

## Geogenic radium mobilization in the iron- and manganese-reducing zone of a DNAPL-contaminated aquifer

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Radium (Ra) occurs naturally in aquifers, produced via decay of parent isotopes (e.g., <sup>232</sup>Th, <sup>238</sup>U). It is a known carcinogen associated with development of bone cancer, and is therefore regulated in U.S. drinking water at 185 mBq (5 pCi/L) for the combined total of <sup>226</sup>Ra and <sup>228</sup>Ra. Radium may be released from rock to groundwater through several mechanisms, including the reductive dissolution of Ra-bearing iron (Fe) and manganese (Mn) (hydr)oxides. At hydrocarbon-contaminated sites the biodegradation of organic carbon is typically coupled with reductive dissolution of Fe and Mn (hydr)oxides, and therefore has potential to release Ra to groundwater.

In this study, Ra cycling was investigated at a field site near Cottage Grove, Wisconsin USA, where the groundwater is contaminated with a mixture of chlorinated solvents, ketones, and aromatics occurring as a dense non-aqueous phase liquid (DNAPL) in the source zone. Groundwater samples were collected from multiple depth-discrete intervals in the Tunnel City Group sandstone of the Midwestern Cambrian-Ordovician aquifer system using a Westbay® multi-level well monitoring network. Well MP-16, located outside the dissolved phase plume, was used as a baseline for “background” dissolved <sup>226</sup>Ra activities. Well MP-24S and MP-19S are located in the plume immediately and ~600 m east of the source zone, respectively, in the direction of contaminant plume migration. Samples were analyzed for <sup>226</sup>Ra and <sup>228</sup>Ra using multi-collector inductively-coupled plasma mass spectrometry. Aqueous <sup>226</sup>Ra activities at MP-24S and MP-19S are heterogeneous with depth and range from 3.2-42 mBq/L, up to eight times higher than background <sup>226</sup>Ra activities. Aqueous Ra activity is strongly correlated with dissolved Fe and Mn concentrations, supporting the hypothesis that Ra is released to groundwater via reductive dissolution of Ra-bearing Fe and Mn- (hydr)oxides. However, aqueous Ra activities in the Fe and Mn-reducing zone are lower than expected, suggesting there is another important Ra sink in the aquifer; for example, some of the mobilized Ra may have re-sorbed to clays or Fe and Mn (hydr)oxides downgradient. This study demonstrates the potential for natural attenuation of hydrocarbon-contaminated aquifers to release geogenic contaminants such as Ra via the reduction of Fe and Mn (hydr)oxides.