

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 #2

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The paper “Land use change affects biogenic silica pool distribution in a subtropical soil toposequence” addresses an important question on which there is so far not much data available, i.e. what changes occur in different Si pools in soil as a consequence of (anthropogenically induced) environmental changes. While the only totally new aspect is the subtropical site studied, the concept is relevant as anthropogenically induced changes in land use and erodibility of soils will, very probably, increase further in subtropical areas. The paper (title, abstract, scope, conclusions) also delivers more or less what it promises: some more work is however needed on the analysis and presentation of the results. The subject is well situated within the scope of the publication. It is to the authors’ credit that they use a fairly novel and under-utilized methodology which makes it easier to differentiate between biogenic and non-biogenic, ecologically and non-geologically speaking potentially relevant Si pools. The description of the methodology is also (to my, admittedly, fairly well prepared) mind quite adequate and would enable reproduction (with the proper equipment). The field study appears to have been well conducted, although the crops cultivated on the steep and less steep slopes should ideally have been more similar: in this section some more effort to ensure that everyone understands where what samples were taken could have been shown (see specific comments below).

We tried to clarify in the text where the sampling points were located. The selected pits were marked in Fig. 2 and the ordering of the Result section has been modified in order to improve the comprehensiveness and the flow of the text. We took the suggestions into account and provide more detailed responses below.

The main weaknesses of the paper are the absence of explicit statistical analyses, the presentation of the results and the discussion of these: the authors could and should make these sections substantially clearer and improve the quality and readability of the paper quite a lot this way. The introduction and most of the methodology sections are quite well written linguistically, but unfortunately the quality of both the text and the structure of the presentation deteriorates towards the end. The results are very likely indeed sufficient to support the interpretations and conclusions, but this should be proven by some kind of statistical analysis (even the use of simple t-tests would improve the paper considerably). These interpretations could also be presented in a clearer fashion (and perhaps the authors would

themselves get more out of the results). The citation of existing literature on the subject is adequate (bearing in mind that it is limited). I think that the paper is well worth publishing as it presents relevant results obtained by appropriate methods and draws fairly good conclusions, but I would recommend that statistical tests are added and quite a lot of the text rewritten.

We answered about the statistics in the response to the previous reviewer:

“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”.

We worked on the flow of the writing and had the manuscript reviewed by a native English speaker that made several changes along the whole manuscript.

Specific, technical comments:

The Abstract and Introduction are both well written. In the Methods-section, it is not quite clear to me what sort of tillage is used (section 2.1 rows 82-).

The tillage used in the crop fields was quite intensive since deforestation occurred. More recently a minimum tillage and a cover cropping system were introduced. We adapted the text in Lines 82-90 in order to give a clearer explanation: “Deforestation occurred around 50 years ago and they have since experienced the same historical agricultural practices. Intensive soil tillage occurred from the time of deforestation to 2003, since when a cover cropping and a minimum tillage practice was introduced (Minella et al., 2014). The actual soil tillage is traditional, based on topsoil mixing and making ridges and furrows. Crops in the gently sloping cropland (maximum 7°) rotate between soybean (*Glycine max*) in summer and black oat (*Avena sativa*) in winter. Some cattle occasionally graze during the vegetative stage, and after the oat is harvested the biomass is left to produce mulch (cover) to soybean seeding based on the no-till system. The cropland of the steep slope (maximum 18°) rotates between tobacco (*Nicotiana sp.*) or maize (*Zea mays*) in summer and fallow or black oat in winter. The winter crop on this slope is also left behind to produce cover for the next crop”.

In section 2.2, please specify how the soils were dried (temperature?). You could perhaps refer to Fig. 2 here?

Included temperature (~40) in Line 98 and the reference to the Fig. 2 in Line 95.

In section 2.3, how were the representative pits selected? How were the rest used and why? Please make this clearer!

We rephrased Lines 102-104 in order to better explain the selection of the 'representative pits': "One pit per position was selected as a representative pit due to the impossibility of carrying out the novel alkaline extraction analyses on such a high number of samples (297), resulting in a total of 81 samples. The selection avoided pits containing large inclusions (visually) or pits shallower than the other two replicas".

I also hate acronyms and question whether you really need the complicated ones you have designed . . . ?

The acronyms used in the study tried to include the information needed to locate the pit in the study schema. Land use type, slope and position in the slope should be given in the acronyms. We opted to change the acronyms for selected pits (removing the replicate number) and marked them in the Fig. 2 as well.

Please also check the overall quality of the language used (starting here); in many instances better words could be used (e.g. not differenced, row 118, smaller THAN or equal to, row 124, nearly linearly dissolving, row 125).

Changed in Lines 135, 141 and 142 respectively. The manuscript was reviewed by a native English speaker.

Starting from section 2.3.1 the readability of the text also deteriorates somewhat: please consider that this is not a list but a paragraph (e.g. line 127). The paragraph starting on line 151 is, in particular, very raw text (with even capitalization completely haywire) and should be entirely rewritten. Similar specific problems occur throughout the discussion e.g. line 288, 306, 325,328,329, etc.): please have a native speaker check the language once more!

Changes suggested were made. We had the manuscript reviewed by a native English speaker, who made several changes along the whole manuscript.

I would also recommend that you re-structure sections 2.3 and 2.3.1 and separate e.g. (laboratory or physicochemical) analyses and calculations into separate sections – it is not that relevant which analyses were made on selected pits and which not. The section on averaging and accumulation is now very hard to follow.

Ordering of the contents in section 2.3 has been changed. Subsections are now as follows: 2.3.1 Physicochemical analyses, 2.3.2. Alkaline continuous extraction and 2.3.3. Post-data treatments.

In the Results-section, I would put the supplementary data on AlkExSi-concentrations either together with the bulk data (isn't what is presented in section 3.1 minearology, besides, rather than physico-chemical characteristics?) or after the main results (the pools) as a "footnote". Now it confuses the reader, or at least me: why first concentrations and then pools?

Concentrations were determined through an extraction and curve modeling afterwards. Pools include interpolated data which makes them an estimation. However, this estimation is needed in order to be able to compare samples to each other, as it includes the humidity of each sample or bulk density.

The listing of averages starting on line 182 is not necessary: you already have them in Table 2! Do a general overhaul of the results section and check that what you mention in the text really is necessary and helps the reader to understand what you are talking about, not only a list of numbers.

List of averages has been removed and Result section has been adapted accordingly. Lines 185-208.

I would put your result summary into the discussion!

We changed the location of the Result summary to the beginning of the Discussion.

Discussion: section 4.1 is really depth distribution within the soil, isn't it? Could the section header reflect this?

Section header was changed for: "Redistribution of AlkExSi concentrations in depth and along the toposequence".

And why don't you start with the comparison to other studies now starting at line 240? Likewise, I would begin section 4.2. with a separate paragraph containing the sections on bulk numbers in cropland and forest now within lines 250-253 and 268-272, with or without a separate heading.

We clearly separate concentrations from pools in the Results and Discussion. As mentioned before, concentrations are directly measured in the samples and pools include some interpolation. Section 4.1 is about concentrations found in depth and along the toposequence. We compare our results with other comparable results from the literature. Section 4.2 is about the pools. We proceed in the same way comparing our result with the existent published works. Then we discuss the differences found between the different studied sites.

The section on crop rotation (line 258-262) is perhaps more land use change than erosion?

We worked again all over the Discussion section with the objective of improving the readability and the flow of the text. We discarded the headings because we consider that both effects, land use change and erosion, are so interrelated that discussing the two effects separately will distort the message that both together have the strongest impact.

Don't you have any references for the effects of erosion on Si pools in forest (line 273-)?

Studies about Si pools in soils under forest analyzed with a comparable method do not exist as far as we know.

Section 4.3. is relevant (but check the language) but would fit better into the discussion later, before the Implications. These are OK; I would perhaps also mention the changes in the non-biogenic pools.

This study is about the effects on the biogenic AlkExSi pool, which is probably the most effective Si pool that supplies DSi into the soil. Thanks to the method used here we are able to distinguish the biogenic from the non-biogenic AlkExSi. Including a section explaining the benefits of this method (and concerns)

is only to demonstrate that all the non-biogenic fractions would have been considered biogenic if other extracting methods were used. Because of that, we give some information about the non-biogenic pool, but it is not the object of study.

Limitations exist in characterizing the non-biogenic AlkExSi. The large variability of compounds that might have been dissolved in NaOH and characterized in the fractions with $Si/Al < 5$ make the interpretation of that pool beyond the scope of this manuscript. We would like to remind that the biogenic and non-biogenic AlkExSi fractions described by the method and discussed in the manuscript are only the fractions that showed a first order dissolving behavior. No minerals or linearly dissolving fractions are commented in this work. This means that the oxides and clay mineral described in that pool are only part of the total pool of oxides and clays present in the soil.

We included Lines 303-305 in order to better introduce the following section about the non-biogenic AlkExSi pool.

I like your figures, but please ensure that all numbers are of readable size! Especially in Fig. 4 with green background the numbers could be clearer.

New graphs were made.

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,

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Minella, J. P. G., Walling, D. E. and Merten, G. H.: Establishing a sediment budget for a small agricultural catchment in southern Brazil, to support the development of effective sediment management strategies, *J. Hydrol.*, 519, 2189–2201, doi:10.1016/j.jhydrol.2014.10.013, 2014.