



Unraveling the partial failure of a permeable reactive barrier using a multi-tracer experiment and Cr isotope measurements



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ABSTRACT

At a Cr(VI) contaminated site in Thun, Switzerland, a permeable reactive barrier (PRB) was installed in 2008. Downstream Cr(VI) concentrations did not indicate any sign of its successful operation more than 2 years after PRB installation. The cause for this potential PRB failure was investigated by performing Cr isotope measurements and a multi-tracer experiment. The combination of reactive (Cr isotopes) and non-reactive tracers allowed characterizing the groundwater flow regime in the vicinity of the PRB in detail. In particular, it could be confirmed that most of the Cr(VI) load is currently bypassing the barrier, whereas only a minor Cr(VI) load is flowing through the PRB. Fitting of observed breakthrough curves using a conventional advection dispersion model resulted in average linear flow velocities of 13–15 m/day for the bypassing Cr(VI) load and 4–5 m/day for the Cr(VI) flowing through the barrier. Using a Rayleigh fractionation model a Cr(VI) reduction efficiency of 77–98% was estimated for the Cr(VI) load that is flowing through the barrier. In contrast, a value of 0–23% was estimated for the current overall PRB reduction efficiency. It is concluded that the PRB bypass and the low overall Cr(VI) reduction efficiency are caused by a limited PRB permeability inherited from skin effects that occurred during PRB emplacement.

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1. Introduction

The installation of permeable reactive barriers (PRB) is an accepted *in situ* technology for the cleanup of a wide range of groundwater contaminants (Blowes et al., 2000; Mayer et al., 2001; Naftz et al., 2002; Flury et al., 2009; Jeen et al., 2011). Among the most widely used reactive materials is metallic iron, Fe⁰ (Powell et al., 1995; Ponder et al., 2000; Naftz et al., 2007). Since it is not stable in water, Fe⁰ is readily oxidized to Fe²⁺ and Fe³⁺ inducing reducing groundwater conditions. Under such conditions some of the most prominent groundwater contaminants (e.g., chlorinated ethenes, Cr(VI)) are degraded or reductively immobilized. At a significant number of field scale PRB installations, however, a complete and long-term contaminant removal was not achieved (ITRC, 2005; Burmeier et al., 2006). This is especially true when referring to a relatively short time span of ca. 15 a, during which such installations have been implemented. The most prominent causes reported for inefficient PRB operations are pore space clogging and a reactivity decrease of the reactive material, both induced by the precipitation of secondary minerals such as Fe-hydroxides and carbonates (Henderson and Demond, 2007; Jeen et al., 2007; Flury et al., 2009; Wanner et al., 2011). A

long-term monitoring of ongoing PRB performance is, therefore, required for any kind of installation.

Quantitative assessment of contaminant removal efficiency of PRBs is not straightforward because conventional concentration measurements do not distinguish between decreasing concentrations due to actual contaminant removal or due to the effects of dilution and/or dispersion. A powerful and quite novel tool is the tracking of stable isotope fractionation along a groundwater flow path within a treatment zone (Blowes, 2002). The concept benefits from the fact that groundwater contaminants such as Cr(VI) and chlorinated ethenes are subject to a significant stable isotope fractionation when they are reduced (Powell et al., 1995; Ponder et al., 2000; Ellis et al., 2002; Naftz et al., 2002; Hunkeler et al., 2008). In the case of Cr(VI) reduction, Cr isotope fractionation occurs because the process favors the lighter of the four stable Cr isotopes (⁵⁰Cr, ⁵²Cr, ⁵³Cr, ⁵⁴Cr) to be accumulated in the reduced form, Cr(III). Hence, if a highly soluble Cr(VI) load is reduced to less soluble Cr(III), remnant Cr(VI) becomes successively heavier. In terms of a more general PRB performance assessment, reducing groundwater conditions can also be tracked by measuring stable S and N isotopes. Such measurements are powerful because the reduction of the major groundwater constituents NO₃⁻ and SO₄²⁻ is accompanied by a significant isotopic fractionation (Green et al., 2010; Gibson et al., 2011). To date, performance assessments of field scale PRBs using stable Cr, N and S isotope measurements have only been performed in a few selected studies (Gibson et al.,

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2011; Wanner et al., 2012b), whereas stable C isotope measurements have been applied in numerous field studies tracking the reductive degradation of chlorinated ethenes (see compilation by Hunkeler et al., 2008). Moreover, Cr isotope measurements have been used to track naturally occurring Cr(VI) reduction (Izbicki et al., 2008, 2012; Berna et al., 2010; Villalobos-Aragon et al., 2012; Wanner et al., 2012a).

This study investigated the cause for the partial failure of a PRB that was demonstrated recently by performing a first series of Cr isotope measurements and reactive transport model simulations (Wanner et al., 2012b). New data include Cr isotope measurements from a second and more extensive sampling campaign as well as data from a multi-tracer experiment using fluorescent dyes. The combination of reactive (Cr isotopes) and non reactive tracers allowed characterizing the groundwater flow regime in the vicinity of the PRB and quantifying the extent of Cr(VI) reduction induced by the PRB.

2. Materials and methods

2.1. Study site

The remediation site is located in Thun, Switzerland, where chromic acid was used for metal processing activities until the mid 1970s. Remnants of spilled chromic acid form the current shallow Cr(VI) source and still induce a groundwater plume with a maximum Cr(VI) concentration of 4 mg/L. Due to acid cleaning of Cu wires, the site is also heavily contaminated with this metal. The source zone covers an area of ca. 30 m² and reaches a maximum depth of about 7 m. The Cr(VI) plume is contained in a carbonate dominated gravel aquifer, where the average groundwater table is 2–4 m below surface. The hydrological situation is strongly influenced by the river Aare that is flowing less than 100 m north of the contamination hotspot (Fig. 1). A hydroelectric power station is located about 100 m downstream of the hotspot causing a sharp drop in the river water table (ca. 2–3 m). Accordingly, the river is recharging the aquifer upstream of the power station (adjacent to the hotspot), whereas the aquifer

discharges into the river downstream of the power station. Shallow groundwater at the contamination hotspot, therefore, almost exclusively originates from the river. The mineralogical composition of the aquifer and the strong hydrologic control by the river leads to a groundwater composition that is almost saturated with respect to O₂ and CaCO₃. Remediation by a PRB was chosen because the Cr(VI) source could not have been accessed due to its location beneath a protected building of historical value. The PRB installation was performed in May 2008 approximately 30 m downstream of the Cr(VI) hotspot. The PRB design was adopted from a pilot scale installation located in Willisau, Switzerland (Flury et al., 2009). It consists of a double array of a total of 62 overlapping vertical piles (Fig. 1). The individual piles are 1.3 m in diameter, reach a depth of ca. 13 m and were filled with a mixture of Fe shavings and gravel. At an Fe to gravel weight ratio of 3:1 a total of 352 tons of Fe was used. The Fe shavings consists of gray cast Fe, which were recently characterized in detail (Wanner et al., 2011).

No reduction of the Cr(VI) plume has been observed since the installation of the PRB (Wanner et al., 2012b): after installation, Cr(VI) concentrations measured at borehole KB08/03, which is located directly downstream of the PRB (Fig. 1) increased from <0.01 to values between 0.05 and 0.1 mg/L, and at boreholes located further downstream (e.g., KB01/05 and KB07/26, Fig. 1) no significant change in Cr(VI) concentrations were observed when compared to those before installation. Preliminary Cr isotope measurements and reactive transport model simulations suggested that these problems occurred because a proper PRB flow through was hindered by limited permeability in the PRB (Wanner et al., 2012b).

2.2. Multi-tracer experiment

Two fluorescent tracers were simultaneously injected into the Cr(VI) hotspot area in September 2010 to validate the suspected low PRB permeability hypothesis using an independent experimental method. Two hundred g of Na-fluorescein (Na₂C₂₀H₁₀O₅) were injected into borehole KB06/16 and 3 kg of Na-naphthionate

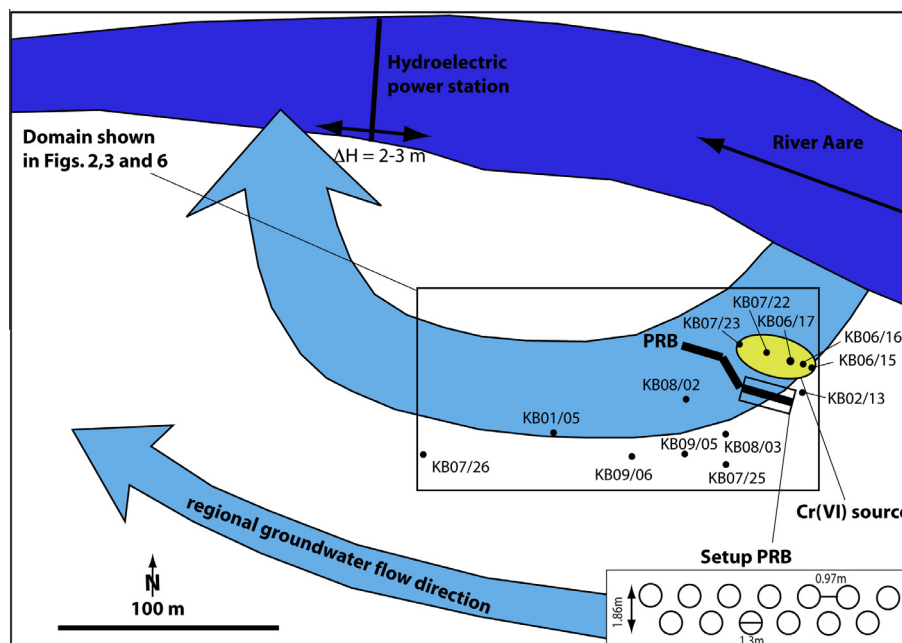


Fig. 1. Overview of the Thun site schematically illustrating the general hydrological conditions and the locations of the Cr(VI) hotspot, the PRB, as well as observation boreholes. Due to the proximity to the river Aare and the presence of a hydroelectric power station, groundwater flow in the vicinity of the PRB is strongly controlled by recharging river water. Also shown is the general setup of the PRB providing the detailed geometry of 13 out of 62 piles (insert on lower right). The central rectangle defines the domain for which results are presented (Figs. 2, 3 and 6).

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