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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Stable water isotopologue ratios in fog and cloud droplets are not size-dependent" *by* J. K. Spiegel et al.

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General Comments:

Overall, this is a very good scientific contribution for ACP. The title of the manuscript summarizes the main finding quite clearly, and the methods and data analysis are sound and well substantiated. The study aimed to test whether the assumption proposed by Bolin (1958) and later modeled by Jouzel (1986)–that droplets of different sizes within a cloud are in isotopic equilibrium with each other–could be confirmed by direct measurement. The study design addresses the question well, and the results show that differences in isotopic composition of cloud water are due more to the length of the sampling interval than to size of the cloud droplets.

The presentation of the manuscript is good overall, and there are a few places, described in the next section, that can be improved for clarity. More notably, the study elicited additional questions for me that remained unanswered in the manuscript's discussion. For example, the study showed that the isotopic composition of cloud water varied significantly over different sample durations (e.g. Fig. 3). It makes sense that this could happen, but some explanation as to why could be helpful for many readers. For example, the "amount effect" might explain why isotopic values became lighter over the event duration if precipitation is occurring simultaneously. Was it raining during these events? The "amount effect" would not explain increasing isotope values, however. Would changes in temperature during the event explain them? Please see comment #6 in "Specific Comments" below that elaborates on this.

Also p. 12665, lines 19-20, state "different droplet sizes could carry isotopic information from different locations in the cloud," based on the assumption that larger droplets typically are at the base of clouds whereas smaller droplets are higher. Do the authors feel their results address this statement? In other words, do they know whether they are sampling different droplet sizes within one region of a cloud, or do the droplet sizes measured over the course of the event represent water collected from different regions in the cloud, which may have occurred if the cloud changed its elevation over the sampling interval? The authors may not be able to answer these questions directly from the data, but the question posed in the introduction could be addressed. Some further discussion about the changes in composition over time is warranted, including the implications of this result, as well as proposing other questions for future work. For example, one of the co-authors (Scholl) has written about isotopic differences between rain and cloud water collected during the same event. Do the "elapsed time" data in this manuscript relate to those prior observations in some way?

Specific Comments: 1. p. 12664, Lines 11-15. The last 2 sentences of abstract state that observations of different isotope ratios for different cloud droplet sizes result from the collector scavenging cloud moisture at different moments in time, which may

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happen as the cloud dissipates. This interpretation also needs to be in the conclusion section of the paper. The term “dissolution period of the cloud” needs to be defined in discussion.

2. p. 12665, lines 25-29: A description of how the paper is structured is unnecessary because it follows the standard format of the journal. Better to replace this text with a summary of the main argument and evidence that supports the conclusion in the first sentence of the paragraph.

3. p. 12667, line 12: Please define the term “50% size cuts.”

4. p. 12671, line 6: Add the word “increasing” to clarify that the t99 decreased slightly with increasing temperature.

5. p. 12671, line 19: The term “time” needs to be clarified as elapsed time. Later in the paper, it is apparent that the elapsed time varies from collection period to collection period, but how the collection time is used in the statistics presented in Table I is not clear. Please elaborate how you tested significant differences between different times.

6. p. 12672, first paragraph under section 3.3 (lines 6-14): Much of this could go in methods section, which would also help clarify statistical analysis presented in Table I. Also, the reader is referred to Spiegel et al. (2012) to understand the temporal evolution of the isotope ratio during cloud events. That’s fine if interpretation warrants a separate publication, however a summary of those interpretations is needed in this paper too (and the authors can refer to Spiegel et al. (2012) for the details and justification). A summary is needed so the paper will read more completely on its own because the variation in isotope ratio is very pronounced in the temporal data (as presented in Table I and Figure 3). Each paper should be able to stand on its own. It is okay to present summary of findings of the complementary manuscript.

7. p. 12673. I appreciated the presentation of the statistical analysis in the first paragraph on this page.

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8. p. 12673, lines 21-26: Too much information is in this sentence. Rewrite to make less awkward, and to improve clarity.

9. p. 12673, lines 27-28: Reference to Fig. 5c. Discussion needs to explain Figure 5 completely; it is difficult to understand from just the figure caption. In particular, I do not understand part d of Fig. 5 and what the hypothetical d2H evolution in cloud droplets represents. Also, make sure the acronym "LWC" (liquid water content) is defined previously in the paper.

10. p. 12674, line 1: Clarify what "least saturated sampling conditions" means.

11. p. 12675, first paragraph. Please clarify use of term "evaporated' in this section. Readers could be confused, thinking that smaller water droplets in the collector may have evaporated, which would change the isotope ratios of the water (and account for the collection artifact this way). Instead, I believe the paper attempts to describe an evaporation process in the cloud that affects whether small water droplets are available to be collected. Be carefully clear in this section because readers need to understand the interpretation of the "collection artifact." This needs to be summarized in the conclusion section (e.g. my comment #1). The interpretation is reasonable; if evaporation in the collector occurred, the data should show a systematically higher isotope ratios for the small droplet size (which wasn't observed).

Technical Corrections:

1. p. 12665, line 1: remove "e.g."

2. p. 12672, line 20: expression measured in fog "so far" is awkward.

3. Figure 3. End of figure caption is missing a period.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 12663, 2012.