

The authors did a good job in revising this manuscript and it reads much smoother now. It will be a useful contribution and potentially valuable new method to be considered. However, I still disagree with the claim of the authors that using high flow rates for the sampling are an advantage and their calibration of isotope data for obtaining soil and xylem isotope values is following the principle of equal treatment. (I attached my original questions and author responses down below).

We thank the reviewer for their favorable evaluation. We also thank the reviewer for being persistent on the important issues of flow rate and the principle of identical treatment. In the revised manuscript we made clearer that the high flowrate was applied to facilitate shorter filling times. While we consider this suitable within the scope of our study, we did not mean to make a general statement devaluating other, equilibrium-based methods. We rephrased the respective sentences.

Regarding the principle of identical treatment, we do think that the calibration and validation standards we prepared in our lab-based experiments were treated identical. Importantly, this refers to gas flow rate, matrix potential and temperature (and thus vapor concentrations). We admit that while the former is easy to control the latter two variables will inevitably change among samples and relative to the standards in the foreseen field application. Certainly, this needs extra attention prior to our method being field-ready. We regret missing this point earlier.

When using high flow rates, no full equilibration inside of the membrane is reached anymore. This membrane has a certain length and exchange area with the tree xylem or soil, and it is true that by changing the membrane length, the chance for equilibrium conditions, a prerequisite for all direct-vapor-equilibration methods in the past, can be reached. The authors claim that they do not need equilibrium conditions in their membrane, because standards are treated the same way and values corrected thereafter. But this argumentation has a major flaw: Yes, same flow rates are used for standards and samples, but in both xylem and soils the membrane sits in a porous media with a limited maximum water content (50% are rarely exceeded). In contrast, vapor exchange in the liquid water standard (or its headspace) will be much faster and isotopic equilibrium reached with much higher flow rates. As a result, semi-equilibrated vapor samples (from xylem or soil) are calibrated with fully or almost-fully equilibrated isotope standards; hence, the kinetic isotope effect on standard and sample is different. And this is why I do not agree that equal treatment principles apply here and suggest initially that this should've been tested. If one key principle of direct-vapor-equilibration methods - isotopic equilibrium - is violated, there needs to be a solid proof-of-concept in my opinion (and reviewer #1 also raised a similar concern in his initial comments). The revised manuscript does still not clearly address this sufficiently in my opinion. The authors mention the validation standards, but these also don't resolve the abovementioned concerns (because those are also based on exchange with liquid water).

Our standards were based on exchange with wet sand, not liquid water. We tried to make this point clearer in the re-revised manuscript. Nonetheless, we see your point of different matrix potentials potentially having different effects on kinetic isotope fractionation. We admit that this issue needs extra attention prior to our method being field-ready. We also stated this point in the latest version of our manuscript. Thank you for being persistent on this.

In the revised manuscript, the authors also say that the good repeatability of their measurements is a proof of the method; however, a good repeatability has nothing to do with obtaining the correct isotope value. On a side note, and because the authors mention the method: the chance for isotopic equilibrium using the stem borehole method is much higher because no constraining membrane is used and the contact area with the matrix is much greater. Also, equilibrium conditions are tested by determining relative humidity of the sample after passage through the borehole. I simply do not

understand why such a test was never performed with the WIP's (other than Volkmann & Weiler 2014) and is not regularly done when taking vapor samples.

We think that the good precision and accuracy was achieved due to similarity of calibration and validation standards in terms of (among others) probe size, thus vapor source area, and gas flow rate, thus vapor pickup time. Unfortunately, these parameters cannot be unified in the context of the borehole method. For that reason, we denied the applicability of our method for the stem borehole method.

Q: Section 2.1.: This chapter is testing the flow rates through the WIP's and the reader gets the impression those can be chosen arbitrarily; but this is not true. For both soils and xylem, equilibrium fractionation inside of the WIP is required.

A: We argue that identical conditions need to be fulfilled which we did. Our chosen flowrate was identical for all standards and samples. Given the reproducibility of isotope values from nonequilibrium vapor samples in our study, we think that flow rates through identically dimensioned WIPs do not need to facilitate isotope equilibrium. However, this would be an issue requiring extra attention when dealing with e.g. the stem borehole method on trees with heterogeneous diameters. We addressed this issue in the revised discussion.

Additional answer A2: In our study, we tested the necessity of equilibrium for obtaining matrix-bound water isotope data. In a controlled lab experiment, we were able to reproduce the isotopic composition of non-equilibrium based vapor using data from non-equilibrium based standards, which were similar in terms of temperature, vapor concentration and matrix potential of its source. This means, we demonstrated the feasibility of our non-equilibrium-based method under well-defined conditions. We see this as an advancement of the purely equilibrium-based approaches as it may potentially yield additional flexibility in the study design (enabling shorter filling times, higher sample throughput, higher temporal resolution). Regarding the stem borehole method we already denied the applicability of our method due to the inevitable inconsistencies in vapor source areas. Regarding soil vapor sampling, further tests should identify the influence of matrix potential (among others) on non-equilibrium sampling and find a suitable correction.

Q: If flow rates are too high, this requirement cannot be fulfilled anymore. Marshall et al. (2020) provided a way to calculate the maximum flow rate possible with their stem borehole method. However, this seems to not have been tested for WIP's, where the membrane might have an influence on the exchange times. If this was tested, it would be great to cite that or at least provide information on this issue.

A: This matter was tested by Volkmann et al. (2014, doi: 10.5194/hess-18-1819-2014). They optimized the probe dimensions and contact area of the porous tip of the probe to the flowrate demanded by the isotope analyzer. As they aimed at developing an equilibrium-based in situ method, they didn't test other flowrates than the one given by the instrument. We added this information to the revised manuscript. We are aware that we were beyond equilibrium in this study, but aimed at shortening the filling times in the field. We therefore additionally investigated the effect of increasing flowrates and were able to reproduce the intermediate values (validation standards)

Additional answer A2: We are aware of the work of Marshall et al (2020). However, in the context of our study, the concept of a maximum flowrate is not applicable, as we did not aim at equilibrium. Rather, we tested the necessity of equilibrium by producing non-equilibrium-based vapor and reproduce its isotopic composition via established calibration schemes. This turned out to be successful in a controlled lab experiment. Prior to field-readiness, further test will (have to) find ways to correct for the effects of inconsistencies in temperature and matrix potential.