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Al and Si oxyanions impact on the structural organization of Fe-OM nanoaggregates

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Natural colloids composed of iron (Fe) and organic matter (OM) are a key factor controlling the mobility of metallic pollutants due to their high adsorption capacity, a consequence of their high density of binding sites. The physico-chemical conditions under which Fe-OM nano-aggregates are formed influence their structural organization, and more particularly the speciation of Fe. In this study, we probe the influence of two major elements of natural systems: aluminum (Al) and silicon (Si). Al is known to have a high affinity with OM but also easily enters the structure of Fe hydroxides. Si, on the other hand, is known to inhibit the growth and crystallinity of Fe hydroxides, although the mechanisms remain unknown. Al and Si are therefore expected to influence the Fe-OM nano-aggregates organization and to have an impact on the Fe speciation.

Fe-OM-Al/Si nano-aggregates, mimicking environmental ones, have been synthesized with different Fe/OM and oxyanion/Fe ratios. In these systems, the Fe speciation is complex and variable, depending on the Fe and oxyanion content relative to OM. The Fe phases appear to be composed of oligomers and ferrihydrite-like nanoparticles (Fh-Np), both integrated in the OM matrix. The Fh-Np form a fractal network whose organization is controlled by the OM. As Fe/OM increases, the oligomer content decreases in favor of the Fh-Np, which increases in size. By adding Al or Si, this phenomenon may strongly differ. Al, forming oligomers and bound to both Fe and OM, clearly allows the growth of the Fh-Np/oligomer ratio, the Fh-Np size and the whole nano-aggregates structure. On the contrary, Si, bound to Fe, has the exact opposite effect. These differences result from the different interactions between Al and Si and the components of the Fe-OM nano-aggregates.

These results clearly highlight the antagonist effect of the major elements, Al and Si, on the structural organization of Fe-OM colloids. They impact all the levels of organization: the Fe speciation and the OM and Fh-Np arrangement. This structural variability has a direct consequence on the ability of Fe-OM nano-aggregates to trap and transport pollutants in the hydrographic network.