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Start

Browse by
Section/Division of
Interest

Author Index

432-12 Effect of Ferrihydrite Crystallite Size on Phosphate Adsorption Reactivity.

See more from this Division: [SSSA Division: Soil Chemistry](#)

See more from this Session: [Advanced Molecular Techniques Characterizing Soil Biogeochemical Processes: II \(Includes Student Competition\)](#)

Wednesday, November 5, 2014: 11:45 AM
Long Beach Convention Center, Room 104C

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Ferrihydrite ($5\text{Fe}_2\text{O}_3 \cdot 9\text{H}_2\text{O}$), a naturally-occurring nanocrystalline iron hydroxide, is common in near-surface environments. Because of its large surface area and high reactivity, nanocrystalline ferrihydrite plays an important role in controlling the toxicity and bioavailability of some contaminants and nutrients through adsorption and coprecipitation. The influence of crystallite size on the adsorption reactivity of phosphate on 2-line to 6-line ferrihydrites was investigated by combining adsorption experiments, structure and surface analysis, and spectroscopic analysis. X-ray diffraction (XRD), transmission electron (TEM) and pair distribution function (PDF) showed that the ferrihydrite samples possessed a similar fundamental structure with a crystallite size varying from 1.6 to 4.4 nm. N_2 adsorption on freeze-dried samples revealed that the specific surface area (SSA_{BET}) decreased from 427 to 234 $\text{m}^2 \text{g}^{-1}$ with increasing crystallite size and micropore volume (V_{micro}) from 0.137 to 0.079 $\text{cm}^3 \text{g}^{-1}$. Proton adsorption (Q_{H}) at pH 4.5 and 0.01 M KCl ranged from 0.73 to 0.55 mmol g^{-1} . Phosphate adsorption capacity at pH 4.5 and 0.01 M KCl for the ferrihydrites decreased from 1690 to 980 $\mu\text{mol g}^{-1}$ as crystallite size increased, while the adsorption density normalized to SSA_{BET} was similar. Phosphate adsorption on the ferrihydrites exhibited similar behavior with respect to both kinetics and the adsorption mechanism. The kinetics could be divided into three successive first-order stages: relatively fast adsorption, slow adsorption and a very slow stage. With decreasing crystallite size ferrihydrites exhibited increasing rate constants per mass for all stages. Analysis of OH^- release and attenuated total reflectance infrared spectroscopy (ATR-IR) and differential pair distribution function (d-PDF) results indicated that initially phosphate preferentially bound to two $\text{Fe-OH}_2^{1/2+}$ groups to form a binuclear bidentate surface complex without OH^- release, with smaller size ferrihydrites exchanging more $\text{Fe-OH}_2^{1/2+}$ per mass. Subsequently, phosphate exchanged with both $\text{Fe-OH}_2^{1/2+}$ and $\text{Fe-OH}^{1/2-}$ with a constant amount of OH^- released per phosphate adsorbed. Also in this stage binuclear bidentate surface complexes were formed with a P-Fe atomic pair distance of $\sim 3.25 \text{ \AA}$.

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[<< Previous Abstract](#) | [Next Abstract >>](#)

