We would like to strongly thank all referees for their thoughtful comments and their appreciation of the paper. The paper has strongly benefited from the suggestions. All minor revisions and rephrasing were accepted as suggested by the reviewers. The text was also checked by a native English speaker. In this response, we present a detailed overview of our responses to all comments.

Anonymous Referee #1

Received and published: 14 March 2017

The study of Unzué-Belmonte et al. is very interesting, well written and based on very new techniques. It combines the old story of soil element depletion by erosion and negative effects of deforestation on soil Si pools shown by Struyf et al. 2010. However, despite the manuscript being well written and the data being well discussed I have two major concerns about the manuscript.

major concerns: The first is the complete missing of any statistical analysis. Without statistical analysis it is a descriptive description but nothing is really proven.

We now include a statistical comparison of average BSi contents of pits from the same position, and also a comparison between the top and the bottom pit from the same slope (accumulation), all for the biogenic AlkExSi pool.

We would like to stress that we opted to study a limited amount of soil pits in detail, rather than studying a larger amount of pits in less depth detail. This way, we can provide first insights in both spatial and depth patterns. This however limits the ability to compare Si pools at certain depths within the profile.

The second concern is that the authors confound the source of available Si in soils (line 34, line 48, line 383). It is only to minor share the phytoliths which have a high silica condensation state and hence a very slow dissolution. But a much more important source as part of the plant material are the amorphous plant silica deposits like the silicon double layer which has a very low condensation state and is thus highly dissolvable. Phytoliths are more or less stable in nature under common soil conditions. You mentioned it in line 389-391 by indicating phytoliths as a permanent sink for carbon as others found. Phytoliths are not easily dissolvable in soils, otherwise they would not be there for geological scales. Phytoliths are paleo indicators!

Although it is true that flora reconstruction based on phytolith preservation is a common paleoecological tool (Kirchholtes et al., 2015; Rovner, 1971) there are several studies that confirm the higher solubility of phytoliths compared to non-biological solid Si phases in soils (Fraysse et al., 2006; Lindsay, 1979; Ronchi et al., 2015; Sommer et al., 2013).

The large study of Piperno (2006) about phytoliths clearly shows how variable and how species dependent phytolith characteristics are. Solubility thus is species dependent (Alexandre et al., 1997; Blecker et al., 2006; Wilding and Drees, 1974). The solubility of phytoliths is further affected by pH, aluminum or Ca²⁺ concentration in the soil and parent material (Melzer et al., 2012). Cabanes and Shahack-Gross (2015) showed how phytoliths are only partially dissolved in soils. Fraysse et al. (2009) described two different Si pools within the plant: the phytolith pool and Si complexed with the organic matter from the cell walls. Moreover, different condensation states were found in phytoliths depending on the location within the plant (Schaller et al., 2013) which, as suggested by the referee, can result in different solubilities in soil. Borba-Roschel et al., (2006) showed a selective dissolution of phytoliths with depth from the Cyperaceae family. Alexandre et al. (1997) described how 92% of the BSi in top soil is rapidly recycled, while only 8 % seems to be permanently stored due to a lower turnover. In our present work we consider the biogenic AlkExSi pool as one single pool, but clearly there will likely be more soluble and less soluble BSi in soil. We have now included these thoughts in the discussion: Lines 332-338.

line 116: Sit and Alt should have the same style like in the formula

Changed in Line 128.

Table 1 should go for supporting information

We moved the Table to the Supplementary data.

line 260: "Highest concentrations were mostly found at the bottom of the slope in every site" this is really no new fact and should not be highlighted that much

Rephrased in Line 215. "In general, the distribution of biogenic AlkExSi shows the same pattern within each pit: the concentration decreases with depth and highest concentrations are found at the bottom of the slope (with the exception of the gentle slope of the cropland)".

Figure 3 and line 379-380 you found no effect of deforestation on biogenic Si pools for gently slop. This is an important outcome contradicting the paper of Struyf et al. 2010. Deforestation is not that bad for soil Si pools, at least for low slopes. This should be highlighted more both in the abstract and conclusion. We rephrased Lines 21-23 from the abstract in order to be more moderate in saying that land use change depletes the biogenic AlkExSi pool in a gentle slope: "Our study shows that deforestation can rapidly (< 50 years) deplete the biogenic AlkExSi pool in soils depending on the slope of the study site (10-53%), with faster depletion in steeper sites."

It is true that the difference between the biogenic AlkExSi pool of the gently sloped forest and the gently sloped cropland is almost absent in our results, which indeed is in contrast with results from Struyf et al (2010). However, we also point to the fact that this might be due to the recent deforestation: 50 years might not be enough to see the decrease in the terrestrial biogenic AlkExSi pool.

We explicitly mentioned the apparent contrast with Stuyf et al. (2010) in Line 272-275: "Our results are apparently in contrast with results from Struyf et al. (2010) who showed a large reduction in DSi export after deforestation in croplands deforested >250 years ago. Nevertheless, the absence of a larger decrease in the gently sloped cropland may indicate that deforestation occurred too recently to see such a decrease, only triggered by harvest."

We would again like to thank you for providing the opportunity to substantially improve our manuscript, and we hope that our paper, which is the first to combine land use change and erosion in the study of terrestrial biogenic Si in subtropical soils, will be accepted for publication in Solid Earth.

Yours sincerely,

Dácil Unzué-Belmonte

Corresponding author

Alexandre, A., Meunier, J.-D., Colin, F. and Koud, J.-M.: Plant impact on the biogeochemical cycle of silicon and related weathering processes, Geochim. Cosmochim. Acta, 61(3), 677–682, doi:10.1016/S0016-7037(97)00001-X, 1997.

Blecker, S. W., Mcculley, R. L., Chadwick, O. A. and Kelly, E. F.: Biologic cycling of silica across a grassland bioclimosequence, Global Biogeochem. Cycles, 20(May), 1–11, doi:10.1029/2006GB002690, 2006.

Cabanes, D. and Shahack-Gross, R.: Understanding fossil phytolith preservation: The role of partial dissolution in paleoecology and archaeology, PLoS One, 10(5), doi:10.1371/journal.pone.0125532, 2015.

Fraysse, F., Cantais, F., Pokrovsky, O. S., Schott, J. and Meunier, J. D.: Aqueous reactivity of phytoliths and plant litter: Physico-chemical constraints on terrestrial biogeochemical cycle of silicon, J. Geochemical

Explor., 88(1-3), 202-205, doi:10.1016/j.gexplo.2005.08.039, 2006.

Fraysse, F., Pokrovsky, O. S., Schott, J. and Meunier, J.-D.: Surface chemistry and reactivity of plant phytoliths in aqueous solutions, Chem. Geol., 258(3–4), 197–206, doi:10.1016/j.chemgeo.2008.10.003, 2009.

Kirchholtes, R. P. J., van Mourik, J. M. and Johnson, B. R.: Phytoliths as indicators of plant community change: A case study of the reconstruction of the historical extent of the oak savanna in the Willamette Valley Oregon, USA, Catena, 132, 89–96, doi:10.1016/j.catena.2014.11.004, 2015.

Lindsay, W. L.: Chemical equilibria in soils, Wiley, New York. [online] Available from: http://soils.ifas.ufl.edu/lqma/SEED/CWR6252/Handout/Chemical equilibira.pdf, 1979.

Melzer, S. E., Cadwick, O. A., Knapp, A. K. and Kelly, E. F.: Lithologic controls on biogenic silica cycling in South African savanna ecosystems, , 317–334, doi:10.1007/s10533-011-9602-2, 2012.

Piperno, D. R.: Phytoliths: A Comprehensive Guide for Archaeologists and Paleoecologists, Altamira Press, San Diego., 2006.

Ronchi, B., Barão, L., Clymans, W., Vandevenne, F., Batelaan, O., Govers, G., Struyf, E. and Dassargues, A.: Factors controlling Si export from soils: A soil column approach, Catena, 133, 85–96, doi:10.1016/j.catena.2015.05.007, 2015.

Rovner, I.: Potential of opal phytoliths for use in paleoecological reconstruction, Quat. Res., 1(3), 343–359, doi:http://dx.doi.org/10.1016/0033-5894(71)90070-6, 1971.

Schaller, J., Brackhage, C., Paasch, S., Brunner, E., Bäucker, E. and Dudel, E. G.: Silica uptake from nanoparticles and silica condensation state in different tissues of Phragmites australis, Sci. Total Environ., 442, 6–9, doi:10.1016/j.scitotenv.2012.10.016, 2013.

Sommer, M., Jochheim, H., Höhn, a., Breuer, J., Zagorski, Z., Busse, J., Barkusky, D., Meier, K., Puppe, D., Wanner, M. and Kaczorek, D.: Si cycling in a forest biogeosystem – the importance of transient state biogenic Si pools, Biogeosciences, 10(7), 4991–5007, doi:10.5194/bg-10-4991-2013, 2013.

Wilding, L. P. and Drees, L. R.: Contributions of forest opal and associated crystalline phases to fine silt and clay fractions of soils, Clays Clay Miner., 22, 295–306, doi:10.1346/CCMN.1974.0220311, 1974.