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Effect of Kaolinite on Adsorption Kinetics of Humic Acids during Ferrihydrite Transformation

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The transformation processes of iron oxides have been proved to significantly affect the retention of organic matter. In soil environment, clay minerals may not only affect the iron oxide transformation, but also participate in the adsorption of organic matter. However, the conventional batch adsorption experiments are difficult to accurately describe the dynamic adsorption processes of organic matter, and it is still unclear how the iron oxide transformation in multi-minerals systems affects the dynamic adsorption process of organic matter. In this study, the adsorption kinetics of humic acids (HA) on pure ferrihydrite (Fh) and kaolinite-doped ferrihydrite (Fh-Kaol) at different aging times was quantified and the spatial distribution of organic C was visualized. With the catalysis of Fe(II), Fh was gradually transformed into lepidocrocite and goethite, while the doping of Kaol reduced the transformation rates of iron oxides and inhibited the formation of goethite. The stirred- flow (SF) experiments showed that, with the continuous introduction of HA, the instantaneous adsorption amounts of HA decreased with the increase of adsorption time due to the “legacy effect”. The total HA adsorption on Fh-Kaol was significantly higher than that on Fh, while HA adsorption on Kaol was minimal. It was worth noting that the transformation of Fh promoted HA adsorption rates and increased HA adsorption amount, while the transformation of Fh-Kaol inhibited HA adsorption at initial transformation stage, and slightly promoted HA adsorption rates at final transformation stage. The scanning transmission electron microscopy combined electron energy loss spectroscopy (STEM-EELS) demonstrated that HA preferentially adsorbed on the surfaces of iron oxides. Compared with the aggregated Fh, the doping of Kaol promoted the dispersion of Fh and exposed the adsorption sites, which further resulted in higher HA adsorption on Fh-Kaol. The transformation of Fh/Fh-Kaol changed the crystal structure and agglomerate states of iron oxides, which further affected the HA adsorption kinetics. Our results help to elucidate the dynamic sequestration of organic matter during the iron oxide transformation in multi-minerals systems, which would be helpful for predicting organic matter cycling in natural environments.