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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Temporal evolution of stable water isotopologues in cloud droplets during HCCT-2010" *by* J. K. Spiegel et al.

J. K. Spiegel et al.

johanna.spiegel@usys.ethz.ch

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Our replies are given in blue color between the reviewer statements. The figures we refer to in the replies are plotted in the supplement. In the revised version of the paper any changes are indicated by the use of blue color.

Final response to the comments from A.L. Rhodes

General Comments:
The authors present a unique study on the temporal evolution of the stable isotopic

composition of cloud water during different cloud events in the mountains of Germany. By focusing on cloud water, the results provide information on the water cycle history of different air masses that supply moisture to the study site. The study is well designed, and the authors do a nice job with considering different possible interpretations of the data within the context of stable isotope theory and results of prior studies. This work should be published after revisions are made to insure that some interpretations are not over stated, and to improve the clarity of the some arguments and explanation of the box model.

1) Reply: We thank A.L. Rhodes for this positive feedback of our MS.

For example, conclusion #1 states that seasonality (or differences in temperature) is reflected in the stable isotope data, and d-excess is an indicator of air mass origin. The short sampling period (6 weeks) isn't really long enough to identify a robust seasonal signal.

2) Reply: We agree with A.L. Rhodes that a 6 weeks data set is too short to identify seasonal changes and that especially the conclusion phrase was stated too explicitly. However, δ values were higher at the beginning of the campaign than at the end, which agrees to what has been observed in European precipitation and water vapor. We therefore changed in the revised MS:

p.15140 l.10: *While seasonality was reflected in decreasing δ values towards the colder season,*

to

Decreasing δ values in the course of the campaign agree with seasonal trends observed in rain in central Europe.

p. 15152 l.14 replace *This fits into the seasonal trend observed in rain (e.g. Dansgaard, 1964) and in water vapor (Jacob and Sonntag, 1991) characterized by decreasing δ values towards the colder season.*

by

Differences in monthly mean values are in the same order as observed in rain and in water vapor (Jacob and Sonntag, 1991, see Table 2). Both the δ values in rain as well

as the ones measured in vapor show a clear seasonal pattern with higher values in summer and lower in winter. Although the samples of this study were collected only during 6 weeks, the δ trend in the collected samples points towards such a seasonal signal, because monthly differences in δ values agree with data sets collected over the entire year.
p. 15152 l.21: we deleted *(seasonality can be neglected in this case)*
p.15155 l.7-8: we deleted *We showed that seasonality was reflected in the δ_c values (Sect. 3.1.1) and air mass origin in the d-excess (Sect. 3.1.2).*
p.15162 l.11-12: we replaced *Seasonality was reflected in δ^2_cH and δ^{18}_cO values while d-excess was an indicator of air mass origin.*
by:
δ^2_cH and δ^{18}_cO values agreed with decreasing values towards winter as known from δ values in precipitation. Changes in d-excess were most probably related to continental moisture recycling.
Only three events (#11-13) occur at a lower temperature, and only one of these (#12) has depleted isotope values relative to other data collected at warmer temperatures (as shown in Figure 2a).
3) Reply: The isotopic fractionation factor α is higher for lower temperatures. So given the same δ value in the water vapor, the δ value of the condensate would be higher (more enriched) at colder than at warmer temperature. So, the fact that δ values in precipitation in central Europe tend to be more depleted in the colder season can not be explained by the lower temperature at which condensation occurs. It is more likely caused by changing meteorological conditions at water vapor formation and due to a stronger rain out, i.e. changes in large scale water vapor transport.
The conclusion could be strengthened, however, if put in better context with the monthly data collected over several years at Heidelberg. Do the Heidelberg results show a strong seasonal isotopic signal? The text states that the results are consistent with the seasonality of the Heidelberg data, but this isn't really presented. Figures 4b and 4d hint at this seasonality, but again the time frame shown is very short.

4) Reply: The δ values measured in vapor in Heidelberg as well as the δ values in precipitation of the closest GNIP station show a clear seasonal trend with highest values in summer and lowest in winter. Moreover, the differences of monthly mean values were comparable. We therefore changed p.15152 ll.14:
This fits into the seasonal trend observed in rain (e.g. Dansgaard, 1964) and in water vapor (Jacob and Sonntag, 1991) characterized by decreasing δ values towards the colder season.
to
Differences in monthly mean values are of the same order as observed in rain and in water vapor (Jacob and Sonntag, 1991, see Table 2). Both the δ values in rain as well as the ones measured in vapor show a clear seasonal pattern with higher values in summer and lower ones in winter. Although the samples of this study were collected only during 6 weeks, the δ trend in the collected samples points towards such a seasonal signal, because monthly differences in δ values agree with data sets collected over the entire year..
Moreover, we add Table 2:

Table 2:Volume weighted monthly mean values for September and October including their difference for the collected cloud water samples at Schmücke. For comparison differences between volume weighted monthly mean values in precipitation for the closest GNIP station (Wasserkuppe Rhön 50°30′ N/ 9°57′ E, 921 m a.s.l.; 60.2 km west of the measurement station) as well as for the water vapor values collected in Heidelberg by (Jacob and Sonntag, 1991) are also given. See Fig. 1 for details on the sites.

Site	Month	Year	δ^{18}O	δ^{2}H
Schmücke	Sep.	2010	-5.8 ± 2.5‰	-32 ± 21‰
Schmücke	Oct.	2010	-7.9 ± 1.2‰	-48 ± 8‰
Schmücke	Sep.–Oct.	2010	2.1‰	17‰
Wasserkuppe	Sep.–Oct.	1978–2007	1.5‰	12‰
Heidelberg	Sep.–Oct.	1981–1988	2.4‰	18‰

Also, it does appear that the polar air masses (sourced from Greenland) generally have higher d-excess values, but one third of the Greenland events measured did not (see events #2, 3, 6 in Figure 3). If someone takes the interpretation literally and only looked at d-excess to identify source area without looking at air mass trajectory data, then he would have missed three "polar" events. The d-excess data presented also don't distinguish a North Atlantic source from a Mediterranean source. As explained in the manuscript, the d-excess is telling us something about the kinetic disequilibrium effects occurring during air mass transport, and the data presented show that this difference will be more likely with polar air masses because their different origin promotes kinetic disequilibrium. But differences in d-excess do not necessarily show differences in air mass origin, which is what conclusion #1 states.

5) Reply: We agree with A.L. Rhodes that our interpretation of the d-excess could be misunderstood. We therefore changed section 3.1.2 to:

D-excess of the cloud water and moisture recycling

The d-excess ($d = \delta^{2}_{C}\mathrm{H} - 8 \times \delta^{18}_{C}\mathrm{O}$) of the Schmücke cloud samples was rather high (10 to 20 ‰, Fig. 4, mean value 14 ‰) as compared to European air moisture d-excess of 7 to 11 ‰ (Gat et al., 2003) and stayed rather constant during most of the cloud events, except for events 1 and 10, which are discussed in more detail in Sect. 3.2.3. D-excess tended to be higher in cloud events that developed directly after rainfall (mean value: 17 ‰, blue dashed line in Fig. 4) than in cloud events that formed after a cloud free period without rainfall (mean value: 13 ‰, red dashed line in Fig. 4). The elevated d-excess could be an indicator of moisture recycling whereas a lower d-excess may represents an early stage condensation. However, as most cloud events

were sampled during nighttime when evaporation rates are closed to zero due to the lack of net radiative energy, moisture recycling due to evaporation of previously fallen precipitation was unlikely to happen directly at the site.

At this time of the year, precipitation at the site is commonly linked to large-scale precipitation occurring during frontal passages. This implies that precipitation at Schmücke is most likely occurring simultaneously with (or slightly delayed after) precipitation upwind of the measurement site. Consequently, the elevated d-excess of those cloud events sampled directly after rainfall was most probably caused by moisture recycling upwind of the Schmücke rather than by different climatic conditions at the initial moisture formation above the ocean. This view is in agreement with measurements of elevated d-excess presented by others both in rain and fog (Gat and Matsui, 1991; Rhodes et al., 2006; Froehlich et al., 2008; Cui et al., 2009).

The new Figure 4 is shown in the supplement (Figure 3 in the supplements)

In the Abstract we changed:

p.15140, l.10-12 to:

The d-excess was higher in clouds developing after recent precipitation revealing episodes of regional moisture recycling.

p.15140, l.25: added: *and regional recycling of moisture.*

In the introduction we changed:

p.15143, l.5: added *D-excess in precipitation also increases due to moisture recycling (Gat and Matsui, 1991, Henderson-Sellers et al., 2002, Rhodes et al., 2006, Froehlich et al., 2008).*

In the introduction we changed:

p.15143, ll.9-12: replaced

However, measuring d-excess in cloud droplets could indeed reveal insights into the initial isotopic signature of the moisture source region, as cloud droplets did not experience any additional kinetic fractionation as rain droplets do.

by

However, measuring d-excess in cloud droplets could indeed reveal insights into

the initial isotopic signature of the moisture source region, as cloud droplets are not affected by additional kinetic fractionation as rain droplets below the cloud.
p. 15152 l.5: we added: *to the water vapor transport and to moisture recycling* In the conclusions we changed p.15162 l.11-12 to:
Changes in d-excess were most probably related to continental moisture recycling.
Additionally, other researchers have discussed that elevated d-excess can indicate an important evapotranspiration flux to air masses (see Gat and Matsui, 1991; Henderson- Sellers et al., 2002; and Rhodes et al., 2006). Could evapotranspiration from the forests of western Europe account for the generally high d-excess values measured during HCCT-2010? I suspect not, given the time of year that the clouds were sampled.
6) Reply: We thank A.L. Rhodes for drawing our attention to these papers. In each of the cited studies d-excess measurements in the tropics and sub-tropics are presented. In addition to the geographic differences, climatic conditions (temperature, rainy seasons, radiation) as well as vegetation differ from the conditions found in central Europe at the time of sampling collection. This is reflected in the δ values as stated by Henderson-Sellers et al., 2002: *This moisture recycling within the Amazon basin leads to a seasonally averaged gradient of only 1.5‰ (1000 km)$^{-1}$ in δ^{18}O going inland on an east to west transect (Fig. 1) as compared with 2.0‰ (1000 km)$^{-1}$ in Europe and elsewhere (Rozanski et al. 1993)*. Because the sampled clouds had a certain horizontal extent and were mostly sampled during night time we assume that local (few kilometers) evapotranspiration could be neglected (see estimations of the evapotranspiration flux presented in the reply 4 to the K.Froehlich comments). An additional analysis of the contribution of moisture recycling on a regional scale was introduced in section 3.2.1 (see reply 5 above for details).
However, on p. 15159, the manuscript does suggest that a "biospheric signal" observed by other researchers may contribute to the observed diel changes in the isotopic composition of the cloud water at HCCT-2010. Please clarify on possible effects of evapotranspiration on the isotopic signals. P. 15160 states the authors'

assumption that total contribution of local evaporation and transpiration to air mass was small. However later in the same paragraph, the authors interpret that "significant uptakes [of water vapor] over land" could have occurred for events 1 and 10 (see p. 15160, lines 5-18 and point #2 in particular). The argument and discussion is contradictory.
7) Reply: Indeed this argument appears to be contradictory, as we did not write it clearly enough. On p.15160, with *local* we mean the measurement site itself including a radius of a few kilometers. However, *over land* stands for a much larger region, i.e. where the airmass passed over land instead of over the ocean.
The box model and its results are difficult to understand. The box model is designed to explain whether changes in the isotopic composition of cloud water during the event is due to differences in condensation or due to changes in the composition of the water vapor.
8) Reply: We are aware of that changed the following passages in order to improve clarity:
p.15149 ll.16-17: deleted *(for the phase change from liquid to vapor as a function of temperature)*
p.15149 ll.21-22: changed *Assuming the cloud droplets in immediate isotope equilibrium with the surrounding vapor (Spiegel et al., 2012)*
to
Assuming that the cloud droplets are in immediate isotopic equilibrium with the surrounding vapor (Spiegel et al., 2012)
p.15149 l.24: deleted *, applied for* ^{2}H *and* ^{18}O*, separately:*
p.15150 l.1: added *Equation (7) applies to* ^{2}H *and* ^{18}O *separately.*
p.15150 l.5: *:* replaced by *. This means that*
p.15150 l.13: deleted *such systems involving*
p.15150 l.17: changed *To evaluate the extent condensation controls the variation of measured δ values for the cloud samples*
to

To evaluate the extent to which condensation controls the variation of measured δ values for the cloud samples

p.15150 l.23: added behind values *at the actual temperature*

p.15150 l.23,24: replaced twice *as* by *because*

p.15150 l.25: introduced: *Details are given in the next sections.*

2.4.1 Model run A: from measured temperature T_{sm} to dew point temperature T_d

p.15150 l.27- p.15152 l.11: changed *In detail: for model run A, the box model was first initialized with the measurements. For this purpose, we calculated the mean values taken over the sampling time of each cloud sample, of the temperature T_{sm}, the liquid water content (*LWC*) and the dew point (T_d) of the Schmücke cloud. We calculated the dew point from the local mixing ratio w, which itself was deduced from the local temperature, air pressure and* LWC*. From these values, the initial box model variables were deduced (framed in red in Fig. 1): $\alpha_0 = \alpha(T_{sm})$ was calculated using the equations of Criss (1999, p.103). R_{v0} (which is the vapor isotope ratio inside the cloud in equilibrium with the condensate) was calculated from the measured δ_C values using Eqs. (6) and (1) (Fig. 1 item 1). X_{v0} was calculated based on the equilibrium water vapor pressure (vapor phase) and the* LWC *(condensed phase). Second, we repeatedly ran the model for every cloud sample from the measured temperature T_{sm} to the dew point T_d, changing X_v from X_{v0} to 1 and T from T_{sm} to T_d. By doing so, we calculated the δ_v value for each measurement point for the moment when the condensation started (Fig. 1 item 2). We refer to these values as $\delta_{v,dew}$.*

to

*First, the box model was first initialized with the measurements. To this end, for every sampling interval we calculated the mean values for the temperature T_{sm}, the liquid water content (*LWC*) and the dew point (T_d) of the Schmücke cloud from the measured data (see Sect. 2.3). Then, we calculated the dew point from the local mixing ratio w, which itself was determined from the local temperature, air pressure and* LWC*. In addition to T_{sm} at Schmücke, X_{v0} and δ_{v0} (corresponding to R_{v0}) are needed as initial values (framed in red in Fig. 2). X_{v0} was calculated based on the equilibrium water*

vapor pressure (vapor phase) and the LWC *(condensed phase). R_{v0} — which is the vapor isotope ratio inside the cloud in equilibrium with the condensate — was calculated from measured δ_C values using Eqs. (6) and (1) (Fig. 2 ①)). Herein $\alpha_0 = \alpha(T_{sm})$ was calculated using the equations of Criss (1999, p.103). Second, we repeatedly ran the model for every cloud sample starting with the corresponding T_{sm} down to the dew point T_d, thereby decreasing iteratively T from T_{sm} to T_d and increasing X_v from X_{v0} to 1. This model run is referred to as model run A. By doing so, we calculated the δ_v value for each measurement point for the instant when condensation started (Fig. 2 ②)). We refer to these values as $\delta_{v,dew}$.*

p.15151 l.12: introduced: *2.4.2 Model run B: from dew point temperature T_d to measured temperature T_{sm}*

p.15151 l.12-16: replaced

For run B, we used the model a second time, from the dew point (taken as initial condition) to the actual measurement temperature T_{sm} (and $X_v = 1$ to $X_v = X_{v0}$). For every cloud event we performed n (= number of samples per event) different model runs B, each starting from a different $\delta_{v,dew}$ as received from model run A (Fig. 1 item 3). This yielded n^2 calculated δ values for the cloud droplets to be compared to the measurements (Fig. 1 item 4)

by

For run B, we used the model a second time in the opposite direction from the dew point T_d to the actual measurement temperature T_{sm}. The initial parameters for model run B are T_d, X_v=1 and $\delta_{v,dew}$ as received from model run A. For every cloud event we performed n (= number of samples per event) different model runs B, each starting from a different $\delta_{v,dew}$ as obtained from model run A (Fig. 2 ③)). This yielded n^2 calculated δ values (=$\delta_{c,mod}$)..

p.15151 l. 15: added

2.4.3 Condensation criteria

In order to evaluate the effect of condensation on the δ_C values, the modeled $\delta_{c,mod}$ values were compared to measured δ_C (Fig. 2 ④)).

The figure caption for the illustrative example in Figure 1 doesn't sufficiently explain how the authors distinguish these differences. This needs to be spelled out better. For example, the last sentence of figure caption #1 doesn't explain why two points meet the condensation criterion, or why the first & second measurement transition is due to changes in water vapor.

9) Reply: The caption of Figure 1 (which is now Figure 2 in the revised MS and is shown in the supplement to these answers as Figure 2) was changed to: *Sketch of a cloud forming at Schmücke including the box model approach **(b)** consisting of the two model runs A and B. The initial values of each model run are framed in red. **(c)** An example of the modeling principle as described in Sect. 2.4 (variables are explained there as well): from the measured δ_C, δ_{v0} is derived (1) leading to $\delta_{v,dew}$ after model run A (2). Starting with each of the $\delta_{v,dew}$, model run B produces a series (three in total for the case shown here) of locally thermodynamically driven $\delta_{v,mod}$ (3), from which $\delta_{c,mod}$ were deduced and in a last step compared to the measured time series of δ_C (4). In this example, the transition between the last two measurement points meet the local condensation criterion, because both measured values δ_C are within the errors of modeled values $\delta_{c,mod}$ (cloud sample 2 is within the black error bar, which shows the uncertainty of the $\delta_{c,mod}$ based on $\delta_{v,dew}$ of cloud sample 3 and wise versa). In contrast, the error bar of $\delta_{c,mod}$ based on $\delta_{v,dew}$ of cloud sample 1 (green) does not overlap with cloud sample 2 (and the red line not with cloud sample 1). So, the transition from the first to the second measurement point is found to be caused by changes in the water vapor isotope composition feeding the cloud.*

Figure 4 summarizes the authors' interpretations of the box model analysis well. Conclusions # 3 & 4 both seem reasonable, and the temporal evolution for the frontal systems is very striking.

10) Reply: We appreciate this positive feedback.

Use of "HCCT-2010" in the title is not broadly descriptive. Consider including the location, geography or season of sampling in the title as a way to provide readers a picture of the key elements of the experimental campaign.

11) Reply: The title was changed to: *Temporal evolution of stable water isotopologues in cloud droplets in a hill cap cloud in central Europe (HCCT-2010)*

A figure with a location map will also be helpful.

12) Reply: Was added a map in the revised MS (see Figure 1 of the supplements). In the revised MS, we refer to the location map on p. 15144 l.23,24, p.15147 l.23, p.15156, l.10 and in the caption of Table 2.

Several passages in the results and discussion are wordy and expressed awkwardly. Aiming to write shorter, concise sentences will improve clarity. Colon punctuation (:) is misused throughout the manuscript, and its misuse obscures the meaning of sentences. Colons should not be used to connect two different ideas in one sentence. In many cases, a semicolon is more appropriate, or one sentence should be broken into separate sentences. Semicolons are used to separate two related, complete sentences. If the sentences are not closely related, separate them with a period.

13) Reply: Was changed in the revised MS.

The word "as" is misused; often, the word "because" is more appropriate. Use "as" when your topic relates to some element of time. Use "because" when something is caused by something else. See p. 15152, line 20; p. 15154, line 12; p. 15160, lines 2-3 for instances where "as" is misused. Other examples may exist.

14) Reply: Was changed in the revised MS.

Other specific comments toward improving writing clarity:

p. 15141, line 25: "fractionation" is misspelled.

15) Reply: Was changed in the revised MS.

p. 15145, line 8: What are "biogenic emissions"? Is this evapotranspiration, wood burning, or fossil fuel combustion?

16) Reply: The main focus of HCCT-2010 was an aerosol-cloud interaction study. Thus, biogenic emissions in this sense comprise aerosol emissions by plants and soils (e.g. bacteria, fungal spores, dust) as well as evapotranspiration.

p. 15147, lines 14-18: Awkwardly phrased text. Break out into separate sentences. The mathematical equation clarifies the text, but text should also be clear.

17) Reply: Was changed in the revised MS to
Then we retrieved the cumulative rain ($\mathrm{Rain_{cu}}$) per cloud sample. This is the total amount of rain that the air masses passing during the sampling interval had formed before reaching Schmücke. $\mathrm{Rain_{cu}}$ is calculated by accumulating $\mathrm{Rain_{tr}}$ of the back-trajectories that started during the sampling interval of each cloud sample.
p. 15150, line 4-6. Awkward sentence structure; misuse of colon.
18) Reply: Was changed in the revised MS to
For $A = 1$, the box model is considered to be closed. This means that the condensate formed completely remains in the box and is in thermodynamic and isotopic equilibrium with the surrounding water vapor.
p. 15151, lines 12 - 15. Awkward. Too much information in one sentence. Please work to clarify explanation of the entire box model section (Section 2.4). Other readers may want to reproduce your methods.
19) Reply: Was changed in the revised MS to
For run B, we used the model a second time in the reversed direction, starting at the dew point T_d and increasing to the actually measured temperature T_{sm}. The initial conditions for model run B are T_d, X_v=1 and $\delta_{\mathrm{v,dew}}$ as obtained from model run A.
For additional changes in the box model section see reply 8 above.
p. 15152, lines 25-27. Awkward expressions, including reference to Rank and Papesch (2005).
20) Reply: Was changed in the revised MS to
So we could only expect differences in isotopic composition if the traveling distance differed sufficiently for different air mass sources. This was for example the case in the study presented by Rank and Papesch (2005).
p. 15152, line 21: Change “Additional water uptake of polar air masses...” to “Additional water uptake by polar air masses...”
21) Reply: Was changed in the revised MS.
On p. 15154, line 14: The paper states that below-cloud evaporation will affect d-excess in rain âĂŤ by how much typically?

22) Reply: This depends on relative humidity and temperatures and can be calculated using the Craig-Gordon model. p.15153 we write: *Decreases of the d-excess caused by below-cloud evaporation have been shown to be in the range of 1 to 4 ‰ for stations in Austria at a similar altitude during September and October, analyzing a 20-yr precipitation data set (precipitation-weighted monthly averages; Froehlich et al., 2008).*
p. 15155, lines 13-15. Awkward sentence. Simplify sentences.
23) Reply: Was changed in the revised MS to
However, looking at the data on a diurnal scale (Fig. 3) revealed highest δ_C values at night and lowest during daytime for at least six of the cloud events (Sect. 3.2.1).
p. 15156, lines 23-25. Dangling phrases make this sentence difficult to follow.
24) Reply: Was changed in the revised MS to
Additional samples also met the local condensation criterion either for δ^{18}O or for δ^{2}H (red in Fig. 5a and c).
p. 15158, line 24-25. Fix structure “...do not allow to determine...”
25) Reply: Was changed to *Based on the DWD synoptic charts (Sect. 2.3) the exact time of the frontal passage can not be determined, because the time resolution of the synoptic charts used in the framework of the HCCT-2010 analysis is 6 h exceeding the duration of cloud event 7.*
p. 15159, lines 19 – 24 (last 2 sentences of paragraph). Please clarify term “free atmosphere” and elaborate on the atmospheric signal and biospheric signals mentioned in the last sentence.
26) Reply: Free atmosphere is the portion of the earth’s atmosphere, above the planetary boundary layer. We consider evapotranspiration (as mentioned in brackets) as a biospheric signal.
p. 15160, lines 2 – 5. Awkward sentence structure. Use of word “as” is confusing.
27) Reply: Was changed in the revised MS to
Fog droplets equilibrate quickly with the advected vapor (Spiegel et al., 2012) and moreover, we could show that condensation only had a minor impact on the temporal evolution of these two events. Consequently, the temporal pattern in δ_C values that we

measured in the cloud water was probably advected by the air mass.
Other awkward passages may exist. Edit carefully for clarity.
28) Reply: The MS was revised carefully for language and clarity. We changed the following passages additional to the ones mentioned in the other replies:
p.15140, l.3: deleted *the course of*
p.15140, l.9: added *other* behind *than*
p.15140, l.14: added *the* behind *in*
p.15140, l.15: replaced *measured* by *investigated*
p.15140, ll.20-22: replaced *Frontal passages led to the highest gradients both in* δ^2H *(≈6 ‰ per hour) and* δ^{18}O *(≈0.6 ‰ per hour) during two of the latter cloud events.*
by
Frontal passages during two of the latter cloud events led to the strongest temporal changes in both δ^2H *(≈6 ‰ per hour) and* δ^{18}O *(≈0.6 ‰ per hour).*
p.15141, ll.6-9: replaced *On the time scale of days to years, stable water isotopologues analysis improved our understanding of the hydrological cycle by measuring their spatial and temporal distribution in precipitation all over the globe (e.g. Rozanski et al., 1993, Gat 1996, 2000).*
by
On the time scale of days to years, measurements of stable water isotopologues in precipitation and analyzing spatial and temporal distribution all over the globe improved our understanding of the hydrological cycle (e.g. Rozanski et al., 1993, Gat 1996, 2000).
p.15141, l.21: replaced *of* by *for*
p.15142, l.13: changed *(Coplen,2011)* to *(IAEA,2009)*
p.15143, l.29: deleted *Additionally,*
p.15144, l.1: added *the* behind *to*
p.15152, l.10: added *More detailed information can be found in the raw data set (http://doi.pangaea.de/10.1594/PANGAEA.788629).*
p.15153, l.8: replaced *are* by *can be*

p.15156, l.1: replaced *This question* by *The possible explanation 1*
p.15156, l.2: added *assuming* before *constant*
p.15157, l.13: replaced *are a valid tool* by *can potentially*
p.15159, l.13: replaced *it* by *they*
p.15160, l.11: replaced *results* by *resulted*
p.15160, l.14: replaced *cause* by *caused*
p.15161, l.25: delete *Furthermore,*
p.15162, l.21: add *characteristics* behind *overflow*

References cited:
Gat, J.R. and E. Matsui (1991) Atmospheric water balance in the Amazon Basin: An isotopic evapotranspiration model, J. Geophys. Res., 96, 13179-13188.
Henderson-Sellers, A., K. McGuffie, and H. Zhang (2002) Stable isotopes as tools for global climate modle predictions of the impact of Amazonian deforestation, J. Clim., 15, 2664-2677.
Rhodes, A.L., A. J. Guswa, S. E. Newell (2006) Seasonal variation in the stable isotopic composition of precipitation in the tropical montane forests of Monteverde, Costa Rica.
29) Reply: We included these references into the revised version of the MS.

Final response to the comments from Referee #2

1) Reply: We thank Referee #2 for his or her comments, which helped us to improve our MS.
The authors present and interpret cloud isotope data collected during a mountain campaign in Germany. The paper is well structured and tries to squezze out as much as possible information the data potentially contain. Sometimes I assume an overinter-

pretation, basically ever, if general conclusions are drawn, simply because of the low number of cloud events and the short duration of the campaign.
2) Reply: The aim of the MS was basically to show the potential of isotope measurements in cloud water. We changed wordings and phrases when general conclusions were drawn in order to avoid over interpretation. Details are given in the answers to A.L. Rhodes (mainly in replies 2 to 5 and 28.)
Moreover, the data may be better embedded in other isotope data sets, e.g. the one from Heidelberg.
3) Reply: We embedded the data better into given data sets - for details see reply 2, 4 and 5 to A.L. Rhodes.
Although the train of thoughts is usually well traceable, due to the complexity of the topic, it is often not simple for the reader to fully understand each step in the interpretation. The situation becomes more complicated by the fact that the nomenclature is often not self-explanatory. For instance, in equation (2) the subscript c is used to "distinguish the collected cloud samples from the calculated vapor samples" (what c is meant?) . . . better write ". . . from the modelled vapour samples" and use c and m as subscripts.
4) Reply: As we use the model to calculate both δ values in the vapor and in the condensate, m can not be used as a subscript. We clarified the use of the subscripts throughout the MS, c always stands for condensed fraction (i.e. the measured δ values in the cloud droplets). If we refer to the modeled values we used c, mod as a subscript.
In equation (6) the subscript c is again introduced differently, with Rc marking the isotope ratio of the condensate. Thus, define well-understandable abbreviations/ subscripts.
5) Reply: We added on p. 15146 l.5: *We used the subscript c (=condensed phase) when referring to the collected cloud water and the subscript v to refer to δ values in the vapor phase (see Sect. 2.4 for details on the modeling procedure).*
Also, the denotation "model run A" and "model run B" is not suitable, as they simply describes two consecutive steps in the model, but not two different model versions (as

one expects if indexing with A and B).
6) Reply: By writing *model run A* and *model run B* instead of *model A* and *model B* we were trying to emphasis that it is one model used in two different model set ups. As we are referring to the model runs in the result and discussion section an abbreviation for the model runs is needed and we consider A and B to be the most simple ones. We also changed parts of the model description section (see reply 8 to A.L.Rhodes) as well as the Figure caption of Figure 1 (see reply 9 to A.L.Rhodes) in order to improve clarity.
Besides this, I have only some minor concerns.
Equation (1). Multiply it by 1000 per mil.
7) Reply: According to Coplen 2011, multiplying by 1000 per mil is no longer the correct writing. We therefore keep the present version of equation (1).
P. 15145, l. 28. What a delta unit? If per mil, then write it.
8) Reply: This is the phrasing which is suggested by Coplen 2011 and Brand and Coplen 2012. We therefore keep the sentence as it is.
Equation (3). Is the uncertainty in the collected volume zero?
9) Reply: There is surely an error in the collected volume as well, however we lack that information. And as we only use the volume in order to work with volume weighted values (as it is the common procedure for GNIP measurements), we think it is appropriate to calculate the error as presented.
P. 15147, l. 7. horizontal instead of spatial
10) Reply: Was changed in the revised version
P. 15147, l. 23. How you maintain such an "upwind sounding"? The sounding site is likely stationary and only during specific synoptic conditions "upwind", right?
11) Reply: In the context of HCCT-2010, we probed two different types of cloud events; the so called full cloud events (FCE) and the cloud only events (COE, in this work number 3, 4 and 5, which will be added in table 1 by a footnote). For the FCE events the following criteria needed to be fulfilled in order to start sampling:

- liquid water content LWC as measured by the Particulate Volume Monitor $>$ 0.1 g m^{-3}
- Wind direction at Schmücke 200° to 250°
- Wind speed 2 to 12 m s^{-1}
- Valley sites Goldlauter (50°38'25" North, 10°45'20" East, 605m a.s.l.) and Gehlberg (50°40'21" North, 10°47'32" East, 732m a.s.l.) free of fog (these two stations have not been mentioned in this MS as no additional information was used from these sites) - see Figure 4 in the supplement.
- No precipitation at any site
- Temperature $>$ 0°C

Soundings were launched in Meiningen 30 km South West of Schmücke. Sounding data were used for the full cloud events in order to calculate the Froude number. So under the condition of Full Cloud Events the soundings are always upwind. We will change: *Meteorological soundings were launched 30 km upwind of the site at 00:00 UTC and 12:00 UTC by the German Weather Service (DWD) (Heinold et al. 2005, Tilgner et al. 2012).*
to
Meteorological soundings were launched in Meiningen 30 km South West (for location see Fig. 1) of the site at 00:00 UTC and 12:00 UTC by the German Weather Service (DWD) (Heinold et al. 2005, Tilgner et al. 2013).
Section 2.4. Introduce two subsections (with numbering 2.4.1 and 2.4.2 or not) for model run A and B so that the reader can better follow the sense of the two runs.
12) Reply: Was implemented in the revised MS, for further changes in this section please see the reply 8 and 9 to comments of A.L. Rhodes.
P. 15151, l.18. add "simply, as other factors, e.g. . . ., which cannot accurately be

described by the model, will not have played a significant role" or something like that.
13) Reply: Was implemented in the revised MS
P. 15152. Indeed, six weeks are not appropriate to retrieve any "seasonal variation". If you come with this argument, you have to embed your data in other studies / observations and have to give typical seasonal trends in "per mil in 6 weeks" or so. Then you can write: "our numbers of . . . agrees with . . .". This badly constrained "seasonal argument" pops up too often in the manuscript.
14) Reply: this was implemented in the revised version of the MS, for details see reply 2 and 4 to to comments of A.L. Rhodes.
Additional references used in this reply:
Brand, W. A. and Coplen, T. (2012), Stable isotope deltas: tiny, yet robust signatures in nature, Isotopes in Environmental and Health Studies, pp. 1-17

Final response to the comments from K. Froehlich

1) Reply: We thank K. Froehlich for his comments, which helped us to improve our MS.
The authors gathered valuable data on the isotopic composition of droplets in fog/clouds, which will help better understand the formation of the isotopic composition of atmospheric water. The samples were collected at a station close to the summit of Schmücke (Thüringer forest) at an altitude of 937 m.a.s.l. The selected sampling period September/October 2010 is within the autumn season, in which in that region often fog develops, so called "Herbstnebel" (autumn fog). The authors consider this fog as orographic cap cloud.
2) Reply: We agree with K. Froehlich that in many regions fog forms in autumn due to radiative cooling. However, the cloud that forms at Schmücke does not form due to radiative cooling. In the context of HCCT-2010, we probed two different types of cloud events; the so called full cloud events (FCE) and the cloud only events (COE, in our MS these are number 3, 4 and 5). For the FCE events the following criteria needed to

be fulfilled in order to start sampling:

- liquid water content LWC as measured by the Particulate Volume Monitor $>$ 0.1 g m^{-3}
- Wind direction at Schmücke 200° to 250°
- Wind speed 2 to 12 m s^{-1}
- Valley sites Goldlauter (50°38'25" North, 10°45'20" East , 605m a.s.l.) and Gehlberg (50°40'21" North, 10°47'32" East, 732m a.s.l.) free of fog (these two stations have not been mentioned in this MS as no additional information was used from these sites) - see Figure 4 in the supplement.
- No precipitation at any site
- Temperature $>$ 0°C

These criteria were needed in order to get connected flow conditions between the three stations which is needed for a Lagrangian-typ cloud experiment (see Figure 4 in the supplement). These criteria have already been developed and used in an earlier Lagrangian-type cloud experiment (FEBUKO in autumn 2001 and 2002, see e.g. Herrmann et al., 2005). Due to the overflow during the cloud experiment the cloud either forms by adiabatic cooling or is at least influenced by the airmass flowing over the ridge and therefore commonly shows orographic characteristics, even if the cloud is not a locally formed isolated one on the top of Schmücke. E.g. the temporal changes of the adiabatic LWC as calculated from the cloud base height and the cloud base temperature typically show the same pattern. Therefore we think that it is appropriate to call the probed cloud an "orographic cloud" .
To define the source of the moisture forming the clouds, backward trajectories were calculated. This concept of backward trajectories is known to be useful in case of

clouds moving above the Planetary Boundary Layer. In case of fog or clouds forming and moving near the ground (including orographic cap clouds), the potential of such calculated back trajectories in identifying the moisture source needs to be discussed.
3) Reply:We would like to point out that the planetary boundary layer should not be considered as a closed box and that there is indeed a mixing between the free atmosphere and the planetary boundary layer. The water vapor at a certain location as e.g. the Schmücke should therefore always be considered as a mixture of both. A backward trajectory analysis takes both the free atmosphere and the potential ground based sources into account. We also think that by presenting these data, we were able to show the potential of this analysis: while for event 1 there were rather localized sources in France, the results from event 10 showed that the sources were more local. We do agree with K. Froehlich that the uncertainty of the moisture source diagnostics method should be given due consideration. That is why we write on p.15161, l.25-p.15162, l.1: *Furthermore, the strong mixing of different moisture sources increases the uncertainty in the identification method (consider the shaded area in Fig. 5b, column 3 representing the estimated moisture source condition), thus changes in the d-excess could not be related to changes in the moisture source relative humidity for event 10.*
In the revised version of the manuscript we added on p.15162, l.1:
Furthermore, due to the limited spatial resolution of the wind analysis data (7 km grid spacing) very localized recycling effects are likely underestimated by this method.
We also added p.15161, l.8: *However, the moisture sources contributing the most (darkest areas) are very closed to Schmücke, suggesting an important moisture contribution of regionally re-evaporated vapor.*
In this context, it should be noted that the sampling station Schmücke is located rather close to a relatively extended upland moor ("Schneekopfmoor" about 500 m north of Schmücke station and "Beerbergmoor" about 2000 m north-north-west from it). Therefore, it can be expected that a certain part of the air moisture (and thus also of the droplets) in the fog/clouds consists of moisture recycled by evaporation of soil

water from the ground near the sampling site.

4) Reply: In the context of HCCT-2010, there were two different cloud events; the so called full cloud events (FCE, see reply 2 for details) and the cloud only events (in this MS these are numbers 3, 4 and 5). The "overflow" criteria were needed in order to get connected flow conditions between the three stations which is needed for a Lagrangian-typ cloud experiment. Due to the wind direction criteria (wind from the south west, see Figure 4 in the supplement), it is highly unlikely that air moisture evaporating from the two fens which were north and north-north-west of the site would significantly contribute to the moisture at the measurement site. For the 5 cloud samples collected during the cloud only events, only the cloud sample collected during event 4 was affected by wind blowing from the 300° to 350°. So only for one out of 41 samples the wind direction may allow moisture to be transported from the fens towards the sampling station.

We did not mention these criteria explicitly as e.g. the sites Goldlauter and Gehlberg were not introduced in this MS as we do not present any data collected there. However, to be more precise about meteorological conditions during sampling we will add on p.15146 l.24:

Wind speed during sampling collection varied between 1 and 12 m s^{-1} for all presented cloud events. The prevailing wind direction at Schmücke was wind blowing from around 200° to 250°.

Also the orographic cap clouds that develop by adiabatic ascent to the summit of the forest, may entrain moisture evaporated from the forest ground. The effect of recycling of evaporated moisture from regions near the sampling station certainly depends on how fast the clouds move.

5) Reply: The evapotranspiration flux depends on net radiation, which is around 0.8 times the global radiation in a cloudy environment. 27 of the 41 cloud samples were collected at night where the evaporative flux can be neglected due to the lack of net radiation. Due to the mentioned overflow criteria presented in reply 2, there is a certain mesoscale wind which influences the uptake in the cloud. We estimate the possible

moisture uptake of the airmass sampled during the 14 cloud samples collected during day time based on the global radiation and the wind speed measurements:

The evapotranspiration flux E [mm h^{-1}] can be deduced from the latent heat flux Q_E [W m^{-2}] (see e.g. Stull, 1988):

$$E \approx 0.0016 \times Q_E \tag{1}$$

From the evaporation rate the moisture added per square meter and second c_{eva} can be calculated. The air volume V per ground surface which is renewed every second depends on the mixing height H (and the vertical extent of the cloud) the wind speed w which is measured at Schmücke. So the added mass per volume m_{eva} due to evaporation during a travel distance t can be written as:

$$m_{\text{eva}} = \frac{t \times c_{\text{eva}}}{w \times H} \tag{2}$$

Assuming saturated conditions (presense of a cloud) the moisture m_{back} in the "background" can be calculated using the Clausius Clapeyron and the ideal gas equation.

$$\frac{m_{\text{eva}}}{m_{\text{back}}} \tag{3}$$

gives the fraction of the background moisture which could be added over certain travel distance.

For an upper limit estimate, we chose a travel distance of 2000 m (which corresponds to the distance of the fen to the site mentioned by K. Froehlich), a mixing height of 200 m and the 0.8 $\times$ global radiation as a estimate for latent heat flux Q_E. Note that the real values of evaporative fluxes will be even lower, because the net radiation is portioned into latent, sensible and ground heat flux.

Based on this estimation the local evaporative contribution is below 1% for 36 of the 41 cloud samples (for 35 it is even below 0.5%). So, we agree with K. Froehlich that a minor effect of very local moisture recycling might exist for some of the cloud events.

However, due to the presented argumentation above, it is highly unlikely that very local moisture recycling is the reason for the observed d-excess in the events analyzed here. We therefore include in the revised version of Section 3.1.2 (see Reply 7 for the full text of the revised Section 3.1.2):
However, as most cloud events were sampled during nighttime when evaporation rates are closed to zero due to the lack of net radiative energy, moisture recycling due to evaporation of previously fallen precipitation was unlikely to happen directly at the site.

Nevertheless, it has been shown (Froehlich et al., 2008) that even a relative small contribution in the order of a few percent of such recycled moisture increases remarkably the deuterium excess.
6) Reply: Froehlich et al. 2008 analysed d-excess values in precipitation and their seasonal changes for a 21 years data set based on monthly mean values. They showed that the seasonal changes in d-excess in precipitation are linked to changes in evaporative fraction. For this purpose a mixing model approach with estimated d-excess values for the evaporative (recycled moisture) d-excess and the advected d-excess was used.
We would like to point out that the presented paper analyses changes of d-excess **on a time scale of hours rather than on a yearly time scale**. For us it is a priori not clear whether the same processes apply for time scales that differ by three orders of magnitude. An increase of evaporated fraction during April to July could reasonably be linked to the changes in leaf area index as it was done by Froehlich et al. 2008. However, this argument does not apply to our data collected within some hours in autumn. Changes of evaporative fractions as a result of changes in radiation are also very unlikely as most of the cloud events were probed over night when it was dark and we do not observe any clear pattern with e.g. higher values at day and lower values at night time due to a higher recycling rate at day time. Moreover, it is not clear whether evaporation rates from steep slopes in the alpine terrain can be compared to evaporation rates in predominantly flatter terrain in Germany (see Reply 7 for the full

text of the revised Section 3.1.2).
For a discussion of these aspects, the authors may also consult a paper, which is not included in their list of references: Jun Cui, Shuqing An, Zhongsheng Wang, Changming Fang, Yuhong Liu, Haibo Yang, Zhen Xu, Shirong Liu (2009) Using deuterium excess to determine the sources of high-altitude precipitation: Implications in hydrological relations between sub-alpine forests and alpine meadows. Journal of Hydrology 373 (2009) 24–33.
7) Reply: We thank K. Froehlich for drawing out attention to this very interesting paper. Cui et. al measured isotope ratios in rain and fog (amongst others) during around two weeks in the rainy season on the Chinese plateau. They found elevated d-excess values of 35‰ (rain) and 38‰ (fog) compared to the monsoon precipitation (10‰). They concluded "that a large part of rain at the alpine meadow derived from secondarily evaporated water and that fog derived from the evaporated water produced shortly after rain events" and that the "contribution from evaporated water to precipitation in the alpine meadow therefore had to be mainly from sub-alpine vegetations in the region."
The large difference between the local rain and fog and the monsoon precipitation ($>$20‰) is striking and a clear sign for strong moisture recycling. However, the d-excess of the cloud at Schmücke (mean value 14‰) is only around 2‰ higher than the d-excess in precipitation at the GNIP station Wasserkuppe (60 km upwind but comparable in altitude) in September and October. However, the location and the climatical conditions are different and therefore we doubt that the findings from Cui et. al can be directly transfered to our site.
The site is 30°N and at 3780 m a.s.l. so the radiation is much higher (Summer, higher altitude and closer to the equator) than at Schmücke. Resulting evaporation rates are therefore also expected to be much higher than evaporation rates in Germany. Moreover, the precipitation during the measurement period was $\approx$120 mm which is twice as much as the precipitation measured at Schmücke in 6 weeks ($\approx$76 mm). So local — or as proposed by Cui et. al — regional moisture recycling will be surely much

more pronounced at the Chinese site compared to Schmücke. Moreover, the 8 fog events presented by Cui et al. mostly formed on the same day after the rain event. This is different for the cloud events at Schmücke: for four events (18 samples) there was no rain at Schmücke since the last cloud event, and for one event (one sample) there was no rain within the last 6 days. 6 Events (19 samples) directly formed after rain and two within some hours after rainfall. Interestingly, the d-excess for the cloud samples formed after rain is higher (mean value: 17‰) compared to the cloud water sampled when there was no rain prior to the cloud event (mean values: 13‰). So, we think that this could indeed be a sign for moisture recycling, however to a smaller extent than presented by Cui et. al. Because we showed that the contribution of local evaporation is small (see reply 5), we therefore assume that the moisture recycling is likely to happen on a larger scale. We therefore changed Section 3.1.2 to

D-excess of the cloud water and moisture recycling

The d-excess ($d = \delta^2_C\mathrm{H} - 8 \times \delta^{18}_C\mathrm{O}$) of the Schmücke cloud samples was rather high (10 to 20‰, Fig. 4, mean value 14‰) as compared to European air moisture d-excess of 7 to 11‰ (Gat et al., 2003) and stayed rather constant during most of the cloud events, except for events 1 and 10, which are discussed in more detail in Sect. 3.2.3. D-excess tended to be higher in cloud events that developed directly after rainfall (mean value: 17‰, blue dashed line in Fig. 4) than in cloud events that formed after a cloud free period without rainfall (mean value: 13‰, red dashed line in Fig. 4). The elevated d-excess could be an indicator of moisture recycling whereas a lower d-excess may represents an early stage condensation. However, as most cloud events were sampled during nighttime when evaporation rates are closed to zero due to the lack of net radiative energy, moisture recycling due to evaporation of previously fallen precipitation was unlikely to happen directly at the site.

At this time of the year, precipitation at the site is commonly linked to large-scale precipitation occurring during frontal passages. This implies that precipitation at Schmücke is most likely occurring simultaneously with (or slightly delayed after) precipitation upwind of the measurement site. Consequently, the elevated d-excess

of those cloud events sampled directly after rainfall was most probably caused by moisture recycling upwind of the Schmücke rather than by different climatic conditions at the initial moisture formation above the ocean. This view is in agreement with measurements of elevated d-excess presented by others both in rain and fog (Gat and Matsui, 1991; Rhodes et al., 2006; Froehlich et al., 2008; Cui et al., 2009).

The new Figure 4 is shown in the supplement (Figure 3 in the supplements)

In the Abstract we changed:

p.15140, l.10-12 to:

The d-excess was higher in clouds developing after recent precipitation revealing episodes of regional moisture recycling.

p.15140, l.25: added: *and regional recycling of moisture.*

p.15143, l.5: added *D-excess in precipitation also increases due to moisture recycling (Gat and Matsui, 1991, Henderson-Sellers et al., 2002, Rhodes et al., 2006, Froehlich et al., 2008).*

p.15143, ll.9-12: replaced

However, measuring d-excess in cloud droplets could indeed reveal insights into the initial isotopic signature of the moisture source region, as cloud droplets did not experience any additional kinetic fractionation as rain droplets do.

by

However, measuring d-excess in cloud droplets could indeed reveal insights into the initial isotopic signature of the moisture source region, as cloud droplets are not affected by additional kinetic fractionation as rain droplets below the cloud.

p. 15152 l.5: we added: *to the water vapor transport and to moisture recycling*

In the conclusions we changed p.15162 l.11-12 to:

Changes in d-excess were most probably related to continental moisture recycling.

All in all, the observed changes in d-excess should be seen as a phenomenon caused by recycling of evaporated moisture from the ground. In this study, recycling from regions close to the sampling station should be given due consideration. The original

isotopic composition of the evaporating water in the ground represents the isotopic composition of precipitation fallen prior to the observation period. Therefore, looking at the isotopic composition of precipitation and its sources is also of interest, at least in connection with the explanation of the observed changes of the δ values in the cloud droplets.

8) Reply: Unfortunately, precipitation event resolved isotopic data are not available. Compared to the closest GNIP station, moisture recycling is indeed possible. Please see the reply 7 for details.

A considerable part of the paper is devoted to the description and application of the model developed by the authors to understand the temporal change of the isotopic composition of the water vapor and the droplets in the cloud. However, the description of the model appears to be rather obscure and difficult to verify.

9) Reply: Changes in δ values in the cloud droplets on the timescales of hours are either linked to changes in local thermodynamics or in changes of δ values in the advected vapor. In order to address this question this model is needed. As it is a rather complicated procedure the model description is long and partly difficult to follow. We therefore rewrote the section with the model description. For details please see the replies 8 and 9 to referee # 1 and reply 6 to referee #2.

It remains to be shown that the model calculations really contributed to a better understanding of the observations.

10) Reply: We do indeed think that this model helps to understand what the drivers of the changes in δ values on the order of hours really are. It is a priori not clear that the contribution of local thermodynamics such as changes in temperature and condensed fraction are not responsible for the observed changes. Figure 4 summarizes these findings (see comments A.L. Rhodes C7308, second paragraph, last sentence). For some events local thermodynamics were more important and for others the δ values of the advected water vapor dominated the temporal evolution of the isotopic composition of the cloud droplets. So these results show that there is no general mechanism, which always explains the temporal evolution of the δ values in cloud droplets. We

carefully edited this section for clarity, for details on the changes, see replies 9, 24, and 28 to A.L. Rhodes.

Unfortunately, the authors missed the unique opportunity, to measure the isotopic composition of both the droplets and the water vapor in the clouds (see also last sentence of the paper, page 15163, line 7-11).

11) Reply: Continuous measurements of δ values in water vapor require expensive laser spectrometry measurements and a laser-based spectrometer was not available for this campaign. Moreover, to the extent of our knowledge no MS has been published on measuring δ values in water vapor in a supersaturated environment such as a cloud. However, because we know from the results of the boxmodel calculations that the changes in δ values due to changes in condensed fraction are rather small, measurements of δ values in water vapor below the cloud base would be sufficient. Therefore, we write in the conclusions that such measurements including additional measurements even further away would be desirable for future measurement setups.

In conclusion, the paper should be re-written taking the above comments into account. The main goal of the revised version should be a presentation of the measured data together with a description of the sampling and measuring procedures and the measured ambient parameters. The discussion and interpretation of the data should be more succinct as in the present version and conclusions should be avoided which hardly can be verified by available data and observations.

12) Reply: We rewrote parts of the methods, results and discussion section taking the presented concerns into account. We also shortened certain passages. For details we refer to the answers given above.

Since a major revision (re-writing) of the paper is recommended, no specific comments on the writing clarity of the present version will be given.

References used in the replies:

Stull, R (1988): An Introduction to boundary layer meteorology, Atmospheric sciences library, published by Kluwer Academics Publishers, The Netherlands

Additional references included in the MS:

Jun Cui, Shuqing An, Zhongsheng Wang, Changming Fang, Yuhong Liu, Haibo Yang, Zhen Xu, Shirong Liu (2009) Using deuterium excess to determine the sources of high-altitude precipitation: Implications in hydrological relations between sub-alpine forests and alpine meadows. Journal of Hydrology 373 24–33.