ORGANIC CONTAMINANT TRANSPORT IN GROUND WATER, SURFACE WATER AND SURFACE WATER SEDIMENTS: A CASE STUDY OF A WOOD-TREATING PLANT

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ABSTRACT

Studies 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 contaminant mixtures. Hazardous waste disposal facilities, chemical plants, oil refineries, etc., usually produce complex waste streams which may contact ground waters or surface waters. This report presents the results of a study conducted at a wood-treating facility located adjacent to the Laramie River in Laramie, Wyoming. Organic contaminants emanating from this facility include phenolics, polyaromatic hydrocarbons (PAH's), and pentachlorophenol. Creosote, which is itself a complex mixture of organic compounds, is the main source of contamination at the site. This study was initiated in FY 1983 and continued with USDI funding for a second year through the summer of 1985.

The objectives of this study were 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. An additional aspect of this project was to supply the Wyoming Department of Environmental Quality (DEQ) with a summarization of these studies and to provide technical assistance and data review for the on-going litigation between the State of Wyoming and the owners of the wood-treating facility.

Currently, mathematical models are not readily available to effectively 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. However, a conceptual model based on laboratory studies and site observations shows: (1) subsurface transport of contaminants as hydrocarbon fluids (oil), oil/water emulsions and dissolved in the aqueous phase; and (2) riverine transport of contaminants dissolved in water, sorbed to sediments, and as oil globules.

Experiments with glass columns packed with a uniform material (2 mm glass beads) indicates that even within a very homogeneous, 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. In addition these experiments have shown that contaminants within an oil/water emulsion move at a rate almost equal to that of chloride, while a single compound (naphthlene) shows a lag in breakthrough. These preliminary results indicate that with an emulsion there is sufficient interaction among the compounds in the fluid to overcome the retardation due to adsorption of the organic compounds to the glass beads.

Field studies and chemical analyses of sediments and fish clearly establish contamination of the Laramie River resulting from off-site migration of creosote from the wood-treating facility. However, comparison of contaminant levels from sediments taken in 1983 and 1985 as well as visual field observations indicate a significant decline in the organic contaminants in the Laramie River over the past three years.

This decline can be attributed to changes in the flow regimes of the Laramie River as well a remedial actions taken by the personnel at the wood-treating facility.

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INTRODUCTION

This report details the work conducted at the Union Pacific's Tie Treatment Plant (TTP) in Laramie, Wyoming (Fig. 1). This facility had been in operation for nearly one hundred years prior to its closure in the spring of 1983. Severe contamination from creosote wastes and pentachlorophenol had occurred over approximately 80 acres of the site. Groundwater contamination ranges from trace level polynuclear aromatic hydrocarbons to levels exceeding single compound solubility limits indicating an oil/water emulsion (CH2M-Hill 1984). In the most severely contaminated areas of the site the groundwater is in fact a hydrocarbon fluid which can be recovered and reused to treat wood products. Therefore, the spectrum of groundwater contamination at this site runs from trace levels in an aqueous media to a hydrocarbon fluid or recoverable creosote.

In addition to on-site groundwater contamination, field investigations have shown that the Laramie River adjacent to the site has at various times shown oily seeps which have contaminated both the water and the river sediments downstream of the site. The implication of downstream contamination has led to the posting of no fishing - no swimming signs until such time as it can be established that no threat to human health through contact with contaminated sediments or consumption of fish exists.

This site is indicative of many if not most contaminant sources in that the pollution involves a complex mixture of chemicals and both groundwater and surface waters are involved. This creates a problem for

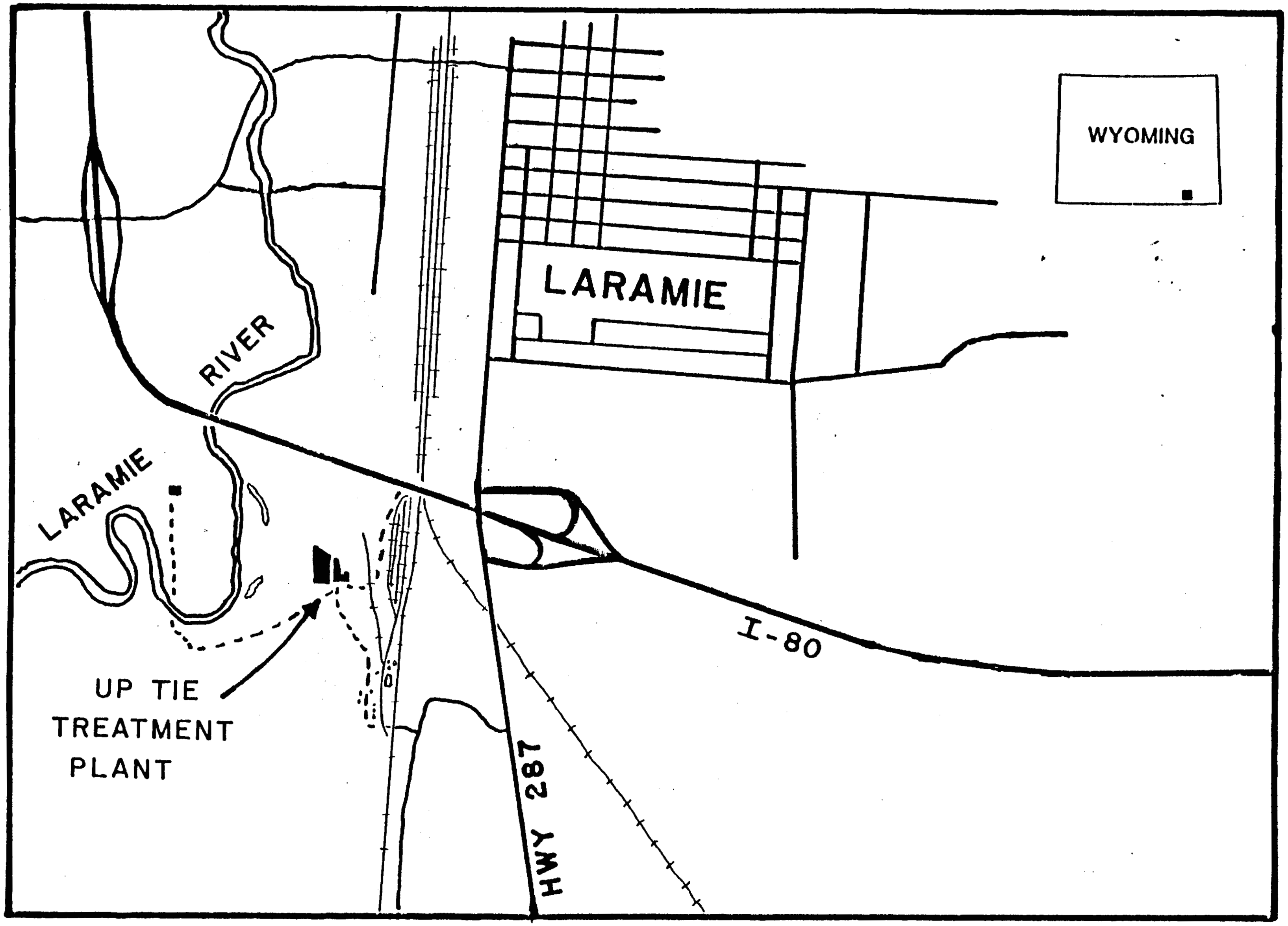


Figure 1. Location of Union Pacific Tie Treatment Plant near Laramie, Wyoming

contaminant hydro-geologists in that the complex interactions among chemicals in the mixture can affect the environmental transport and fate of any single chemical. For example, the soil adsorption/desorption rate constants for a hydrophobic compound derived experimentally using water as the fluid medium will greatly overestimate the retardation seen in a situation where solvents such as benzene, or toluene are also present in the mobile fluid. Modeling such complex and variable interactions is difficult at best. The ability of state and federal regulatory agencies to detect, monitor and predict the movement and fate of complex contaminant mixtures, as well as plan and implement remedial measures, has been hampered by the lack of knowledge concerning multi-phase transport phenomenon. While no two sites are exactly alike there are certain general principles discussed here which may be applicable to other similarly contaminated sites.

OBJECTIVES

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

1) Transport model evaluation: Literature review and evaluation of contaminant transport and fate models dealing with complex organic mixtures.
2) Contaminant transport in ground water: Laboratory soil column experiments to investigate movement of contaminated oils, oil/water emulsions and dissolved compounds in simulated groundwater conditions.
3) Contaminant transport in surface water: Field sampling and analysis of Laramie River sediments downstream from the Union Pacific Tie Treatment Plant.
4) Report to regulatory agencies: Summarize site study and literature review to provide assessment of current methods to model transport of complex contaminant mixtures in groundwater and surface water.

Through the course of the FY83 project these objectives were modified slightly and expanded to add more intensive sampling at our case-study field site. These modifications were incorporated into the FY84 objectives. The principle additions in the FY84 study were to expand the groundwater 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 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 finding 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 Department of Environmental Quality (DEQ) by the 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 organic 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).

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 by M. Crossey at the public hearing in February, 1984 pointed out the danger of on-site storage of solidified organic wastes due to the possible infiltration of groundwater as well as the questionable interaction of clay liners and organic fluids such as those found at the wood-treating facility. Reviews of the Phase III, Remedial

Action Feasibility Study, and a Risk Assessment have also been submitted. However, these two documents have not been issued in final form as of the time of this report. A brief presentation of the results of the river mini-piezometer study and the oil body underlying the Laramie River was given to the State, the EPA and the Union Pacific at an advisory meeting in February 1985. Recommendations were made to move the river as a first step in any remedial action plan.

TASK 1: TRANSPORT MODELS

Although the development and use of contaminant transport models for single chemicals in ground and surface waters and 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 multi-phase 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 knowledgeable researchers in North America confirmed that no routine transport models for complex contaminant mixtures apparently exist. With regard to this question, most of the relevant literature discovered dealt only with the conceptual framework necessary to approach such a modeling problem (e.g., Bergman and Meyer 1982, Makay et al. 1985), or was a mathematical representation of an idealized situation (Ambriola and Pinder 1985a and 1985b). Villaume (1985) reviews some recent case studies and multi-phase modeling attempts.

Laboratory models are currently being used which demonstrate empirically the important parameters controlling the flow of immiscible contaminants in aquifers (Scwille 1984, Barker 1982). Many physical properties of the fluid such as solubility, density, viscosity, surface tension, vapor pressure and relative density all exert a marked effect on the migration of an immiscible contaminant. In addition, the ability of a model to adequately predict the migration of a complex mixture is limited by lack of knowledge concerning the chemical interactions taking place among chemicals within the solution matrix. Single organic compounds such as trichloroethene can be modeled adequately in simple systems. However, any anisotropies not accounted for in the model (whether in the rock matrix or the flow patterns) can drastically effect the actual field movement of the compound. This points out an obvious difference between dissolved and immiscible contaminant flow modeling. Dissolved contaminants can usually be modeled almost as accurately as the groundwater itself can be modeled. On the contrary, immiscible contaminants do not necessarily follow groundwater flow paths. In addition, the aqueous and non-aqueous relative permeabilities are controlled by the multi-phase composition of such solutions. The percent aqueous/organic composition of the fluid can change over space and time and thus drastically change the fluid flow solely to changes in solution regardless of the homogeneity of the rock matrix (Schwille 1984, Marle 1981, Villaume 1985).

Although no models in the current literature are applicable to a situation as complex as the wood-treating facility there are several general contaminant models. We have classified the models and literature into three categories: (1) single-contaminant groundwater transport models (Javandel et al. 1984), (2) single-contaminant surface water transport models, and (3) multi-phase oil/water transport models that are usually applied to oil reservoir dynamics problems (Marle 1981).

Groundwater 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 groundwater for domestic and agricultural uses. Models such as these are now fairly refined and most situations can be adequately modeled (Freeze and Cherry 1979). Groundwater contaminant transport modeling is a more recent development and has built upon the previously established resource modeling techniques. There are three general types of models available; advection, advection-dispersion, and geochemical. These types of models allow one to theoretically incorporate the effects of both the aquifer matrix and chemical interaction 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 groundwater 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 will require multi-phase flow modeling (Marle 1981). In general, the theoretical/mathematical development of contaminant transport models is limited by the amount of field data that goes into the model. Multi-phase systems are controlled by many more interactions and consequently require a more detailed set of field parameters. Most situations would require a prohibitively large data set in order to model effectively. However, Ambriola and Pinder (1985a, 1985b) have developed a finite difference model which closely approximates an ideal situation and could, with sufficient field data, be tested against an actual field situation.

Surface water contaminant modeling presents many of the same short-comings as groundwater 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 field studies and observations are required to detect and define the important controlling processes upon which the models are to be based. A discussion of the fate, effect and modeling of complex effluents in surface water system can be found in Bergman, Kimerle and Maki (1985).

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 as well as with the solid matrix through which the fluid is moving. 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). Given that creosote is a complex mixture of over one hundred compounds (EPA 1984), we have approached this problem in a more general way. The studies presented below were designed to treat creosote as a whole and to define properties that would describe the behavior of this mixture in ground water ignoring any environmental partitioning for the time being. This approach seems valid for the TTP because, as described below, some of the contamination reaching the river at the TTP is creosote as an undiluted, undifferentiated oil.

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, 2 mm glass beads, site soil, and washed sand. After column packing, the test material 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 flourescence (described below) and HPLC (described in Task 3). Specific

TABLE 1. Summary of Column Parameters Used in the Laboratory Migration Studies

Parameter	Value
Packing	2mm glass beads
Column Length	60.5 cm
Column Diameter	14.0 cm
Volume	10,691 cm^3
Pore Volume	3,650 cm^3
Porosity %	34.14%
Discharge (Q)	2 - 20 mls/min (Variable*)
Cross-sectional Area (A)	176.71 cm^2
Specific Discharge v (Q/A)	Ca. 10^{-2} - 10^{-4} cm/sec (Variable*)
Temperature	27°C

* These parameters can be varied by controlling the flow through the column using a peristallic pump.

column parameters are described in Table 1. The results reported below for the laboratory column study are preliminary and further experimentation and results should be completed within the next year.

In addition to this lab column study 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 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. 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.). A geologic cross-section of the site is shown in Appendix 1.

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 extractred 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.

Results and Discussion

Only the preliminary observations from the column studies can be presented in this report. In the first experiment, no flow through the column was used. Once packed with glass beads, 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/cm^3 but remained in this unstable equilibrium. This phenomenon had 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 decend 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 ceased following the initial movements (over a two month period no movement was recorded). This phenomenon has been described previously (Schwille 1984, Villaume 1985, Marle 1981). As the non-aqueous fluid (creosote) moves downward in the column its saturation declines until a point of residual saturation is reached and no further movement can occur.

Following the initial creosote experiment we have begun a series of single compound experiments. In this way we can build from a background of single compounds and compare these migration patterns to those seen in a complex mixture such as creosote. The results from the first experiment are show in Figure 2. Chloride was used as the conservative tracer and naphthalene was the first single compound organic. Naphthalene shows retardation in break-through due to interaction with the matrix. The concentration of naphthalene was 15 ppm, one half of its saturation concentration. The breakthrough for naphthalene occurs at approximately 1.5 pore volumes. The final experiment reported here for the column studies involved the movement of naphthalene in an emulsion mixture. A 50:1 mixture of creosote and distilled water was vigorously mixed in order to form a stable emulsion. The solution was transferred to an 8 l beaker using a siphon which drew solution from the center of the mixing container. Using this procedure we were able to avoid the oil film which formed on the top of the mixing container as well as the free oil droplets which sank to the bottom. The emulsion was cloudy white but not opaque. As can be seen from Figure 2 the naphthalene in the emulsion appears to move at a faster rate than the naphthalene in the single compound experiment. The greater variability in the naphthalene concentrations in the emulsion experiment reflect the analytical difficulties in detecting and quantifying a single compound in a complex mixture such as creosote. The concentration of naphthalene in the emulsion was 60 ppm almost twice its saturation concentration. This preliminary data indicates that compounds in solution at concentrations high enough to form emulsions could possibly travel with groundwater at rates exceeding dissolved single compounds. One explanation for this phenomenon would be that the small micelles forming the emulsion interact less with the solid matrix than did the dissolved

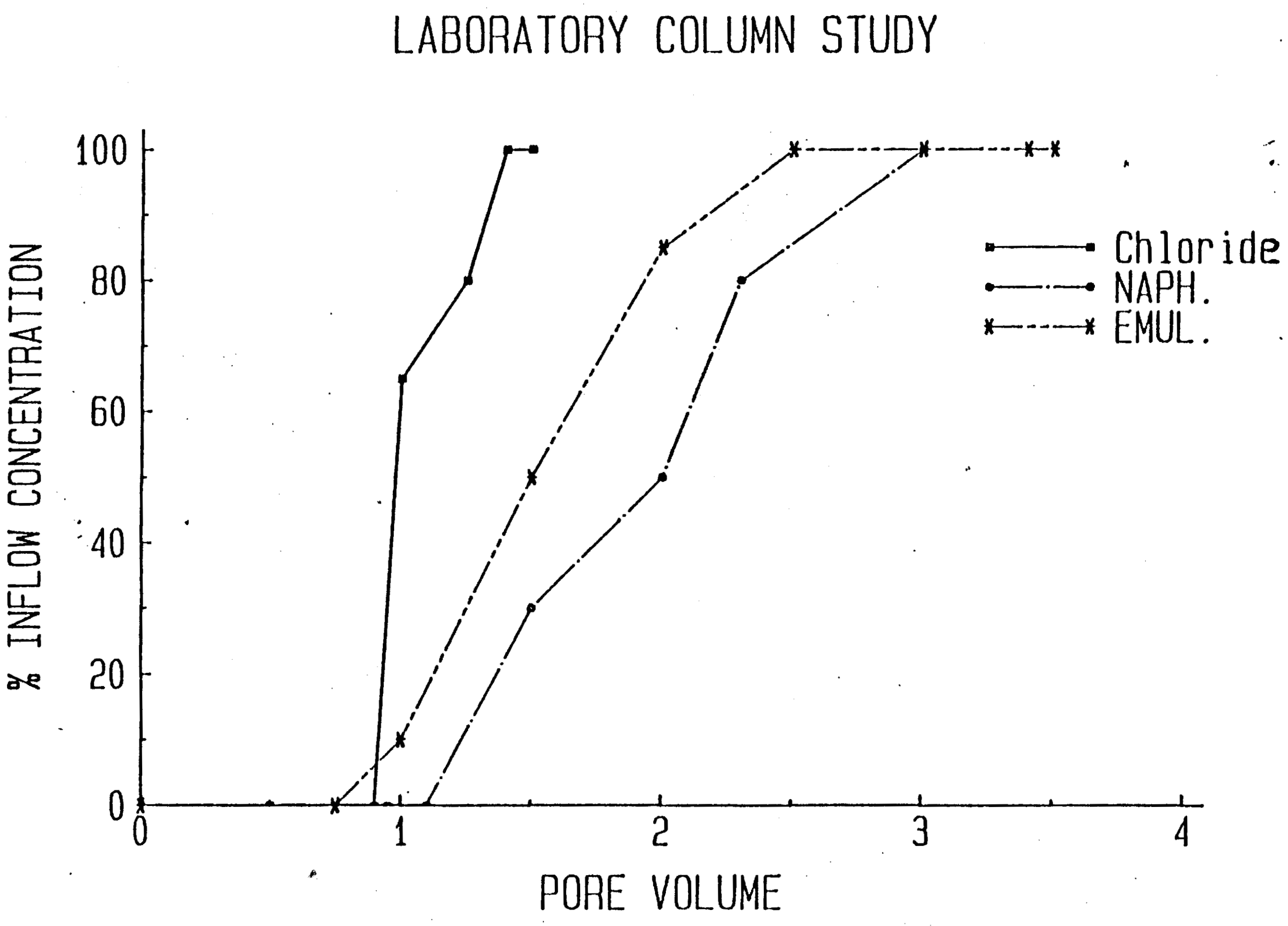


Figure 2. Elution patterns for laboratory columns. Experiment 1 ; Chloride = 5 ppm Cl^- NAPH.= naphthalene (15 ppm) in distilled water, EMUL.= naphthalene concentration in a creosote emulsion (inflow concentration approx.=60ppm). See text for experimental details.

single compound. Both of these preliminary experiments will be repeated and several other single compounds including phananthrene and chrysene will be tested as well. These initial results indicate that the design of the columns is satisfactory and that future column experiments will provide useful information regarding the flow of multi-phase liquids through a porous media.

Results from the field study using mini-piezometers have shown conclusively that creosote is leaving the site and entering the Laramie River. The first set of mini-piezometers were sampled in February 1984. The location of each of the mini-piezometers is shown in Figure 3.

As can be seen from the data for total fluorescence of piezometer water extracts (Fig. 4), there is an obvious seep at sample location 10. This sampling point lies east and slightly north of well 23 (Fig. 3). Sample 11 which is mid-river directly east of sample 10 also shows high fluorescence. Sample 10 was, in fact, black, oily fluid which had a strong creosote odor. Other samples had fluorescence values that indicated that they may have been near other seeps (samples 3, 16, 32, 36 for example). Total fluorescence of sample 36 would seem to indicate that there may have been a seep at the northern end of the old river oxbow near well D-32.

These data confirmed that organic contaminants were reaching the Laramie River as oily seeps. The fluorescence data indicated 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

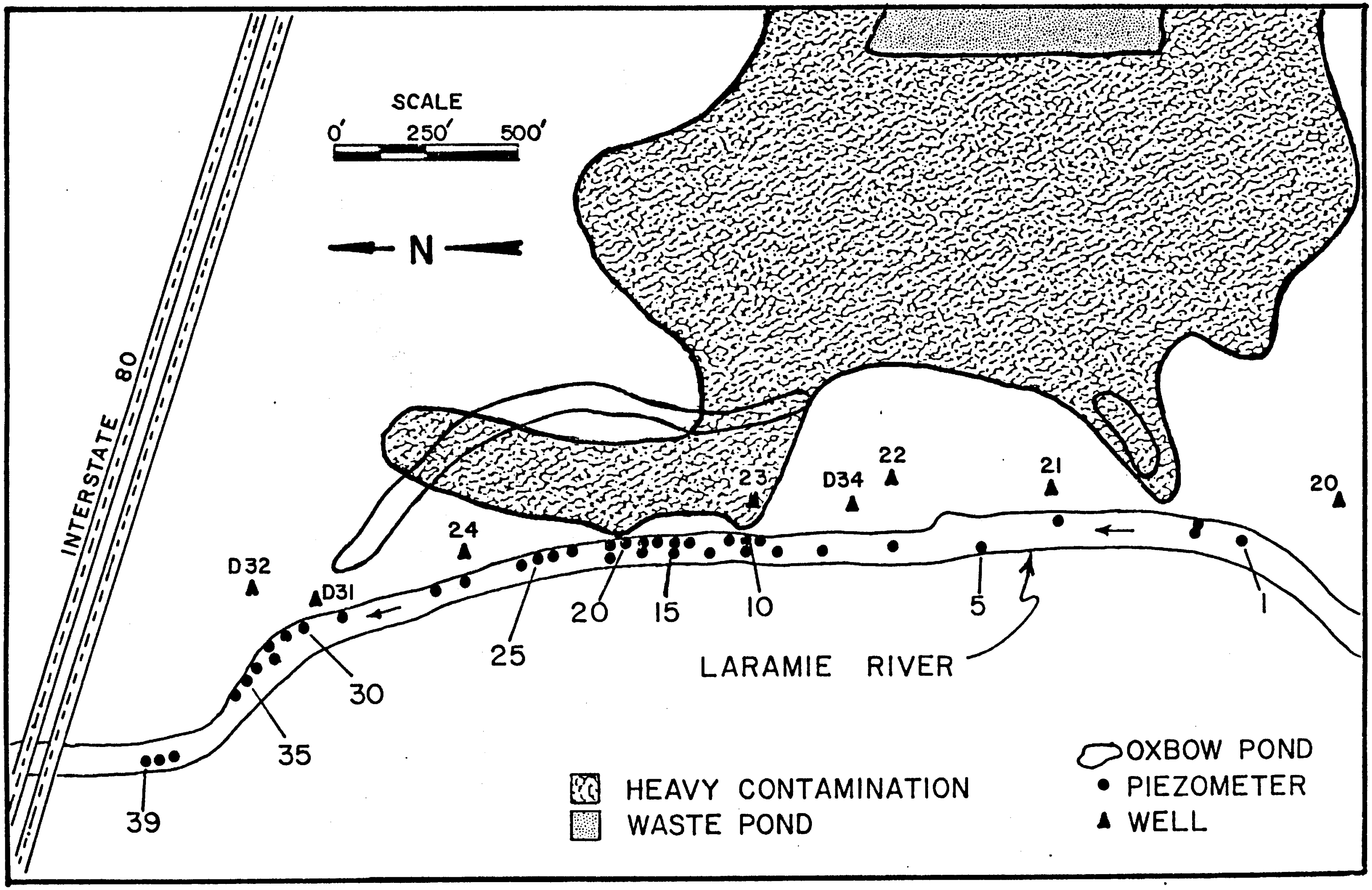


Figure 3. Location of mini-piezometer sampling points in the Laramie River adjacent to the Union Pacific Tie Treatment Plant near Laramie, Wyoming.

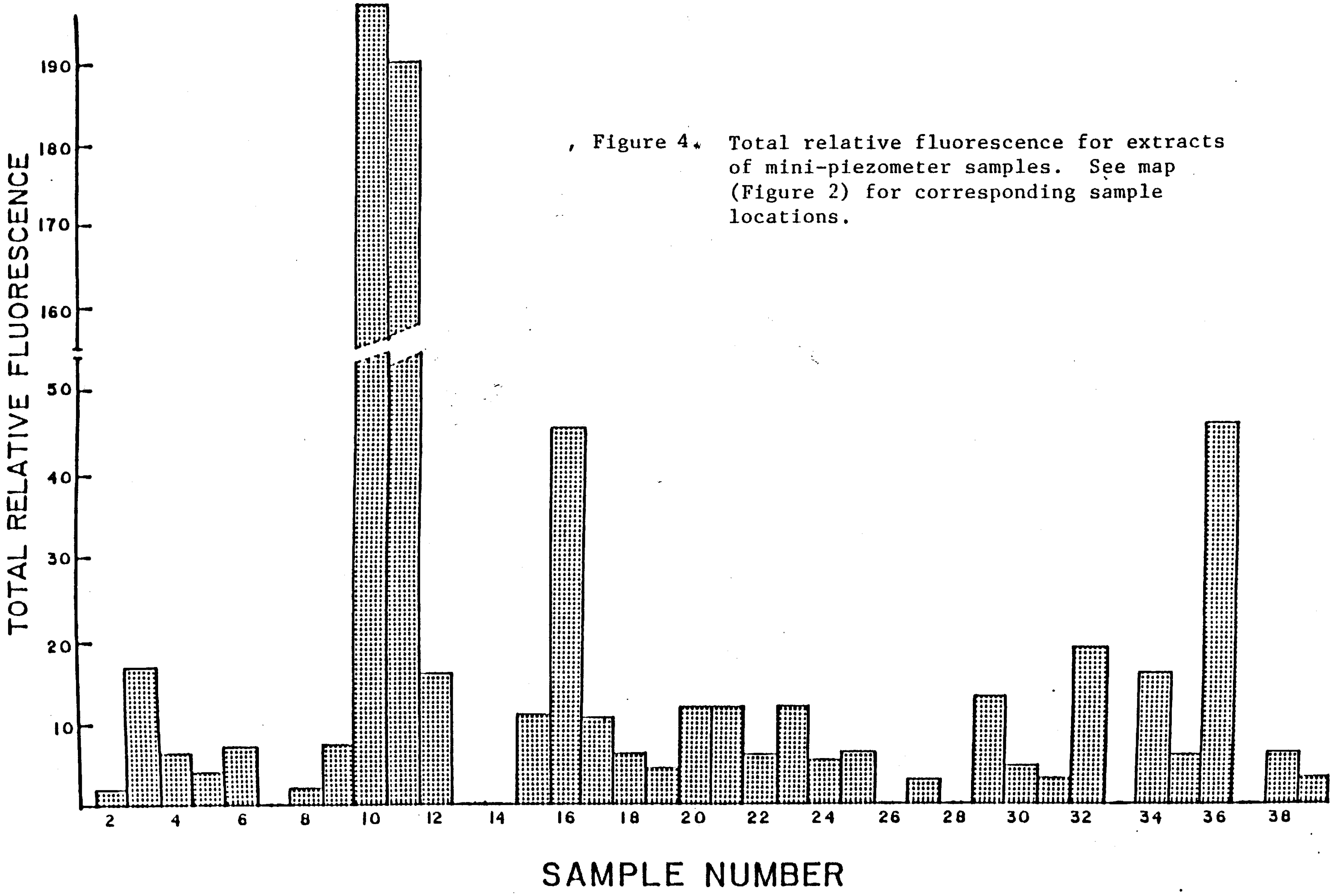


Figure 4. Total relative fluorescence for extracts of mini-piezometer samples. See map (Figure 2) for corresponding sample locations.

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 centimeters to several meters, and the sampling grid of 10 to 100 meters may have missed a number of contaminant seeps.

In order to better define the movement of creosote related contaminants from the site to the Laramie River we resampled the river in October 1984 and again in February 1985. At both these times the spacing interval was shortened to 1 meter to better define the oily seeps. During both these sampling periods a marked change was discovered from the previous (February 1984) sampling. The oily seep at piezometer number ten, Figure 2 and Figure 3, had increased. There was now an oil body underlying the river approximately three to four feet below the bed surface. This oil body was approximately 90 feet long and extended approximately 30 feet into the river channel (Fig. 5). This oil body was an average of two feet thick and given that the bed sands would have a porosity of about 30% this would mean that approximately 6,000 gallons of creosote were underlying the Laramie River. This was the only area where any significant changes from the February 1984 sampling period could be seen. No other sampling points contained free oil. The presence of this oil body has necessitated the moving of the river channel prior to remedial actions. This has recently been completed and the current channel runs approximately 50 yards west of the old channel. The mini-piezometers have remained in place and will be used to monitor the oil body until the site is hydrologically secured.

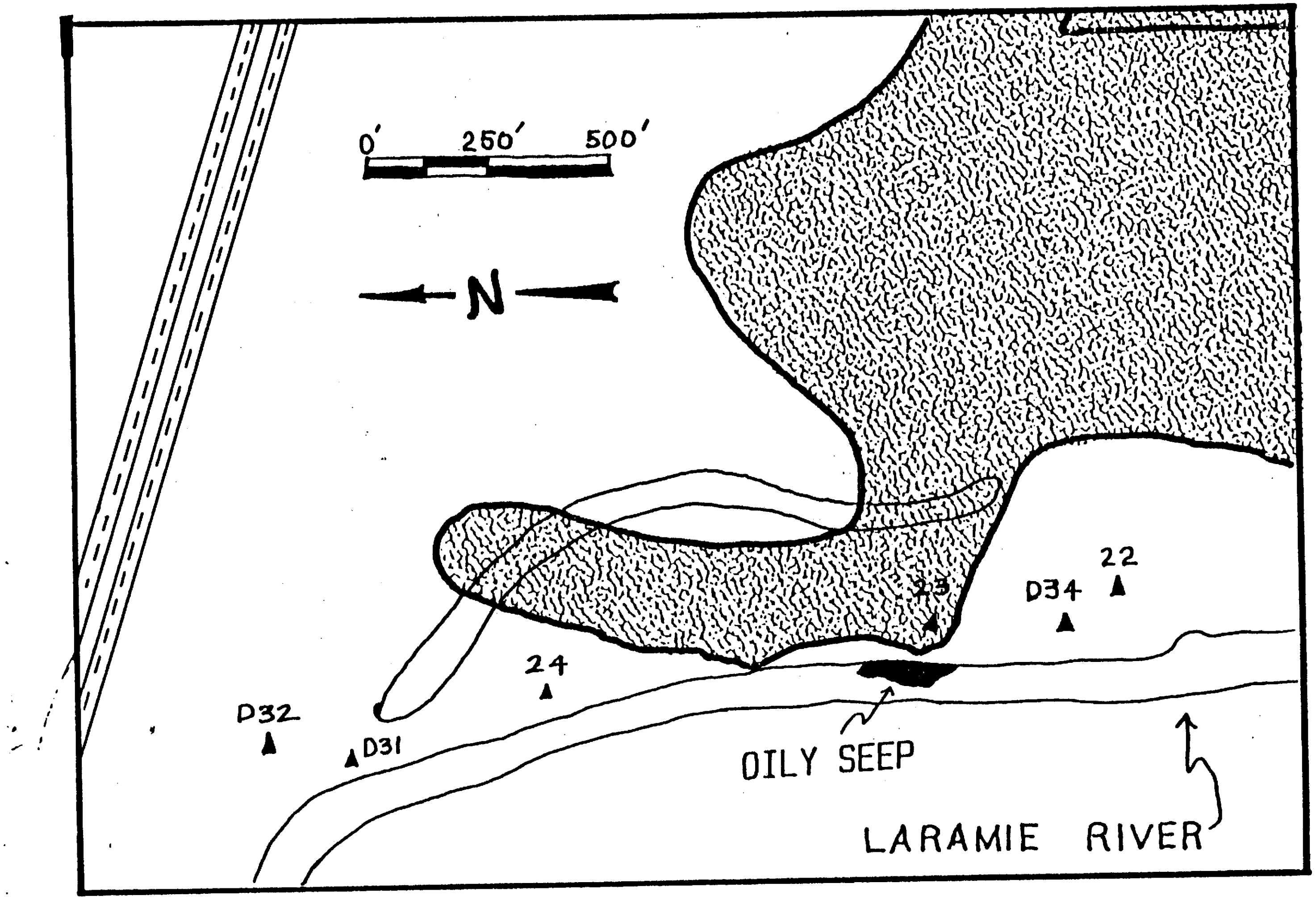


Figure 5. Creosote oil body underlying the Laramie River adjacent to the Union Pacific Tie Treatment Plant near Laramie , Wyoming, 1984-1985.

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 were 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 (Urching 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 analyzed fish taken from 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 course 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 provided approximately 500 mls of "organic rich" sediments. Sticks, leaves and other course 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). Eleven of these sites were resampled in March 1985 in order to compare the concentrations and detect any changes that may have occurred over a two year period.

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 hours. Following extraction, samples were allowed to settle and a 2 ml aliquot of the extraction solvent was removed, filtered through a 0.2 um 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/280nm) 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.

In addition to HPLC analysis, two sites were analyzed using gas chromatography/mass spectroscopy (GC/MS). These samples were quantitatively analyzed and corroborated the HPLC qualitative analysis.

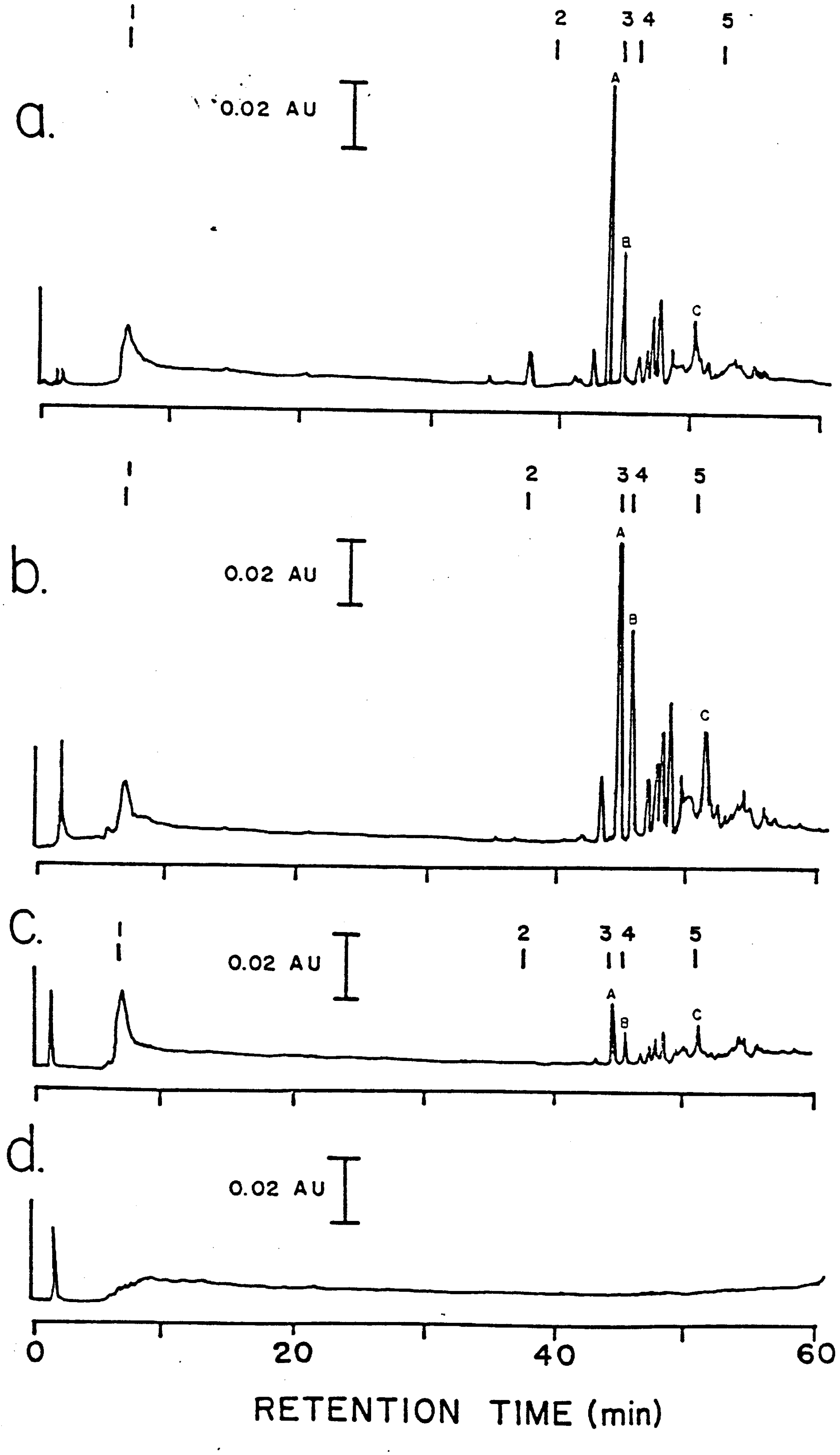
Fish were sampled on May 9, 1983 using electroshocking equipment. Four composite samples were analyzed. After sampling, fish were eviscerated, decapitated, and individually wrapped in aluminum foil and frozen. Analyses were performed at the U.S. Fish and Wildlife Service, Columbia National Fisheries 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 gell 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 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. 6). 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 contaminated soil and Laramie River sediment (Figs. 6b, c). This pattern is not seen in

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: 1 = pentachlorophenol ($C_6H_5Cl_5OH$), 2 = naphthalene ($C_{10}H_8$), 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.


a.
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2
3 4
5
A
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C
d.
0.02 AU
0
20
40
60
RETENTION TIME (min)

upstream sediment samples (Fig. 6d). 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. 6a). 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 (1gm/20 mls). Clearly the pattern shown in Figure 6 a,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). In general it appears from the samples taken from the same locations on both sampling times (1983 and 1985) that the levels have decreased from 1983 to 1985.

The contaminants detected using quantitative GC/MS analysis also indicate a decline from 1983 to 1985 samples (Table 2).

The results presented in Table 3 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 2. Gas Chromatography - Mass Spectroscopy (GC/MS) Analysis for Base/Neutral Organic Compounds in Laramie River Sediments[a]

Parameter (ppb)[b]	Curtis Street Bridge 1983[c]	Curtis Street Bridge 1985[c]	"Swimming Hole" 1983[d]	"Swimming Hole" 1985[d]
Acenaphthene	220	BMDL[e]	450	BMDL
Anthracene	300	BMDL	200	78
Benz(a)Anththracene	540	86	140	85
Benzo(a)Pyrene	400	120	120	BMDL
Benzo(b)Fluoranthene	320	BMDL	100	84
Chrysene	520	140	160	110
Fluoranthene	1600	120	720	190
Fluorene	240	BMDL	370	BMDL
Naphthalene	180	BMDL	BMDL	BMDL
Phenanthrene	1000	75	510	110
Pyrene	1300	120	460	190

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[a] For complete laboratory report see Appendix 2.

[b] Parameter concentration (ppb) = ug/kg.

[c] Curtis Street Sampling Station 3.5 km downstream from TTP. Total organic carbon (TOC) for sediments; 2.49% 1983, 2.31% 1985.

[d] "Swimming Hole" sampling station 1 km downstream from TTP. Total organic carbon (TOC) for sediments; 1.15% 1983, 2.26% 1985.

[e] BMDL = Below Minimum Detection Limit (64 ppb).

TABLE 3. Summary of Fish Tissue Analysis Results From the Laramie River Downstream From the Union Pacific Tie Treatment Plant, Laramie, Wyoming. Samples were

Sample	Description	Distance Downstream (km)	Compound[a]	Concentration[a] (ppb)[b]
1	Composite of 3 white suckers; edible portion; 608, 794 and 907 gm	0	Acenaphthalene	142
			Dibenzofuran	65
			Fluorene	56
			Naphthalene	117
			Pentachlorophenol	147
2	Composite of 2 white suckers; whole fish; ~ 200 gm each	0.25 - 1.5	Methylnaphthalene	114
			Naphthalene	58
			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	73
			Pentachlorophenol	119

[a] Only those compounds found at concentrations greater than 50 ppb are reported.

[b] ppb = parts per billion = ng/gm.

SUMMARY AND CONCLUSIONS

1) To date it appears that no modeling effort could 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. However, the contaminant levels over the past two years appear to be declining.

4) Droplets of free oil (creosote) were probably an important component of the mass flux of contaminants from the site to the river system during the 1983-1984 sampling period. The 1985 samples did not indicate any free oil downstream from the TTP.

5) The soil columns, mini-piezometers, sediment sampling methods, and analytical methods (flourescence and HPLC) developed in this study have proven to be satisfactory and will be continued to monitor this site.

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APPENDIX 1

Geologic Cross-Section of the Union Pacific
Tie-Treatment Plant South of Laramie, Wyoming
(Source: CH2M-Hill 1984).

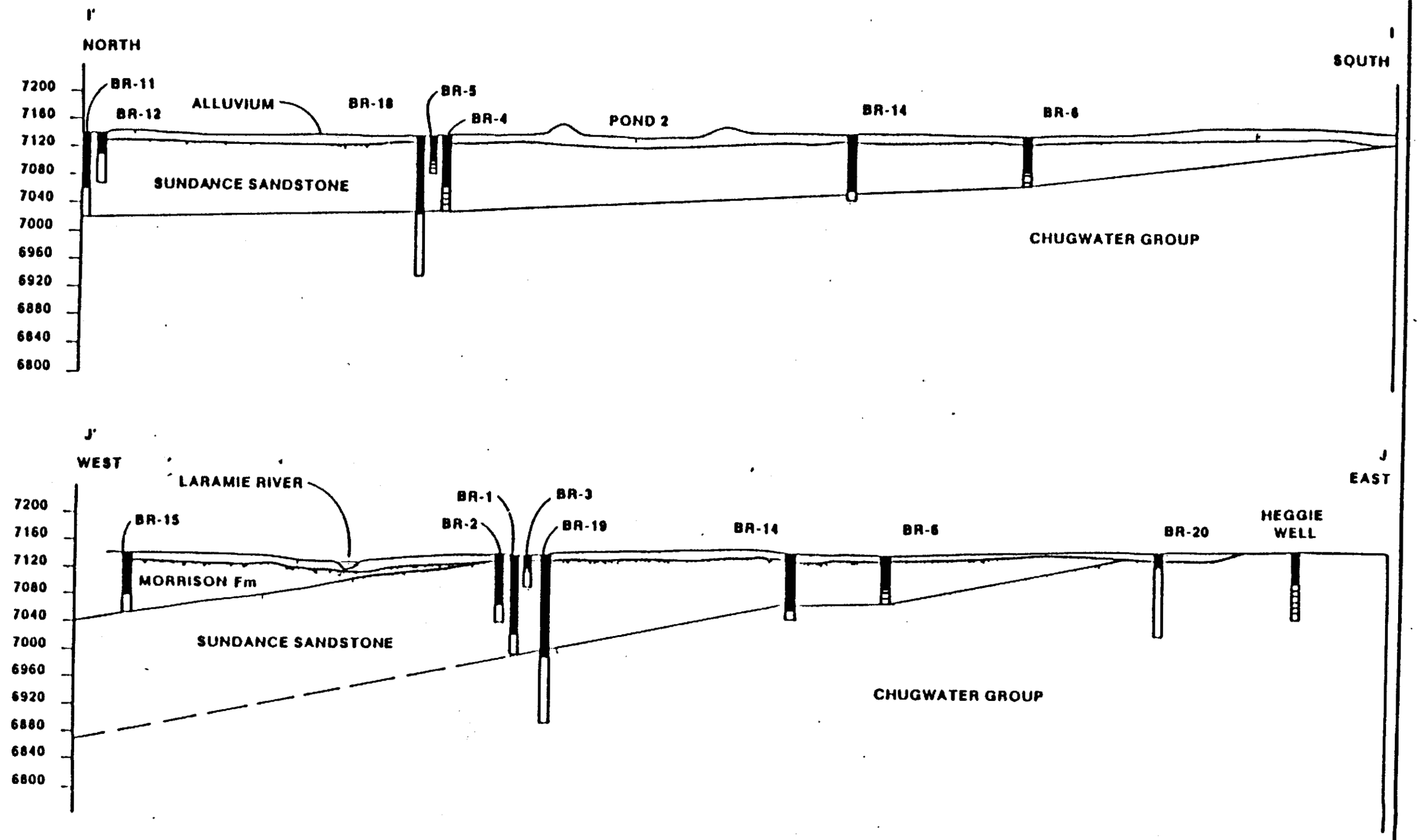

FIGURE 3-12
GEOLOGIC CROSS SECTIONS I—I' AND J—J'
OF THE BEDROCK AT THE
UPRR TIE TREATING PLANT
LARAMIE, WYOMING

(source CH2M-HILL, Denver CO.)

APPENDIX 2

Laboratory Report for Base/Neutral Organics Analysis for Curtis Street and "Swimming Hole" Sediments 1983 and 1985.

Rocky Mountain Analytical Laboratory

August 13, 1985

Mike Crossey
Fish Physiology & Toxicology Lab
Department of Zoology and Physiology
University of Wyoming
University Station, Box 3166
Laramie, Wyoming 82701

Dear Mike:

The analyses of your samples are complete. Enclosed is the report of our findings. We attempted to push the general analytical method detection limits to the best achievable. Therefore, you may notice some results reported below the quantification detection limits given. Those results are accurate qualitatively but may present some quantitative error because they are below the well defined detection limit. Overall, many polyaromatics were detected in the samples.

Please call me if you have any questions.

Best Regards,

Michael Brooks

for O. John Logsdon
Technical Director
Organic Chemistry

OJL/cc
Enclosures

RMAL # 51161

Rocky Mountain Analytical Laboratory 5530 Marshall Street, Arvada, CO. 80002 (303) 421-6611

SUMMARY OF ANALYTICAL RESULTS AND METHODOLOGY

for

University of Wyoming

On July 15, 1985, five samples were received for analysis for polyaromatics by gas chromatography/mass spectrometry (GC/MS). The samples were labeled as shown in the attached Sample Description Information sheet. The water sample was filtered to 0.45 microns before analysis. Sample 51161-05 was received in a broken container and it was transferred to another container immediately in the sample receiving facility.

Results

The results for the analysis are shown in the attached data tables. Low levels of phthalates, acenaphthene and diphenylamine appeared in the water sample (51161-01). Because we pushed the detection limits to below the normal levels, the phthalates and diphenylamine are probably preparation artifacts. We often see them at these low levels, that are below our normal detection limits. The acenaphthene however is not a typical artifact.

The soil/sediment results showed many polyaromatics. Fluoranthene, phenanthrene and pyrene were the most concentrated species. Diphenylamine appeared in sample 51161-02 but again it should be considered an artifact of the analysis.

Methodology

The samples were extracted and analyzed by GC/MS for polyaromatics using procedures similar to the EPA Contract Laboratory Program under the Superfund program. Briefly, the water sample was extracted with methylene chloride and the extract concentrated to 1 ml for analysis. The soil/sediment samples were extracted by sonication with methylene chloride. The extracts were then concentrated to as small a volume as feasible before GC/MS analysis. All samples were spiked with surrogate and internal standards for reference throughout the analysis. The results for these quality control materials were acceptable.

The analyses were conducted by GC/MS. The instrument was optimized for the lowest detection limit achievable. The resultant detection limits were 5 to 10 times lower that the nominal detection limits for the methods. Chromatograms for the samples are attached to illustrate the level of interference in the samples. Many aliphatic hydrocarbons were detected in the samples preventing measurement of the polyaromatics at even lower levels.

SAMPLE DESCRIPTION INFORMATION

for

University of Wyoming

RMA Sample No.	Sample Description	Sample Type	Date Sampled	Date Received
51161-01	River Piezometer #2	Water	6/14/85	7/15/85
51161-02	Curtis St. East Side	Sediment	11/11/83	7/15/85
51161-03	Curtis St. Bridge	Sediment	3/18/85	7/15/85
51161-04	Swimming Hole	Sediment	11/11/8~~5~~3	7/15/85
51161-05*	Swimming Hole	Sediment	3/20/85	7/15/85

August 13, 1985

ANALYTICAL RESULTS

for

University of Wyoming

BASE/NEUTRAL ORGANICS - WATER

Parameter	Units	51161-01	
1B Acenaphthene	ug/L	0.8J	(1)
2B Acenaphthylene	ug/L	ND	(1)
3B Anthracene	ug/L	ND	(1)
4B Benzidine	ug/L	ND	(10)
5B Benzo(a)anthracene	ug/L	ND	(1)
6B Benzo(a)pyrene	ug/L	ND	(1)
7B Benzo(b)fluoranthene	ug/L	ND	(1)
8B Benzo (ghi) perylene	ug/L	ND	(1)
9B Benzo (k) fluoranthene	ug/L	ND	(1)
10B Bis(2-chloroethoxy)methane	ug/L	ND	(1)
11B Bis (2-chloroethyl)ether	ug/L	ND	(1)
12B Bis(2chloroisopropyl)ether	ug/L	ND	(1)
13B Bis(2-ethylhexyl)phthalate	ug/L	ND	(1)
14B 4-Bromophenyl phenylether	ug/L	ND	(1)
15B Butyl benzyl phthalate	ug/L	ND	(1)
16B 2-Chloronaphthalene	ug/L	ND	(1)
17B 4-Chlorophenyl phenylether	ug/L	ND	(1)
18B Chrysene	ug/L	ND	(1)
19B Dibenzo(a,h) anthracene	ug/L	ND	(1)
20B 1,2-Dichlorobenzene	ug/L	ND	(1)
21B 1,3-Dichlorobenzene	ug/L	ND	(1)
22B 1,4-Dichlorobenzene	ug/L	ND	(1)
23B 3,3'-Dichlorobenzidine	ug/L	ND	(4)
24B Diethyl phthalate	ug/L	5	(4)
25B Dimethyl phthalate	ug/L	1	(1)
26B Di-n-butyl phthalate	ug/L	3	(1)
27B 2,4-Dinitrotoluene	ug/L	ND	(1)
28B 2,6-Dinitrotoluene	ug/L	ND	(1)
29B Di-n-octyl phthalate	ug/L	ND	(1)
30B 1,2-Diphenylhydrazine*	ug/L	ND	(1)

ND = Not detected. *Measured as azobenzene. Detection limits in parentheses.
J = Concentration detected is less than the required quantitative detection limit but mass spectrum confirms presence of the component.

ANALYTICAL RESULTS

for

University of Wyoming

BASE/NEUTRAL ORGANICS - WATER(Cont.)

Parameter	Units	51161-01	
31B Fluoranthene	ug/L	ND	(1)
32B Fluorene	ug/L	ND	(1)
33B Hexachlorobenzene	ug/L	ND	(1)
34B Hexachlorobutadiene	ug/L	ND	(1)
35BHexachlorocyclopentadiene	ug/L	ND	(1)
36B Hexachloroethane	ug/L	ND	(1)
37B Indeno(1,2,3-cd)pyrene	ug/L	ND	(1)
38B Isophorone	ug/L	ND	(1)
39B Naphthalene	ug/L	ND	(1)
40B Nitrobenzene	ug/L	ND	(1)
41B N-Nitrosodimethylamine	ug/L	ND	(1)
42B N-Nitrosodi-n-propylamine	ug/L	ND	(1)
43B N-Nitrosodiphenylamine*	ug/L	1	(1)
44B Phenanthrene	ug/L	ND	(1)
45B Pyrene	ug/L	ND	(1)
46B 11,2,4-Trichlorobenzene	ug/L	ND	(1)

ND = Not detected. *Measured as azobenzene. Detection limits in parentheses.

ANALYTICAL RESULTS

for

University of Wyoming

BASE/NEUTRAL ORGANICS - SOILS

Parameter	Units	Curtis St. 1983	Curtis St. 1985	Swimming Hole 1983	Swimming Hole 1985
		51161-02	51161-03	51161-04	51161-04
1B Acenaphthene	ug/kg	220 (64)	ND (64)	450 (64)	ND (64)
2B Acenaphthylene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
3B Anthracene	ug/kg	300 (64)	ND (64)	200 (64)	78 (64)
4B Benzidine	ug/kg	ND (640)	ND (640)	ND (640)	ND (640)
5B Benzo(a)anthracene	ug/kg	540 (64)	86 (64)	140 (64)	85 (64)
6B Benzo(a)pyrene	ug/kg	400 (64)	120 (64)	120 (64)	ND (64)
7B Benzo(b)fluoranthene	ug/kg	320 (64)	ND (64)	100 (64)	84 (64)
8B Benzo (ghi) perylene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
9B Benzo (k) fluoranthene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
10B Bis(2-chloroethoxy)methane	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
11B Bis (2-chloroethyl)ether	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
12B Bis(2chloroisopropyl)ether	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
13B Bis(2-ethylhexyl)phthalate	ug/kg	220 (64)	93 (64)	120 (64)	160 (64)
14B 4-Bromophenyl phenylether	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
15B Butyl benzyl phthalate	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
16B 2-Chloronaphthalene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
17B 4-Chlorophenyl phenylether	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
18B Chrysene	ug/kg	520 (64)	140 (64)	160 (64)	110 (64)
19B Dibenzo(a,h) anthracene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
20B 1,2-Dichlorobenzene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
21B 1,3-Dichlorobenzene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
22B 1,4-Dichlorobenzene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
23B 3,3'-Dichlorobenzidine	ug/kg	ND (260)	ND (260)	ND (260)	ND (260)
24B Diethyl phthalate	ug/kg	ND (260)	ND (260)	ND (260)	ND (260)
25B Dimethyl phthalate	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
26B Di-n-butyl phthalate	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
27B 2,4-Dinitrotoluene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
28B 2,6-Dinitrotoluene	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
29B Di-n-octyl phthalate	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
30B 1,2-Diphenylhydrazine*	ug/kg	ND (64)	ND (64)	ND (64)	ND (64)
31B Fluoranthene	ug/kg	1600 (64)	120 (64)	720 (64)	190 (64)

ND = Not detected. *Measured as azobenzene. Detection limits in parentheses.

ANALYTICAL RESULTS

for

University of Wyoming

BASE/NEUTRAL ORGANICS - SOILS(Cont.)

Parameter	Units	51161-02		51161-03		51161-04		51161-05	
32B Fluorene	ug/kg	240	(64)	ND	(64)	370	(64)	ND	(64)
33B Hexachlorobenzene	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
34B Hexachlorobutadiene	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
35B Hexachlorocyclopentadiene	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
36B Hexachloroethane	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
37B Indeno(1,2,3-cd)pyrene	ug/kg	160	(64)	ND	(64)	ND	(64)	ND	(64)
38B Isophorone	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
39B Naphthalene	ug/kg	180	(64)	ND	(64)	ND	(64)	ND	(64)
40B Nitrobenzene	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
41B N-Nitrosodimethylamine	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
42B N-Nitrosodi-n-propylamine	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)
43B N-Nitrosodiphenylamine*	ug/kg	220	(64)	ND	(64)	ND	(64)	61J	(64)
44B Phenanthrene	ug/kg	1000	(64)	75	(64)	510	(64)	110	(64)
45B Pyrene	ug/kg	1300	(64)	120	(64)	460	(64)	190	(64)
46B 1,2,4-Trichlorobenzene	ug/kg	ND	(64)	ND	(64)	ND	(64)	ND	(64)

ND = Not detected. *Measured as azobenzene. Detection limits in parentheses.
J = Concentration detected is less than the required quantitative detection limit but mass spectrum confirms presence of the component.