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Use of 222Rn as natural tracer for LNAPL quantification and recovery efficiency in a crude-oil contaminated aquifer

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In august 2009, five hectares of the pristine gravel aquifer of Crau in southern France were contaminated by 5,100 m3 of crude oil due to the sudden break of a pipeline. The remediation of this site is still ongoing and consists in replacement and off-site disposal of contaminated topsoils, plume management by hydraulic groundwater barriers with re-injection of activated charcoal-treated waters, and dual-phase LNAPL extraction in the source zone. It is anticipated to stop these remediation actions when the rate of hydrocarbon extraction becomes inefficient. The volume of LNAPL is estimated between 100 and 1000 m3. A more accurate estimation is needed for the implementation of natural attenuation once physical treatment is discontinued. 222Rn has been introduced as a natural tracer for the quantification of LNAPL saturation in porous media under natural gradient conditions (Hunkeler et al., 1997; Semprini et al., 2000; Schubert et al., 2007).
The objective of this study was to investigate whether 222Rn in groundwater can be used as a tracer for LNAPL quantification at a field site treated by LNAPL removal. To this end, groundwater samples were obtained in pristine monitoring wells from upgradient the contamination using submersible electric pumps, and in LNAPL recovery wells. There, samples were obtained from the tap on the hard PVC tubing used for pumping groundwater to the treatment facility. For 222Rn analysis, flasks of 250 mL were gently filled and were capped thereafter without permitting air bubbles. The flasks were analysed within 6 to 24 hours. The 222Rn activity of groundwater was measured by a Rn detector (RAD7-Durridge, Co. Inc.). The measurements were spaced over more than 15 months in order to account for seasonal changes. Each well was sampled at least 3 times.
In pristine groundwater, the radon activity was relatively constant and remained always > 14 Bq/L. The radon activities in the groundwater of source zone wells were also relatively constant and the mean activities were generally significantly lower than upgradient. This is due to partitioning into the oil phase. Decreases were correlated with NAPL recovery efficiency. The laboratory-determined crude oil-water partitioning coefficient of $38{,}5 \pm 2{,}9$ was used for estimating LNAPL saturation in each recovery well. However, extrapolations of LNAPL saturations to whole-site oil volume estimations are difficult since at low water tables, the volume in the capillary fringe is not assessed. Nevertheless, we find that 222Rn is a useful and cheap groundwater tracer for finding zones of good LNAPL recovery in a heavily pumped aquifer.

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