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Research article

Redox-induced mobilization of copper, selenium, and zinc in deltaic soils originating from Mississippi (U.S.A.) and Nile (Egypt) River Deltas: A better understanding of biogeochemical processes for safe environmental management

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A B S T R A C T

Studies about the mobilization of potentially toxic elements (PTEs) in deltaic soils can be challenging, provide critical information on assessing the potential risk and fate of these elements and for sustainable management of these soils. The impact of redox potential (E_H), pH, iron (Fe), manganese (Mn), sulfate (SO_4^{2-}), chloride (Cl^-), aliphatic dissolved organic carbon (DOC), and aromatic dissolved organic carbon (DAC) on the mobilization of copper (Cu), selenium (Se), and zinc (Zn) was studied in two soils collected from the Nile and Mississippi Rivers deltaic plains focused on increasing our understanding of the fate of these toxic elements. Soils were exposed to a range of redox conditions stepwise from reducing to oxidizing soil conditions using an automated biogeochemical microcosm apparatus.

Concentrations of DOC and Fe were high under reducing conditions as compared to oxidizing conditions in both soils. The proportion of DAC in relation to DOC in solution (aromaticity) was high in the Nile Delta soil (NDS) and low in the Mississippi Delta soil (MDS) under oxidizing conditions. Mobilization of Cu was low under reducing conditions in both soils which was likely caused by sulfide precipitation and as a result of reduction of Cu^{2+} to Cu^{1+}. Mobilization of Se was high under low E_H in both soils. Release of Se was positively correlated with DOC, Fe, Mn, and SO_4^{2-} in the NDS, and with Fe in the MDS. Mobilization of Zn showed negative correlations with E_H and pH in the NDS while these correlations were non-significant in the MDS. The release dynamics of dissolved Zn could be governed mainly by the chemistry of Fe and Mn in the NDS and by the chemistry of Mn in the MDS. Our findings suggest that a release of Se and Zn occurs under anaerobic conditions, while aerobic conditions favor the release of Cu in both soils. In conclusion, the release of Cu, Se, and Zn under different reducing and oxidizing conditions in deltaic wetland soils should be taken into account due to increased mobilization and the potential environmental risks associated with food security in utilizing these soils for flooded agricultural and fisheries systems.

1. Introduction

Deltaic soils are distributed globally, primarily located where the world's major rivers reach the sea. The Nile River is the longest river in the world and it originates near the equator and flows nearly 7000 km northward from 4° S to 32° N with a catchment area of about 3.1 million km^2 (Bruggers, 2010). Consequently, the Nile Delta is one of the world's largest river deltas and covers the area between Cairo and the shoreline of the Mediterranean Sea. The delta comprises the two branches of the Nile River, the Damietta branch in the east and the Rosetta branch in the west (Bruggers,

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2010). The delta begins approximately 20 km north of Cairo and extends north for about 150 km. At the coast, the delta is about 250 km wide, from Alexandria in the west to Port Said in the east with an area of ~20,000 km^2 (Bruggers, 2010). The soils of the Nile Delta region are predominantly Fluvisols. The Nile Delta has very fertile soils, is a rich agricultural region and is the largest area of vegetation in Egypt. The delta represents an important sector for the Egyptian national economy. However, the soils of the floodplains have become less productive since the delta does not receive an annual supply of nutrients and sediments from upstream due to the construction of the Aswan High Dam. Therefore, large amounts of fertilizers and agrochemicals are now applied to the soils. As a consequence, the various anthropogenic activities, as well as the use of fertilizer, pesticides, and agrochemicals in the farmlands are dispersing contaminants including potentially toxic elements (PTEs) into the river sediments, soils, and waters (Shaheen and Tsadilas, 2009).

The Mississippi River has the largest discharge and drainage basin in North America (Day et al., 2005). The Mississippi delta is the largest contiguous coastal ecosystem in the U.S.A and was formed over the past 6000–7000 years as a series of overlapping delta lobes (Roberts, 1997). Among the world's largest coastal and river basins, the Lower Mississippi River Alluvial Valley is one of the most disturbed by human activities (Ouyang et al., 2013). Currently, only two of the distributaries of the river are functioning, the lower Mississippi River and the Atchafalaya River which carries about one-third of the total flow of the Mississippi River (Day et al., 2005). The Atchafalaya River is a distributary of the Mississippi River and receives ~30% of the flow of the Mississippi River. The area of the coast comprising the Atchafalaya River delta complex, which is composed of the Atchafalaya River sub delta and the Wax Lake sub Delta, is one of the very few places where active land building is occurring along the Gulf of Mexico coastline (DeLaune and White, 2012; Roberts et al., 2015).

There has been an increase in the area of wetlands over the past several thousand years (Day et al., 2005). Wetlands can be net sinks for PTEs. However, wetland soils and sediments may act as a source for pollutants resulting in adverse impact on the agricultural environment due to fluctuating reducing and oxidizing conditions in the soil (Shaheen et al., 2014; Rinklebe et al., 2016a,b). Wetland ecosystems have, a unique aerobic and anaerobic soil environment, both spatially and temporally. Redox processes play an important role in biogeochemical cycling of PTEs in flooded ecosystems (Frohne et al., 2011). The intensity of soil reduction can be rapidly characterized by soil oxidation–reduction (redox) potential (E_H), which allows for the prediction of the stability and dynamics of PTEs in soils (DeLaune and Seo, 2011). Deltaic wetland soils are particularly susceptible to shifting redox conditions as these systems typically receive river flood pulses (Roy et al., 2013). The solubility and release of PTEs under flooded conditions is largely controlled by soil E_H and pH, composition of dissolved organic carbon (DOC), iron (Fe)-manganese (Mn) oxides, chloride (Cl), and sulfate (SO_4^{2-}) (Shaheen et al., 2014; Rinklebe et al., 2016a,b). Thus, we hypothesized that oscillations from oxidizing to reducing conditions in soils originating from two major world rivers deltas would affect the release dynamics of three PTEs i.e., copper (Cu), selenium (Se), and zinc (Zn) due to changes in E_H/pH- values, aliphatic dissolved organic carbon (DOC), and aromatic dissolved organic carbon (DAC), and the related chemistry of sulfur (S), Cl^-, Fe, and Mn.

Copper and Zn are essential micronutrients for plants, animals, and humans; however, they can also be toxic pollutants if they occur at high soluble concentrations in soils (Shaheen and Rinklebe, 2014; Shaheen et al., 2015a; Shaheen and Rinklebe, 2015a). Therefore, knowledge about release dynamics of Cu and Zn in soils under different reducing-oxidizing conditions is required for assuring proper plant nutrition as well as understanding the possible environmental risks associated with Cu and Zn pollution (Shaheen et al., 2014). Selenium (Se) contamination in the environment has recently become a major issue world-wide, with both elevated and deficient Se concentrations found in groundwater, surface water, soils, and associated cultivated crops (Johansson et al., 2016). Selenium has attracted substantial interest because it is an essential nutrient to humans at low levels, but can be a toxin at elevated levels (Navarro-Alarcon and Cabrera-Vique, 2008; Yadav et al., 2008). The oxidation status of Se, controlled by the soil redox conditions, differs in its mobility and toxicity in the aquatic environment. Selenium is subject to transformation when oxidation–reduction conditions shift ranging from Se(+6) → Se(+4) → Se(0) → Se^{2-}, dependent on E_H (He et al., 2010). The release of Cu, Se, and Zn in deltaic wetland soils is a concern for surface- and groundwater quality since these elements might be released from soil solid phase to soil solution, particularly under alternating wetting and drying conditions. There have been studies which have shown the release dynamics of PTEs including Cu and Zn under different reducing-oxidizing conditions in acidic wetland systems (e.g. Frohne et al., 2011; Shaheen et al., 2014; Rinklebe et al., 2016a). Rinklebe et al. (2016b) studied the release dynamics of dissolved As, Ba, Cd, Cu, Pb, and Sr in seven different paddy soils from the U.S.A. and Asia under controlled-redox conditions. However, our knowledge regarding specific soil biogeochemical factors and mechanisms on the release dynamics of Cu, Se, and Zn in alkaline, periodically flooded soils in the Nile and Mississippi River Delta is limited.

The ecosystem services of river deltas often support high population densities and agricultural activities (Islam et al., 2015). Therefore, there is a need for detailed knowledge about the behavior of inorganic pollutants such as Cu, Se, and Zn in deltaic wetland soils under dynamic reducing-oxidizing conditions which could serve as a precondition for the development of innovative technologies for management of rice and deltaic wetland systems aimed at avoiding pollutant exposure to plants, water, soil, and riverine ecosystems. Since, the impact of reducing-oxidizing conditions on the mobilization of Cu, Se, and Zn and related governing factors in Nile and Mississippi Rivers deltaic soils have not been systematically studied to date, our goal was to study the impact of E_H and governing factors i.e., pH, Fe, Mn, Cl^-, DAC, DOC, and SO_4^{2-} on the release dynamics of Cu, Se, and Zn in two different soils collected from the Mississippi and Nile Rivers deltaic plains.

2. Materials and methods

2.1. Study site and soil sampling

2.1.1. Nile River Delta Soil (NDS)

The Nile delta is located along the southern coast of the Mediterranean Sea (30° 00′ − 31° 40′ N and 30° 00′ − 32° 30′ E; Fadlelmawla and Dawoud, 2006). Our study site at the Nile Delta is located at the Kafr El-Sheikh governorate (Supplemental 1). The Nile Delta has a Mediterranean climate, with dry mild summers and fairly cool and wet winters with an annual precipitation of <100 mm (Bruggers, 2010). The Nile Deltaic Soil was collected from fluvial deposits which are under rice production each summer, as they are periodically flooded for five months every year. Four soil profiles were collected from wetland rice soils existing in the area. The profiles were collected after harvesting the rice plants. From the surface (0–25 cm) and subsurface (25–50 cm) layer of each profile, ten samples were collected, pooled, and well-mixed. All samples were sealed in polyethylene bags and transported to the laboratory on ice where they were stored at 4 °C prior to analysis.