

Adsorption of arsenite and arsenate onto muscovite and biotite mica

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Abstract

Arsenite and arsenate sorption was studied on two silt-sized phyllosilicates, namely muscovite and biotite, as a function of solution pH (pH 3–8 for muscovite, and 3–11 for biotite) at an initial As concentration of 13 μM . The amount of arsenic adsorbed increases with increasing pH, exhibiting a maximum value, before decreasing at higher pH values. Maxima correspond to $3.22 \pm 0.06 \text{ mmol kg}^{-1}$ As(V) at pH 4.6–5.6 and $2.86 \pm 0.05 \text{ mmol kg}^{-1}$ As(III) at pH 4.1–6.2 for biotite, and $3.08 \pm 0.06 \text{ mmol kg}^{-1}$ As(III) and $3.13 \pm 0.05 \text{ mmol kg}^{-1}$ As(V) at pH 4.2–5.5 for muscovite. The constant capacitance surface complexation model was used to explain the adsorption behavior. Biotite provides greater reactivity than muscovite toward arsenic adsorption. Isotherm data obeyed the Freundlich or Langmuir equation for the arsenic concentration range 10^{-7} – 10^{-4} M. Released total Fe, Si, K, Al, and Mg in solution were analyzed. Calculation of saturation indices by PHREEQC indicated that the solution was undersaturated with respect to aluminum arsenate ($\text{AlAsO}_4 \cdot 2\text{H}_2\text{O}$), scorodite ($\text{FeAsO}_4 \cdot 2\text{H}_2\text{O}$), and claudetite/arsenolite (As_4O_6).
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1. Introduction

Various hypotheses on arsenic mobilization in groundwater of the Ganges delta, Bangladesh and India, have been proposed [1–7]. These hypotheses consider iron oxyhydroxides, arsenopyrite, and clay minerals from which arsenic is being released in response to changes in redox potential. Local contamination by, e.g., high-arsenic pesticide input to irrigated fields cannot account alone for the size of the phenomenon (6×10^6 people potentially exposed in West Bengal over an area of 23,000 km^2 and $\sim 3 \times 10^7$ people in Bangladesh over an area of 150,000 km^2) [8] and references therein]. One must therefore consider the mineralogy of the delta sediment to trace the source of arsenic contamination. The Ganges sediments are silt- and sand-dominated and consist of micaceous minerals (mainly muscovite and biotite); these micas are highly degraded in nature and are good adsorbents of metals [9–11]. Due to their degraded nature, mica minerals can presumably adsorb arsenic in the presence of ferrous iron and transform aqueous arsenic

into surface Fe(III) and As-rich precipitates [12]. Adsorbed arsenic was detected in natural muscovite particles collected from contaminated Ganges sediment by X-ray photoelectron spectroscopy (XPS) [13]. Pal et al. [14] found 9 mg kg^{-1} of arsenic in sand-sized biotite separated from sediment of the Baruipur area in West Bengal.

A number of studies have been made with arsenate and arsenite sorption onto clay minerals such as montmorillonite, kaolinite, and illite. Frost and Griffin [15] observed that arsenate sorption onto kaolinite and montmorillonite first increased with pH, then exhibited a maximum at about pH 5.0, and finally decreased at high pH; the arsenite sorption peak on montmorillonite occurs near pH 7, while no peak appears for arsenite sorption on kaolinite in the pH range 4–9. These researchers also found that montmorillonite sorbed both As(V) and As(III) more strongly than kaolinite and that As(III) was sorbed much less than As(V) by both clay minerals. In contrast, Goldberg and Glaubig [16] found that the magnitudes of arsenate sorption maxima are similar for both clays. The shapes of the arsenate sorption curves for montmorillonite and kaolinite agreed well according to both Frost and Griffin [15] and Goldberg and Glaubig [16]. Goldberg [17] found no competitive effect of

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the presence of equimolar arsenite on arsenate adsorption onto clay minerals. Manning and Goldberg [18] studied competitive adsorption of arsenate–phosphate and arsenate–molybdate on kaolinite, montmorillonite, and illite. They found As(V) adsorption maxima at pH \sim 5.0 for kaolinite, 6.0 for montmorillonite, and 6.5 for illite, which ranged from 0.15 to 0.22 mmol As(V) kg^{-1} . Besides arsenic, other anion adsorption studies have been performed. For example, boron adsorption showed a peak at pH range 9–10 on montmorillonite, 8–9 for kaolinite, and near 9 in the case of illite [19]. Phosphate sorption onto kaolinite and illite has a maximum at pH 4–5, while sorption increased with increasing pH for montmorillonite. Montmorillonite has a phosphate adsorption capacity 8.2 times greater than kaolinite, whereas illite has greater adsorption capacity 27.6 times than kaolinite and 3.4 times that of montmorillonite [20].

Phyllosilicates possess three types of surface hydroxide groups associated with ruptured bonds along the crystal edge: aluminol, silanol, and Lewis acid groups [21]. Several researchers have investigated the greater reactivity of the edge sites of micaceous minerals compared to basal surfaces for adsorption. Charlet et al. [12] found As(V)–Fe(II) reaction products in the presence of phlogopite, a Fe-free mica, to be located predominantly on the layer edges. Preferential adsorption of uranyl complexes (UO_2^{2+}) at the micaceous mineral edges and optically visible steps was observed by Lee and Jackson [22]. Farquhar et al. [21] studied sorption of Cu(II), Cd(II), and Pb(II) onto muscovite and biotite in mildly acidic solution and concluded that biotite provides greater reactive surface than muscovite. Precipitates of PbCl_2 were found on the basal surfaces, steps, and edges of muscovite, with precipitation preferentially occurring along the broken steps, compared to the basal surfaces in an AFM study by Zhang and Bailey [23]. They suggested that the edges of the freshly cleaved muscovite are more reactive than the basal surfaces. Johnsson et al. [24] have shown that the basal surface of muscovite is also highly reactive, particularly with respect to water adsorption and subsequent dissolution and precipitation. Proton-induced X-ray emission (PIXE) arsenic elemental mapping has shown an accumulation of arsenic both on the mica edge and, to a lesser extent, on the basal surface of muscovite [13]. Relatively few published articles are available on arsenic adsorption on muscovite and biotite. Huang [25] studied the relative significance of arsenic retention by hydroxy-Al on external and interlamellar surfaces of micaceous mineral colloids and observed that K-depletion of muscovite and biotite resulted in decreased arsenic retention; also, hydroxy-Al on the external surfaces of micaceous mineral colloids is significant in retention of arsenic for $<0.2\text{-}\mu\text{m}$ fractions.

Muscovite and biotite dissolution and alteration were investigated by Kalinowski and Sachweda [26] (at pH 1–4), Malmström et al. [27] (near neutral to near alkaline region), Turpault and Trotignon [28] (acidic pH region), and more recently Samson et al. [29] (alkaline pH region).

The purpose of the present study is to understand the role of silt-sized (2–50 μm) muscovite and biotite mica fractions in arsenate and arsenite adsorption with varying initial As con-

Table 1

Particle size distribution and specific surface areas of muscovite and biotite studied

Mica(s)	SSA ($\text{m}^2 \text{g}^{-1}$)	Clay ($<2 \mu\text{m}$) (%)	Silt (2–50 μm) (%)	Sand ($>50\text{--}100 \mu\text{m}$) (%)
Muscovite	14.28	3.61	85.69	10.7
Biotite	8.34	2.02	86.08	11.92

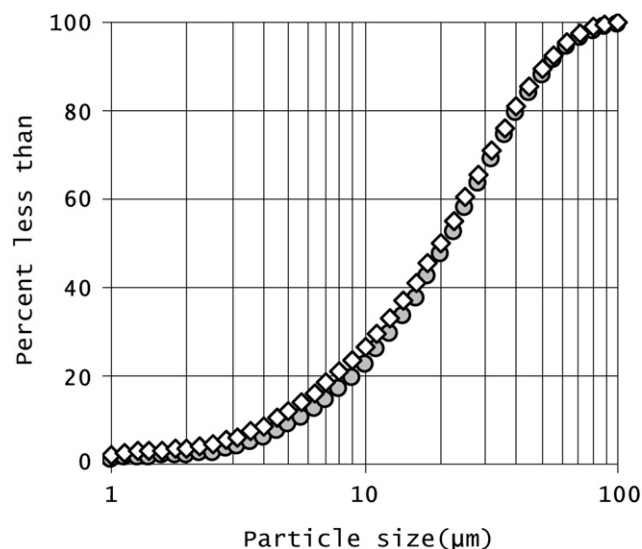


Fig. 1. Particle size distribution curve (gray filled circles and open diamonds for biotite and muscovite respectively).

centration (isotherms) and solution pH (edges). Release of different elements from micas due to dissolution was studied in the presence of arsenic. The initial arsenic concentration chosen to study the pH dependence of sorption was 13 μM , since this is the naturally realistic concentration for contaminated Bengal groundwater.

2. Materials and methods

2.1. Reagents

Muscovite [$\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH},\text{F})_2$] and biotite [$\text{K}(\text{Mg}, \text{Fe}^{2+})_3(\text{Al}, \text{Fe}^{3+})\text{Si}_3\text{O}_{10}(\text{OH},\text{F})_2$] samples (Refs. 46E5470 and 46E1190, respectively) were obtained from Ward's Natural Science Establishment. They were used without any chemical pretreatment. The purity of the samples was verified by XRD using Cu- $K\alpha$ radiation operating at 40 kV and 37.5 mA (Ona Siemens 5000 diffractometer). The fresh mineral samples ($\sim 0.5 \text{ g}$) were initially disaggregated using an ultrasonic probe. Specific surface areas (SSA) of the micas were determined by single-point Brunauer–Emmett–Teller (BET) N_2 adsorption method and are given in Table 1. Mica samples were suspended in deionized water (Millipore Milli-Q 18 M Ω) for particle size determination by a laser-diffraction-based particle size analyzer (Mastersizer 2000, Hydro-G). For both minerals, the silt fraction is the dominant granulometric fraction (Fig. 1 and Table 1).

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