Field Sampling and Modelling of Creosote-Derived Contamination in a Tidally Forced Aquifer

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ABSTRACT We describe research into the fate and transport of a creosote-derived groundwater contaminant plume found in an aquifer adjacent to and beneath the Fraser River. The site, located in Coquitlam, B.C., has been an active wood preserving facility since the 1920's. In the source zone, a dense non-aqueous phase liquid (DNAPL) has penetrated into the Fraser Sands aquifer. A pumping well has been operated since 1997 to contain and capture the contaminant source. Previous research at the site has demonstrated that the primary component of the dissolved phase plume is naphthalene and that biodegradation of naphthalene is taking place, likely through a combination of iron reduction and methanogenesis. Groundwater samples collected in 2002 along the axis of the plume from the portion of the aquifer beneath the Fraser River show contaminant concentrations significantly reduced from 1996 levels. Aqueous concentrations of the two dominant electron acceptors are sufficient to account for mass loss between 1996 and this most recent sampling round. Sorption in the aquifer was assessed through analysis of organic carbon content of soil collected from the plume core. Although the net average gradient within the aquifer is towards the Fraser River, gradients within the aquifer are observed to vary both daily, with tidal fluctuations in river stage, and monthly, with seasonal variations in river stage elevations. A groundwater flow model was constructed to assess the effect of seasonal and tidal fluctuations in groundwater gradients on plume concentrations. We present chemical concentration data, soil core data and model results to explain the plume behaviour.

Introduction

The motivation for this research is to understand the fate and transport of a creosote-derived groundwater contaminant plume located adjacent to and beneath the Fraser River. Any explanation of contaminant fate must account for the complex geochemical and biological processes and the tidally-driven groundwater hydrology at the site. Here we present the results of a field investigation into these processes.

Creosote is the product of the coal tar distillation process and is used as a wood preservative. Because of the widespread historical use of creosote, creosote-derived plumes at wood-preserving facilities are common. Creosote consists of a complex mixture of polycyclic aromatic hydrocarbons (85%), monoaromatic hydrocarbons (MAHs) (10%), and nitrogen, sulphur, and oxygen (NSO) containing heterocyclics (5%) (Mueller et al., 1989).

Research into the evolution of creosote plumes has shown that the different components of creosote travel at different speeds in groundwater, with more highly sorbed components moving more slowly and dominating at later times of plume evolution (Fowler, 1994, King and Barker, 1999). Previous research has suggested that when biodegradation is active, concentrations of compounds are expected to increase to steady state, when the rate of supply equals the rate of degradation and then recede as the flux from the source zone changes over time (King et al., 1999). Recent data has shown that this hypothesis may not necessarily be the case for all components (Blaine et al., 2001). As a highly sorbed component of creosote, naphthalene has been identified as a dominant component at later times in plume evolution.

Although naphthalene is readily degraded in aerobic conditions (Mueller, 1989), anaerobic degradation is less well-documented and is generally believed to be slower. Naphthalene degradation under iron-reducing conditions has been documented in the field (Anderson and Lovley, 1999, Robinson et al., 2001). Recent anaerobic enrichment cultures have demonstrated that complete naphthalene degradation by methanogenesis is also possible where long-term exposure to PAHs exists (Chang et al., 2001).

Considerably less is known about the fate and transport processes involved in the evolution of heterocyclic components of creosote than PAHs. Recent research has shown that benzothiophene, a sulphur-containing heterocyclic component of creosote, is an inhibitor of naphthalene degradation (Annweiler et al, 2001, Meyer, 2000). Although degradation of benzothiophene under sulphate-reducing conditions has been documented (Annweiler et al., 2001), degradation of sulphur-containing heterocyclics has been shown to be much slower than degradation of PAHs (Meyer, 2000). Column experiments designed to study the sorption of heterocyclic compounds have found that benzothiophene has a similar retardation to naphthalene (Broholm et al., 1999).

Tidal forcing can have significant impacts on the distribution of contaminants in a plume as it evolves. Diurnal fluctuations in groundwater flows due to tidal forcing increases the dispersion of contaminants (Yim and Mohsen, 1992) and may increase the potential for biodegradation due to increased mixing of nutrients (El-Kadi, 2001).

Site Description

The site that is the focus of this study is located adjacent to the Fraser River in Coquitlam, British Columbia. The site has hosted an active wood preserving facility since the late 1920's. The source of contamination of the site is a creosote-derived dense non-aqueous phase liquid (DNAPL).

The geologic setting consists of hundreds of metres of recent age glacio-marine, alluvial, swamp and flood plain deposits overlying Tertiary sedimentary bedrock (Monahan, 1997). The Fraser Sands aquifer is the aquifer of interest at the site. It is confined by overlying silt and clay overbank deposits and is underlain by the low permeability Pre-Fraser Sands unit. Flow in the aquifer is primarily horizontal, except for the region beneath the Fraser River, where a vertical component is present as the aquifer discharges to the river.

Extensive site characterization has been undertaken previously (Golder, 1997, Golder, 1998, Anthony, 1998, Bianchin, 2001) as part of efforts to delineate site contamination. Hydrostratigraphy and the DNAPL source at the site have been well characterized by boreholes, cone penetrometer tests, and testpits.

Continuous water level monitoring data collected at the site (Golder, 1997, 1998) show that although the dominant direction of groundwater flow in the aquifer is towards the Fraser River, gradients within the aquifer are observed to vary both daily, with tidal fluctuations in river stage, and monthly, with seasonal variations in river stage elevation. In addition, in the southern region of the site, average aquifer gradients have an east-west component that follows the direction of flow in the river.

The source of contamination at the site is a creosote-derived DNAPL plume which extends into the Fraser sands aquifer in a small localized area to approximately 22 m depth. The deep DNAPL source is located approximately 120 metres from the shore of the Fraser River (Fig. i). Borehole and test pit observations used to map the extent of the DNAPL at the site suggest that this is the only area where the NAPL penetrates into the deep zone of the Fraser Sands aquifer, although it extends in the upper region of the aquifer and in overbank deposits over a much larger region.

In past groundwater sampling efforts, the dissolved phase plume has extended from the region of the deep DNAPL source zone to where it discharges beneath the Fraser River approximately 200 m downgradient. The aqueous plume is composed of PAHs, MAHs and chlorophenols. By far, the predominant component of the plume is naphthalene, although other PAHs have been detected in significant concentrations at the site. All previous groundwater sampling has shown that the plume is anaerobic.

The plume was first mapped offshore where it discharges beneath the Fraser river in 1996 (Anthony, 1998). Contaminant concentrations recorded along the plume axis were found to greatly decrease in the offshore zone of the

Fig. i. Site Map
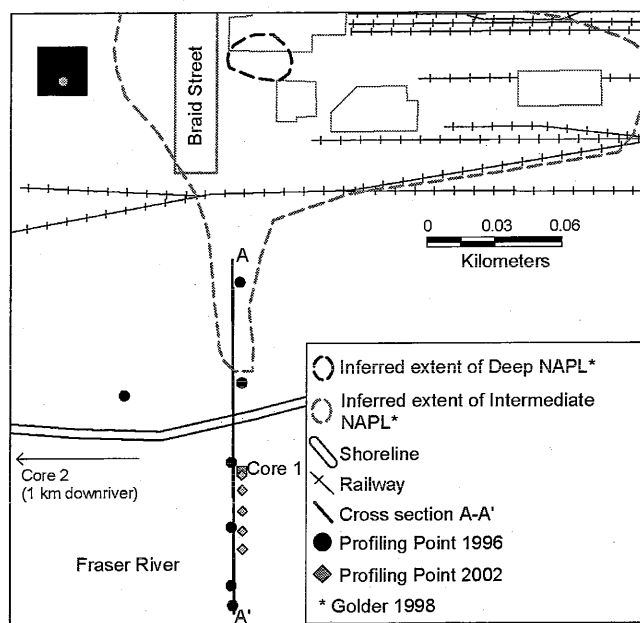
plume (Fig. ii). In addition, the ratio of naphthalene concentrations to other PAHs sampled was found to decrease in the offshore zone.
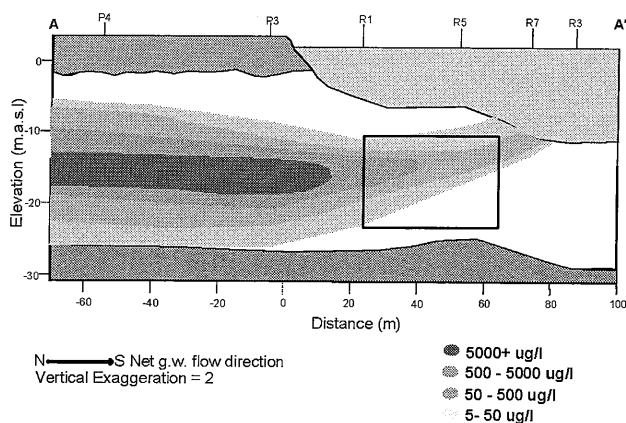
Reactive transport modeling (Anthony 1998) suggested that significant mass loss must occur in the offshore region of the plume to explain the reduced contaminant concentrations found in this zone. Increasing concentrations of methane and dissolved iron along the plume axis suggested that degradation by methanogenesis and iron reduction might explain this loss (Anthony 1998). The hypothesis that biodegradation of the plume in the offshore region takes place by a combination of methanogenesis and iron reduction was confirmed by a radiolabeled ^{14}C-naphthalene and bromide tracer experiment conducted in 2000 (Bianchin 2001).

As part of the site management plan proposed for the site, a pump and treat program to contain the DNAPL source has been in operation since 1997 (Golder, 1999). In 1999, the original pumping well was replaced by another well as groundwater modeling at the site showed that better capture of the source could be obtained with a well in this location (Golder, 1999).

Methods

An environmental review by the Fraser River Estuary Management Program (FREMP) and approval for temporary work from the Fraser River Port Authority (FRPA) were obtained prior to commencement of offshore profiling work. As with all previous groundwater sampling efforts in the offshore region of the plume (Anthony, 1998, Bianchin, 2001), the Waterloo Drive Point Profiler (Pitken, 1994) was used to obtain groundwater samples representative of discrete depth intervals over several vertical profiles. The profiling system was deployed from a 22 metre seiner (salmon fishing boat) with 5 point anchoring capability. The profiler was advanced using a

Fig. ii. Results of profiling in 1996 (Anthony, 1998). The rectangular outline represents the region sampled in 2002 (see Fig. iii.).

fluctuations.

Samples to be analyzed for dissolved gases and PAHs were collected in an anoxic environment using a sampling manifold and were subsequently stored on ice to limit post-sampling microbial alteration. Samples were collected in 40 ml glass screw top bottles with a Teflon lined septa. Samples to be analyzed for PAHs were preserved with 0.4 ml of sodium azide.

Samples to be analyzed for cations were filtered using a 0.45 um polysuppor filter and collected in 60 ml polyethylene bottles. Samples were acidified with nitric acid to pH 2 and kept on ice until analysis. Alkalinity was determined onsite by titration.

Analysis for the polycyclic aromatic components of creosote was performed at the University of Waterloo Organic Geochemistry Laboratory. Samples were solvent extracted, and a gas chromatograph equipped with a flame ionization detector was used to determine concentrations.

Analysis for dissolved methane and carbon dioxide was performed using a gas chromatograph (GC-8A Shimadzu). 4 ml of Argon, the carrier gas of the GC, was added to each sample while 4 ml of sample water was simultaneously removed from the vial to maintain atmospheric pressure within the vial. The vial was then shaken at room temperature for 5 minutes to allow the liquid and gas phases within the vial to equilibrate. 0.4 ml of the gas phase within the vial was removed and injected into the GC.

Analysis for cations was performed using inductively coupled plasma - optical emission spectroscopy (ICP-OES EPA Method 6010B) at ALS Environmental, Vancouver, British Columbia.

The Waterloo Drive Point Piston Sampler (WDPPS) (Starr and Ingleton, 1992) was used to obtain soil core samples both inside the plume and at a point downstream from the plume considered to be representative of similar uncontaminated soils. The WDPPS is a direct push instrument well suited to taking shallow soil core samples in sand, silt and soft clays.

In this sampling investigation, the WDPPS system was driven by the same pneumatic hammer suspended from the ship's 25 ton boom as was used in the groundwater profiling. Using the WDPPS on the river presented challenges not faced on land. Once a core had been collected and the system was removed from the soil at the river bottom, care was taken to minimize the time the core was submerged in the river. Sample loss would have occurred if the sample was left submerged in free water.

Organic Carbon was analyzed on soil samples collected from the two cores. Core 1 was collected along the plume axis from a depth of 7.8 to 8.8 metres below the bottom of the river bed (Fig. i.). Core 2 was collected downstream, outside of the plume at a comparable distance offshore from a depth 5.3 to 6.1 metres below the river bed. Sediments collected in both cores consisted of coarse to medium sand.

Representative samples for organic and inorganic carbon

pneumatic hammer suspended from the seiner's boom.

The coordinates of groundwater sampling points were determined by triangulation, using measurements of distances to three nearby dolphins which are normally used for anchoring log booms. Accurate depths of groundwater sampling locations were determined using a water gauge to record tidal fluctuations of water levels during the course of sampling and relating water level fluctuations to the initial depth of water when profiling began.

The profiling system consists of a profiling tip equipped with screened ports connected to a common reservoir, telfon-lined tubing connecting the common reservoir to the sampling system on the deck of the seiner and drill rods used to advance the profiler with a pneumatic hammer. The sampling system on deck consists of a sampling manifold, peristaltic pump and flow-through cell. The sampling manifold consists of two sampling heads each of which accommodate 40 ml glass septa vials. The flow-through cell is a glass cell equipped with pH, temperature and conductivity probes.

To collect a sample, the profiler was first driven to the desired sampling depth. Next, a minimum of three pore volumes was purged from the system. Purging continued until parameters monitored in the flow-through cell had stabilized.

Initial profiling of the plume was performed from June 9th to 13th, 2002. Five profiling locations from approximately 25 to 60 metres offshore were sampled along the axis of the plume. Between 5 and 11 discrete interval groundwater samples were taken at each profiling location.

This profiling took place during a period of high water levels in the Fraser River and correspondingly lower average aquifer gradients directed towards the river. Based on hourly river elevation data recorded at the nearest hydrographic station to the site, Annacis Island, the average river water level over the period of sampling was 1.28 metres above mean sea level (amsl). During sampling, the river water level recorded at Annacis Island varied by up to 1.81 metres in a single day due to tidal

analysis were obtained by homogenizing subsamples collected at several depth intervals along the cores. Samples were dried overnight at between 50 and 60 °C. Once samples were dry, an automatic grinder was used to grind the samples into a fine powder. Samples were analyzed for inorganic carbon using a CM5130 UIC inc. coulometer. Samples were analyzed for organic carbon using a Carlo Erba NA-1500 Analyzer (Verardo et al., 1990).

Results of Groundwater Profiling

Naphthalene concentrations observed along the cross section A-A' (Figure iii) are significantly attenuated from concentrations observed in 1996. Because this sampling round included detailed data collection in a region over which concentrations were previously interpolated rather than directly sampled, the amount of attenuation over the entire flow path cannot be constrained. Instead, concentrations sampled in this round can be directly compared to concentrations sampled previously at specific locations. By this method, concentrations at sampling point R-5 have decreased from 166 µg/l to 5 µg/l between 1996 and 2002.

Although naphthalene was previously identified as the dominant contaminant in the offshore region of the plume, two contaminants that were not within the suite of volatile aromatic compounds analyzed, indane and benzothiophene, were the most prominent components observed in this sampling round. They have not previously been identified in the onshore or offshore regions of the plume. Although the concentrations were not determined, they were found to have the two most prominent peaks on the GC chromatogram.

Previous sampling in 1996 showed iron and methane concentrations that increase with distance along the flow path of the plume. In this round of sampling, iron concentrations were observed to remain relatively constant over the flow path while methane concentrations were observed to increase. The concentrations of methane and iron sampled are sufficient to account for the mass loss observed between 1996 and this sampling round, although degradation of net organic matter could also be responsible.

pH values within the plume range from 6.07 to 7.66. Generally, lower pH values were observed in the northern, or upgradient, region of the area profiled. At all points profiled, the greatest pH values were observed in the deepest intervals sampled. pH values have not previously been profiled in the offshore region of the plume. Anthony (1998) stated that the pH of the inland groundwater was found to be 6.2 +/-.2.

Decreased alkalinity along the flow path of the plume was suggested by data collected in 1996 (Anthony, 1998). Alkalinity values observed in this round of sampling appear to confirm this hypothesis, as a region of low alkalinity was observed in the moderate depth intervals at each profiling point. Alkalinity values sampled range from 3.12 to 13.0 meq/l.
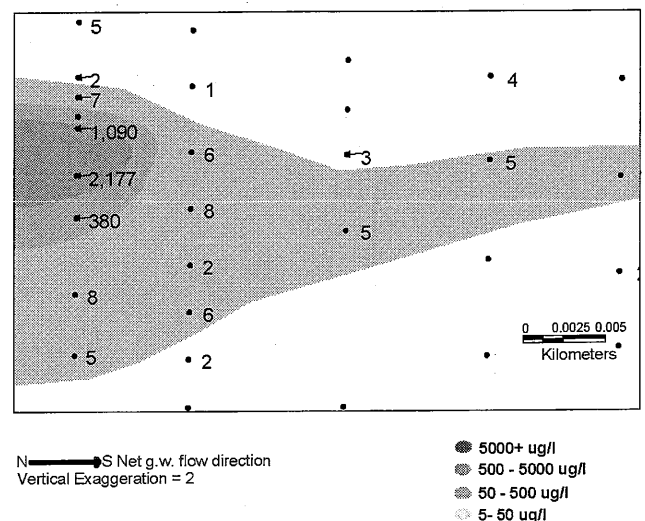
Results of Organic Carbon Analysis

Organic carbon contents range from 0.09 to 0.52 % in core 1 and from 0.11 to 0.14 % in core 2. These results can be related to Kd values using an empirically-derived correlation equation appropriate to non-polar organics such as naphthalene (Schwarzenbach and Westall, 1981). Kd values for core 1 calculated with this equation range from 0.91 to 4.3. Kd values for core 2 range from 0.93 to 1.17 and average 1.0.

Organic carbon contents analyzed by Anthony (1998) in two cores collected north of the site had an average Kd of 1.17. The agreement between organic carbon contents analyzed in cores collected onshore at the site (Anthony 1998) and in this research implies that mass loss in the region of the plume that is beneath the river is likely not due to increased sorption.

Fig. iii Results of profiling 2002.

N ━━━━●S Net g.w. flow direction
Vertical Exaggeration = 2

- ● 5000+ ug/l
- ◉ 500 - 5000 ug/l
- ◍ 50 - 500 ug/l
- ◌ 5- 50 ug/l

Groundwater Flow Model

A groundwater flow model was constructed to obtain a better understanding of the hydrogeology of the site and of the effects of seasonal changes in river stage elevation on fate and transport of the contaminant plume. The model used to simulate the hydrogeological conditions at the site is MODFLOW (McDonald and Harbaugh, 1988). MODPATH (Pollock, 1989) was used to determine the path of a conservative tracer in the aquifer.

The model was constructed using average monthly water levels and aquifer gradients to simulate seasonal variations in river stage elevations and recharge to the aquifer. The

model represents the time period from January 1st, 1999 to Dec 31st, 2000. The daily tidal variations in river stage were not explicitly simulated over the model's two-year time horizon.

The domain of the model is 800 metres east-west by 780 metres north-south. The grid is most refined in the regions of the two source containment wells and at the tracer injection location.

Based on the conceptual model (Golder 1997); the aquifer was represented by several model layers with a cumulative thickness of 27m. Flat layer horizons were used since the topographic variations at the site are generally less than 2 metres. Eleven model layers were used to resolve the vertically upward flow as groundwater discharges to the river.

Boundary conditions in the model were derived from monthly averages of water level data recorded at the hydrographic station located at Annacis Island and from continuous water level monitoring data collected at the site (Golder, 1997). Bathymetric data collected in the south-eastern portion of the model domain was used to delineate the bottom of the river boundary (Bianchin 2001). An average of 7 surveys of bathymetric data collected between September 1998 and August 1999 was used.

The northern and southern boundaries of the model were represented with time varying specified head boundaries. The eastern and western boundaries of the model were, for the most part, represented with no flow boundaries since the dominant direction of flow in the aquifer is north-south. Because flow develops an east-west component beneath the river in response to the downstream river gradient, the constant head boundaries representing the river at the southern edge of the model were wrapped around the east and west edges for approximately 50 metres beyond the natural bounds of the river. Beneath the specified head boundary representing the river at the southern edge of the model, a no flow boundary was used as suggested by migration of the contaminant plume. The lower boundary of the aquifer was represented with a no flow boundary since the Pre-Fraser River sands unit which underlies the aquifer is considered to be of very low permeability (Golder 1997).

Recharge to the model was estimated as a percentage of average monthly precipitation. A previous site water balance suggests that a greater percentage of precipitation infiltrates from April to September of each year than during the fall and winter (Golder, 1997); therefore recharge in the model was set to a greater percentage of precipitation from April to September than from October to March.

Because of the unavailability of spatially distributed continuous water level monitoring data and the small area of transport covered by the tracer plume, a single averaged value of transmissivity and storage were used to represent the model domain. The transmissivity and storage values used in the model were derived from pumping tests performed at wells source containment wells (Golder 1997).

Average monthly pumping rates for the wells used to capture the plume source were input into the model.

The model was calibrated by correlating particle tracking results with the transport of the bromide tracer released in February 2000 (Bianchin 2001). This plume was monitored offshore for 249 days after release. These criteria were chosen because, unlike instantaneous water-level measurements, the cumulative transport and distribution represents average hydraulic gradients and, therefore, the effects of tidal fluctuations would be averaged out.

Groundwater Model Results

Groundwater modelling shows that flow in the aquifer oscillates on a seasonal time scale, due to changing river stage elevations. Changes in river stage elevation throughout the year cause the influence of the source containment well on the contaminant plume to be highly seasonally dependent. During periods of high river stage elevation, the well influence extends to the region of the aquifer beneath the Fraser River and causes migration of contaminants to slow or become directed away from the river.

Figure iv compares groundwater flow directions along the plume axis in June with groundwater flow directions in February. The relative sizes of arrows in this figure reflect the magnitudes of groundwater velocities over the cross section. In February, flows in the region of the aquifer beneath the river are directed towards the river and velocities are much greater than in June. In June, groundwater velocities beneath the river are directed away from the river or very low.

Fig. iv. Model Results: Typical seasonal changes in groundwater flow direction under the influence of source containment wells. Top: Groundwater flow directions in June. Bottom: Groundwater flow directions in February.
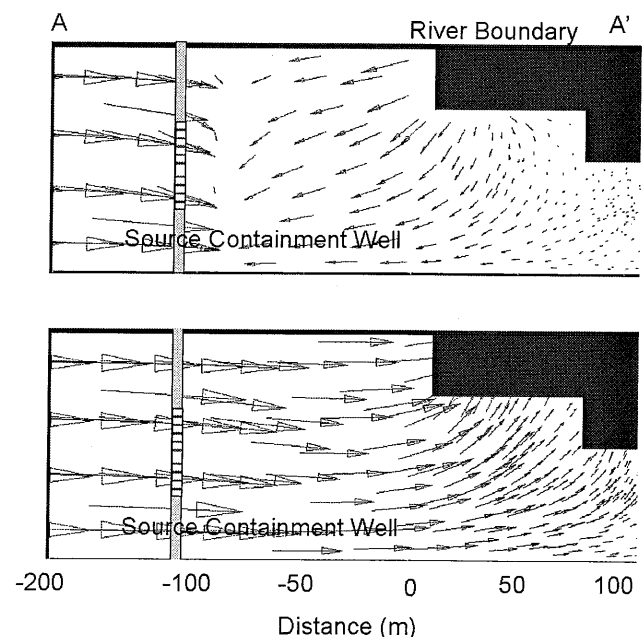
Vertical Exaggeration = 4

Particle tracking shows that particle movement is directed downwards and away from the river during periods of high river stage. Particles were observed to migrate up to 3 metres away from the discharge point of the plume during the freshet in the offshore region of the plume.

Discussion of Results

The effects of natural attenuation, mass stored in the aquifer matrix due to sorption, and dynamic site hydrogeology must all be accounted for in order to explain the contaminant fate and transport of the creosote-derived plume at this site. Processes of natural attenuation such as degradation and dispersion may explain the reduced contaminant concentrations observed at the site, while continuing desorption of contaminant mass from aquifer matrix may offset this loss. The complex site hydrogeology caused by variations in aquifer gradients on seasonal and diurnal time scales is expected to increase the effects of natural attenuation processes at the site.

The results of groundwater flow modelling show that a low flow zone exists in the aquifer due to the influence of the capture well at the site and seasonally varying aquifer gradients. Due to movement of the well influence with the spring freshet, contaminants at the fringes of the capture zone are expected to be transported towards the river at times of low river stage and away from the river at high stage. This variance in the capture zone will extend the residence times of contaminants in the aquifer, increasing the opportunity for attenuation of contaminants before discharge to the river.

Desorption of contaminant mass stored in the aquifer matrix is expected to buffer reductions in aqueous concentrations due to natural attenuation as contaminant mass desorbs into the aqueous phase. A direct measurement of sorption in the aquifer via extraction of sorbed naphthalene from sediment collected from core 1 yielded K_d values between .03 and .96, with an average value of .41 (Bouchard, 2003). Because of the consistency between organic carbon content in cores collected onshore at the site (Anthony 1998) and in this research, sorption within the aquifer is considered to be relatively homogeneous and K_d values obtained from this core are considered to be representative of aquifer properties onshore as well as offshore.

By comparing data collected at R-5 in 1996 with data collected in this study, it can be seen that aqueous concentrations of naphthalene have decreased by up to 161 µg/l. Assuming equilibrium sorption and using the average K_d of .41, mass loss from the sorbed phase can also be estimated. Taking into account mass loss from both the sorbed and aqueous phases, concentrations at this point have been reduced by up to 623 µg/l.

Naphthalene degradation rates derived from a zero order fit to tracer experiment data (Bianchin, 2001) and laboratory microcosms (Lesser, 2000) can be used to estimate the maximum reduction in concentrations that could be expected to occur from biodegradation alone (Bekins, 1998). The observed reduction in concentrations is within the range of degradation rates predicted by a zero order fit to these data sets.

Dispersion may also be a factor contributing to dilution, but because of the low dispersivity determined from an onshore tracer test (Bianchin, 2001), dispersion alone is not expected to significantly contribute to the decrease in concentrations observed. Tidally driven mass loss is expected to significantly contribute to decreased concentrations of naphthalene in the portion of the aquifer underneath the river (Yim and Mohsen, 1992). Because of the low flow zone caused by the source containment well at the site, this parameter could not be determined from an offshore tracer test (Bianchin, 2001) and is not well quantified.

The increased prominence of indane and benzothiophene in the samples collected in this research, despite source containment, provides further evidence that mass loss of naphthalene is occurring preferentially by degradation. Because of the resistance of benzothiophene to degradation, concentrations of this compound in further sampling may be useful to estimate mass loss due to tidal forcing.

Summary

Contaminant concentrations sampled along the plume profile in June 2002 are significantly reduced from previously sampled concentrations. From current data, it is not possible to resolve the magnitude of this reduction that is purely due to biodegradation of contaminants.

To resolve this issue, further field sampling in the on- and off-shore regions of the plume is planned. Further sampling will also focus on identifying products of anaerobic naphthalene degradation and obtaining concentrations of benzothiophene and indane in groundwater samples.

Acknowledgements

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