



Transformation of silicon in a sandy beach ecosystem: Insights from stable silicon isotopes from fresh and saline groundwaters

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ABSTRACT

Dissolved silicon isotope compositions ($\delta^{30}\text{Si}$) have been analysed for the first time in groundwaters of beach sediments, which represent a subterranean estuary with fresh groundwater discharge from a freshwater reservoir and mixing with recirculated seawater. The fresh groundwater reservoir has high and variable dissolved silica concentrations between 136 and 736 μM , but homogeneous $\delta^{30}\text{Si}$ of $+1.0 \pm 0.15\%$. By contrast, the seawater is strongly depleted in dissolved silica with concentrations of 3 μM , and consequently characterised by high $\delta^{30}\text{Si}$ of $+3.0\%$. The beach groundwaters are variably enriched in dissolved silica compared to seawater (23–192 μM), and concentrations increase with depth at all sampling sites. The corresponding $\delta^{30}\text{Si}$ values are highly variable ($+0.3\%$ to $+2.2\%$) and decrease with depth at each site. All groundwater $\delta^{30}\text{Si}$ values are lower than seawater and most values are lower than dissolved $\delta^{30}\text{Si}$ of freshwater discharge indicating a significant amount of lithogenic silica dissolution in beach sediments. In contrast to open North Sea sediments, diatom dissolution or formation of authigenic silica in beach sediments is very low (ca. 5 $\mu\text{mol Si g}^{-1}$). Silica discharge from the beach to the coastal ocean is estimated as approximately 210 mol Si yr^{-1} per meter shoreline. Considering the extent of coastline this is, at least for the study area, a significant amount of the total Si budget and amounts to ca. 1% of river and 3.5% of backbarrier tidal flat area Si input.

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1. Introduction

Dissolved silica is one of the major nutrients in the oceans to sustain growth of diatoms and other siliceous organisms. Most of the dissolved silica enters the oceans via rivers and benthic silica fluxes from the sediments (Tréguer and De La Rocha, 2013), whereby the amount of benthic flux is largely controlled by the dissolution of biogenic silica at the sediment-water interface. Studies from the southern bight of the North Sea show that especially during summer, biogenic silica dissolution in the sediments can contribute up to 50% of the total dissolved silica input to the water (Rutgers van der Loeff, 1980; Gehlen et al., 1995). In contrast, the amount of biogenic and lithogenic silica dissolution in marine sediments and its influence on the total dissolved silica budget is not well constrained. Studies suggest that in general lithogenic material deposited along ocean margins may release considerable amounts of dissolved silica (Tréguer and De La Rocha, 2013, and references therein). However, experimental and field studies show that the dissolution of silica is initially rapid, but the increasing dissolved silica concentration in pore

waters results in saturation of the solution. In addition, together with aluminium (Al), iron (Fe), potassium (K) and magnesium (Mg), silica can be removed by precipitation of authigenic aluminosilicates (e.g., Mackin, 1987; Loucaides et al., 2010; Michalopoulos and Aller, 2004), which also reduces the diffusive benthic flux of dissolved silica from marine sediments. In permeable sandy marine sediments of the North Sea, however, the frequent high water flow rates of several centimetres per hour (Huettel and Gust, 1992) allow dissolved species to be transported out of the sediments at least $50\times$ faster than via molecular diffusion (Huettel and Webster, 2001), and therefore reduce the accumulation of regenerated nutrients in pore waters. This effect potentially enhances dissolution rates of (biogenic) silica by increasing the degree of silica undersaturation (Ehrenhauss et al., 2004).

In permeable beach sediments tidal and wave forcing increase advective flow and salt transport in the near shore aquifer (Robinson et al., 2007), which enhances the exchange between the aquifer and ocean and the mixing between terrestrial-derived fresh groundwater and seawater in a subterranean estuary. Therefore, beaches represent potentially important reaction zones in the near shore aquifer, where land- and ocean-derived particles and solutes might undergo important biogeochemical transformation before they are discharged to the ocean

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(Moore, 2010). For example, Anschutz et al. (2009) could show that permeable intertidal beach sediments of the Aquitanian coast accelerate the remineralisation of coastal organic matter and supply high amounts of dissolved inorganic phosphorus and nitrogen to the coastal ocean representing up to 5% relative to the supply of the nearest estuary. In addition to seawater recirculation, beaches represent a zone of freshwater discharge to the ocean. Because nutrient concentrations in coastal groundwater may exceed surface seawater concentrations by several orders of magnitude, groundwater input may be a significant source of nutrients to sustain benthic and planktonic productivity in the coastal regions (Hays and Ullman, 2008; Moore, 2010). For example, Georg et al. (2009a) reported that annual dissolved silica fluxes by groundwater discharge into the Bay of Bengal are equivalent to 40% of the total river input. However, it often becomes difficult to determine the fraction of nutrient input originating from freshwater and its subsequent transformation processes within beach sediments, when coastal groundwater, which is affected by *in situ* remineralisation of organic matter of marine origin, is admixed (Hays and Ullman, 2008).

In this study we present for the first time the stable silicon isotope compositions ($\delta^{30}\text{Si}$) of fresh and saline groundwaters along a beach transect on Spiekeroog Island (Fig. 1) in order to investigate mixing and transformation processes associated with the transfer of dissolved silica from land to ocean in a subterranean estuary. In general, the $\delta^{30}\text{Si}$ of fresh groundwater was reported to be systematically lower than in rivers or ocean water due to dissolution of isotopically light secondary silica phases like clays and silcretes (Georg et al., 2009a, 2009b; Pogge von Strandmann et al., 2014). Therefore, fresh groundwater may potentially provide a distinct isotope signature for groundwater discharge and mixing with isotopically heavier seawater along the beach transect. On the other hand, transformation processes like lithogenic silica dissolution, incorporation of biogenic silica, mostly from marine diatoms, into the beach sediments and subsequent dissolution as well as secondary authigenic silica formation through precipitation will affect the groundwater dissolved $\delta^{30}\text{Si}$ (e.g., Georg et al., 2009a, 2009b; Ehlert et al., 2016). Therefore, $\delta^{30}\text{Si}$ in the groundwater in combination with other water constituents and salinity can provide information about the dominant processes in such environments.

2. Study area

Sampling took place on Spiekeroog, which forms part of the East Frisian barrier island chain along the German North Sea coast (Fig. 1). Together with tidal sand and mud flat areas, they represent the highly protected ecological system of the World Heritage Site “Wadden Sea”. Spiekeroog has a length of ca. 9.8 km and a maximum width of ca. 2 km and is subject to a semidiurnal mesotidal regime with a tidal

range of approximately 2.6 m at the western head of the island (Reuter et al., 2009). The island morphology is characterised by large dry dunes, which are supplied and shaped by prevailing westerly winds. The sediments mostly consist of fine- to coarse-grained quartzitic sands with a few separate clay lenses (Streif, 1989). Regular rainwater infiltration into the permeable sediments has led to the formation of a freshwater reservoir below the main dune area in the western part of the island (Fig. 1) (Röper et al., 2012). Average annual precipitation on Spiekeroog is about 750 mm (OOWV, 2009). Due to the high permeability of the sandy sediments about 300–400 mm rapidly drains into the subsurface (Röper et al., 2012); the remaining precipitation evaporates and no significant surface runoff (rivers, ditches or creeks) exists. A clay layer at approximately 50 m below mean sea level is assumed to act as an aquitard (OOWV, 2009). It divides fresh from underlying salty groundwater, although it might be permeable to a certain degree. The outer parts of the freshwater lens consist of a transition zone towards brackish water due to gradual exchange with the surrounding salty groundwater (Röper et al., 2012).

The seawater in the southern North Sea is generally characterised by a strong influence of river discharge by the rivers Scheldt/Rhine and Elbe, whereas the area around Spiekeroog is also affected by the smaller rivers Ems and Weser (Röper et al., 2012).

A detailed description of the beach morphology and hydrodynamics of the adjacent North Sea is also available in Beck et al. (under review).

3. Material and methods

3.1. Sampling and analyses of water samples

All samples were collected during a joint sampling campaign in May 2014 (see also Beck et al., under review for details). The groundwater samples from the freshwater lens were taken at seven sites in the dune area (Figs. 1, 2, Table 1). At each site three different well depth intervals were sampled (Fig. 2), whereby A and B are the shallower wells with fresh- or brackish water and C is the deeper well with saline water. The water was pumped from the respective wells/depths and filtered immediately through 0.8/0.2 μm AcroPak filter cartridges (Supor filter membrane) and stored in acid pre-cleaned polyethylene (PE) bottles.

The beach groundwaters were sampled along a Southwest-Northeast cross-shore transect from the dunes towards the low-water line, which represents a distance of approximately 350 m at five different main sites from 0.5 m to 5.2 m depth below sediment surface (Figs. 1, 3, Table 1). Additional samples for major ions and nutrients were taken at four additional sites to achieve higher resolution (Fig. 3, see also Beck et al., under review). Seawater samples were taken during low tide at ca. 0.5 m water depth by submerging PE-beakers. The

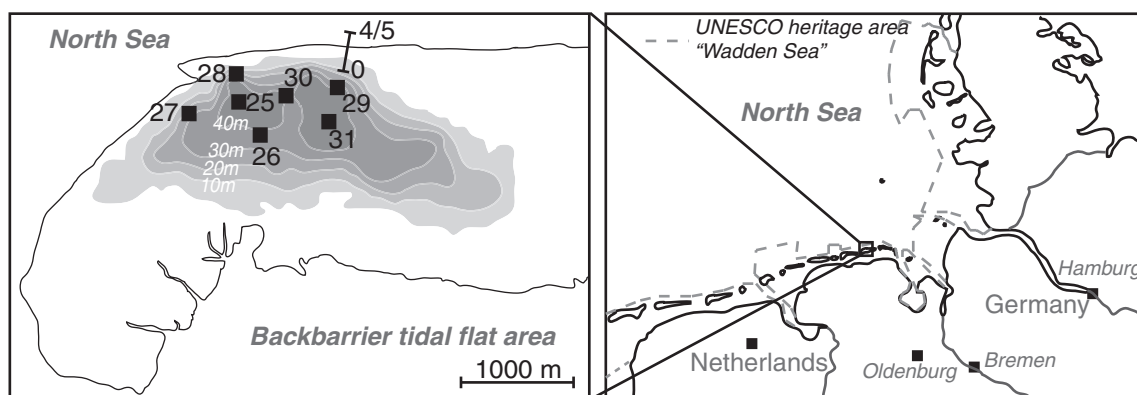


Fig. 1. Location of Spiekeroog in the southern North Sea and the sampling sites in the western part of the island (stations 25–31 – drinking water well sites from the OOWV for freshwater lens sampling, stations 0–4/5 – beach transect for groundwater and sediment sampling). The grey shading indicates the approximate extension depths of the base of the freshwater lens (after Röper et al., 2012).

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