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Interactive comment on “Trend in ice moistening the stratosphere – constraints from isotope data of water and methane” by J. Notholt et al.

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We thank all three reviewers for their positive and constructive comments. Below we have addressed all major and minor comments from the three reviewers. For clearness, all comments are enumerated.

Reviewer 1: General comments: The authors present measurements of isotopic ratios of H₂O and CH₄ retrieved from balloon borne FTIR observations. These results are used to determine H₂O, HDO, and Delta D at the entry point. Finally a trend of particulate water transported through the tropopause is derived from these observations. The topic is very important to understand trends of stratospheric water vapor and hence to investigate climate change. Balloon borne infrared observations are one of very few techniques which allows to measure isotopic ratios with good vertical resolution and

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precision. However, the number of flights is limited. The paper is well written. The subject is fully appropriate for publication in ACP. I would recommend publishing it after minor revisions.

Specific comments:

(1) The number of flights performed at 35 and 65 North should be given in chapter 2. In principle it can be taken from the Figures. However, a statement about the number of samples might be helpful for the reader.

Response: A detailed list describing all flights is given in the supplement. In the main text we have included a short statement about the number of flights in chapter 2.

(2) The retrieval of the isotopic ratios is described very briefly. Do you use onion peeling or optimal estimation? In case of optimal estimation an interspecies constraint may even improve the precision of the delta D profiles.

Response: We use optimal estimation in the sense that vmr profiles are retrieved simultaneously for all levels. We did not apply an inter-species constraint of HDO to H₂O. An inter-species constraint is a good idea when doing a ground-based profile retrieval and the problem is under-determined. In this case, a priori information is essential to make the problem well-posed. But in the balloon limb-viewing geometry, the retrieval problem is fully determined and so an a priori HDO/H₂O constraint is less useful. This is now shortly explained in the text in chapter 2, second paragraph.

Technical corrections:

(3) - To my memory SMOW is about 3 * 10-4?

Response: The SMOW ratio of [D] to [H] is about 1.55e-4, and consequently that of [HDO] to [H₂O] is twice that ratio, the value the reviewer mentions. It does not matter whether SMOW is expressed as one or the other (as long as all calculations are carried out consistently!), and we have chosen to express it as the ratio of deuterium to hydrogen atoms.

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(4) - Brackets are missing for the term $([H2tr]-[H2])$ in eq. 4.

ACPD

Response: The equation has been corrected.

(5) 'as described in Sect. 5.1': Do you mean Section 4.1 since there is no section 5.1.?

Response: The reviewer is correct, it is section 4.1, has been corrected.

Reviewer 2:

This paper addresses the important question about the trend of water vapour in the stratosphere and gives important input to the problem. This paper is well written, however, I am not totally happy about the outline in the first sections. More about that in the comments below. The paper does address relevant scientific questions within the scope of ACP. I recommend the paper to be published after the following comments have been addressed.

Comments.

(6) In the introduction and in section two the delta-notation is used and figure one shows profiles of delta-D values, but the delta notation is not explained until section three. The authors might consider putting section three before section two. In the introduction a reference to the explaining section should be included.

Response: The delta notation is now given in chapter 1 and in chapter 3 we discuss the specifics of the stratospheric fractionation.

(7) Section 2: What retrieval method is used?

Response: We use a line-by-line algorithm for the forward model. The retrieval is performed using the optimal estimation method in the sense that vmr profiles are retrieved simultaneously for all levels. This is now explained in more detail the text.

(8) In section two the paragraph starting at line 4, page 16977, and the paragraph starting at line 13, should switch places. First discuss the measured spectra, then the

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retrievals.

Response: We have rearranged the order and included a statement on the retrieval method.

(9) Line 5, page 16979: ...neither methane NOR molecular hydrogen ...

Response: Has been corrected.

(10) Line 7, page 16983: dependant should be dependent

Response: Has been corrected.

(11) Line 25, page 16983: Section 5.1?

Response: Has been corrected, it should be section 4.1.

(12) Line 4, page 16984: 'The closer the isotopic composition of this additional ice is to that of the average total water entering the stratosphere' I don't really understand this sentence. Why would it be, and how sensitive to δD of the entering ice is the method?

Response: We want to state that in the (unrealistic) case where the isotopic composition of the vapour and ice would be identical, it would not be possible to measure an effect from ice evaporation on δD . We have added this explanation in the text.

Reviewer 3

General comments

The authors use an interesting set of balloon MkIV FTIR solar occultation data to derive trends in isotopic ratio values (delta D values) in water when entering the stratosphere, and from these trends in the transport of particulate water through the tropopause, for the period 1991-2007. The objective is to verify whether trends in the partitioning between water vapour and ice entering the stratosphere can explain (part of) the stratospheric water trends. This is an interesting paper, in which the balloon data set is exploited in an original way. The paper is well written in general but a little careless

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as to figures and details (missing elements in the captions / legends, etc; see specific comments below). Also the reasonings are sometimes hard to follow since they are given only very briefly.

Specific comments

(13) - pg 16976, line 5: the delta notation should be defined here

Response: Done, see also response (6).

(14) - pg 16976, line 21: delete 'at an altitude'.

Response: Has been corrected.

(15) - pg 16977 line 20-21: have the micro-windows been fitted simultaneously or not? it is not clear since the authors say 'individual results have been averaged'. Please clarify.

Response: In each spectrum all suitable microwindows are analysed individually. Since these microwindows belong to one measurement, the same atmospheric air path, they are averaged to give one vmr-value. Subsequently, the vmr-data for different altitudes (different spectra) are averaged. The averaging is performed using error propagation laws, as described in the supplement. This is now described in the text.

(16) - Pg. 16978 eqs (3) and (4): the superscript 'tr' has not been defined. Then the question rises what the VMRs without superscript stand for exactly.

Response: 'tr' denotes tropospheric. Has been corrected.

(17) - Pg. 16979 line 11: add 'in' before '(Rahn et al., 2003)'.

Response: Has been corrected.

(18) - Pg. 16981, line 4: the authors argue that the lack of detection in the ATMOS profiles may be due to their different vertical resolutions and accuracies: how do these characteristics compare between ATMOS and the presently used balloon data? On the

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other hand, they mention clear evidence in MIPAS data which 'I suspect' have about the same vertical resolution as ATMOS data. What about the characteristics of MIPAS versus ATMOS as to vertical resolution and accuracy? Are these parameters really the reason for the differences in detection?

Response: The signal that we detect is relatively weak (order 50% and requires an excellent signal-to-noise ratio of the measurement, which the balloon data have. As for MIPAS, the SNR of an individual profile is much lower, but there are many profiles available (between 100 and 600 each month in the inner tropics). First results of the MIPAS data, as presented at the EGU 2009 in Vienna, show that the results from MIPAS and our MkIV data are consistent, supporting the hypothesis that the ATMOS measurements were not able to detect the signal because of insufficient SNR and too few measured profiles.

(19) - Section 4.1: I have a problem with the fact that this Section considers the results of Fig. 2 as 'short-term variations'. The Table in the supplement clearly shows that the data set spans several seasons but spread over different years. The authors used only 23 balloon profiles covering 17 years, so on average they do not even have 2 profiles per year. How then can you disentangle short- from long-term variations ? So don't we see a mixture of short- and long-term variations in Fig. 2?

Response: The text is, admittedly, very dense on these aspects. The argument we make relies on the fact that from the tropical tropopause up to about 450K there is relatively fast quasi-isentropic meridional transport associated with large-scale wave (breaking). Consequently, variations in the entry conditions on the timescale of weeks to months are still observable at our observation altitude 380-425K. In contrast, in the upper layer (500-800K) the broad age spectrum (with mean age of order years, and a similar width of the distribution), measurements can only resolve variability at entry on the timescales of years (i.e. one can observe interannual variability at entry in the upper layer, whereas in the lower layer one can observe seasonal and even sub-seasonal variability at entry). This is the point we make here, not that we have a highly

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resolved timeseries at the position of observation. We modified the text to make this more clear.

(20) - Pg. 16981, line 13: why would the contribution from isotopically heavy ice increase with increasing H₂O ?

Response: Due to the isotopic difference between ice and vapour, it is possible to produce the observed slope if the amount of evaporated ice varies just a little. Relatively small seasonal variations in the deepest convection – e.g. height and/or how much detrained ice evaporates at the highest levels – could easily produce the observed signal. In fact, one would expect from the non-linearity of Clausius-Clapeyron more evaporation during the warmer period (when there is also more vapour) for a given contrast in frostpoint temperature between the detraining convective cell and the ambient air masses. We modified the text to make this more clear.

(21) - Pg. 16981, line 23: should read 'partitioning between vapour and ice' instead of 'partitioning between water and ice'.

Response: Has been corrected.

(22) - Pg. 16981, line 25: I guess that 'delta D = -300' should be 'delta D for ice= -300'? The units are missing.

Response: Has been corrected, units are %.

(23) - Pg. 16982 lines 3 and following: I don't understand how the conclusions in the summary follow from the paragraph just above it. The authors should be more explicit in their explanations. In fact, it is not clear how the last paragraph on pg. 16981 contributes to the findings in the summary at the start of pg. 16982.

Response: We have reformulated the last paragraph on page 16981. Now the connection to the summary on page 16982 is more clear.

(24) - Pg. 16982, line 22: the delta value trend of 11.1 plus/min 12.3 per mille per

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decade is not statistically significant, and neither is the trend calculated in Fig. 3 (black line) so can we conclude anything?

Response: We agree that the trend is statistically not significant, as can be seen from the uncertainties. However, we would like to point out that our result is only, to give an upper limit for the ice contribution. The upper limit depends on the trend and its uncertainty, but not on the question whether the trend is significantly significant or not. We now mention this at the beginning of section 5.

(25) - Pg. 16984, line 21: Should read 'The changes in the calculated amount of particulate water are within the uncertainty'.

Response: Has been corrected.

(26) - Fig. 2: vertical axis of middle plot not fully readable; legend not explained in the caption and not fully comprehensive.

Response: We have changed the size of the legend on the right side to make it more readable, and explain the Figure in more detail in the caption.

(27) - Fig. 3: I don't see the black symbols in plot (c). Open and closed circles in plots (a) and (b) are not identified. The first sentence in the caption is not complete ([HDOentry] is missing).

Response: We used indeed brown symbols, and not black ones. This has been corrected, and the symbols are now explained in the Figure caption.

(28) – 'Vapor' and 'vapour' are both appearing in the manuscript: English or US spelling should not be mixed.

Response: We now use English throughout the manuscript.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 9, 16973, 2009.

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