



Documentation of time-scales for onset of natural attenuation in an aquifer treated by a crude-oil recovery system



Violaine Ponsin^{a,b}, Joachim Maier^c, Yves Guelorget^c, Daniel Hunkeler^d, Daniel Bouchard^d, Hakeline Villavicencio^d, Patrick Höhener^{a,*}

^a Aix-Marseille Université-CNRS, Laboratoire Chimie Environnement FRE, 3416 Marseille, France

^b French Environment and Energy Management Agency, 20 avenue de Grésillé, BP 90406 Angers Cedex 01, France

^c ICF Environnement, 14/30 rue Alexandre Bâtiment C F, 92635 Gennevilliers, France

^d Centre for Hydrogeology, University of Neuchâtel, Rue Emile-Argand 11, CH-2000 Neuchâtel, Switzerland

HIGHLIGHTS

- One of the world largest terrestrial oil spills is studied for 4 years.
- Initially pristine aerobic groundwater turns anoxic in 8 months.
- Sulfate reduction is the most important redox process thereafter.
- Biologically enhanced dissolution of toluene and benzene is evidenced.
- Stable carbon isotopes prove the degradation of benzene and ethylbenzene.

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ABSTRACT

A pipeline transporting crude-oil broke in a nature reserve in 2009 and spilled 5100 m³ of oil that partly reached the aquifer and formed progressively a floating oil lens. Groundwater monitoring started immediately after the spill and crude-oil recovery by dual pump-and-skim technology was operated after oil lens formation. This study aimed at documenting the implementation of redox-specific natural attenuation processes in the saturated zone and at assessing whether dissolved compounds were degraded. Seven targeted water sampling campaigns were done during four years in addition to a routine monitoring of hydrocarbon concentrations. Liquid oil reached the aquifer within 2.5 months, and anaerobic processes, from denitrification to reduction of sulfate, were observable after 8 months. Methanogenesis appeared on site after 28 months. Stable carbon isotope analyses after 16 months showed maximum shifts in $\delta^{13}\text{C}$ of $+4.9 \pm 0.22\%$ for toluene, $+2.4 \pm 0.19\%$ for benzene and $+0.9 \pm 0.51\%$ for ethylbenzene, suggesting anaerobic degradation of these compounds in the source zone. Estimations of fluxes of inorganic carbon produced by biodegradation revealed that, in average, 60% of inorganic carbon production was attributable to sulfate reduction. This percentage tended to decrease with time while the production of carbon attributable to methanogenesis was increasing. Within the investigation time frame, mass balance estimations showed that biodegradation is a more efficient process for control of dissolved concentrations compared to pumping and filtration on an activated charcoal filter.

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1. Introduction

Most of petroleum hydrocarbon-contaminated sites in Europe are industrial sites or storage sites (Van Liedekerke et al., 2014). These sites are often subjected to progressive and slowly emerging contamination, created for example by leaking tanks, and once the pollution is discovered, it is difficult to assess when the groundwater ecosystem switched from a pristine system to a contaminated one because historical

data are rarely available. A major industrial accident such as a pipeline rupture is different because the moment when the pollution occurs is generally clearly identified by a loss of pressure followed by discontinuation of delivery. The United States have more than 240,000 km of oil pipeline (Trench, 2003), and the average annual number of crude-oil pipeline spills per year per 1000 miles was 1.5 (i.e. 0.9 per 1000 km) for the 2007–2009 period, for a total amount of oil released of 111 m³ (Delin and Herkelrath, 2014). In comparison, oil pipelines in Europe represent a total length of 36,000 km and the average annual number of pipeline oil spills (including crude oil and oil products) per 1000 km was 0.22 for the 2008–2012 period, for a total amount of 371 m³ of oil released

* Corresponding author.

E-mail address: patrick.hohener@univ-amu.fr (P. Höhener).

(Davis et al., 2013). These numbers suggest that the consequences of pipeline bursts on the environmental vulnerability of aquifers merits further research.

The crude-oil spill in Bemidji (Minnesota, USA) is a textbook case regarding pipeline spill research. A pipeline rupture in 1979 contaminated a shallow aquifer with 1700 m³ of oil. The United States Geological Survey (USGS) Toxic Substances Hydrology Program sponsored a long-term and interdisciplinary project that started in 1983 (Delin et al., 1998). The objective of the project was to improve the understanding of the mobilization, transport, and fate of crude oil in the shallow subsurface. Work at the Bemidji site has ranged from characterization of microscopic-scale microbes-mineral interactions (Hiebert and Bennett, 1992; Bennett et al., 2000; Roberts, 2004; Rogers and Bennett, 2004) to plume-scale geochemical and microbial evolution (Baedeker et al., 1993; Bennett et al., 1993; Eganhouse et al., 1993; Cozzarelli et al., 1994; Bekins et al., 1999), and has included testing of complex models of multiphase flow, reactive transport, and biodegradation (Essaid et al., 1995, 2003; Curtis, 2003; Molins et al., 2010). More recent work focused on processes occurring in the vadose zone (Chaplin et al., 2002; Amos et al., 2005; Sihota and Mayer, 2012). One of their most important conclusions is that transport and fate of hydrocarbons in the subsurface is a spatially and temporally complex problem, and that only long-term monitoring can help to understand and predict the evolution of subsurface hydrocarbon plumes. Although work at Bemidji gave a very complete insight into processes occurring on the long-term at such a site, the very initial phases of the evolution of natural attenuation were not studied in great detail due to the absence of research programs in the initial years.

Very few studies actually described the early stages of aerobic and anaerobic degradation after a sudden pollution. Most of those studies were performed on dedicated sites deliberately contaminated to avoid problems associated with poorly defined sources and plumes. The mass of pollutants spilled at those sites are orders of magnitudes smaller compared to the masses at crude-oil pipeline bursts. Käss and Schwillie (1992) poured about four m³ of heating oil into the alluvial aquifer of the Rhine valley in order to study the formation of a hydrocarbon plume but they were not able to document complete geochemical reactions at the time of their pioneering work that started before 1980. The sandy Borden research aquifer in Ontario (Canada) was another such a site: secured distinct parts of this sandy aquifer were used for numerous studies on artificial pollutant release. Natural attenuation of coal tar creosote mixed with aquifer material to create a source zone was thoroughly studied in Borden (King et al., 1999; Fraser et al., 2008), and the first sampling campaign for monitoring of soluble compounds such as phenol, *m*-xylene and naphthalene was done 55 days after source emplacement (King and Barker, 1999). However, most of the processes that control the fate of hydrocarbons, such as aerobic degradation, were already occurring at Borden, and mass release and study duration were often not sufficient to document the change of redox state in this aquifer.

On August 7th, 2009, a pipeline transporting crude oil buried at 1 m depth broke in a steppe grassland nature reserve of restricted access, 50 km northwest of Marseille (France), and about 5100 m³ of oil under pressure were projected to the surface where they spread across 5 ha of flat arid landscape. The crude oil later infiltrated through consolidated material via fractures, and formed a floating oil body in the underlying aquifer. The largest extension of the pool was covering about 4 ha. The underlying aquifer is the most important regional aquifer (520 km²) and is under high pressure for water supply (Roux, 2006). It has been thus thoroughly studied since 1975 and a model was developed by the French Geological Survey to achieve optimal water management (Berard et al., 1995). In view of potential application of natural attenuation as management option at this site, research on the evolution of attenuation processes was initiated from the very beginning. Monitoring in La Crau started as soon as wells were drilled, while oil had not yet reached the aquifer and was still migrating through the vadose zone.

The objectives of this study were: (1) to monitor the temporal and spatial evolution of redox processes in the saturated zone during a four-year period following a massive oil spill in a pristine aquifer (2), to assess when and where hydrocarbon degradation and especially benzene degradation was occurring (3) to quantify ongoing biotic degradation processes through mass balances and carbon fluxes, and (4) to identify the dominant redox processes.

2. Material and methods

2.1. Site description

The site is in the “La Crau” alluvial plain, on 16 m above sea level (Fig. 1). This plain was formed by the former river Durance during the Pleistocene (Naudet et al., 2004). Deposits are made of coarse gravel of various alpine rocks with fine gravel and sand in the interstices. They are cemented in the upper meters by calcareous concretions formed by evaporation, in the form of a Puddingstone with fractures (Figure S1, Sup. mat). The deeper parts of the gravel deposits are free from concretions, have hydraulic conductivities of $0.2\text{--}2 \times 10^{-3} \text{ m s}^{-1}$, and form the most important regional aquifer used for irrigation and drinking water supply. Gravel deposits extend over approximately 15 m and overlay silt or sandy-silt deposits of low permeability. The groundwater table is at 8–11 m below surface and shows important seasonal variations of up to 3 m caused by local rainfall and irrigation upgradient. The groundwater quality is well documented, and concentrations of aqueous species and pH values in background water are summarized in Table 1.

2.2. Remediation

The first remedial actions consisted in excavation and off-site disposal of oil-soaked surface soils. 19 monitoring wells were installed within two months, and the site was characterized in detail by hydrogeological and geophysical methods. The presence of Light Non Aqueous Liquid Phase (LNAPL) was observed in three monitoring wells 2.5 months after the spill and a dissolved plume was detected thereafter. Subsequently, 17 new wells devoted to hydraulic plume management and 30 more wells for tentative LNAPL recovery or monitoring were then installed (Fig. 1, information about the wells named throughout the paper in Table S1, Sup. Mat). Recovery wells were equipped with an electric submersed pump positioned deep below water table delivering continuously $0.5 \text{ to } 2 \text{ m}^3 \text{ h}^{-1}$ of groundwater, and a pneumatic pump just below the water table filled gravitationally by LNAPL. This latter was delivered by pressurized air in cycles to a recovery tank. Recovery started in January 2010 in three wells and was extended progressively as a function of LNAPL arrival in other wells. LNAPL recovery was operated in a maximum of 22 wells until the end of March 2014. A total of 37 m³ of LNAPL was recovered from the aquifer within four years. Water pumped in recovery wells, together with $20\text{--}25 \text{ m}^3 \text{ h}^{-1}$ pumped downgradient of the site to cut the dissolved plume, was mixed in an oil separator, analyzed weekly, treated by an activated charcoal filter and re-injected upgradient in seven wells.

2.3. Routine monitoring and targeted sampling campaigns

The routine site monitoring requested by authorities included a monthly survey of piezometric heads, basic water quality parameters (T, O₂, pH, EC) and hydrocarbons in 17 monitoring wells positioned all outside, downgradient and upgradient of the source zone. These monitoring wells included Pz4 and Pz17 (Fig. 1) and 15 other wells distant from 50 to 700 m of the border of the source zone (not pictured in Fig. 1). This monitoring was completed by a bi-weekly hydrocarbon analysis (BTEX, aliphatics C5–C35, other aromatics) of the pumped water at the inflow and the outflow of the activated charcoal filter. For both monthly and weekly monitoring, hydrocarbon analyses were performed by an accredited lab (Eurofins) by GC–MS.

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