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The early fate of hydrothermal Fe nano-colloids at the Rainbow Hydrothermal Vent Field

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Motivated by the goal to increase our knowledge of the impact of hydrothermal iron (Fe) nanoparticles on ocean chemistry and to explore their unique catalytic capabilities, we sampled suspended and dissolved matter in the water column above the Rainbow (36°-33°N) hydrothermal vent field at the Mid-Atlantic Ridge. Innovative sampling techniques were used to constrain the (trans)formation of hydrothermal iron-based nanoparticles. Instead of filtration of plume particles, freezing, and later resuspension, which is commonly used to separate particles from their surrounding solution and preserve them¹, we immediately drop cast small amounts of the fluid on transmission electron microscopy (TEM) grids and plunge-freeze them, resulting in vitrification of dissolved compounds and preservation of containing nanoparticles. Using an array of (micro)spectroscopic techniques, TEM, and a machine learning approach, we can characterize the Fe nanoparticles and unravel their fate in the ocean biogeochemical cycle.

Initial results show that the new sampling approach allows us to successfully collect Fe colloids with minimal artifacts – specifically avoiding aggregation of various suspended phases during filtration, which can result in spurious spatial correlations. The hydrothermal plume samples collected closest to an active vent show crystalline spherical Fe-nanoparticles that predominantly consist of poorly-ordered Fe-oxyhydroxide and are in parts enriched in P, S, Ni, and/or Cu. Using the machine learning model SIGMA² further allows us to explore the distribution of distinct Fe phases and reveals the local occurrence of reduced Fe as chalcopyrite and pyrite. On the outside, the Fe-nanoparticles are covered with an amorphous phase enriched in Mg, Cl, ± P, and S. Amorphous silica clusters are omnipresent and often co-occur with the Fe colloids. Notably, our results do not show associations of Fe with (organic) carbon.

These observations suggest that a higher local concentration of P within the Fe-colloids is potentially a crucial component affecting the Fe-nanoparticle's properties and environmental fate. Furthermore, this shows that C-rich phases do not significantly affect, at least in the early stages, the particles at the Rainbow vent field, contrasting previous studies, which suggest that organic compounds play a key role in stabilizing and transporting hydrothermal Fe^{1,3}. While Si is abundant in the hydrothermal fluid and often interacts with Fe precipitates similar to P, we show spatial

decoupling suggestive of a distinct precipitation mechanism. Neither in the hydrothermal plume away from the active vent nor in the sediment did we observe much transformation of the poorly-ordered Fe-colloids, suggesting that these were stable early on. However, we do observe an enrichment in organic compounds associated with the Fe-colloids further up in the buoyant plume.

Our research presents the first indications that during the early formation of hydrothermal Fe colloids, the properties of the Fe-based nanoparticle and, subsequently, the environmental fate and impact are more likely affected by P and Si than by organic carbon compounds.

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