Response to comments by Anonymous Referee #1

We thank Referee #1 for reviewing our paper and their positive feedback on the methods and analysis that we present in our manuscript. We are pleased to see that we could convey the message of the presented study in a convincing way, given the synopsis Referee #1 provides at the beginning of their review. We will first comment on the major issues raised by Referee #1 and further respond to each specific comment by Referee #1 below.

The manuscript describes evaporation dynamics of soil water in podzolic soils in the Scottish Highlands that are then related to surface vegetation and aspect. The authors sampled soils monthly and analzyed the δ18O and δD stable isotope values of soil water from four different depths for one year using an isotopic equilibrium method. The authors inferred evaporative losses from the soil water profile as well as explored relationships between isotope fractionation and deuterium excess (normalized to the local meteoric water line) with soil organic matter characteristics and corresponding vegetation (forest and heather). They hypothesized that their isotopic patterns were related to precipitation inputs and mixing processes in the soil. They found the unique characteristics of the research site to dominate the isotopic patterns they detected, especially at the near surface. In particular, the high organic matter content of the soil served as an important storage pool that potentially dampened an evaporative signal in the water isotopic patterns. During summer, when evaporative potential is high, the evaporative signal was strongest in the upper 15 cm.

The experimental design is tailored for testing vegetation influence on site hydrology, the methods are appropriate and the analyses were exhaustive. However, a justification or more information is needed regarding the use of hydrocalculator. The issue at hand is that at the core of hydrocalculator is the Craig-Gordon model, which is designed for open water surfaces – which the soil clearly is not. There are other models that consider diffusion in the unsaturated zone (i.e., Barnes and Allison, Journal of Hydrology, 1988) and they are able to model the profile while estimating an evaporative flux. In the study under consideration, estimating evaporation is not the focus per se, so removing these potentially erroneous estimates wouldn't be a problem. The figures are in general useful, however, I am afraid figure 8 is too busy to extract the point being addressed without investing a significant amount of time.

Response: We agree that the application of the Hydrocalculator is a first approximation and uncertain, since it was developed for isotope mass balances of open waters. We therefore revisited the evaporation estimates and changed the approach as follows to justify its application.

We do not use the Hydrocalculator anymore, where the exponent of the diffusion coefficient ratio (see Eq. 1 in Horita et al. (2008) or Eq. 6b in Gat (1996)) is assumed to be $n = 0.5$, but use $n = 1$, which is more representative for more stagnant interfaces like soil water (Horita et al., 2008).

We further now calculate the evaporation losses as a fraction of the original water source, rather than correcting the soil water isotope signal for seasonal variable precipitation input (as criticized in a comment by Referee #1 further below). We calculated the isotopic composition of the original water source as the intercept between the evaporation line of the soil water isotope data in the dual-isotope space and the local meteoric water line according to Javaux et al. (2016). This isotopic signal represents δ_P in Eq. 3 of our manuscript and the measured soil water isotopes represent δ_S. We are grateful for the suggestion to weight the isotopic composition according to the soil moisture and applied this in the revised calculations.

In conclusion, we believe that this revised approach to estimate the fraction of evaporated water is a better representation of the physical processes and we thank Referee #1 for their suggestions.

We are aware of the methods discussed by Barnes and Allison (1988), but we think that their presented approaches cannot be applied to our setting. The wet environment in the Scottish Highlands prevents a development of exponentially shaped δ^2H depth profiles. Quoting Barnes and Allison (1988): "infiltration of rainfall in this period will invalidate the approach (page 169)".

As discussed in section 4.3 and in accordance to Braud et al. (2005), we believe that a representation of soil water isotope concentration profiles needs to account for non-steady conditions and time variant atmospheric boundary conditions. However, we will add the following sentence in section 4.3: "Potential transient numerical modelling approaches that account for isotopic fractionation of the soil water isotopes are available (Braud et al., 2005; Rothfuss et al., 2012; Mueller et al., 2014)."

We also decided to take out Figure 8, since the graph is relatively difficult to understand and Referee #2 asked for shortening the manuscript.

Despite the issue of the evaporation estimates, it is clear that evaporation occurs within these soils and the role of vegetation is highly relevant. And while this is interesting, it is not entirely unexpected. Emphasis is also placed on the high frequency of the sampling (11 campaigns), but with advent of portable CRDS lasers even monthly samples are considered a rather low frequency sampling strategy (for example, see Volkmann et al., New Phytologist, 2016). Modelling is mentioned in the discussion (L198-213; pg- 22-23) although no modelling is performed and the results from the study are not really brought into the modelling discussion directly. There is a long history of investigating and modelling the soil water isotopic profile, and again while the results are interesting for this particular site, the title seems to promise more than the study can deliver. The merit in the study lies in the site-specific nuances such as the role of organic matter in effecting soil water capacity and the subsequent isotopic mixing that occurs.

Response: To our knowledge, there has not been an investigation of isotopic fractionation dynamics in pore waters over an entire year covered by almost monthly (11 sampling campaigns) sampled at the same date at four locations in parallel. In addition, our study site is a headwater catchment, with relatively difficult access, as well as being representative of other low energy, humid northern environments. We therefore believe that the presented results are not of limited interest restricted only to this particular study site, but are more widely relevant for stable isotope dynamics in soils of humid northern environments that have not yet been well studied.

Despite the new possibilities that come with in-situ measurements of pore water stable isotopes, only the recently published study by Oerter and Bowen (2017) applied it to cover almost one year limited to one particular location. However, for the most part, experimental studies used the high frequency sampling with in-situ stable isotope analysis are short term investigations covering few days (Volkmann et al., 2016; Beyer et al., 2016). Our motivation of the experimental set up was to cover the dynamics that occur within an entire year at a number of dominant landscape units. We will include the discussion above in the introduction of the revised manuscript. We will also include in the introduction the following to show the importance of studying the seasonal variability of soil water isotopes: "While studies based on two sampling campaigns (Goldsmith et al., 2012; Evaristo et al., 2016) were supportive of the ecohydrological separation, as presented by Brooks et al. (2010), newly published work with higher temporal resolution of *soil water and xylem water isotope sampling suggests that there are seasonal differences with regard to ecohydrological separation (McCutcheon et al., 2016; Hervé-Fernández et al., 2016).”*

Specific comments: Title: It is not exactly clear what is meant by “the soil-plant- atmosphere interface of the critical zone”. In this study soil water is measured, why not just state this?

Response: We consider changing the title to: Soil water stable isotope reveal evaporation dynamics at the soil-plant-atmosphere interface of the critical zone.

Abstract: Page1, L13: Because this paper is not a test of the method, it is necessary to report it here.

Response: We would prefer to keep this information in here, since recent findings (see Orlowski et al. (2016)) showed that different methods can potentially result in different findings.

Page1, L22-25: I would argue that this sentence can be deleted.

Response: We would not agree with that as we aim to provide a wider relevance of the findings in that sentence.

Introduction:

Page1, L30: remove “well” from well understood so that it reads simply “insufficiently understood”

Response: Will be changed as suggested.

P2 L2: I think it is the age distribution of the water that is used in evapotranspiration that is meant here, and not the age of the flux.

Response: Yes, changed to “evaporating water”.

P2 L10: perhaps introduce the term isotopic fractionation here, not all readers will understand the relevance of this process.

Response: We were hoping that the lines 10 to 15 would introduce the term of isotopic fractionation. We will rephrase the sentence to: “However, evaporation leads to isotopic fractionation, where…”

P2 L32: This sentence needs to be restructured.

Response: We will change the sentence as follows: However, recent findings about kinetic fractionation in the water pools and tracks of an extended drainage network in a raised bog within the Bruntland Burn showed that evaporation can have a fractionating effect on the stable isotopes of peatland waters, despite the relatively low energy available (Sprenger et al. 2017) .”

Page 3: Research question 1: Instead of “critical zone”, I suggest soil profile.

Response: Will be changed as suggested.

Research question 2: This has already been done before. Better to restate your question/goal as to estimate evaporation of the site based on the water stable isotope values within the soil profile. This section might need to be removed if the hydrocalculator approach cannot be justified.

Response: We are not aware of a similar study that observes the evaporation fractionation in the field at this temporal resolution over a year for four sites. Therefore, we see a clear research gap in understanding the dynamics of soil evaporation. We hope that we better highlight this research gap with the following statement in the introduction: "The temporal variability of the isotopic fractionation in the field has not yet been studied. While Rothfuss et al. (2015) sampled the soil water isotopes in a soil column undergoing evaporation in the laboratory, field studies are usually limited to few sampling campaigns or short period (Twining et al., 2006; Gaj et al., 2016). Additionally, soil…". We will slightly change the research question as follows: "How can one infer soil evaporation dynamics from measured soil water isotopic fractionation?" As discussed above, we believe that the revised estimates of the fraction of evaporation losses are a better representation of the physical processes and should therefore remain as part of the manuscript.

Research question 3: This is very vague. What is meant by feedbacks? And, how do you expect them to vary spatially and temporally?

Response: We will change the research question to be more specific as follows: "How do soil characteristics, vegetation cover and aspect drive evaporation fractionation dynamics?

P3 L20: catchment "is" covered

Response: Will be changed as suggested.

Page 5, lines 21-25: Please briefly explain how the 'equilibrium method' works.

Response: We will include more details on the applied method as suggested.

P5 L26: Please explain how the standard water was "sampled"?

Response: In the newly included information on the applied method, we will also add the info that the standard waters were sampled the same way as the soil samples. 10ml of standard water was added into an airtight bag and then heat sealed and allowed to equilibrate for 2 days. The analysis was done the same way as for the soil samples.

P7 L3: Please discuss the "effect of antecedent conditions". What might we expect that occurs during the time window you suggest?

Response: We will clarify that we decided to investigate the antecedent condition (in terms of precipitation input averages over 7 and 30 days and PET over 30 days) as follows in the revised manuscript: "To understand the potential atmospheric drivers for the soil water isotopic composition, we investigated the effect of antecedent conditions. We calculated average values of PET over 30 days prior to each sampling campaign (PET_{30}) to account for the potential soil evaporation dynamics. Additionally, the precipitation sums and the amount weighted isotopic signal of the daily precipitation isotope samples were computed for the 7 days and 30 days period prior to the sampling (P_7 and P_{30}, respectively) to assess the mixing processes. Weekly and monthly averages were chosen to see if the relatively young water input or the average over the last month better relate to the observed soil water isotopic signal."

Page 7, Lines 4-6: Could you briefly explain why exactly you used these number of days (7 and 30)? Furthermore, how were the isotopic signals weighted? This whole approach is rather unclear.

Response: Please see response above.

Page 7, Line 13: Did you mix the isotopic results from 5 and 10 cm for achieving f?

Response: Thanks, this is a good point. We have now weighted the 0-5 cm and 5-10 cm samples by their gravimetric water content.

Page 7, Lines 15-17: I have some concerns about this approach. What is the purpose of applying this correction to δS? This is not fully clear. Secondly, did you correct the δP for the net precipitation (mm), which was different between from month to month? This would affect the average δ18O and δ2H of precipitation.

Response: Please see response above. This section will be changed according to the above outlined revised approach to estimate the fraction of evaporation losses.

P8 L16-17: What is the reason for testing for significance along such a small range? Isn't 0 and 1‰ already in the measurement precision limits? What value is relevant to determine significant evaporation?

Response: We will add in section 2.5 the following to account for the measurement precision of lc-excess: "The accuracy for the liquid water isotope analysis and the soil water isotope analysis result in a precision limit for lc-excess of about 1.1 ‰ and 3.4 ‰, respectively.

Given that the Figure 8 is difficult to understand, while the differences between few permill are not that relevant for the evaporation dynamics considering the measurement limits, we will take this graph out. In the text, we will refer to if lc-excess is significant different from +-3.4 ‰.

P8 L21: Pearson not Parson

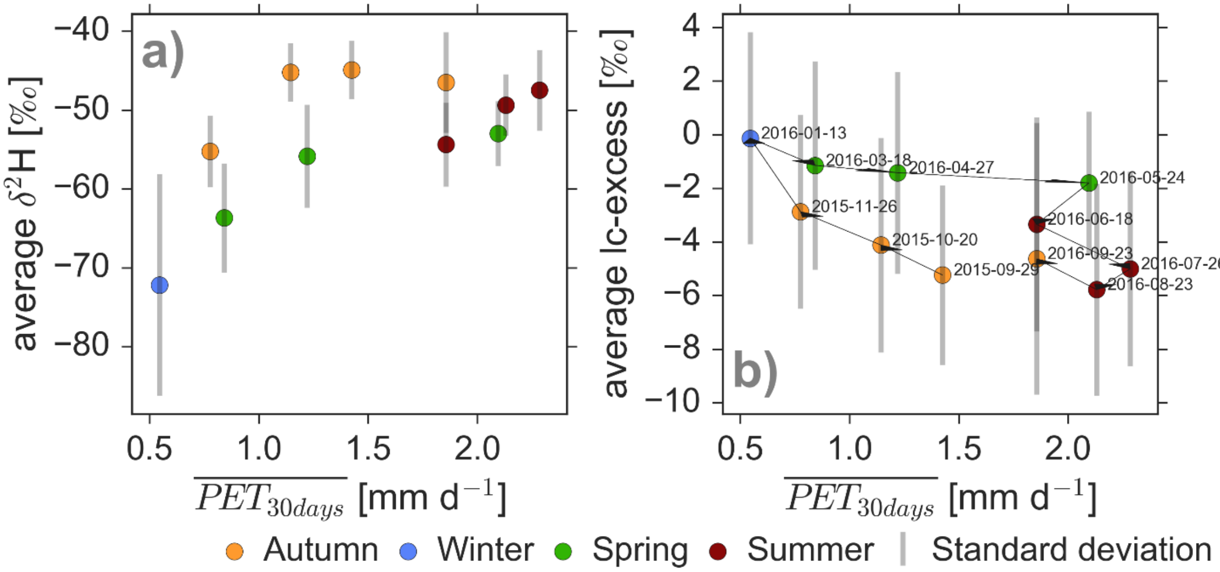
Response: Will be changed as suggested.

Figure 3. I really like this figure, especially with the temporal patterns along the secondary plots.

Response: Thanks, this way of presenting additionally boxplots to the dual isotope plot was inspired by Hervé-Fernández et al. (2016).

Figure 5 b. The pattern in lc-excess over the season is interesting but what qualifies this as hysteresis and not just seasonal changes in PET? This plot needs error bars along both axes.

Response: We do not see how the variability of PET as shown with error bars along the x-axis is relevant for the interpretation of the data. Error bars representing the standard deviation of PET 30 days prior to sampling would surrogate that the PET is relatively variable. But instead, we chose the 30 day average to represent the long term variability of the PET and not short term variability. Therefore, we do not see how error bars of the standard deviation of PET over 30 days prior to the sampling would improve the Figure. We show here the Figure with error bars representing the standard deviation of lc-excess within the data set for each sampling day. The information gained is relatively small, since the standard deviation does not vary in time. We would therefore prefer to keep the original figure. The high variability within the depth profiles is shown in Figure 9.

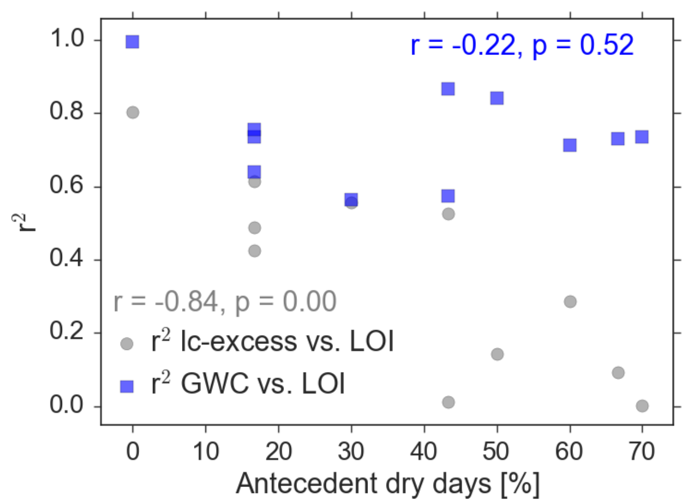


Page 14, Lines 22-23: What is ‘organic content’? Maybe “organic matter content” is more appropriate.

Response: We will change as suggested for the entire manuscript.

Figure 7. Is there a reason why LOI should change in such a short time period? The x label of this figure is really confusing.

Response: In Figure 7, we assume that LOI stays relatively constant over time. The correlation between LOI and GWC or lc-excess is only due to changes in GWC or lc-excess. We hope that the x-axis is better understandable as “Antecedent dry days [%]” and the following figure caption: “Relationship between coefficient of determination of the relationship between lc-excess and LOI (grey) and GWC and LOI (blue) with the percentage of dry days during the 30 day period prior to the soil sampling.”



P16 L74: I think this is in reference to 0‰ lc-excess

Response: That is correct and we will include the missing information as follows: "The soil water lc-excess at 15 – 20 cm was usually not…"

Page 17 L85: Should it be "depths" instead of "sites"?

Response: Thanks, this should be depths and will be changed accordingly.

Discussion:

P21 L159: Be careful, technically even precipitation has undergone fractionation!

Response: We will rephrase this sentence for clarification as follows: "With increasing new precipitation input (lc-excess close to zero), the sites…"

P21 L170: I assume the dynamics at 5cm are being described here. Is the replacement of autumn water the only possibility here? Is it not also possible that mixing (which the authors advocate elsewhere) is responsible? Are they mutually exclusive?

Response: Mixing and replacement is not mutually exclusive, since both will happen in parallel, but it is difficult to assess which process is dominating. We changed the sentence to: "However, the significantly lower δ^2H values in the top 5 cm indicate that, despite the potential occurrence of preferential flow, most of the depleted precipitation input was stored in the very top soil and the more enriched soil water from autumn was partly mixed with and partly replaced by the event water."

P21 L171: "further special" is a bit awkward

Response: We will replace "further" with "also".

P22 L172-174: Time reference is lost here. Is the rest of the year for any time that does not occur in January? It looks like this pattern was also emerging in November of 2015 (probably also established in December).

Response: Yes, we refer here to the vegetation period and replace therefore "rest of the year" with "vegetation period".

P22 L190-197: I don't think this part of the discussion warrants a whole paragraph.

Response: We believe that this paragraph is highly relevant, given the recent call for higher spatial soil sampling to address the Two Water World Hypothesis (Berry et al., 2017). We therefore rephrased the paragraph as follows and hope that the Referee agrees that this paragraph is relevant: "In contrast to Geris et al. (2015), who sampled the soil-vegetation units with duplicates, our sampling design with five replicates allowed for a clearer assessment of the spatial heterogeneity of the subsurface. For example, the standard deviation of the SW δ^2H and $\delta^{18}O$ values for the 5 cm depth increments at each site was - for the entirely sampled upper 20 cm of soil - always higher than the measurement accuracy of 1.13 ‰ and 0.31 ‰, respectively. At Bruntland Burn – as in most Northern temperate and boreal biomes (Jackson et al., 1996)- topsoils contain almost all the root biomass. For the interpretation of potential sources of root water uptake this means that the uncertainty of the potential water source signal due to the heterogenous isotopic composition at particular depths within the rooting zone is higher than the error due to the measurements. Our field measurements underline, therefore, the need for an improved spatial resolution of soil water sampling when studying root water uptake patterns with stable isotopes, as recently called *for by Berry et al. (2017). Further, this high variability will potentially impact the application of soil water isotopes for the calibration of soil physical models and the resulting interpretation (Sprenger et al., 2015b).* *"*

Page 22, Lines 192-195: This statement is not fully clear. What is the importance of the accuracy? Please remember that you are only talking about the top 5 cm. So root water uptake will depend on the plant species and rooting depth.

Response: Please see above comment and changes of the paragraph.

P23 L217-218: "kinetic fractionation dynamics" is a little cumbersome; it may be easier to the reader if you simply refer to evaporation when referencing the isotopic effect.

Response: We will replace "kinetic fractionation" by "soil evaporation".

P24 L246: citation

Response: We combined the two sentences so that (Sprenger et al., 2015a) is the reference for the mobility of water sampled with the two different methods.

P24 L255: Do you mean isotopically depleted infiltration water? I don't immediately see how this study shows that the "the legacy of evaporation losses" allows for separating pools of different water mobility in the SPA interface. Can you make this more apparent?

Response: We added: "This means that old (more tightly bound) water might not only have a distinct $\delta^{2}H$ or $\delta^{18}O$ signal compared to mobile water due to seasonally variable precipitation inputs, but also evaporative enrichment signal from periods of high soil evaporation. In conclusion, when relating isotope values of xylem water to soil water to study root water uptake patterns, an evaporation signal (that is lc-excess of xylem water < 0) would not be paradoxical, but simply represent the range of available soil water in the subsurface."

P25 L279: Where is the number 3 coming from? I think a few citations or reviews might help back this up. I think it is also important to keep the context of the research question in mind when assessing another study's design.

Response: We agree that the research question should be considered. We are referring here to studies that investigate the root water uptake patterns and do not cover the temporal variability of soil water isotope dynamics. We will also refer now to the Berry et al. (2017) who called for a higher temporal resolution of soil water isotope sampling when investigating root water uptake pattern with stable isotopes. "In line with the call for a higher temporal resolution of soil water isotope sampling (Berry et al., 2017), the highly dynamic isotopic signal during the transition between the dormant and growing seasons underlines the importance of not limiting the soil water isotope sampling to a few sampling campaigns (usually $n \leq 3$ in Brooks et al. (2010), Evaristo et al. (2016), Goldsmith et al. (2012)), when investigating root water uptake patterns."

P25 L288: Isn't the storage capacity referred to earlier relevant here as well?

Response: Yes, we meant to refer to the storage capacity differences here and clarify this by changing the sentence to: "However, these differences in water storage capacity seemed to only influence the *evaporation signal during periods of high precipitation input, when evaporation is already likely to be low (Figure 7).”*

P25 L302-303: citation

Response: We will add (Allison et al., 1983; Barnes and Allison, 1988) as follows: “The fractionation signal was shown to be more pronounced for evaporation losses from drier soils compared to wetter soils (Allison et al., 1983; Barnes and Allison, 1988).”

P26 L345: I don’t think the word “exceptionally” is warranted here, although the study is data rich. This sentence is also structured in a strange manner (i.e., “but also” when there isn’t a contrast to begin with). I don’t think the final statement is justified. Are modelers calling for higher spatial and temporal isotope data? Is this listed as a research priority in the literature? Do the data presented here help realize a realistic representation of “soil-vegetation” interactions? Statements like these are beyond the scope of this study.

Response: We will split this into two sentences and will rephrase them for more clarity. We further will include Vereecken (2016) and McDonnell and Beven (2014) as reference, who are calling for soil water isotope data in high temporal and spatial resolution to calibrate soil physical models and isotope tracer data from within the catchments to foster process understanding, respectively. “The presented soil water isotope data covering the seasonal dynamics at high spatial resolution (5 cm increments at four locations) will allow us to test efficiencies of soil physical models in simulating the water flow and transport in the critical zone (as called for by Vereecken (2016)). The data can further provide a basis to benchmark hydrological models for a more realistic representation of the celerities and velocities when simulating water fluxes and their ages within catchments (McDonnell and Beven (2014)).

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