

Identification of the mechanism for fluoride and phosphate release during managed aquifer recharge

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During managed aquifer recharge (MAR) the injection of purified wastewater can create a geochemical disequilibrium that generally triggers various water-rock interactions. For selected sedimentary aquifer types this also induces the risk of mobilizing geogenic fluoride.

Carbonate-fluorapatite (CFA = $\text{Ca}_{10}(\text{PO}_4)_5(\text{CO}_3\text{F})\text{F}_2$) is by far the most common autochthonous phosphate mineral in sedimentary environments [1]. However, aquifers containing CFA do not necessarily contain high dissolved fluoride concentrations. Dissolution of calcium apatites is complex involving rapid exchange processes and the formation of a surface layer of dicalcium phosphate (CaHPO_4) composition that controls dissolution at circum-neutral pH [2].

In sedimentary aquifers significant fluoride release from fluoride-bearing phosphate minerals has been found where equilibrium with the surface layer is disturbed. This especially occurs in conjunction with calcium removal due to exchange reactions along natural groundwater flow paths [3]. Such a disequilibrium can also be induced by MAR with highly purified, recycled wastewater. However, to date the risk of fluoride and phosphate mobilisation by MAR with highly purified water and the mechanisms by which the CFA surface-layer adjusts to lower ionic strength water has not been recognized and adequately assessed.

In this study we use the data collected during a comprehensive field injection trial with low ionic strength, purified wastewater into an otherwise low fluoride (<0.3 mg/L) sedimentary aquifer to identify and quantify the mechanisms controlling the fate of fluoride and phosphate. Our data from a large-scale field trial document the linkage between the injectant breakthrough and rising fluoride and phosphate concentration. Complementary mineral characterization and laboratory experiments verified CFA as the main source of fluoride and phosphate release.

[1] Föllmi, K.B., *Earth-Sci. Rev.*, 1996. 40(1–2): p. 55-124.

[2] Chairat, C., et al., *GCA*, 2007. 71(24): p. 5901-5912.

[3] Edmunds, W.M. in *Ess. of Med. Geol.*, 2013. p. 311-336.