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Applicability of stable Cr isotope variations in marine sediments for the reconstruction of environmental changes through the geologic record

R. Schoenberg (1), I.C. Kleinhanns (1), M. van Zuilen (2), V.A. Melezhik (3,4), and R.B. Pedersen (4)

(1) Dept. of Geosciences, University of Tuebingen, Wilhelmstrasse 56, D-72074 Tuebingen, (2) Institut de Physique du Globe de Paris, 1 rue Jussieu, F-75238 Paris cedex 5, (3) Geological Survey of Norway, Postboks 6315 Sluppen, NO-7491 Trondheim, (4) Centre for Geobiology, University of Bergen, Postboks 7803, NO-5020 Bergen

In nature chromium occurs in two stable oxidation states: Cr(III) which is immobile and particle-reactive, and Cr(VI) which is soluble, highly mobile and toxic. Partial redox-cycling of chromium has been shown to be accompanied by large changes in the ^{53}Cr/^{52}Cr ratios of up to several per mill (usually expressed in δ^{53}Cr values; [1, 2]), preferentially enriching the oxidized Cr(VI) species in heavy isotopes. Recent studies show that the change in the Cr isotopic composition of Cr(VI) polluted groundwater along aquifers is a powerful tool to investigate natural or forced remediation of toxic Cr(VI) through reduction to Cr(III) on mineral surfaces in the soil or reductive materials in reactive barriers [3]. Accurate quantification of Cr(VI) reduction by Rayleigh-type Cr isotopic fractionation models is possible, because the Cr(VI) content in contaminated groundwater is high, while the concentrations of Ti, V and Fe, which all share one isobaric isotope with chromium, are very low, thus enabling accurate Cr isotope ratio determinations.

The application of Cr isotopes as paleo-redox tracer in marine sedimentary archives, such as banded iron formations [4] or carbonates [5], however, is analytically much more challenging. Here the observed Cr isotopic variations are often very small - in the order of tenths of ‰ in δ^{53}Cr values. Furthermore, these sediments normally contain trace amounts of chromium at the ppm level, but are highly-enriched in Ti, V, and especially Fe, requiring excellent Cr purification methods to achieve accurate Cr isotopic determinations by MC-ICP-MS.

The applicability of stable Cr isotopes as sensitive paleo-redox tracer will be demonstrated by the variations in the Cr isotopic compositions of 2.2 to 2.0 Ga old lacustrine carbonates, marine stromatolites and near-shore deposits of jaspers from the Pechenga Greenstone Belt and organic-rich, siliceous deposits from the Onega Basin, both situated in the NW Fennoscandian Shield. The variable Cr isotopic signatures in these sediments, with δ^{53}Cr values ranging from approximately +1.4 ‰ to -1.2 ‰ are most likely the result of changes in the oxidation states of chromium on the continents and in the oceans at this time. Models for chromium redox-cycling and accompanied isotopic fractionation will be discussed.

[1] Ellis et al. (2002), Science 295, 2060-2062; [2] Zink et al. (2010), Geochimica et Cosmochimica Acta 74, 5729-5745; [3] Berna et al. (2010), Environmental Science & Technology 44, 1043-1048; [4] Frei et al. (2009), Nature 461, 250-U125; [5] Bonnand et al. (2011), Journal of Analytical Atomic Spectrometry 26, 528-535.