Anonymous Referee #1

Received and published: 28 July 2016

General comments:

The study "Response of water vapour D-excess to land-atmosphere interactions in a semi-arid environment" by Parkes et al. analyses the interplay between changes in atmospheric moisture isotopic compositions and the impact of local scale forcing of evapotranspirative vapor isotopes. As recently the isotopic composition of atmospheric moisture has been proposed as a tracer of large scale moisture recycling, this is an interesting topic. Also, apparently the impact of isotopic compositions was often studied using modelling approaches, which is rather surprising to me, given the increasing amount of isotopic ET studies recently! The overall quality of the paper notwithstanding, I see quite some space for improvement both technical and content related.

We appreciate the reviewer's comments and thank them for their time. Our comments and responses are shown in italics and line references are for the revised manuscript. In the revised manuscript, specific changes are in red. More general changes have not been highlighted but are referenced in response to the reviewer's comments.

With respect to the reviewer 1's general comment "Also, apparently the impact of isotopic compositions was often studied using modelling approaches, which is rather surprising to me, given the increasing amount of isotopic ET studies recently!"*, we are unsure of the reviewers meaning or the section of the paper they are referring to. Perhaps they refer to the sentence in the introduction (lines 115-116) that states "The studies of the dv diurnal cycles have largely relied on isotopic models to assess the contribution of ET fluxes, but a lack of dET measurements make it difficult to draw robust conclusions." This is true, as the papers we refer to (which study the diurnal cycle of* d_v *and the role of ET) have used process based models (Welp et al., 2012), empirical models (Simonin et al., 2014), or provided an interpretation of their data* (Zhao et al., 2014) *without providing any direct measure of D-excess in ET.*

1) In my opinion the study campaign was rather short, only 2 weeks roughly, and to my mind the significance of the interpretation is hence limited.

While the study period was relatively short, our study was completed in a remote semi-arid environment. These ecosystems are largely under-represented in isotope literature, especially within Australia, and also in using in-situ analysers. Given the duration, we do not make any major conclusions or claims about impacts on the hydrological cycles. Indeed, we deliberately focused the conclusions so that they relate to the specific meteorological conditions observed during the experiment (quiescent meteorology and extended dry periods, which are common to semi-arid environments) and what these might mean more generally for d_v *and* d_{ET} *variability over the longer term, as well as for other conditions and environments. We believe that this is an important and quite novel set of results.*

Beyond the ecological setting of our study, providing direct measurement of d_{ET} *is also novel, and shows that for our location and meteorological conditions, ET does not cause the relatively high D-excess values. We believe this is a useful and interesting finding, especially when in terms of providing context to other studies of longer duration and in different locations (e.g. Bastrikov et al., 2014; Simonin et al., 2014; Welp et al., 2012), we observed a very similar diurnal cycle. We have used section 4.2 in the discussion to elaborate on the context of our work: in particular, the long dry period leading to very low soil moisture D-excess values and how this may be applicable elsewhere.*

We have modified section 4.2 to further emphasise the context of our measurements. In particular lines 576-587 deal directly with the context of measurements relative to key studies. In addition, section 4.3 includes reference to the context of our d_{ET} values and we have tried to make this clearer for the reader.

We have also made sure that throughout the discussion our conclusions are related to our specific conditions (i.e. semi-arid environment with extended dry periods between rain events, and low rates of moisture recycling).

2) I have some methodical concerns regarding laser spec calibration and chamber construction (see detailed comments).

   Fully addressed below for relevant specific comments.

3) To my mind both results and discussion section are rather long and very detailed. Moreover, quite often results are repeated within the interpretation section, making the manuscript rather hard to follow at that point (very unlike the intro and M&M part btw.). I suggest to focus on the main results and shorten both parts to make it easier to follow.

   Comments noted and we have streamlined results and discussion sections where appropriate. Some re-ordering of text has been carried out to help make the flow of the manuscript easier to follow. In addition to minor rewording of passages, sections 3.2 and 4.2 have been reworded and reordered, section 4.1 has been split into 2 sections.
   We have ensured that results are not over-repeated within the discussion section. While this has made the manuscript easier to read, the results, interpretations and conclusions remain unchanged.

Specific comments:

48ff: Be more specific! How?

Sentence modified – "Spatial and temporal variability of D-excess in ET fluxes therefore needs to be considered when using dv to study moisture recycling and during extended dry periods may act as a tracer of the relative humidity of the oceanic moisture source." – lines 47-50.

60ff: I think this is a bit overstated, there are surely some examples here!

We have clarified this sentence to reflect that datasets directly quantifying land-atmosphere exchange processes are rare – "To do this effectively, a range of datasets that directly quantify a variety of processes represented within these models are required. Unfortunately, datasets that directly measure land-atmosphere exchange at the process level are limited." Line 58-61.

63ff: Shouldnt this be 2 sentences?

We are not sure what the reviewer is referring to here, as this is already two sentences.

73ff: how about transport processes? i.e. kinetic fractionation?

We have changed the sentence to include 'equilibrium and kinetic isotopic fractionation' i.e. "The utility of water isotope ratios for tracing sources of moisture derives from the characteristic equilibrium and kinetic isotopic fractionation that occurs when water undergoes a phase change, causing light water molecules to preferentially accumulate in the vapour phase."- line 72-74.

81ff: Again, doubt there are so few. How about Berkelheimer, Simonin, Welp and others?

Our statement is that there are relatively few studies using vapour, relative to precipitation focused studies. The references mentioned by the reviewer do indeed discuss land atmosphere exchange for vapour isotopes, and we have referred to these in other sections of the manuscript. We have added (e.g. Aemisegger et al. 2014; Risi et al. 2013) to indicate some of these related studies - line 81.

98: Suggest to change "given this" to therefore

Noted and adjusted – line 99.

140: have has? omit has?

Noted and adjusted line 141.

168: If you indeed did not calibrate or drift check the LGR i think your values have a high uncertainty. I.e. the average difference to the Picarro might be small but you standard deviation suggests there was a high point to point difference. At the least it would be nice here to see the time evolution of the difference between laser specs throughout the campaign!

We are not sure if the reviewer is questioning whether we present raw LGR data (i.e. no calibration corrections applied) or whether there were no calibrations run during the campaign period. As stated in the text (section 2.2.1), we calibrated the LGR in the lab before and after the campaign to develop corrections for water vapour cross-sensitivity and linearity. This was completed simultaneously with the Picarro. During the campaign no calibration experiments were completed for the LGR, but to determine the importance of instrumental drift for our measurements, we regularly ran the two analysers simultaneously sampling ambient vapour (lines 180-190).

Reviewer 1 raises an important point regarding the drift for the LGR. While on average the Picarro and LGR agreed over the campaign, there was some shorter term drift that led to differences between analysers: most likely the result of the LGR's large temperature dependence. We did not include a comparison of the two analysers, as the differences between them is defined mostly by scatter with no clear trends. Interestingly though, there is no relationship between differences between the analysers and the major shift in H_2O concentration (i.e. wet vs dry period). This indicates that we have accurately characterised the water vapour cross-sensitivity of the two analysers and that this correction was stable throughout the campaign.

In line with Reviewer 1's comments we will include a figure showing the time series of differences between the two analysers over the campaign (see figure 1 below). So as not to detract from the main message of the paper we will place the figure and discussion of its consequences for our interpretations in the supplementary section S1 (as indicated on line 184). We have also added section headings to supplementary section, as it now has more than one section.

In constructing this figure we realised the biases listed in the paper were for the whole comparison period, which included nocturnal hours. This is not what we have indicated in the text (line 180) and is not a fair comparison for our measurements, as the LGR showed a very big temperature dependence that led to nonsensical values at night (hence our restriction of chamber measurements to 09:00 to 17:00). The comparison is more favourable during the day when the LGR cavity temperature was relatively stable and chambers measurements were made. We have updated the bias statistics to the values shown in figure 1 below (line 201 in revised manuscript).

In some cases the differences between ambient d_v and d_{ET} were quite small, so I_{ET} calculated for the D-excess could be strongly influenced by LGR instrumental drift. However, this does not affect our interpretation, as for all chamber measurements that passed our QC requirements D-excess decreased during the measurement, illustrating that ET always had a negative forcing on d_v. To deal with the uncertainty caused by the relative instrumental drift of the two analysers, we have included *statements in the text emphasising that while I_{ET} would be influenced by drift, our interpretation of negative forcing remains the same – lines 436-442.*

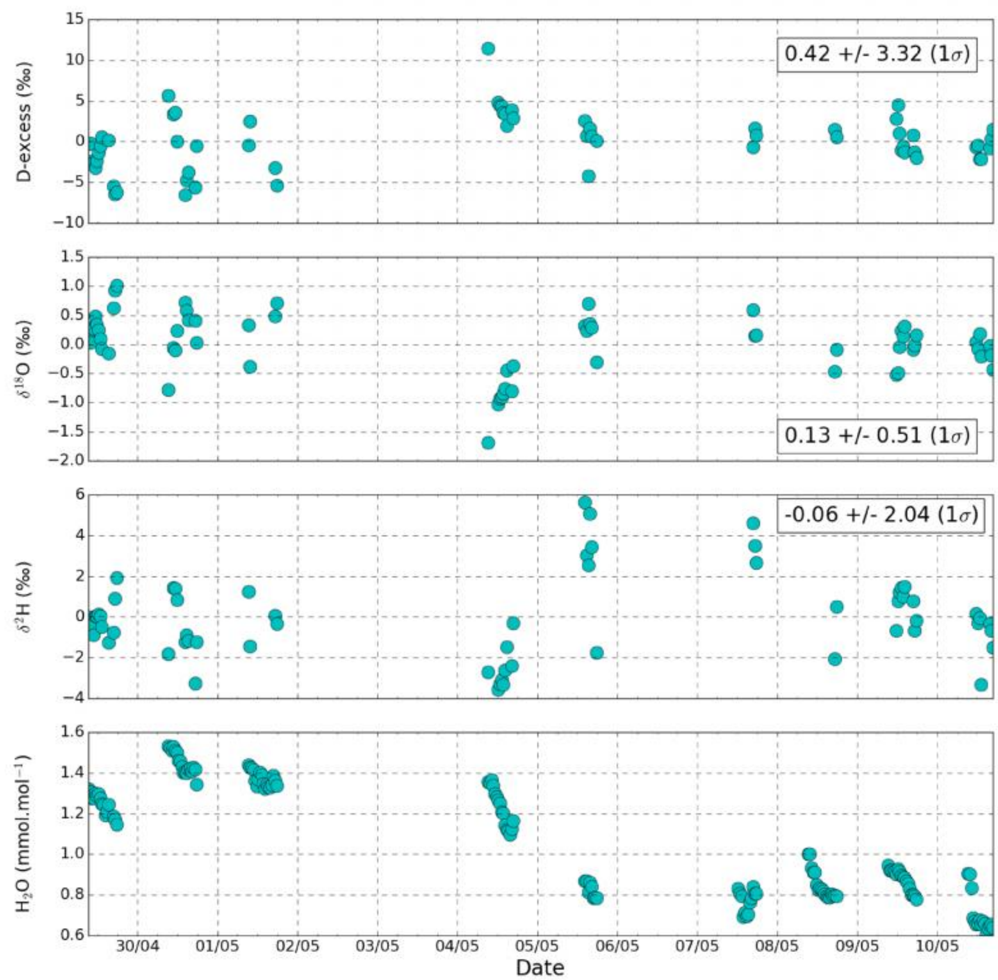

Figure 1: Time series of daytime differences between isotopic measurements of Picarro and LGR for periods when the LGR and Picarro were simultaneously sampling from the meteorological tower. The H_2O concentrations measured by the Picarro for these periods are shown.

207: Strong doubts concerning you placement of collars only 2 days prior to measurements! this will surely cut roots and there will be some affects in that direction.

We agree with the reviewer that collars were installed a relatively short time before the study period. However, the vegetation consisted of grasses with shallow roots (~5cm), so while near the edge of the chamber, roots may have been cut, the vast majority of the vegetation cover was unaffected. As with all chamber measurements, the apparatus can influence the environment and thus fluxes, but these would not change our interpretations here. We can add a sentence to include the reviewer's comment on this issue (lines 211-212).

211: Did you coat the chamber in some ay? It is well known that Plexi exchanges water and acts like a sponge creating a smearing effect in background chamber and vice versa transitions. This could actually affect you keeling plots quite much.

No, we did not coat the chamber to reduce memory effects. We assume the reviewers comments are related to memory effects influencing ET isotopic compositions calculated from the Keeling plots of chamber measurements. As the reviewer correctly notes, memory effects could have a major effect on the determined ET isotopic compositions. Indeed, we considered this and to combat memory effects we employed high flow rates, as the high turnover rates will reduce such memory effects. We also developed quality controls for Keeling plots, ensuring linearity and a significant H_2O concentration change was observed.

While memory effects are unavoidable and can influence ET isotopic compositions, we believe by ensuring linearity of the Keeling plot their effect was small and they do not change our interpretation. Memory effects are likely to attenuate the slope of Keeling plots, thus reducing the disparity between ambient vapour and ET isotope composition determined from the intercept of these plots. This is because chamber walls retain the isotopic composition of the ambient vapour being mixed with the ET flux. So, while memory effects would cause a high bias for the determined d_{ET} (i.e. Keeling plots for the D-excess always had a negative slope), they would still give a negative isoforcing. As such, minimal evidence of memory effects through our quality control procedures and that ET would still cause negative forcing on d_v, means memory effects would not change the interpretation of our results.

We have added specific comments regarding attempts to minimise memory effects in lines 222-225. Regarding using quality control of Keeling analysis to minimise memory effects, a comment is added between lines 251-254.

230ff: Why did you choose the Keeling method? Why not a mass balance approach?

Studies comparing the two methods have shown they are comparable (Lu et al., 2016; Wang et al., 2013), which is not surprising, as they are based on the same assumptions (i.e. that background concentrations and isotopic compositions of source and background water vapour does not change during a measurement). The main difference is that the Keeling plot requires extrapolation to determine the intercept of the $\delta_{chamber}$ vs $1/q_{chamber}$ plot. Comparisons in the literature have shown they agree well in practice. Considering the focus of our work was not to evaluate the two methods, we only present data using the Keeling model. In addition, as discussed in lines 246-262, we developed a filtering approach for the Keeling model focussing on ensuring linearity of our Keeling plots.

We have added a comment to indicate that we considered mass balance, but based on literature findings, decided it would not have made a major difference on results (lines 237-241).

256: Did you not measure soil water isotopes directly ? What is the uncertainty of the model approach?

Soil water isotopes were measured, as presented in section 2.2.5.

The uncertainty of the model approach is governed by the uncertainty of the chamber measurements of ET isotopic compositions and the parameterisation of Craig-Gordon (GG) model. While it is difficult to assess the accuracy of the CG model without direct observations, we did try different parameterisations (i.e. using Cappa et al (2003) vs Merlivat (1978) diffusion coefficients, and different values for the diffusion exponent). This had a large effect compared to uncertainty in ET isotopic compositions, but does not change the interpretation that soil water at the evaporation front was very enriched with very low D-excess values. While assessment of the CG model was not our focus, we have provided some mention of the uncertainty of the model and how this may impact upon our results/interpretations in the discussion of water pool isotopic compositions (results section 3.2 – lines 381-388).

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Anonymous Referee #2

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Summary:

The authors present an analysis of two weeks of atmospheric water vapour stable isotope measurements in a semi-arid environment. They focus on understanding the potential drivers of D-excess variability they observed in the near surface atmosphere. They use the short-term Keeling plot method to calculate the isotopic composition of the ET flux and find that under these conditions, ET cannot explain the increase in mid-day D-excess which has been observed in many other locations and studies. They use radon concentration measurements to constrain the influence of entrainment of moisture with a different isotopic composition from the free troposphere and don't find much support for an anomalous signal from the free troposphere. In the end, they conclude that the fact that mid-day D-excess is correlated with local RH, means that an oceanic evaporation signature is unchanged as the air mass passes over the dry land mass.

We thank the reviewer's for their comments. Our comments and responses are shown in italics and line references are for the revised manuscript. In the revised manuscript, specific changes are in red. More general changes have not been highlighted but are referenced in response to the reviewer's comments.

Major comments:

This paper is appropriate for HESS, but there are major flaws in the discussion and analysis that need to be addressed before publication.

1. The authors should provide more details of their methods. They should discuss analytical uncertainty of their measurements, especially the dET calculations. Small ET fluxes make measuring the dET values difficult. Were the plexiglass chambers tested for isotopic effects?

As noted in our response to Reviewer 1, we can include comments about analytical uncertainty into the results and methods sections, in particular, the Keeling plot intercepts and CG for soil evaporation front modelling. See response to reviewer 1 (lines 97-147, and lines 148-170) indicating relevant sections where measurement uncertainty, chamber memory effects and quality control are discussed along with the relevant changes.

For the chamber measurement, whether the ET fluxes are small or not is irrelevant for determination of d_{ET}. Our method for determining d_{ET} was based on using flux chambers and the Keeling plot method, so the change in H_2O concentration during a chamber measurement and the difference between the isotope composition of ET fluxes and ambient vapour are the variables that influence d_{ET} uncertainty. As discussed in the text in lines 246-262, we used a quality control routine to ensure that the assumptions of the Keeling method were met.

2. Throughout the discussion of the results, the authors comment on how their results contradict previous studies. The results are in fact different, but I believe they represent very different environmental conditions and the discussion should be prefaced with that in mind.

In fact our results are not different, as we observe a very similar D-excess diurnal cycle as other studies (e.g. Bastrikov et al., 2014; Simonin et al., 2014; Welp et al., 2012). *So in this* *sense we do not contradict other studies. However, by adding* d_{ET} *measurements we are able to provide a more conclusive role for ET fluxes in the D-excess diurnal cycle. While we contradict the conclusions of Simonin et al.* (2014) *and Zhao et al.* (2014) *(as noted lines 588-589), they do not provide direct measurements of* d_{ET}*. Others have been more circumspect* (Bastrikov et al., 2014; Welp et al., 2008). *Regardless, our results are very similar, but are able to provide different (or more conclusive) interpretations through directly measuring* d_{ET}*.*

As reviewer 2 indicates, it is certainly possible (likely) that we are observing different environmental conditions to the other studies referenced above. We agree with this statement and provided context of our findings in the discussion (4.2) and also mention this in the abstract. We can further modify section 4.2 to make this clearer: in particular in paragraph 2 of section 4.2 where we can add more direct reference to the literature for context of our results. These comments are similar to Reviewer 1's (lines 27-28 of response to their comments). We have made adjustments to the manuscript in line with their comments (lines 29-52 response to reviewer 1) which also addresses reviewer 2's concerns.

3. The discussion of using dv as a tracer of RH of the oceanic moisture source region contains many errors and is a misrepresentation of Aemisegger et al. The original application is to use dv along with d18O and dD to solve for temperature and RH of the oceanic source region, not to assume that RH near the ocean surface in 100%. Ocean surface humidity is more like 75% on average anyway. A strong correlation between local dv and local RH does not necessarily imply a preserved signature of the oceanic moisture source region. This would require that local and source RH are tightly coupled. Or, that changes in local RH are driven by mixing with a constant isotopic source of moisture (e.g. the free troposphere). The authors do not describe the Aemisegger approach correctly. Their aim was to estimate terrestrial evapotranspiration based on assumptions about the oceanic moisture source informed by back-trajectories and climate observations.

Reviewer 2 is indeed correct that the main aims of Aemisegger et al (2014) was to estimate terrestrial evapotranspiration using d_v *as a tracer. However, within their paper they use the precise methodology described in our section 4.1 to estimate the D-excess of the average liquid moisture source. We refer the reviewer to page 14 of section 5.1 and Appendix A in Aemisegger et al (2014). Please also refer to figures 7, 10 and 11 from Aemisegger et al (2014) where the methodology is applied.*

Reviewer 2 appears to have misunderstood the application of our methodology, which was taken from Aemisegger et al (2014). This methodology does not assume the RH near the ocean surface is 100% and it does not model the vapour D-excess of the moisture source. Instead the method uses the closure assumption of Merlivat and Jouzel (1979) and shows that for RH=100% the C-G model reduces to $R_v = R_l/\alpha$ *(*R_v*=vapour isotope ratio,* R_l*=liquid isotope ratio and* α*=equilibrium fractionation factor). By definition* α *for equilibrium processes is very close to 1, so that* $R_v=R_l$ *for RH=100%. Based on this derivation, Aemisegger et al (2014) use the relationship between RH and* d_v *and extrapolate to an RH of 100%. This reflects a weighted average of D-excess values for contributing liquid moisture sources.*

As the reviewer points out, this implies tight coupling between local and source RH. Exchange between the ABL and free troposphere could impact upon this relationship. There is no way we can determine if this was the case from our dataset (which we discussed in the same section). However, to produce the strong relationship we see between RH and d_v*, the free troposphere source of moisture and ABL moisture must have a relatively constant D-excess and RH, otherwise the relationship would be weakened. Likewise, for multiple moisture sources from the surface, as reviewer 2 surmises, are likely to significantly weaken the relationship between RH and* d_v*. So while we cannot rule out the influence of these effects, we conclude that the* d_v *during the day indicates a large unchanging remote moisture source: most probably a large reservoir such as the ocean.*

To accommodate the misunderstanding and concerns of reviewer 2, we have provided some additional details and discussion of the methodology of Aemisegger et al (2014). In particular, reference to the closure assumption of Merlivat and Jouzel (1979) is made (lines 565-566). We have also made it clearer that we are not aiming to calculate the D-excess of vapour at the remote moisture source, but the liquid source D-excess (lines 564-575). Additionally, in our discussion of the methodology we included details to address concerns about coupling between local and source RH, with direct reference to multiple sources and not accounting for ABL/free tropospheric exchange (lines 553-563).

3. This study is too short to examine synoptic variability with any depth.

We have not examined synoptic variability in depth: we simply refer to synoptic conditions to provide context for our short study. As outlined in addressing reviewer 1's comments (lines 29-52 of that response), given the relatively short duration of the campaign, providing some synoptic context was appropriate. In doing this, we refer to the specific conditions evident during the campaign, but also examine what conclusions may be relevant in a wider context. This is the purpose of section 4.2.

Specific comments:

ln 31: citation missing

We prefer to leave references out of the abstract as we feel it infers we are directly evaluating the referenced paper, which we are not. Relevant references are included in the Introduction.

ln33-35: there are a fair number of dET measurements published, which you discuss later in fact.

There are a number of studies presenting d_v *measurements, but only Huang et al. (2014) presents actual* d_{ET} *measurements, which is referenced in our paper.*

ln 126-127: Welp et al. measured dET

They measured d_v *(see abstract and methods) and modelled the D-excess of transpiration (see section 4.3). As we stated in the text,* d_{ET} *measurements were not made.*

ln 144: lat/lon

Done.

section 2.2.1: Please comment on the non-linearity of the delta values with respect to water vapor mixing ratio of the LGR analyzer and the stability of the calibration before/after the field experiment. The Picarro calibration method does not correct for water mixing ratio dependence of the analyzer. At what water levels were the analyzer uncertainties characterized?

We explicitly corrected for water vapour cross-sensitivities for both analysers, since this is one of the major contributors to measurement uncertainty. We have mentioned this on line 163 and lines 172-173.

ln 191: how long was the tubing and what was the flow rate in them?

We have added this information (line 195-197) – "Approximately 20m of tubing was required to connect the tower inlet to the analyser. A vacuum pump (MV 2 NT, Vacuubrand, Wertheim, Germany) was used to draw air through all inlets to the analyser at a flow rate of 10 l.min^{-1}*."*

ln 289: what modifications were made to West et al.?

Our modifications were minimal, simply using our own vacuum line. We have removed 'similar' from the text (line 304).

ln 374: significant periods of the day were excluded to characterize a diurnal cycle.

We agree that 'diurnal cycle' is misleading, so have changed the wording to indicate that we refer to the transition between the stable nocturnal and convective boundary layers. This section was modified in streamlining of the results, but is now included between lines 402 and 405..

ln 377-381: Is there any evidence that this much difference between soil water and the evaporation front could be real?

We believe this difference is entirely possible and not at all surprising. Dubbert et al. (2013) observed a large enrichment in soil moisture $\delta^{18}O$ values near the surface in their soil profile measurements, as did the seminal work of Allison et al. (1983). Besides literature evidence, our 0-5 cm soil measurements showed low D-excess compared to the LMWL indicating evaporative enrichment. It can be presumed that moisture at the evaporation front would be much more enriched and D-excess much lower. We have added further reference to the literature to support our measurements and expanded on uncertainty of modelled soil isotope values (lines 381-388).

ln 401-406: Are you referring to Fig 7 here? It's very difficult to see these features in the data as it is plotted.

Yes, we are referring to figure 7, as indicated at the start of this paragraph. We believe the drier mixing ratios observed from May 5th are quite clear in the plot. However, we can attempt to make this clearer to the reader. This has been included in streamlining results section, specifically lines 408-412.

ln 458-460: I'm not sure about this. I think you have to make a stronger case that it's not entrainment of air from above the boundary layer.

Indeed. We discuss this precise issue later (now in section 4.1 and section 4.2 –lines 553-563) and the fact that we cannot rule out entrainment as a possible explanatory mechanism.

ln 485: typo? 'encroachment'

Encroachment mixing is common terminology used in boundary layer meteorology, referring to the process where the mixed layer encroaches upwards as the layer warms.

ln 537-546: This paragraph has major problems. See #3 above. The authors come to some unsupported conclusions here based on a misunderstanding of many of the processes controlling vapor isotopes.

We disagree that there are any unsupported conclusions in the text and refer the reviewer to the comments above (lines 67-98).

ln 566-569: under what conditions was this observed?

The wording of this section has been changed as part of streamlining results and discussion sections, but we have made direct reference to Figure 8, which shows the drying trend the reviewer is questioning. Now reads - "Drying and depleting trends for water vapour, δ^2H and $\delta^{18}O$ throughout the day, particularly during the dry period (**Error! Reference source not found.**), indicate an important role for free troposphere entrainment." – lines 530-532.

ln 608-609: the two processes have very different fractionation factors as well

We have discussed this in more clarity in section 4.3.

ln 632: Didn't you screen out nighttime dET measurements? Consider showing a plot of dET time series.

Yes this is true. We have changed the terminology to indicate more explicitly that we are referring to transitional periods between the stable and nocturnal boundary layers – lines 624-626.

Fig 6: This figure needs more discussion.

We have discussed this figure across three separate paragraphs in section 3.2. If the reviewer could be more specific about their concerns we would be happy to address them.

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