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Colloid and heavy metal transport at landfill sites in direct contact with groundwater

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ABSTRACT

Colloids are ubiquitous in aquatic systems and are suspected of facilitating contaminant transport. At sites where the disposed waste is in direct contact with ground water, two main prerequisites for colloidal transport are fulfilled: these two prerequisites are a high concentration of colloids and many different contaminants, some that are very unlikely to be transported in an aqueous solution.

In our investigation, three landfill sites with different historical background and hydrogeological conditions were examined. The colloids upstream, downstream, and inside the waste disposal sites were characterized with respect to their size distribution and chemical composition. The particle concentration upgradient and downgradient of the sites were 2–23 and 8–80 mg/L, respectively. Inside the waste disposal sites the particle concentration was 160–870 mg/L. The particles upgradient reflected the mineral composition of the aquifer, with calcareous colloids and silicates dominating the composition. Downgradient of the sites, we saw an increase of iron-precipitates and salt colloids, together with organic colloids. However, the downgradient colloids were significantly different from the colloids and particles inside of the disposal sites with respect to their size and chemical composition. Colloids inside the disposal sites reflected the waste composition and degradation.

The association of heavy metal ions to colloids and particles showed a surprisingly high fraction of dissolved metal ions. We determined that the lowest metal ion concentrations (less than 20% of the total concentration) were associated with the colloid size class between 10 nm and 1 μm, which is considered most mobile in porous systems. The association of Fe and Mn to colloids was dominated by the redox conditions inside the disposal sites, where there was a reducing environment, Fe and Mn were dissolved. Outside the disposal sites, where there was an oxidizing environment, these metals formed colloids and particles >1 μm. Together with these particles, As was precipitating. For other metal ions (Cd, Co, Cu, Ni, Pb, Zn), we determined an association to colloids coinciding roughly with the colloid size distribution.

The results suggest that the change of hydrochemical conditions at the interface, from a reducing, high ionic strength environment inside of the disposal sites to an oxidizing, low ionic strength environment in the groundwater together with physical filtration effects for the larger particles, is an effective chemical barrier for colloids. Field observations suggest, that the colloids form a rather persistent coating around the aquifer matrix that reduces the hydraulic conductivity and enhances the sorption capacity of the aquifer close to the waste disposal sites.

In every case, there was an increase of the contaminant concentrations downstream of the waste disposal sites, but the increase was less than expected from the initial transport

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calculations, which were under the assumption that there was a direct contact between the waste disposal site and the groundwater. It seems more than likely, that under the given conditions, colloids are contributing to a self sealing layer at the landfill bottom. The results of this study are relevant for the assessment of many landfills without appropriate landfill sealing systems.

1. Introduction

There are still many landfills without proper sealing systems, which are emitting toxic substances such as heavy metals into groundwater. Heavy metals are well known to bind to colloidal matter present in leachate and groundwater (Gounaris et al., 1993; Baun and Christensen, 2004). When associated with colloids, heavy metals may show significantly different transport properties compared to dissolved heavy metals. However, the transport of heavy metals in the presence of colloids may be enhanced or retarded (McCarthy and Zachara, 1989; Kretzschmar et al., 1999), depending on the hydrogeological and hydrochemical conditions. Thus, the presence of colloids in landfill leachate that contaminates the groundwater may alter the propagation of heavy metals in the contamination plume.

Colloids and colloid transport are very sensitive to hydrochemical and hydrodynamic conditions (Roy and Dzombak, 1997a,b; Bergendahl and Grasso, 2000; Bradford et al., 2002; Ryan and Elimelech, 1996a). These conditions are likely to occur in the vicinity of landfills (Bjerg et al., 1995). Therefore, both mobilization of natural colloids and precipitation of colloids from landfill leachate are likely to occur downgradient of landfills.

In leachate from four Danish landfills, generally more than 60% of the total heavy metal concentrations in the leachate were associated to colloids (Jensen and Christensen, 1999). In another study, contaminated groundwater close to landfills was spiked with heavy metal ions, which were then found to be associated to colloids (Jensen et al., 1999). Thus, at disposal sites with direct contact to groundwater, it seems likely that colloids and colloid bound contaminants are emitted into the aquifer. The above results also suggest that the heavy metal–colloid interaction is stable in contaminated groundwater.

A recent review (Baun and Christensen, 2004) lists only eight field studies addressing the speciation of heavy metals in landfill leachate; five of them address the distribution of heavy metals between colloids and dissolved fractions. This review indicates a significant association between heavy metal ions and organic colloids. However, the association between heavy metal ions and precipitates (iron, sulphide, carbonate, and phosphate) is less significant. The authors did not comment on the association of heavy metals to natural colloidal matter (clay, quartz), due to a lack of data.

One issue that remains open in the current literature is the transport from the landfill into the groundwater. From the current state of knowledge, it seems likely that colloids and heavy metals are emitted into the groundwater together (Jensen and Christensen, 1999). The fact that a large fraction of the heavy metals in landfill leachate, as well as in groundwater, are bound to colloids does not necessarily mean that there is colloidal transport between the landfill and groundwater, although this seems plausible. It might also be possible that colloid bound heavy metals are filtered at the interface, between the landfill and groundwater; thus, leading to a retardation of colloids and heavy metals (Ryan and Elimelech, 1996b; Grolimund et al., 1996; Kretzschmar et al., 1999). Finally, dissolved heavy metals released from the landfill might bind to colloids in the groundwater (Jensen et al., 1999).

This study examines colloids in the leachate, in the groundwater upgradient, and in the groundwater downgradient of three different landfill sites in Bavaria (Germany). The size distribution of colloids, the chemical/mineral composition of colloids, and the distribution of heavy metals between the aqueous solution and colloid size fractions are analyzed to gain insight into the transport pathways.

2. Materials and methods

2.1. *Sampling and analysis*

Sampling was done at monitoring wells in the vicinity of the landfill sites. The wells were usually 114 to 150 mm in diameter. The PVC filter screen reached from the groundwater table to the bottom of the aquifer. The screen width was at least 1 mm. Filter gravel (2–4 mm) was filled between the well and the aquifer matrix. Prior to sampling, the wells were pumped at 0.5–0.8 L/s until the electrical conductivity, the pH-value, and the redox potential reached steady state. Then, the pumping rate was lowered to ≈100 mL/min to avoid remobilization of colloids (Degueldre et al., 1999). Samples were stored in a dark and cool place (4–5 °C) and usually prepared and analyzed on the same day.

Samples were sequentially filtered through polycarbonate filters (10.0–1.0–0.1–0.01–0.001 μm, INFILTEC, Speyer), using ultrafiltration in a stirring cell (see Klein and Niessner, 1996). Filters were weighed before and after filtration to obtain colloid concentrations and colloid size distributions. Heavy metal concentrations were detected in filtrates and from colloids on the filters (after acid treatment, HNO_3 65% Selectipur, Merck Darmstadt) by atomic adsorption spectroscopy (Perkin Elmer 4100, Überlingen). This procedure obtained the heavy metal concentrations associated with the colloid size classes.

2.2. *Particle characterization*

The colloids were characterized using scanning electron microscopy (SEM, Leica Cambridge Stereoscan 360, Bensheim) that was combined with an energy dispersive X-ray fluorescence detector (EDX, Röntec, Berlin). First, colloids were