The iron isotopic imprint of benthic iron release in suspended particles from the African oxygen minimum zone

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Continental margin sediments are increasingly being recognized as an important source of bioavailable iron to the open ocean. Benthic iron fluxes are particularly high in oxygen deficient ocean regions, such as the major Oxygen Minimum Zones (OMZs) off the African and South American west coasts. In these highly productive regions of the oceans, benthic (diagenetic) iron sources may be of similar importance to atmospheric iron sources. Iron isotopes are emerging as a powerful tool to trace the influence of reducing margins as a supply of iron to the open ocean. Redox recycling of reactive iron in the organic-rich shelf sediments imparts a characteristically light iron isotope signature on the benthic iron efflux. Aerosol iron, in contrast, typically resembles crustal material and shows no distinct isotope fractionation relative to average igneous rock. To examine the isotopic imprint of benthic iron release in oxygen deficient ocean regions we have measured the iron concentrations and isotope composition of suspended particles along two transects through the southern African OMZ at 13°S and 26°S. Dissolved and particulate iron concentrations are strongly elevated along the northern transect, which passes near the core of the OMZ. This pattern of high iron concentrations at intermediate depth suggests significant release of iron from the shelf sediments at bottom water oxyen concentrations <40µM. Despite this strong benthic release signal, iron isotope compositions of highly reactive Fe (0.5 M HCl leach at 60°C overnight) does not vary significantly from 0 ‰. At the southern sites, in contrast, overall sediment iron fluxes are smaller, but isotope compositions as low as -0.54 ‰ ($\delta^{56/54}$Fe, normalized to igneous rocks) are consistent with a diagenetic iron source from suboxic or anoxic sediments. Previous porewater studies have revealed a reversal in iron isotope fractionation during progression from suboxic/anoxic to sulfidic early diagentic reactions, potentially producing an iron efflux with isotope compositions close to 0 ‰ or even slightly positive. Consequently, rather than invoking an atmospheric signal at the northern site, we suggest a greater role for sulfides in the reduction and release of Fe^{2+} in the most oxygen-depleted regions of the OMZ. Light particulate isotope compositions at the southern site suggests that a significant proportion of dissolved Fe^{2+} is adsorbed or forms authigenic iron phases, and that particles are potentially an important vector for the transfer of reduced iron from the shelf to the open ocean. Results from this study provides a useful framework for the investigation of particles and their ispotope composition in the forthcomming GEOTRACES Pacific Section, which cosses the Peru OMZ.

Disturbance of the U/Pb and Th/Pb chronometers during low-T alteration of monazite.

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Low-temperature alteration of monazite is documented in three centimetric monazite crystals from Norway (Arendal), Madagascar (Ambato), and Srilanka. The three crystals have different chemical compositions, especially regarding U, Th, Y and Pb contents and have ages ranging from 491 to 900 Ma. Optical and Electron microscope (SEM and TEM) images and electron microprobe analyses (EPMA) show that all of them share the same texture, suggesting an alteration reaction following which unaltered monazite (Mnz1) reacts to form a secondary, Th-U(-Y)-depleted, high-Th/U-monazite (Mnz2) associated with variable proportions of thorite ($ThSiO_4$), thorianite (ThO_2) and xenotime (YPO_4), depending on the initial composition of Mnz1. Images reveal variably intense fracturing, with cracks filled with Th-rich +/- Fe-rich phases. Monazite-xenotime thermometry demonstrates that Mnz1 interacted with a low temperature fluid. The alteration process is interpreted to follow a mechanism of fluid-present coupled dissolution-precipitation. Chemical dating with EPMA show no isochron age difference between primary and secondary monazite, except for the Ambato monazite, where altered domains are apparently older (750 Ma). U/Pb and Th/Pb isotope dating using LA-ICP-MS give dates consistent with EPMA dates, in pristine zones. However, in Mnz2, the systems are disturbed. In the case of Srilanka and Arendal, only ^{232}Th/^{208}Pb dates give a reasonable estimate of alteration age, respectively 450 and 864 Ma. U/Pb systems are disturbed due to common Pb contamination (up to 40%) and U fractionation relative to Th, responsible for depletion of U in altered monazites (and increase of Th/U). In contrast, for the Ambato monazite, both U/Pb and Th/Pb systems were affected and give inconsistent older dates for altered zones. This is attributed to significant common Pb contamination (up to 80%), also affecting all Pb isotopes and explained why electron probe ages were disturbed as well. Secondly disturbance results from Th-U-silicate contamination during measurement, due to the presence of high density of nano-phases and nano-fractures filled with Th-U-silicates, only visible using TEM. Finally, these results demonstrate the important role of radiation damage effects, in particular swelling-induced fracturing, and the essential role of porosity and cracks, which allow fluid (charged with elements) migration through the sample during this process.