Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-1313-AC1, 2019

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Vertical profile observations of water vapor deuterium excess in the lower troposphere" *by* Olivia E. Salmon et al.

Olivia E. Salmon et al.

lwelp@purdue.edu

Received and published: 9 May 2019

See attached pdf for formatted version with author responses in color.

Authors' Responses to Reviewer 1 âĂć Author responses are indicated in blue font. âĂć Locations of our edits in the "strike-through" and final versions of the manuscript are provided with the following convention: ([strike-through version] pg. #, ln # / [final version] pg. #, ln #).

1 Major comments

1. I am not convinced by the way the Rayleigh model is used as an explanation in flights RAY and STC. Even though a Rayleigh model can match much your data, it does not necessarily indicate that the guiding processes are correctly represented. In

particular, the presence of a quite homogeneous mixing line of water vapour below the inversion, topped by a pronounced inversion layer, and relative humidity below 10% is in stark contrast to the moist adiabatic profile that would be associated with a Rayleigh model. I recommend to include the possibility of long range advection of air with subsequent mixing into the picture, as well as considering the role of subsidence of air as a contribution to the inversion layer. Can several mixing processes in time and in the vertical combined look like a Rayleigh curve? This analysis should be placed in the context of the critique of Taylor (1984) on Rozanski and Sonntag (1982), and the very valuable study by Gedzelman (1988) to analyze Taylor's (1972) data set. Maybe a conclusion of your study could be a critique rather than a statement of consistency with the Rayleigh model? The Reviewer makes an excellent point that our flight conditions during the RAY (now CLR) flights are not consistent with moist adiabatic conditions that would be required for Rayleigh rainout processes to be actively occurring. However, the Rayleigh prediction is most consistent with our observations, compared to mixing scenarios as we now show in Figure 7. We have changed this discussion to indicate that observed profile is a fingerprint of previous airmass dehydration conditions that is retained by transport and downward mixing of dehydrated higher altitude FT air, consistent with past studies (Taylor, 1984; Gedzelman, 1988). (pg 19, ln 2-28 / pg 8, ln 10-33)

2. The interpretation of the stratocumulus case study as evaporating cloud drops needs further corroboration. The literature cited on the kinetic fractionation of evaporating droplets, such as Stewart (1975) considers rain drops, which are orders of magnitude larger, and have a vertical downward motion. This provides the potential of partial evaporation, leading to a kinetic fractionation signal in the dexcess. Cloud drops, however, will rapidly evaporate completely as they become unstable at smaller radii, leaving no sign of kinetic fractionation in the d-excess. Furthermore, they are suspended in the atmosphere, thus allowing for equilibrium fractionation. Without the consideration of rain evaporation in addition, or other processes, the explanation of the STC case is thus not viable. What is the potential of the occurrence of rain evaporation from these stra-

tocumulus clouds? Can you estimate how much cloud water specific humidity would have to be added to produce the negative d-excess signal, and is this consistent with the saturation specific humidity in the cloud layer? Could there potentially have been a potential for rain evaporation or ice processes in the airmass upstream earlier instead? The Reviewer again brought up some excellent details to consider. Our calculations using the equations in Stewart likely apply to cloud droplets evaporating just like rain droplets. Similarly, we realize that the assumptions of the Worden equations are not valid either because that is for a case of a dehydrating airmass (closed system Rayleigh). However, there are 2 important timescales to consider when exploring the influence of liquid droplet evaporation in the inversion layer. (1) The timescale at which a liquid droplet isotopically equilibrates with its surrounding vapor. (2) The speed at which droplets move through the inversion layer. We calculated the first timescale using Eqn. B5 is Bolot et al., 2013 which is $\sim$2 seconds for a cloud droplet size of 15 microns. For our observations of vertical wind speeds, the time for a droplet to move from the bottom to the top of the inversion is 19 seconds. These calculations do indicate that cloud droplet evaporation starting in the lower inversion and finishing in the top of the inversion during the flight conditions in unlikely. However, if previous inversion conditions were colder, if the droplets were larger than 50 microns (like drizzle), or if the inversion had faster vertical wind speeds, these values could converge. We have edited the discussion of the STC flight day to reflect these new considerations. (pg 24, ln 19 – pg 25, ln 39 / pg 10, ln 17 – pg 11, ln 2)

3. Section 3.3 (DBL case) describes and interesting case of BL development, but is particularly difficult to follow. Consider reorganising the material in a more logical order, and formulate a clearer take-away in the end. We have consolidated the DBL case study's results (originally Sect. 3.3) and discussion sections (originally Sect 4.3) so that the case study's key isotope features are identified, and their cause(s) immediately discussed. Furthermore, we have extensively revised Section 3.3 (now Section 4.3), which presents the DBL case study results.

4. The material included in the Supplement only seemed marginally useful. It is also confusing to have both an appendix and a supplement in a manuscript. Consider merging the supplement into the Appendix, keeping only Fig. S1. This is a good suggestion. We have merged the Appendix and the important components of the Supplemental Information (SI) into the SI in order to further limit the length of the paper.

5. It is difficult to follow the description of the cases without additional context of the flight situations. Maybe Fig. 3 could be split up for each case, and supplemented by a weather chart similar to Fig. S6. We have split up Fig 3a-c to make three plots for the three case studies (Fig 5, 8, 10) so that they can be placed closer to the text where they are being discussed. Weather charts have been added to the SI for each case studies (Fig. S5.1 – S5.3).

6. The discussion sections 4.3 and 4.4 appear repetitive to what has already been presented earlier and can be deleted. We have consolidated the Results and Discussion sections so that discussion paragraphs directly follow their respective results sections. This reorganization has reduced repetitive parts of discussion sections 4.3 and 4.4. We have also reorganized the manuscript so that discussion of the campaign-wide observations (previously Sect 4.4, now Sect 3) now precedes discussion of the case study observations.

7. The second half of the first paragraph of the Conclusions section should be moved to the introduction/motivation. We have made the suggested change.

8. Important studies that should be referred to are Taylor (1972, 1984), and He and Smith (1999) and Ehhalt (1975, 2005) and Tsujimura et al., 2007. We did not focus on these studies because our primary interest was high-frequency deuterium-excess observations, but the reviewer is right that they provide great context for our work. This is why we now point to the the detailed overview of airborne water vapor isotope studies reviewed in the introduction of Sodemann et al., 2017 (pg. 3, ln. 37/ pg. 3, ln. 9).

9. I found the structure of the manuscript somewhat confusing, in that first the flights

around both Washington DC and Indianapolis are presented, and then almost all analysis focuses on flights from only one site, before returning to a wider overview in section 3.4. It would be more logical for the reader to move Sec. 3.4 ahead of the case studies, and then zoom into individual aspects. Consider adding a more intuitive way to present an overview of all data than a comparison to Rayleigh in 7. This is a great suggestion. We have followed the Reviewer's suggestion by reorganizing the manuscript so that the campaign-wide results/discussion precede the case studies' results/discussion sections. We have added a new figure (Fig. 3) which shows dD and d18O vs H2Ov along every VP descent conducted during the campaign. We no longer present our results relative to Rayleigh by removing the right panel of Fig 7 (now Fig 4 post reorganization).

10. Several times in the writing, wording such as "Fig. x shows ..." is used. For conciseness, consider rephrasing this to sentences talking about what is shown in the figure, added by a figure reference "(Fig. x)". This is a good suggestion, we have made changes where appropriate.

2 Detailed comments P3, L8: "However, relatively few...": As far as I am aware there is only one published dataset of airborne d-excess measurements. Why the citation of Schmidt et al., 2005 here? We have reworded the sentence and deleted the reference to Schmidt et al., 2005, which provides modeled d-excess.

P3, L34: Please state if the inlet or parts of it were heated. We have added a sentence stating the TWVIA inlet was not heated during the calibrations. (pg 5, ln 27-28 / pg 4, ln 35-36)

P4, L9: How sensitive are the measurements to variations of cavity pressure and temperature during flight? Once the isotope analyzer has warmed up (i.e. reached the manufacturer recommended pressure and temperature values for operation), cavity pressure and temperature only vary (1σ) by $\pm$0.02 Torr and $\pm$0.08oC, respectively, over a vertical profile descent on average. This is within the operating specification

given by the manufacturer. (pg. 5 ln 13-15 / pg 4, ln 21-22)

P9, L21: Worden et al. 2007 formulate their models for rain evaporation (falling condensate), not cloud evaporation, see their supplementary information. We have removed all discussion of the Worden et al., 2007 model (see our response to major comment #2 for more explanation).

P10, L10: "Rayleigh-consistent observations": Consider rephrasing in light of major comment 1 This is a great suggestion. We have renamed the RAY case study to CLR, which is now representative of the case study's clear sky conditions rather than a possible interpretation of the day's dominant isotopic process.

Fig.4: Consider adding a panel that shows more mixing lines, e.g. between the bottom and top of the inversion, or the bottom and top of the BL, how do these compare with the complete mixing line? We have split up Fig. 4 into three figures for the three case studies (Fig. 7 CLR, Fig. 9 STC, and Fig 12 DBL). Each new case study figure has an additional panel showing mixing lines for scenarios relevant to the flight day. The new mixing panels are discussed in each case study's respective sections (Sect 4.1-4.3).

P12, L1: Consider using a standard symbol such as q or m for specific humidity or mixing ratio of water vapour, rather than H2O. We respectfully choose to retain H2Ov to represent water vapor mole fraction, as it is a common convention in the atmospheric chemistry field.

P14, L8: "relative to Rayleigh" rephrase as e.g. "Rayleigh model predictions" The sentence now reflects this suggestion.

P14, L16: "d-excess switches to tracking" rephrase This sentence has been rephrased.

P14, L13-22: hard to follow, rewrite this section for clarity We thank the Reviewer for identifying this paragraph that could be improved. We have revised this paragraph to include a summarizing, take-away sentence, which highlights the unique d-excess characteristics of the first vertical profile flown on STC (pg 23, ln 17-20 / pg 9, ln 36-40).

P16, L6: "Differences" - difference to what is shown? Are these not absolute values? This sentence has been rephrased.

Fig.6: How are RL and INV defined? You could provide a quantitative comparison of averaged quantities for both layers that supports their distinction. To address this comment, we have added a sentence that reads, “We define the base of the RL using the same approach described in Section 2.4 ($d\theta/dz$ and $|d(H2Ov)|/dz$ threshold values) for determining the base of the INV (zINV).”

P17, L14: Not clear what the take-away from this section is. Revise paragraph. We thank the Reviewer for identifying this paragraph as an opportunity for improvement. We have revised this paragraph about our campaign-wide vertical profile observations. (Sect. 3, paragraph 1-2)

P17, L17: "Figure xxx shows": revise according to major comment 11 We have reworded sentences that begin with “Figure # shows. . .” throughout the manuscript.

P18, L12: Not clear what the take-away from this section is. Revise paragraph. We have deleted this paragraph because we decided its inclusion does not benefit the overall story about the campaign-wide observations section.

P18, L16: Why the citation of Lee et al. (2006)? Compare Gedzelman (1988) and the discussion of the Taylor (1972) data (major comment 1). During our consolidation of the results and discussion sections for each case study, we have removed the indicated sentence. We now discuss the possibility of Rayleigh-consistent condensation occurring upwind of the CLR measurements (see response to major comment #1). We also reference the suggested paper (pg 19, ln 17-28 / pg 8, ln 22-33)

P19, L3: The thermodynamic characteristics of the profiles are not consistent with a Rayleigh processes, even though the isotope composition may be – what is the conclusion from that finding? We now discuss the possibility of Rayleigh-consistent condensation occurring upwind of the CLR measurements (see response to major comment

#1). (pg 19, ln 17-28 / pg 8, ln 22-33)

P19, L15: How would cloud droplets be lofted but not the surrounding vapour? If vapour and droplets move together, how can non-equilibrium fractionation result? The vapor and droplets do not necessarily have to remain in isotopic equilibrium under changing conditions, including mixing of airmasses. We have revised the indicated sentence so that cloud droplet evaporation is discussed from the framework of timescales, e.g. the time required for condensate and vapor to isotopically equilibrate vs. time required to transport a cloud droplet from the bottom to the top of the inversion. Please see our response to major comment #2 for more detail. (pg 24, ln 19 – pg 25, ln 39 / pg 10, ln 17 – pg 11, ln 2)

P20, L16-20: Consider the possibility of rain evaporation upstream (major comment 2). We now discuss how rain droplet evaporation in dry, cold conditions upwind of the STC flight measurements could possibly explain the minimum in d-excess at the top of the inversion layer. (pg 25, ln 33-36 / pg 10, ln 36-38)

P21, L10: What relative roles could vertical motion and entrainment of dry air from aloft play in the evaporation of the cloud layer? This is an interesting question that we believe that stable isotope observations can inform about the occurrence of cloud evaporation, but are not sure that it’s possible to distinguish the driver of that evaporation (cloud drops moving into the FT versus entrainment of dry air into the cloud) using our observations downwind of clouds. This would require a sophisticated cloud microphysics model, similar to Bolot et al. (2013). Respectfully, it is outside of the scope of this paper, but an exciting future application. We do, however, cite papers that give evidence for stratocumulus cloud droplet evaporation occurring within and above the cloud layer due to differences in entrainment efficiency (pg 24, ln 36- pg 25 ln 2 / pg 10, ln 12-15).

P21, L20-29: Rephrase for clarity. Could ice-phase processes have been relevant further upstream, earlier in time? We have rephrased the paragraph about the effect condensation in the presence of ice has on vapor d-excess for clarity. We also now include the following sentence, “It is possible, however, that condensation under ice-supersaturated conditions occurred prior to the STC flight, and that the resulting isotopic imprint was maintained during transport to Indianapolis.” (pg 26, ln 28-30 / pg 11, ln 15-17)

P22, L12: What is the Rapid Refresh Model? Could be deleted here. Reference to the model name has been deleted.

P22, L7-34: reorganize and rewrite this section for clarity We have revised the entire results/discussion sections related to the DBL case study to improve their clarity. (now Sect 4.3) P22, L36: "so we look" rephrase This phrase has been deleted as part of the manuscript reorganization.

Fig. A1: What is the reason for the large scatter between both instruments in the mid-range of humidities? Does longer averaging of the data provide a better agreement? Was the scatter similar for each of the flight? How is the scatter for the downward profiles only? The scatter corresponds to few points relative to most of the points that show good agreement. The mid-range humidities in Fig. A1 (now Fig. S1 due to the Appendix-SI merger) correspond to vertical profiles. These few points that do not show as good agreement result from the TWVIA data being low pass filtered relative to the Picarro due to a longer residence time, as we mention in Sect 2.2.2 (SI pg 5, ln 23 / SI pg 4, ln 31).

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-1313/acp-2018-1313-AC1-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1313, 2019.