Nanoscale titanium dioxide (nTiO$_2$) transport in water-saturated sand columns: Influence of phosphate, illite colloid, and Fe-oxyhydroxide coating

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Nanoscale titanium dioxide (nTiO$_2$) is an important emerging contaminant due to its mass production in recent years. While extensive research has been performed using simplified laboratory systems to study the stability and transport of nTiO$_2$, much fewer investigations are conducted to examine the behaviour of nTiO$_2$ in complicated systems similar to natural aquifers with heterogeneous transport media and multiple water-borne components. To better understand the fate and transport of nTiO$_2$ in natural environments, we performed laboratory column experiments to measure nTiO$_2$ transport in water-saturated sand columns in the co-presence of multiple components including dissolved phosphate, suspended illite colloid, and Fe-oxyhydroxide coating on sand grains. Results showed that pH plays a crucial role in how phosphate, illite colloid, and Fe-oxyhydroxide coating influence nTiO$_2$ transport. At pH 5 in the absence of phosphate and illite colloid, despite favorable deposition conditions, nTiO$_2$ was found in the effluent of the quartz column after $\sim$4.5 PV (pore volume) due to saturation of deposition sites, after which effluent nTiO$_2$ concentration quickly increased and then levelled off. For the Fe-oxyhydroxide coated sand column under the same conditions, whereas the shape of the BTC (breakthrough curve) was similar to that for the quartz sand column, nTiO$_2$ was detected in the effluent much earlier ($\sim$1 PV), owing to the reduction in favorable deposition sites caused by Fe-oxyhydroxide coating. When illite colloid was co-present with nTiO$_2$ in the influent at pH 5, nTiO$_2$ concentration in the effluent gradually increased to C/C$_0$ = $\sim$0.08 and $\sim$0.22 respectively for the quartz sand and Fe-oxyhydroxide coated sand columns. The low transport was attributed to physical straining caused by formation of the nTiO$_2$-illite aggregates during the experiment. For all the experiments with phosphate in the influent at pH 5, nTiO$_2$ concentration in the effluent gradually increased to C/C$_0$ = $\sim$0.08 and $\sim$0.22 respectively for the quartz sand and Fe-oxyhydroxide coated sand columns. The low transport was attributed to physical straining caused by formation of the nTiO$_2$-illite aggregates during the experiment. For all the experiments with phosphate in the influent at pH 5, nTiO$_2$ was not detected in the effluent regardless of the presence of illite colloid or Fe-oxyhydroxide coating, presumably due to straining of the large nTiO$_2$ aggregates formed via phosphate bridging. At pH 9, for both the quartz sand and Fe-oxyhydroxide coated sand columns with or without phosphate, high nTiO$_2$ transport was observed for all but one experiment. The high transport was due to the repulsive forces between nTiO$_2$ and the transport media. For the Fe-oxyhydroxide coated sand column without phosphate or illite colloid at pH 9, nTiO$_2$ transport was considerably lower (C/C$_0$ $\sim$0.07), as a result of pH buffering by the Fe-oxyhydroxide coating, which made the Fe-oxyhydroxides positively charged, and therefore providing favorable sites for nTiO$_2$ deposition. This study demonstrated that common water-borne components and minerals in transport media could significantly alter nTiO$_2$ transport and the interactions between each component and nTiO$_2$ as well as the synergy effects of these components must be considered in order to predict the behaviour of nTiO$_2$ in natural environments.