



Spatial and temporal distributions of sulfur species in paddy soils affected by acid mine drainage in Dabaoshan sulfide mining area, South China



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ABSTRACT

In paddy soils polluted by acid mine drainage (AMD), heavy metals contamination has been studied intensively, but little information has been gathered regarding the associated speciation of sulfur. Therefore, the concentrations of various sulfur species in paddy soil profiles were investigated at sites downstream of the Dabaoshan sulfide mining area, South China. No significant differences in water-soluble sulfate (WS), adsorbed sulfate (AS), or total sulfur (TS) were observed between upstream and downstream paddy soil profiles. A surprising finding was that seasonal change of WS in deeper soil may have contributed to the gradual decline in TS over the past 10 years. The largest concentrations of AS was found in 20 to 30 cm soil horizons, where a strong influence of large surface area soil particles, and abundant amorphous iron hydroxides and fine clays likely enhanced sulfate adsorption. Reduced inorganic sulfur (RIS) was found in highest concentrations at depths of 0 to 20 cm and was significantly positively correlated with total soil C and N contents. Furthermore, ester sulfate (ES) was the dominating sulfur species in every layer, which may result from (i) abundant WS and AS promoted ES formation, and (ii) the presence of active Al and Fe oxides inhibited ES mineralization. Concentrations of sulfur species and other soil properties were used in principle component analysis to identify three geochemically distinct layers in these paddy soils (0 to 20 cm, 20 to 30 cm and 30 to 80 cm). Due to AMD irrigation, differences in the speciation of sulfur among these layers illustrates how the chemical imprint of AMD irrigation has influenced and been affected by the background biogeochemistry of these soils. The present examination of the speciation and vertical and horizontal distribution of sulfur species provides valuable information for the continued management and remediation of these and other AMD-contaminated paddy soils.

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1. Introduction

Soils contaminated by acid mine drainage (AMD) have been referred to as “the nastiest soils in the world”, reflecting their acidity and toxicity (Boman et al., 2008; Conesa et al., 2014; Johnston et al., 2011). AMD has not only resulted in elevated concentrations of sulfur and heavy metals in soils (Goienaga et al., 2015; Mirzaei et al., 2014; Scheckel and Ryan, 2004), but has also damaged the growth of plants and the health of human beings (Rosicky et al., 2004). Heavy metal contamination of soils affected by AMD has been studied intensively (Du et al., 2015; Monroy et al., 2014; Sobrino-Plata et al., 2013), but relatively little

information has been reported regarding the speciation, spatial distribution, migration and transformation of associated concentrations of sulfur. Although sulfur is relatively nontoxic, sulfur redox plays a significant role in soil acidity and the mobilization of toxic metals (Hodson and Langan, 1999; Prakongkep et al., 2012). Sulfur chemistry in aquatic environments is important not only because sulfur is a major nutrient next to nitrogen and phosphorous (Cao et al., 2011), but also because it is an important vehicle for biochemical electron transfer under anaerobic conditions (Sheng et al., 2011). Moreover, although sulfur in soils is present mainly in organic form (Tabatabai et al., 2005), very little is known about the identities of organic sulfur compounds. Therefore, the investigation of the species and spatial and temporal distributions of sulfur is important when considering soil pollution and reclamation.

Sulfur exists in a variety of inorganic and organic chemical species in soil whose concentrations depend on soil types and depth. Soil sulfur is

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fractionated into water-soluble sulfate (WS), adsorbed sulfate (AS), ester sulfate (ES), carbon-bonded sulfur (CS), and Residual sulfur (RS) (Aguilera et al., 2002). The sulfur cycle in soils includes the processes of mobilization, immobilization, mineralization, oxidation and reduction (Tabatabai et al., 2005; Tanikawa et al., 2014b; Wilhelm Scherer, 2009). Inorganic SO_4^{2-} in soil solution is highly mobile, and it can strongly affect the remediation of anthropogenic acidified soil (Mitchell et al., 2011). The average export of sulfate from agricultural soil during February through May was estimated to be 70% of the total annual export and was inversely correlated with the capacity of the soil to adsorb SO_4^{2-} (Lynch and Corbett, 1989). The capacity of soil to adsorb SO_4^{2-} depends on the soil's properties, including the concentrations of Al and Fe-hydrous oxides, secondary minerals, clay minerals, soil pH and organic matter (Acosta et al., 2011; Jennings and Driese, 2014; Liu et al., 1999). The mechanisms for AS in plough layer soils have been investigated by many researchers (Ishiguro and Makino, 2011; Mikkelsen and Norton, 2013; Takahashi and Higashi, 2013). However, there have been few reports on the behavior of SO_4^{2-} in AMD-contaminated soil.

There remains considerable uncertainty regarding the abundance, vertical distribution and species of reduced inorganic sulfur (RIS) in paddy soils contaminated by AMD (Burton et al., 2006a; Johnston et al., 2014). Reformation and accumulation of RIS species are greater in surface layers of organic-rich soils than at other depths (Zhuang et al., 2014). In wetland acid sulfate soils, chromium reducible sulfide (CRS) and elemental sulfur (S^0) are the dominant RIS species, while acid volatile sulfide (AVS) is a minor component (Johnston et al., 2014). CRS and AVS are used to assess the health of acid sulfate soils because these RIS species may be at risk of rapid oxidation and acid generation during future drought episodes (Burton et al., 2006b; Johnston et al., 2014).

Organic sulfur in soil is present as a variety of complex compounds from crop residues and other sources including ES and CS (Kertesz and Mirleau, 2004), which are continually transformed between organic and inorganic forms by microbial action. In soil, both CS and ES can be mineralized to SO_4^{2-} , however, mineralization of ES can be inhibited by fine clays (Bettany et al., 1980; Hu et al., 2007). Studies have shown that immobilization of SO_4^{2-} into the ES fraction is more rapid than into CS forms (Ghani et al., 1992). Despite this knowledge, very little is known about the distribution and transformation of ES and CS in AMD-contaminated soils with high concentrations of WS.

Sulfur redox status and spatial distribution can influence the mobility and toxicity of trace elements. Therefore, the main objectives of this study were to characterize the vertical distribution of sulfur species in paddy soil in a sulfide mine area and to illuminate the relationships between the species and distribution of sulfur and the physicochemical properties of the soil.

2. Materials and methods

2.1. Study sites

This study was carried out in rice paddies within the Dabaoshan mining area (24°34'28"N, 113°43'42"E) (Fig. 1) near the largest polymetallic sulfide mine in southern China, which has been in operation for >50 years. Soils were polluted by enormous amounts of S^{2-} , SO_4^{2-} , Cu, Pb, Cd, Zn and Fe-rich mine wastes during the early years of mining operations (Chen et al., 2007; Zhou et al., 2007). As a result, water from the upper reaches of the Hengshi River just downstream of the mine, has extremely low pH (2.39–2.80) and high concentrations of sulfate and sulfite (Chen et al., 2015). Farmers in the Hengshi River watershed who were unaware of this contamination used the river for flush irrigation resulting in the severe contamination of adjacent rice paddies and their accumulation of great quantities of sulfur (Lin et al., 2005; Ye et al., 2014; Zhao et al., 2012a; Zhao et al., 2012b; Zhuang et al., 2009).

2.2. Sampling and pre-treatment

Soil sampling was carried out at previously studied sites in the Dabaoshan mining area (Chen et al., 2015; Lin et al., 2005; Zhou et al., 2007). Soil samples were collected at sites S1, S2, S3, S4, S5 and S6 in November 2012, July 2013, November 2013 and July 2014 (Fig. 1). Sites S1 to S3 were chosen because they were near a mining waste mud impoundment where rice paddy soil was contaminated by irrigation with AMD-polluted water (Li et al., 2009; Lin et al., 2005; Zhuang et al., 2014), which resulted in serious damage to the health of local inhabitants. Sites S4 and S5 were chosen because they were downstream of points along the river. Site S6, located along the Taiping River, a tributary of the Hengshi River just upstream of Site S3 (Shangba Village), was taken as the control site.

Three 1-m wide soil pits were dug to a depth of approximately 0.8 m at each site. Soil samples (about 2.0 kg) were collected from six soil layers (0–10 cm, 10–20 cm, 20–30 cm, 30–40 cm, 40–60 cm and 60–80 cm) in each paddy field, stored at 4 °C, and transported to the laboratory for determination of physicochemical properties. Some samples were immediately freeze-dried in the laboratory; others were stored at –20 °C to forestall sulfur transformation.

2.3. Sulfur species determinations

The sulfur species determined in the paddy soils included water-soluble sulfate (WS), adsorbed sulfate (AS), acid volatile sulfide (AVS), chromium reducible sulfide (CRS), water-soluble sulfite (sulfite), ester sulfate (ES), and carbon bonded sulfur (CS) (Chen et al., 2015; Shan et al., 1997). AVS includes amorphous monosulfides of metals (e.g. Fe, Zn, Cd or Pb) (Backlund et al., 2005; Morse and Luther, 1999), while CRS mainly consists of pyrite (Burton et al., 2008; Hsieh et al., 2002). AS represents phosphate-exchangeable sulfate (Boukhalfa, 2010).

2.3.1. Soil physical and chemical properties

Soil pH was measured using 4 g of freeze-dried soil in 10 mL deionized water. Particle-size distribution (20 g sample) was determined using a pipette method (Pansu and Gautheyrou, 2007). Available Mn was determined in 20 g soil samples by atomic absorption spectrophotometry after extraction with diethylene triamine pentacetate acid. Alkali-hydrolyzable nitrogen concentration was determined in 10 g soil samples using the KCl extraction-indophenol blue colorimetric method. Available K was measured in 5 g soil samples by the neutral ammonium acetate extraction method (Bao, 2005). Additionally, complex and amorphous Fe oxides were quantified in 1 g soil samples by using sodium pyrophosphate and ammonium oxalate extraction procedures, respectively (Bao, 2005). The BET-SA surface area was measured by nitrogen adsorption and desorption analysis (Micromeritics, ASAP 2020 M). Total carbon (TC) and nitrogen (TN) concentrations were measured in 0.05 g soil samples using an Elementar Vario EL III element analyzer.

2.3.2. Available sulfate in soil profile

WS and AS were determined in 8 g soil samples by extraction with deionized water (soil:water = 1:5.0) and phosphate solution (0.032 M sodium dihydrogen phosphate, pH 6), respectively (Boukhalfa, 2010). Sulfate was analyzed by ion chromatography (IC, Dionex-1000) with AG14-AS14 anion exchange columns and an eluent of 4.5 mM Na_2CO_3 -2.0 mM NaHCO_3 , delivered at a flow rate of 1.2 mL min⁻¹ (Chen et al., 2015).

2.3.3. Reduced inorganic sulfur in soil profile

Reduced inorganic sulfur (RIS) represented the sum of AVS, CRS and sulfite (water soluble sulfite). AVS was detected by purge-and-trap

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