

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

Anonymous Referee #2

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This manuscript is very well written, innovative and promising for the future global Si cycle studies and their relation with the other elemental biogeochemical cycles. It addresses an aspect of the Si cycling on Earth's critical zone that has not yet studied: the impact of land use management on the Si pool distribution in soils. This BGD paper is a valuable contribution to the recent interest in the Si cycle in terrestrial environments. The authors present a very interesting and original point of view about the terrestrial Si cycle, influencing by a sequence of anthropogenic disturbances which largely impact the distribution of the alkali-soluble Si content in soil. Besides primary and secondary crystalline silicates, Si also occurs as a readily soluble Si pool, including dissolved Si, adsorbed Si, amorphous silica (biogenic and pedogenic opal) and short-range ordered aluminosilicates (allophane and imogolite). The quantification of this pool is very important to better constrain the DSi output from terrestrial environments to hydrosphere.

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This land-ocean transfer of Si is crucial for the homeostasis of global elemental cycles such as N, P and C as demonstrated in numerous studies (sink of CO₂ through diatom bloom and silicate weathering). The objectives of this manuscript are thus essential for a better understanding of the biogeochemical cycle of Si and global change processes. The authors assess the decrease of “alkali-extractable” and “plant available” Si due to anthropogenic modifications of the land use management (forest, grazed forest, pasture and arable lands). The original results and the global model of the anthropogenic impacts on Si distribution fits very well into a growing scientific conscience that terrestrial silicon fluxes are largely influenced by the different soil-plant systems and opens interesting outlooks about the anthropogenic impacts on the Si dynamics in these soil-plant systems. However, I point some poor interpretations of the pedogenic processes influencing Si dynamic in the soil-plant system, besides the fact that there is no detailed soil description and classification allowing us to be sure that all the soils are identical. Some improvements must also be done in the conceptual description of the “biogenic Si” pool since the specific extractions do not allow deciphering exactly what we extract. Indeed, the alkaline extraction correspond to a “readily soluble pool” (adsorbed, amorphous and poorly crystalline Si) and the CaCl₂ extraction correspond to a “plant available Si” (dissolved Si) pool which is also extracted by alkaline dissolution. Moreover, it is well-known that the type of soil-plant system strongly influence the dynamic of Si between solid and aqueous phase via the source of Si (Si mobility/dynamic is dependent on soil type) and Si biocycling (Si recycling rates in the biotic system are dependent on the Si uptake rates). The scientific quality of the manuscript is suitable for Biogeosciences if the authors are willing to make a major revision of the manuscript.

Here below, some detailed comments for each part of the manuscript:

Abstract

-p4392, lines 1-5: The link between Si mobilization/storage and the dynamic of BSi is not well established. Please, explain the impact of the biological Si cycling on Si

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transfer from continents to oceans.

-p4392, lines 7-9: 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 make such assumption for all the temperate climate as the Si global cycle is strongly impacted by soils (Henriet et al. 2008, *Biogeochemistry*, 90, 209-223; Bartoli 1983 *Environ. Biogeochem. Ecol. Bull.*, 35, 469-476) and by vegetation (Hodson et al. 2005, *Annals of Botany* 96, 1027-1046; Cornelis et al. 2010, *Biogeochemistry*, 97, 231-245). In your manuscript, you have to tone down your conclusions/assumptions about the impact of the anthropogenic disturbance on the BSi dynamic in a scale of the temperate regions. The type of vegetation (as a pump which recycles Si in the soil-plant system) and soil (as a source for the pump) greatly influence the chemical equilibrium between solid and aqueous phases in soil and thus the dynamic of BSi in the soil-plant system.

-p4392, lines 15-17: In your interpretation of the different stock of alkali-extractable Si and plant available Si, you have to mention the other factors, such as preservation and particulate exportation, soil surface erosion, secondary mineral formation, Si adsorption onto Fe oxyhydroxydes, precipitation of pedogenic opal. . . , which can also influence the Si solubility and dynamic. Indeed, thanks to your data, it is impossible to decipher the origin of the decrease of BSi and Si(CaCl₂), besides the decrease of the input of phytoliths. 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)? Thus, please tone down your assumptions/conclusions as your observations are not sufficient to assume that the mobilization is the only principle responsible of Si dynamic in soil system.

Introduction

-p4392, lines 24-25: the first sentence is not clear, please rephrase. What do you mean by “biological processes”, in continents (plant uptake) or oceans (diatom uptake)?

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-p4393, line 6: for the importance of the Si biological cycle, please add the reference of Derry et al. 2005, *Nature* 433, 728-731

-p4393, lines 10-13: I don't agree with the fact that the Si uptake by vegetation and BSi dissolution controls the DSi outputs in rivers. Indeed, numerous recent studies show that the Si transfer to oceans is clearly influenced by bio/pedo/geo-logical processes. Here you have to see/mention reviews of Sommer et al. 2006 *Journal of Plant Nutrition and Soil Science* 169, 310; Street-Perrott et al. 2008, *Earth Surf. Proc. Land* 33, 1436-1457; Cornelis et al. 2011 *Biogeosciences* 8, 89-112; and the paper of Meunier et al. 2010 *Geoderma*, 159, 431-439) which demonstrated that the impact of BSi on DSi fluxes is specific to some environments (for instance, tropical environments where the stock of weatherable minerals is depleted), and other environments can be influenced by the weathering of primary minerals.

Materials and methods

-p4395, line 3: the assumption about the impact of land use could be right if you assume that soil type, parental material and climate is strictly identical between the experimental sites. Thus that is crucial to have more detailed pedological analyses to allow classifying your soils with the WRB classification system.

-p4397, lines 7-11: there are some confusing about what you extract with Na₂CO₃ dissolution. The alkaline digestion, used here, extracts biogenic opal (phytoliths and micro-organisms remains), pedogenic opal, adsorbed Si, poorly crystalline aluminosilicates and dissolved Si. Please, see in detailed the classification of Si components by Sauer et al. 2006, *Biogeochemistry* 80, 89-108 and Cornelis et al. 2011, *Plant and Soil*, 342, 369-378 and rephrase by changing the explanation of the pools extracted.

-p4397, line 11: here you mention correctly “alkali-extractable Si” as the pool extracted by Na₂CO₃ digestion and which incorporates the components here above mentioned. Why do you use the term BSi in the rest of the manuscript? Please standardize in your manuscript or re-define your BSi term.

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-p4397, lines 22-23: “extrapolating the Si release...(Clymans et al. 2011)” is not the right reference for this method. Please, for the extrapolation, mention De Master (1981) and/or Koning et al. 2002, *Aquat Geochemistry* 8, 37-67.

-p4398, lines 11-12: I don't agree with your definition of the Si extracted by CaCl₂. The Si-CaCl₂ is not from dissolution of phytoliths. This pool of Si represents the Si available for plant and thus soluble in soil solution. This mistake influences the interpretation of this pool in your manuscript. Please correct the definition and the subsequent conclusions.

-p4398, line 15: what's the reference of the CaCl₂-extraction?

Results

-p4400, line 14: I don't understand this sentence: “CSie in the top layer were lower in CSie than below”, mistake with the two CSie?

- Fig 3d: please use the same scale for X-axis.

Discussion

-p4402, lines 9-11: PSie is not only linked to BSi dynamic (polymerization/dissolution) but also to dissolved Si content, and equilibrium with Si adsorption and formation of secondary solid phases. Your interpretation is thus too weak.

-p4402, lines 18-19: the distribution can also be explained by reaction of the solid phases (organo-mineral complexation, adsorption onto the exchange complex of minerals and incorporation in the secondary clay solid phases).

-p4402, line 25: please add Alexandre et al. 1997; Sacconne et al. 2007 ; Cornelis et al. 2010 (*Biogeochemistry*) after Blecker et al. 2006. Please also add Meunier et al. 2008 *Mineralogical Magazine* 72, 263-272 “Terrestrial plant Si and environmental changes” for your interpretations about “soil-phytolith depletion due to agriculture?”

-p4403, line 13. . . : the OC content is a proxy of the rate of organic matter input in

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topsoil and is not a direct chemical proxy of the BSi presence or absence.

-p4403, lines 14-15: factors influencing DSi in soil solution: please see review of Sommer et al. 2006; review of Cornelis et al. 2011 and the book chapter Drees et al. 1989: *Silica in soils: quartz and disordered polymorphs in “Minerals in soil environments”*.

-p4403, lines 20-22: 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)

-p4404: lines 4-7: you forget to mention that a part of BSi can be dissolved and subsequently precipitated as secondary clay solid phases (see Lucas et al. 1993, *Science* 260, 521-523)

-p4405, line 24: your references for natural ecosystems are not complete.

-p4406, lines 4-6: please delete Henriot et al. 2008 which is not the accurate reference here. Please add Ziegler et al. 2005, *Geochimica et Cosmochimica Acta* 2005, 69, 4597-4610; Opfergelt et al. 2010 *Geochimica et Cosmochimica* 74, 225-240; and Cornelis et al. 2011 (review dealing with the impact of various soil-plant systems on the Si transfer to hydrosphere and using geochemical tracers such as Ge/Si ratios and stable Si isotopes).

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