

We thank Reviewer 1 for pointing out the weaknesses of the manuscript. Comments are answered below and a revised draft taking into all the reviewer's comments is presented as a companion file.

The reviewed manuscript presents an important work, which examines the robustness of the ^{17}O -excess of phytoliths as a proxy for RH. However, more work is needed to make the manuscript more readable. At the present form, I had troubles even verifying that the main conclusion are supported by the data. Moreover, I am not sure if there are quantitative conclusions, as vague terms like "most" are used throughout the text.

The text was rewritten to make the objectives, results, interpretation, conclusions and abstract clearer. Vague terms were avoided.

- The two questions to be dealt with are more clearly stated in the introduction: 1) whether grass anatomy diversity impacts the ^{17}O -excess of phytoliths vs RH relationship, 2) whether RH changes from day to night and from leaf elongation to leaf senescence should be considered when interpreting the ^{17}O -excess of phytoliths as a RH proxy.
- Two section (6.3 and 6.4) modelling the triple oxygen isotope composition of leaf water and phytoliths along the blade are added. They show that evolution with length of ^{17}O -excess of *F. arundinacea* leaf water and phytoliths can be predicted using the Farquhar and Gan (2003) model and considering a $\lambda_{\text{silica-water}}$ value decreasing from 0.522 to 0.520 from the sheath to the apical part of the blade. Despite of this heterogeneity, the ^{17}O -excess values of bulk leaf water and phytoliths can be estimated and are not length-dependent. Impact of the diversity in grass anatomy on the triple oxygen isotope composition of phytoliths is discussed in more details leading to the conclusion that it should not impact the triple oxygen isotope composition of bulk grass phytoliths.
- We now make clearer that as most of silica polymerizes at the end of the elongation stage (58 % in the present case), RH conditions leading to leaf senescence in nature should be considered in addition to RH condition during leaf elongation, when interpreting the ^{17}O -excess of phytoliths.
- Tracks for future research are given when inconclusive results are acquired (i.e. potential kinetic fractionation during phytolith formation, impact of day/night alternation on the triple oxygen isotope composition of phytoliths).
- A conclusion section entitled '7. Conclusions for interpreting the ^{17}O -excess of grass phytolith assemblages' is added to clearly answers the two questions presented in the introduction.

Tables 1 and 2, contains full details of the results, and as such are hard to follow. So additional figures and summary tables are needed for easier access to the data (and then the full data can be moved to an appendix).

- Tables 1 is simplified. Table 2 is moved to supplementary material. Two tables showing the modelled calculations for the triple oxygen isotope composition of leaf water and phytoliths along the blade are added (Tables S3 and S4).

Figure 2 should be divided into four different figures, each with its own caption. Will be good to explain in the caption what the filled versus empty markers stand for (this is also not explained in the legend).

Figure 2a, correct the Y-axis.

Figures were reworked. Only 3 figures are now presented:

- **Figure 1.** Growth chamber experiment 1 : SEM pictures of phytoliths from young, adult and senescent leaf blades : silicified Trapeziform short cell (1, 2 and 3), silicified Trapeziform sinuate short cell (4), undefined silicified short cell or broken Elongate cylindric long cell (5), silicified Elongate cylindric long cell (6a, 6b, 7 and 8), silicified cell wall also reported as silica sheets (9 and 10).
- **Figure 2. Growth chamber experiment 1 : a)** Phytolith concentration vs Long Cell phytolith proportion. Error bars represent the 5% error on counting (refer to text for details).
- **Figure 3. Growth chamber experiment 1. ^{17}O -excess vs $\delta^{18}\text{O}$ observed for leaf water (LW) and phytoliths (Phyto) in young adult and senescent leaves and along adult leaf (sheath, proximal blade, apical blade).** Predicted curves depicted for leaf water along the blade according to the Farquhar and Gan (2003) model (Table S4), assuming two possibilities for the evaporation process starting point: irrigation water (IW) and leaf sheath water (LSW). Predicted curves depicted for leaf phytoliths along the blade using $^{18}\alpha_{\text{silica-water}}$ after Dodd and Sharp (2010), $^{17}\alpha_{\text{silica-water}}$ calculated using $^{17}\alpha = ^{17}\alpha^{\lambda_{\text{Phyto-LW}}}$ with $\lambda_{\text{Phyto-LW}}$ set at 0.522 and decreasing from 0.522 to 0.520 from base to tip (Table S4). Craig and Gordon estimates for leaf water (LW) and phytoliths (Phyto)

(Table S3) assuming $\lambda_{\text{Phyto-LW}}$ of 0.522 and 0.521, and two source waters : irrigation water (IW) and sheath leaf water (SLW).

The second half of the abstract needs to be made clearer. Start with: Lines 29 and 31: "most" – give percentage. Line 32: "at that time" – replace with "during this growth phase" for clarity Line 33 – Replace "At least", by "when RH was fixed during day/night" However, when day/night alternations are characterized by significant changes in RH, the lowest RH conditions favoring evaporation and silica polymerization should be considered when calibrating the phytolith proxy." – Not clear. Do you mean the proxy is for the minimum diurnal RH? Also maybe need to add here what is the percentage of silica polymerization that happens under the lowest RH conditions.

The last sentence of the abstract should be removed or rewritten.

The abstract was rewritten for more clarity/accuracy:

Continental relative humidity (RH) is a key-climate parameter but there is a lack of quantitative RH proxies suitable for climate model-data comparisons. Recently, a combination of climate chamber and natural transect calibrations laid the groundwork for examining the robustness of the triple oxygen isotope composition ($\delta^{18}\text{O}$, $\delta^{17}\text{O}$, ^{17}O -excess) of phytoliths as a new proxy for past changes in RH. However, it was recommended that besides RH, additional factors that may impact $\delta^{18}\text{O}$ and $\delta^{17}\text{O}$ of plant water and phytoliths be examined. Here, the effects of grass length, leaf development stage and day/night alternations are addressed from the growth of the grass species *F. arundinacea* in climate chambers. Plant water and phytoliths are analyzed in $\delta^{18}\text{O}$ and ^{17}O -excess. Silicification patterns are assessed. In the experimental conditions, day/night alternations do not modify the triple oxygen isotope composition of leaf phytoliths. However, additional monitoring for low RH conditions should be performed before withdrawing any generalizable conclusions. Evolution with length of ^{17}O -excess of *F. arundinacea* leaf water and phytoliths can be predicted using the Farquhar and Gan (2003) model and considering a $\lambda_{\text{silica-water}}$ value decreasing from 0.522 to 0.520 from the sheath to the apical part of the blade. Despite of this heterogeneity, the ^{17}O -excess values of bulk leaf water and phytoliths can be estimated and are not length-dependent. Overall, diversity in grass anatomy should not impact the triple oxygen isotope composition of bulk grass phytoliths. Our experiment additionally shows that as most of silica polymerizes at the end of the elongation stage (58 % in the present case), RH conditions leading to leaf senescence in nature should be considered, in addition to RH conditions during leaf elongation, when interpreting the ^{17}O -excess of phytoliths. These outcomes contribute to a more precise identification of the parameters to take into consideration when using the ^{17}O -excess of phytoliths as a proxy of RH.

Section 2: The authors give there two different definition for theta. Since they are not identical, one of them must be wrong. Indeed the second one is the definition for lambda (or slope) which they give later. Papers cited by the authors give these definitions: 1) Angert, A., Cappa, C.D., DePaolo, D.J.: Kinetic O-17 effects in the hydrologic cycle: Indirect evidence and implications. *Geochim. Cosmochim. Acta* 68, 3487–3495, 2004. 2) Luz, B., Barkan, E.: Variations of $^{17}\text{O}/^{16}\text{O}$ and $^{18}\text{O}/^{16}\text{O}$ in meteoric waters. *Geochim. Cosmochim. Acta* 74, 6276–6286. <https://doi.org/10.1016/j.gca.2010.08.016>, 2010. Please correct.

The section was corrected as follows:

In the oxygen triple isotope system ($\delta^{18}\text{O}$, $\delta^{17}\text{O}$), the fractionation factors ($^{17}\alpha$ and $^{18}\alpha$) are related by the exponent θ where $^{17}\alpha = ^{18}\alpha^\theta$ or $\theta = \ln^{17}\alpha / \ln^{18}\alpha$. For the silica-water couple and according to the Sharp et al. (2016) empirical equation 10, $\theta_{\text{silica-water}}$ equals 0.524 for the 5-35°C temperature range. For the water liquid-water vapour couple at equilibrium, θ_{equil} equals 0.529 for the 11-41°C range (Barkan and Luz, 2005). When evaporation occurs, a fractionation due to the vapour diffusion in air is added to the equilibrium fractionation, as conceptualized by the Craig and Gordon model (Craig and Gordon, 1965; Gat, 1996). θ_{diff} associated with this diffusion fractionation equals 0.518 (Barkan and Luz, 2007). When RH decreases, amplitude of the diffusion fractionation governed by θ_{diff} increases. While θ applies to a particular well constrained physical process, the term λ is used when several processes occur at the same time, the resulting fractionation in the triple oxygen isotope system can be formulated as following: $\lambda = \Delta'^{17}\text{O}_{\text{A-B}} / \Delta'^{18}\text{O}_{\text{A-B}}$ with $\Delta'^{17}\text{O}_{\text{A-B}} = \delta'^{17}\text{O}_{\text{A}} - \delta'^{17}\text{O}_{\text{B}}$, $\Delta'^{18}\text{O}_{\text{A-B}} = \delta'^{18}\text{O}_{\text{A}} - \delta'^{18}\text{O}_{\text{B}}$, $\delta'^{17}\text{O} = \ln(\delta^{17}\text{O} + 1)$ and $\delta'^{18}\text{O} = \ln(\delta^{18}\text{O} + 1)$. δ and δ' notations are expressed in ‰ vs VSMOW. In the $\delta'^{18}\text{O}$ vs $\delta'^{17}\text{O}$ space, λ represents the slope of the line linking $\Delta'^{17}\text{O}_{\text{A-B}}$ to $\Delta'^{18}\text{O}_{\text{A-B}}$.

Also in Line 117, a reference line is defined not only by slope, but also by a point it is going through – add

this info.

The equation of the Global Meteoric Water Line was added (L112-113): (Global Meteoric Water Line expressed as $\delta^{17}\text{O} = -0.528 \times \delta^{18}\text{O} + 0.33$ per meg, Luz and Barkan, 2010).

Line 287 – " Isotope data from experiment 2b (Table 2) shows that D'18OPhyto-IW and 17OexcessPhyto-IW values obtained for the same tank are very close when constant day conditions of light, temperature and RH or when day/night alternation conditions are set." – This seems to contradict what appear in the abstract? So which one is correct? Do day/night variations in RH have an effect or not? Also, throughout the results, please don't just use the terms "very close" without giving the numbers, or indicating if the difference is significant or not.

Text and abstract were made clearer: in the experimental conditions, day/night alternations do not modify the triple oxygen isotope composition of leaf phytoliths. However, additional monitoring for low RH conditions should be performed before withdrawing any generalizable conclusions.

Line 374 – If it is only "52%", why this will dominate the signal? Line 399 – "mainly occur" – give a number. Line 414 Quantify "most" and point to a figure or reference showing this. Line 426 – "Likely Favor"? – Still not clear to me. So what is the 17Oexcess of phytoliths actually tell us? If the bottom line of this study is that more research is needed to answer this question, then this should be clearly written in the abstract and conclusions.

As previously stated, the text was rewritten to make the objectives, results, interpretation, conclusions and abstract clearer. Vague terms were avoided. Overall, the presented data and estimates contribute to a more precise identification of the parameters to take into consideration when using the ^{17}O -excess_{Phyto} vs RH relationship previously obtained (Alexandre et al., 2018).

