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# Nitrate impacts on heavy metal retention in heterogenous aquifers

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Alluvial aquifers are essential for meeting global water demands, with groundwater quality controlled by a unique balance of hydrological and biogeochemical processes. The composition of sediments comprising the aquifer, as well as on their spatial heterogeneity are the critical determinants of water chemistry. Coarse-grained materials dominate the primary (conductive) aquifer, but fine-grained materials, rich in clay, organic matter, Fe and Mn, are also widespread within alluvial aquifers and have a disproportionate influence on the biogeochemistry. The spatial distribution of fine-grained materials gives rise to redox gradients that play a profound role in the aquifer's reaction to contaminants such as heavy metals and fertilizers. Therefore, we simulated a heterogenous alluvial aquifer by embedding two fine-grained sediment lenses in coarse-grained aquifer sand. We then investigated the attenuation of heavy metal contaminants Ni and Zn under nitrate-rich conditions for a total of 77 days. The fine-grained lenses supported reducing conditions that extended to the downgradient sand. These conditions supported accumulation of nitrite and the ultimate oxidation of ferrous iron. Mössbauer spectroscopy identified a distorted Fe(III) phase in the lenses, implying that the enhanced iron cycling fueled by denitrification is responsible for this newly formed iron phase. The lenses retained up to 6-times more Ni and Zn than the sand, demonstrating their superior role in heavy metal sequestration. Accumulated amounts of the two metals were similar, yet differences were identified in their mechanisms of retention. While both Ni and Zn retention were dominated by metal silicates and metal sulfides, organic matter associations were more important for Ni retention compared to Zn, which was more prone to form layered double hydroxides. The metal sulfide pool was the most sensitive solid phase to nitrate introduction – Ni, Zn, and Fe concentrations were on average 20% lower under nitrate-rich conditions, likely on account of sulfide oxidation and/or inhibition of sulfate reduction. This work demonstrates the outsized importance of fine-grained materials for heavy metal retention and the profound influence of nitrate on the biogeochemical activity and consequent limitation of heavy metal retention.