ORGANIC CONTAMINANT TRANSPORT IN GROUND WATER, SURFACE WATER, AND SURFACE WATER SEDIMENTS. YEAR 1 PROGRESS REPORT.

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Research Project Technical Progress Report (USGS G-879, Project No. 03)

Prepared for: U.S. Department of the Interior U.S. Geological Survey

The research on which this report is based was financed in part by the United States Department of the Interior as authorized by the Water Research and Development Act of 1978 (P.L. 95-467).

Contents of this publication do not necessarily reflect the views and policies of the United States Department of the Interior, nor does mention of trade names or commercial products constitute their endorsement by the U.S. Government.

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ABSTRACT

Questions concerning the transport and fate of contaminants in the environment have typically been limited to single compounds. However, the greatest danger to water resources is most often posed by sites releasing complex mixtures containing many, even hundreds, of hazardous chemicals. Hazardous waste disposal facilities, chemical plants, oil refineries, etc., typically produce complex waste streams which may contact ground water or surface waters. This progress report presents results of a study on movement through ground and surface water of a complex mixture of organic compounds emanating from a wood-treatment facility located adjacent to the Laramie River in Laramie, Wyoming. This study, initiated in FY 1983, is continuing with USDI funding for a second year, through summer 1985.

The objectives of the study are to (1) initiate a literature review on contaminant transport models and develop a conceptual model of contaminant transport at the Laramie River site; (2) conduct laboratory (soil column) and field experiments to investigate oil/ water emulsion migration through porous media; and (3) begin a field study analyzing contaminant fate in sediments downstream from the wood-treating facility. A fourth objective, to supply the Wyoming Department of Environmental Quality (DEQ) with a summarization of contaminant transport models, was deferred to FY 84 based on requests from DEQ officials to incorporate the Year 2 study and submit a single, final report.

Currently, mathematical models are not readily available to

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simulate the transport and fate of complex mixtures of organic compounds. This is especially true when these contaminants are in high concentrations producing a multi-phase solution. But a conceptual model summarizing laboratory and site observations shows: (1) subsurface transport of contaminants in oil, oil/water emulsions, and groundwater; and (2) riverine transport of contaminants in water, sorbed to sediments, and as oil globules.

Experiments with glass columns packed with a uniform material (1 mm glass beads, sand, or gravel) indicates that in even the most homogenous, isotropic matrix, oil/water mixtures cannot be expected to flow in a uniform manner. Creosote sludges and oils break up into globules and stringers with no uniform front of contamination. Data from a field study using mini-piezometers in the Laramie River adjacent to the site confirm this observation.

Chemical analyses of sediments and fish, as well as field observations indicating oily droplets moving along the river bed, clearly establish contamination of the Laramie River resulting from offsite migration of creosote from the wood-treating facility.

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INTRODUCTION

Contaminant transport modeling in ground water, surface water, soils, and sediments has been restricted primarily to single compound interactions with the environment. However, the greatest danger to water resources is more often posed by sites releasing complex mixtures of chemicals. The problem that arises is that interactions among chemicals in the mixture can affect the environmental transport and fate of any single chemical. This is especially true when organic contaminants are present in high concentrations. A simple example would be that the soil adsorption/desorption rate constants for a hydrophobic compound derived from an experiment using water as a solvent do not apply to a system which also contains high concentrations of an organic solvent such as phenol, toluene or benzene.

Industrial plants, oil refineries, drilling sites and wood-treating plants produce and may release such complex mixtures of organic compounds. At sites such as these, as well as at hazardous waste disposal sites and chemcial spill sites, hazardous chemicals often can be released into soils, ground water, surface waters, and surface water sediments as an emulsion or hydrocarbon fluid. Once released in this way, contaminants may be transported through these systems in three different fluid phases: oil, oil/water emulsions, and water. The ability of state and federal regulatory agencies to detect, monitor, and predict the movement and fate of complex contaminant mixtures, as well as their ability to implement remedial action, has been hampered by

a lack of knowledge concerning multi-phase transport problems such as these.

This report details the work to date and currently in progress at the Union Pacific's tie treatment plant (TTP) In Laramie, Wyoming (Fig. 1). This plant had been in operation for almost one hundred years prior to its closure in the spring of 1983. Severe contamination from creosote wastes including pentachlorophenol has occurred over approximately 80 acres of the site. Groundwater contamination ranges from trace level polynuclear aromatic hydrocarbons to levels exceeding solubility limits, indicating presence of an oil/water emulsion (CH2M-Hill 1984). In fact, some on-site recovery wells produce creosote in sufficient quantity and quality to warrant a product recovery program (CH2M-Hill 1984). In addition to on-site groundwater contamination, field investigations have shown that the Laramie River adjacent to the site has oily seeps which are contaminating both the water and the river sediments downstream from the site.

Further studies have been funded through the Wyoming Water Research Center (WWRC)/U.S. Department of Interior program for fiscal year 1984 (FY84). This continuation in funding will be used to continue and expand portions of the study presented below. Completion of some of the original FY83 objectives of this study have been postponed to the FY84 period based on recommendations of WWRC personnel and the Wyoming Department of Environmental Quality (DEQ).



Figure 1. Location of Union Pacific Tie Treatment Plant near Laramie, Wyoming.

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OBJECTIVES

Briefly the four original objectives in the FY83 study were as follows.

- <u>Transport model evaluation</u>: Literature review and evaluation of contaminant transport and fate models dealing with complex organic mixtures.
- <u>Contaminant transport in ground water</u>: Laboratory soil column experiments to investigate movement of contaminated oils and oil/water emulsions in ground water.
- 3) <u>Contaminant transport in surface water</u>: Field sampling and analysis of Laramie River sediments downstream from the Union Pacific tie treatment plant.
- 4) <u>Report to regulatory agencies</u>: A summary of existing knowledge and experience providing practical guidance to regulatory agencies and industry for assessing transport of complex contaminant mixtures in ground water and surface water.

Through the course of the FY83 study these objectives were modified slightly and expanded to add additional information about contaminant transport and fate phenomena at our case-study field site. These modifications were then incorporated into the FY84 objectives. The principal additions in the FY84 objectives are, briefly, to expand the ground-water contaminant transport studies using mini-piezometers and seepage meters on and adjacent to the field site, and to expand the surface water contaminant transport and fate studies with fish and additional sediment collection and analysis from the river downstream from the site.

RELATED ACTIVITIES

Due to on-going litigation involving the State of Wyoming, the U.S. Environmental Protection Agency (EPA), and the Union Pacific Railroad, we have been asked by the State of Wyoming to review data related to the site as well as report any significant findings from our own studies to the State. In this capacity we have attended private and public meetings as well as reviewed technical reports submitted to the Wyoming DEQ by Union Pacific's consultant CH2M-Hill.

Following the initial site investigations a contaminant assessment was submitted to the State by the Union Pacific (CH2M-Hill, 1983). Evaluation of this draft document led to the conclusions that the tie treatment plant was severely contaminated, having contaminants in the groundwater above their solubility in aqueous solutions (Bergman and Crossey, 1983), and that the evaporation ponds probably leak (Huntoon, 1983). More recent investigations have shown this to be the case (CH2M-Hill, 1984). A second draft report (CH2M-Hill, 1984) for which comments were requested and submitted (Crossey, 1984) has not as yet been released in final form.

In addition to site investigations the EPA requested comments on the evaporation pond closure plan (CH2M-Hill, 1983 and 1984) which is regulated under the Resource Conservation and Recovery Act (RCRA). Comments presented at a public hearing in February, 1984 pointed out the danger of on-site storage of solidified organic wastes due to the possible infiltration of ground water as well as the questionable interaction of clay liners and organic fluids such as those found

at the site.

Currently we are reviewing the Phase III, Remedial Action Feasibility Study, and a risk assessment of the Union Pacific tie treatment plant for the Wyoming DEQ.

TASK 1: TRANSPORT MODELS

Although the development and use of contaminant transport models for single chemicals in ground and surface waters has been increasing markedly in the last 10 to 15 years, there have been few, if any, attempts to modify these models for complex contaminant mixtures and contaminated multi-phase fluids. Thus, our aim in this task was to determine whether recent work by contaminant transport modelers included such modifications and to evaluate the utility of existing single-contaminant transport models for complex mixture and multiphase fluid transport problems.

Procedures

Hand searches and computerized bibliographic database searches were conducted to identify recent published literature on the application of contaminant transport modeling to complex contaminant mixtures and contaminated multi-phase fluids. Additionally, key researchers and model users in North America were contacted to determine whether such models were currently under development in the contaminant modeling field.

Results and Discussion

Literature searches and contacts with knowledgable researchers in North America confirmed that no working transport models for complex contaminant mixtures apparently exist. With regard to this question, all of the relevant literature discovered dealt only with the conceptual framework necessary to approach such a modeling problem (e.g.,

Bergman and Meyer, 1982). All of the potentially applicable models or literature discovered can be classified into three categories: (1) single-contaminant ground-water transport models, (2) singlecontaminant surface water transport models, and (3) multi-phase oil/ water transport models that are usually applied to oil reservoir dynamics problems.

Ground-water modeling was initially developed as a resource monitoring tool. Models were used to calculate flow, specific yield, drawdown rates, etc., to enable agencies to manage ground water for domestic and agricultural uses. Models such as these are now at a refined state and most situations can be adequately modeled (Freeze and Cherry, 1979). Ground-water contaminant transport modeling is a more recent development and has built upon the previously established resource modeling techniques. There are three types of models available, advection, advection-dispersion, and geochemical. These types of models allow one to theoretically incorporate the effects of the aquifer matrix as well as chemical interactions on the concentrations of contaminants through distance and/or time. However, in practice only the simplest, the advection models, are routinely used (Anderson, 1981). These models are limited by the simplifying assumptions of no dispersion or chemical interaction between the contaminant and the aquifer matrix. However, these models can be used to estimate the "worst possible case" scenario for contaminant transport.

Modeling complex organic contaminants in ground-water systems requires not only incorporation of the aquifer interactions mentioned

above but, in addition, the contaminant chemistry itself, since it can greatly affect migration rates. Heavily contaminated sites in which hydrocarbon fluids and aqueous fluids are present require multi-phase flow modeling (Marle, 1981). In general, the theoretical/mathematical development of contaminant transport models is ahead of the knowledge about the controlling processes from the field.

Surface water contaminant modeling presents many of the same short-comings as ground-water modeling. The fate and transport of complex organic mixtures is not nearly as well researched as dissolved single compound contaminant models. The complex nature of organic contaminants does not necessarily preclude modeling but, obviously, field studies and observations are required to detect and define the important controlling processes on which the models are to be based.

TASK 2: CONTAMINANT TRANSPORT IN GROUND WATER

The movement of organic contaminant mixtures in ground water and surface water is often difficult to predict due to the numerous interactions among compounds within the mixture. Often physical/ chemical constants derived for single compounds may not reliably predict the chemical behavior of a compound in a complex mixture (Banerjee et al., 1984). With this in mind, the studies presented below were designed to treat a complex mixture (creosote) as a whole and to define properties that would describe, in a general way, the behavior of this mixture in ground water.

Procedures

Glass soil columns, 60 cm long x 15 cm diameter, with teflon ends were constructed with four glass piezometers extending into the column packing at various depths to sample pore fluids. Three column packings are currently being tested, 1 mm glass beads, site soil, and washed sand. After column packing, creosote is introduced through an opening in the cap. Water flow, either top to bottom or bottom to top, is maintained with teflon tubing fitted into the center of each cap. Pore fluids and eluents from the columns are analyzed using total fluorescence (described below) and HPLC (described in Task 3).

In addition to this lab column study, which will be continued through FY84, a field study was initiated to investigate the flow of creosote and water within the alluvial aquifer at the TTP. For

this purpose mini-piezometers were constructed following the design of Lee and Cherry (1978). Teflon tubing (3/8" O.D., 1/4" I.D.) was perforated (1/16" holes) and screened on one end and inserted into the river sediments. In this way a water sample could be taken below the bottom of the riverbed representing subsurface water. Depending on the hydraulic gradient, this water would be either coming from the site into the river, moving from the river to the site or moving downstream with the interflow. Thirty-nine mini-piezometers were placed in the Laramie River adjacent to the west side of the TTP. The depth to which the piezometers penetrated the river bottom ranged from one to three feet. Typically, the piezometers were driven through the fine sands making up the river bottom until the Morrison formation was reached. This formation underlies the Laramie River in this area and subcrops just east of the river on the TTP site (Charles Feast, CH2M-Hill, Denver, Colorado pers. comm.). The location of each of the mini-piezometer samples is designated on Figure 2. Samples were collected on February 23-25, 1984, during a period when the river was ice-covered.

Samples from the subsurface were obtained by applying a vacuum to the end of the piezometer, and the samples were drawn into 100 ml glass jars. Jars were capped with teflon-lined lids and taken to the lab for analysis. At the lab the samples were extracted into freon (30 mls sample to 5 mls freon). Total fluorescence of the extracts was then measured (excitation wavelength = 360 nm, emission wavelength = 410 nm). The total fluorescence is proportional to the amount of aromatic organic chemicals in the sample.

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Figure 2. Location of mini-piezometer sampling points in the Laramie River adjacent to the Union Pacific Tie Treatment Plant near Laramie, Wyoming.

Results and Discussion

Only the preliminary observation from the column studies will be presented in this report. Delays in column manufacture and the extremely slow movement of creosote in water-saturated columns has slowed progress in this phase of the study. In the first experiment, no flow through the column was used. Once packed with glass beads, washed sand, or site soil, the columns were filled (bottom to top) with water. Creosote was then introduced to the top of this static column. Initially no movement could be detected. The creosote appeared to "float" on top of the column of water. This creosote sample had a density of 1.01 gm/cm³ but remained in this unstable equilibrium. This phenomenon has been described previously in multi-phase flow situations (Marle, 1981). However, once perturbed by mixing the top 2 cm of packing, the creosote began to flow in small stringers and globules for several centimeters. This has also been observed in the field at the TTP site (CH2M-Hill, 1984) and in the mini-piezometer study presented below. The flow of these stringers and globules virtually ceased following the initial movements (over a two month period no movement was recorded). Other studies to be completed in FY84, will provide a better understanding of the processes controlling the flow of creosote through these types of matrices.

As can be seen from the data for total fluorescence of piezometer water extracts (Fig. 3), there is an obvious seep at sample location 10. This sampling point lies east and slightly north of well 23 (Fig. 2). Sample 11 which is mid-river directly east of sample 10 also shows high fluorescence. Sample 10 was, in fact, black, oily



SAMPLE NUMBER

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fluid which had a strong creosote odor. Other samples had fluorescence values that indicate that they may be near other seeps (samples 3, 16, 32, 36, for example). Total fluorescence of sample 36 would seem to indicate that there may be a seep at the northern end of the old river oxbow near well D-32.

These data confirm that organic contaminants are reaching the Laramie River as oily seeps. The fluorescence data indicate that there may have been as many as four or five seeps present during the sampling period in February 1984. More seeps may have been present, but they could have been missed due to the 10-meter spacing of the mini-piezometers. For example, sample 10 was a thick, oily fluid but samples 9 and 12, which were collected from within 10 meters of sample 10, show low to moderate fluorescence. For this to occur, the seeps were probably small, on the order of several inches to several feet, and the sampling grid of 10 to 100 meters may have missed a number of contaminant seeps.

TASK 3: CONTAMINANT TRANSPORT IN SURFACE WATER

A full understanding of contaminant transport in riverine systems requires knowledge of contaminant transport in water, in suspended particulates, and in the sediments. Compounds having very low water solubility, such as the compounds of principal concern at the tie treatment plant (TTP) site, will be found mainly sorbed to the river bed sediments. For that reason, this study concentrated on contamination of river bed sediments downstream from the TTP site as an indicator of contaminant release from the site. Analyses of fish taken from the river adjacent to and downstream from the site was added to the study as an additional indicator of off-site movement of contaminants.

Typically aquatic sediments have been thought of as a sink rather than source of contamination. Recently research has been directed toward identifying the processes that control the transport, fate, and bioavailability of contaminants bound to the sediments (Baker, 1980). The importance of sediment as a transport mechanism for organic compounds has been expressed with regard to environmental fate modeling (Urchin and Weber, 1980; Burns, 1982). Field studies as well have stressed the importance of sediment binding and transport of organic compounds resulting in extensive low-level contamination of aquatic systems (Black, 1982; Pierce et al., 1980). Data presented below describes the methods developed to sample, extract, and analyze sediments downstream from the TTP to evaluate off-site contaminant migration.

Hydrophobic compounds such as those found in creosote can be bioaccumulated. To assess the contaminant concentrations in fish living near the TTP we sampled a four kilometer stretch immediately downstream from the site.

Procedures

In the fall of 1983, 16 sediment samples were collected in slow backwater areas over a distance of 16 km downstream from the TTP. Surface sediment was scraped and collected into a two liter glass beaker. This sediment and river water were stirred vigorously to suspend the organic fines, clays, and particulate organic matter. This solution was then decanted into another two liter beaker, leaving behind the coarse sands and gravel which were discarded. The second beaker containing fine sediments was allowed to settle for five to ten minutes, and the water was then decanted and discarded. Following this procedure several times at a given site allowed us to collect approximately 500 mls of "organic rich" sediments. Sticks, leaves, and coarse particulate organic matter were removed and discarded. Samples were then frozen until analyses were performed. This sampling scheme was developed because several authors have stressed the importance of the organic fraction in the transport of contaminants (Black, 1982; Lopez-Avila and Hites, 1980; Pierce et al., 1980).

A batch extraction method was developed to rapidly process samples for High Performance Liquid Chromatography (HPLC). The extraction solvent consisted of acetonitrile, freon, and acetic acid (1000, 10, 2 mls, respectively). One gram of air-dried sediment was

placed in a 250 ml Erlynmeyer flask to which 25 mls extraction solvent was added. The flasks were capped with teflon-lined screwcaps and placed on a shaker table (150 rpm) for 24 hrs. Following extraction, samples were allowed to settle and a 2 ml aliquot of the extraction solvent was removed, filtered through a 0.2 µm teflon filter, and injected directly into the HPLC without further processing.

Chromatographic analyses were performed on a Waters liquid chromatograph using a one hour linear gradient, 100% HPLC grade water containing 0.3 mls/l acetic acid to 100% acetonitrile, at a flow of 2 mls/min. Absorbance at 254 nm and 280 nm was used for detection. In addition to retention time, the ratio of absorbance at 254 nm to 280 nm (254 nm/280 nm) was used for compound identification. Retention times and absorbance ratios were compared to a mixture of purified standards as well as an extract of on-site contaminated soil.

Fish were sampled from a boat using electroshocking equipment. Four composite samples were analyzed. After sampling, fish were eviscerated, decapitated, and individually wrapped in aluminum foil and frozen. Analyses were performed at the U.S. Fish and Wildlife Service, Columbia National Fisheries Research Laboratory. For determinations of polynuclear aromatic hydrocarbons (PNAs) and phenols, each sample was subjected to column extraction with methylene chloride of a dried powder mixture of the fish tissue and sodium sulfate, followed by two fractionation steps using gel permeation chromatography. Each sample was spiked prior to the extraction at 100 ng/g with a series of internal reference compounds, six deuterated PNAs and 4-iodophenol. Additionally

one sample was processed in triplicate, with two of the triplicate samples spiked with the internal standards and the third spiked with both the internal standards and a mixture of eleven representative PNAs and eleven representative phenols (the EPA Priority Pollutant phenols). Four control samples (no fish tissue) were also included in this set of samples for purposes of providing recovery and procedural background data. The analyses of the latter four samples were not completed at the time of this report.

Determinations of the target compounds were by high resolution gas chromatography/low resolution mass spectrometry using a 30 meter DB-5 capillary column and a Finnigan 4023 mass spectrometer. Individual confirmations of component identity were made for each compound in each sample.

Results and Discussion

The methods developed during the course of this study have proven to be satisfactory and only minor modifications are planned for FY84 sampling, extraction and analysis.

The complexity of the organic contaminants at the TTP precludes identification of all the compounds adsorbed to the sediments. However, based on identical retention times and 254 nm/280 nm absorbance ratios, phenanthrene, anthracene, and chrycene were tentatively identified on the HPLC chromatograms for a creosote extract and extracts of TTP site soil and Laramie River sediments 3.5 km downstream from the site (Fig. 4). The chromatograms also can be used to describe the general distribution of compounds ("fingerprint") in the mixture. Illustrations of this "fingerprint" are the chromatograms of on-site

Figure 4. Reverse-phase High Performance Liquid Chromatography (HPLC) gradient chromatograms for solvent extracts of (a) creosote saturated glass beads, (b) contaminated site soil, and (c) sediment sample taken 3.5 km downstream from the Tie Treatment Plant (TTP), (d) sediment sample taken .5 km upstream from TTP. UV absorbance intensity at 254 nm is shown for each sample, and an absorbance scale (AU = absorbance units) is indicated beside each chromatogram. Retention times of five major compounds are indicated above each chromatogram, as follows: $l = pentachlorophenol (C_6H_5Cl_5OH), 2 = naph$ thalene $(C_{10}H_8)$, 3 = phenanthrene $(C_{14}H_{10})$, 4 = anthracene $(C_{14}H_{10})$, and 5 = chrysene $(C_{18}H_{12})$. Peaks A, B, and C in the chromatograms are tentatively identified on the basis of retention time and 254nm/280nm absorbance ratios as phenanthrene, anthracene, and chrycene respectively. See text for details of extraction procedures and chromatographic conditions.



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contaminated soil and Laramie River sediment (Fig. 4b,c). This pattern is not seen in upstream sediment samples (Fig. 4d). A second interesting comparison is the chromatogram from an extract of 1 mm glass beads which had been contaminated with creosote from the TTP (Fig. 4a). These glass beads were used in the column experiments described above. Free oil was allowed to drain from the beads prior to extraction. The extraction ratio was similar to the sediments and soils (1 gm/20 mls). Clearly the pattern shown in Figure 4a,b,c is very similar, the differences being only quantitative and not qualitative.

Not all of the downstream samples show this contaminant fingerprint. This indicates that the downstream areas are not uniformly contaminated nor do they show decreasing contamination with distance from the TTP as seen by Black (1982). This heterogeneous contaminant distribution may be a function of the hydrologic regime at the site which causes sporadic seeps to appear and a pulse of contaminated sediments may move down the river and settle in one area. Additionally, as discussed in the preceding section, the presence of oily droplets or globules may cause this type of contaminant distribution. Seasonal or day to day changes in the flow of the river may govern where these oil droplets settle and how far they travel. If the contamination downstream is a secondary effect due to clean sediments contacting these oily droplets this may increase the variability in contaminant concentrations in downstream sediments. This phenomenon has been noted in an estuary system (Lopez-Avila and Hites, 1980).

The results presented in Table 1 indicate contamination of

fish living downstream from the TTP. Pentachlorophenol has been shown to be depurated within a few days if a fish is returned to clean water (EPA, 1980; Lech et al., 1978; Akitake and Kobayashi, 1975). Therefore, the pentachlorophenol present in the fish indicates either an exposure event within one to two days prior to sampling or a chronic low-level exposure leading to the observed body burden of pentachlorophenol in the fish. The latter case would seem the most plausible. Table 1. Summary of fish tissue analysis results from the Laramie River downstream from the Union Pacific Tie Treatment Plant, Laramie, Wyoming.

Sample	Description	Distance downstream (km)	Compound ^a Com	ncentration ^a (ppb)
1	Composite of	0	Acenaphthalene	142
	3 white suckers;		Dibenzofuran	65
	edible portion;		Fluorene	56
	608, 794 and		Naphthalene	117
	907 gm		Pentachlorophenol	147
2	Composite of	0.25-1.5	Methylnaphthalene	114
	2 white suckers;		Naphthalene	58
	whole fish; ∿ 200 gm each		C-2 Naphthalene	51
3	Composite of 3 brown trout; edible portion; 597, 793 and 1,134 gm	2	Pentachlorophenol	79
4	Composite of 2 brown trout; edible portion; 294 and 1,250 gm	3.5	Naphthalene Pentachlorophenol	73 119
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^aOnly those compounds found at concentrations greater than 50 ppb are reported.

bppb = parts per billion = ng/gm.

SUMMARY AND CONCLUSIONS

- To date it appears that no models exist that adequately describe the transport and fate of complex organic mixtures in surface water and/or groundwater systems.
- 2) Lab and field studies indicate that, for multi-phase contaminant transport in ground water, there is rarely a uniform front or plume but rather small stringers and isolated globules.
- 3) Organic contaminants can be traced several kilometers downstream from the Union Pacific Tie Treatment Plant, and there appears to be heterogeneous distribution of contaminants in downstream backwater areas.
- 4) Droplets of free oil (creosote) are probably an important component of the mass flux of contaminants from the site to the river system.
- 5) The soil columns, mini-piezometers, sediment sampling methods, and analytical methods (fluorescence and HPLC) developed in FY83 have proven to be satisfactory and will be continued in the FY84 study.
- 6) The FY84 studies will require increased sampling of sediments and mini-piezometers and the use of seepage meters to better quantify the rate of contaminant movement to the Laramie River from the Tie Treatment Plant.

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