## Historical land use change has lowered terrestrial silica mobilization (to the North Sea and the Scheldt Estuary)

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It has recently been shown that intense biogeochemical cycling of silica occurs in terrestrial soils. The annual ecosystem biogeochemical cycling of Si exceeds the annual export from the continents to the coastal zone by two orders of magnitude and effectively buffers weathering Si fluxes. Given the importance of the ecosystem biological Si filter, human impacts on vegetation cover through land use changes will significantly alter the biogeochemical silica cycle, although the impacts on the magnitude and fluxes of silica from land to coastal zones are presently unknown. Addressing this knowledge gap is essential as the terrestrial silica cycle is closely connected to i) important terrestrial and ocean carbon sinks and the ii) occurrence of harmful algal blooms in adjacent coastal ecosystems.

We showed that land use is the most important controlling factor on baseflow Si mobilisation in the river basin of the Scheldt Estuary, with historical soil disturbance and sustained cultivation (> 250 years) of formerly forested areas leading to two- to threefold decrease in baseflow delivery of Si from the land surface to the estuary. We propose a new conceptual model where initial deforestation leads to increased Si fluxes, with sustained soil disturbance depleting soil pools of amorphous silica, eventually leading to reduced Si fluxes. The Scheldt basin has been one of the most densely populated areas in Europe already since the 13th century. As early as 1250, only 10% woodland cover remained; in a pristine state the Scheldt basin was almost fully forested (>90 %). In this conceptual model, the present Scheldt basin therefore represents a new equilibrium state.

Despite limiting our study to base-flow conditions (60-90% of total yearly water fluxes) and the uncertainty associated with the timeframes in the new conceptual model, our data clearly indicate that long-term soil disturbance and a millennium of agricultural development has strongly changed biogeochemical Si dynamics in the Scheldt River basin (and likely other temperate river basins). Modern agricultural practices also result in increased input of N and P to the aquatic continuum, resulting in Si limitation in the Scheldt Estuary, with negative effects on ecosystem quality including the development of harmful algal blooms. Our new hypothesis suggests this DSi limitation in aquatic systems could be counteracted initially by increased Si export from cultivation of former forest soils. As the soil biogenic Si pool gradually declines, this counter-effect diminishes and Si mobilization becomes lower compared to pristine conditions. This implies that Si depletion events in more recently cultivated areas could currently still be masked by increased DSi export fluxes from recyclable biogenic Si pools. In the Scheldt watershed, lowered Si fluxes were already attained long before intense fertilization started, and no such masking was observed.

Our observations emphasize the need to increase our understanding of land use impacts on biogeochemical Si cycling. A millennium of soil disturbance after deforestation lead to 2-fold to even 3-fold decreases in TSi flux from a watershed where the adjacent coastal zone has experienced significant eutrophication problems due to changed N:P:Si river deliveries in the last three decades. Our results are the first to show that locally factors controlling terrestrial Si mobilization can be separated from factors important at continental and global scales, where controlling factors largely include lithology, precipitation and slope. We clearly show that land use should be included in watershed scale models for baseline Si mobilization. Our results shed new light on how historical cultivation has affected the terrestrial silica cycle, and indicate yet another anthropogenic reduction of silica fluxes through the aquatic continuum, adding to globally important reductions in riverine Si transport by deposition in reservoirs and in eutrophicated rivers and estuarine sediments.

The observation that long-term soil disturbance has lowered baseflow mobilization of Si from landscapes is novel and part of a complex puzzle linking Si, N, and P continental biogeochemical cycles to the ecological quality of coastal waters and C burial in the ocean. Transport of nutrients to the coastal zone and the ocean will be strongly dependent on processing within estuaries. The yearly average filtering capacity (the ratio of the net process rate to the total inputs) of the Scheldt Estuary is 0.12 for DSi, with strong seasonal fluctuations. In addition, tidal marshes will play an important role in estuarine Si fluxes, especially in summer. Ratios of processed nutrients in the estuary may also vary with discharge. Increasing discharge lowers diatom uptake of Si within the estuary, while increasing the net amount of TSi transported through the estuary. Yet, the average filtering capacity demonstrates that riverine inputs of Si are an important determining factor for eventual fluxes to the coastal zone, and a good understanding of the effect of land use in Si dynamics is therefore essential.