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## Interactive comment on "Anthropogenic impact on biogenic Si pools in temperate soils" by W. Clymans et al.

## Anonymous Referee #1

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The study attempts to assess the anthropogenic perturbation of soil silica pools within the temperate zone and their effect on land-ocean silica transfer. Soil storages of particulate biogenic silica (BSi) can be substantial and have been identified to be of potential importance for land-ocean silica transfer as silica cycling within terrestrial ecosystems, i.e. between soil and plant storages, can be as high or even higher than the net release of dissolved silica (DSi) from the terrestrial to the fluvial system, that in most study has merely been attributed to the chemical weathering of bedrock. The assessment of net changes in these silica pools and their contribution to contemporary fluvial DSi fluxes are now an important research question. The topic of this study fits well within the scope of Biogeosciences. Generally, the manuscript is not too long. The language is fluent and precise. Abbreviations used are clearly defined. However, before it can be considered for publication some modifications and clarifications of

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the present manuscript are necessary. With regard to methodology, the study can be divided into two main parts. The first part comprises field and lab work and attempts to assess the anthropogenically induced reduction in soil BSi stocks for two close by study sites in Southern Sweden. In the second part of the study, rough first order estimates of the anthropogenically induced reduction in soil BSi stocks within the whole temperate climate zone and their contribution to global fluvial DSi fluxes are made by applying the results of this local scale study to a data set on modeled global land use change over the last 5000 years. For this second part, the rather non-realistic assumption had to be made that the study area in Southern Sweden would be representative for the whole temperate zone. However, as this is the first study which attempts such an assessment, the authors' considerations in this regard are of interest for the scientific community. In the following, I'd like to comment on these two parts separately before I give a some technical comments.

1st Part: Antropogenically induced soil BSi loss on the study sites On the two study sites, BSi pools have been determined for altogether 29 soil profiles covering four different land use types for which different degrees of anthropogenic impact on the soil BSi stocks could be assumed. One site merely covers continuous forest which is assumed to represent a rather pristine ecosystem and the climax of the natural succession. The other site covers the three different land-use types, in the following order with increasing anthropogenic impacts assumed: grazed forests, pasture and arable land. The field work and lab methods are clearly described and are based on a well established methodology (DeMasters). Promising seems the approach to determine the pool of total armorphous silica PSia, which is thought to represent soil BSi, and the pool of easily soluble amorphous silica PSie. The anthropogenic impact on the soil BSi pools is assessed comparing average concentrations and stocks of PSia and PSie of the four land use types assuming that all other important environmental parameters, i.e. soils, hydrologic setting and climate, are similar. Further, it is assumed by the authors that soil erosion had no significant effect on the soil profiles. The authors hypothesized anthropogenic land use results in long-term decreases in BSi stocks due increased

BSi dissolution and leaching while replenishment by plant litter is hampered. Indeed, results show that for the "continuous forests" the BSi stock in the soils is more than twice as high than for grazed forest, pasture and arable land. However, for that latter three land use types no significant differences in PSia (=BSi) stocks which could be interpreted in terms of different degrees of anthropogenic impact on these BSi stocks could be identified. Within the topsoil down to 25cm, for arable land, for which the decreased in BSi replenishment should be highest, the BSi stock is even substantially higher than those of pasture and grazed forest. By the authors it was hypothesized that this was due to BSi stemming from a root network of the crops which would be comparably dense. I would like to suggest adding following information:

- What crops are grown on the arable land in the study area? What is the common crop rotation? - As soil BSi stocks are replenished by plant litter and BSi contents of plant tissue might vary substantially between plant species, it would be appreciable if the authors could state the dominant plant species for each of the land use types (in the manuscript only the two dominant tree species of the continuous forest are given). - How are the soils in the study area to be classified according to an international soil classification system (FAO)?

The difference between the average BSi stocks of the rather pristine continuous forest and the anthropogenically impacted land use classes is assumed to represent the anthropogenically induced depletion. Further assuming that the anthropogenic impact started about 5 centuries ago and caused constant annual depletion rates since then, the authors deduce a decrease in soil BSi stocks of  $86.7\pm51$  kg SiO2 ha-1 yr-1. As the authors expect that soil erosion is not of significance, they conclude that the loss of BSi is due to net dissolution and flushing of DSi to the fluvial systems. As it can further be assumed that net dissolution of soil BSi represents only a part of the DSi flushed into rivers (silica weathering is the other source), the resulting fluvial DSi fluxes should be even higher. However, compared to what is known from the literature, this net-release of DSi from the soil BSi stocks seems to be unreasonably high. The highest specific

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DSi flux for Swedish rivers has been reported to be about 2300 kg Si km-2 yr-1 (Humborg et al., 2008), what is less than 50 kg SiO2 ha-1 yr-1. This disagreement with the study by Humborg et al. (2008) is striking. I suggest the authors to discuss this in their study. As the annual BSi loss calculated in the manuscript seems surprisingly high, some alternative explanations must be taken into account:

- The net release rate of BSi had not been constant over the 5 centuries and is currently rather low. - There are other sinks for the net release of DSi from the soil BSi pools within the terrestrial systems (e.g. precipitation of opals or adsorption in underlying sediments). - Soil erosion did play an important role in the land use history of the study area. (Are there any alluvial sediments of the Holocene which might stem from the study sights or similar areas?)

I suggest considering these points in the discussion as well.

2nd Part: Estimating the anthropogenic impact on the soil BSi stocks of the temperate zone and the resulting alteration of fluvial DSi fluxes at the global scale In the manuscript, methods and results of this part are given in the discussion (section 4.3). However, the results of this part play a central role in the conclusion and the abstract of the manuscript. Thus, I suggest describing the methods of this part in the methods section and the results in the results section as it was done for the first part of the study. When giving the results in the abstract, it should be shortly stated that this is a rough first order estimate that is only valid for the assumption that the study site in Sweden would be representative for the whole temperate region. However, extrapolating the results from part 1 is questionable, as the estimated annual contribution from anthropogenically induced net decrease in soil BSi stocks to fluvial DSi fluxes seems doubtful. As already discussed by the authors, the annual contribution of net soil BSi dissolution to fluvial DSi fluxes is more than five times higher than the increase in DSi fluxes described for Hubbard-Brook experimental forest in the first years after clearcutting (Conley et al., 2008). Under these circumstances, it is hardly comprehensible why the findings from the Swedish study sites should be regarded as representative for

the whole temperate region. I suggest to apply the land-use change related increase rate of fluvial DSi fluxes from Conley et al. (2008) as well and compare it with the results based on the findings from the Swedish study sites. For the Swedish study sites, soil erosion was not expected to have a significant impact on soil BSi stocks. However, soil erosion plays a significant role in extensive parts of the temperate zone (eg. Syvitski et al., 2005) and has partly resulted in substantial soil losses over land use history. I suggest considering this very important fact in the discussion. In the manuscript it is stated that net dissolution of soil BSi in temperate zones contributes 20% of GLOBAL fluvial DSi fluxes. It would be interesting for the reader to know the proportion the net soil BSi dissolution and subsequent DSi mobilization within the temperate zone takes on the total fluvial DSi fluxes from TEMPERATE watersheds. The total fluvial DSi exports from temperate watersheds can be calculated from data given by (Dürr et al., 2011).

Please, state how the temperate zone is exactly defined in the study.

Technical comments: - p. 4397, row 8: Replace "non-crystalline amorphous silica" by e.g. "pedogenic opal" - p. 4400, line 14: there is something wrong with this sentence, omit one CSie - Fig. 4: It is hard to read the subscripts in the legend. I suggest to make the legend more readable. - Table 1: Continuous forest, 0-0.1 m: Does this represent surface layer of leaf litter? In case it does, I suggest addressing this layer separated from the soil profile. - Referring to soil BSi stocks, the authors give their results in mass per area (kg SiO2 ha-1 yr-1). Referring to global fluxes they give the numbers in moles (Tmole Si). I suggest using either molar or mass units. This would increase the interpretability and comparability for the reader.

Conley, D.J., Likens, G.E., Buso, D.C., Saccone, L., Bailey, S.W. and Johnson, C.E., 2008. Deforestation causes increased dissolved silicate losses in the Hubbard Brook Experimental Forest. Global Change Biology, 14(11): 2548-2554. Dürr, H.H., Meybeck, M., Hartmann, J., Laruelle, G.G. and Roubeix, V., 2011. Global spatial distribution of natural riverine silica inputs to the coastal zone. Biogeosciences. Humborg,

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C., Smedberg, E., Medina, M.R. and Mörth, C.M., 2008. Changes in dissolved silicate loads to the Baltic Sea - The effects of lakes and reservoirs. Journal of Marine Systems, 73(3-4): 223-235. Syvitski, J.P.M., Vorosmarty, C.J., Kettner, A.J. and Green, P., 2005. Impact of humans on the flux of terrestrial sediment to the global coastal ocean. Science, 308(5720): 376-380.

Interactive comment on Biogeosciences Discuss., 8, 4391, 2011.