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Evaluation of ferrolysis in arsenate adsorption on the paddy soil derived from an Oxisol

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HIGHLIGHTS

- Transformation of Oxisol to paddy soil decreased free Fe oxides and increased amorphous ones.
- Ferrolysis decreased arsenate adsorption by paddy soil.
- Increasing amorphous Fe oxides increased arsenate adsorption on paddy soil.
- Increasing pH and incorporating phosphate were unfavorable for arsenate adsorption.

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ABSTRACT

Iron oxides are dominant effective adsorbents for arsenate in iron oxide-rich variable charge soils. Oxisol-derived paddy soils undergo intensive ferrolysis, which results in high leaching and transformation of iron oxides. However, little information is available concerning the effect of ferrolysis on arsenate adsorption by paddy soil and parent Oxisol. In the present study, we examined the arsenate affinity of soils using arsenate adsorption/desorption isotherms, zeta potential, adsorption kinetics, pH effect and phosphate competition experiments. Results showed that ferrolysis in an alternating flooding–drying Oxisol-derived paddy soil resulted in a significant decrease of free iron oxides and increase of amorphous iron oxides in the surface and subsurface layers. There were more reactive sites exposed on amorphous than on crystalline iron oxides. Therefore, disproportionate ratios of arsenate adsorption capacities and contents of free iron oxides were observed in the studied Oxisols compared with paddy soils. The Gibbs free energy values corroborated that both electrostatic and non-electrostatic adsorption mechanisms contributed to the arsenate adsorption by bulk soils, and the kinetic adsorption data further suggested that the rate-limiting step was chemisorption. The zeta potential of soil colloids decreased after arsenate was adsorbed on the surfaces, forming inner-sphere complexes and thus transferring their negative charges to the soil particle surfaces. The adsorption/desorption isotherms showed that non-electrostatic adsorption was the main mechanism responsible for arsenate binding to the Oxisol and derived paddy soils, representing 91.42–94.65% of the adsorption capacities. Further studies revealed that arsenate adsorption was greatly inhibited by increasing suspension pH and incorporation of phosphate.

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1. Introduction

Paddy soils are widely distributed in Asia and other parts of the world (Zhang and Gong, 2003). The solid components of paddy soil, including clay minerals, organic matter, microorganisms as well as metal oxides are strongly affected by repeated water-submerging cultivations (Kogel-Knabner et al., 2010). For paddy soil derived from variable charge soils, the parent upland soils are rich in iron oxides as a result of intensive weathering and eluviation. Cyclic reduction and oxidation of iron oxides in an alternating flooding–drying system, predominantly characterized by bleaching and mottling, is conceptualized as ferrolysis (Brinkman, 1970).

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Ferrolysis was proposed as a major pedogenesis for paddy soil and aquorizem in Southeast Asia and Japan (Brinkman, 1970). Ferrolysis theory was extensively used to explain how iron oxides undergo segregation, gleying and chloritization (Brinkman, 1970; Singh et al., 1998). There are large areas of variable charge soils in the tropical and subtropical regions in the south of China and they are rich in iron and aluminum oxides as a result of intensive weathering and eluviation (Yu, 1997; Jiang et al., 2011; Xu et al., 2016). There are also extensive plantations of rice in this region. The iron oxides in paddy soils derived from variable charge soils change greatly when the soil suffers intensive ferrolysis (Brinkman, 1970); however, there is little information about the effect of ferrolysis on the chemical characteristics of these paddy soils. This is important because of their associated properties of adsorbing or binding contaminants (e.g. heavy metals, organic pollutants and pesticides) as well as nutrients and bacteria (Jiang et al., 2008; Xu et al., 2012).

Arsenicosis is one of the most serious environmental chemical diseases and affects many populations throughout the world (Sun, 2004). The potential risk of arsenic contamination in rice planting areas is a major environmental concern (Smith et al., 2002; Ma et al., 2008). Accumulated arsenic in soils can derive mainly from agriculture practices (pesticides and manure) and mining and smelting activities (Matschullat, 2000). Apart from mining and pedologic environments, aquifers under strongly reducing conditions are known to be linked to natural arsenic enrichment in surface and ground water systems, where the release of arsenic is related to reductive dissolution of arsenic-bearing iron (hydr)oxides in rocks and arsenic-contaminated soils (Takahashi et al., 2004; Rodríguez-Lado et al., 2013). Water for irrigation of rice is primarily dependent on surface and ground water, leading to a higher possibility of arsenate accumulation in paddy soil, toxicity to rice and increased levels of arsenic in rice grain and straw (Panaullah et al., 2009). Thus, paddy soils are more likely to suffer arsenic contamination than dry land due to greater irrigation water demand. Moreover, several arsenic resources and mining areas occur in the tropical and subtropical area of China, and the ability of soils to adsorb arsenic is highly variable (Smith et al., 2002). Thus, determining the adsorption mechanisms of arsenate on the variable charge soils and their derived paddy soils is of paramount importance in research on the nature and electrochemical properties of the soils in this area.

Variable charge soils show a high affinity for inorganic oxyanions, such as phosphate (Antelo et al., 2005; Jiang et al., 2015), chromate (Jiang et al., 2008) and arsenate (Smith et al., 2002). The sorption mechanisms of multivalent oxyanions are generally recognized as electrostatic interaction of the oxyanions with positively-charged hydroxide surfaces, ligand exchange or precipitation (Antelo et al., 2005; Jiang et al., 2008). Although the chemical behaviors of arsenate on the surfaces of oxide minerals, paddy soils as well as variable charge soils have been extensively studied (Manning and Goldberg, 1996; Smith et al., 1999; Ladeira and Ciminelli, 2004; Takahashi et al., 2004; Barrow, 2012), the adsorption behavior and related mechanisms of arsenate on the paddy soils, compared with the original soils remain unclear. However, this knowledge is necessary to ensure quality and safety of food, and to explore arsenic detoxification in these soils. Substantial areas of paddy soils over Southeast Asia have experienced arsenic contamination and will remain contaminated, as it is not cost-effective to remediate polluted paddy fields (Meharg, 2004). Little information is available on the evolution of chemical properties of intensively used paddy soils (Sumfleth and Duttmann, 2008), which is crucial to the fate, transport and mobility of arsenate. In the present research, the contents and morphologies of iron oxides in a paddy soil, compared with the original upland soil were studied.

The mobility of arsenate in soils depends on several factors, including but not limited to soil pH and the presence of other oxyanions that compete with arsenate for retention sites (Manning and Goldberg, 1996). Disclosing the factors affecting arsenate mobility in soils is of great importance to better assess the potential consequences of its fate, transport and mobility in soils. Previous studies showed that arsenate adsorption decreased with increasing the system pH, mainly due to increasing negative charge on the soil surface and the amount of negatively charged arsenate species present in soil solution (Smith et al., 1999; Antelo et al., 2005; Jiang et al., 2015), in turn enhancing the electrostatic repulsions. In addition, phosphate can compete with arsenate for adsorption sites on the positively-charged surfaces of soils and iron oxides (Violante and Pigna, 2002). However, the competitive adsorption of arsenate and phosphate on Oxisols compared with the derived paddy soil is little understood and, with current knowledge, it is impossible to quantitatively assess the role of ferrolysis in the competitive adsorption.

Thus, the purposes of present study are to (1) compare selected properties between an upland Oxisol and its derived paddy soil, (2) determine the adsorption and desorption of arsenate by these soils and gain insight concerning the involved mechanisms, and (3) reveal the effects of pH and phosphate, as a competitive oxyanion, on adsorption of arsenate at the soil–water interface.

2. Materials and methods

2.1. Soil samples

Two soils were collected from experiment fields of Guangdong Ocean University (GDOU), Zhanjiang City, Guangdong Province, China (21°9′N, 110°17′E). The sampling sites were in a south subtropical region with an average temperature of 23.2 °C and average annual rainfall of 1750 mm. The original soil is classified as an Oxisol, derived from basalt. Rice cultivation has occurred for more than 80 years, as confirmed by memories of local farmers and by documentation from researchers in GDOU. Soil samples were taken from the surface (0–20 cm) and subsurface (20–40 cm) by the serpentine sampling method. After air drying, the collected soils were ground to pass a 0.25-mm sieve prior to use. The basic properties of the studied soils are presented in Table 1.

The clay (<2 μm diameter), silt (2–20 μm diameter) and sand (>20 μm diameter) fractions were separated from soil samples by

Table 1
Basic properties of the studied soils.

Soil	Depth	Clay	Silt	Sand	pH	EC	Organic matter	CEC
	cm	g kg^{-1}	g kg^{-1}	g kg^{-1}		μS cm^{-1}	%	cmol kg^{-1}
Paddy soil	0–20	372.8	196.8	430.4	7.33 b	444.45 a	2.70 a	18.0 a
Paddy soil	20–40	437.7	164.1	398.2	7.45 a	219.30 b	1.38 d	17.4 b
Oxisol	0–20	672.4	73.1	254.5	5.04 d	135.41 c	2.06 b	15.1 c
Oxisol	20–40	660.7	143.2	196.1	5.12 c	78.06 d	1.50 c	14.0 d

Means followed by the same letter within columns are not significantly different (Duncan at $P = 0.05$).

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