Interpretation of Zn isotope ratio measurements in a complex geochemical system

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Zinc isotope ratios were measured for pore water samples collected from a pilot-scale remediation system designed to asssess the potential benefits of promoting bacterial SO_4 reduction and precipitation of metal sulfides. Samples were collected from three test cells at the Greens Creek mine (Alaska, USA), including a control cell and two treatment cells. Cells TC4 and TC7 were amended with organic carbon. The first treatment cell (TC4) contained 5 vol.% organic carbon as peat (2.5 vol. %) and spent brewing grain (2.5 vol. %), and the second treatment cell (TC7) contained 10 vol.% organic carbon as peat (5 Vol. %), spent brewing grain (2.5 vol. %) and municipal biosolids (2.5 vol. %). High concentrations of dissolved Zn (97 to 320 mg L^{-1}) and SO_4 near the tailings surface indicate Zn release by sphalerite [(Zn,Fe)S] oxidation. Zinc isotope ratios near the tailings surface in all three cells were similar and ranged between +0.25 and +0.35 ‰ ($\delta^{66}Zn_{avg}$ = +0.3 ±0.05 ‰). At depths equal or below 1 m below surface, Zn concentrations were generally below 2.7 mg L^{-1} in TC4 and TC7 and below 7.1 mg L^{-1} in TC2. This decline in Zn concentrations in TC4 and TC7 is attributed bacterial SO_4 reduction and concomitant alkalinity production, leading to extensive precipitation of Zn sulfide phases and potentially Zn carbonate phases. Zinc isotope measurements indicate $\Delta^{66}Zn$ values of up to -0.35 ‰. Laboratory studies indicate precipitation of Zn sulfide phases results in preferential incorporation of ^{64}Zn, resulting in increasingly positive $\delta^{66}Zn$ values, whereas precipitation of Zn carbonate leads to increasingly negative $\delta^{66}Zn$ values. These observations suggest that precipitation of a combination of secondary sulfide and carbonate phases controls Zn mobility and isotope ratios under SO_4-reducing conditions within the amended cells.