as follows.

P 8904 L1-2: "The smoothing error dropped off at 57 km. This might be due to the values of $\mathbf{S_x}$ being multiplied by two above 55 km." → "The smoothing error seems to be oscillated, which is due to introducing cross terms in $\mathbf{S_x}$."

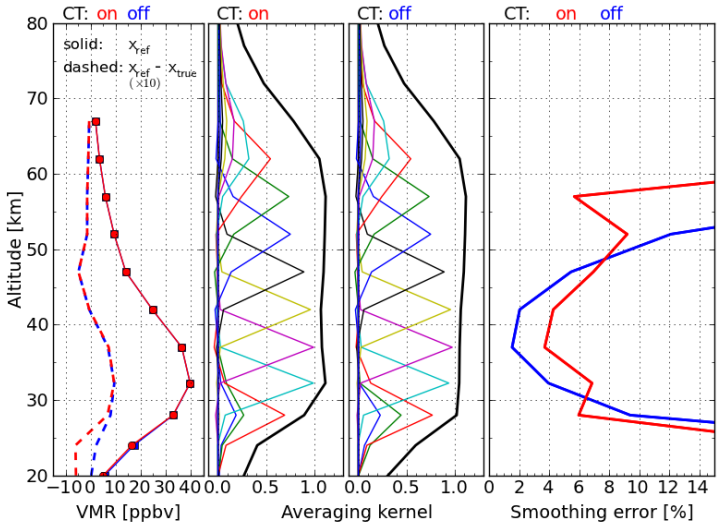

Fig. SC 1-11: Comparison of averaging kernel and smoothing error of b2 ^{18}OOO VMR with and without cross terms (CT) in $\mathbf{S_x}$. "CT: on" is results with CT (same as TOROROS). "CT: off" is results without CT.

SC 1-12:

p8904 line7: The information in figure 3 looks just like the information in figure 2. Is this suppose to be the case?

AR to SC 1-12:

Figure 3 shows the errors in δ^{18}OOO that is calculated from the errors in O_3 and ^{18}OOO VMRs (VMR errors are shown in Fig. 2.). Hence, it makes sense that Fig. 3 seems like Fig. 2. But please note that information in Fig. 3 is the errors in δ^{18}OOO and is different from that in Fig. 2 of the errors in VMR.

SC 1-13:

p8904 line8: Is the consistency you are talking about the similarities between total b2 and c1 errors. Because if that's the case, please clarify what you mean until line 11. line16: The minimum value is 5%.

AR to SC 1-13:

Yes, we improved the manuscripts as follows. Also, the minimum value in line 16 was corrected.

P 8904 L 7-11: "The systematic errors using the b2 and c1 ^{18}OOOs were consistent within 2–3%

above 45 km and increased from 6% (45 km) to more than 10% (> 60 km). At altitudes between 25 and 40 km δ^{18}OOO using the b2 ^{18}OOO had larger systematic error (14%) than that of the c1 ^{18}OOO (4–6 %)." → "The systematic errors using the b2 and c1 ^{18}OOOs showed similar values of 6% at 45 km and increased to more than 10% at altitudes higher than 60 km. But the errors in δ^{18}OOO from the b2 ^{18}OOO were larger than those from the c1 ^{18}OOO of 4–6% at altitudes between 25 and 40 km."

P 8904 L 16: "… values of 4% between …" → "… values of 5% between …"

SC 1-14:

p8906 line23: From 45 to 50 km V215 also shows a decrease please comment.

AR to SC 1-14:

P 8906 L 23-24: "The enrichments of TORORO S showed a decrease at altitudes above 45 km, which is discussed in Sect. 4." → "A decrease of δ^{18}OOO was observed in both TORORO S and V215 between 47 and 52 km, which is discussed in Sect. 4."

MC 1-1:

p8892 line 12: Change "instrument to observe atmospheric" to "instrument that observes atmospheric"

line13: remove "quite"

line15: Change "The signal to noise ratios …" to "For a single scan, the signal-to-noise ratios …"

p8893 line 13: Change "vertical profile observations of delta18OOO …" to "a vertical profile of delta18OOO …"

AR to MC 1-1:

These comments are already taken into account in AR to GC 1-1.

MC 1-2:

p8894 line 5: Consider changing "We used the tangent height after correcting it by a bias offset in TORORO S. The bias offset was estimated by …" to " A tangent height offset was estimated

by …"

AR to MC 1-2:

Referee #2 also commented to these statements. We modified the manuscript as follows.

P 8894 L 5-6: "We used the tangent height after correcting it by a bias offset in TOROROS. The bias offset was estimated by comparing the brightness temperatures observed by …" → "A tangent height offset was estimated by comparing the brightness temperatures observed by …"

MC 1-3:

p8895 line 13, 14 and 15 change "The VMR of X" to "X VMR"

AR to MC 1-3:

P 8895 L 13: "The VMR of O_3" → "O_3 VMR"

P 8895 L 14: "the VMR of ^{18}OOO" → "^{18}OOO VMR"

P 8895 L 15: "The VMR of ^{17}OOO" → "^{17}OOO VMR"

MC 1-4:

p8896:

line 1: Change "We employed the forward model …" to "We employed the v215 forward model (F) with the following improvements."

line 22: change "which was half that of the …" to "which is half of the …"

AR to MC 1-4:

P 8896 L2: "We employed the forward model (F) in V215 with the following improvements." → "We employed the V215 forward model (F) with the following improvements."

P 8896 L 22: "…which was half that of …" → "… which is half of …"

MC 1-5:

p8897:

line 11: Change "Sy is the diagonal matrix with the diagonal elements: : :" to "Sy is a diagonal matrix with elements (0.5K)ˆ2".

line 22: Change "The apriori profiles of pressure and temperature were taken from …" to "The pressure and temperature apriori profiles were taken …"

AR to MC 1-5:

We changed the statements following your suggestions.

P 8897 L 11-12: "**S**y is the diagonal matrix with the diagonal elements of $(0.5K)^2$." → "**S**y is a diagonal matrix with elements $(0.5K)^2$."

P 8897 L 22: "The a priori profiles of pressure and temperature were taken from …" → "The pressure and temperature a priori profiles were taken …"

MC 1-6:

p8898

line 20: instead of having (m=16,18) in the line above and then: i and j in square brackets ... just say: where m is either 16 or 18, i and j indicate vector or matrix indices, h is the altitude vector, and h_c is the correlation length set to 6 km.

AR to MC 1-6:

We changed the manuscript following your comment.

P 8898 L 19-21: "(m = 16,18)

i and *j* in square brackets indicate the index of a matrix or a vector. ***h*** is the vector of the altitude. h_c is the correlation length and was set to 6 km." → "where m is either 16 or 18, *i* and *j* in square brackets indicate the index of a matrix or a vector. ***h*** is the vector of the altitude. The correlation length h_c was set to 6 km."

MC 1-7:

p8902 line25-27: change "The same retrieval grid was employed for all retrieval windows for obtaining the isotopic ratio without any vertical interpolation in TORORO S, while that of V215 was adjusted to optimize each molecule (see Fig. A1)." to: "The same retrieval grid was employed for all retrieval windows to obtained the isotopic ratio without any vertical interpolation in TORORO S, while V215 adjusted the retrieval grid to optimize each molecule (see Fig. A1)."

AR to MC 1-7:

Referee #2 also commented to this sentence. We changed it as follows combining comments

from both reviewers.

P8902 L 25-27: "… windows for obtaining the isotopic ratio without any vertical interpolation in TOROROS, while that of V215 was adjusted to optimize each molecule (see Fig. A1)." → "… windows in order to obtain the isotopic ratio without recourse to any vertical interpolation in TOROROS, while V215 adjusted the retrieval grid to optimize each molecule (see Fig. A1)."

MC 1-8:

p8903 line7: The total systematic o3 is more between 2% and 3% than around 2%.

line22: change: "in window b2 was 5-15%" to "in window b2 varied from 5 to 15%"

p8904 line27: change: "is essential for error in remote sensing..." to "is essential to reduce erros in remote sensing ..."

p8905 line 3: Just say performed using a profile by profile comparison. No need to quote other paper that did the same thing.

AR to MC 1-8:

P 8903 L7: "…error of the b1 O_3 was about 2% …" → "… error of the b1 O_3 was 2–3% …"

P 8903 L22: "…in window b2 was 5-15% …" → "… in window b2 varied from 5 to 15% …"

P 8904 L27-28: "…is essential for error in remote-sensing …" → "… is essential to reduce errors in remote-sensing …"

P 8905 L3-4: "This comparison was performed using an individual profile comparison approach (e.g., Sagawa et al., 2013; Kasai et al., 2013)." → "This comparison was performed using a profile by profile comparison."

MC 1-9:

p8905 Line21: are 1145 the number of profiles for b1 and 1377 the number of profiles for c1? If thats the case please change "The numbers of profiles of 18OOO calculated from the b1 O3 and the c1 18OOO with "good quality" were 1145–1377 in an altitude range between 28 and 57 km."

to

"The numbers of profiles of 18OOO selected for b1 O3 and the c1 18OOO with "good quality" in an altitude range between 28 and 57 km were 1145 and 1377, respectively."

otherwise clarify.

In figure 4 in the VMR difference add the values for the systematic errors, individually and then added, to see if the difference is between those lines (ie. Two blue lines on either side of zero,

two red lines and two total).

AR to MC 1-9:

The numbers of 1145 and 1377 indicate the minimum and maximum numbers of averaged profiles of δ^{18}OOO, filtered by χ^2 and measurement response thresholds, among seven altitude levels in 28–57 km. The numbers of profiles of the b1 O_3 and c1 ^{18}OOO are the same for each altitude level. The values of measurement response vary in altitudes, as shown in Fig. 2, thus the numbers of profiles which satisfy the measurement response criteria are different at each altitude level.

Following your suggestion, we changed Fig. 4 to clarify the relationship between systematic errors and VMR difference of TORORO S and V215. Systematic error in TORORO S VMR was included in Fig. 4. Corresponding this change, Figs. 5, A3 and A4 were similarly updated.

MC 1-10:

p8906 line27: Change "This was in good..." to "This is in good..." line29: please add reference for the atmos observation.

p8907 line16: Change "(the reaction R1)" to "(reaction R1)" line17: Change "the temperature dependence." to "a temperature dependence." line 20: Remove "the" from "Only the nighttime date ..." line 21: change "excepting" to "except at" line 22,23: change: "the positive correlation between the delta 18OOO and the temperature was clearly obtained that the ozone isotopic enrichment is increased as the temperature increases." to: "Clearly, there is a positive correlation between delta18OOO and temperature strongly suggesting the ozone isotopic enrichment increases with temperature."

p8908 line14: either define the wall effect or just say "due to an apparatus artifact."

p8909 line:13: are this results for c1 18OOO? Table 6 says 16% at 32km.

P8910 line5: change: "to the mesosphere" to "to the lower mesosphere"

AR to MC 1-10:

P 8906 L 27: "This was in good agreement with …" → "This is in good agreement with …"

P 8906 L 29: "… with the ATMOS observation." → "…with the ATMOS observation (Irion et al., 1996)."

P 8907 L 16: "… (the reaction R1) …" → "… (reaction R1) …"

P 8907 L 17: "the temperature dependence." → "a significant temperature dependence." (combined with comment from Referee #2, SC 2-21)

P 8907 L 20: "Only the nighttime data …" → "Only nighttime data …"

P 8907 L 21: "… excepting 57 km …" → "…except at 57 km …"

P 8907 L 22-24: "The positive correlation between the δ^{18}OOO and the temperature was clearly obtained that the ozone isotopic enrichment is increased as the temperature increases." → "Clearly, there is a positive correlation between δ^{18}OOO and temperature strongly suggesting the ozone isotopic enrichment increases with temperature."

P 8908 L 14: "… an apparatus effect caused by the wall effect." → "… an apparatus effect."

P 8921 Table 6: "16%" → "13%" (δ^{18}OOO from c1 at 32 km)

P 8910 L 10: "… from the stratosphere to the mesosphere." → "… from the stratosphere to the lower mesosphere."

[Corrections by ourselves (CO)]

CO 1:

A full width at half maximum of the rows of the averaging kernel matrix was added in Figs. 2, A1 and A5 (column a). Corresponding to these updates, the following descriptions were added.

P 8903 L 6: "The FWHM of rows of **A** for the b1 O_3 VMR was about 5 km at altitudes from 20 to 80 km, and those for ^{18}OOOs were increased from 5 to 10 km at altitude levels of 28-57 km. "

P 8921 Table 6 column of VR: "VR^5" → "FWHM of **A**^{5}", "5 km, 5 km, 5 km" → "5 km, 6 km, 9 km" (at altitude level of 32, 42 and 52 km, respectively), "^{5} Vertical resolution" → "^{5} Full width at half maximum of the averaging kernel matrix"

CO 2:

In Fig. 6, a conversion of isotopic enrichment from atmospheric O_2 typical value to SMOW for the data from Liang et al., (2006) was incorrectly performed, thus it was corrected. The error bars of data from Krankowsky et al., (2007) was changed from 2-σ standard deviation to 1-σ to be identical to the others. The legend for the TOROROS data were changed from "SMILES" to "TOROROS".