Provenancing arsenic release and organic matter in shallow groundwaters of South and South East Asia

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The release of arsenic (As) in the shallow reducing groundwater extensively utilized as drinking waters in South and South East Asia has resulted in high levels of exposure to over 100 million people [1]. The As is mobilized through the microbially mediated reductive dissolution of Fe (III) bearing minerals coupled to the oxidation of organic matter (OM), which is therefore critically implicated in the process [2]. Currently there are several oustanding issues that limit our ability to predict distribution of groundwater As hazard both now and in the future, namely (1) the location of As release within the subsurface, (2) the source of OM driving these common biogeochemical processes, and (3) how the present As hazard will change in the coming years and decades.

We present here data collected from two As hotspots in West Bengal and Cambodia. In West Bengal, δ^{18}O, δD and tritium data of shallow groundwater suggest a contribution of modern evaporated surface waters to As rich groundwaters at depths of up to 80 m [3]. In Cambodia, preliminary ^{14}C ages of dissolved inorganic and organic carbon within the aquifer sands show a depth dependent trend of increasing age along a flow path, with residence times of between 1000 and 4000 years at depths of 20 and 50 m respectively. Flow-path integrated rates of As release decrease with increasing depth, suggestive of a surface or near surface source of OM driving microbially mediated As release. The decreased As release rate at depth along a flow path suggests a potential role of abiotic desorption processes and groundwater flow regimes in controlling the spatial distribution of As within the subsurface. Comparisons between these two sites suggests that groundwater pumping practices may play a key role in contributing labile OM to the subsurface and in controlling the spatial distribution of As in this region.

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Experimental fractionation of Ni stable isotopes between metal and silicates at 500-950°C and 10 kbar

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We determined the equilibrium nickel stable isotope fractionation between metal and silicate phases at 500-950°C and 10 kbar, using piston-cylinder expeirments and the three isotope method [1, 2]. We combined NiO enriched in ^{58}Ni with natural quartz, terrestrial Ni metal and H_2O in sealed Au capsules, and held the experiments at P and T for different times. A final invariant assemblage of Ni metal, Ni talc, quartz and H_2O was produced. BSE imaging revealed that metal and quartz were consumed to produce talc, consistent with the reaction: $3Ni^{\circ}+4SiO_2+H_2O+1.5O_2 \Leftrightarrow Ni_3Si_4O_{10}\,(OH)_2$. Our spiked starting NiO was isotopically shifted from the terrestrial mass fractionation line (δ^{61}Ni & δ^{62}Ni $\approx\sim$ -28‰). Analysis by MC-ICPMS yielded Δ^{62}Ni (metal-silicate) values of -0.76±0.01‰ at 500°C, -0.38±0.01 ‰ at 800°C, and -0.15±0.01 ‰ at 950°C. A best fit to the data gives $\Delta^{62}\text{Ni} = 0.45\pm0.03 \times 10^6/T^2$.

Parallel quartz-free experiments containing similarly isotopically-spiked NiO and terrestrial metal did not react to a new assemblage and yielded no isotopic shifts between oxide and metal. This suggests that, despite the substantial isotopic gradient between phases, intergranular diffusion was not an important exchange mechanism in our experiments. We instead propose that recrystallization processes such as nucleation, growth and annealing caused the observed isotopic exchange. Such processes occur as a system chemically re-equilibrates to a stable mineral assemblage [1]. Our talc-bearing experiments began with a metastable assemblage, but the Ni-NiO experiments began at or near chemical equilibirum.

Our preliminary data, coupled with the similar Ni coordination environment among nickel silicates, suggests that there may be a measurable high-temperature metal-silicate fractionation of Ni isotopes. Core forming processes could lead to Ni isotopic fractionation on a planetary scale. Redistribution of Ni during metal growth attending serpentinization may also be accompanied by strong fractionation of stable Ni isotopes.

[1] Shahar, Young & Manning (2008) *EPSL* **268**, 330–338. [2] Matsuhisa, Goldsmith & Clayton (1978) *GCA* **42**, 173–182.