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Comment

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

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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.

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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. 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). 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.

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.

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. However, on p. 15159, the manuscript does suggest that a "biospheric signal"

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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.

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. 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. 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.

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. A figure with a location map will also be helpful.

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

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into separate sentences. Semicolons are used to separate two related, complete sentences. If the sentences are not closely related, separate them with a period.

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.

Other specific comments toward improving writing clarity:

p. 15141, line 25: “fractionation” is misspelled.

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

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.

p. 15150, line 4-6. Awkward sentence structure; misuse of colon.

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.

p. 15152, lines 25-27. Awkward expressions, including reference to Rank and Papesch (2005).

p. 15152, line 21: Change “Additional water uptake of polar air masses...” to “Additional water uptake by polar air masses...”

On p. 15154, line 14: The paper states that below-cloud evaporation will affect d-excess in rain — by how much typically?

p. 15155, lines 13-15. Awkward sentence. Simplify sentences.

p. 15156, lines 23-25. Dangling phrases make this sentence difficult to follow.

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p. 15158, line 24-25. Fix structure “. . .do not allow to determine. . .”

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.

p. 15160, lines 2 – 5. Awkward sentence structure. Use of word “as” is confusing.

Other awkward passages may exist. Edit carefully for clarity.

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 model 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.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 12, 15139, 2012.

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