RESUMEN

El arsénico (As) es un oligoelemento y es considerado actualmente como indispensable para la vida. Sin embargo, su acumulación en altas concentraciones en organismos vivos se traduce en una elevada toxicidad para los mismos. En general las aguas subterráneas son las que contienen mayor concentración del mismo y las que representan la mayor amenaza para la salud humana.

Como ocurre con la mayoría de los elementos trazas, la concentración de arsénico en aguas naturales es probablemente controlada por algún tipo de interacción agua/mineral. Muchos sistemas son regulados por procesos de adsorción/desorción en la superficie de óxidos y otros minerales, y la concentración de arsénico depende fuertemente de la reactividad de los grupos funcionales de la superficie y de la presencia de especies que compitan con el arsénico por los sitios de adsorción.

En este trabajo de tesis se estudió la adsorción de arseniato en ferrihidrita, se realizó un estudio de las propiedades ácido-base del sólido y se aplicó el modelo de complejación superficial CD-MUSIC para describir el comportamiento de la superficie de la ferrihidrita. Aparentemente, dos tipos de complejos superficiales de esfera interna se forman por la reacción entre arseniato y los grupos superficiales, un complejo binuclear bidentado protonado y un complejo binuclear bidentado deprotonado. El carbonato presente como contaminante en la muestra forma también un complejo superficial de esfera interna y compite con el arseniato por los sitios de adsorción.

También se estudiaron los procesos cinéticos de adsorción/desorción de arseniato en una montmorillonita intercambiada con Fe(III), la cual mostró que las especies de Fe(III) presentes en la montmorillonita poseen una elevada eficiencia de unión de arseniato.

Se presenta un estudio cinético de la competencia entre arseniato y fosfato en la superficie de goethita, en el cual el fosfato se adsorbe primero y luego el arseniato es agregado para promover la desorción de fosfato. La disminución de fosfato adsorbido es cuantificada en función del tiempo, y los efectos de la concentración inicial arseniato, el fosfato inicial adsorbido, el pH y la temperatura sobre la velocidad de desorción son evaluados. La desorción de fosfato en goethita es inducida por arseniato y se produce en dos etapas: una etapa rápida, que tiene lugar entre el momento de la adición de arseniato y el primer punto de desorción medido a los 5 min de reacción, y

una etapa más lenta que toma varias horas. Las etapas determinantes de la velocidad de intercambio están relacionadas a la captación de arseniato por la superficie y no a la liberación de fosfato por la misma.

Por último, se evaluaron posibles procesos de control de la concentración de As en el agua intersticial de los sedimentos de la Cuenca del Arroyo Napostá Grande. Se aplicó el modelo de complejación superficial CD-MUSIC para describir la adsorción de aniones en la superficie de los sedimentos. Los cálculos indican que las concentraciones de As en el agua están principalmente controladas por procesos de adsorción/desorción en la superficie de los minerales del sedimento, donde el pH y la concentración de especies competidoras como el carbonato juegan un papel preponderante.

ABSTRACT

Arsenic (As) is a trace element and is actually considered essential for life. However, their accumulation at high concentrations in living organisms results in a high toxicity. Among different drinking water sources, groundwaters are those that in general contain higher concentrations of this element and those that represent the greatest threat to human health.

As it happens with most of the trace elements, the concentration of arsenic in natural waters is probably controlled by some water/mineral interaction. Many natural systems are regulated by adsorption/desorption processes at the surface of oxides and other minerals, and the arsenic concentration in water strongly depends on the reactivity of the functional surface groups and the presence of species that compete with arsenic by the adsorption sites.

In this thesis the adsorption of arsenate on ferrihydrite was studied under different conditions. A study of the acid-base properties of the solid was performed and the CD-MUSIC surface complexation model was applied to describe the behavior of the ferrihydrite surface. Apparently, two types of inner-sphere surface complexes are formed by the reaction between arsenate and the surface groups, a bidentate binuclear protonated complex and a bidentate binuclear deprotonated complex. The carbonate present as a contaminant also forms a inner-sphere surface complex and competes with arsenate for the adsorption sites.

The adsorption/desorption kinetics of arsenate on a Fe(III)-modified montmorillonite was also studied, and it was stablished that that Fe(III) species located in montmorillonite have high efficiency in binding arsenate.

The competition between phosphate and arsenate on the goethite surface was kinetically explored. In these systems, phosphate was adsorbed first and then arsenate was added to promote phosphate desorption. The decrease in adsorbed phosphate was monitored as a function of time, and the effects of arsenate concentration, initial adsorbed phosphate, pH and temperature on the desorption rates were investigated. The rate-controlling steps of the phosphate-arsenate exchange reaction are related to the arsenate uptake by the surface and not to the release of phosphate by it.

Finally, possible processes controlling As concentration in the pore water of sediments of the Arroyo Napostá Grande watershed were evaluated. The CD-MUSIC model was applied to describe the adsorption of anions on sediment minerals. Calculations indicate

that As concentrations in water are mainly controlled by adsorption/desorption processes on the minerals surface, where pH and concentration of competing species such as carbonate play a key role.

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