Reply to Referee #2

Dear Referee #2

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
SC: Specific comments
MC: Minor comments
AR: Our answer and revision

SC 2-1:

—- Title

Suggest "... from *the* middle ... derived *using* a retrieval ..."

AR to SC 2-1:

The improvement of the title was also pointed out by Referee #1. We changed the title as:

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

SC 2-2:

—- Abstract

Line 4/5: "...algorithm specific for the isotopic..." is unclear and clumsy wording. Does it mean you're retrieving the isotope ratio as a state vector element, or, more simply, that this whole retrieval system is geared towards retrievals of isotopic species?

AR to SC 2-2:

It means the latter. In our retrieval, the isotopic ratio is not retrieved as a state vector element and the VMRs of ozone isotopomers and isotopologues are retrieved. We changed the sentence as

P 8890 L 4-5: "… algorithm specific for the isotopic …" → "… algorithm optimized for the isotopic …"

SC 2-3:

Line 14: Add comma after "agreement"

Line 16/17: Reword to "The delta18OOO peak, of 18%, is located at the stratopause"

Line 20: suggest "... wide *altitude* range ..."

AR to SC 2-3:

P 8890 L 14: "… in good agreement with …" → "…in good agreement, with …"

P 8890 L 16-17: "Stratopause is the peak-height of the δ^{18}OOO value, and it rose to 18%." → "The δ^{18}OOO peak, of 18%, is located at the stratopause."

P 8890 L 20: "… a wide range …" → "… a wide altitude range …"

SC 2-4:

—– Page 8891

Line 8: Not sure that "trend" is the right word here, as it's altitude not time we're considering. I suggest "behavior" instead.

Line 18: "Using spectroscopic techniques, aymmetric..."

—- Page 8892

Line 10: "the importance of photochemistry..." Not sure what this is trying to say, was it a surprise that photochemistry was important? Surely everyone would have expected that. Was there something specific about the photochemistry that was important?

Line 14: Suggest "...low-noise spectra, enabling observations at high altitudes with good signal to noise."

—- Page 8893

Line 3: "spectrum" → "spectral"?

Line 5: "above the middle stratosphere" → "in the middle stratosphere and higher" (or do you really mean to exclude the middle stratosphere?)

AR to SC 2-4:

We also received the comments from Referee #1 to improve the introduction. The introduction was revised following the suggestions from both reviewers.

SC 2-5:

—- Page 8893

Line 21: Consider changing "functions" to "model"?

AR to SC 2-5:

We changed the manuscripts following your comments.

P 8893 L21: "functions of SMILES. …" → "models of SMILES. …"

SC 2-6:

—- Page 8894

Line 5/6: Fuse two sentences to give: "...after correcting it by a bias offset estimated by comparing ..."

AR to SC 2-6:

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 …"

SC 2-7:

—- Page 8894

Line 12-14: Not sure this last sentence adds anything, consider deleting.

AR to SC 2-7:

P 8894 L 12-14: The sentence "The corrected … limited frequency ranges." is deleted.

SC 2-8:

Line 16: suggest "As mentioned in Sect. 1, only two of the three bands can be simultaneously observed..."

Line 19: suggest adding comma after "study"

Line 20/21: "depending on the other band" - this discussion feels unclear, at least to me.

AR to SC 2-8:

P 8894 L 16-22 A paragraph that starts with "As mentioned in Sect. 1, …" is modified as follows.

"As mentioned in Sect. 1, only two of the three bands can be simultaneously observed. The AOSs assigned to Bands B and C are fixed, i.e., the observations of Bands B and C are always performed by AOS 2 and AOS 1, respectively. Band A is observed by either AOS 1 or AOS 2. We used only data of the observation from Bands B and C for this study, so as not to cause any undesirable errors due to instrumental differences with the band configuration."

SC 2-9:

—- Page 8895

Line 1-2: I actually don’t understand why "retrieving the VMRs of O3 and 18OOO with different weights due to the difference of the spectral line intensities" would be a problem? Why do you not want to do this. A more detailed explanation and motivation here would help.

AR to SC 2-9:

As we can see from SMILES observation spectrum (Fig. 1), the spectral intensity of O_3 is much larger than that of ^{18}OOO (i.e., higher measurement sensitivity to O_3) and the ^{18}OOO line is located at the wing slope of the O_3 line. If O_3 and ^{18}OOO are simultaneously retrieved, a solution tends to preferably fit the O_3 spectral line rather than ^{18}OOO because of difference in their spectral intensities mentioned above. We revised the explanation as follows.

P 8894 L 24-P8895 L 2: "Setting windows with ... line intensities." → "Setting windows with a small frequency range has an advantage of reducing contaminations from transitions of molecules different from the target. In case of spectral lines of O_3 and ^{18}OOO in Band B, the spectral intensity of O_3 are much larger than that of ^{18}OOO and the ^{18}OOO line is located at the wing slope of the O_3 line. If their VMRs are simultaneously retrieved, a retrieval solution is converged to one that is optimized to O_3 rather than ^{18}OOO. Hence we set retrieval windows individually for O_3 and ^{18}OOO (window b1 and b2, respectively). The retrieval processes of the four windows were independent of each other to prevent any error propagation from window to window."

SC 2-10:

—- Page 8895

Line 5: suggest moving "were" to after "retrieved"

Line 6-7: suggest "the spectrum..." -> "a first-order polynomial representing a spectral baseline"

Line 13-14: suggest "spectral" rather than "spectrum"

Line 17: again "spectrum" -> "spectral"?

AR to SC 2-10:

P 8895 L 4-5: "Other parameters were simultaneously retrieved; …" → "Other parameters simultaneously retrieved were ..."

P 8895 L 6-7: "…the spectrum baseline of the first-order polynomial function." → "a first-order polynomial representing a spectral baseline."

P 8895 L 13-14: "… retrieved to fit a spectrum baseline." → "… retrieved to fit a spectral baseline."

P 8895 L 17-18: "… retrieved for spectrum baseline …" → "… retrieved for spectral baseline …"

SC 2-11:

—- Page 8896

Line 14-16: Took me a while to realize you're talking about accounting for the movement of the antenna during integrations. Consider rewording along those lines. Just calling it a "widening" doesn't make it clear where it's coming from.

AR to SC 2-11:

We changed the statements as follows.

P 8896 L 14-16: "Second, R_{ANT} was widened taking into account the accumulation of atmospheric limb emissions over 0.5 s (six steps of the antenna moving with a stepping rate of 12 Hz) to generate a spectrum at one tangent height." → "Second, considering a movement of the antenna over 0.5 s, which is the data integration time for one limb emission spectrum, the instantaneous antenna response pattern R_{ANT} was modified to represent the actual SMILES limb scan mechanism."

SC 2-12:

—- Page 8897

Equation 2: Your definition of chi-squared is unconventional, as you point out. If you'd wanted to weight the a priori more, why not simply have a tighter a priori covariance matrix? Furthermore, it's not clear that your retrieval is really minimizing this chi-squared. If it were, I would have expected to see 1/nx and 1/ny terms in equation 13. If it is truly equation 13 you're using to arrive at the solution, then I would contend that you've found the minimum in the conventional chi-squared (i.e., the one without the division by nx and ny). Please clarify this discussion. If indeed there are additional 1/nx 1/ny terms lurking about in the algebra, have you checked that they don't extend to things like the averaging kernels also.

AR to SC 2-12:

We agree with your comment, thank you for pointing it out. Our retrieval strategy is more constrained by a priori knowledge, thus we set a tight $\mathbf{S_x}$ (for example, introduction of cross terms). Our retrieval calculation is performed as Eq. (13), thus it leads a solution that minimizes χ^2:

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

The normalization of χ^2 in Eq. (2) was quite misleading and the Eq. (2) was corrected as the one without normalization.

We are sure that, for the iteration process in retrieval, χ^2 is utilized in a relative sense not an

absolute value. We confirmed that a way to normalize χ^2 is almost independent to a retrieval solution for the case of the data presented in this study. We changed the manuscript as follows.

P 8897 Eq. (2):

$$\chi^2 = \{y - F(x,b)\}^T S_y^{-1}\{y - F(x,b)\} + \{x - x_a\}^T S_x^{-1}\{x - x_a\}. \quad (3)$$

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

P8905 L10-19: "In this study ... were also removed." → " In this study we selected data, that is regarded as "good quality", by χ^2 and measurement response $\boldsymbol{m}$. About 20% data with larger χ^2 values were removed, and only the retrieved data at altitude levels that satisfy $0.9 < \boldsymbol{m} < 1.2$ were used. Data from L1b data that included any visual field disturbances were also removed."

SC 2-13:

—- Page 8898

Line 6: Not sure what "dispersion" means in this case, would "divergence" be better?

AR to SC 2-13:

We used "dispersion" with a meaning of variation due to measurement errors from spectrum noise and other sources of random error.

P 8898 L6 "… dispersion." → "… variations due to measurement errors."

SC 2-14:

—- Page 8900

Equation 13: So, the Gamma, Marquardt-Levenberg parameter doesn't change from iteration to iteration then? That seems quite unconventional to me. Is the retrieval really that non linear that you have to use such an unconventional approach to reach your solution. Granted, if the chi-squares are good, then the solution is probably a perfectly good one, even if you got there a round about way.

AR to SC 2-14:

Thank you for pointing out our incorrect description. Actually, the Γ varies in the iteration process to be increased and decreased by multiplying a factor of 2 and 1/2 for Band B and 5 and 1/5 for Band C. We changed the sentence as follows.

P 8900 L 9-10: "The Levenberg–Marquardt parameter Γ was tuned to 2 or 1/2 and 5 or 1/5 for Band B (windows b0, b1 and b2) and Band C (window c1), respectively." → "The Levenberg–Marquardt parameter Γ was increased or decreased during the retrieval iteration by being multiplied by 2 or 1/2 and 5 or 1/5 for Band B (windows b0, b1 and b2) and Band C

(window c1), respectively."

SC 2-15:

—- Page 8901

Line 27: "... conservatively estimated as being twice the size of those for the O3 line, considering..."

—- Page 8902

Line 26: "for obtaining" → "in order to obtain the isotopic ratio without recourse to any vertical..."

—- Page 8903

Line 12: Suggest "This is because of" → "This improvement derives from"

AR to SC 2-15:

P 8901 L 27-28: "…conservatively estimated by multiplying by two for those of the O_3 line considering …" → "… conservatively estimated as being twice the size of those for the O_3 line, considering …"

P 8902 L 26: "…for obtaining the isotopic ratio without any vertical interpolation …" → "…in order to obtain the isotopic ratio without recourse to any vertical interpolation …"

P 8903 L12-13: "This is because of the different treatment of the tangent height in the VMR retrieval." → "This improvement derives from the retrievals of O_3 and ^{18}OOO using the same tangent height."

SC 2-16:

—- Page 8904

Line 1: "might" is weak here, surely that would be easy enough to check and verify for sure.

AR to SC 2-16:

Referee #1 also commented to this point. When this manuscript was submitted to AMTD, we considered that the smoothing error was dropped at 57 km. But now we recognize it as a part of oscillation through altitudes from 25 to 60 km. We investigated the cause of the 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 50km. Thus 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 $\mathbf{S_x}$."

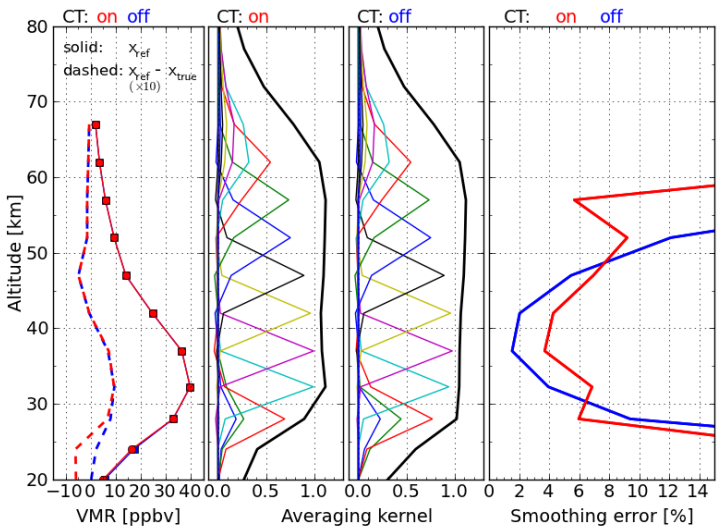

Fig. SC 2-16: Comparison of averaging kernel and smoothing error of b2 ^{18}OOO VMR with and without cross terms (CT) in $\mathbf{S_x}$. "CT: on" is results with CT (same as TOROROS). "CT: off" is results without CT.

SC 2-17:

—- Page 8904

Line 24: "We encourage determining" is clumsily worded. How about "We recommend that a laboratory study be undertaken to determine"

Line 28: "overcame" → "overcome"

AR to SC 2-17:

P 8904 L 24-27: "We encourage determining γ_{air} of ^{18}OOO transitions at an accuracy of at least the same order of that of the O_3 transition (3%), although both laboratory experiments and theoretical predictions have large difficulties that must be overcame." → "We recommend that a laboratory study be undertaken to determine γ_{air} of ^{18}OOO transitions at an accuracy of at least the same order of that of the O_3 transition (3%), although both laboratory experiments and theoretical predictions have large difficulties that must be overcome."

SC 2-18:

—- Page 8905

Line 16: What do you mean by "comparable" here? Was it that roughly the same fraction of points were rejected?

AR to SC 2-18:

Yes, we intended to mean that rates of data rejection by χ^2 thresholds for TOROROS and V215 are roughly the same. We changed the description of data selection by χ^2 not by absolute threshold values such as 2.5 and 1.0 but by a relative rate of numbers of the selected data.

P8905 L10-19: "In this study ... were also removed." → " In this study we selected data, that is regarded as "good quality" data, by χ^2 and measurement response ***m***. About 20% data with larger χ^2 values were removed, and only the retrieved data at altitude levels that satisfy $0.9 < \boldsymbol{m} < 1.2$ were used. Data from L1b data that included any visual field disturbances were also removed."

SC 2-19:

—- Page 8905

Line 18: This kind of filtering can be dangerous (TOMS ozone hole etc.) Could you at least say how many points were rejected by this filter? Did they still have reasonable chi-squares?

AR to SC 2-19:

We compared numbers of data of δ^{18}OOO from b1 O_3 and c1 ^{18}OOO selected with the VMR threshold and those without the threshold. Data selection by χ^2 and **m** was performed. In the condition: Feb. - Mar. (2010), 20°N-40°N, daytime (SZA < 80°), 28-57 km (7 levels), numbers with the VMR threshold are 1145-1377, as mentioned in page 8905 lines 20-21, and are identical to those in the case without VMR threshold. In fact, this VMR threshold is very conservative one and only less than 0.1 % data are removed by this VMR threshold in the all SMILES observations. Therefore, we deleted the descriptions about the VMR threshold.

SC 2-20:

—- Page 8906

Line 17: At 28km - what about lower altitudes, or are there no data produced lower down?

Line 18: At 57km - what about higher altitudes, or are there no data produced above that altitude?

AR to 2-20:

The lowest altitude level in TOROROS is 20 km, and there are also retrieval data at altitudes above 57 km. But data at these altitude levels are filtered out by the measurement response threshold.

SC 2-21:

—- Page 8906

Line 18: suggest "a significant" before "temperature dependence"?

Line 24-25: The sentence "We also discuss" is clumsily worded

—- Page 8911

Line 16: suggest "trends" → "behavior" as trends is typically thought of as being time related.

AR to SC 2-21:

P 8907 L 18: "… the temperature dependence." → "… a significant temperature dependence."

P 8908 L 24-25: "We also discussed …" is removed.

P 8911 L 16: "They showed similar trends as those in the daytime." → "They showed similar behaviors as those in the daytime."

SC 2-22:

—- Figure 1

Caption: For last sentence suggest: "Green, red and blue shading represent the b1, b2 and c1 frequency windows, respectively."

AR to SC 2-22:

P 8923 Fig. 1 Caption: "Shaded area represents the frequency region of the spectral window b1, b2 and c1 by green, red and blue color, respectively (see Table 1)." → "Green, red and blue shading represent the b1, b2 and c1 frequency windows, respectively (see Table 1)."

SC 2-23:

—- Figure 2

Not sure that having symbols on the lines really adds anything. Consider removing them to make the plots less cluttered. Also, I suspect the fonts are a bit small.

—- Figure 3

Again, not sure the symbols add anything.

AR to SC 2-23:

We removed symbols. We considered that the font is readable when they are set in the AMT layout.

SC 2-24:

—- Figure 4

The last sentence in the caption is unclear (at least to me). Do you mean divergence rather than dispersion? Also, does "which is the same" really mean "as for"?

AR to SC 2-24:

P 8926 Fig. 4 Caption L 4-8: "In column (a), the left and right panels display the median values and their differences, respectively. Column (b) shows the same as column (a) but for ^{18}OOO VMRs in Bands B and C. The dotted line represents data that the deviation is large or the number of profiles is small."

SC 2-25:

—- Figure 5

The overlapping shadings are a little hard to distinguish, though not impossible. Might showing error bars be clearer? It would probably be more cluttered though, so what you have here may be OK.

AR to SC 2-25:

We have tested the plot with error bars, but there is no clear improvement in the visibility (at least our eyes). We decided to keep the style with shading. Note that Fig. 5 was updated following Referee #1's comment (MC 1-9) to clarify the consistency among TOROROS δ^{18}OOOs.

SC 2-26:

—- Figure 7

Suggest showing a larger y-range, to at least take in the error bars for the averages, if not every point. Also, consider using darker colors for the points, they’re too light to see clearly on my

screen.

AR to SC 2-26:

We increased the range of y-axis, and the yellow and light blue colors are replaced to darker ones.

SC 2-27:

—- Figure A1

Again, symbols don't add much, fonts too small.

—- Figure A2

Again, consider dropping symbols

—- Figure A4

Same comment on shading as for Figure 5.

—- Figure A5

Again, font size and need for symbols.

AR to SC 2-27:

We also updated these figures in a similar way to Figs. 2-5.

[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".