



## Are hypersaline tidal flat soils potential silicon sinks in coastal wetlands?

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### ABSTRACT

Hypersaline tidal flats (HTF) are transitional ecosystems which play an important role over Si biogeochemistry in wetlands. Sources and sinks of Si in these ecosystems are directly dependent on pedogenesis and are strongly influenced by the readily soluble Si components, such as dissolved Si, adsorbed Si, and amorphous silicates. The aim of this study was to identify and assess the distribution of readily soluble Si pool in two HTFs on the Brazilian coast. We based our studies on sequential extractions of iron and silicon, modeling of the soil aqueous phase, and X-ray diffraction of the fine clay fraction. Our results show that mobile Si and adsorbed Si are present at very low levels in HTF soils, comprising < 3% of the readily soluble Si pool. Silicon associated with Fe oxyhydroxides is the second largest component of the readily soluble Si pool, making up 8.7–40.8% of the Si extractions. The correlation between the ferrihydrite fraction and Si associated with Fe oxyhydroxides indicates a large control of Si by ferrihydrite. Amorphous Si makes up the dominant fraction, comprising 59–90% of the Si extractions. SEM images show the presence of diatoms, sponges, and phytoliths in the soils. Precipitation of pedogenic opal seems to occur in the soils due to a dry season at both sites. The clay assemblages are composed of the mixed-layer kaolinite-smectite (K-S), kaolinite-illite (K-I), and illite-smectite (I-S), and lesser amounts of kaolinite (K) and smectite (S) end-members. The presence of these clays indicates that HTF soils are conducive to the formation of mixed-layer minerals, which partially control Si dynamics. Our data indicate that HTF soils tend to accumulate Si by adsorption, (co)precipitation, and reactions involving clay minerals.

### 1. Introduction

Silicon (Si) is the second most abundant element in the Earth crust and an essential nutrient for most living organisms in marine and terrestrial ecosystems (Conley, 2002; Sommer et al., 2006). This element is ubiquitous in soils and a basic constituent of soil liquid, adsorbed, and solid phases (Sauer et al., 2006). Even though most soils contain Si, the distribution of the different Si components within the Si pool varies widely depending on a broad array of factors, such as the influence of pedogenetic processes, parent material, soil chemical and physical properties (e.g. pH, redox potential, temperature, particle size distribution), intensity of leaching, biological cycling, and mineral formation and alteration (Monger and Kelly, 2002; Cornelis et al., 2014).

Regardless of which factors are involved, it is well known that the pedosphere plays a major role in controlling Si dynamics in environments (Derry et al., 2005; Sommer et al., 2006; Cornelis et al., 2011).

About 80% of dissolved Si input to the ocean is transported by rivers (Tréguer et al., 1995), and Si export to rivers is largely dependent on the soil formation processes (Conley, 2002; Borrelli et al., 2010; Struyf and Conley, 2012). Therefore, soils can promote the release of Si to marine and terrestrial ecosystems (Sommer et al., 2006; Struyf and Conley, 2009), or even retard Si output to rivers and oceans (Meunier et al., 1999).

The interplay of some specific soil conditions is essential to understand the Si dynamics in terrestrial and aquatic ecosystems (Cornelis et al., 2014; Georgiadis et al., 2017), such as ion activity, pH, and salinity (Drees et al., 1989). In general, the solubility of silica increases in soils affected by salts and under high pH values (Loucaides et al., 2008). pH values between 2.5 and 8.5 do not favor the release of Si from the soil solid phase. This process is enhanced in higher pH values due to the dissociation of monomeric silicic acid ( $\text{H}_4\text{SiO}_4$ ) to deprotonated forms (e.g.  $\text{H}_3\text{SiO}_4^-$  and  $\text{H}_2\text{SiO}_4^{2-}$ ). Nevertheless, the dissolution

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of silica in near-neutral pH can also take place due to the nucleophilic reaction of water at the solid-solution interface (Dove and Crerar, 1990). Dissolved silicon availability is also favored in high saline systems because of the higher catalytic effect of salt solutions (e.g. seawater, Loucaides et al., 2008). These conditions are commonly found in evaporative environments (Albuquerque et al., 2014a; Furquim et al., 2010b). In these ecosystems, the evaporative concentration of waters can cause an increase in soil salinity and pH values.

Hypersaline tidal flat soils (HTF) are evaporative environments distributed worldwide and commonly found on the Brazilian coast. These soils have very high salinities and commonly pH values above 7 (Albuquerque et al., 2014b) and therefore may be of high importance for the biogeochemical cycling of nutrient silicon across the land to ocean transition. Thus, research is needed to provide insights into the role of these soils in controlling the Si dynamics in coastal ecosystems. This study aims to 1) identify the main Si pools in HTF soils and estimate the distribution of its components, and 2) define the processes involved in sources and sinks of Si in HTF soils. To address these objectives, two HTFs were studied based on sequential extractions of Si and Fe, modeling of the soil aqueous phase, and X-ray diffraction of the fine clay fraction.

## 2. Environmental settings

The hypersaline tidal flats (HTFs) selected for this study are located in different regions on the Brazilian coast, with distinct climatic and geological conditions (Fig. 1). The site located in the city of Acaraú (Ceará State, NE Brazil) is within the Acaraú River Basin and is referred as HTF-CE in this study. The climate is Bw (mild semiarid according to Köppen), with an irregular rainfall distribution throughout the year and a dry season taking place between July and December. The average annual precipitation is approximately 1200 mm and the evapotranspiration of about 1700 mm. The regional geology of Acaraú River Basin consists of Pre-Cambrian metamorphic rocks (gneisses, schists, and phyllites). The sediments reaching the Acaraú estuary are composed of Cenozoic sand- and clay-size from the Barreiras Tablelands, consisting of well-crystallized kaolinites and quartz. This geologic

formation encompasses the study area (Bezerra et al., 2001; Hesp et al., 2009). Another source of sediments contributing to the soil formation in the HTF-CE is the poorly-developed soils formed under semiarid conditions (Luvisols, Planosols, Leptosols, Cambisols, Calcisols, and Durisols), which is rich in Al-rich 2:1 clay minerals.

The second study site is in the city of Rio de Janeiro (Rio de Janeiro State, SE Brazil). This site is part of the Sepetiba-Guaratiba coastal complex and is referred as HTF-RJ throughout the paper. The climate is Aw (tropical wet-dry according to Köppen) with a short dry season between June and August. The average annual precipitation is approximately 1016 mm and the evapotranspiration of about 1450 mm. The regional geology is composed of Quaternary coastal plains and Pre-Cambrian crystalline basement, the latter composed of granites, gneisses, and migmatites (Borges and Nittrouer, 2015). The inland soils encompassing the estuary have been formed under intense weathering and leaching, leading to the formation of soils rich in kaolinite and gibbsite, been the main source of sediments to the HTF-RJ soils (Furian et al., 2002; Mendonça-Santos et al., 2007).

Both HTFs have an irregular distribution of halophyte plants on the soil surface and are flooded several days throughout the month with major tidal flooding during spring tides.

## 3. Materials and methods

Three soil profiles were sampled at each site during low tide (Fig. 1). Soil profiles were labeled as P1CE, P2CE, and P3CE (HTF on the NE coast, HTF-CE), and P1RJ, P2RJ, and P3RJ (HTF on the SE coast, HTF-RJ). Detailed soil morphological descriptions were made of each soil profile at both sites before sampling the selected soil horizons.

Three soil horizons of each soil profile were chosen for the sequential extractions of Fe and Si: i) surface horizons (A horizons of P1CE, P2CE, P3CE, P1RJ, P2RJ, and P3RJ), ii) intermediary horizons (C3 of P1CE, 2C2 of P2CE, 2C1 of P3CE, C2 of P1RJ, C1 of P2RJ, and C1 of P3RJ), and iii) deeper horizons (3C4 of P1CE, 3C3 of P2CE, 3C2 of P3CE, 2C3 of P1RJ, 3C3 of P2RJ, and 3C3 of P3RJ). Collected samples were maintained at 4 °C during transportation to the laboratory. A portion of the samples was air dried in preparation for sequential

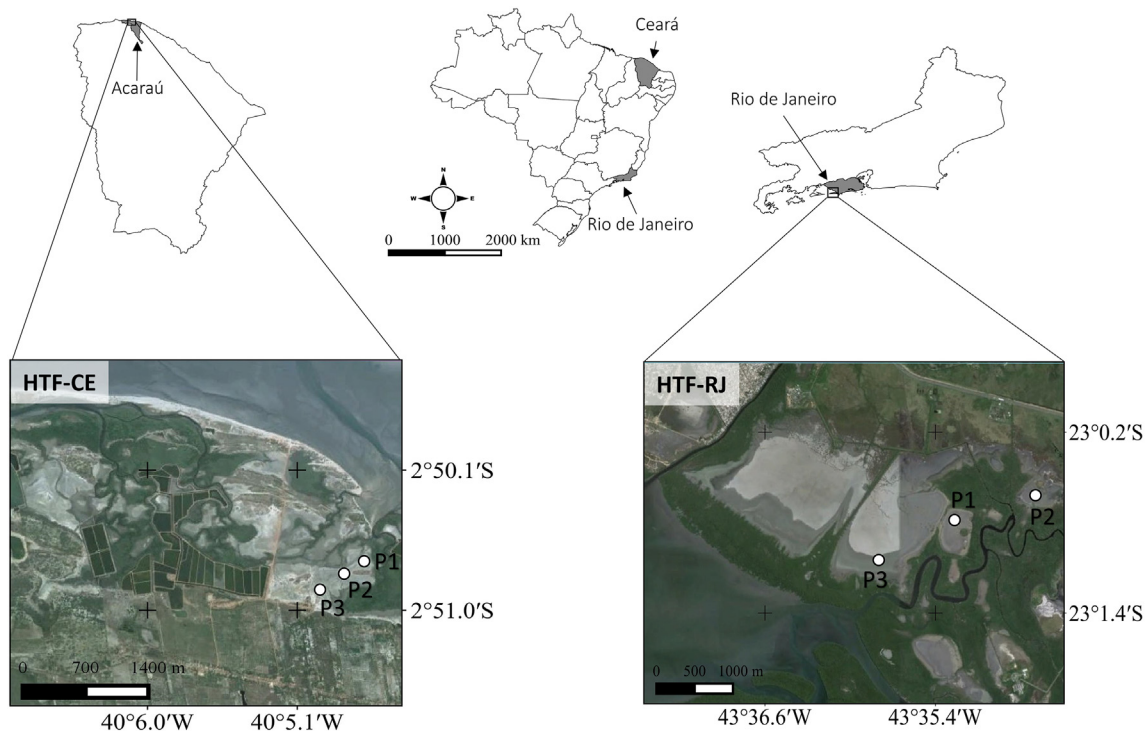


Fig. 1. Location of the study sites and soil profiles. Images source: Google™ Earth (2015).

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