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Large-scale semi-distributed modelling of dissolved silica fluxes and assessment of their role in coastal eutrophication

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ABSTRACT

Dissolved silica (DSi) can play a significant role in primary production in coastal and marine ecosystems. This paper describes the silica module developed for the hydrology and water quality catchment model HYPE, and its application in Sweden. The model simulates weathering of silicate-containing minerals and processes in the surface water system where DSi may be used in the production of biogenic silica. The model was able to reasonably simulate levels of DSi across Sweden (mean relative error = 26.9%) while also capturing some of the temporal dynamics (mean Pearson correlation coefficient = 0.44). The calibrated model was used to quantify DSi fluxes across Sweden as well as to perform an assessment using the Indicator of Coastal Eutrophication Potential (ICEP). ICEP values calculated for 25 large Swedish rivers were all negative, albeit with a large spatial variability, indicating that Swedish rivers contribute to a favourable nutrient ratio in Swedish coastal waters.

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1 Introduction

Although nitrogen (N) and phosphorus (P) have historically been the focus of nutrients in surface water management, dissolved silica (SiO_2 , hereafter referred to as DSi) also has a significant impact on coastal and marine aquatic systems. The ratios of N and P to DSi impact not only primary production, but also the quality and succession of algal species (Turner *et al.* 1998, Danielsson *et al.* 2008). In contrast to N and P, DSi in aquatic systems is less influenced by anthropogenic activities. Whereas N and P in eutrophic surface waters stem primarily from treated or untreated waste water or excess fertilizers in agriculture, DSi originates to a large extent from natural land processes. However, climate change and human activities (e.g. agriculture, industrialization, urbanization, and the construction of dams and reservoirs) are altering the nutrient delivery ratios and nutrient limitation patterns of coastal systems, and this can have significant ecological and socioeconomic implications.

DSi fluxes originate from the physical and chemical weathering of silicate-containing minerals (Bluth and Kump 1994, Conley *et al.* 2000). Weathering rates are influenced by a variety of physical, chemical, and climatic factors, including: lithology (Meybeck 1986, Bluth and Kump 1994), mineral composition and grain size (Bray *et al.* 2015, Israeli and Emmanuel 2018), pH (Drever and Stillings 1997, Bray *et al.* 2015), physical erosion rates (West *et al.* 2005), continental weathering intensity (Murnane and Stallard 1990), temperature (White and Blum 1995, West *et al.* 2005, Li *et al.* 2016), and precipitation (White and Blum 1995, West *et al.* 2005). In

addition, weathering may be affected by vegetation both directly through the generation of chelating ligands, modification of pH, and alteration of physical soil properties, and indirectly by changing regional precipitation distributions (Drever 1994, Carey and Fulweiler 2012a). Depending on physical erosion rates, silicate weathering can become either transport limited or kinetically limited. When erosion rates are low relative to chemical weathering rates, chemical weathering is complete (i.e. minerals have been completely broken down through processes such as acidification, carbonation, dissolution, hydration, hydrolysis, and oxidation) and the weathering rate becomes dependent upon the supply of solids by erosion, or “transport limited.” However, when erosion rates are high relative to chemical weathering rates, there is an abundant supply of material and chemical weathering is incomplete. Thus, the weathering rate becomes dependent on reactions regulated by factors such as temperature, runoff, and vegetation, or “kinetically limited” (West *et al.* 2005).

Several processes affect the amount and speciation of silica as it is transported from headwaters to the sea. Of particular importance is the growth of diatoms, algal species which convert DSi into biogenic silica (BSi) as they construct their siliceous skeletons. Diatoms represent a significant source of riverine silica loading, especially during spring and summer diatom blooms in which BSi can account for 50–70% of total riverine silica export (Conley 1997). The growth of diatoms is highly affected by the availability of other macro nutrients, such as N and P, with nutrient enrichment leading to increases in diatom production and corresponding increases in total DSi

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consumption (Conley *et al.* 1993). Because BSi has a low remineralization rate and accumulates easily within sediments, the consumption of DSi by diatoms can cause an increase in the retention of silica within river sediments and a subsequent reduction in silica export to coastal areas (Muylaert *et al.* 2009). Silica fluxes are also strongly affected by the sedimentation and trapping of diatoms within lakes and reservoirs, with riverine DSi concentrations decreasing as the fraction of the lake/reservoir area within a watershed increases (Conley *et al.* 2000). In addition, inundations due to the construction of dams decreases soil–water contact and, consequently, reduces weathering rates and DSi export (Humborg *et al.* 2002). Further decreases in DSi fluxes to coastal zones are expected as the number of dams increases globally (Humborg *et al.* 2000).

The amount of silica delivered to the coastal zone may have a profound influence on the coastal ecosystems by affecting algal species composition and food web structures. As primary producers, phytoplankton, such as diatoms, is critical to marine ecosystems by serving as the base of the food web (Humborg *et al.* 2000). In total, diatoms have been estimated to account for 20–75% of marine primary production (Nelson *et al.* 1995, Aumont *et al.* 2003, Tréguer *et al.* 2018). As DSi fluxes to coastal zones decrease, diatoms are replaced by phytoplankton species that do not require DSi for growth, and this can result in an increase in the proportion of nuisance algae and the prevalence of harmful algal blooms (Humborg *et al.* 1997, 2000, Turner *et al.* 1998, Ittekkot *et al.* 2000, Cugier *et al.* 2005, Makareviciute-Fichtner *et al.* 2020). Furthermore, reductions in diatom biomass can lead to more complex planktonic food webs in which there are a greater number of trophic levels and where microzooplankton has a more important role (Makareviciute-Fichtner *et al.* 2020). Diatoms are also recognized as primary drivers of carbon export, so changes in DSi fluxes can also have important implications for biogeochemical carbon cycling (Uitz *et al.* 2010, Tréguer *et al.* 2018).

An indicator for assessing the potential contribution of riverine inputs to coastal eutrophication was proposed by Billen and Garnier (2007). The Indicator of Coastal Eutrophication Potential (ICEP) is an estimate of the potential for production of non-siliceous algae by comparing the fluxes of N or P (depending on which of them is limiting) delivered in excess of DSi. Calculating the indicator for a large number of rivers, they found the indicator to be predominantly positive (indicating potential for eutrophication) in the temperate European and North American rivers of their study and negative in the Nordic rivers (including many rivers in Finland and Sweden).

DSi fluxes are of particular interest in Sweden, where the eutrophication of coastal areas and the presence of harmful algal blooms have had significant socioeconomic impacts. Regular monitoring of river discharge and nutrients has taken place in Sweden since the 1960s. Sweden has nearly 100 000 lakes, and considerable decreases in DSi concentrations have been observed along rivers as the percentages of lakes and reservoirs in the upstream basins increase. In addition, the construction of dams and reservoirs in Sweden for hydropower production and flood protection has substantially reduced DSi export to the coast (Conley *et al.* 2000, Humborg *et al.* 2000). Further decreases in DSi export may exacerbate

the effects of eutrophication and algal blooms in Sweden. Harmful algal blooms have been linked to mortality in fish and shellfish and have occasionally resulted in massive losses for the Northern European aquaculture industry (Karlson *et al.* 2021). Moreover, biotoxins from algal blooms can accumulate in bivalve molluscs and present a human health risk if ingested (Visciano *et al.* 2016). Along the Swedish west coast, fish die-offs and high concentrations of diarrhetic shellfish toxins in mussels have been attributed to algal blooms (Karlson *et al.* 2021). Furthermore, tourism in Sweden has suffered as a result of eutrophication and the presence of algal blooms in the Baltic Sea (Söderqvist 2000, Nilsson and Gössling 2013).

Various modelling approaches have been developed to assess and quantify DSi fluxes. Studies by Humborg *et al.* (2008) and Beusen *et al.* (2009) used linear regression models to estimate DSi export of major river basins to coastal zones as a function of inputs such as hydraulic load, soil bulk density, precipitation, slope, and volcanic lithology area. However, these linear regression models do not provide information on intra-annual fluxes and are not suitable for estimating loads for small scales (i.e. individual rivers). In contrast, process-based models represent physical processes more accurately and can offer improved spatial and temporal resolution, but require much more knowledge and data to develop. The process-based RIVERSTRAHLER model has been successfully used to model DSi fluxes in several large watersheds including the Seine, Danube, Kalix, and Lule rivers (Garnier *et al.* 2002, Cugier *et al.* 2005, Sferratore *et al.* 2005, 2008). However, a major limitation of the model is that the drainage network is represented by tributaries which are aggregated according to given stream orders and assigned mean morphological characteristics rather than describing each tributary as a separate river with its own individual characteristics. Thus, to improve the spatial resolution when modelling large domains, it becomes necessary to separately model several sub-basins and connect the results to a model of the main branch of the drainage network (Garnier *et al.* 2002). Machine learning has been used to predict global river DSi yields (Phillips 2020), but few spatially-explicit models have been developed to assess DSi transport at large scales. Jansen *et al.* (2010) developed a lumped continental-scale, process-oriented, empirical model to evaluate DSi mobilization in the US, but the model did not account for anthropogenic impacts such as those of dams/reservoirs, and outputs were limited to the annual time scale. At the global scale, Harrison *et al.* (2012) created the spatially explicit Silica Retention in Reservoirs and Lakes (SiRReLa) model to estimate the global removal of DSi in lentic water bodies, but outputs of the model are also limited to the annual scale. Thus, there is a need for spatially explicit, process-based models which can simulate DSi fluxes and anthropogenic factors across large domains at short time steps.

In this study, a silica module was implemented in the semi-distributed, catchment-based Hydrological Predictions for the Environment (HYPE) model (Lindström *et al.* 2010). The design of the process-based HYPE model allows for multi-variable (hydrology and water quality constituents) simulations of domains ranging from a single river basin to continental, or even global, coverage. The HYPE model has been applied for the whole of Sweden to simulate hydrology

and water quality variables (e.g. nitrogen, phosphorus, sediments, water temperature) at a high spatial resolution and a daily time step. This Swedish HYPE model provided an ideal test bench for (1) developing the HYPE silica routines, (2) quantifying and exploring the drivers of riverine DSi transport, and (3) assessing the multi-nutrient effect on aquatic systems. Specifically, the objectives of the study were to:

- Derive a robust calibration approach for large-scale simulation of soil mobilization and fate of DSi in surface waters;
- Quantify the riverine transport of DSi from land to the Swedish coasts and retention in the surface water system;
- Explore the use of multi-nutrient simulations to assess the role of riverine DSi in coastal eutrophication, following the ICEP approach.

2 Methods

2.1 The HYPE model

HYPE is a semi-distributed hydrological rainfall-runoff model with integrated calculations of several water quality constituents. First described by Lindström *et al.* (2010), the model simulates how water and constituents move through soil profiles, water courses, lakes and reservoirs. The modelled domain

is split into sub-basins, and the land in each sub-basin is further divided into hydrological response units (HRUs), which are typically combinations of specific land uses and soil types. The soil profile within an HRU is divided into up to three soil layers. The model is driven by meteorological data, primarily precipitation and air temperature, but other meteorological data are needed for more complex evapotranspiration routines. Flow of water between the soil layers and out of the profile is simulated for each time step, and water and constituents leaving the soil profile are routed through a network of rivers, lakes and reservoirs (Fig. 1). For an updated version of the model description the reader is referred to the HYPE wiki (<http://hype.smhi.net/wiki/doku.php>).

2.2 Implementation of silica routines

The silica routines described in this section were implemented in version 5.15.10 of the HYPE modelling software. The development was split into two main parts: (1) soil chemical weathering and (2) surface water processes. Soil chemical weathering is the sole source of silica in the model. Weathered silica is transported with runoff to the modelled river network, where it may be converted from DSi into biogenic silica through diatom uptake. Silica is returned to the dissolved phase during diatom decomposition, but may also be permanently removed

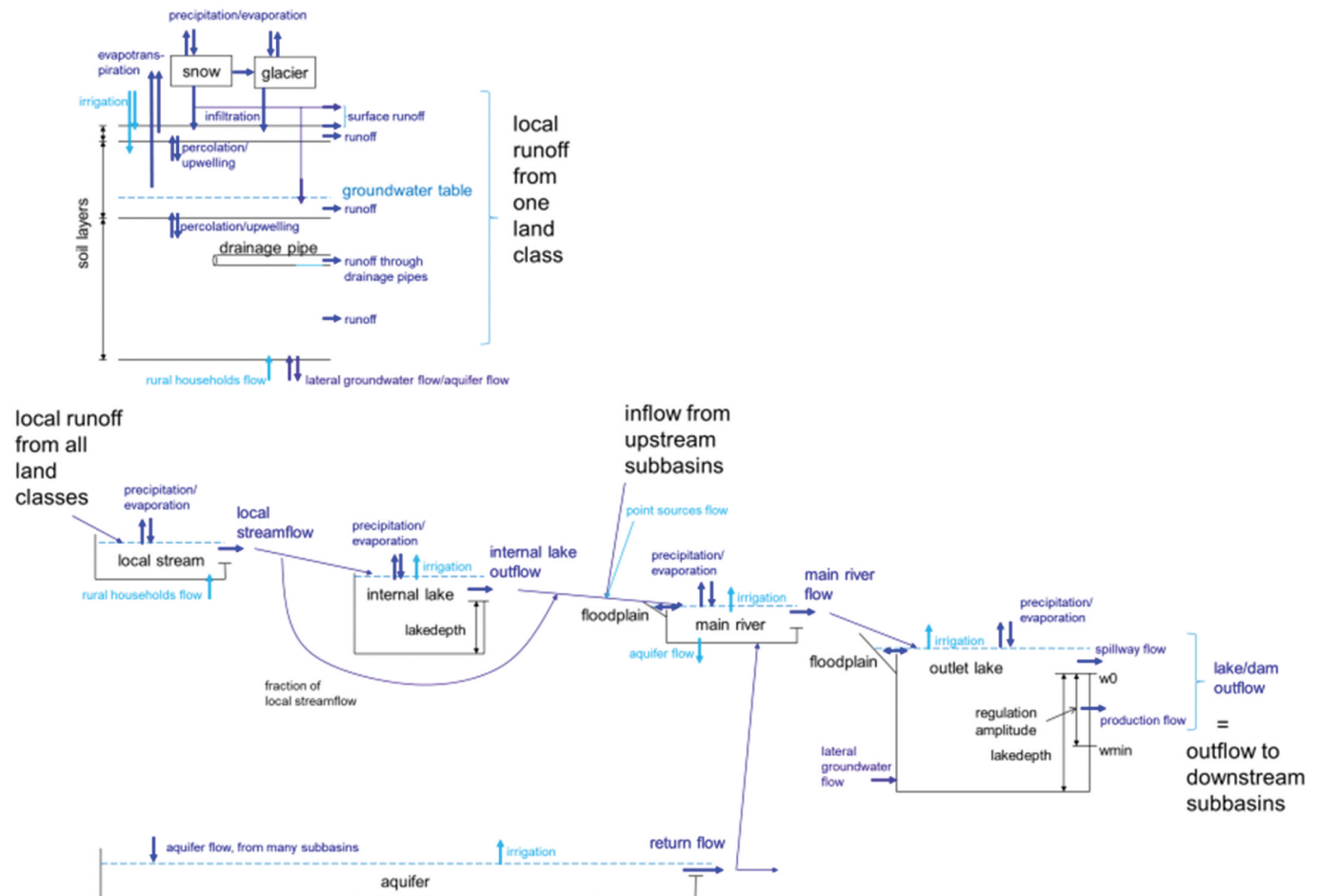


Figure 1. Overview of fluxes of water in soils (upper) and surface water (lower) in the HYPE model. The aquifer routine was not used in this work. Light blue arrows indicate water flows that may occur if certain water quality constituents or irrigation are simulated.

from the system as diatoms settle to the bottom of lakes/reservoirs.

2.2.1 Weathering of dissolved silica

Silica weathering is assumed to occur in the soil profile according to Equation (1). The equation attempts to encompass key factors affecting weathering while acknowledging the HYPE model structure, simulated variables, and available data:

$$W_i = \text{weathrate} \cdot \Delta z_i \cdot f(T_{\text{soil},i}) \cdot f(z_i) \cdot \text{weathcorr} \quad (1)$$

where W_i is the weathering ($\text{mg}/\text{m}^2/\text{d}$) of soil layer i , weathrate is the soil type dependent weathering rate parameter ($\text{mg}/\text{m}^3/\text{d}$), Δz is the thickness of the soil layer (m), T_{soil} is the (simulated) soil temperature of the soil layer ($^{\circ}\text{C}$), z is the depth from soil surface to the middle of the soil layer, and weathcorr is a dimensionless weathering correction factor. The parameter weathcorr may be linked to any relevant geographical feature of the sub-basin (e.g. land use or bedrock geology) that has the potential to affect weathering.

DSi weathering varies according to soil temperature, increasing with higher temperatures. The temperature effect on weathering is implemented in the model based on an Arrhenius-type equation, used for example by Hartmann *et al.* (2014), including a normalization against a reference temperature (Equation 2):

$$f(T_{\text{soil},i}) = e^{-\frac{\text{weathKT}}{R} \left(\frac{1}{T_{\text{soil},i} + 273.15} - \frac{1}{T_{\text{ref}} + 273.15} \right)} * \text{frozensoilcorr} \quad (2)$$

where weathKT (J/mol) is the rock activation energy, R is the universal gas constant ($\text{J}/\text{mol}/^{\circ}\text{K}$), T_{ref} is a reference temperature ($^{\circ}\text{C}$), and frozensoilcorr is a dimensionless correction factor to account for reduced connectivity between soil particles and soil water in partly frozen soil layers. This correction factor is calculated for negative soil temperatures (Equation 3):

$$\text{frozensoilcorr} = 1 - \frac{T_{\text{soil},i}}{T_{\text{soil},i} - 1} \quad (3)$$

The silica weathering is assumed to decline with the depth of the soil layer to account for, for example, the influence of organic acids and plants. An exponential decline of weathering is implemented as a function of soil depth (Equation 4):

$$f(z_i) = e^{-\ln(2) \cdot z_i / \text{weathdep}} \quad (4)$$

where the model parameter weathdep (m) is the depth where the activity is half of that at the surface.

2.2.2 Surface water processes

The weathered DSi is carried by the runoff water from the various HRUs in each sub-basin to the local rivers from where it is transported further downstream through the network of main rivers and lakes (Fig. 1). In the surface water compartments of the river, the DSi may be transformed into BSi (immobilization) and vice versa (mineralization). Sedimentation is a sink of BSi in lakes and reservoirs.

2.2.2.1 Biogenic silica (BSi). Net uptake of DSi and production of BSi occur during spring and early summer in a similar way as for the other nutrients in the model (Lindström *et al.*

2010). The process is reversed during late summer and early autumn. This (potential) net production is simulated as a function of water temperature, total P concentration in the water body, and volume of water in the water body (Equations 5–8):

$$\text{Prodmin} = \text{wprodSi} \cdot f_1(T_{\text{water}}) \cdot f_2(T_{\text{water}}) \cdot f(C_{\text{totP}}) \cdot V_{\text{water}} \quad (5)$$

$$f_1(T_{\text{water}}) = (T_{\text{water}}/20)^{\text{sitmpexp}} \quad (6)$$

$$f_2(T_{\text{water}}) = \frac{T_{10} - T_{20}}{5} \quad (7)$$

where Prodmin is a potential net production, T_{water} is the water temperature ($^{\circ}\text{C}$), T_{10} and T_{20} are the average water temperatures of the last 10 and 20 days ($^{\circ}\text{C}$), respectively, and V_{water} is the volume of water in the water body (m^3). The exponent sitmpexp , a model parameter, affects the temperature–process rate relationship. The production is assumed to be spurred by higher levels of P in the surface water compartment (Equation 8):

$$f(C_{\text{totP}}) = \frac{\text{MAX}(0, C_{\text{totP}} - \text{plimsi})}{\text{MAX}(0, C_{\text{totP}} - \text{plimsi}) + \text{hsatTP}} \quad (8)$$

where C_{totP} (mg/L) is the current simulated total phosphorus (TP) concentration in the water body (mg/L) and plimsi is a model parameter that represents the part of TP that is not bio-available and hsatTP (mg/L) is the bio-available phosphorus concentration where the value of the function is reduced by half.

2.2.2.2 Lake/reservoir diatom sedimentation. BSi may settle to the bottom of lakes/reservoirs (Equation 9). As it reaches the bottom it is assumed to be buried in sediments and permanently removed from the model.

$$\text{sed}_{\text{lake}} = \text{sedSi} * C_{\text{BSi}} * a_{\text{lake}} * 10^{-3} \quad (9)$$

where sed_{lake} is lake sedimentation (kg/d), sedSi is a model parameter representing the settling velocity of BSi (m/d), C_{BSi} is the concentration of BSi in the water body (mg/L) and a_{lake} is the surface area of the water body (m^2).

2.3 The HYPE model set-up for Sweden (S-HYPE)

A HYPE model for the entirety of Sweden has been set up and calibrated for flow, nutrients, suspended solids, and water temperature. This model application, referred to as “S-HYPE,” is used for example in the national hydrological warning service and by national, regional and local water managers in their work related to the EU Water Framework Directive. The first version of S-HYPE was released in 2012 and is described by Strömquist *et al.* (2012). Since its initial release, S-HYPE has undergone continuous development, and different water management tools have been built to visualize S-HYPE results (e.g. Strömbäck *et al.* 2019). This work is based on S-HYPE version 2016e, which also covers the parts of Norway and Finland draining into Sweden. In this version, the model domain is divided into nearly 40 000 sub-basins

with an average size of 13.2 km² (median 6.5 km²). The model simulations for this work are based on a daily time step, although HYPE can also be run on a sub-daily time step. The median Nash-Sutcliffe efficiency (NSE) for simulated discharge is 0.76 for 505 sites with flow gauging data. Sources of N and P include diffuse sources (such as agricultural runoff) simulated dynamically by the model and direct input to the surface water system such as point sources (e.g. discharge from sewage treatment works) and atmospheric deposition. The median absolute relative errors for total P and total N concentrations are 24% and 14%, respectively, based on data from 1052 monitoring stations.

2.4 S-HYPE model calibration

The time series of measured DSi concentrations in watercourses used for the S-HYPE model calibration were obtained from the Swedish national water body monitoring data repository (Swedish University of Agricultural Sciences 2019). Sampling stations with more than 12 data records during the calibration period (200 records on average) were selected and linked to the corresponding sub-basin in the S-HYPE model.

A step-wise calibration was performed in which the model parameters related to weathering were calibrated first, followed by those related to surface water processes. The calibration period was 1998–2017, following a 15-year model spin-up period. The calibration time period was also used for the analyses presented here. Parameters in the model are not linked to specific rivers or river basins but are instead general for the entire model domain (e.g. those related to surface water processes) or linked to specific landscape features such as soil type (e.g. weathering parameters). The calibrated set of model parameters thus represents a compromise solution aimed at achieving a good overall model performance and is not optimized to a specific site. Overall, this calibration approach favours having a limited

number of landscape features to calibrate as this reduces the risk that some categories will be poorly represented in calibration catchments. Furthermore, limiting the number of landscape features helps to reduce the problem of co-variation between the prevalence of certain landscape elements and hydro-climatic factors.

2.4.1 Weathering

HYPE's weathering parameters were calibrated using a catchment mass-balance approach comparing simulated riverine fluxes of silica against those observed at river sampling sites representing a large variety of catchment characteristics. This allowed for the estimation of weathering rates and subsequent upscaling to the whole of Sweden. The calibrated weathering parameters were the soil-type dependent rate parameter (*weathrate*) and the correction factor (*weathcorr*). According to analyses of model biases for initial simulations, it was observed that lithotectonic regions provided a suitable division of the country to account for the spatial variations in weathering not accounted for by the other weathering drivers included in the model. The parameter *weathcorr* was thus applied for the different lithotectonic areas of Sweden (Fig. 2) but was not calibrated for areas outside of Sweden. Figure 3 illustrates the relationship between the distribution of soil types and the lithotectonic units. Till is the dominant soil type across every lithotectonic region, except for the Platformal sedimentary cover rocks area. Thin soils are relatively more common in the western part of the country, including the mountainous areas in the north.

Calibration was performed for 119 stations in smaller rivers (upstream area ≤1000 km²) with low influence of lakes and reservoirs (water surface <2% of the catchment area) to reduce the risk of interference by surface water processes. The objective of the calibration of the weathering parameters was to minimize the relative error (RE) between the average simulated and observed DSi concentrations at the calibration sites (see Section 2.4.3).

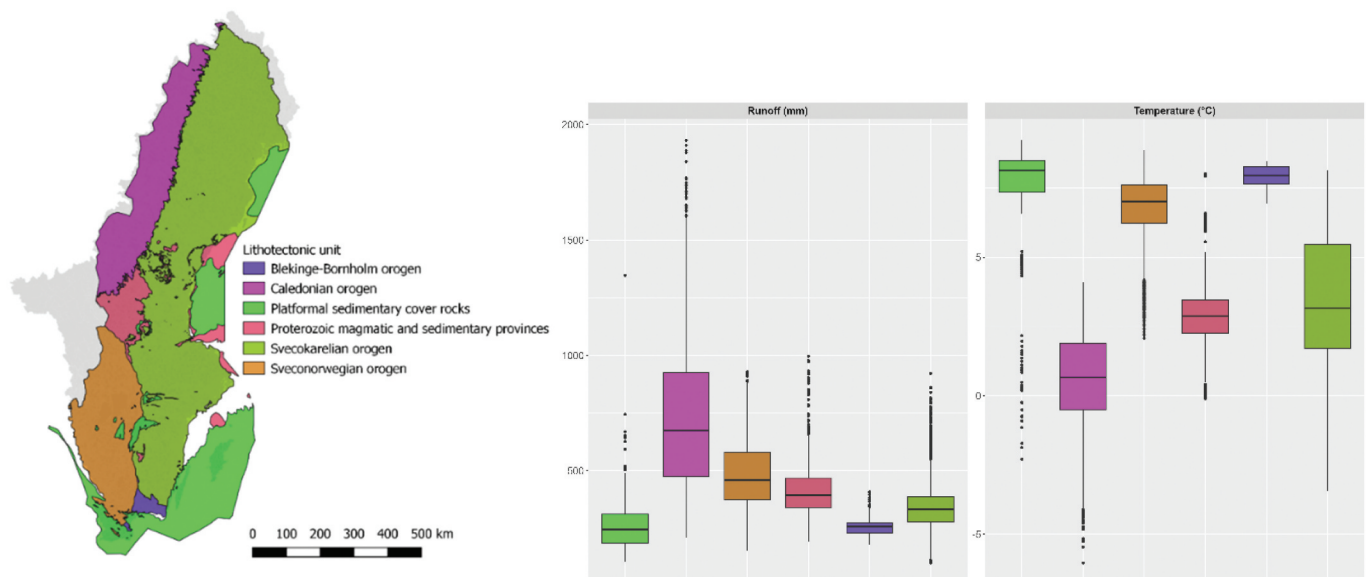


Figure 2. Lithotectonic regions of Sweden, with accompanying box plots showing simulated mean annual runoff and mean annual air temperature for the sub-basins contained in each region. Note that the S-HYPE domain also includes small portions of Norway and Finland (grey areas).

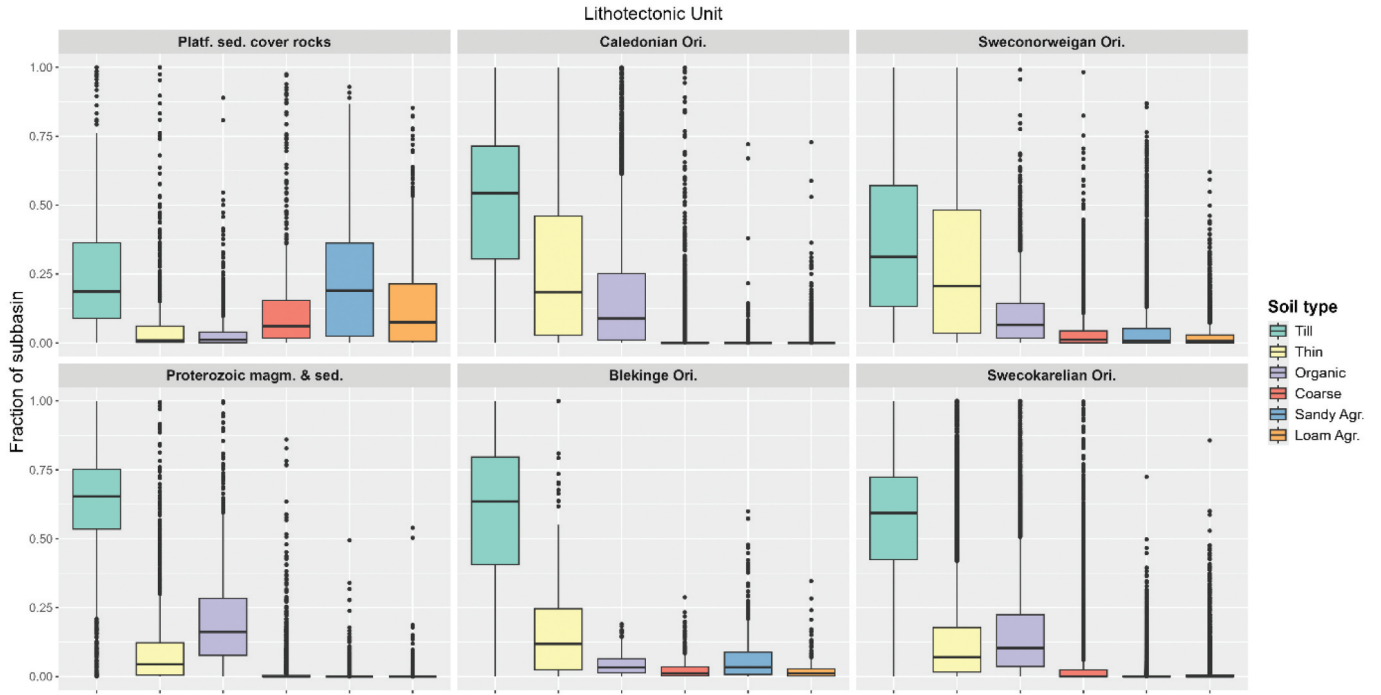


Figure 3. Distribution of soil types within sub-basins across Sweden's lithotectonic regions.

2.4.2 Surface water processes

Lake process parameters were then calibrated against observed DSI concentration data downstream of 10 selected lakes with different characteristics. The focus of the calibration was to realistically simulate the level and seasonal dynamics of DSI concentrations using the correlation and RE criteria (see section 2.4.3), and visual time series inspection. BSI values were only calibrated indirectly, as no concentration data other than that for DSI were available. The simulated effect of lakes on overall DSI transport was more closely explored in the basin of the Motala Ström River (Fig. 4) which passes through several larger lakes before reaching the coast. The basin area of Motala Ström is approximately 15 400 km² and 20% of the area is covered by lakes or dams.

2.4.3 Model performance indicators

Several performance indicators were used to assess model performance. RE indicates the average percent differences between mean simulated and observed DSI concentrations relative to the mean of the observations. To assess potential biases related to specific catchment characteristics, we also computed weighted REs (Equation 10):

$$RE_{weighted} = \frac{\sum_{i=1}^n RE_i x_i}{\sum_{i=1}^n x_i} \quad (10)$$

where x is a specific catchment characteristic, in this case fractional area of the soil type or lithotectonic unit upstream of the observation site i , or other characteristics such as catchment area, lake volume and a climatic factor. Because RE ranges from -100 to infinity, a few sites with very large positive RE values could disproportionately influence model-skill estimates. To mitigate this, we defined a new metric using Equation (11), here referred to as an

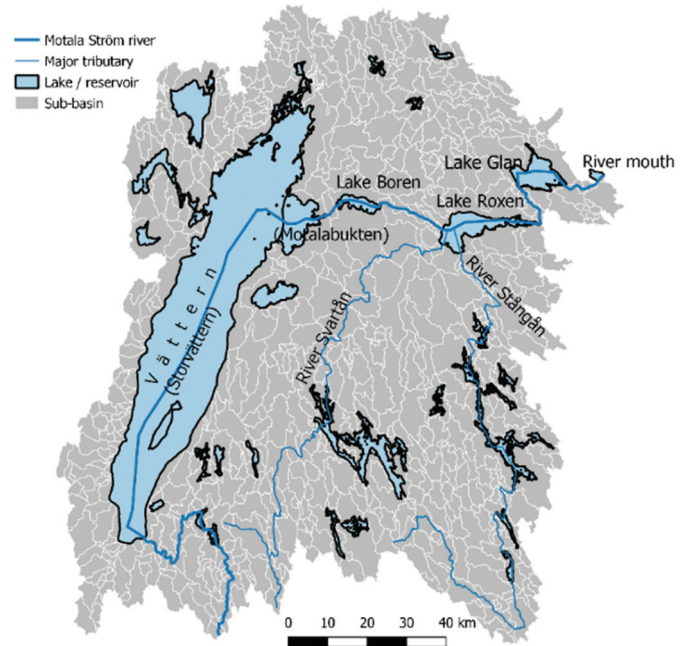


Figure 4. Overview of basin of the Motala Ström River.

adjusted relative error (RE*), which ranges from -100 to 100:

$$RE^* = \begin{cases} 100 * \frac{\bar{C}_{sim} - \bar{C}_{obs}}{\bar{C}_{obs}} & \bar{C}_{sim} \leq \bar{C}_{obs} \\ 100 * \frac{\bar{C}_{sim} - \bar{C}_{obs}}{\bar{C}_{sim}} & \bar{C}_{sim} > \bar{C}_{obs} \end{cases} \quad (11)$$

where $\bar{C}_{sim}$ and $\bar{C}_{obs}$ are the average simulated and observed concentrations of DSI for days with observations, respectively.

The RE statistic was calculated for both concentrations and flow-weighted average concentrations. Flow-weighted

concentrations were included because long-term weathering is linked more to riverine DSi transport than the average DSi concentration. The flow-weighted averages were calculated using Equation (12):

$$\bar{C}_{qw} = \frac{\sum_{i=1}^n C_i \cdot Q_i}{\sum_{i=1}^n Q_i} \quad (12)$$

where $\bar{C}_{qw}$ is the flow-weighted average concentration, C_i is either the observed or simulated DSi concentration at dates of DSi measurement and Q_i is the simulated flow at the dates of DSi measurement. Simulated flows were used because observed flow data were unavailable for the majority of sites with DSi observations.

The Pearson correlation coefficient (CC), calculated using Equation (13), was also used to assess model performance:

$$CC = \frac{\frac{1}{mi} \cdot \sum_{i=1}^{mi} (r_i \cdot c_i) - \bar{c} \cdot \bar{r}}{\sigma_c \cdot \sigma_r} \quad (13)$$

where mi is the number of values in the time series of a station, c_i and r_i are computed and recorded values, $\bar{r}$ and $\bar{c}$ are the mean of the observations and simulation results at days with observations, and σ_c and σ_r are the standard deviation of the computed and recorded concentrations.

Finally, the Kling-Gupta model efficiency (KGE) indicator was used for model evaluation and was calculated according to Equation (14) (Gupta *et al.* 2009):

$$KGE = 1 - \sqrt{(CC - 1)^2 + \left(\frac{\sigma_c}{\sigma_r} - 1\right)^2 + \left(\frac{\bar{c}}{\bar{r}} - 1\right)^2} \quad (14)$$

2.5 Parameter sensitivity analysis

A parameter sensitivity analysis was carried out to gain a better understanding of the relative importance of the model's parameters with respect to DSi concentrations in surface water in the model domain. Relative changes in simulated surface water DSi concentrations as a result of changes in the calibrated parameter values were evaluated from two different perspectives:

- How does the model respond to changes in model parameters affecting weathering (parameters *weathrate*, *weathdep*, *weathKT*) and sedimentation (*sedSi*)? Here, model parameter values were altered one at the time, either increasing or decreasing their baseline values by 10%. The change in the *weathrate* parameter was applied to the values for all soil types simultaneously.
- How does the model represent spatial variability in weathering due to different soil types and lithology? The model allows the user to define the *weathrate* parameter individually for different soil types. Similarly, the *weathcorr* factor can be used to introduce geographic variability in weathering potential. The relative effect of soil type and bedrock on weathering in the model set-up was evaluated by individually increasing weathering rates by 10% for the three most abundant soil types (covering 84% of the area) and for the five most dominant litho-tectonic units of the model domain (99% coverage).

The analyses were conducted by examining the spatial patterns of change resulting from the sensitivity runs across the entire modelled domain. In addition to analysing concentration changes, we also calculated the changes in riverine DSi loading to the sea for the five EU River Basin Districts (RBDs) in Sweden, which are administrative river basin management areas with associated coastal areas. This analysis aimed to provide an indication of how uncertainties in parameterization affect the input to coastal areas.

2.6 Indicator of Coastal Eutrophication Potential (ICEP)

By adding silica to the nutrients already simulated by S-HYPE (N and P), it was possible to calculate the ICEP index (Billen and Garnier 2007) for selected rivers. The ICEP was proposed as a way to estimate the amount of additional carbon biomass potentially produced in a receiving coastal water body from the flux of the N or P (whichever is the limiting nutrient) delivered with the riverine inflows in excess over silica. The ICEP is derived from the Redfield ratio for marine phytoplankton and therefore is less relevant for eutrophication in inland waters. The ICEP is calculated according to Equation (15):

$$ICEP = \begin{cases} \frac{1}{A} \cdot \left(\frac{NFlx}{14 \cdot 16} - \frac{SiFlx}{28 \cdot 20} \right) \cdot 106 \cdot 12 & \frac{N}{P} < 16 \\ \frac{1}{A} \cdot \left(\frac{PFlx}{31} - \frac{SiFlx}{28 \cdot 20} \right) \cdot 106 \cdot 12 & \frac{N}{P} > 16 \end{cases} \quad (15)$$

where *PFlx*, *NFlx* and *SiFlx* are fluxes of total nitrogen, total phosphorus and dissolved silica (kg/d), respectively, delivered to the basin outlet, and *A* is total upstream area (km²). N/P is the molar ratio of nitrogen and phosphorus. The unit of ICEP is kg[C]/km²/d.

Simulated riverine fluxes of N, P and DSi were used to calculate the ICEP for 25 coastal water bodies receiving inflows from 25 larger rivers in Sweden (Fig. 5) representing a wide range of climatic regions, land cover types, and the influence of lakes and reservoirs. The ICEP was calculated from riverine fluxes delivered to and thus affecting coastal water bodies, assessing only the new production in coasts from river inflows. Data on other nutrient inputs to the coastal zone such as direct point source emissions, internal nutrient loading from sediments or exchange with the outer sea would be required together with a coastal zone model to fully assess the eutrophication of the coastal zones.

3 Results

3.1 Calibrated weathering parameters and simulated weathering rates

Some differences in calibrated weathering rate parameter values were detected for various soil types (Table 1). Note that the agricultural land has its own soil type classes in the S-HYPE model as a different, more detailed, soil map based on soil texture was used for defining HRUs on the agricultural land. The *weathrate* weathering rate parameter varies between 7.5 mg/m³/d (thin soils) and 16.7 mg/m³/d (agricultural silty soils). Thin soils co-vary to some degree with the Caledonian orogen in the mountainous northwest part of the country characterized by lower temperatures and higher precipitation



Figure 5. Rivers in Sweden for which the Indicator of Coastal Eutrophication Potential (ICEP) was calculated.

than the country average (Figs. 2 and 3). For the agricultural soil there was no apparent correlation between particle sizes and the calibrated weathering parameter values, but these soil classes only cover a small fraction of the land and the calibrated model parameters may not be very robust for these soil types.

The calibrated weathering correction factors (*weathcorr*) varied across the tectonic regions, with values ranging from 0.5 to 1.3. In the two largest tectonic regions, Svecokarelian orogen and Caledonian orogen, both dominated by metamorphic rocks, no weathering correction was required (i.e. *weathcorr* = 1). The most pronounced deviation from the mean was found for the Platformal sedimentary cover rocks unit where *weathcorr* = 0.5. This low value indicates lower weathering rates that cannot be explained by the other factors accounted for in the simulation (e.g. temperature or soil type).

The calibrated S-HYPE model allowed for the assessment of DSi weathering patterns across Sweden (Fig. 6). Overall,

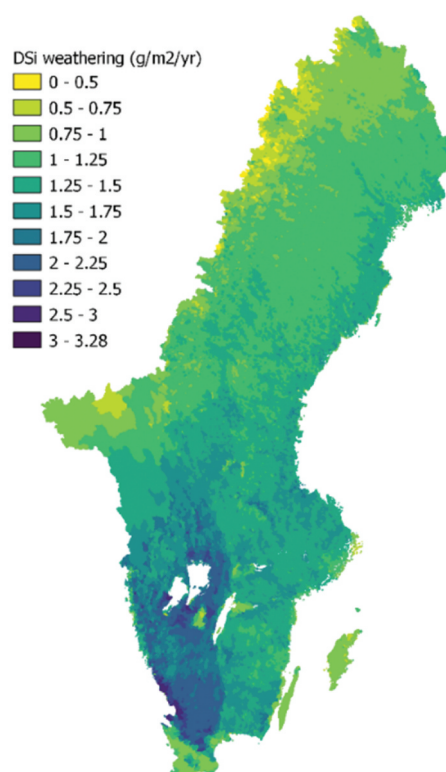


Figure 6. Simulated annual average dissolved silica (DSi) soil weathering rates in the S-HYPE model set-up.

the simulated weathering pattern in Sweden was clearly linked to the average air temperature with lower weathering in the north, especially in the mountainous areas in the west. Differences in weathering rates were visible between the lithotectonic units, especially in the southern part of the country where the weathering rates create distinct patterns matching the boundaries between the Sveconorwegian orogen in the west, the Svecokarelian orogen to the east and the Platformal sedimentary cover rock in the far south and on the two larger islands (Öland and Gotland) off the Swedish east coast.

3.2 Model evaluation

Modelled DSi concentration levels generally matched observations across Sweden (Fig. 7), although the model tended to

Table 1. Model soil weathering parameter (weathrate) values for different soil types and lithotectonic regions. Water surface is excluded from the table, thus the total does not add up to 100%.

Soil type	% of Sweden	weathrate	Lithotectonic region	% of Sweden	weathcorr
Till	46.2%	10.2	Platformal sedimentary cover rocks	4.0%	0.5
Thin soils	15.9%	7.5	Caledonian orogen	17.1%	1
Organic	14.8%	9.2	Sveconorwegian orogen	15.9%	1.3
Glacial deposits	2.5%	10.9	Proterozoic magmatic and sedimentary rocks	5.3%	1.2
Coarse	2.6%	14.5	Blekinge-Bornholm orogen	1.0%	0.95
Silt	0.6%	13.9	Svecokarelian orogen	56.7%	1
Clay	1.8%	11.1			
Sandy (agricultural)	2.1%	12.5			
Loam (agricultural)	1.8%	11.1			
Silty (agricultural)	0.8%	16.7			
Medium clay (agricultural)	1.1%	11.6			
Heavy clay (agricultural)	1.1%	10.5			

underpredict values in the far north. This bias was diminished when flow-weighted concentrations were analysed, suggesting that the underprediction occurs mainly during low-flow periods, typically occurring during the winter months in northern Sweden. Figure 7 includes results for all sites including those that have been used in calibration. Average Pearson correlation coefficients were typically above 0.25 for most sites (mean = 0.44), indicating that the model was also capable of representing some of the temporal variations in riverine DSi concentrations. KGE values were predominantly above -0.41 (mean = 0.2), which indicates that the model provides better performance than simply using the mean observed value as a predictor (Knoben *et al.* 2019).

Higher correlation and KGE values were noted for the validation sites (Table 2) than for the calibration sites. The catchments of

the validation sites were generally larger and with higher lake fractions than those of the calibration sites. Typically, the influence of site-specific conditions on riverine concentrations decreases with catchment area, resulting in better model performances.

Figure 8 illustrates the simulation bias related to different characteristics of the upstream area of each sampling site based on long-term averages. Overall, the mean adjusted RE for all of the sampling sites was 1.1%. No clear relationship was observed between bias and the size of catchment. However, there was a tendency towards overpredicting concentrations for sites with higher mean air temperature. These sites, located in the south and central parts of Sweden, contrast the higher runoff sites in the mountainous region in the northwest, where there is a small tendency for model underprediction. No apparent trend was noted with

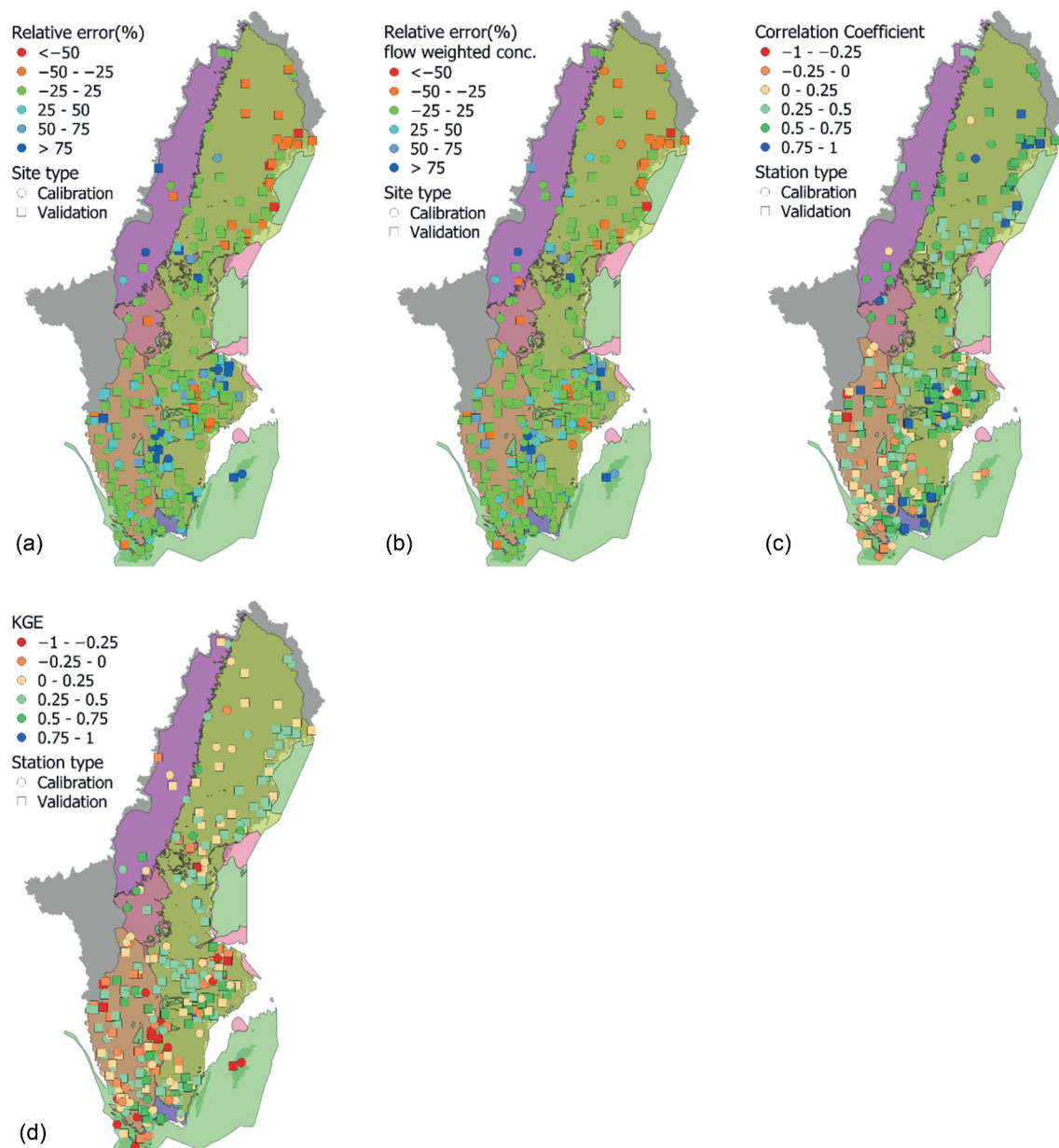


Figure 7. Model performance at dissolved silica concentration sites: (a) Relative error of the mean, (b) average relative error of the mean for flow-weighted concentrations, (c) correlation coefficient, (d) Kling-Gupta efficiency (KGE). The lithotectonic units of Sweden are shown as background. Both calibration (circles) and validation (squares) sites are shown in the figures.

Table 2. Model performance at calibration and validation sites.

	Calibration	Validation
Number of stations	129	239
CC mean	0.33	0.49
CC median	0.34	0.54
KGE mean	0.21	0.25
KGE median	0.15	0.29
abs (RE%) mean	26.1	27.4
abs (RE%) median	16.8	20.0
abs (RE%, flow weighted) mean	22.2	22.5
abs (RE%, flow weighted) median	16.2	16.9

CC: Pearson correlation coefficient; KGE: Kling-Gupta Efficiency; RE: relative error.

increased lake influence. The dominant types of soils in the model domain are till and organic soils together with land with a thinner soil cover, and only small biases were noted for these soil types. It is more difficult to draw a clear conclusion about some of the other soil types as they make up a relatively small fraction of the area upstream of the observation sites, but overall, the model appears to be “balanced” with regard to soil type. The same analysis shows that only a little of the bias can be attributed to the lithotectonic units. For illustration, examples of typical time series plots from the range of model performances are given in Fig. 9.

3.3 Sensitivity analysis

The leaching of DSi from soil in the model is proportional to the value of the *weathrate* parameter (Equation 1).

Consequently, DSi concentrations in surface waters show a nearly linear response to changes in this parameter (Fig. 10). Similarly, the model is sensitive to changes in the *weathdep* parameter, which describes how weathering activity declines with depth. The effect of changing the *weathKT* parameter is smaller and exhibits opposite signs of change in northern and southern Sweden. Changes in the *sedSi* parameter, which affects the sedimentation of BSi, affect specifically DSi concentrations in those rivers that are strongly influenced by lakes.

The spatial distribution of the major soil types and lithotectonic units in Sweden is apparent from Fig. 11, showing the effect of combined changes to these parameters on DSi concentrations. In particular, the spatial extents of the lithotectonic units are clearly visible, although the effects are also observed in downstream river reaches.

The patterns observed in the analyses above are also evident in Fig. 12 showing the load delivered to the coastal areas of individual RBDs of the Baltic Sea from Sweden. Uniform changes in the model parameters do not lead to uniform changes in the DSi transport from land to sea as different soil types and lithotectonic units are not uniformly distributed across space. The relatively highest effect is seen for the Skagerrak and Kattegat RBD, then South Baltic RBD, and it gradually decreases as we move to the northern RBDs. Changing *weathcorr* for the most prevalent soil type, till (46% in Sweden), affects all RBDs (5–8% increase in DSi transport). While the sedimentation parameter *SedSi* can have a large effect locally, changing it within 10% affects the DSi transport to the sea by less than 2%. Similarly, *weathKT*

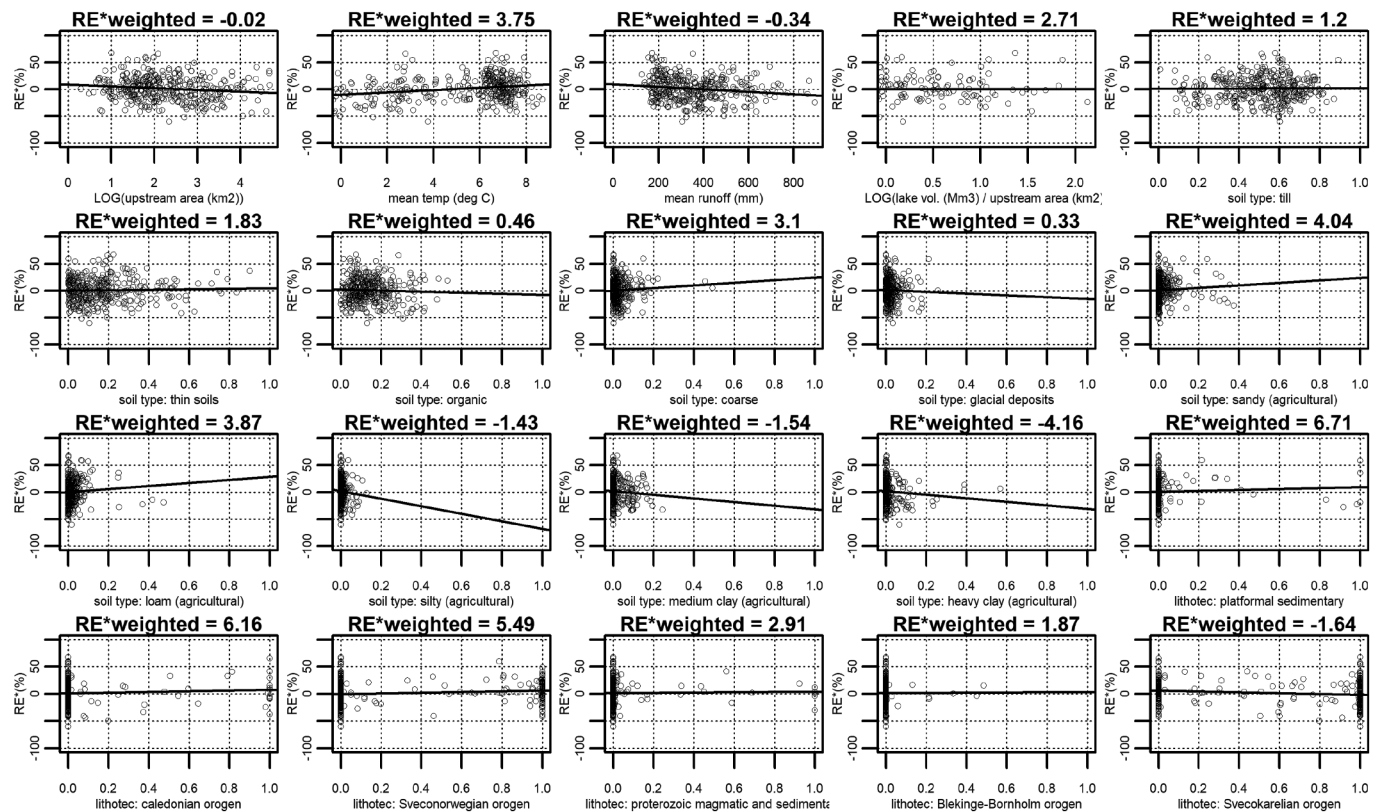


Figure 8. Characteristics of catchments (upstream of dissolved silica concentration sampling sites) versus adjusted relative error with linear interpolation lines. Numbers in the charts show mean adjusted Relative Error for sampling sites weighted against the catchment characteristic (Equation 10). The soil and lithotectonic sub-plots illustrate the bias versus the upstream areal fraction of the different categories.

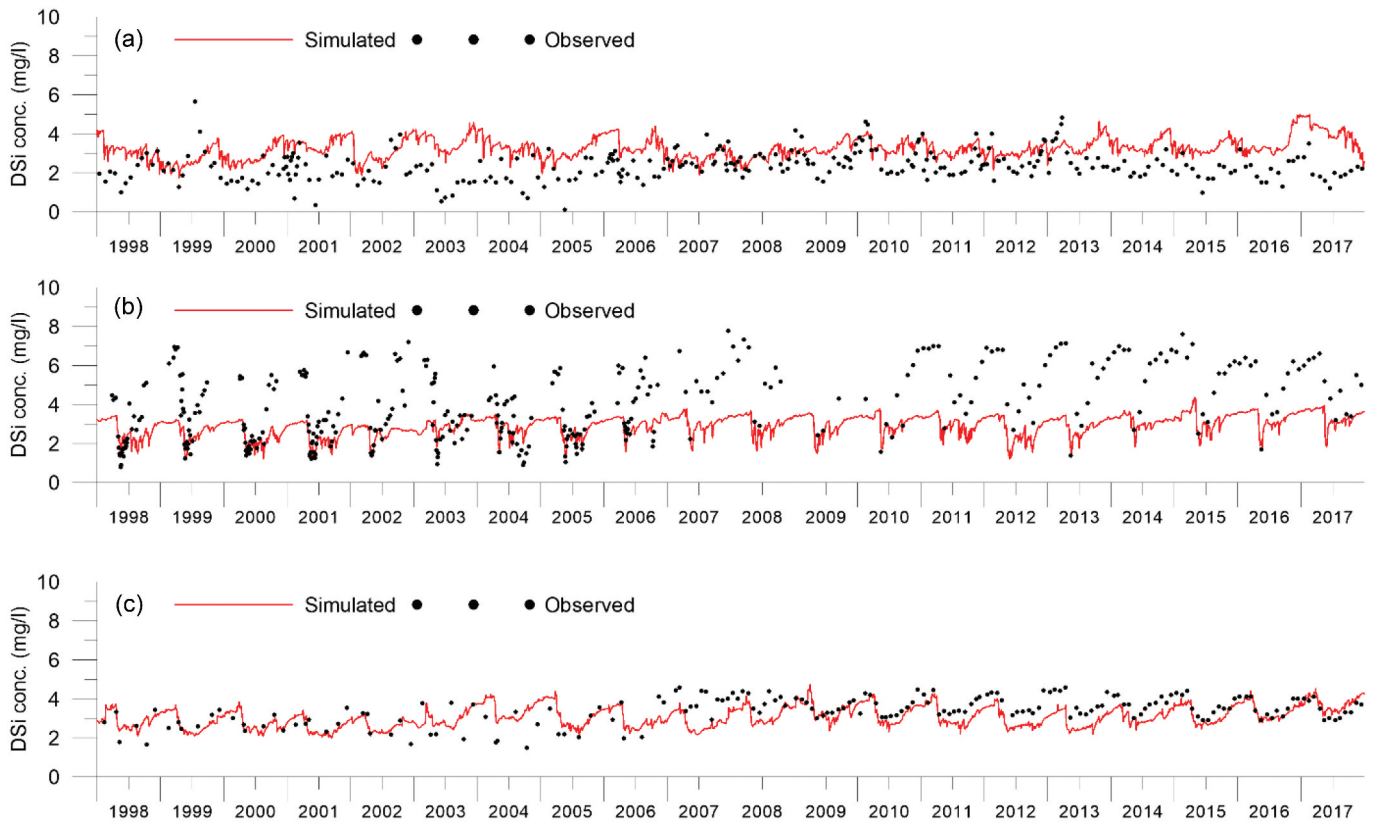


Figure 9. Plots of simulated and observed dissolved silica concentrations at the sites with the (a) 75th (KGE = 0.05), (b) 50th (KGE = 0.25) and (c) 25th (KGE = 0.42) percentile highest KGE values. KGE: Kling-Gupta Efficiency.

has only marginal impact on total DSi transport. Weathering parameters *weathrate* and *weathdep* are determining factors for DSi transport from all RBDs.

The Caledonian orogen unit is the second largest lithotectonic region in Sweden (17% of Sweden). It affects the headwaters of rivers in the two northernmost RBDs in Sweden, where it represents about 30% of the drainage area. Here, increasing the *weathcorr* factor by 10% results in an increase by only 2–3% in DSi transport from Bothnian Bay and Bothnian Sea RBDs due to retention processes on the way from headwaters to the Baltic Sea. The same change in *weathcorr* for Swecokarelian orogen unit, which represents the remaining 70% of these two RBDs, leads to an increase by 6–8% in DSi transport from the same RBDs. However, for North Baltic RBD where it is prevalent, this change leads to an increase by over 10% in total DSi transport from the RBD.

3.4 Silica transport and retention

The amount of silica transported in the hydrological network is a function of both the input from land, calculated in the model as a function of DSi mobilization (weathering) and soil runoff processes, and transport and transformation processes in surface waters. The input of DSi to the surface water system (Fig. 13a, aggregated to larger areas representing whole river basins or larger tributaries) reflects the pattern of the simulated soil weathering (see Figure 6). Lower values were noted in catchments with

large areal proportions of lakes and reservoirs. This is an aftereffect of averaging the mobilized DSi over the whole catchment, as no weathering is simulated in the inundated areas but the area itself is included in the catchment area.

The retention of DSi in the surface water system and its corresponding effect on transported amount is seen in Fig. 13b–c. The effect of large lakes is clearly visible as locations with high retention show low yields. This is further illustrated in Fig. 14, where we follow the DSi generated in headwaters of Motala Ström as it passes through a number of larger lakes on its way to the coast. One of these lakes, Lake Vättern, is the sixth largest lake in Europe, with a mean depth of around 40 m and a very long turnover time (approximately 60 years), retaining much of the silica generated upstream. Most of the retention occurred in the two lake parts of Lake Vättern. Smaller but still significant retention (ca. 10–15%) occurred in the lakes downstream of Lake Vättern (Boren, Roxen and Glan with turnover times of 1.4, 2.6 and 3.6 months, respectively). A significant amount of silica enters Motala Ström from the tributaries downstream of Lake Boren, especially from the two larger rivers Svartån and Stångån. The total DSi outflow from Lake Roxen is about 5 times higher than the total outflow from Lake Vättern, while Roxen drains an area about 2 times larger than Vättern. The total DSi outflow increases slightly after additional inputs from tributaries to Lake Glan that exceed the retention in the lake.

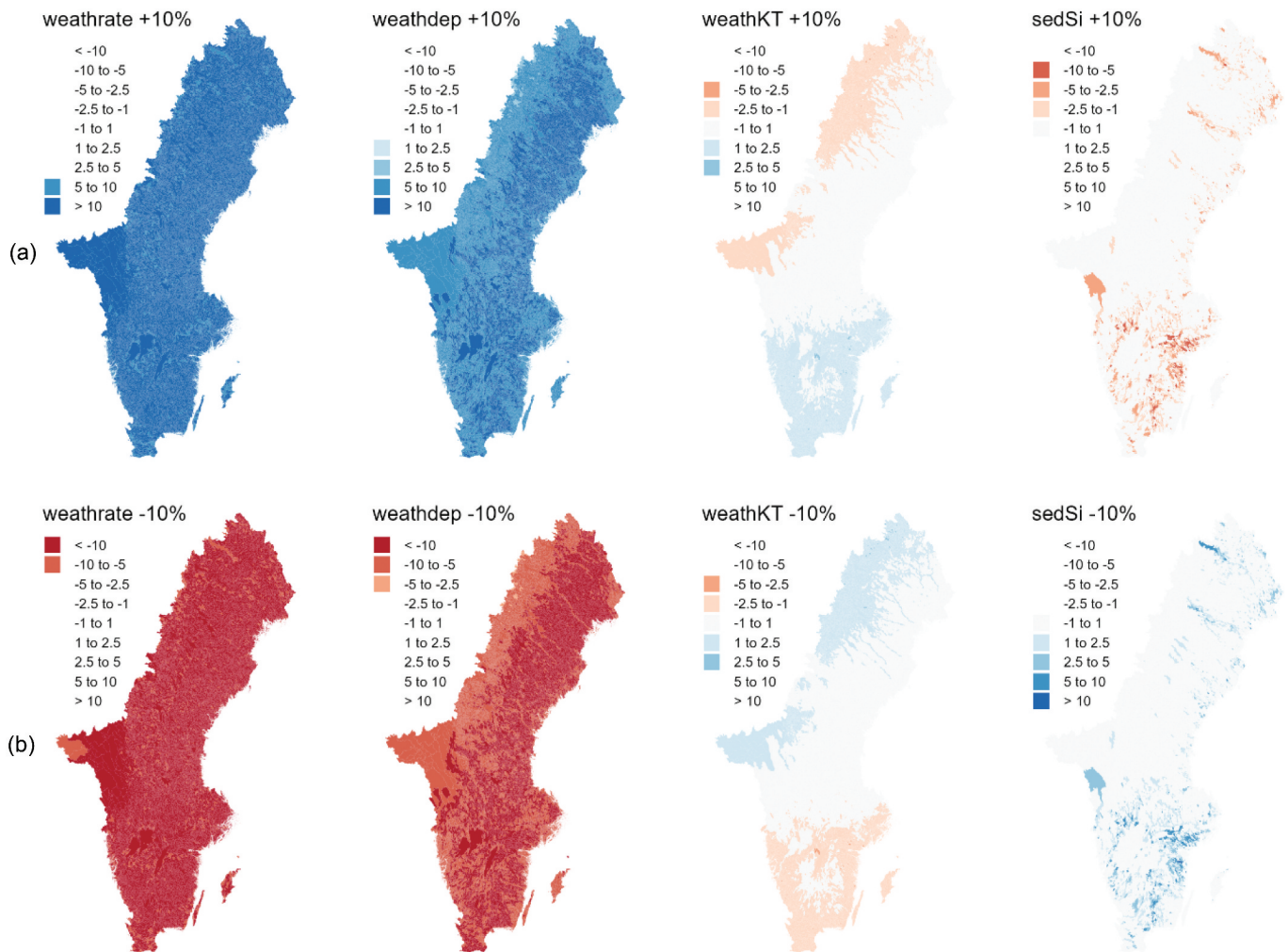


Figure 10. Percentage change in simulated riverine dissolved silica concentrations resulting from (a) 10% increases or (b) decreases in parameter values.

3.5 River mouth assessment of ICEP

All 25 selected large Swedish rivers (see Section 2.5) were shown to be clearly phosphorus limited with a negative ICEP score (Table 3), although it was close to zero for several rivers. The negative ICEP indicates that the nutrient composition of the contribution from land sources is not favourable for harmful algal blooms as DSi is present in excess over the other nutrients and can support the diatom communities. Overall, most rivers have low ICEP scores, with only two rivers having an ICEP score higher than -1 , six rivers higher than -2 , and 16 rivers higher than -5 .

The northern rivers were characterized by low fluxes of all nutrients, including DSi. These rivers are important for hydro-power production, with many of the dams located upstream in the river system, thus only retaining DSi from a small part of the total river basin area. Despite overall lower retention rates, total nutrient fluxes stay relatively low due to a lower weathering rate as well as a low level of human activity. In contrast, the river basins in the southwest are all characterized by high DSi fluxes due to the elevated weathering in this region. This provides a sufficient amount of excess DSi and explains the negative ICEP values even in basins heavily dominated by agriculture.

DSi fluxes in the rivers Norrström, Motala Ström and Göta Älv were all heavily affected by retention in large lakes within their basins. The importance of the lakes in Motala Ström is discussed above in Section 3.3. Lake Vänern, the third largest lake in Europe with a turnover time of 16 years, is in the Göta Älv river basin. The relatively high DSi yield in this basin compared to the other two is due to higher weathering rates in the southwest and further inflows of DSi from tributaries downstream of Lake Vänern. Lake Mälaren, the third largest lake in Sweden, is at the outlet of River Norrström. Much of the DSi generated upstream was retained here, resulting in the lowest DSi yield and the highest (albeit negative) ICEP value.

4 Discussion

4.1 Implementation of silica routines in HYPE

A strengths, weaknesses, opportunities, and challenges (SWOC) analysis was performed for the HYPE model and its new silica routines (Fig. 15). We discuss strengths and weaknesses of the existing implementation while opportunities and challenges explore the future possibilities.

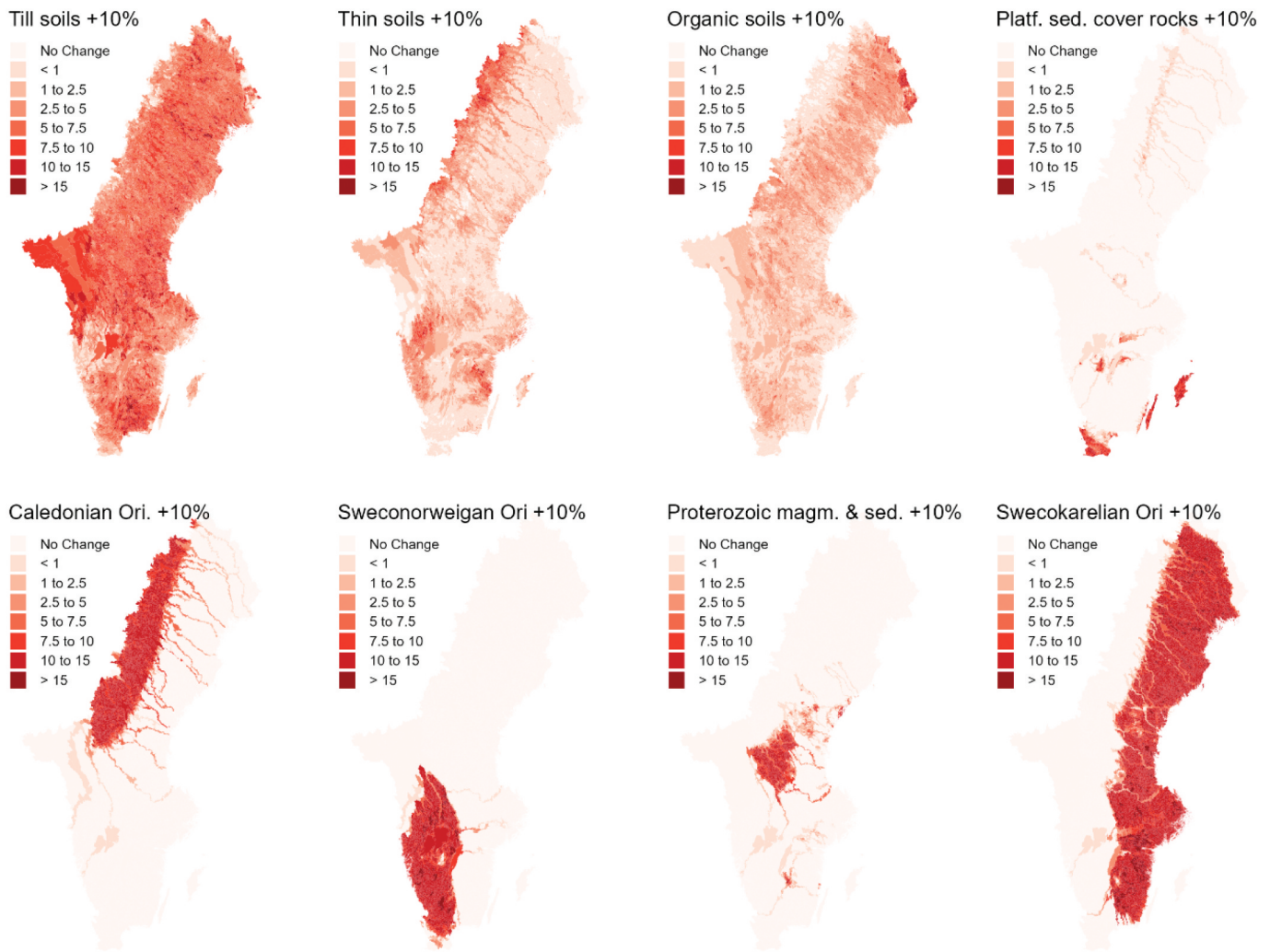


Figure 11. Percentage change in simulated riverine dissolved silica concentrations resulting from 10% increases in *weathrate* parameter values for soil types and *weathcorr* values for the lithotectonic units.

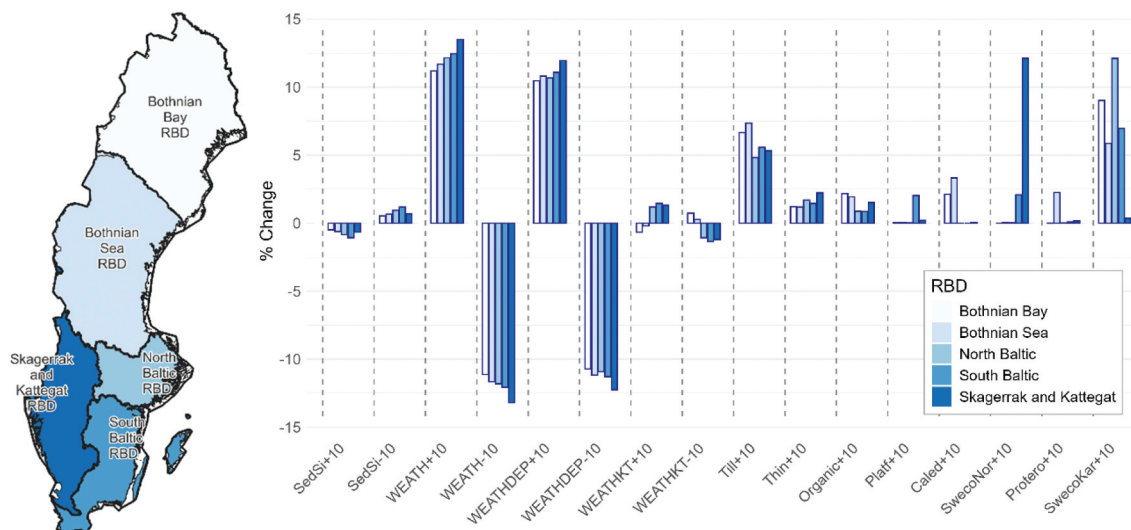


Figure 12. Percentage change in simulated riverine dissolved silica transport to the coast of the five River Basin Districts of Sweden as a result of changes in parameter values in the sensitivity analysis.

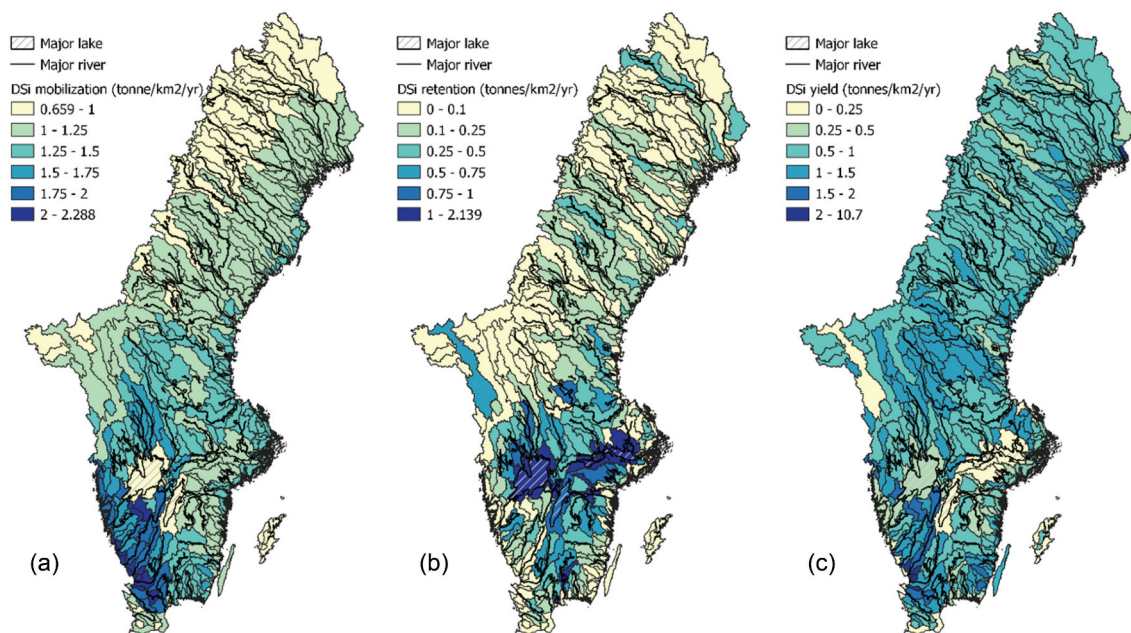


Figure 13. Simulated yearly dissolved silica (DSi) mobilization (a), simulated retained silica in surface water (b), and simulated yearly DSi load per upstream area (c). Results are aggregated from sub-basin to main river basin or large tributary scale. Results for load and yield represent model results at basin outlets but are rendered in each basin polygon.

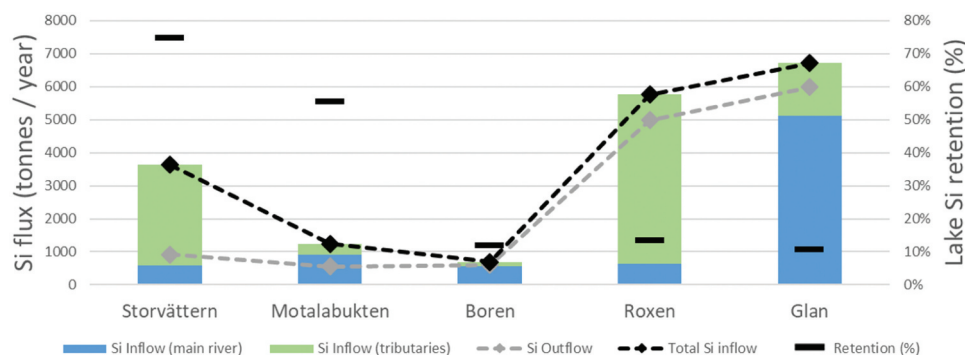


Figure 14. Lake silica inflow, outflow (left y-axis) and retention (right y-axis) in the major lakes that the Motala Ström River passes on its way to the coast, as simulated by the HYPE model. Storvättern and Motalabukten are two lake basins of Lake Vättern.

4.1.1 Strengths

The SWOC analysis identified several key strengths. The implemented silica simulation routines align with the overall complexity of the HYPE model, allowing for the simulation of large spatial domains at small spatial and temporal scales (i.e. individual rivers and lakes at daily time steps) while remaining responsive to the main processes that drive silica mobilization and delivery. The model explicitly simulates important sinks of DSi such as lakes, reservoirs, and dams including regulations associated with, for example, hydropower production (Conley *et al.* 2000).

In comparison to existing models for assessing and quantifying DSi fluxes, HYPE is a spatially and temporally explicit, integrated hydrological and multi-substance water quality model simultaneously calculating discharges and water quality concentrations for a large domain. The impacts of individual substances on each other (e.g. how

P fluxes are tied to sediment fluxes) can be assessed in a consistent way because all relevant substances – such as N, P, silica, and sediments – are integrated directly in HYPE.

Furthermore, HYPE's HRU spatial discretization (see Section 2.1) allows HYPE to be calibrated to gauged basins and then applied to ungauged basins, making the model suitable for use in data-scarce regions as well as simulations of large-scale domains. Finally, set-up, calibration, analysis, and visualization of HYPE models is straightforward thanks to the accompanying HYPETOOLS R package (Brendel *et al.* 2024), which provides an extensive suite of tools designed specifically to support HYPE model development.

4.1.2 Weaknesses

There are several notable limitations to the implementation of DSi simulations in the version of HYPE used in this paper (version 5.15.10). This version only supports constant land use, vegetation type, and crop management during the

Table 3. River basin characteristics and calculate Indicator of Coastal Eutrophication Potential (ICEP) values for 25 Swedish rivers. The rivers are listed in order from the north coast of the Baltic Sea, following the Swedish coastline to the south and west. River Basin Districts: BB – Bothnian Bay, BS – Baltic Sea, NB – North Baltic, BS – South Baltic, SK – Skagerrak and Kattegat.

Name	River basin district	Basin area (10 ³ km ²)	Land use (%)		Flux (kg km ⁻² yr ⁻¹)			Molar ratio (mol:mol)			Lim. nut.	ICEP (kg C km ⁻² day ⁻¹)
			Lakes/ dams	Arable	N	P	DSi	Si/N	Si/P	N/P		
Torneälven	BB	39.8	4.5	0.2	89	7	436	2.5	69	28	P	-1.9
Kalixälven	BB	18.1	3.3	0.2	188	10	793	2.1	92	43	P	-3.9
Råneälven	BB	4.2	3.3	0.4	136	6	809	3.0	141	47	P	-4.3
Piteälven	BB	12.3	7.1	0.6	122	7	555	2.3	94	41	P	-2.7
Luleälven	BB	25.2	9.1	0.3	120	4	740	3.1	209	68	P	-4.2
Skellefteälven	BB	11.7	12.7	0.5	136	3	779	2.9	306	107	P	-4.5
Umeälven	BB	26.8	7.1	0.5	126	4	783	3.1	210	68	P	-4.4
Ångermanälven	BS	31.8	8.1	0.5	122	4	739	3.0	216	71	P	-4.2
Indalsälven	BS	26.7	9.0	1.4	136	3	926	3.4	360	106	P	-5.4
Ljusnan	BS	19.8	5.1	1.5	106	4	994	4.7	313	67	P	-5.8
Dalälven	BS	28.9	6.5	2.4	157	5	883	2.8	179	64	P	-4.9
Norrström	NB	22.6	11.0	17.0	132	5	165	0.6	34	54	P	-0.4
Motala Ström	SB	15.4	20.0	13.5	149	6	283	1.0	54	57	P	-1.1
Söderköpingsån	SB	0.8	8.6	18.9	240	21	648	1.4	35	26	P	-1.7
Emån	SB	4.5	5.9	7.2	224	6	908	2.0	177	87	P	-5.0
Mörrumsån	SB	3.4	12.5	7.1	223	7	421	1.0	72	76	P	-1.9
Helge Å	SB	4.7	4.7	14.4	539	14	1438	1.3	118	88	P	-7.4
Kävlingeån	SB	1.2	2.1	57.1	1417	25	793	0.3	35	124	P	-2.1
Vege Å	SK	0.5	0.1	58.0	1769	56	1127	0.3	22	70	P	-0.7
Rönne Å	SK	1.9	3.2	23.7	891	24	1546	0.9	72	84	P	-7.0
Lagan	SK	6.4	8.5	7.9	368	10	1162	1.6	131	83	P	-6.1
Ätran	SK	3.3	5.5	10.1	539	15	1445	1.3	108	81	P	-7.3
Göta Älv	SK	48.1	18.3	9.4	271	5	748	1.4	155	112	P	-4.1
Bäveån	SK	0.3	5.4	10.9	444	24	1388	1.6	63	40	P	-5.9
Örekilsälven	SK	1.3	3.8	14.0	449	28	1497	1.7	59	35	P	-6.1

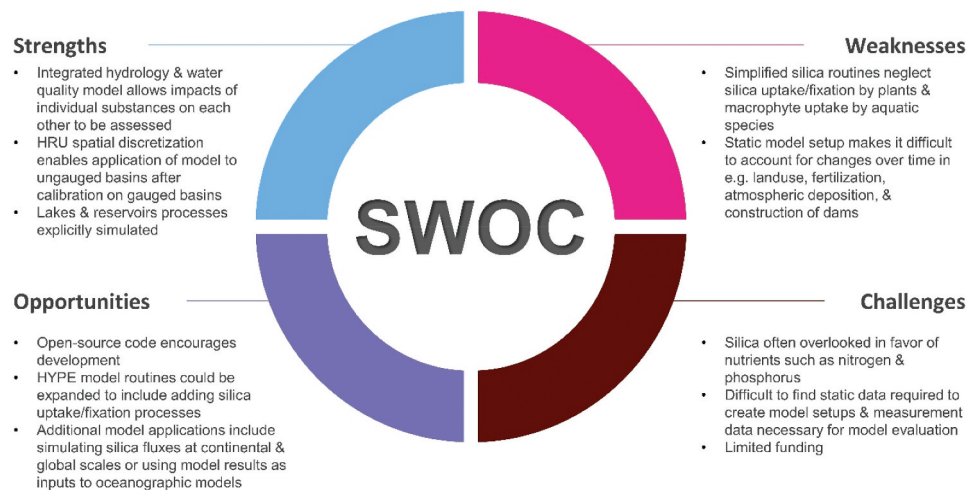


Figure 15. Strengths, Weaknesses, Opportunities, and challenges (SWOC) analysis of the implementation of silica routines in S-HYPE.

simulated time period. Consequently, the impacts of temporal changes in landscape or physical soil properties on DSi fluxes are not reflected in the model simulations in this study. This limitation may introduce biases in model calibration and validation, particularly in cases where the model's physical representation does not accurately represent the actual situation when the DSi measurements were taken. It is worth noting that the ability to simulate time-dependent land use and crop management changes was recently implemented in HYPE code version 5.31.0 (released 10 April 2025), but this release occurred after the work in this paper was already performed. Besides land use, there are other static data in S-HYPE (e.g. crops, point sources, dams, etc.) making it difficult to account

for changes over time such as altered fertilization, changes in atmospheric deposition, and construction of dams.

While HYPE can be described as a process-based model, many of the processes it simulates are necessarily represented in a simplified form, either due to data limitations or to maintain computational efficiency for large scale applications. A choice to exclude certain processes from the HYPE silica routines may limit basins where HYPE can be successfully applied. Specifically, HYPE is expected to underperform in areas where silica uptake and fixation by plants (Carey and Fulweiler 2012b) or macrophyte uptake by aquatic and wetland species (Schoelynck *et al.* 2010) significantly affects the DSi budget.

4.1.3 Opportunities

Several noteworthy opportunities exist in terms of the future development and application of the HYPE silica routines. Information on silica concentrations in freshwater is of potential interest to scientists and managers studying e.g. plankton communities in freshwater, coastal areas, or seas. A major advantage of the HYPE model is that the source code is open-source, which encourages development from the HYPE and modelling communities. All users of HYPE could include additional processes that are ignored in the current model routines (see Section 4.1.2) as it becomes relevant for their applications. Similarly, the ICEP indicator can be implemented directly into HYPE as an output variable to simplify the postprocessing.

The implementation of time-dependent land use in the newly released HYPE version 5.31.0 presents a novel opportunity to use HYPE to assess how riverine transport of DSi and coastal eutrophication potential changes in time or how it may be impacted under various development scenarios and Shared Socioeconomic Pathways (SSPs). Additional potential applications of HYPE silica simulations include using the model outputs as input to oceanographic and coastal models and simulating silica fluxes at the continental or global scales.

4.1.4 Challenges

The most significant challenge related to the HYPE silica routines is that silica is often overlooked by management and monitoring agencies in favour of nutrients such as N and P. Because of this, it can be increasingly more difficult to find not only the information needed to describe sources of silica, but also the measurement data needed to calibrate and validate the models. This is especially challenging for large-scale continental models such as E-HYPE (Brendel *et al.* 2023) and global models such as WWH (Arheimer *et al.* 2020), which require data for large domains in standardized formats.

Future studies focusing on silica may also be limited as it can be more difficult to find funding for projects focused on silica than for projects focused on nutrients. Addition of silica may not always be a cost-effective use of resources to study (for example) eutrophication, as its importance in aquatic community composition may vary with local conditions.

4.2 Quantification of DSi sources and transport in Sweden

The national-scale simulation of DSi mobilization and transport provides insights into the spatial patterns of weathering, delivery, and retention across Sweden's river basins. By implementing a process-based representation of silica mobilization, driven by chemical weathering, and simulating its downstream transport, we were able to generate spatially resolved estimates of DSi fluxes to coastal waters. The model output includes a national map of weathering rates (Fig. 6), representing a rare attempt to spatially quantify silica mobilization at this scale. Previous efforts to simulate chemical weathering in Sweden used models such as PROFILE (Sverdrup and Rosén 1998, Akselsson *et al.* 2004) and ForSAFE (Akselsson *et al.* 2019), a dynamic ecosystem model that builds on the steady-state PROFILE by incorporating time-dependent processes like

vegetation growth, hydrology, and nutrient cycling. The MAGIC model, estimating weathering indirectly using a mass balance approach rather than mechanistic simulations, was also applied extensively in Swedish catchments to estimate weathering (Moldan *et al.* 2013). These studies were primarily focused on acidification recovery and base cation fluxes, with little direct emphasis on silica. Similarly, observational studies have provided weathering estimates in specific catchments, but systematic national mapping of silica weathering remains limited.

Calibrating weathering parameters using a mass-balance approach enhances confidence in the accuracy of the simulated weathering rates. While some correlation exists between landscape characteristics (such as dominant soil types and geological features, Fig. 3) and meteorological variables (Fig. 2), these are not strong enough to invalidate the calibration method or to suggest parameter equifinality. This is further demonstrated in the sensitivity analysis, which shows that altering individual factors results in spatially distinct effects on DSi levels (Fig. 11).

The accuracy of the simulated national-scale patterns of DSi transport and retention (Fig. 13) is best evaluated through comparison of modelled and observed concentration time series (Table 2; Fig. 7). Few stand-out spatial patterns are seen in the map, or patterns that can be attributed to a specific landscape attribute (Fig. 8). A tendency towards underprediction at sites with low mean annual temperatures, particularly in the northern stations, is evident in Fig. 7a–b. This bias may be reduced by recalibrating the temperature sensitivity parameter *weatKT*, as suggested by the sensitivity analysis results (Fig. 10).

The contribution of different landscape units (Fig. 12) to the total DSi load to coastal areas reflects not only their relative areal extent but also their position within the catchment. Interestingly, the changes in DSi transport exceed the changes in mobilization resulting from adjustments to the *weathrate* parameter. This may be explained by the fact that DSi concentrations are close to zero during parts of the year in some water bodies. When silica input to these environments increases, it can begin to exceed the systems' retention capacity, leading to a disproportionately higher relative throughput.

Lakes and reservoirs can have a significant impact on overall DSi transport through retention processes, as seen in the national simulations (Fig. 13). This is in line with other studies such as Humborg *et al.* (2006, 2008). The large impact of lakes on DSi transport is also demonstrated in the modelling results from the Motala Ström basin (Fig. 14). The retention estimate for Lake Vättern falls within the range reported by Conley *et al.* (2000), based on observational measurements. The model evaluation indicates that the HYPE model can simulate surface water retention processes reasonably well. Lakes and dams have a smoothing impact on the riverine concentration signal and induce a stronger seasonal pattern, which the model's surface water processes appear to be able to replicate. This can also explain why the model in some aspects performs better for the validation sites compared to the calibration sites (Table 2), as calibration sites are less affected by large water bodies. Figure 8 also indicates that an increased volume

of water accumulated in lakes within the upstream drainage area does not introduce any bias in the model results at the observation stations. The model has, however, a rather simplistic representation of key surface water processes, and the model's utility in freshwater management could be further enhanced by incorporating the ability to simulate the dynamics of key algal groups, as suggested by Pyo *et al.* (2019).

4.3 Implications for coastal management

The importance of silica for coastal ecosystems is well established, particularly in relation to the growth of diatoms, which are essential components of marine food webs (e.g. Conley *et al.* 1993). In many cases, it is critical for coastal management to consider terrestrial inputs, given the potentially large inflows of freshwater and nutrients from rivers into coastal environments. The magnitude of the inflows as well as ratios between key nutrients – such as N, P, and silica – can significantly influence the coastal response to eutrophication.

Dynamic models, such as HYPE, can be used to assess changes at river mouths resulting from catchment alterations, including impoundments, agricultural intensification, and shifts in nutrient deposition patterns. Bartosova *et al.* (2019) quantified nutrient loads to the Baltic Sea from individual RBDs, in this case from all contributing countries, with a Baltic HYPE model and compared them with the Fifth Baltic Sea Pollution Load Compilation (PLC5.5, HELCOM 2015). Both PLC5.5 and Baltic HYPE also indicated the river outflows were phosphorus limited, albeit their N/P ratios were lower than in this study (18–28) for the Bothnian Bay, Bothnian Sea, and Baltic Proper (sub-divided into Baltic North and Baltic South in this study).

The ability to estimate not only N and P, but also DSI fluxes in a consistent manner is critical in our understanding of impacts of riverine fluxes on the receiving sea or coastal areas now or under future scenarios. ICEP can serve as a valuable tool to evaluate and quantify the impacts of these changes relative to a baseline, offering preliminary insights into their effects on coastal waters. Furthermore, ICEP can help link land-based nutrient sources with other contributors, especially in areas with known eutrophication issues. These may include internal nutrient loading from sediments, direct point source discharges, biological N fixation, and net nutrient exchange with offshore waters. However, due to the complexity of additional pressures and internal biogeochemical processes affecting coastal water bodies, the ICEP indicator should not be used as a standalone screening tool for identifying areas at risk of eutrophication-driven water quality degradation. Coastal models such as Edman *et al.* (2018) provide deeper insight into local biogeochemical processes and potential impacts.

For many of the rivers the ICEP scores are comparable to those reported for Swedish rivers by Billen and Garnier (2007), but slightly higher for the northernmost ones due to lower estimates of DSI fluxes in this study. This is also a region where DSI concentrations are underpredicted by the model (Fig. 7). Several rivers show ICEP close to zero; these rivers should be closely evaluated, especially in the context of climate change or changes in hydropower regulations. For

example, Bartosova *et al.* (2019) showed that P load may increase much more than N load due to climate change by the 2050s, changing the proportion unfavourably under the majority of the evaluated future socioeconomic scenarios. Changes in weathering and retention could also potentially lead to an increase in ICEP and threaten the existing uses of coastal waters.

5 Conclusions

This study presents a comprehensive modelling framework to support large-scale quantification of DSI transport. The framework, built around the concepts of mobilization and delivery, is implemented in the HYPE model. An applied calibration strategy is designed to make optimal use of available data while maintaining confidence in the model's predictive capacity – particularly in ungauged basins. The approach integrates a step-wise calibration method, linking parameter values to landscape characteristics, such as soil type and geology, using data from numerous observation sites. This approach enhances the model's robustness against overfitting and reduces the risk of equifinality, as evidenced by our results where the model performance does not deteriorate at validation sites compared to the calibration sites.

The open-source integrated hydrology and water quality model, HYPE, provides a tool for the spatially explicit simulation of water, nutrients, and DSI fluxes across large domains. This paper describes the development of HYPE's silica simulation routines as well as a first application of these routines to assess riverine transport of DSI and coastal eutrophication potential in Sweden. The primary conclusions drawn from this work are summarized as follows:

- The step-wise calibration scheme in which the silica weathering parameters were calibrated first, followed by the surface water process parameters, provided a robust approach to capitalize on available monitoring data while allowing for the assessment of the relative importance of various DSI weathering drivers. However, there is an inherent risk that co-variation between different factors affecting weathering could impact the quantification of their relative importance.
- The application of HYPE's silica routines to the Swedish domain demonstrated that the calibrated model could reasonably simulate the spatial and temporal patterns of DSI concentrations in Swedish watercourses over a wide range of hydro-meteorological conditions.
- Assessment of silica fluxes in Sweden indicated that DSI transport was lowest in areas greatly influenced by lakes and dams due the retention of DSI in lakes and reservoirs as well as a lack of weathering in inundated areas.
- The Indicator of Coastal Eutrophication Potential (ICEP) values calculated from multi-nutrient simulations for 25 large rivers in Sweden were all negative, which indicates that Swedish rivers contribute to a favourable nutrient ratio in Swedish coastal waters.

Although Sweden was used as an example to test and evaluate HYPE's silica routines, the methodology and calibration

scheme described here can be applied to other geographic domains and scales. Integration of the HYPE silica model routines with nutrients and hydrological routines supports the assessment of potential changes to the aquatic environment as a result of, for example, (1) climate change, (2) nutrient reduction scenarios, (3) building or removal of dams, or (4) changes in hydropower regulations. In addition, silica may modulate the effect of N and P loading on coastal environments with regard to eutrophication, and is thus worth considering in assessments of nutrient load–response studies.

Disclosure statement

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