

## Editor's comments to the Author:

Dear Dr Sprenger and co-authors,

Based on two reviews of your paper, I recommend that it may be suitable for publication after major revisions. Please consider the reviewers comments carefully in your revision. In particular, it is recommended that the paper is significantly shortened, including a reduction in the number of figures, and the title is changed to be more accurate.

Kind Regards,

Hilary McMillan

## Response to the Editor

*Dear Hilary McMillan,*

*Thank you for handling our manuscript and considering the study for publication in HESS after major revision. We addressed all the referee's comments by a reply to each comment and changed the original manuscript accordingly. Please see the replies to the referee's comments below and the track changed manuscript for the major revisions undertaken in the manuscript.*

*As recommended by both referees, we changed the manuscript title to: "Soil water stable isotopes reveal evaporation dynamics at the soil-plant-atmosphere interface of the critical zone", which clarifies that soil water isotopes have been measured, from which we infer evaporation dynamics at the soil-plant-atmosphere interface. We believe that the term "soil-plant-atmosphere interface" is appropriate in the context of our study, since we cover with our sampling design the major rooting depth and thus the part of the critical zone, where the interlinkages between the soil, vegetation and atmosphere takes place.*

*As recommended by Referee #1, we revisited the evaporation estimates using the Craig-Gordon model. We now apply the Craig-Gordon approach adjusted for evaporation from soils using  $n=1$  for the exponent of the diffusion coefficient ratio (see Eq. 1 in Horita et al. (2008) or Eq. 6b in Gat (1996)), 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). 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  $\delta_p$  in Eq. 3 of our manuscript and the measured soil water isotopes represent  $\delta_s$ . We also included the suggestion of Referee #1 to weight the isotopic composition according to the soil moisture and applied this in the revised calculations.*

*Another major change is the shortening of the manuscript, where we reduced the number of figures by 2 and revised the text with special focus on repetitive paragraphs to shorten the volume.*

*We hope that the revised version meets the Editor's and Referee's concerns and that the study will therefore still be considered for publication in HESS.*

## 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 analyzed the  $\delta^{18}\text{O}$  and  $\delta\text{D}$  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  $\delta_p$  in Eq. 3 of our manuscript and the measured soil water isotopes represent  $\delta_s$ . 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  $\delta^2\text{H}$  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 added 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. Furthermore, we did not include Figure 4 in the revised version to shorten 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 changed 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: 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 rephrased the sentence to: "However, evaporation leads to isotopic fractionation, where..."*

P2 L32: This sentence needs to be restructured.

*Response: We changed 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: 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 changed 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 changed 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: Changed as suggested.*

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

*Response: We included 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 also added the info that the standard waters were sampled the same way as the soil samples. 10 ml 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 clarified 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  $\delta S$ ? This is not fully clear. Secondly, did you correct the  $\delta P$  for the net precipitation (mm), which was different between from month to month? This would affect the average  $\delta^{18}O$  and  $\delta^2H$  of precipitation.

*Response: Please see response above. This section was 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 added in section 2.5 the following to account for the measurement precision of  $I_c$ -excess: "The accuracy for the liquid water isotope analysis and the soil water isotope analysis result in a precision limit for  $I_c$ -excess of about 1.1 ‰ and 3.4 ‰, respectively.*

P8 L21: Pearson not Parson

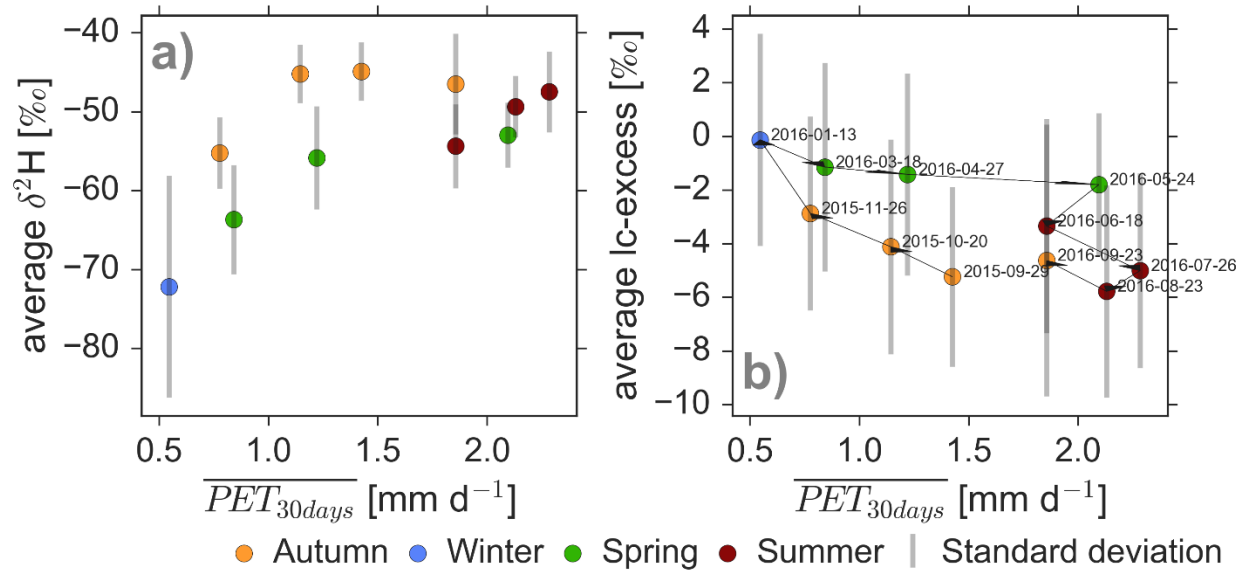
*Response: Changed as suggested.*

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  $I_c$ -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  $I_c$ -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 7 of the revised manuscript.*

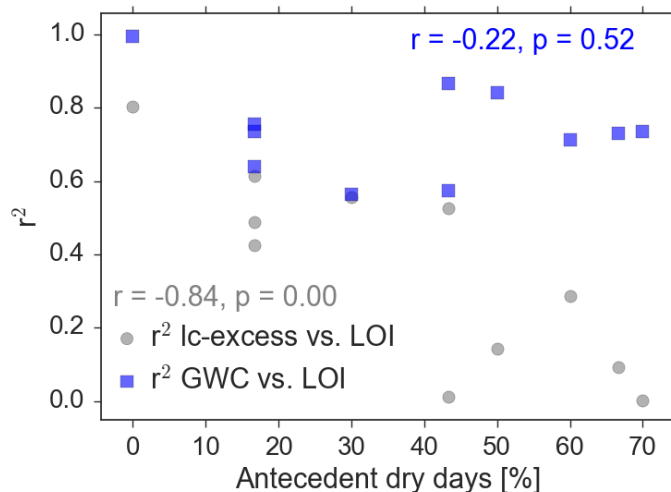


Page 14, Lines 22-23: What is 'organic content'? Maybe "organic matter content" is more appropriate.

*Response: 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 included the missing information as follows: “The soil water  $\delta^2\text{H}$ -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 was changed accordingly.

Discussion:

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

Response: We rephrased this sentence for clarification as follows: “*With increasing new precipitation input ( $\delta^2\text{H}$ -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  $\delta^2\text{H}$  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 replaced “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 replaced 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  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  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 heterogeneous 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 replaced “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  $\delta^2\text{H}$  or  $\delta^{18}\text{O}$  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  $\text{Ic-excess of xylem water} < 0$ ) 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 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 clarified 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 6)."*

P25 L302-303: citation

*Response: We added references 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 split this into two sentences and will rephrase them for more clarity. We further included 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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## Response to comments by Anonymous Referee #2

*We thank Referee #2 for reviewing our paper, the positive feedback on the visualization of the presented data and that the study will be a good addition to the soil water isotope literature*

*We will first comment on each of the major issues raised by Referee #2 and further respond to each comment by Referee #2 below.*

### General comments

The authors studied the influence of vegetation on water fluxes in the upper soil compartment of the Scottish Highlands by means of stable water isotopes. Soil samples were taken eleven times over the course of a year and analyzed for their isotopic composition using the direct equilibration method. The authors nicely visualized their results. However, they should consider cutting down the number of figures.

I think the paper length should also be reduced by at least five pages, which would help focus on the most important points.

*Response: We revised the manuscript with a special focus on repetition of results and discussion to shorten the manuscript. We took out the Figure 4 and Figure 8 of the original manuscript, since also Referee #1 was commenting on the difficulty to understand it. However, Referee #1 asked for a more detailed description of the direct-equilibration method, which will add a few paragraphs to the manuscript.*

There are many repetitions, which unnecessarily blow up the manuscript. What did the authors really expect to find?

*Response: We refer to the literature reviews by Evaristo et al. (2015) and Sprenger et al. (2016) who showed that northern environments have not yet been widely studied regarding their soil water isotope dynamics. Especially the intensity of evaporation fractionation was the main question, since surface waters in an adjacent peatland drainage network showed kinetic a fractionation signal (Sprenger et al. 2017b), but soil water with suction lysimeters in the same experimental catchment showed only limited fractionation (Geris et al. 2015). We therefore believe that the presented results are not just of limited interest to the particular study site, but are more widely and generally relevant for stable isotope dynamics in soils of humid northern environments that have not yet been well studied.*

I think there could also be a more compelling title that illustrates immediately to the potential reader what exactly the paper is about. The title should focus more on the actual findings as the manuscript does not present atmospheric data or detailed data on vegetation (e.g. rooting depth and density) anyway.

*Response: We changed the title to “Soil water stable isotope reveal evaporation dynamics at the soil-plant-atmosphere interface of the critical zone” in order to clarify what compartment was measured. However, we present rooting depth for the heather and present and discuss the atmospheric driver of the isotope dynamics of the soil water.*

The authors pose three research questions. In my opinion, these questions could be more precise. In particular, the third research question cannot really be answered by the results – especially not the atmospheric component.

*Response: We changed the research questions as follows to be more specific:*

*How do precipitation input and the soil water storage mix and affect the soil water isotope dynamics over time?*

*How can one infer soil evaporation dynamics in the field from soil water isotopic fractionation?*

*How do soil characteristics, vegetation cover and aspect drive evaporation fractionation dynamics?*

With regard to the soil samplings, I would not consider the sampling strategy as high frequent, especially against the background of portable laser spectroscopes which can indeed measure water isotopic composition in-situ with high frequency.

*Response: Despite the new possibilities that come with in-situ measurements of pore water stable isotopes, only the recently published study by Oerter, 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 short term investigations covering only a few days (Volkman*

*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, capturing seasonal variability at a number of dominant landscape units. We included the discussion above in the introduction of the revised manuscript.*

The authors describe the soil texture of the upper 20 cm as mainly loamy sand. A table, which compiles all soil properties, would be helpful at this point. Soil properties have been shown to affect the extraction method's isotope results. Do the authors have data on the soil mineralogy (clay mineral composition)? The applied direct equilibration has several downsides: It is less precise for more clayey soils and soils with low water content; storage time is also an issue as it can lead to evaporative water loss through the bag (How long were the bags stored prior to analysis in the present study?) Furthermore, soil organic matter content has been proven to have an effect on gained isotope results. The authors should consider these aspects when discussing their data.

*Response: Please see Table 1 for a detailed list of the soil properties. You see that the clay content was generally very low for the studied soils. As referred to in the manuscript, we have tested our method for soil water isotope analysis for its sensitivity of CO<sub>2</sub> emissions during the equilibration period of 2 days. The results have been published elsewhere (Sprenger et al. 2017a). This is why we do not pick up this issue in the discussion. We included in the methods section comments that the conditions at the study site (low clay content and generally high volumetric water content) are in favor of the applied direct-equilibration method. We will further mention that the analysis was done within one week after the sampling and that the evaporation losses through the bag can be neglected. A test between different bags (as reported by Sprenger et al. (2015)) showed that the used bag (Weber packaging) loosed less than 0.15% of its stored water over 30 days (details [www.hydro.uni-freiburg.de/publ/pubpics/post229](http://www.hydro.uni-freiburg.de/publ/pubpics/post229)).*

In sum, I think this paper will make a good addition to the soil water isotope literature, although it does not contain much novel or surprising findings. However, the authors did a great job in data analyses and presentation.

*Response: Thanks for the positive feedback, however we strongly disagree that the study lacks novelty. We have not found a study that showed the soil water isotope fractionation in the field over an entire year at sites with contrasting land cover; this alone makes our study highly novel.*

Specific comments

P 1 L 13:  $\delta^{18}O$

*Response: Changed as suggested.*

P 3 L 1: Thus, . . .

*Response: Changed as suggested.*

P 4 L 14-29: Described in too much detail; consider compiling important soil data in a Table

*Response: We shortened it. Please also see the Table 1.*

P 5 L 3-8: Far too detailed

*Response: We shortened it.*

P 5 L 24: Not necessary to state model and serial number of the isotope analyzer

*Response: Was removed.*

P 6 L 7: Not necessary to reference the python module; please change throughout the manuscript

*Response: Changed as suggested.*

P 8 L 4 –P9 L3: This whole section is again too long. Please condense

*Response: We believe that details of the applied statistical analysis are of relevance for the reader.*

P 9 L 6: Different font used

*Response: Changed as suggested.*

P 11 Fig. 2: for a) I would suggest to plot the rainfall amount data inversely (top-down) and either change the scale of the axis or the size of the blue star so that they are not cut off; for b) consider including moving averages through the soil data (e.g., moving average for the top and subsoil); describe the color code of the soil data (light brown dots stand for. . .)

*Response: We changed the bar plot in a way that the y-axis is inversely and adjust the axis scale to prevent cut off. We will not consider including moving averages, since the physical meaning of such a moving average is questionable, given that we do not know the soil water isotopic composition of up to 30 days between two sampling campaigns.*

P 13 Fig. 4: This figure does not add much information; consider deleting this figure. Does the average precipitation input signal represent a 1-yr mean?

*Response: We removed this figure.*

P 16 chp. 3.2.2.: Include this section in results section 3.1 as it does not add too much new information

*Response: We prefer to keep the structure of the results section with a focus on temporal (3.1) and spatial (3.2) variability. The differences in depth would therefore be part of section 3.2. We removed a paragraphs in 3.1, which dealt with the former Figure 4 and 8. Thus, there is no repetition anymore on the variability over the soil depth.*

P 20 chp. 3.2.4 Delete this section. There are no sig. differences in isotopic signatures when considering the aspect.

*Response: We believe that also a negative result is worth mentioning.*

P 20 L n134 ff: Repetition; consider deleting

*Response: We rephrased this section according to the new evaporation loss estimates.*

P 21 L 167-68: bypass flow, really; not so much differences over depth here

*Response: We agree that it is difficult to infer from hydrometric data (here GWC) to mixing and removed that sentence.*

P 22 L 175 : Is throughfall data available for these sites to underline this statement?



*Response: Yes, we now refer to the throughfall study here as follows: “The higher variability of the SW  $\delta^2H$  values beneath Scots pine compared to the SW beneath heather (Figure 9) cannot be explained by differences of the throughfall isotopic signal, since they are minor for the two vegetation types (Braun 2015). The higher variability in the isotopic signal therefore indicates that flow paths are generally more variable in the forest soils.”*

P 24 L 260: Add Gaj et al. (2017b)

*Response: Gaj et al. (2017) was added.*

P 24 L 265: This is not a new finding and not really surprising.

*Response: If this is not surprising, why is it ignored in studies dealing with root water uptake pattern with the means of stable water isotopes?*

P 24 L 269: The authors compare their study with results by Geris et al. (2015a) quite frequently. Is the vegetation cover comparable in both studies?

*Response: Yes, here, we refer to their samples taken at site NF, but we state that in the sentence.*

P 24 L 277: Debatable that the authors state to see highly dynamic isotope signals.

*Response: We refer to Fig. 3 which shows that.*

P 25 L 290: What exactly is the angle in your case?

*Response: We added in brackets (4°).*

P 25 L 293: grammar: . . .are mainly due to . . .

*Response: Was changed as suggested.*

P 26 L 318: In my opinion, the present study does not really unravel interactions occurring in the soil-plant-atmosphere continuum but adds to process understanding of water fluxes through the soil compartment.

*Response: Here we simply state that the presented soil water isotope data can be used to improve hydrological models.*

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# Soil water stable isotopes reveal evaporation dynamics at the soil-plant-atmosphere interface of the critical zone

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## Abstract

Understanding the influence of vegetation on water storage and flux in the upper soil is crucial in assessing the consequences of climate and land use change. We sampled the upper 20 cm of podzolic soils at 5 cm intervals in four sites differing in their vegetation (Scots Pine (*Pinus sylvestris*) and heather (*Calluna sp.* and *Erica Sp.*)) and aspect. The sites were located within the Bruntland Burn long-term experimental catchment in the Scottish Highlands; a low energy, wet environment. Sampling took place on 11 occasions between September 2015 and September 2016 to capture seasonal variability in isotope dynamics. The pore waters of soil samples were analysed for their isotopic composition ( $\delta^2\text{H}$  and  $\delta^{18}\text{HO}$ ) with the direct equilibration method. Our results show that the soil waters in the top soil are, despite the low potential evaporation rates in such northern latitudes, kinetically fractionated compared to the precipitation input throughout the year. This fractionation signal decreases within the upper 15 cm resulting in the top 5 cm being isotopically differentiated to the soil at 15 – 20 cm soil depth. There are significant differences in the fractionation signal between soils beneath heather and soils beneath Scots pine, with the latter being more pronounced. But again, this difference diminishes within the upper 15 cm of soil. The enrichment in heavy isotopes in the topsoil follows a seasonal hysteresis pattern, indicating a lag time between the fractionation signal in the soil and the increase/decrease of soil evaporation in spring/autumn. Based on the kinetic enrichment of the soil water isotopes, we estimated the soil evaporation losses to be about 5 and 10 % of the infiltrating water for soils beneath heather and Scots pine, respectively. The high sampling frequency in time (monthly) and depth (5 cm intervals) revealed high temporal and spatial variability of the isotopic composition of soil waters, which can be critical, when using stable isotopes as tracers to assess plant water uptake patterns within the critical zone or applying them to calibrate tracer-aided hydrological models either at the plot to the catchment scale.

## 1. Introduction

Processes in the soil-plant-atmosphere continuum exert a major influence on water partitioning into evaporation, transpiration and recharge fluxes. Therefore, the outer part of the Earth's terrestrial surface, where the subsurface is closely coupled with the atmosphere and vegetation, is often referred to as the critical zone (Brooks et al., 2015). However, the dynamics of feedbacks between soils and vegetation remain insufficiently well-understood (Werner and Dubbert, 2016). Consequently, there is an increased interest in improving the conceptualization of the upper boundary of soils, as the important interface between soils-plant-atmosphere. For example, it has been shown that evapotranspiration dynamics can affect travel times of percolating water in the unsaturated zone (Sprenger et al.,

2016c; Heße et al., 2017) and catchment outflows (van der Velde et al., 2015; Rinaldo et al., 2011). Additionally, understanding the age distributions of evapotranspiration ~~on fluxes themselves~~ water has recently gained interest in the literature (Harman, 2015; Soulsby et al., 2016a; van Huijgevoort et al., 2016; Queloz et al., 2015). However, estimates of these evapotranspiration ages and catchment travel times require a sound understanding of the storage and mixing dynamics of the subsurface and surface water pools which form the sources of evapotranspiration within catchments. Disentangling these atmospheric losses into evaporation and transpiration is particularly challenging.

Stable isotopes of water ( $^2\text{H}$  and  $^{18}\text{O}$ ) are a powerful tool for the analysis of the partitioning of water (see review by Kool et al., 2014). They are often seen as ideal tracers, since they are part of the water molecule. During root water uptake, the isotopic composition of the remaining soil water is usually not altered (Wershaw et al., 1966; Dawson and Ehleringer, 1991). However, evaporation ~~leads to isotopic fractionation~~ is a fractionating process, where the remaining water is generally enriched in heavy isotopes (equilibrium fractionation). Additionally, in natural open systems with a humidity of  $< 100\%$ ,  $\delta^2\text{H}$  is more likely to be evaporated than  $\delta^{18}\text{O}$ , because of their different atomic weights, leading to kinetic non-equilibrium fractionation (Craig et al., 1963). Therefore, evaporation losses result in an isotopic signal in the residual water that is distinct from the original isotopic composition of the precipitation waters that were formed in isotopic equilibrium (Dansgaard, 1964).

Such enrichment from kinetic fractionation was found in soil water isotopes across various climatic regions; with more pronounced evaporative signals reaching deeper into the soils in arid and Mediterranean environments than in temperate regions (Sprenger et al., 2016b). ~~However, the temporal variability of the isotopic fractionation in the field has not yet been studied, despite recent technical developments that enabled easier analysis of stable isotopes in soil water (see review by Sprenger et al., 2015a). 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 few days~~ (Twining et al., 2006; Gaj et al., 2016; Volkmann et al., 2016). ~~Only recently, Oerter and Bowen (2017) applied in-situ soil water isotope measurements to cover almost one year, but their study was limited to one particular location. Comparison of the soil water stable isotope dynamics that occur within an entire year at a number of dominant landscape units are yet missing. So far~~ Additionally, soil water isotopes have been studied much less extensively in the colder regions of the northern latitudes (but see Tetzlaff et al., 2014; Geris et al., 2015b; Geris et al., 2015a). However, the knowledge of soil water isotopic composition is of fundamental importance when studying the root water uptake pattern of plants (Rothfuss and Javaux, 2017). While comparisons of the isotopic signal in soil waters with waters of plant tissues have been reported for decades (see review by Ehleringer and Dawson, 1992), ~~recent technical developments have enabled easier analysis of stable isotopes in soil water (see review by Sprenger et al., 2015a).~~ Recently posed research questions on which water of the soils pore system is used by plants (Brooks et al., 2010) are enhancing ecohydrological studies on soil – plant interactions in the critical zone ~~(Barbeta et al., 2015; Evaristo et al., 2016; Volkmann et al., 2016; McCutcheon et al., 2016; Hervé Fernández et al., 2016; Oerter and Bowen, 2017). 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).

A comparative study by Evaristo et al. (2015) showed that the isotopic composition of plant waters - just like soil waters in the upper horizons – are usually kinetically fractionated. However, Evaristo et al. (2015) did not cover the northern latitudes in their review, since there has been only ~~one~~ preliminary study, looking into the root water uptake of the vegetation in this low energy region (Geris et al., 2015a; Geris et al., 2017). Geris et al. (2015a) found relatively little to moderate fractionation in the soil waters at -10 cm soil depth and xylem waters in the Bruntland Burn catchment in Northern Scotland. 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 ~~despite the low energy environment,~~ evaporation can have a fractionating effect on the stable isotopes of peatland waters. despite the relatively low energy available (Sprenger et al., 2017b). While such isotopic fractionation of open waters in peatlands of the northern latitude were found by others (Carrer et al., 2016; Gibson et al., 2000; Isokangas et al., 2017), the potential fractionation dynamics of the water in the upper soil layer in these cold regions have not previously been studied.

Thus, there is a particular need to better understand such ecohydrological processes in the higher latitudes (Tetzlaff et al., 2013). These environments are known to be especially sensitive to future climate change projections, since relatively little warming could cause intense changes in the water balance when snowfall and snow melt dynamics (Mioduszewski et al., 2014) and vegetation phenology shift (Shen et al., 2014). The upper soil layers (top 30 cm), where about 90 % of the root mass is usually present in the Northern temperate and boreal biomes (Jackson et al., 1996), are of special interest to better understand how vegetation influences the partitioning of soil water into evaporation, transpiration and recharge. The marked seasonality in the northern environments and its impact on the evaporation signal in soil water stable isotopes are fertile areas for investigation.

Here, we address the following research questions in order to improve understanding of the evaporation dynamics at the soil-plant-atmosphere interface and their influences on the water storage and mixing in the critical zone:

How do precipitation input and the ~~critical zones~~soil water storage mix and affect the soil water isotope dynamics over time?

How can one ~~derive~~infer soil evaporation dynamics in the field from soil water isotopic fractionation?

How do soil characteristics, vegetation cover and aspect drive evaporation fractionation dynamics~~How does the soil-plant-atmosphere feedbacks vary in space, depending on the soil, vegetation, and aspect?~~

## 2. Methods and study sites

### 2.2 Environmental conditions

Our study was conducted in the Bruntland Burn (BB) experimental catchment (3.2 km<sup>2</sup>) in the Scottish Highlands; a sub-catchment of the Girnock burn, a long-term ecohydrological research site. A detailed description of the soil and vegetation characteristics follows in section 2.3. The underlying geology of the BB is characterized by granitic and metamorphic rocks (Soulsby et al., 2007). About 60 % of the catchment ~~are-is~~ covered by up to 40 m of glacial drift deposits which maintain a high groundwater storage (Soulsby et al., 2016b). The climate is temperate/boreal oceanic with mean daily air temperatures ranging between 2°C in January and 13°C in July. Annual precipitation is about 1000 mm yr<sup>-1</sup>, which is fairly evenly distributed and occurs mainly as rainfall (usually < 5 % as snow) of low intensities (50 % of rainfall at intensities of < 10 mm d<sup>-1</sup>) (Soulsby et al., 2015). The annual potential evaporation (PET) is about 400 mm yr<sup>-1</sup> and the annual runoff is around 700 mm year<sup>-1</sup>. While the runoff of the BB shows limited seasonality, though lower flows tend to be in summer, the PET estimated with the Penman-Monteith approach follows a strong seasonal dynamic with average PET rates of 0.3 to 0.7 mm d<sup>-1</sup> from November to February and 2.3 to 2.7 mm d<sup>-1</sup> from May to August (Sprenger et al., 2017b).

The seasonality of the climate is also reflected in the variability of the isotopic signal of the precipitation with depleted values being more common during winter (dropping frequently below – 80 ‰ δ<sup>2</sup>H from November to March) and more enriched values dominant in summer. The weighted averages for the precipitation isotope signal over 5 years (2011-2016) were P<sub>avg</sub> δ<sup>18</sup>O = -8.5 ‰ and P<sub>avg</sub> δ<sup>2</sup>H = -61 ‰. The regression (~~calculated via scipy.stats.linregress in Python~~) between δ<sup>18</sup>O and δ<sup>2</sup>H values of daily precipitation data sampled between June 2011 and September 2016 describes the local meteoric water line (LMWL):

$$\delta^2H = 7.6 \times \delta^{18}O + 4.7 \quad (1)$$

### 2.3 Study sites

Soil sampling focused on four different sites within the BB, where the soils are characterized as freely draining podzols (Figure 1a). The study sites differed ~~with regard to~~regarding their vegetation cover and their aspect: At two sites, Scots Pine (*Pinus sylvestris*) forest is the dominant vegetation and at the two other sites, heather (*Calluna sp.* and *Erica sp.*) shrubland is dominating (Figure 1b). Each of the two soil-vegetation landscape units were studied at a north facing and a south facing slope (sites had gentle slopes), leading to the following four different sites: North facing heather (NH), north facing forest (NF), south facing heather (SH), and south facing forest (SF).

The podzols are shallow soils and frequent large clasts within the glacial drift deposit usually inhibit soil sampling below 20 to 30 cm. The soils at the four study sites were relatively similar ~~with regard to~~regarding their colour (Figure 1c) and texture (Table 1)-(Figure 1e). The texture ~~of the upper 20 cm~~ was determined ~~for each site in 5 cm depth increments. After~~ ignition of the soil samples to free the soil of organic matter content, ~~the~~ The coarse and medium

sand fractions were determined by dry sieving, and the fine sand, silt, and clay fractions were estimated with the hydrometer method (Gee and J. W. Bauder, 1986). The ~~upper 20 cm soil~~ consist mainly of loamy sand and ~~only for SH, the top 10 cm indicated a higher contribution of the silt fraction leading to sandy loam (Table 1). For NH and SH, the gravel content as well as the sand content generally increases with depth (Table 1). The texture analysis proved infeasible for the upper 5 cm at SH, NF, and SF, since there was not enough soil material left after ignition of the organic material. Coarse gravel (> 20 mm diameter) were only present in the soil below 5 cm at SF, where also the highest fine gravel content was also present.~~ The bulk density of the podzols in the BB is about  $0.74 \text{ g cm}^{-3}$  for the top 20 cm (Geris et al., 2015b).

The organic ~~matter~~ content ~~in the upper 20 cm~~ of the soils was determined for each study site in replicated 5 cm depth increments ( $n = 5$  per site and depth) by loss on ignition (LOI) of about 10 g soil material at  $550^\circ\text{C}$  in a furnace over 2h according to Ball (1964). The LOI decreased linearly with soil depth at all study sites. ~~Pearson correlations between LOI and soil depth were strong and significant at the 99 % confidence with NH, SH, and SF and weaker for NF (Table 1). The LOI and also~~ showed generally a strong relationship with the gravimetric water content (~~generally  $r \geq 0.7$ ,  $p < 0.01$  at all four study sites (Table 1Table 1).~~ We determined the gravimetric water content (GWC) of all soil samples by relating the weight loss after oven drying at  $105^\circ\text{C}$  over night to the dry soil mass.

Hemispheric photos taken during the vegetation period revealed that the median of the canopy coverage (CC) of the heather was slightly lower (NH: CC = 65 %,  $n = 9$ ; SH: CC = 62 %,  $n = 9$ ) than for the forested sites (NF: CC = 67 %,  $n = 36$ ; SF: CC = 69 %,  $n = 46$ ) (Braun, 2015). The forested sites were both plantations, but with larger trees (mean diameter at breast height (DBH) = 21.8 cm) and lower tree density at SF compared to NF (mean DBH = 13.8 cm), where the tree ages were more variable (Braun, 2015). The height of the heather vegetation was limited to about 0.5 m and tree height was 12 – 15 m in the plantations.

Fine root (defined as 0.5 – 2 mm according to Zobel and Waisel (2010)) density was determined by wet sieving of the fine roots from soil cores ( $100 \text{ cm}^3$ ) ~~taken from the upper 20 cm in 5 cm increments. Afterwards, the roots were dried and subsequent oven drying at  $70^\circ\text{C}$  overnight. The root density was then derived by relating the dried root mass at each depth to the total root mass in the profile.~~ This analysis was limited to the heather sites, since the roots and boulders at the forested sites inhibited sampling of undisturbed soil cores. The roots of heather were limited to the upper 15 cm at the heather sites with almost exponential decrease at NH and a more linear decrease at SH (Table 1Table 1).

## 2.4 Sampling design and analysis

Soil sampling at each site was conducted at monthly intervals between September 2015 and September 2016 (except for Dec., Feb., March;  $n = 11$ ). For each sampling campaign, soils at all four sites were sampled with a spade across five profiles in 5 cm increments down to 20 cm soil depth. For each sampling depth, five replicate samples were taken to account for the high subsurface heterogeneity (Figure S 1). Each soil sample contained of about 80 to 250 g of soil and was stored in air tight bags (Weber Packaging, Güglingen, Germany), ensuring - by manually furling the bags - that as little air as possible was inside them. The used bags ensured that there were no evaporation losses through bags.



since less than 0.15% of water was lost over 30 days in an experiment as reported by Sprenger et al. (2015a). For the sampling campaign in May, five additional sites with heather vegetation were sampled on the north (n=2) and south facing (n=3) slopes to increase the sample size for comparisons between the two slopes and get an idea of the general variability in space. In addition to the monthly sampling, two extra sampling campaigns for the two heather sites were conducted in August (4<sup>th</sup> and 9<sup>th</sup> August 2016) to investigate short time changes on both slopes and to further increase the sample number for comparison between the two slopes.

The soil samples were analysed for their stable isotopic composition ( $\delta^2\text{H}$  and  $\delta^{18}\text{O}$ ) according to the direct equilibration method suggested by Wassenaar et al. (2008). The analyses were conducted within one week after the sampling to prevent microbial activity within the bags. The analyses were done by adding dry air to the bags that contained the soil samples, heat sealing the bags and letting the soil water equilibrate with the dry atmosphere in the bag for two days at constant temperature in the laboratory. The same was done in parallel with bags each filled with 10 ml of one of three different standard waters covering the range of the soil water isotopic signals: seawater ( $\delta^{18}\text{O} = -0.85\text{‰}$  and  $\delta^2\text{H} = -5.1\text{‰}$ ), Aberdeen tap water ( $\delta^{18}\text{O} = -8.59\text{‰}$  and  $\delta^2\text{H} = -57.7$ ), condensate of distilled tap water ( $\delta^{18}\text{O} = -11.28\text{‰}$  and  $\delta^2\text{H} = -71.8$ ) and for the sampling in January Krycklan snow melt ( $\delta^{18}\text{O} = -15.36\text{‰}$  and  $\delta^2\text{H} = -114.4$ ). After the equilibration over 2 days, the vapour in the headspace of each bag was sampled directly with a needle connected to an off-axis Integrated Cavity Output Spectroscopy (OA-ICOS) (TWIA-45-EP, Los Gatos Research, Inc., San Jose, CA, USA). The  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  composition was continuously measured over 6 minutes, of which the last 2 minutes, where the water vapour pressure in the cavity was constant (standard deviation < 100 ppm), were used to calculate average values. The standard deviation for the  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  measurements were usually < 0.25 ‰ and < 0.55 ‰, respectively. The standards, which were treated the same way as the soil samples and measured at the beginning, the middle and the end of each sampling day, were then used for calibration to derive the isotopic composition of liquid soil waters from vapor measurements. For a detailed description of the soil water isotope analyses in the lab of the Northern Rivers Institute at the University of Aberdeen with the direct equilibration method using off-axis Integrated Cavity Output Spectroscopy (OA-ICOS) (triple water vapour isotope analyser TWIA-45-EP, Model#: 912-0032-0000, Serial#: 14-0038, Manufactured: 03/2014, Los Gatos Research, Inc., San Jose, CA, USA), we refer to Sprenger et al. (2017a). To assess the precision of the analysis, we derived the standard deviation of in total 81 measurements of the standard water (Aberdeen tap water) sampled along with the soil samples at the beginning, the middle and the end of each of the on 27 days of laboratory analyses over one year. The standard deviation of the standard water analysis was 0.31 ‰ for  $\delta^{18}\text{O}$  values and 1.13 ‰ for  $\delta^2\text{H}$  values. Recently reported potential effects of  $\text{CO}_2$  on the isotope analysis of vapour with wavelength-scanned cavity ring-down spectroscopy (Gralher et al., 2016) have been shown to not apply to the OA-ICOSO that we used, as shown by Sprenger et al. (2017a). Potentially fractionating effects of interactions between soil water and surfaces of clay minerals (Oerter et al., 2014; Gaj et al., 2017a; Gaj et al., 2017b; Newberry et al., 2017) are of minor relevance for our study, since clay contents were low in the sampled soils (Table 1). We can further ensure that the sampled soil volumes always contained much more than 3 g of water as suggested by Hendry et al. (2015).

In addition to the soil water analysis, precipitation at the field site was sampled with an auto sampler on daily basis at the catchment outlet (location shown in Figure 1a). The auto sampler was emptied at least every two weeks and evaporation from the sampling bottles was prevented by adding paraffin. The precipitation isotopic composition ( $\delta^2\text{H}$  and  $\delta^{18}\text{O}$ ) was determined with the above mentioned OA-ICOS running in liquid mode with a precision of 0.4 ‰ for  $\delta^2\text{H}$  values and 0.1 ‰ for  $\delta^{18}\text{O}$  values, as given by the manufacturer.

**Figure 1 Location of the four sampling sites within the Bruntland Burn catchment on (a) a soil map and (b) an aerial photo. The precipitation sampling location is indicated by a blue triangle in (a). (c) The four photos on the right show exemplary soil profiles for the four study sites.**

## 2.5 Data analysis

We calculated the evaporation line (EL) as a regression line through the soil water isotope data of each sampling date in the dual isotope space (~~scipy.stats.linregress in python~~). The EL is characterized by its slope and intercept with the  $\delta^2\text{H}$  axis. All regressions for EL presented here were significant at the 95 % confidence interval. For each soil water and precipitation sample, we further calculated the line conditioned excess (lc-excess) as a function of the slope ( $a = 7.6$ ) and the intercept ( $b = +4.7$  ‰) of the LMWL (Equation 1) as suggested by Landwehr and Coplen (2006):

$$lc - excess = \delta^2H - a \times \delta^{18}O - b \quad (2)$$

The lc-excess describes the deviation of the sample's  $\delta^2\text{H}$  value the LMWL in the dual isotope space (Landwehr et al., 2014), which indicates non-equilibrium kinetic fractionation processes due to evaporation after precipitation. Therefore, the lc-excess is similar to the well-established deuterium-excess (Dansgaard, 1964) that relates the deuterium composition to the global meteoric water line (GMWL). However, we found that lc-excess was advantageous over the deuterium-excess (or single isotope approaches with  $\delta^2\text{H}$  or  $\delta^{18}\text{O}$ ) for inferring evaporation

fractionation, because the lc-excess of the precipitation input is about 0 ‰ and with relatively little seasonal dynamics, while  $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$ , and d-excess can have an intense seasonal variability (Sprenger et al., 2017b). The accuracy for the liquid and soil water isotope analysis result in a precision limit for lc-excess of about 1.1 ‰ and 3.4 ‰, respectively.

To infer dynamics of potential evaporation rates, we estimated potential evapotranspiration (PET) with the Penman-Monteith-Equation adjusted for the Scottish Highlands by Dunn and Mackay (1995). Note that we focus in our study on the PET dynamics and that the absolute values could vary depending on the aerodynamic and roughness parameter of different vegetation covers. We further did not partition PET into evaporation and transpiration fluxes, since PET was primarily used as a proxy for potential soil evaporation rates, and evaporation and transpiration usually show a linear relationship in temperate regions (Renner et al., 2016; Schwärzel et al., 2009). 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 ~~were calculated~~ (PET<sub>30</sub>) 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 over 7 days and 30 days period prior to the sampling ~~was calculated~~ (P<sub>7</sub> and P<sub>30</sub>, respectively) to assess the mixing processes between precipitation input and soil water. Weekly and monthly averages were chosen to see if the relatively young water input or the average over the last month relate differently to the observed soil water isotopic signal.

We estimated ~~the evaporative water losses  $f$  and evaporation/input ratios ( $E/I$ ) based on the Craig-Gordon model (Craig and Gordon, 1965) with the *Hydrocalculator* provided by Skrzypek et al. (2015). For a detailed description of the calculations, we refer to Skrzypek et al. (2015) and will only briefly introduce the main characteristics of the approach and assumptions we used in our application. The estimate of the evaporative losses  $f$  [%] is based on the Craig-Gordon model (Craig and Gordon, 1965) and formulations introduced by Gonfiantini (1986) and adapted in the *Hydrocalculator* for isotope mass balance as follows:~~

$$f = 1 - \left[ \frac{(\delta_S - \delta^*)}{(\delta_P - \delta^*)} \right]^m \quad (3)$$

~~For our estimates of  $f$ , we defined  $\delta_S$  defined as the average weighted isotopic signal of the soil water in the upper 10 cm. The upper 10 cm were chosen, because this was the depth with the highest evaporation signal in the soil water isotopes (see results section) and where most evaporation are usually observed in laboratory experiments (Or et al., 2013), as shown in the results section. The sampled soil water isotope signal  $\delta_{SW}$  was corrected for the seasonality of the input signal by the average isotopic signal of P<sub>30</sub> as  $\delta_S = \delta_P + (\delta_{SW} - \delta_{P30})$ .  $\delta_P$  was defined as the isotopic signal of the original water source by calculating the intercept between the evaporation line of the soil water isotope data in the dual-isotope space (see Figure S 2) and the LMWL according to Javaux et al. (2016). in the long-term precipitation input (-61 ‰ for  $\delta^2\text{H}$  and -8.5 ‰ for  $\delta^{18}\text{O}$ ),  $\delta^*$  as is the limiting isotopic enrichment factor, and  $m$  as is the enrichment slope, both described by Gibson and Reid (2014) (Equation (8) and (9) therein).  $\delta^*$  is a function of the air humidity  $h$ , the isotopic composition of the ambient air  $\delta_A$ , and a total enrichment factor  $\varepsilon$  (Gat and Levy, 1978). For humidity, we averaged over 30 days prior to each soil water sampling date the measured humidity at a meteorological station less than 800 m away from the study sites  $h_{30}$ .  $\delta_A$  was derived as function from the weighted average precipitation~~

input of the 30 days prior to the soil sampling  $\delta_{p30}$  and the equilibrium isotope fractionation factor  $\varepsilon^e$  (Gibson et al., 2008), ~~which with the latter depends depending~~ on the temperature as given by Horita and Wesolowski (1994). We used air temperature data from the aforementioned meteorological station and computed values averaged over 30 days prior to soil water sampling  $T_{30}$ . The total enrichment factor  $\varepsilon$  is the sum of the equilibrium isotope fractionation factor  $\varepsilon^e$  and the kinetic isotope fractionation factor  $\varepsilon_k$ , which is a function of  $h_{30}$ , the exponent of the diffusion coefficient ratio  $n$  and a kinetic fractionation constant, which has a value of 28.4 ‰ for  $\delta^{18}\text{O}$  and 25.0 for  $\delta^2\text{H}$  (Gonfiantini, 1986). Here, we define  $n = 1$  in accordance to Barnes and Allison (1983), representing diffusional transport in soil pores. The enrichment slope  $m$  was calculated as a function from  $h_{30}$ ,  $\varepsilon$ , and  $\varepsilon_k$  in accordance to Welhan and Fritz (1977).

~~The ratio between evaporated water  $E$  [%] to the net precipitation input  $I$  can be calculated with the Hydrocalculator in accordance to Allison and Leaney (1982) as:~~

$$E/I = \frac{(\delta_s - \delta_p)}{[(\delta^* - \delta_s) \times m]} \quad (4)$$

~~Both  $E/I$  and  $f$  can be derived either by  $\delta^2\text{H}$  or  $\delta^{18}\text{O}$ , indicated as  $E/I_{d2H}$ ,  $f_{d2H}$ ,  $E/I_{d18O}$ , and  $f_{d18O}$ . Note that our approach is based on the assumption that 1.) the soil evaporation mainly happens from the soil water in the upper 10 cm (justified by the observations given in the results section), 2.) that isotopic enrichment of soil waters was defined as the enrichment compared to the long term isotopic signal of the precipitation input, and 3.) that the averages over 30 days prior to the soil water sampling for humidity ( $h_{30}$ ), air temperature ( $T_{30}$ ), and precipitation isotopes ( $\delta_{p30}$ ) represent the atmospheric drivers for the isotopic enrichment.~~ Note that  $\delta_{p30}$  represents in these calculations the net precipitation infiltrating the soil, which has the same isotopic composition as the measured rainfall isotope values, since no evidence for isotopic enrichment was found for the throughfall and stem flow in heather and Scots pine stands in the Bruntland Burn catchment (Braun, 2015).

Statistical analyses for the soil water isotopes ( $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$  and lc-excess) for individual sites, depths, and dates were done with non-parametric tests, since the null-hypothesis that the data was drawn from a normal distribution was rejected for several sampling campaigns using the Shapiro-Wilk test for normality (~~scipy.stats.shapiro.test in Python~~).

We also tested whether there were significant differences between the sites (each site with  $n > 200$ ) and sampling dates (each date with  $n > 75$ ) or at different depths (each depth with  $n > 200$ ) with the Kruskal-Wallis test (~~kruskal.test in R~~). When significant differences were present at the 95 % confidence interval, a post-hoc Dunn test (~~posthoc.kruskal.dunn.test in R~~) with p-value adjustment “bonferroni” was applied to see which of the sampling dates or sampling depths were significantly different. For pairwise tests (e.g., aspect (north- and south-facing) or vegetation (soils beneath heather and soils beneath Scots pine;  $n > 35$  for each vegetation type on each sample day and  $n > 100$  for each vegetation type at each sample depth), the nonparametric Mann-Whitney-Wilcoxon test was used (~~pairwise.wilcox.test in R~~). We assessed if the soil water lc-excess values for different sites were significantly lower than 0, ~~1, 2, 3, 4, 5, 6, and 7~~ ‰ by using the Wilcoxon signed-rank test (~~scipy.stats.wilcoxon in Python~~).

Mean values for each of the 11 sampling dates of  $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$ , and lc-excess of soil water and  $P_7$  or GWC,  $PET_{30}$ ,  $P_7$ , and the lc-excess of  $P_{30}$  were normally distributed according to the Shapiro-Wilk test. Therefore, we used the T-test for the mean of one group of samples (~~scipy.stats.ttest\_1samp in Python~~) to test if the average soil water lc-excess deviated significantly from zero for any sampling campaign. We further calculated the Pearson correlation coefficients (r) to describe linear relationships between mean values (~~scipy.stats.pearsonr in Python~~). Since  $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$  of  $P_{30}$  were not normally distributed, the Spearman rank correlation ( $\rho$ ) was applied to describe relationships with these two variables (~~scipy.stats.spearmanr in Python~~). For all statistical analyses, the 95 % confidence interval was defined as significance level ( $p < 0.05$ ). Visualizations with boxplots generally show the interquartile range (IQR) as boxes, the median as line within the box, the 1.5 IQR as whiskers and data points  $\geq 1.5$  IQR as points (~~matplotlib.boxplot in Python~~). We further make use of violin plots, where a kernel density estimation of the underlying distribution describing the data is visualized (~~seaborn.violinplot in Python~~). Statistical differences derived with the post-hoc Dunn test are visualized by either letters or coloured markers within the boxplots, where the same letter or marker color indicate that the samples are not significantly different to each other. Statistical differences between two groups derived with the Mann-Whitney-Wilcoxon test are indicated by either an asterisk or “X”.

### 3 Results

#### 3.1 Temporal dynamics in soil water isotopes

The temporal dynamic of the seasonally variable precipitation input signal between September 2015 and September 2016 was generally imprinted on the soil water (SW) isotope data at all sites.  $P_{30}$   $\delta^2\text{H}$  values usually were most similar to the lowest (most depleted) SW  $\delta^2\text{H}$  values (Figure 2b). An exception to this were the soil samples from January, because the sampling took place after a period of intense rainfall ( $P_{30} = 434$  mm) that occurred at the end of December and beginning of January (Figure 2a). The SW  $\delta^2\text{H}$  (and  $\delta^{18}\text{O}$ ) values averaged over all sites and depths for each sampling campaign correlated significantly with  $P_{30}$   $\delta^2\text{H}$  ( $\rho = 0.92$ ,  $p < 0.01$ ) and  $P_{30}$   $\delta^{18}\text{O}$  ( $\rho = 0.97$ ,  $p < 0.01$ ) and  $P_7$   $\delta^2\text{H}$  ( $r = 0.74$ ,  $p = 0.01$ ) and  $P_7$   $\delta^{18}\text{O}$  ( $r = 0.83$ ,  $p < 0.01$ ). However, the soil water averages were usually more enriched than the precipitation input (except for the January sampling). There is relatively little variability in the P lc-excess, which had a weighted average of  $-0.58$  ‰. The average SW lc-excess was usually more negative than the  $P_{30}$  lc-excess, but the sampling campaigns on 13 January and 18 March in 2016 were an exception (Figure 2c, Table 2). In contrast to  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$ , there was neither a relationship between the SW lc-excess with  $P_{30}$  lc-excess ( $r = 0.33$ ,  $p = 0.32$ ) nor with  $P_7$  lc-excess ( $r = 0.18$ ,  $p = 0.59$ ). Thus, the dynamics of SW lc-excess cannot be explained by variation of the lc-excess in the input.

The seasonally variable input of P  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  led to significantly more depleted SW isotope values during January and March compared to the other sampling days (indicated by the letter “a” at the boxplots in Figure 3). The sampling in November and April represented a transition period, where the SW isotopic composition was significantly different to the Winter and Summer samples. The SW  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values between May and August did not differ significantly. The soil water samples from January were the only ones that plotted along the LMWL with a slope of the EL of 8.2

(cyan dots in Figure 3). The other soil water samples followed ELs of slopes between 3.7 and 5.4 with the lowest slopes at end of summer and beginning of autumn (Table 2). The variability of  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values was highest for the sampling during winter and generally higher for  $\delta^{18}\text{O}$  compared to  $\delta^2\text{H}$  (note that the axes are scaled according to the GMWL in Figure 3).

~~The SW isotopes were usually more enriched towards the soil surface. Only the  $\delta^2\text{H}$  depth profiles for the November and January sampling showed more depleted values in the topsoil (Figure 4a). The lc-excess depth profiles revealed that the deviation from the LMWL decreased with soil depth for all sampling campaigns (Figure 4b). While the values varied over the year, with more positive lc-excess values in winter, the shape of the lc-excess profiles remained relatively persistent throughout the year. In addition, the GWC across the upper 20 cm of soil had a fairly persistent pattern over the year with generally highest values in the top 5 cm and decreasing values with soil depth (Figure 4c).~~

The enrichment in SW  $\delta^2\text{H}$  ~~in the upper 20 cm of the soil~~ during spring (green dots in ~~Figure 4~~Figure 5a) followed an increase in  $\text{PET}_{30}$ . Highest  $\text{PET}_{30}$  in summer correspond with enriched  $\delta^2\text{H}$  values. While  $\text{PET}_{30}$  decreased at the end of summer, the SW stayed enriched in  $^2\text{H}$  until late autumn (orange dots in ~~Figure 4~~Figure 5a). Thus, the SW  $\delta^2\text{H}$  (and also  $\delta^{18}\text{O}$ , not shown) to  $\text{PET}_{30}$  relationship can be described by a linear correlation ( $r = 0.65$ ,  $p = 0.03$  for  $\delta^2\text{H}$  and  $r = 0.64$ ,  $p = 0.03$  for  $\delta^{18}\text{O}$ ). However, ~~Figure 4~~Figure 5a shows that the delayed response in the SW isotopic signal to changes in  $\text{PET}_{30}$  resulted in a hysteresis pattern. The error bars in ~~Figure 4~~Figure 5a, representing standard deviations of all the samples for each sampling campaign, show again that the SW  $\delta^2\text{H}$  values were most variable for the sampling in January, becoming less variable during spring and lowest in autumn.

The hysteresis pattern was very pronounced for the relationship between SW lc-excess and  $\text{PET}_{30}$ , since the SW lc-excess only increased little with onset of  $\text{PET}_{30}$  in spring time (green dots in ~~Figure 4~~Figure 5b). During summer,  $\text{PET}_{30}$  remained high, and SW lc-excess values became lower and stayed low, even when  $\text{PET}_{30}$  started to decrease (~~Figure 4~~Figure 5b). However, the relationship between SW lc-excess and  $\text{PET}_{30}$  could statistically be also described by a linear relationship ( $r = -0.64$ ,  $p = 0.04$ ). ~~Note that since there was no relationship between SW lc-excess and  $\text{P}_{30}$  lc-excess, the SW lc-excess to  $\text{PET}_{30}$  relationship also showed a hysteresis pattern when applying the correction for seasonality using the difference between SW lc-excess and  $\text{P}_{30}$  lc-excess as described in the methods.~~ It is further worth noting that the average SW lc-excess in the upper 20 cm was for all sampling campaigns, apart from the January sampling, significantly  $< 0 \text{ ‰}$ , (T-test,  $p = 0.63$ ). When limiting the analysis to samples from the upper 5 cm, there was also a hysteresis pattern in the relationship between SW lc-excess and  $\text{PET}_{30}$ .

The average GWC for each sampling campaign correlated significantly with the average SW lc-excess for that sampling day, with lower SW lc-excess when soils were drier ( $r = 0.67$ ,  $p = 0.02$ ). The GWC also correlated with SW  $\delta^{18}\text{O}$  ( $r = -0.75$ ,  $p = 0.01$ ) and  $\delta^2\text{H}$  ( $r = -0.74$ ,  $p = 0.01$ ) with isotopically more enriched values when the soil was drier.

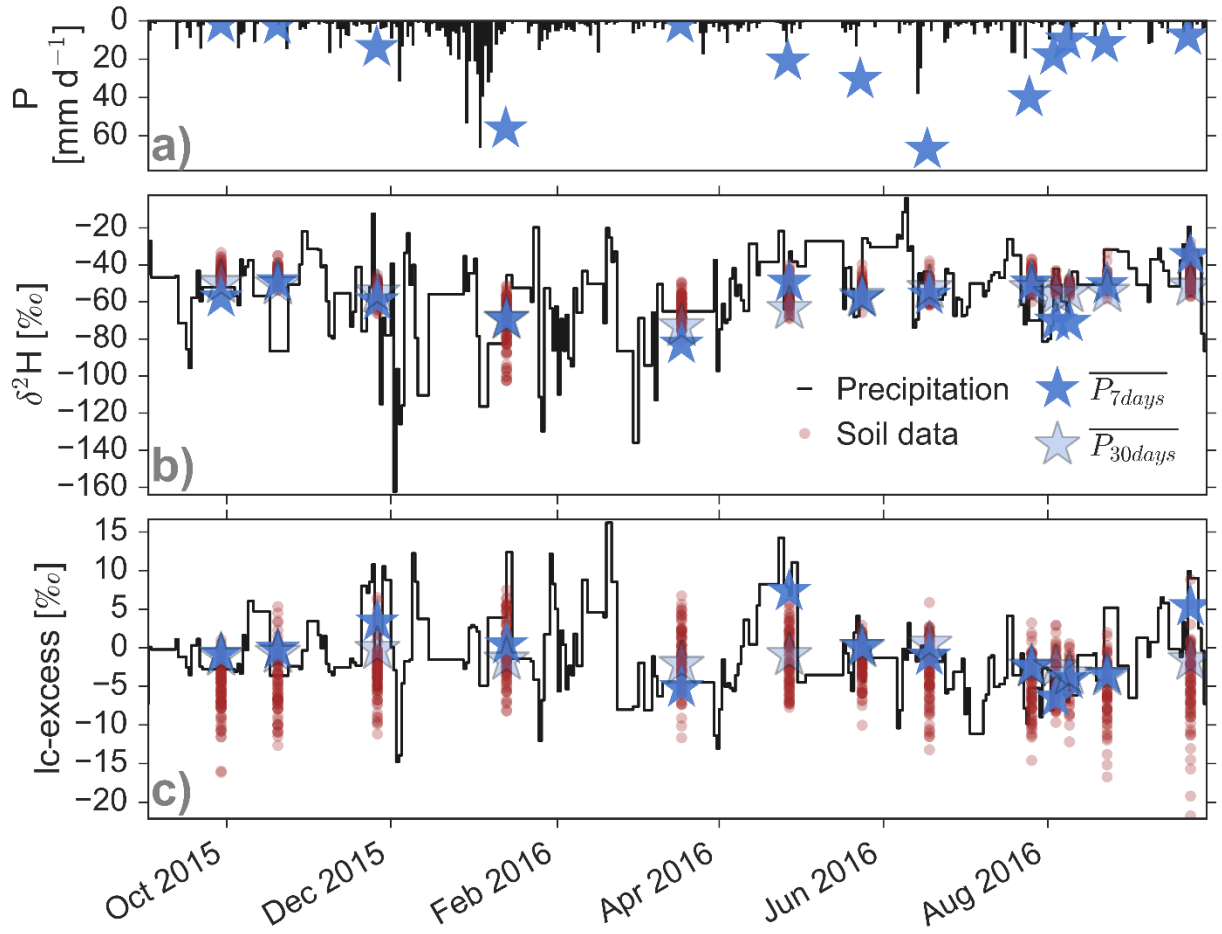


Figure 2 (a) Daily precipitation sums shown in a bar plot and precipitation sums over the 7 days prior to the soil sampling  $P_7$  as a blue star. (b) Dynamics of the  $\delta^2\text{H}$  and (c) lc-excess in the precipitation input (black line) and soil waters (half transparent brown dots). Stars indicate values as weighted averages over 7 days ( $P_7$ , blue) and 30 days ( $P_{30}$ , light blue) before the day of soil sampling.

5



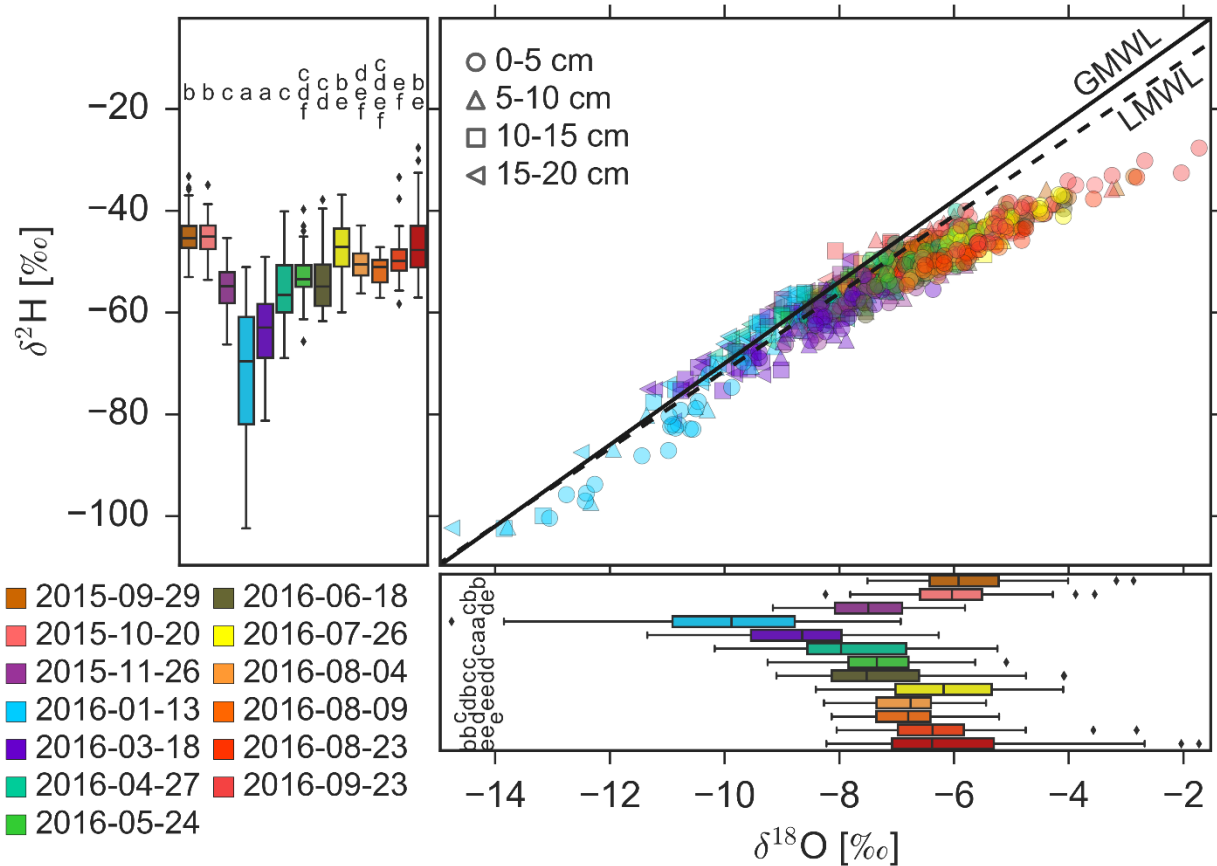
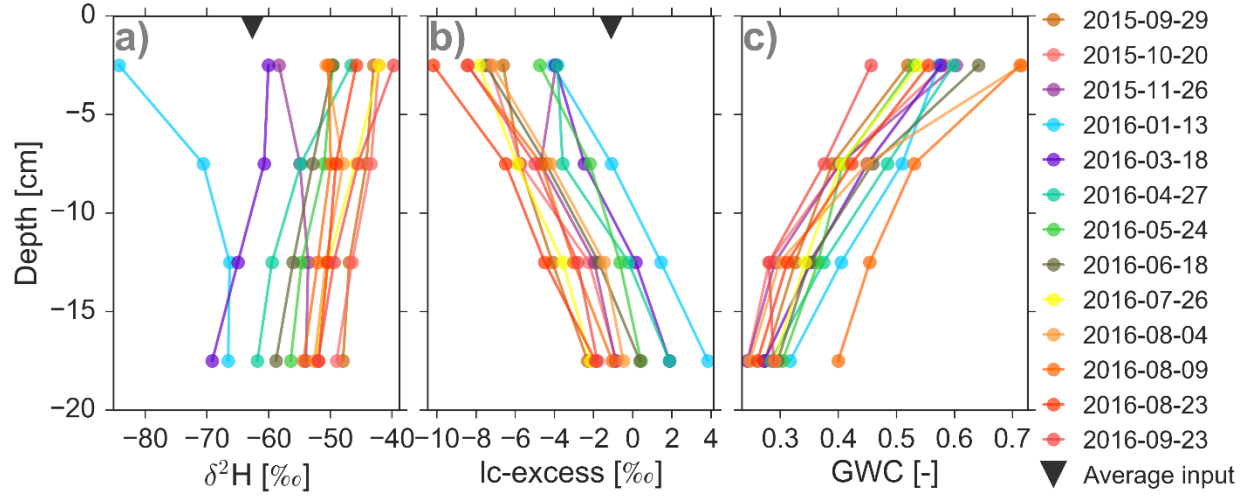
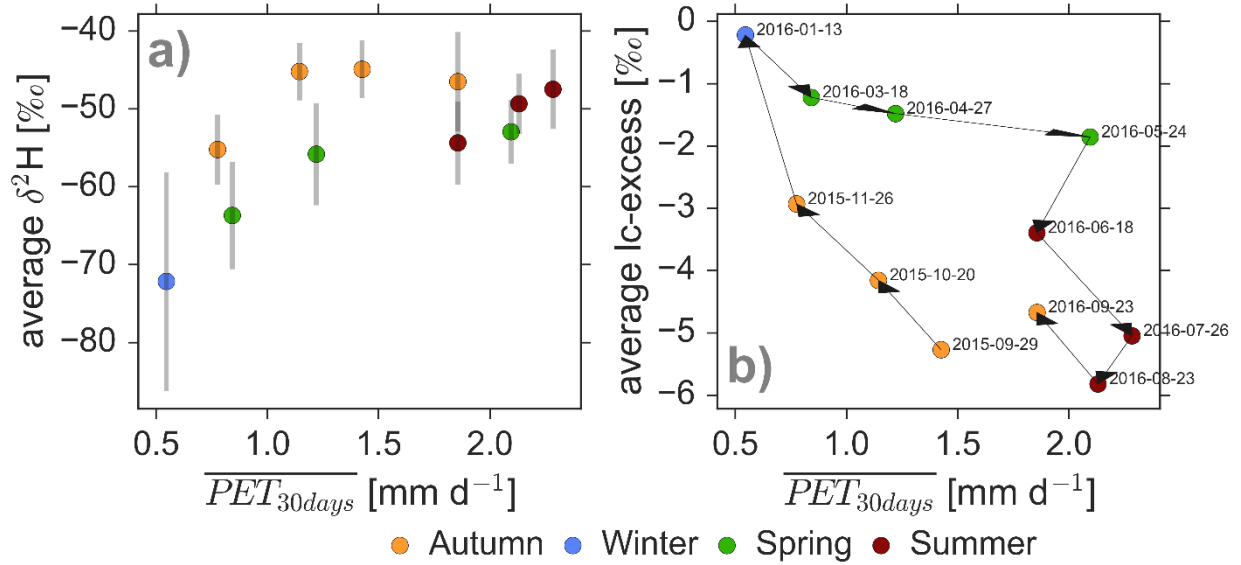


Figure 3 Dual isotope for all soil sampling campaigns and boxplots for  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  bulked over all four sampling sites. The date of each soil sampling is indicated by colours. The depth of the soil samples is shown by different symbols in the dual isotope plot. Sampling dates which do not have significantly different isotope data according to the post-hoc Dunn test are indicated by the same letter next to the boxplots. For example, the letter “a” at the box plots from sampling day 13 January 2016 and 18 March 2016, indicate that the soil water isotopes do not differ significantly from each other, but to all other sampling days. The global meteoric water line (GMWL,  $\delta^2\text{H} = 8 \times \delta^{18}\text{O} + 10$ ) is shown with a solid line and local meteoric water line (LMWL,  $\delta^2\text{H} = 7.6 \times \delta^{18}\text{O} + 4.7$ ) is plotted with a dashed line.



**Figure 4** Depth profiles of (a)  $\delta^2\text{H}$ , (b) lc-excess, and (c) gravimetric water content (GWC) for each sampling day pooled across all sites. Color code indicates sampling days and the black triangle represents the weighted average precipitation input signal during the sampling period ( $\delta^2\text{H} = -62.6$  ‰ and lc-excess = -1.1 ‰).



**Figure 4** Variation of the (a)  $\delta^2\text{H}$  and (b) lc-excess in the top 20 cm of the soil as a function of the potential evaporation averaged over the 30 days prior to the sampling day ( $\text{PET}_{30}$ ). The colours of the scatter points indicate the season of the sampling. The error bars in (a) indicate the standard deviation of  $\delta^2\text{H}$  in the soil waters. The arrows in (b) indicate the order of sampling and sampling dates are given.

## 3.2 Spatial soil water isotopes patterns

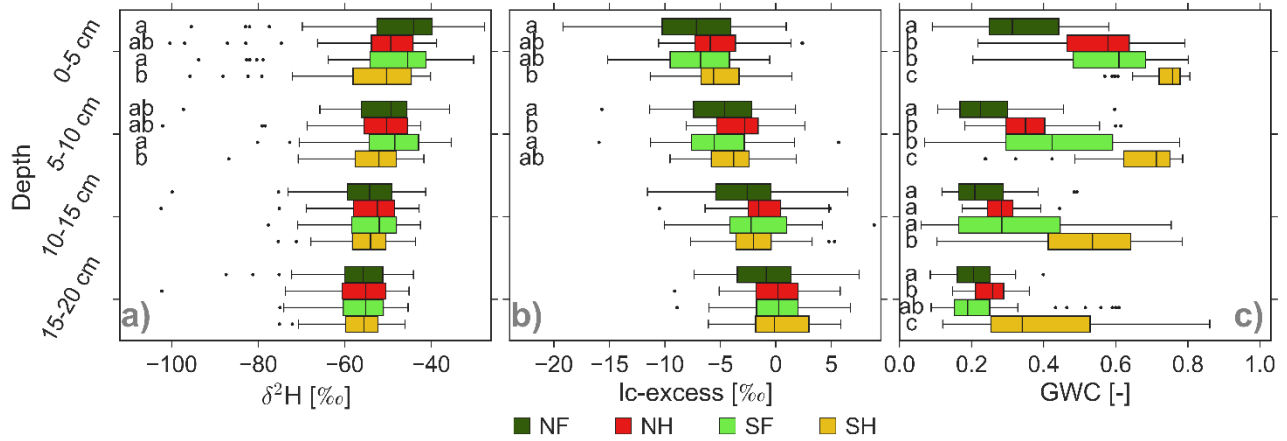
### 3.2.1 Differences between study sites

A comparison of the SW  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values within the upper 20 cm at the sites for different sampling showed no significant differences between the sites (not shown). Differences of the SW isotope values between sites were limited to the upper 10 cm, with a significantly more depleted  $\delta^2\text{H}$  signal for soil water at SH compared to NF (at 0-5 cm) and SF (at 0-10 cm), when looking at values bulked over the entire sampling period (Figure 5Figure 6a). For lc-excess, on average soil water samples at NF and SF showed lower values than NH and SH and differences were significant between NF and SH at 0-5 cm depth and for NF and SF compared to NH at 5-10 cm depth (Figure 5Figure 6b). At all four sites, the SW lc-excess averaged over the upper 20 cm was significantly  $< 0 \text{ ‰}$  between May and October and not significantly  $< 0 \text{ ‰}$  in January. However, the SW lc-excess for NF and SF was often lower than for NH and SH and the lc-excess for NF and SF was significantly  $< -5 \text{ ‰}$  for some sampling campaigns, while NH and SH was usually not significantly  $< -3 \text{ ‰}$ . ~~With regard to~~Regarding GWC (Figure 5Figure 6c), SH had significantly higher values than the other sites and NF had significantly lower values than the other sites in the upper 15 cm. The median GWCs at NH and SF were very similar, but variability was higher in the latter. The GWC was significantly linearly correlated to the organic matter content in the soil (Table 1Table 4) at all sites and this relationship explains partly the significant higher GWC at SH. ~~As already shown in Table 1, GWC is influenced by the organic matter.~~ This influence of organic matter content on GWC was evident during each sampling campaign: the sites with higher LOI tended to have higher average GWCs for the 11 sampling days. The coefficients of determination for the relationship between average LOI and average GWC on the sampling day was generally  $>0.56$  (blue points in Figure 6Figure 7). Note that these coefficients of determination are limited to a sampling number of 4, given by four sampling sites, while the correlations given in Table 1Table 4 are based on  $n = 20$  for each site. This relationship between GWC and LOI persists independently of how many dry days occurred prior to the soil sampling (Figure 6Figure 7).

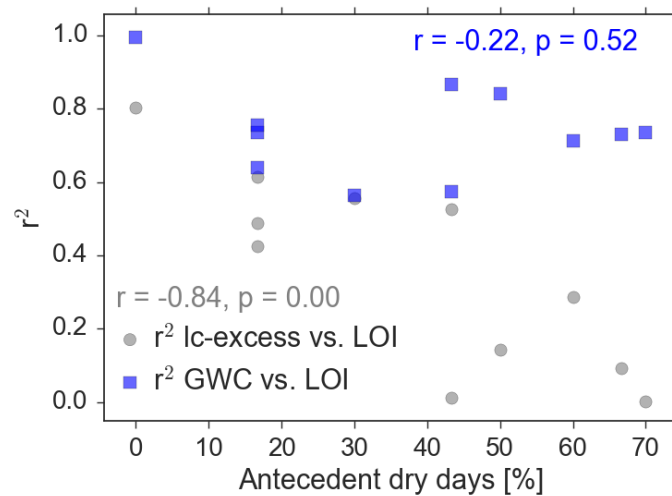
While there was no effect of organic matter content on  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values, average LOI at the sites showed a relationship with lc-excess on several sampling dates. These sampling campaigns, when the  $r^2$  for the relationship between lc-excess and LOI ranged from 0.42 to 0.80, were all characterized by having at least 13 dry days during the 30 day period (40 %) prior to the sampling. For sampling campaigns with  $>40 \text{ ‰}$  dry days more than 13 dry days over the 30 days prior to the sampling, there was no relationship between lc-excess and LOI ( $r^2$  ranged between 0.00 and 0.29, Figure 6Figure 7). Therefore, there is a significant correlation between the correlation coefficient between lc-excess and LOI and the number-percentage of dry days prior to the sampling ( $r = -0.84$ ,  $p < 0.01$ ).

~~The SW lc-excess in the upper 20 cm was usually not statistically different between the sites, since they generally follow a similar pattern (bars in first four columns in Figure 8). At all four sites, the SW lc-excess averaged over the upper 20 cm was significantly  $< 0 \text{ ‰}$  between May and October and not significantly  $< 0 \text{ ‰}$  in January. However, the SW lc-excess for NF and SF was often lower than for NH and SH and the lc-excess for NF and SF was significantly  $< -5 \text{ ‰}$  for some sampling campaigns, while NH and SH was usually not significantly  $< -3 \text{ ‰}$  (background colour in first four columns in Figure 8).~~

Further, SH, the site with the highest GWC, had in 6 out of 11 sampling campaigns the lowest slope of EL, while NF, the site with the lowest GWC, had 6 times the highest slope of the EL (Figure S 2).



**Figure 5** Site specific (a)  $\delta^2\text{H}$ , (b) lc-excess, and (c) gravimetric water content (GWC) for the soils at the four study sites summarized for each site over all sampling dates as function of depth. For depths, where letters are shown, different letters indicate significant differences between the sites for the specific depth.



**Figure 6** Relationship between coefficient of determination of the relationship between lc-excess and LOI (grey) and GWC and LOI (blue) with the percentage number of dry days during the 30 day period prior to the soil sampling.

Figure 8 Average lc-excess values for each study site and each sampling campaign (first four columns), averaged over the 20 cm depth profiles and separated into the 5 cm sampling intervals. The length of the bars represents the lc-excess value (red bars indicate negative lc-excess and blue bars indicate positive lc-excess values). The color of the background provides information if the values are significantly lower than -7 to 0 ‰ at the 95 % confidence interval. White background shows that these samples do not deviate significantly from 0 ‰ or the lc-excess is on average > 0 ‰.

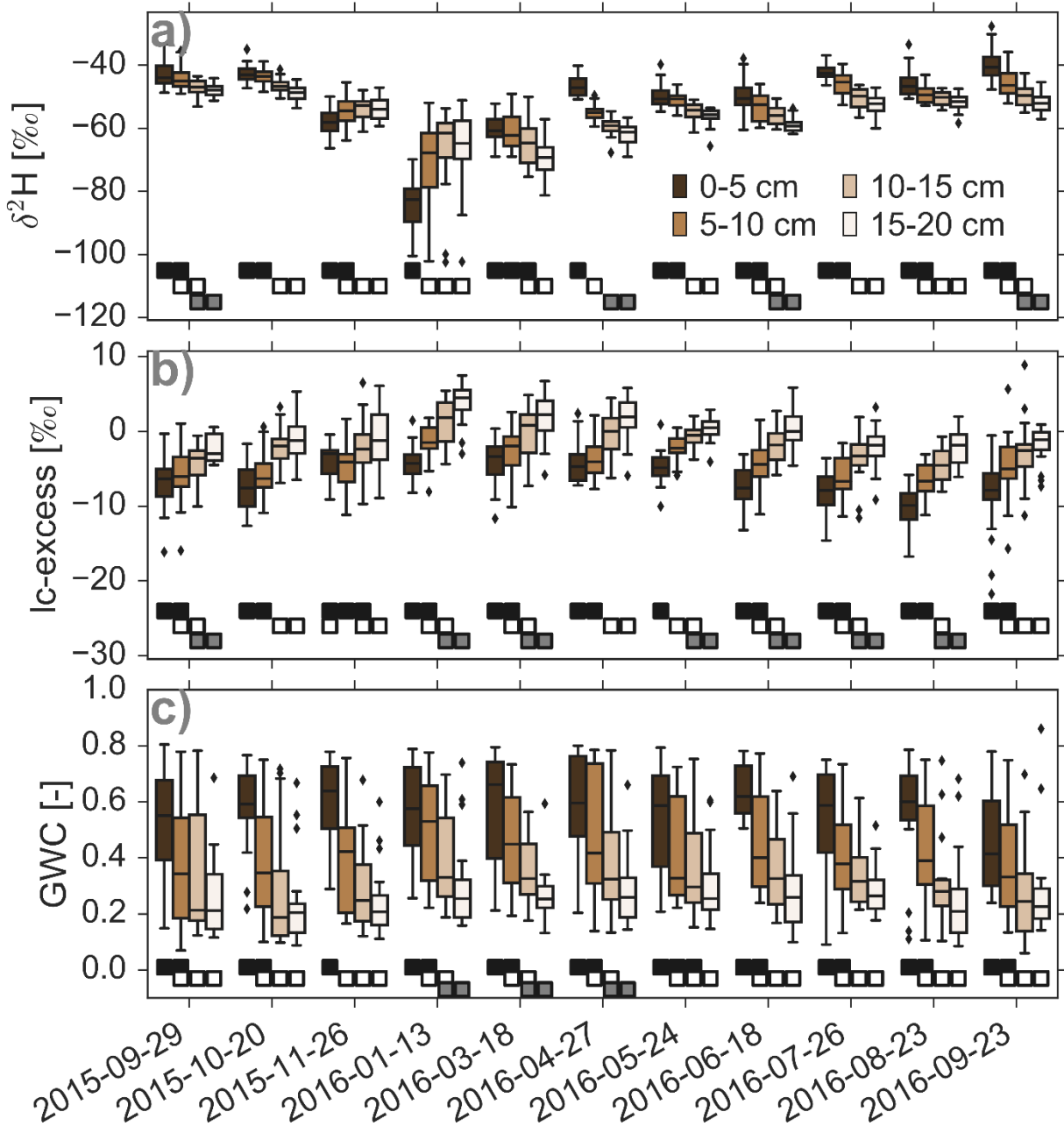
### 3.2.2 Differences with soil depth

During most sampling dates  $\delta^2\text{H}$  values became more depleted with increasing soil depth (Figure 7Figure 9a; SW  $\delta^{18}\text{O}$  very similar and therefore not shown). The  $\delta^2\text{H}$  values for the top 5 cm were always significantly different compared to the values at 15-20 cm. The  $\delta^2\text{H}$  values at 0-10 cm were for most sampling campaigns significantly enriched compared to the soil water at 15-20 cm. However, during the November and January sampling, the upper 0-5 cm were more depleted due to the depleted precipitation  $\delta^2\text{H}$  input (Figure 1b). The highest variability of SW  $\delta^2\text{H}$  within the sampling depths was found for the samples taken in January and March. Importantly, the variability of SW  $\delta^2\text{H}$  generally decreased with soil depth.

The lc-excess for the upper 5 cm was always significantly more negative than at 15-20 cm (Figure 7Figure 9b). In addition, the soil signature at 5-10 cm was significantly more negative than at 15-20 cm; exceptions were November 2015 at 0-5 cm and September 2016 at 5-10 cm. The lc-excess depth profiles had a persistent pattern of steadily decreasing lc-excess values with depth, approaching SW lc-excess of 0 ‰ at 15 – 20 cm depth. The SW lc-excess variability usually decreased with depth, but not for the November sampling (Figure 7Figure 9b).

The soil water SW lc-excess at 15 – 20 cm was usually not significantly < 0 ‰ over the entire sampling period (see also Figure 8). At 10 – 15 cm depth, the SW lc-excess was significantly lower than at least 0 ‰ only between September and November 2015. There were more sampling dates, when the SW lc-excess at 5 – 10 cm was significantly lower than -2 ‰ and usually < 0 ‰ (except for January and August 2016). For, but at the top 5 cm, the SW lc-excess was partly significantly < -7 ‰ and at least significantly lower < 0 ‰ throughout the year; with only few exceptions (January and April 2016).

The upper 5 cm soil had always significantly higher GWCs than the soil between 15 and 20 cm depth. This pattern of decreasing soil moisture with depth was persistent over time. The range of the GWC was always much lower below 15 cm than above (Figure 7Figure-9).



**Figure 7** Differences of (a)  $\delta^2\text{H}$ , (b)  $\text{lc-excess}$ , and (c) gravimetric water content (GWC) over depth for each sampling day. Black, white and grey squares below the boxplots indicate significant differences between the sites-depth for each sampling day according to the Dunn test (i.e. same colour reflects similarities; different colours differences). E.g., for 2015-09-29, samples from 0-5 cm depth were significantly different from samples from the depth 10-15 cm and 15-20 cm, but not significantly different to samples from 5-10 cm depth.

### 3.2.3 Differences due to vegetation cover

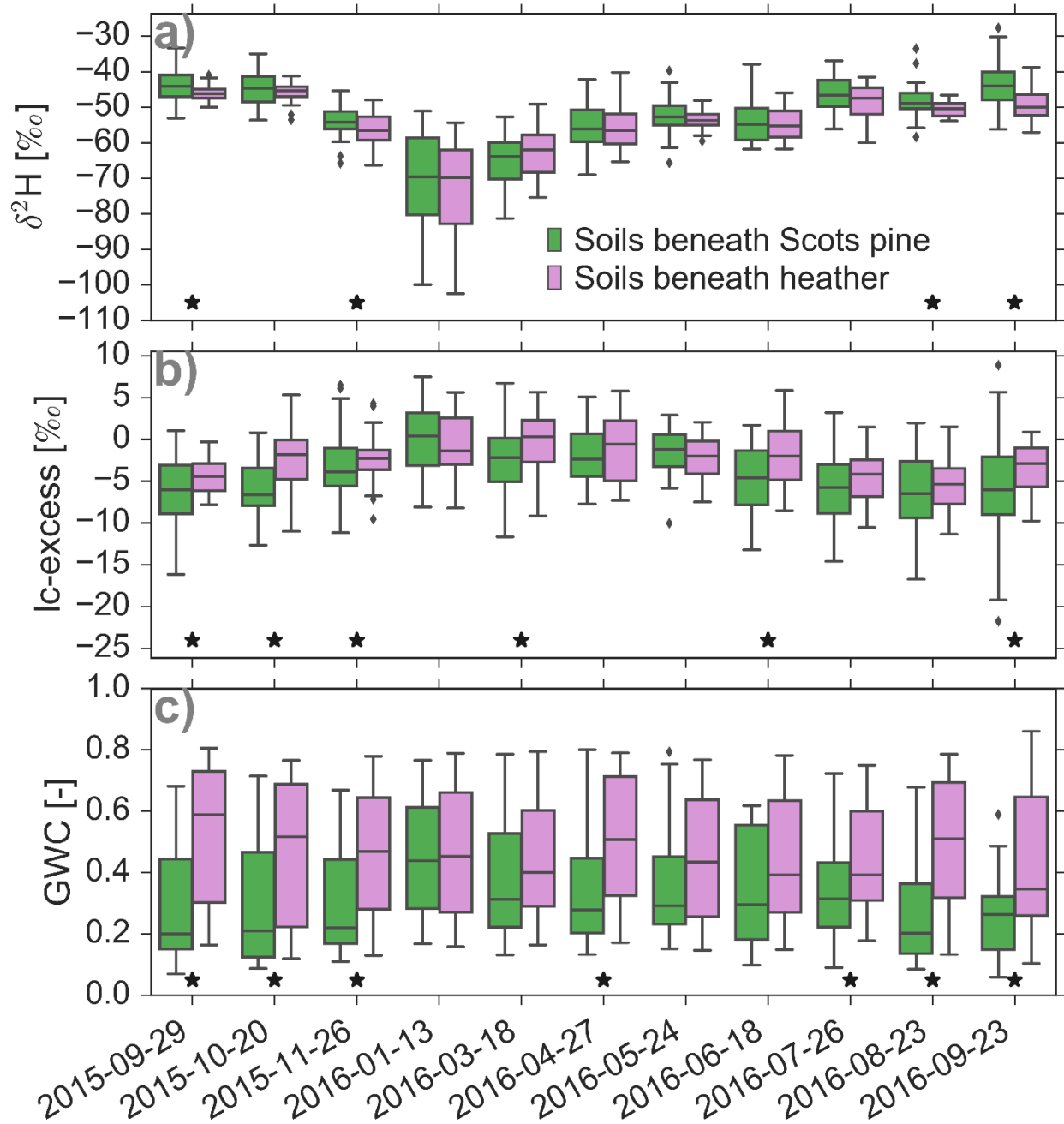
Comparing all soil samples bulked over depth and sampling campaigns, SW beneath Scots pine was significantly more enriched in  $\delta^2\text{H}$  (Mann-Whitney-Wilcoxon Test,  $p=0.038$ ) and  $\delta^{18}\text{O}$  (~~Mann-Whitney-Wilcoxon Test~~,  $p=0.0062$ ) than the SW beneath heather sites. In addition,  $\text{Ic-excess}$  was significantly lower for the SW at the forested sites compared to the heather sites (~~Mann-Whitney-Wilcoxon Test~~,  $p < 0.01$ ).

The temporal dynamics of the differences of  $\delta^2\text{H}$  (and also SW  $\delta^{18}\text{O}$ , but not shown) values for SW beneath the different vegetation sites show that the SW beneath Scots pine was (except for the sampling in March for  $\delta^2\text{H}$ ) always more isotopically enriched than the soil water beneath heather (~~Figure 8~~~~Figure 10a~~). However, the differences were only significantly different for four out of the eleven sampling campaigns (~~i.e., Sept. 2015, Nov. 2015, Aug. 2016, Sept. 2016~~). SW  $\delta^2\text{H}$  values showed usually higher variability for soils beneath Scots pine for the sampling campaigns between May and October, but during Winter, the variability is generally high for soils beneath both vegetation types.

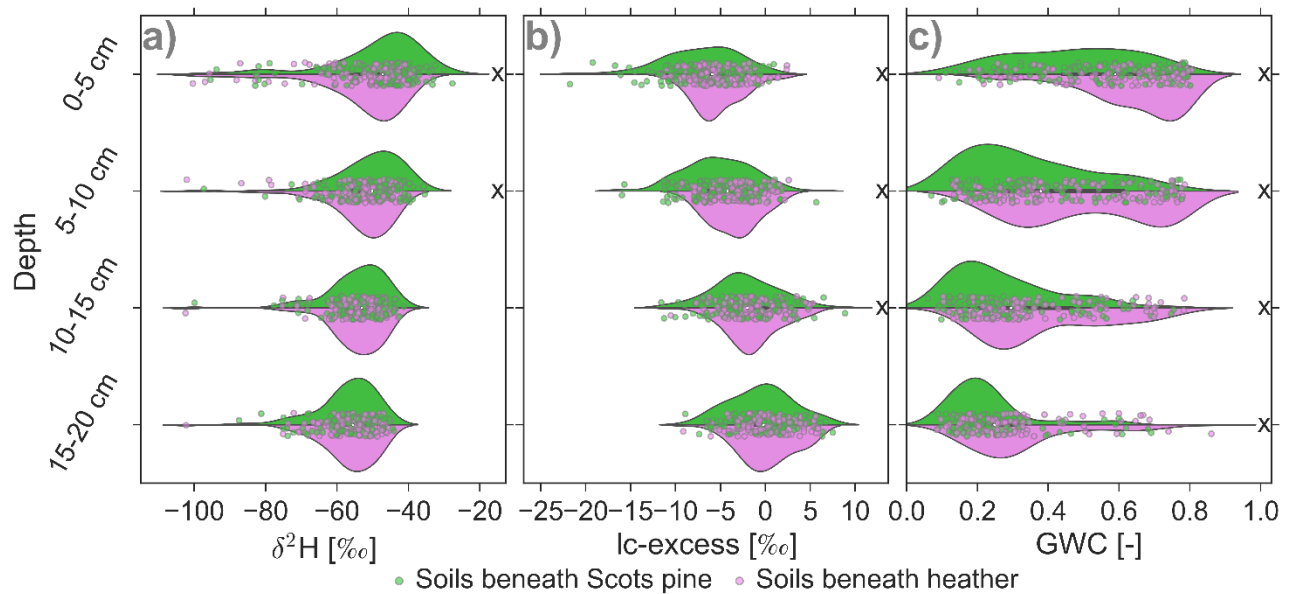
The SW  $\text{Ic-excess}$  ~~in the upper 20 cm~~ was - except for the January and May sampling campaigns - more negative in the soils beneath Scots pine compared to soils beneath heather (~~Figure 8~~~~Figure 10b~~). ~~This difference was significant for more than half of the sampling campaigns.~~ The GWC was also always lower in the soils beneath Scots pines. These differences were most pronounced at the end of the summer and during autumn (~~Figure 8~~~~Figure 10c~~).

The differences between the SW isotopic composition under the two different vegetation types mainly stemmed from differences in the shallow soils. Despite being classed as similar podzolic soils, the SWs beneath Scots pine were significantly more enriched in the upper 10 cm in  $\delta^2\text{H}$  (and also  $\delta^{18}\text{O}$ , not shown) than the SW beneath heather (~~Figure 9~~~~Figure 11a~~). SW beneath Scots pine had a significantly more negative  $\text{Ic-excess}$  signal for the upper 15 cm soils. ~~Below 15 cm, the differences between SW under Scots pine and SW under heather were not significant (Figure 11b).~~ The GWC of soil beneath Scots pine were over the entire soil profile significantly lower than for the soils beneath heather (~~Figure 9~~~~Figure 11b~~).





**Figure 8** Differences of (a)  $\delta^2\text{H}$ , (b) lc-excess, and (c) gravimetric water content (GWC) for soils under Scots pine (green) and soils under heather (violet) during one year. Significant differences estimated with the Mann-Whitney-Wilcoxon test between soils under Scots pine and soils under heather for a particular day are indicated by a star.



**Figure 9** Violin plots showing the distribution as a kernel density estimation of the soil water (a)  $\delta^2\text{H}$ , (b) lc-excess, and (c) gravimetric water content over all sampling campaigns with differentiation between soils beneath Scots pine (green) and soils beneath heather (purple). The symbol “X” at the depths indicates significant differences between the soils under Scots pine and soils under heather for the particular depth as estimated with the Mann-Whitney-Wilcoxon test. Dots indicate the individual sample points.

### 3.2.4 Differences due to aspect

No significant differences were found when splitting the samples according to their aspect. There were neither differences when looking at all sampling depths, nor for any individual sampling depth (not shown). A comparison of the more intensive spatially distributed sampling focusing on the top 10 cm of heather soils in May 2016 at the north facing sites ( $n = 26$ ) and south facing sites ( $n = 33$ ) showed also no significant differences between aspects ~~with regard~~ ~~to~~ regarding isotopes and soil moisture. Increasing the sampling numbers for the heather sites when the two additional sampling campaigns in August were included, also did not result in significant differences between the two studied slopes. The median SW  $\delta^2\text{H}$  values for the samples taken at the south and north facing slopes were  $-50.6 \text{ ‰}$  and  $-50.9 \text{ ‰}$ , respectively. The median SW lc-excess was  $-3.4 \text{ ‰}$  and  $-3.3 \text{ ‰}$  for south and north facing slope, respectively. The median GWC was 0.62 and 0.54 for the samples at the south and north facing slope, respectively.

## 3.3 Evaporation estimates

Our findings clearly showed an influence of the vegetation on the evaporation signal in the soil water between April and October. Therefore, ~~for that period we applied the Hydrocalculator, we estimated the fraction of evaporation losses in the SW beneath Scots pine and heather~~ as described in the methods section, ~~to estimate the evaporative water losses  $f$  and evaporation to input ratios ( $E/I$ ), for the soil water beneath Scots pine and heather~~ for the nine samplings in this period. For the heather sites, the median ( $\pm$  SD ~~over the period~~) values of  $f_{d2\text{H}}$  and  $f_{d18\text{O}}$  were  $4.55.0 \pm 14.0 \text{ ‰}$  and  $3.7 \pm 1.6 \text{ ‰}$ , respectively, and for the forested sites, the values were  $f_{d18\text{O}} = 7.79.9 \pm 1.73.4 \text{ ‰}$  and  $f_{d2\text{H}} = 8.46.3 \pm 2.19 \text{ ‰}$ . Thus, the SW fractionation signal indicates that about 8 % and 4.5 % of the originally infiltrated water (with lc-excess = 0

~~%) went back into the atmosphere by soil evaporation. This indicates that between April and October, usually 5 % and 8 % of the soil water was evaporated beneath heather and forests, respectively.~~

~~This translates to median values for the ratio between the evaporation and the infiltration (net precipitation) of  $E/I_{d2H} = E/I_{d18G} = 5 \pm 2.4$  % for heather and  $E/I_{d2H} = 10.4 \pm 3.8$  % and  $E/I_{d18G} = 9.3 \pm 2.0$  % for Scots pine during the growing season. Thus, of 20 mm throughfall and stemflow reaching the soil, 1 and 2 mm will evaporate from the soils beneath heather and Scots pine, respectively. The residual water will be taken up by the roots or percolate into the subsoil.~~

## 4. Discussion

### 4.1 Mixing of precipitation input and the critical zone water storage

Our uniquely detailed data set from 11 sampling campaigns over one year revealed the isotopic response of soil water to variable precipitation inputs. An understanding of the isotopic variability of the precipitation input signal is crucial for the interpretation of the soil water isotopes in terms of soil evaporation dynamics at the soil-plant-atmosphere interface. Therefore, we will first discuss how mixing of the infiltrating precipitation within the topsoil provides the basis for the isotopic enrichment in the soil due to evaporative losses.

Given that the soil at the four sites consists of similar texture, it is not too surprising that the SW generally responded similarly to the seasonal P  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  input signal. However, we would expect differences in the soil physical properties due to the differences in the organic matter contents. Our data showed that higher organic matter resulted in higher GWCs at the sites. That means that the soil water storage is higher for the sites with higher organic material, which is in line with several other studies (e.g., Hudson, 1994).

While this relationship between GWC and LOI persisted independently of how many dry days occurred prior to the soil sampling, LOI showed a relationship with lc-excess depending on the dryness. This suggests that during dry periods, the external atmospheric drivers such as evaporative demand may have a high influence on the SW lc-excess. In contrast, during wet periods, the soil water storage capacities – here, mainly controlled by organic material – gain importance. With increasing new ~~usually unfractionated~~ precipitation input (lc-excess close to zero), the sites with higher organic matter content have a different SW lc-excess than the sites with lower organic matter content, because there is a higher mixing volume for the sites with the high LOI compared to the soils with lower LOI (Figure 6Figure 7). Hence, future and current changes in the organic matter content of soils with high organic matter due to, for example, land use or climate changes (Foley et al., 2005; Rees et al., 2011), would have an impact on the mixing of event and pre-event water in the podzols.

Interestingly, the highest variability of the  $\delta^2\text{H}$  values in the soil water was found for the sampling after the intense rainfalls in January 2016 (Figure 4Figure 5a, Figure 7Figure 9a), which indicates that the unsaturated zone was not homogeneously wetted by the exceptionally high precipitation input. ~~The GWC corroborates this, because its variability for the sampling campaign in January was not lower than for other sampling days (Figure 9b).~~ This suggests that bypass flow occurred and the stored soil water was not necessarily well-mixed. However, the significantly lower

$\delta^2\text{H}$  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. The sampling in January was ~~further also~~ special, because the GWC was about as high in the soils beneath Scots pine than in the soils beneath heather. Hence, differences in GWC between soils beneath the two vegetation types during the ~~rest of the year~~vegetation period as discussed below would stem from different transpiration and soil evaporation rates and not from differences in the soil water storage capacities.

The higher variability of the SW  $\delta^2\text{H}$  values beneath Scots pine compared to the SW beneath heather (~~Figure 9~~Figure 44) cannot be explained by differences of the throughfall isotopic signal, since they are minor for the two vegetation types (Braun, 2015). The higher variability in the isotopic signal therefore indicates that flow paths are generally more variable in the forest soils. This is probably due to preferential flows via larger macropores formed by tree roots that also reach deeper than the shallower roots of heather. While the high variability of SW  $\delta^2\text{H}$  values indicate preferential flow paths, there was also a clear signal that much of the input water percolates through the pore matrix, since we see that the depleted winter precipitation signal is still evident at -20 cm soil depth during the March sampling campaign (~~Figure 7~~Figure 9a).

The relatively high soil water storage volumes in the podzols result in a damping of the  $\delta^2\text{H}$  signal in the SW compared to the precipitation input. There was also a more damped isotopic signal with soil depth due to increased mixing and cumulatively larger soil water storage. Because of this damping effect, the differences between the sites (~~Figure 5~~Figure 6a, ~~Figure 9~~Figure 11a) and sampling times (~~Figure 7~~Figure 4a) decreased with depth.

Our data support the findings of Geris et al. (2015b), who showed - with soil water samples extracted with suction lysimeters - a more damped signal in deeper layers of the soils beneath heather than beneath Scots pine. However, we did not see a more delayed response beneath Scots pine compared to podzols beneath heather as observed by Geris et al. (2015b). This discrepancy could arise from the fortnightly sampling frequency by Geris et al. (2015b) and monthly sampling frequency in the current study, as well the effect of an unusually dry summer in former study.

In contrast to Geris et al. (2015b), who sampled the soil-vegetation units with duplicates, our sampling design with five replicates allowed ~~to for~~ a clearer assessment of the spatial heterogeneity of the subsurface. ~~Our results emphasize the importance of the need to account for the spatial variability of the pore water isotopic signal.~~ For example, the ~~variability standard deviation~~ of the SW  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  values ~~at for the -5 cm depth increments for 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. This variability of soil water isotope signals will also affect~~ For the interpretation of potential sources of root water uptake this means that the uncertainty of the potential water source signal – caused by the heterogeneous isotopic composition at particular depths within the rooting zone – is higher than the measurement errors. 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). (for examples with replicates see McCutcheon et al., 2016 and Goldsmith et al., 2012). 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).

The observed mixing processes have implications for modelling of water movement within the critical zone. The soil water isotope response seems to follow a replacement of the pore waters by a mixture of newly infiltrated precipitation and older water, which could probably be described by the advection dispersion equation as frequently applied for isotope modelling in the unsaturated zone (Adomako et al., 2010; Stumpp et al., 2012; Mueller et al., 2014; Sprenger et al., 2015b). However, during intense rainfall, the precipitation input seems to partly bypass the matrix leading to rapid changes in the tracer signal in the subsoil and a generally high variability of the soil water isotopes within the entire soil profile. Such preferential flow processes might be better represented by soil physical models that take different mobility of water within the pore space into account, like the dual-permeability (Gerke and van Genuchten, 1993) or dual-porosity (van Genuchten and Wierenga, 1976) approaches. For example, Gouet-Kaplan et al. (2012) showed with column experiments that a significant amount of pre-event (or “old”) water stayed in a sandy pore system. They found that these processes could be described by a dual-porosity (mobile-immobile) model. Such incomplete mixing within the unsaturated zone could have an impact on catchment transit time estimates (van der Velde et al., 2015). Given the high storage volumes of the periglacial deposits (Soulsby et al., 2016b) and the high water mixing volume in the riparian zone (Tetzlaff et al., 2014) in the Bruntland Burn, it is yet unclear, which role the relatively slow moving and partly mixed soil waters in the soil matrix at the hillslopes could play regarding long tails of transit times in the Scottish Highlands, as reported by Kirchner et al. (2010).

#### **4.2 Evaporation dynamics within the soil-plant-atmosphere interface**

The correlation between the seasonally variable P and SW isotopic signals inhibited an assessment of soil evaporation processes from either  $\delta^2\text{H}$  or  $\delta^{18}\text{O}$ . Instead, the SW lc-excess values were independent from the P input and were therefore indicative of kinetic fractionation due to soil evaporation. The monthly soil water sampling revealed **kinetic fractionation** dynamics (in terms of lc-excess) during times of highest potential evaporation for the Scottish Highlands. The evaporation fluxes were not expected to be high given the relatively low energy environment at the study site. The soil waters showed, nevertheless, a clear fractionation signal in terms of their lc-excess (25th percentile  $< -6\text{‰}$  between June and October) and the evaporation line (3.6 and 4.5 between June and October, Table 2). This fractionation signal in the soil water was of the same magnitude as for surface waters in the peatland drainage network of a raised bog in the Bruntland Burn, where the lc-excess reached values  $< -5\text{‰}$  and the EL slope ranged from 3.9 to 4.9 between May and September (Sprenger et al., 2017b). In comparison to arid and Mediterranean environments, where SW lc-excess can fall below  $-20\text{‰}$  (McCutcheon et al. (2016) and reviewed in Sprenger et al., (2016b), the SW lc-excess in the Scottish Highlands remained relatively high. While the lc-excess was usually not significantly different from zero at 15 – 20 cm soil depth in the studied podzols of the study site, soils studied by McCutcheon et al. (2016) in a much drier environment (Dry Creek, Idaho) showed that the SW lc-excess from the surface down to -70 cm was significantly lower than zero.

The high soil water storage contributes to the SW lc-excess dynamics shown in the hysteresis pattern for the relationship between SW lc-excess and  $PET_{30}$ , which revealed that there was a delayed response of the SW lc-excess to the onset and offset of soil evaporation in spring and autumn, respectively. We explain the hysteresis by generally low soil evaporation fluxes, relatively high humidity in the Scottish Highlands, and high soil water storage. The high soil water storage means that relatively high evaporation losses are needed in spring to change the  $^2H$  to  $^{18}O$  ratios by kinetic fractionation. The variability of SW lc-excess values from July to September was relatively small, indicating that a steady state between soil water fractionation by soil evaporation and input of unfractionated precipitation was reached. In autumn, when the soil evaporation ceased, relatively high unfractionated precipitation input was needed ~~in order to~~ dilute the evaporation fractionation signal of the soil water again (Figure 4Figure 5).

In contrast to our findings of a pronounced evaporation fractionation in the soil water, Geris et al. (2015b), who sampled the mobile soil water for its isotopic composition with suction lysimeters, saw little to no fractionation. The sampling by Geris et al. (2015b) took place at the same/similar sites (podzols beneath Scots pine and heather within the Bruntland Burn), but during different years. However, the sampling period of Geris et al. (2015b) covered summer 2013, which was exceptional dry (~10 year return period) and would have been therefore very likely to induce evaporation fractionation in soil waters. We conclude that the differences between the two studies most likely relate to the different sampling methods. Suction lysimeters, as applied by Geris et al. (2015b), are known to be limited to sample the mobile water within the pore space, ~~but, if~~ The direct equilibration method applied in our study also samples more tightly bound waters (Sprenger et al., 2015a). Consequently, we can infer that the mobile waters sampled with suction lysimeters will be relatively young waters that percolated without experiencing pronounced water losses due to soil evaporation. In contrast, the soil water data presented in our study, represent a bulk pore water sample, where more tightly bound waters will be integrating older ages and therefore, affected by kinetic fractionation during periods of atmospheric evaporative demand. However, comparisons between mobile and bulk soil water isotopic signals showed that tightly bound water at soil depth deeper than 10 cm was usually more depleted than the mobile waters during spring or summer, due to filling of fine pores of a relatively dry soil with depleted precipitation several months earlier (Brooks et al., 2010; Geris et al., 2015a; Oerter and Bowen, 2017). Our study shows that, in contrast to old isotopically distinct infiltration water, also the legacy of evaporation losses over previous months allows for separating between pools of different water mobility in the soil-plant-atmosphere interface. This means that old (more tightly bound) water might not only have a distinct  $\delta^2H$  or  $\delta^{18}O$  signal compared to mobile water due to seasonally variable precipitation inputs, but also an 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.

So far, mostly cryogenic extraction was used to analyse the isotopic composition of bulk (mobile and tightly bound) soil waters and relate these data to for example root water uptake pattern. However, recent experimental studies indicated potential for fractionation during the cryogenic extraction (Orlowski et al., 2016), probably induced by water-mineral interactions (Oerter et al., 2014; Gaj et al., 2017a; Gaj et al., 2017b). The direct-equilibration method –



as a potential alternative analysis method - we applied in our study was so far usually limited to study percolation processes (e.g., Garvelmann et al., 2012; Mueller et al., 2014; Sprenger et al., 2016a). To our knowledge, only Bertrand et al. (2012) used the direct-equilibration method to study the water use by plant water use and the influence of evaporation fractionation, but they bulked the soil data of the upper 20 cm.

However, our results emphasize the importance of sampling the upper most soil layer when studying plant-water interactions and soil evaporation dynamics. The evaporation was predominantly taking place at the interface to the atmosphere, where the lowest SW lc-excess values were found throughout the sampling period. Due to the relatively wet conditions of the studied soils, the evaporation signal dropped sharply within the first 15 cm soil depth and was always significantly lower at the top 5 cm compared to the SW at 15 – 20 cm depth. Geris et al. (2015a) sampled with cryogenic extraction the isotopic composition of the bulk soil water at -10 cm soil depth at the site NF during the summer of 2013. They found soil water isotopes of the same magnitude at -10 cm ( $\delta^2\text{H}$  between -40 and -55 ‰), but their sample size was too little to assess evaporation fractionation patterns.

The pronounced differences of the soil water samples in the dual isotope space over little soil depth is crucial when assessing potential water sources of the vegetation. The lc-excess signal could therefore provide additional information as an end-member when applying mixing models to derive root water uptake depths and times, since  $\delta^2\text{H}$  and lc-excess do not necessarily correlate. So far, usually either  $\delta^2\text{H}$  or  $\delta^{18}\text{O}$ , but not lc-excess are being considered to delineate water sources of the vegetation (Rothfuss and Javaux, 2016). In line with the call for a higher temporal resolution of soil water isotope sampling (Berry et al., 2017), the relatively-highly dynamic of the 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) ~~published literature~~), when investigating root water uptake patterns. In the light of recent studies dealing with potential water sources of vegetation that showed how variable the plant water isotopic signal can be over a year (Hervé-Fernández et al., 2016; McCutcheon et al., 2016), a proper understanding of the temporal variability of the potential water sources (i.e., bulk soil water, groundwater, stream water) appears to be critical.

#### 4.3 Vegetation affects critical zone evaporation losses

The vegetation was the main reason for differences in the soil water evaporation signal between the four study sites. While we limited the study to one soil type, we still saw differences between the soils in terms of their organic matter content leading to different soil water storage capacity. However, these differences in water storage capacity seemed to only influence the evaporation signal during periods of high ~~wetness~~precipitation input, when evaporation is already likely to be low (Figure 6~~Figure 7~~). The aspect and exposition of the slopes did not affect the soil water evaporation signal, which probably can be explained by the low angle (4°) for the north- and south-facing slopes at our site. Differences in net radiation for the two studied slopes were found to be small with the north facing slope receiving about 4 % less radiation than the south facing slope between April and October (Ala-Aho et al., 2017).

The significant differences of the SW lc-excess in soils beneath Scots pine and heather are mainly due ~~mainly~~ to the differences of the vegetation structure. The heather forms a dense soil cover just about 20 cm above the ground with



an understory of mosses and lichens. Thus, the vegetation shades most of the soil and generates a microclimate with little direct exchange between soil vapour and atmospheric vapour. The soil climatic conditions beneath heather are therefore characterized by higher relative humidity and less radiation input than for the soils beneath Scots pine. In addition, the soils beneath the Scots pine are not densely vegetated by understory, which allows for more open exchange between soil vapour and atmospheric vapour at the forested sites. A similar conclusion of microclimatic conditions driving the differences in the evaporation signal was drawn by Midwood et al. (1998), who also reported a higher isotopic enrichment of soil surface soil waters under groves and woody clusters than under grassland.

The fractionation signal ~~will generally was shown to~~ be more pronounced for evaporation losses from drier soils compared to wetter soils (Allison et al., 1983; Barnes and Allison, 1988) ~~a smaller water pool than a larger one~~. Higher canopy storage, higher interception losses and reduced net precipitation in the forested stands, where throughfall is about 47 % of precipitation, compared to the heather with throughfall being about 35 % of precipitation (Braun, 2015) will influence the GWC of the soils beneath the two vegetation types. The higher transpiration rates of the trees (Wang et al., 2017) compared to the heather further influences the soil moisture dynamics, leading to significantly lower GWC in the forested sites. In consequence, soil evaporation taking place from the drier soils beneath Scots pine will result in higher evaporation fractionation than for soil evaporation from soils beneath heather.

The evaporation estimates are limited to the period of highest evaporation fluxes in the Bruntland Burn and are likely to be lower than what we present, since we relate the isotopic enrichment to the long term average input signal in our calculations. However, the assumption of relating the enrichment to the average input seems to be valid, since we have seen the steady state balance of unfractionated input and kinetic fractionation due soil evaporation during the summer months. Nevertheless, detailed estimates of the evaporative fluxes require transient modelling of all water fluxes (i.e., precipitation, transpiration, evaporation, recharge) and their isotopic composition, which will be subject of future work. 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).

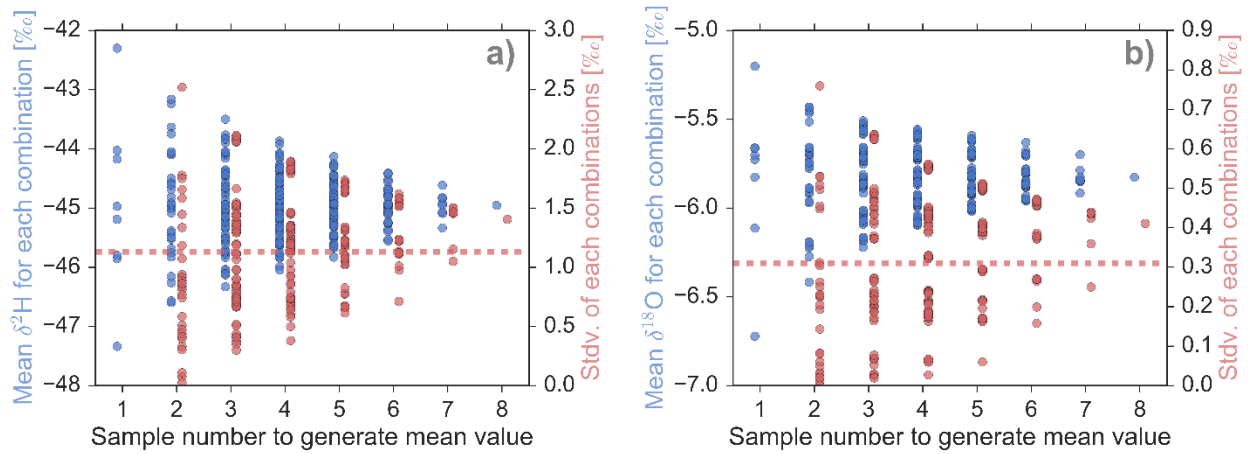
The frequently measured soil water isotope data we have presented and the inherent evaporation signal can help to calibrate or benchmark the representation of soil-vegetation-atmosphere interactions in tracer aided hydrological modelling from the plot (Rothfuss et al., 2012; Sprenger et al., 2016b) to the catchment scale (Soulsby et al., 2015; van Huijgevoort et al., 2016). So far, the isotopic composition of mobile water has been used to better constrain semi-distributed models (Birkel et al., 2014; van Huijgevoort et al., 2016). Including the isotopes of the bulk soil water, could allow for an improved conceptualization of the soil-plant-atmosphere interface, which is crucial for an adequate representation of evaporation and transpiration. Especially the marked differences of the evaporation signal within the first few centimetres of the soil depth will significantly affect the age distribution of transpiration (Sprenger et al., 2016c), evaporation (Soulsby et al., 2016a) or evapotranspiration (Harman, 2015; Quéloz et al., 2015; van Huijgevoort et al., 2016).

## 5. Conclusions

Our study provides a unique insight into the soil water stable isotope dynamics in podzolic soils under different vegetation types at a northern latitude site. We showed that, despite a relatively low energy environment in the Scottish Highlands, the temporal variability of soil water isotopic enrichment was driven by changes of soil evaporation over the year. The monthly frequency of the soil water isotope sampling corroborates the importance of covering transition periods in a climate with seasonally variable isotopic precipitation input signals and evaporative output fluxes. Missing sampling these periods of higher temporal variability in spring and autumn could pose problems when referring plant water isotopes to potential soil water sources or when using soil water isotopic information for calibrating hydrological models. Especially the delayed response of the soil water  $\delta^{18}O$ -excess to evaporation (hysteresis effect) provides valuable insight into how unfractionated precipitation input and kinetic fractionation due to soil evaporation both affect mixing processes in the upper layer of the critical zone. The fact that the evaporation signal generally disappears within the first 15 cm of the soil profile emphasises the importance and the spatial scale of the processes taking place at the soil-vegetation-atmosphere interface of the critical zone.

The vegetation type played a significant role for the evaporation losses, with a generally higher soil evaporation signal in the soil water isotopes beneath Scots pine than beneath heather. Notably, these differences - as indicated in the soil water  $\delta^{18}O$ -excess - remain limited to the top 15 cm of soil. The vegetation cover directly affected the evaporation losses with soil evaporation being twice as high as beneath Scots pine (10 % of infiltrating water) compared to soils beneath heather (5 % of the infiltrating water) during the growing season.

The presented soil water isotope data covering the seasonal dynamics of exceptionally high sampling frequency in time (monthly) and at high space-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 by for Vereecken (2016)), but The data can further provide also a basis to benchmark hydrological models to realize for a more realistic representation of the soil-vegetation-interactions celerities and velocities when simulating water fluxes and their ages within catchments (McDonnell and Beven, 2014).



**Figure S 1** Spatial variability of (a)  $\delta^2\text{H}$  and (b)  $\delta^{18}\text{O}$  in soil water and the effects of taking several samples in parallel to get average values (blue dots and primary y-axis) for the samples if only having one sample (1 on the x-axis) or averaging over 2 to 8 samples (2 to 8 on the x-axis). Red dots indicate the standard deviation of the different combinations and the red dotted line shows the precision of the isotope analysis. All 8 samples were taken in the upper 5 cm of soil within 10 m distance to each other.

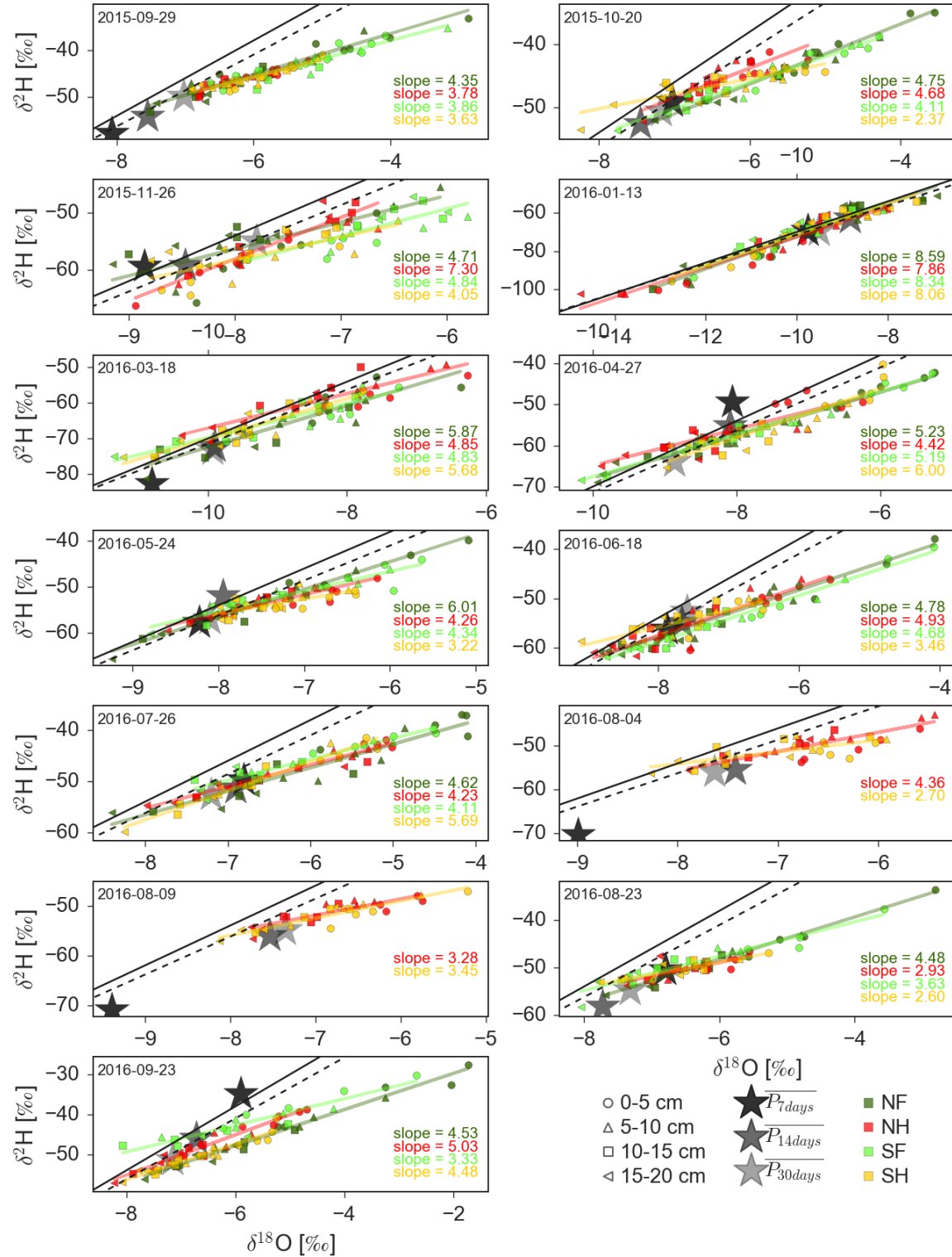


Figure S 2 Dual isotope plots for each sampling day showing the pore water isotopic composition and the isotopic signal in the precipitation averaged over the 7 days (black stars), 14 days (grey stars), and 30 days (light grey stars) prior to the sampling date. Colors indicate the sampling site and marker indicate the sampling depth. Solid line shows the GMWL and the dotted line represents the local meteoric water line (LWM:  $\delta^2\text{H} = \delta^{18}\text{O} \times 7.6 + 4.7$  ‰).

**Author contribution**

D.T., C.S. and M.S. established the sampling design together, M.S. conducted the field and laboratory work and statistical analysis, all authors were involved in the data interpretation, M.S. prepared the manuscript with contributions from both co-authors.

**Competing interests**

The authors declare that they have no conflict of interest.

**Data availability**

The data is available upon request.

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**Table 1** Vegetation and soil profile characteristics of the four study sites: Percentage of coarse gravel (>20 mm diameter) and fine gravel (20-2 mm diameter) of the soil sample, percentage of sand (S, 2-0.6 mm), silt (Si, 0.06-0.002 mm), and clay (C, <0.002 mm) of the fine soil matrix. Percentage of organic matter content in the soil as loss on ignition (LOI), correlation characteristics for LOI with depth and with gravimetric water content (GWC), and fine root density as percentage of total root mass. Note that for SH, NF, and SF, not enough fine soil was left for soil texture analysis after ignition of organic material at 550 °C and that for NF and SF, no root density could be measured due to root thickness and stone content. ± indicate standard deviations out of five replicates for LOI values and range of two and three replicates for root density, respectively.

Site	Vegetation	Depth [cm]	Coarse gravel [% of soil]	Fine gravel [% of fine soil]	Sand [% of fine soil]	Silt	Clay	LOI [% of soil]	LOI vs. Depth	LOI vs. GWC	Fine root density [% of total roots]
NH	Heather	0-5	0	0	74	22	4	26 ± 15			82 ± 7
		5-10	0	5	77	19	4	11 ± 5	r = 0.66;	r = 0.73;	15 ± 5
		10-15	0	11	78	18	4	8 ± 2	p < 0.01	p < 0.01	3 ± 1
		15-20	2	10	80	16	4	9 ± 6			0
SH	Heather	0-5	0	0	-	-	-	92 ± 4			56 ± 18
		5-10	0	1	57	35	8	79 ± 14	r = 0.72;	r = 0.87;	24 ± 9
		10-15	0	5	78	18	4	54 ± 27	p < 0.01	p < 0.01	16 ± 6
		15-20	0	18	81	16	3	50 ± 39			4 ± 2
NF	Scots pine	0-5	0	1	-	-	-	28 ± 11			
		5-10	0	2	80	17	3	31 ± 20	r = 0.42;	r = 0.70;	
		10-15	0	0	77	18	5	20 ± 18	p = 0.06	p < 0.01	
		15-20	0	2	77	20	3	14 ± 10			
SF	Scots pine	0-5	0	1	-	-	-	95 ± 1			
		5-10	12	27	84	12	4	58 ± 36	r = 0.69;	r = 0.91;	
		10-15	15	22	85	12	3	40 ± 23	p < 0.01	p < 0.01	
		15-20	18	18	81	16	3	45 ± 39			



**Table 2** For each soil sampling campaign: the PET on that sampling day; PET as average over the 30 days prior to the sampling day (PET<sub>30</sub>); lc-excess of the precipitation input weighted averaged over 7 (P<sub>7</sub>) and 30 (P<sub>30</sub>) days, respectively; precipitation summed over 7 (P<sub>7</sub>) and 30 (P<sub>30</sub>) days prior to soil sampling; total soil sample number; minimum, median, maximum, and interquartile range (IQR) of the soil water lc-excess data; slope and intercept of the evaporation line (note that regression is significant at the 99 % confidence interval); and mean gravimetric water content (GWC).

	PET		P lc-excess		P		Sample	Soil water lc-excess				Evaporation line		GWC
	[mm d <sup>-1</sup> ]		[‰]		[mm]		[-]	[‰]				Slope [-]	Intercept [‰]	[-]
Date	PET	PET <sub>30</sub>	P <sub>7</sub>	P <sub>30</sub>	P <sub>7</sub>	P <sub>30</sub>	n	min.	median	max.	IQR			mean
2015-09-29	2.7	1.4	-0.8	-1	1	66	91	-16.1	-5	1.1	4.2	4	-21.4	0.39
2015-10-20	1.2	1.1	-0.2	-0.7	2	60	78	-12.6	-4.1	5.3	5.2	3.6	-23.3	0.37
2015-11-26	1.1	0.8	3.4	-0.2	14	94	80	-11.1	-3	6.5	3.9	4.6	-21.1	0.37
2016-01-13	0.4	0.6	0.4	-1.6	56	434	77	-8.2	-0.8	7.5	6	8.2	10.8	0.42
2016-03-18	0.6	0.8	-5.2	-2.1	2	33	79	-11.6	-1.3	6.7	5.4	5.5	-15.2	0.37
2016-04-27	0.9	1.2	7.3	-1	21	80	79	-7.7	-1.8	5.8	6.4	5.1	-16.2	0.40
2016-05-24	1.6	2.1	0.1	0.1	31	66	80	-10	-1.4	2.9	3.7	4.9	-17.1	0.36
2016-06-18	2.6	1.9	-1.1	0.5	67	105	80	-13.2	-3.2	5.9	5.2	4.5	-21.6	0.38
2016-07-26	2.2	2.3	-2.5	-2.5	40	98	72	-14.6	-4.5	3.2	5.1	4.6	-19.1	0.37
2016-08-23	1	2.1	-3.4	-3.6	12	54	64	-16.7	-6	2	5.1	3.8	-25.4	0.35
2016-09-23	0.4	1.9	5.3	-1.6	8	51	77	-21.7	-4.1	8.9	6.2	4.3	-19.9	0.33