

COMMENT OF THE REFEREE:

This is a well written paper on an interesting and topical subject. The authors have very clearly defined their objectives and demonstrated the quality of their observations and analysis. Their careful attention to error analysis enhances their arguments. While I believe that most readers will not follow all the matrix algebra, the comprehensive treatment of the data and attention to detail present a convincing argument. Their techniques should find wider use in the remote sensing of atmospheric isotopologues. Their results should prove to be a valuable test for atmospheric models that include isotopologues and for satellite validation. Also their detailed and careful analysis of the ground-based FTIR data adds considerable value to those NDACC observations (already shown to be of high quality by their presentation of the CO₂ retrievals). I offer a few minor observations that may make the presentation clearer.

REPLY:

Many thanks for your encouraging remarks!

COMMENT OF THE REFEREE:

Page 3, Line 9 I do not believe that other work or the evidence here (Figure 3 A'') really support a vertical resolution as good as 2 km.

REPLY:

We offer two products (see page 5371, line 5-9). The first product offers the best possible estimation of the Humidity state (the second product the best possible consistency between Humidity and δD). We are sorry if the Referee got confused between the two different products.

The upper left graph of Figure 3A' shows the kernels if we aim on a best possible estimation of the Humidity state (it is not Figure 3A'' as commented by the Referee). The Figure shows the row kernels and documents that the values obtained at 2.4km represent real atmospheric Humidity between 2.4 and 3.4km. Atmospheric variability at altitudes above 3.4km hardly affects the value retrieved at 2.4km. In our opinion it is therefore justified talking about a vertical resolution of 2km (which is often defined as the FWHM – Full Width at Half Maximum – of the kernel). Please consider that the vertical resolution is that good only close to the observer. In the upper troposphere it is about 8km.

We will revise the text and try to avoid any confusion between Figures 3A, A', and A''.

COMMENT OF THE REFEREE:

P3,L15 "interference from humidity" is a bit odd here since after all humidity is what is being measured. The retrieval may have a dependence on humidity but that is not interference.

REPLY:

Ok, we will replace the term "interference from humidity" by "artifact cross-dependency/sensitivity of retrieved δD on atmospheric Humidity".

COMMENT OF THE REFEREE:

P4,L2 cycle is comprised of the

REPLY:

Ok!

COMMENT OF THE REFEREE:

P9,L13 Line 7 is not an equation. An equation contains an equal sign.

REPLY:

Ok, we will replace Eq. (2) by cost function (2) throughout the paper.

COMMENT OF THE REFEREE:

P10,L15 Are tropospheric water vapour concentrations logarithmic in the horizontal dimension or do they just vary so dramatically in the vertical?

REPLY:

The pdf of water vapour mixing ratios at a certain altitude is rather log-normal, meaning that there is a large variability. For instance At 2.4 km (at Izaña) we observe values that range between 300 and 30000 ppmv.

COMMENT OF THE REFEREE:

P12,L9 What is the justification for the first step in equation 8, likewise for equation 9?

REPLY:

$\partial \ln[x] = \partial x / x \rightarrow \Delta \ln[x] \approx \Delta x / x$

COMMENT OF THE REFEREE:

Figure 2: the radiance scale has a unit. Were the radiances really calibrated in these FTS observations?

REPLY:

Yes, the Izaña FTIR experiment regularly records radiances of a known laboratory blackbody radiation source, which allows for absolute calibration of the measured solar radiances (modest precision).

COMMENT OF THE REFEREE:

P15,L14 and Figure 3. Figure 3 does not make the discussion clearer. I believe that few readers will know what "row kernels of the water vapour state" means. The plots in Figure 3 show four colored lines labeled with altitudes and a number of unlabeled grey lines; more description is required for this figure to be useful.

REPLY:

The kernels presented in Figure 3 describe what we actually measure (what altitude is represented in the FTIR Humidity and δD product). It makes a difference if the Humidity or δD product represents the lower or the upper troposphere. Honestly, we think that remote sensing products should always be provided with their kernels otherwise the product might be misinterpreted. Figure 3 is of fundamental importance and we will make this clearer in the revised paper.

COMMENT OF THE REFEREE:

P16,L19 In Figure 4 the H₂O smoothing error seems to range from 6 to 90% through the troposphere. How can the smoothing error for the column, the sum of tropospheric layers, be only 0.1%?

REPLY:

Figure 4, like Figures 5, 6, 8, and 9, only depicts the square root values of the diagonal of an error covariance matrix (see, e.g., page 5372, line 15). However, there is a strong correlation between the errors at different altitudes, e.g., a positive error at 3km occurs together with a negative error at 7km. Consequently, the total column amount error cannot be assessed from the diagonal elements of the error covariance matrices as shown in the Figures. Therefore, we had to look on the outer diagonal elements, e.g., calculate eigenvectors and eigenvalues and depict the so-called error patterns. In the revised manuscript we will explicitly clarify this.

COMMENT OF THE REFEREE:

P16,L17 Is the natural variability of δD less than that of H₂O because of the strong correlation between [H₂O] and [HDO] ?

REPLY:

Yes! Please see also page 5362, line 5-12.

COMMENT OF THE REFEREE:

P17,L13 I tend to regard the word "assume" to mean that there was no real justification. I think that your case for adopting some of the error estimates is much stronger than that. For some assume may be the correct word.

REPLY:

Ok, we will revise this paragraph accordingly. For instance, instead of "based on the white noise that we observe in the spectra we assume a measurement noise of 0.4% (defined as noise to signal ratio)", we will simply write "we typically observe a white noise (noise to signal ratio) of 0.4% in the FTIR spectra".

COMMENT OF THE REFEREE:

P18,L26 If the random error is about 5% throughout the troposphere, how is the total precision better than 1%?

REPLY:

You have to consider that the errors at different altitudes are correlated (see also our reply above that deals with the error covariance matrix).

COMMENT OF THE REFEREE:

P29,L5 Have other effects, that might be specific to only the high altitude sites, been eliminated?

REPLY:

What do you mean by other effects?

Do you mean atmospheric processes that additionally remove HDO (e.g., rain recycling)? Then the answer is: no. Our paper is a technical paper and we cannot provide a scientifically comprehensive interpretation of the observed isotopologue ratios.

Do you mean measurement uncertainties that affect high altitude sites differently than low altitude sites? Our extensive error estimations indicate that the error patterns are very similar for low and high altitude sites. On the other hand there might be a secondary uncertainty behavior, which can cause that errors at humid and dry stations are somehow inconsistent: The spectroscopic line parameter uncertainty of weak lines might be significantly different from the parameter uncertainty of strong lines. At very dry sites the line parameter error is significantly determined by the uncertainties of the strong lines and at very humid sites by the uncertainties of the weak lines. This might lead to some inconsistencies in the errors. Therefore, within the MUSICA project we recently setup continuous in-situ water isotopologue observations (applying Picarro systems) at two FTIR sites: the dry site of Izaña and the humid site of Karlsruhe. In case there are secondary inconsistencies between dry and humid sites we will be able to constrain them in the future by comparing the FTIR data to the in-situ data.

In the conclusion Section we could give a very brief outlook on this comparison to in-situ data, which we plan for the near future.

COMMENT OF THE REFEREE:

P29,L20 What do the arrows and dashed lines in Figure 14 represent?

REPLY:

With the arrows we try to indicate the typical δD -H₂O distribution of the datasets (black for FTIR and red for IsoGSM). We will explain this in the Figure caption.

COMMENT OF THE REFEREE:

P29,L22 isotopologues

REPLY:

Ok!

COMMENT OF THE REFEREE:

P31,L24 I believe that the method presented here is complex, clear but complex, and not straightforward.

REPLY:

We called it straightforward since our proposal of transforming the atmospheric {H₂O, HDO}-state onto a {Humidity, δ D}-proxy state allows for a formalism that is analog to the “Rodgers formalism” (Rodgers, 2000), which is widely used by the remote sensing community. Maybe we should specify this a bit better. Instead of being “straightforward”, we can say that our method allows for characterizing the isotopologue ratio data in analogy to the widely used “Rodgers formalism”.