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How does Ca modify the surface reactivity of Fe-OM aggregates against Arsenic binding?

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Iron-Organic Matter (Fe-OM) aggregates produced by redox alternation in wetlands are a key factor in the control of metallic pollutants mobility. Their ability to adsorb metal(loid)s depends on the size, morphology and structural arrangement between Fe and OM phases, which are mainly controlled by the OM occurrence. The physical, chemical and morphological organization of such aggregates is influenced by the physico-chemical conditions prevailing in the environment. Calcium (Ca) is a common major ion in natural waters which exhibits high affinity for OM. It can thus modify the size and the structural organization of Fe-OM aggregates and, subsequently, their ability to bind metal(loid)s. Among metal(loid)s, arsenic (As) is of major importance because of its high toxicity and its high affinity towards Fe(III)-oxyhydroxides. Moreover, Fe-OM aggregates are an important factor controlling the mobility of arsenic (As) in the environment.

Mimetic natural Fe-OM aggregates were synthesized at various Fe/OM and Ca/Fe ratios. After a fine characterization of the size and structural organization, Fe-OM-Ca associations were used to perform As binding sorption experiments at 2 As/Fe ratios. The suspensions were stirred during 24h and subsequently filtered and ultra-filtrated.

Our study demonstrates that Ca strongly influences the Fe-OM aggregates physical organisation. For low Ca/Fe ratio, Fe phases exhibit a fractal organization in which Fe phases are composed of oligomers, and primary nano-aggregates (around 6 nm) which aggregate in larger Fe secondary aggregates (>200 nm). Both are embedded in the OM matrix composed of isolated molecules and OM aggregates. For high Ca/Fe ratios, OM, Fe oligomers and primary nano-aggregates form a large continuous network where Fe phases are connected by OM large molecules. With the increasing Ca/Fe ratio, the amount of Fe oligomers decreases to the benefit of larger primary nano-aggregates (increase of their geometrical radius). Ultrafiltration experiments demonstrated that DOC, Fe, Ca and As follow the same size distribution. Surprisingly, As sorption increases with the increasing size and amount of primary nano-aggregates and the formation of the large network. SAXS analyses revealed that in such network, the distance between primary nano-aggregates

increases as compared to their distance in secondary aggregates. All this results suggest that, with the increasing Ca/Fe ratio, although the primary nano-aggregates size increase, their structural distance allows to rise the availability of their binding site for As.

This study demonstrates that Ca not only controls the Fe-OM structural organization but also its subsequent capacity to bind toxic elements such as As. These results are of major importance since such parameter was never so clearly evidence. They show that the actual representation of the physical organisation of Fe-OM aggregates and its reactivity have to be renewed as well as the geochemical models.