Numerical evaluation of arsenic mobilisation during deepwell injection of aerobic groundwater into a pyritic aquifer

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Managed aquifer recharge is being widely promoted as an attractive technique to meet growing water demands. It involves the injection of, for example, treated or reclaimed water into permeable formations for later withdrawal.

An impediment for all applications, where oxygenated water is recharged into anoxic aquifers, can be the mobilization of trace metals, including arsenic.

While conceptual models for the fate of arsenic under such circumstances exist, they are generally not rigorously tested through translation into numerical modelling approaches and subsequent application to field data sets.

In this study, geochemical data from a deepwell injection trial in The Netherlands, where arsenic mobilization resulted from the introduction of oxygenated water into an anoxic aquifer, was used to test several conceptual models of arsenic mobilization under natural flow conditions.

A reactive transport model was developed to explore physical, chemical, and biochemical interactions that influence arsenic mobility under transient geochemical conditions at the field scale.

The first part of the study focussed on the simulation of the non-reactive transport behaviour. In the subsequent part of the study, a calibration for the major ion and redox chemistry was performed, where pyrite oxidation and the formation of amorphous iron-oxides were shown to be key chemical processes for water quality changes. In the final part of the study various models for arsenic release and sorption were tested.

In the model that best reproduced field observations the fate of arsenic could be explained by (i) release/mobilisation via co-dissolution of arsenopyrite, stoichiometrically linked to pyrite oxidation (ii) kinetically controlled oxidation of dissolved As(III) to As(V) and (iii) As adsorption via surface complexation on neo-precipitated amorphous iron oxides.

Links between planetary degassing, gold and the redox state of Late Archean Oceans?

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Mineral systems that created the Earth's major mineral deposits and provinces were thermo-chemical engines with roots deep in the mantle, driven by a degassing Earth. Such models predict links between metallogenesis and secular changes in architecture and geochemistry of the planet over some 4.5 billion years.

Recent studies of ~2.65 Ga gold systems in the Yilgarn Craton, Western Australia, have emphasised the interaction of intrinsically reduced ($CH_4 \pm H_2 \pm N_2 \pm HCl$) and oxidized ($CO_2 \pm SO_2$) anhydrous fluids of mantle ± lower crustal origin. Inferred $\delta^{13}C$ CO_2 (-4 to -6 ‰) and $\delta^{34}S$ SO_2 (~ 0 ‰) fluid-values are consistent with mantle sources. By analogy with modern island-arc vlocanism, $CO_2 \pm SO_2$ fluids probably originate in the sub-continental lithosphere. Differences in noble gas chemistry [1] between CO_2 fluids ($^{40}Ar/^{36}Ar \leq 21000$) and CH_4 fluids ($^{40}Ar/^{36}Ar$ of $\leq 50,000$) confirms different sources for the reduced and oxidized fluids. The reduced fluids may have been derived from serpentinisation reactions in the deep-crust or mantle wedge. Thermodynamic studies, and inclusion mineralogy of diamondiferous kimberlites and lamproites suggest hydric fluids could dominate in the Earth at depths greater than 300 - 400 km. Ne isotope data track the crustal contribution. Elevated $^{21}Ne/^{22}Ne$ (0.24 - 0.55) in distal carbonate alteration is linked to in-mixing of aqueous crustal fluids in upper crustal sites of Au deposition. $^{20}Ne/^{22}Ne$ from proximal pyrite (8.5-11.4) indicates mixing of mantle and crustal sources, inferred to have occurred in the mid to lower crust.

The variation of $\delta^{13}C$, $\Delta^{33}S$ in sedimentary sulfides and phenomena such as banded iron formation are sensitive indicators of redox variations in the Earth's hydrosphere and atmosphere and may be used to map the impact of deep-Earth degassing. Levels of $CH_4 \pm H_2$ and $CO_2 \pm SO_2$ fluctuated in the hydrosphere/atmosphere prior to ~ 2.7 with H_2 dominating post ~ 2.5Ga. Formation of Late Archean Au deposits, at ~ 2.7 to 2.63 Ga, can be interpreted as one manifestation of planetary degassing of highly reduced volatiles from ~ 2.7 to ~ 2.5 Ga as evidenced by a decline in BIF deposition after ~ 2.7 Ga and an excursion in $\Delta^{33}S$.

[1] Kendrick *et al.* (2008) *AESC Abstracts* **89**, 153.