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Atmospheric Measurement Techniques Discussions

Interactive comment on "Global stratospheric measurements of the isotopologues of methane from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer" *by* E. M. Buzan et al.

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We thank the referee for their constructive comments and provide our responses below.

GENERAL COMMENTS

However, what is missing are some key numbers, indicating retrieval uncertainty, and how it varies in space and time, what does the error budget look like, random versus systematic error components, etc.

We believe that problems with the spectroscopic constants are dominating the errors, and a detailed error analysis would reflect this rather than show sources of error from ACE. The most improvement in ACE retrievals would come from better constants derived from laboratory measurements. Therefore, the ACE data presented in the paper can be considered a "first look" to show the potential of ACE isotopologue measurements, such as its nearly global coverage, as well as a call for new measurements of methane's isotopologues. A few comments on sources of error in the retrieval process have been added to the text.

. . .how important is the a priori information that was used in the retrieval? To which extent might the latitude-height distributions reflect the prior rather than the measured spectra? When comparing the retrievals with the model, shouldn't the averaging kernels of the retrievals have been applied to the model?

A priori information is used as a first guess for pressure and temperature retrievals. We use a least squares fitting rather than optimal estimation, so we do not have averaging kernels for the retrievals. Ultimately, the retrieved profiles are fully derived from the measured spectra and no prior information is present in the VMR profiles. This also means no averaging kernels are available to apply to the model results. The text in the Methods has been changed to explicitly state that optimal estimation has not been used.

SPECIFIC COMMENTS

Page 11178, line 10: 'pockets. . .spring months' Which 'pockets' are meant here exactly? In Figure 3 I see such pockets during 'MAM' and 'SON' at 40km elevation, but in the autumn hemisphere, rather than the spring hemisphere. Please clarify.

Spring is the incorrect season here; the text has been updated to say the summer and fall months.

Figure 4, bottom left panel: Is the dark box at 80-90S and 27km dark blue (=dD de-

pleted) or out of the color range (=dD highly enriched). If it is the latter, please fix the contour colors.

The color is correct. The very low value of δD is an artifact due to the fact that measurements taken above 80 degrees in both hemispheres are few in number and low in quality due to the satellite's non-polar orbit. The text on page 11178, line 11 has been amended to point out this artifact.

Page 11178, line 14: 'Step function at 12 km' Actually horizontal lines are visible also at higher altitudes in the tropics.

These lines are also present in δ13C and are discussed in the paragraph below. Text pointing out this shared artifact has been added.

Figure 6, bottom plots: What is the vertical line at 70S above 30 km altitude?

The WACCM data in Figures 6-8 are sampled to match the ACE data shown in Figures 3-5. In other words "profile" of WACCM data is taken at each time and location as measured by ACE and compiled into figures in the same manner as ACE. This introduces the same bias in measurement location that ACE has due to its orbit. In particular, the line below 70°S in the two plots is due to the low number of measurements over the poles. These measurements were also all in the month of August and September, whereas other locations have data from several months.

Page 11179, line 23: 'These seasonal trends. . ..' It is worth mentioning here that for dD the model is more symmetric between the hemispheres than the measurements. The July – November enrichment is see only in the Southern Hemisphere, which is not in the model.

The model shows that the polar enrichment in δD is somewhat symmetric, but the southern hemisphere is stronger and reaches lower into the stratosphere. This enrichment is detectable by ACE in the southern hemisphere, but it does not spread low enough in the northern hemisphere to be picked up by ACE. This has been noted in

the text.

Figure 9, Caption: I suppose you mean 10 ppm-1?

This text has been corrected.

Page 11180, line 12: 'd13C data show more variance' It is important to distinguish variance in signal from variance from measurement uncertainty. The first should be good for a Keeling curve, the latter bad. Here presumably measurement uncertainty is meant.

The text has been changed based on this suggestion.

Page 11181, line 21: 'If WACCM is assumed. . .' If this condition is indeed satisfied, then we wouldn't need measurements anymore. However, it is sufficient to assume that tropospheric values are accurate. This is easily justified, because gradients in the troposphere are relatively small and measurements are available to confirm whether the model is indeed accurate or not. It would be worth explaining this more clearly.

"Assumed" was a poor choice of words here. We did compare WACCM's output in the troposphere to in-situ measurements for validation. For δ13C, WACCM predicts a tropospheric value of -47‰ which agrees with surface measurements (e.g., (Conny and Currie, 1996)). Tropospheric δD measurements have a larger range, between -100‰ and -75‰ (Rice et al., 2003; Röckmann et al., 2011; Umezawa et al., 2012). WACCM lies on the high end of this, between -81‰ and -78‰ with more a negative δD in the northern hemisphere. These citations have been added to the text as justification for using WACCM as a calibration standard.

Page 11185, line 2: 'Adjustment of spectroscopic parameters. . .' No material has been presented in support of this conclusion. Please provide more details about the spectroscopic adjustments that were made and the impact on the retrieval, if this finding is considered important enough to be kept (which seems to be the case).

The adjustments mentioned here refer to the improved research product referred to on

page 11175, line 25. The preliminary results from before this adjustment are not shown in the paper. The text here has been changed to refer to all the improvements in this new research product.

TECHNICAL CORRECTIONS

For figures showing delta values (4,5,7,10-14) a per mil sign is needed as unit. The figures have been updated.

REFERENCES

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