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Discussions

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

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First of all, we are pleased that both reviewers agree that the manuscript is innovative and delivers important insights in a yet understudied aspect of global silica cycling: the impact of land use change and management on Si pool distribution in soils. The positive comments on the methodology applied, on the scientific interest of many biogeochemists and the conclusion that it is a valuable contribution to the scientific domain, encourage us to improve the manuscript according to all suggestions. All the suggestions of both reviewers will be taken into account in the final revised version of the manuscript. Below we have compiled our responses. We hope that the future revised version will be considered as responsive to the concerns of both reviewers and suitable for publication in Biogeosciences.

Comment1 R1: "As soil BSi stocks are replenished by plant litter and BSi contents of plant tissue might vary substantially between plant species" We agree with reviewer 1 that BSi content varies significantly between plant species (Epstein, 1999). For example, grass species are known to have high concentrations of silica, and therefore could be important for the dynamics of BSi in soils after deforestation or other human intervention (Blecker et al., 2006). Nevertheless, Kelly (1998) showed that forest stores a larger amount of plant-available silica compared to grasslands. We will add information on dominant plant species and crops on the arable fields, pasture and grazed forest of Råshult, and crop rotation.

Comment2 R1&R2: "Soil classification system (FAO)?" We understand the referees concern that conclusions of the effect of land use change on the Si-storage can only be true, if the assumption that soil type, parent material and climate are identical is correct. Therefore we have conducted extra analysis (Cation Exchange Capacity, CEC and Base Saturation, BS) on soil profiles for each land use to make classification possible. We classified each soil as a Cambisol according to FAO standards: typical sandy loamy texture, no profile development, presence of a cambic horizon, decreasing CEC with depth and a BS lower than 50% somewhere in the 1m below the soil surface. This proves that our results are not influenced by differences in soil type: we already excluded the influence of parent material and climate in the original manuscript. We will add the new results in table 1, and adapt the manuscript throughout.

Comment3 R1&R2: "Annual BSi loss calculated in the manuscript seems surprisingly high, 86.7±51 kg SiO2 ha-1 yr-1. ", "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." & "I can understand your assumptions about the magnitude of change in temperate continental BSi, in which the climatic impact factor is isolated. But, you cannot made such assumption for all the temperate climate as the Si global cycle is strongly impacted by soils and by vegetation." We have defined the temperate regions as: the land area in the North, between the Arctic Circle and the Tropic of Cancer, and the South, between the Antarctic Circle and the Tropic of Capricorn. This covers about 70% of the land surface. We will add this information to the manuscript. A major comment of both referees is the representativeness of our data for the whole temperate region, and the assumption that the BSi decrease in the soil pool can be regarded as a simple excess in exported DSi. We agree, for several reasons stated below, that this is a simplification of the reality as some important secondary pedological & biological processes responsible for dissolved silica concentration in soil solution and other sinks or loss processes are neglected. We will therefore emphasize these potential uncertainties in the discussion. Nevertheless, our first crude estimate is meant to be an eye-opener, providing valuable information for the ongoing discussion on the human influence on terrestrial Si-dynamics and the inherently changing land-ocean Si flux. Data on Psia and Psie are mostly available for natural systems, and little attention has been paid to human influenced systems. Therefore our study is the first to thoroughly address the effect of changing land use on biogenic Si-pools over a specific land use gradient. Here we summarize how we will address concerns regarding our estimate of the influence of land use change on global Si fluxes: We have included several factors that contribute to uncertainty in the discussion: (1) storage of both alkali-extractable silica (Psia) and easily soluble silica (Psie) in Swedish soils are considered representative for the whole temperate region (2) a decreasing Psia pool doesn't necessarily imply conversion to dissolved silica (DSi) or (3) net flux of DSi towards oceans. A major comment indicates that we extrapolated our data towards the whole temperate region considering land use changes from the HYDE 3.1 database (Klein Goldewijk et al., 2011). This implies that our data are representative for the whole temperate region. In the revised version we will now thoroughly discuss and emphasize that rather than delivering an exact number, the goal of our approach is to emphasize the potential magnitude of the influence of land use changes on terrestrial Si dynamics. Our data are collected for sites with similar climatalogical (e.g. vegetation, precipitation), geological (e.g. parent material) and pedological (e.g. soil type, hydrology) characteristics. All these parameters have

an influence on biological Si-cycling and Si-storage in temperate ecosystems (reviews of Sommer et al., 2006 and Cornelis et al., 2011 for a more extensive list). Our broad definition of the temperate region, and the resolution of the available land use data (i.e. per country) implies that climatology, geology and pedology can't be considered spatially uniform. This will be thoroughly emphasized throughout the manuscript Total land-ocean fluxes of DSi were estimated based on the annual loss of Psia in the soil. This implies that all BSi has been dissolved and leached out of the soil, but this is a simplification of the processes present. Other processes can be regarded responsible for losses in BSi; (re)-precipitation of dissolved BSi as secondary clay minerals or less dissolvable pedogenic silica fractions (Lucas et al., 1993; Van Cappellen, 2003), direct uptake in the agricultural cycle by annual harvest, timber logging, hay-meadow and grazing (Vandevenne et al., 2011), losses through severe land degradation (Smis et al., 2010; Triplett, 2008) and soil erosion as well as sequestration within rivers & lakes (Humborg et al., 2002 & 2005; Schelske et al., 1983). Furthermore the aqueous chemistry of Si is regulated by a number of linked processes (overview Cornelis et al., 2011). The fact that not all "disappearing" BSi is exported as DSi, will be clearly mentioned in the revised version and the implications of this will be discussed.

We want to stress that the purpose of our up-scaling exercise was meant to illustrate the possible importance of reduced Si-storage on the land-ocean Si flux and the order of magnitude of the impact on Si dynamics. We delivered a first estimate based on available data. This is also appreciated and acknowledged by both reviewers in their comments. Because we can't make a more accurate estimation, we will tone down the conclusions with respect to the effect on land-ocean fluxes on temperate and global scale. We are convinced that the magnitude is representative for how much BSi has been lost and/or converted in temperate soils, and that this illustrates the importance it could have on Si-delivery to mainly aquatic systems.

Comment4 R1: "I suggest to apply the land-use change related increase rate of fluvial DSi fluxes from Conley et al. (2008)" A simplified calculation of excess silica loss based on the rate of Conley (et al., 2008) for temperate regions could result in an average annual net increase of 0.16 Tmol Si y-1. This value corresponds with 12% of the temperate load and 3% of the global load. The value retrieved from our Swedish study suggests significantly higher annual net losses of ca; 1.1Tmol Si yr-1 corresponding on temperate and global scale with 84% of the temperate load and 20% of the global load respectively. The large discrepancy can be explained by several factors (1) a smaller Psia pool present in the HBEF catchment (Saccone et al., 2007) (2) losses in HBEF are based on riverine measurements of DSi so precipitation and biomineralization are taken into account but particulate BSi transport is neglected and (3) perturbations in HBEF were only persistent for 3 years, after which regeneration was allowed. We now will discuss these important differences in the dynamics of Psia within the temperate region, as well as the processes that may induce differences between changes in BSi stocks and BSi delivery to rivers such as (re-)precipitation and bio-mineralisation etc. Our study indicates that large uncertainties exist mainly due to lack of studies concerning this topic, and we encourage further research towards the effect of human influences on bio-reactive Si pools. Hereby we suggest to take in account changes in dissolved silica fluxes, biogenic silica fluxes and changes in distribution and total silica pools in soils to disentangle the effect of human influence on terrestrial Si cycle and land-ocean flux.

Comment 5 R2: “Is it an increase of BSi exported and/or BSi dissolution, or is it a change of Si form (BSi dissolution and Si precipitation with other elements as secondary solid phases)?” The reviewer indeed makes an appropriate comment about the fact that it is difficult to decipher the processes responsible for the redistribution. In our manuscript we mention all different processes like preservation and physical transport, secondary mineral formation, Si adsorption and precipitation of pedogenic opal. We specifically target changes due to reduced input of BSi from vegetation in the soil as the plant-soil cycle is perturbed, and the mobilisation of BSi within the soil profile (e.g. leaching, physical transport and (re)-precipitation. These are important drivers for long-term losses. Transformation to other Si forms will play also a role, but

this correlates with a perturbation of the soil-vegetation continuum. A complex linkage of different processes, which needs to be investigated further, is beyond the scope the paper. Still we will clarify the presence of other controlling factors or processes to make to reader aware of the complexity.

Comment 6 R2: "alkali-extractable Si" or BSi, please standardize in your manuscript or redefine” We will adapt the BSi term, because it does not only constitute phytoliths and microscope plant residues (i.e. real biogenic fraction, see Sauer et al., 2006), but also a poorly-crystalline fraction and amorphous lithogenic and pedogenic forms (e.g. silica glass, silica sorbed on Fe and Al oxi/hydroxides). It could be assumed that the importance of poorly-crystalline fraction in the total pool can be neglected. The synthesis of allophane and imogolite fractions, mainly occurring in volcanic soils, is hampered due to the acidic conditions (pH: 3.3-4.7). Although other alkali-extracted silica fractions are not present in a biogenic form, they have generally a higher solubility then the mineral phase. This makes them more easily available for biological (re-)cycling on shorter time scales (Sommer et al., 2006). Moreover Si adsorbed on Fe- and Al oxi-/hydroxides can be cycled by plants before sorption. This fraction partly controls the concentration of H_4SiO_4 in aqueous phase (Cornelis et al., 2011). If leached, it would be replenished by Si from phytoliths (Farmer et al., 2005). As the alkali-extracted silica here mainly corresponds with the amorphous silica fraction, we will standardise the alkali-extracted silica with the term amorphous silica (ASi) throughout the manuscript.

Comment 7 R2: “I don’t agree with your definition of the Si extracted by $CaCl_2$.” We will rephrase our definition of this easily soluble silica to plant-available silica without referring to phytolith origin. However, as hypothesized by Farmer et al. (2005), we do consider phytoliths as the main Si source controlling the final equilibrium concentrations of DSi in soil water. We agree that the control of phytoliths on the silica concentration in the aqueous phase depends on climatology and weathering stage (Cornelis et al., 2011), indicating that in non-weathered soils adsorbed silica also plays in important role. For our study site the lower CSie concentrations, and associated lower

Psie, indicate that the magnitude of BSi dynamics is less under arable land then under other sites, so that a lower Si-equilibrium stage is reached. We will change/rephrase our conclusions, and will indicate that we hypothesize that this mainly represents only dissolution of phytoliths, acknowledging other potential pathways.

Comment 8 R2: “the OC content is a proxy of the rate of organic matter input in topsoil and is not a direct chemical proxy of the BSi presence or absence.” We agree with the comment of reviewer 2. OC content can’t be regarded as a proxy for the presence or absence of BSi and we will not longer use the term ‘proxy’ in the revised manuscript. However, it makes sense to investigate how OC content and CSIa are related because we know that litter fall, and decomposition is an important source of both organic matter and biogenic silica, e.g. phytoliths, to the top-layer. We will argue in the discussion two reasons exist why this can’t be true for deeper layers: (1) there is no strict coupling between OM and biogenic silica and (2) different processes are responsible for the redistribution of OC and CSia. This implicitly means that once pedogenic processes are involved OC can’t be regarded as a proxy nor predictor. We will rewrite the paragraph to clarify this misunderstanding.

Comment 9 R2: “phytolith from roots decomposition? Other studies in specific plant species shown a very low content of Si in roots (see for instance Gérard et al. 2008).” In the present estimates, only the contribution of the above-ground biomass is considered due to the lack of data on below-ground productivity. This omission will inevitably lead to an underestimation of the total input. We wanted to address in our paper the possibility of extra input at various depths. Little is known about concentrations in roots and their turnover rate. Turnover rates have shown to be significantly different, between 10-56% annually, for roots in different vegetation types (e.g. trees, shrub, grassland) and for different root sizes (Gill & Jackson, 2000). Gérard et al. (2008) refers to a value measured in tree roots by Gordon & Jackson (2000), which we however couldn’t retrieve in this original paper. Still, Webb and Longstaffe (2000) estimated that roots and rhizomes contained up to 34% of the silica content of two grass species. Another paper

on grass species showed that the amount of opal deposited in the soil annually by root systems and above-ground parts is approximately equal in magnitude (Geis, 1977). An older review paper on ‘Silicon and plant growth’ (Lewin & Reimann, 1969) states that dependent on plant type, silicon can be uniformly distributed between shoots and roots, or accumulate in shoots or roots preferentially. Accurate measurements of root Si input for different plant species and for larger scale ecosystems are lacking. We will include the root discussion because we want to clarify different processes which can be regarded responsible for difference in distribution. In current state-of-the-art, as shown above, roots cannot be excluded.

Technical comments: We will address all smaller concerns of both reviewers as well as technical comments (rephrasing, references, figure adaptions and smaller clarifications) in the final revised version.

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