DELINEATION OF NEAR-SURFACE PREFERENTIAL PATHWAYS OF CONTAMINANT TRANSPORT WITHIN THE UNSATURATED ZONE USING SOIL-GAS: A NEW METHOD FOR HYDROGEOLOGICAL SITE CHARACTERIZATION ¹

By

Frederick E. Kirschner, Jr. and Roy E. Williams²

<u>Abstract.</u> Preferential pathways for contaminant transport between the ground surface and the underlying watertable have been identified by injecting a gaseous-phase tracer and subsequently sampling for the tracer in numerous locations. Zones of anomalously higher tracer concentrations constitute zones in which preferential gaseous transport occurs from a deeper injection well to a sampling point located directly below the surface of the ground. Sulfur hexafluoride constitutes the gaseous tracer. Injection and subsequent sampling were accomplished with Paradise Creek Industries Soil-Gas Probes. Samples were analyzed on-site with an SRI 8610 gas chromatograph equipped with an electron capture detector. The results of the test identify two preferential pathways for gaseous transport that connect the ground surface of the site to the underlying aquifer. The presence of both pathways was confirmed by drilling.

Additional Key Words: ground water monitoring networks; gaseous-phase tracer; vapor injection well; vapor monitoring well; soil-gas probe; mine and mill wastes; sulfur hexafluoride; gas chromatograph.

Introduction

When confronted with the task of characterizing a site with respect to placement of

monitoring wells or siting a hazardous waste ¹ Paper presented at the 1993 National Meeting of ASSMR entitled: "The Challenge of Integrating Diverse Perspectives In Reclamation," Spokane, Washington, May 15-19, 1993. Publication in this proceedings does not preclude authors from publishing their manuscripts, whole or in part, in other publications.

² Frederick E. Kirschner is a Post Doc Fellow of Hydrogeology and Manager of the College of Mines and Earth Resources Waste Management Studies Laboratory, University of Idaho. Roy E. Williams is Professor of Hydrogeology and Director of Waste Management Studies in the College of Mines and Earth Resources at the University of Idaho, Moscow, Idaho 83843. facility, the hydrogeologist is charged with testing the subgrade to determine whether preferential pathways for contaminant transport exist between the site and the watertable (unsaturated zone).

Methods used detecting preferential pathways within the vadose zone are limited to: (1) in-situ saturated permeability tests, (2) core analyses, (3) borehole geophysical tests, and (4) surface geophysical tests. In-situ saturated permeability tests, core analyses, and borehole geophysical tests are of limited value because these methods provide only point measurements in the horizontal dimensions. Surface geophysical techniques identify anomalies in the response of a given parameter. Depending on the nature of the response, transport via such anomalies is then inferred--not observed. A new method for identifying and delineating preferential pathways for mass transport in which actual transport is observed is described herein.

The purpose of this investigation is to test the feasibility of delineating preferential

Proceedings America Society of Mining and Reclamation, 1993 pp 89-99 DOI: 10.21000/JASMR93010089

89

https://doi.org/10.21000/JASMR93010089

pathways for mass transport within the unsaturated zone. In order to accomplish this objective a gas-phase tracer test was used to identify areas within the unsaturated zone that transport the tracer more rapidly than other areas within the same medium. We hypothesize that such pathways are capable of transmitting contaminants (gaseous, aqueous, and nonaqueous phases) in the event that a release occurs.

Soil-gas surveying has been employed as a prospecting tool for over 100 years (Chavez, 1988; Farwell et al., 1984; Philip and Crisp, 1982; Taylor et al., 1982; Hinkle et al., 1978; Bristow and Jonasson, 1972). Recently, gas surveying techniques have been used to map plumes of volatile organic compounds (VOCs) that leak from underground storage tanks, surface impoundments, and landfills (Barrows, 1987; Devitt et al., 1987; Kerfoot and Barrows, 1987; Marrin, 1987, 1987; Reisenger et al., 1987; Thompson and Marrin, 1987; Balfour et al., 1986; La Breque et al., 1985; Marrin and Thompson, 1984; and Voorhees et al., 1984). More recently, soil-gas has been used to delineate preferential pathways for ground water flow (Kirschner and Williams, 1990; Kirschner, 1989).

Our method differs from those described above in that we sample for an innocuous tracer that we have introduced into the subsurface instead of sampling for a specific gas or suite of gases that has diffused from liquid-phase contaminants at depth. Also, our tracer is injected under a slight pressure; therefore, mass transport is governed by advection and diffusion whereas transport described by the previous researchers is governed by diffusion alone.

A portion of a floodplain covered by mine and mill wastes located at the the Kellogg, Idaho Superfund site constitutes the field-test area (Figure 1). This site was selected because: (1) preferentially permeable pathways have been identified in an adjacent area during other investigations; and (2) the hydrostratigraphy within the study area has been characterized (Kirschner, 1990).

<u>Generalized Hydrostratigraphy of</u> <u>the Study Area</u>

Four hydrostratigraphic units are present within the study area: (1) channel gravel, (2) brown silt, (3) gray silt, and (4) red sandy silt (Figure 2). The channel gravel and the brown silt units are natural deposits. The other two units were deposited in an impoundment that occupied the valley from the early 1900's to the early 1930's. Stumps of trees that had once grown in the gravel and brown-silt unit and logs deposited on the overlying brown silt unit by the South Fork of the Coeur d' Alene River protrude through the gray and red sandy-silts.

The channel gravel is Pleistocene in age and constitutes the uppermost aquifer of the region. The hydraulic conductivity (K) of the aquifer is estimated to be on the order of 1-10 cm/s (Ralston and Kunkel, 1989; Dames and Moore, 1990). The aquifer is unconfined much of the year.

The brown silt deposit overlies the gravel conformably and is interpreted to be a buried forest soil. The brown silt deposit acts as an upper aquitard for the system during high water-table conditions (Kirschner, 1990). Kirschner (1990) has shown that this unit is highly heterogeneous with values of field saturated hydraulic conductivity (K_{FS}) ranging from 8.7 x 10⁻³ to 3.9 x 10⁻⁶ cm/s.

The gray silt sized deposit consists of mill wastes that conformably overlie the brown silt deposit. Kirschner (1990) has shown that this unit also is highly heterogeneous with values of K_{FS} ranging from 2.5 x 10⁻⁴ to 1.2 x 10⁻⁶ cm/s. Norton (1980) reports that this unit is associated with relatively high concentrations of lead and zinc (up to 7.0 and 4.0 percent by weight, respectively).

The red sandy-silt sized deposit of mine/mill wastes overlies the gray silt deposit conformably. Kirschner (1990) also has shown that this unit is highly heterogeneous with values of K_{FS} ranging from 1.0×10^{-2} to 3.2×10^{-4} cm/s. This unit also is associated with relatively high concentrations of lead and zinc (up to 5.0 and 3.0 percent by weight, respectively; Norton, 1980).

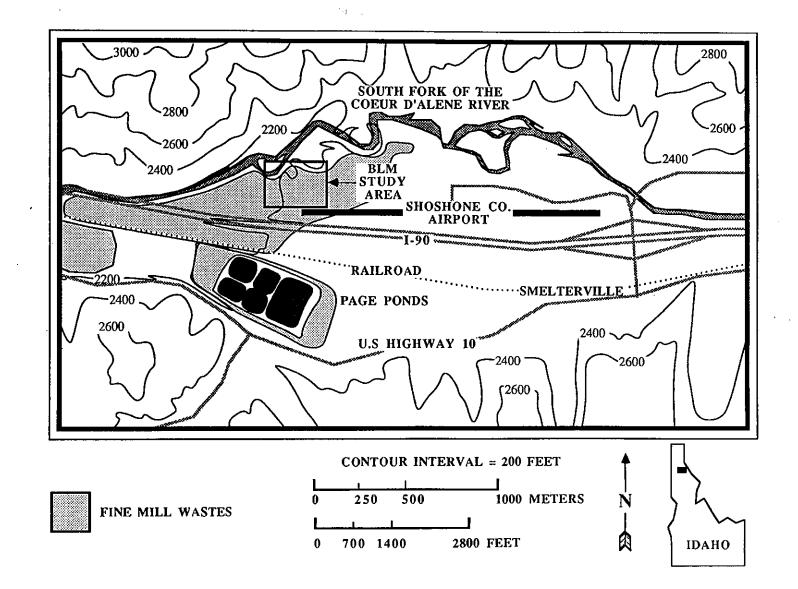


Figure 1. Location of the study area within the Kellogg Superfund site and surficial distribution of fine-grained wastes.

16

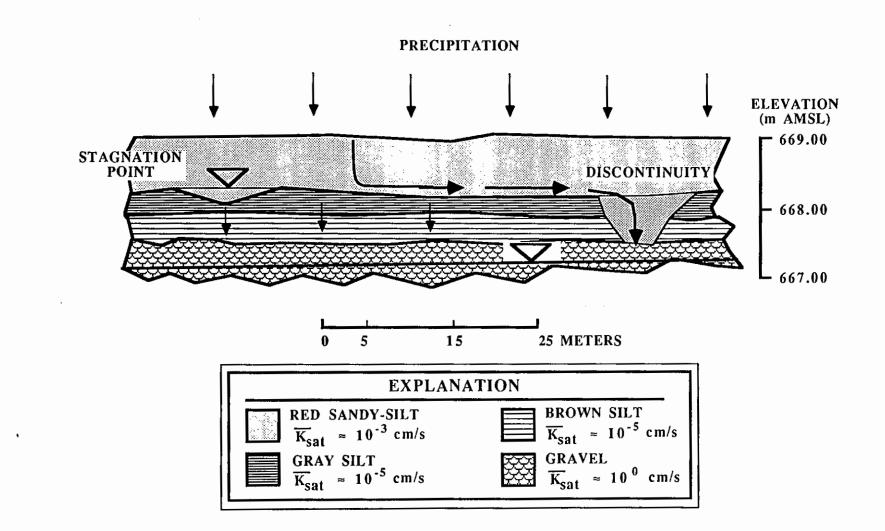


Figure 2. Generalized hydrostratigraphy and conceptual model of semi-saturated flow (arrows) within the four units. Length of arrows represent relative amounts of flow.

92

<u>Conceptual Model of Semi-Saturated Ground</u> <u>Water Flow Within the Four</u> <u>Hydrostratigraphic Units</u>

Our conceptual model of semi-saturated flow within the aforementioned units during a major infiltration event is depicted in Figure 2. The conceptual model was developed while conducting hydraulic stress tests and liquidphase tracer tests in an area immediately adjacent to the study area (Kirschner, 1991). The model is based on the contrasting values of hydraulic conductivity the different hydrostratigraphic units. We hypothesize that the brown silt and gray silt act as a semi-permeable boundary in areas where these two units are present.

Our conceptual model of semi-saturated flow is as follows: (1) water infiltrates vertically in the red sandy-silt unit and ponds on the underlying gray silt unit; (2) the major portion of flow begins to occur horizontally in the red sandy-silt unit as ponding increases with time; (3) the direction of horizontal flow in the red sandy-silt unit is controlled predominantly by the topography of the upper surface of the gray silt and the local hydraulic conductivity distribution of the red sandy-silt; (4) horizontal flow occurs until stagnation is achieved or until a preferential pathway connecting the red sandysilt unit to the top of the aquifer is encountered; (5) a small fraction of flow occurs vertically through the gray clayey silt and brown silt units to the unsaturated portion of the aquifer; and (6) stagnant water captured in subterranean basins is removed predominantly by evaporation once infiltration has ceased (the downward flux must be very low owing to the low value of K associated with the gray and brown silts and the amount of hydraulic head that can build).

Methodology and Rationale for Using a Soil-Gas Technique to Identify Preferential Flowpaths Within the Unsaturated Zone at the Site

From the aforementioned conceptual model of downward preferential ground-water flow from the red sandy-silt unit to the aquifer, it follows that such pathways can be identified by reversing the direction of flow using a gasphase tracer. Gaseous-phase flow in this system, sampling/analysis procedures, and quality assurance/quality control are detailed by Kirschner (1992). The general methodology and its rationale is described below.

Sulfur hexafluoride (SF₆) is continuously injected into the unsaturated portion of the aquifer for the duration of the study. Sulfur hexafluoride was selected as the tracer because it: (1) is anthropogenic; (2) exhibits a relatively high gas/liquid partition coefficient; and (3) is amenable to detection at concentrations below one part per billion. Injection occurs: (1) through a Paradise Creek Industries (PCI) vapor injection well, (2) under a very slight pressure, and (3) at a constant rate (2 l/s). The vapor injection well constitutes a point source for advective and diffusive flow of the tracer within this unit.

Samples from the unsaturated gravels are obtained from eight PCI vapor sampling wells arranged on a radially symmetrical grid centered on the vapor injection well (Figure 3). Once concentrations measured at these locations approach that of the source, the unsaturated gravel constitutes a planar source for upward advective/diffusive mass transport. We hypothesize that: (1) zones of higher concentrations of the tracer measured within the red sandy-silt will occur directly over preferential pathways and (2) the zones will expand with time of tracer injection.

A radial grid was selected to facilitate the comparison of advective-diffusive radial flow velocities within this unit. The results of this portion of the study are given in Kirschner (1992).

The red sandy-silt unit is sampled on a rectangular grid centered on the radial grid (Figure 3). Samples are obtained via PCI soil-gas probes driven 0.5 meters into this unit. A rectangular grid was selected in order to facilitate a statistical procedure described by Gilbert (1989) and applied to a field situation by Kirschner (1989). Sampling on the rectangular grid is initiated after full break-through is observed at all vapor wells completed within the unsaturated gravel. Off-node sampling occurs near nodes where concentrations of the tracer are anomalously high.

All samples are contained in 0.280 liter Vaccutainers[®]. Samples are analyzed on-site in a mobile laboratory with an SRI-8610 gas chromatograph equipped with an electron capture detector.

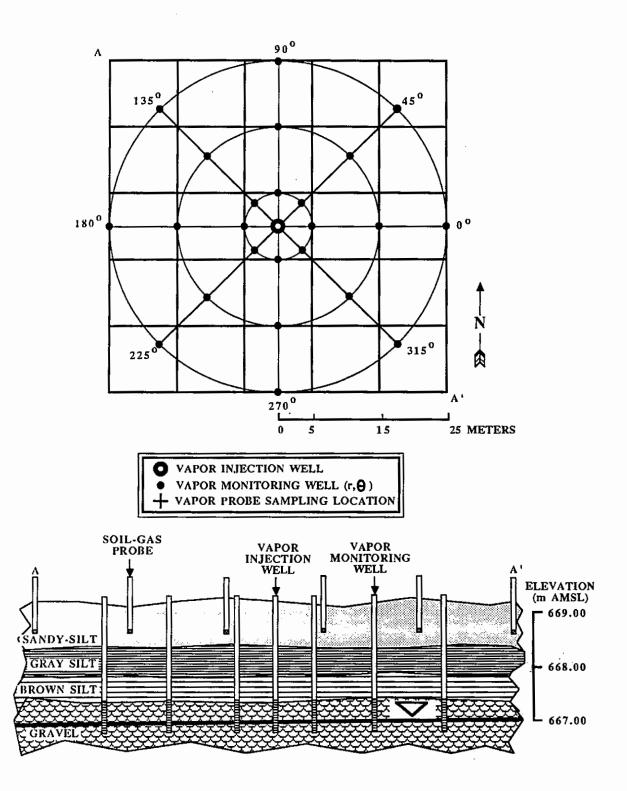


Figure 3. Map view and cross-section of both sampling grids. Vapor injection well and vapor monitoring wells (radial grid) are completed in the unsaturated portion of the gravel unit. Soil-gas probes (rectangular grid) are completed in the red sandy-silt unit.

<u>Results</u>

Two soil-gas surveys were conducted on the rectangular grid. The first soil-gas survey (SGS-1) was conducted in order to determine whether anomalous concentrations of SF₆ could be identified. The second soil-gas survey (SGS-2) was conducted later time to determine whether or not such anomalies increased in size.

<u>SGS-1</u>

Sampling in the sandy-silt was initiated 5.0 hours after full breakthrough within the unsaturated gravel occurred (approximately 30 hours after injection began). Sampling proceeded outward from the injection well because radial flow within the unsaturated gravel causes full breakthrough to occur nearest the injection well first. This portion of the study took approximately three hours to complete.

Two preferential pathways that connect the red sandy-silt to the unsaturated gravel are present within the study area (Figure 4). Eight nodes associated with the northeastern anomaly had concentrations of SF₆ that were higher than background. Only one node associated with the southwestern anomaly had concentrations of SF₆ that were higher than background. Off-grid sampling occurred immediately after sampling on the grid was completed. This portion of the study took 1.5 hours to complete.

<u>SGS-2</u>

The second soil-gas survey was initiated 5.0 hours after SGS-1 had been completed to determine whether: (1) any new anomalies were present and (2) the two previously identified anomalies had expanded appreciably. No new anomalies were identified. Comparison of Figures 4 and 5 indicate that both anomalies increased in size, but did not shift position. This suggests that the probability of locating such anomalies given a certain nodal spacing increases with the duration of injection.

Investigation of the Two Anomalies

Thirty-three holes were drilled to verify the structural features hypothesized to be responsible for the SF₆ anomalies. Ten holes were drilled in areas in which SF₆ was not detected (Figure 5).

A buried log that protrudes through the gray silt and brown silt units directly underlies the axial trace of the southwestern anomaly. This preferentially permeable flowpath is interpreted to result from the decay of the stump prior to deposition of the gray silt. The lateral extent of the stump is much less than that of the anomaly.

The gray and brown silt deposits are absent below the axial trace of the northeastern anomaly. The lateral extent of this discontinuity also is much less than that of the anomaly.

All of the hydrostratigraphic units were identified in eight of the ten boreholes drilled over areas in which SF_6 was not detected. Competent logs or stumps protrude through the gray silt and brown silt units in two of the 10 boreholes.

<u>Conclusions</u>

Preferentially permeable pathways that connect the upper aquifer to the ground surface through the unsaturated zone are amenable to detection using the described method. The probability of locating such pathways given a certain nodal spacing increases with the duration of injection within the 2500 square meter study area. Two identify two pathways in which mass can be transported preferentially at greater rates. The method is: (1) cost-effective, (2) timeefficient, (3) non-invasive, and (4) relatively easy to implement. The method should prove useful to evaluate subgrades at other existing or proposed waste disposal facilities.

<u>Acknowledgements</u>

The authors acknowledge the financial support of the United States Department of the Interior, Bureau of Mines. The project was funded under a Cooperative agreement between the Bureau of Mines and the

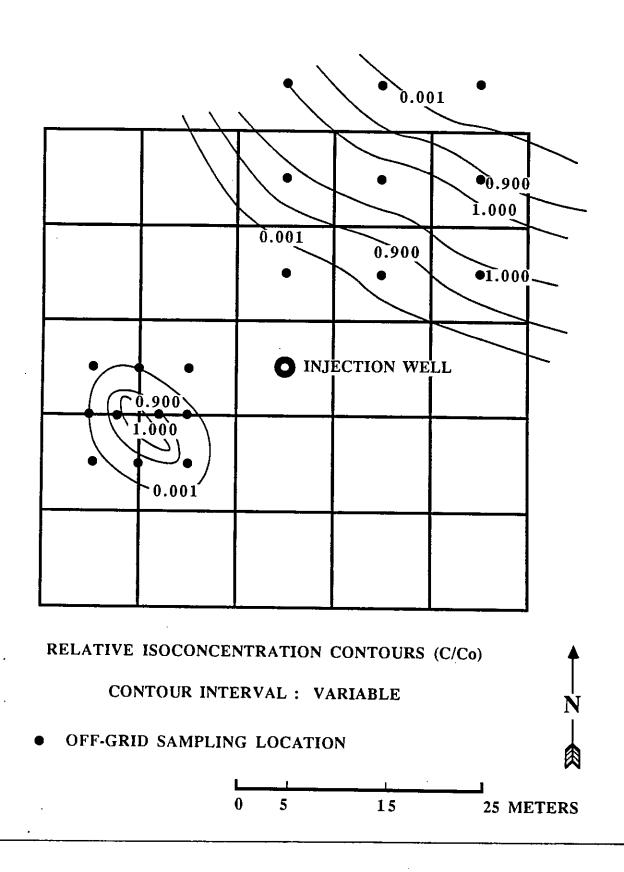
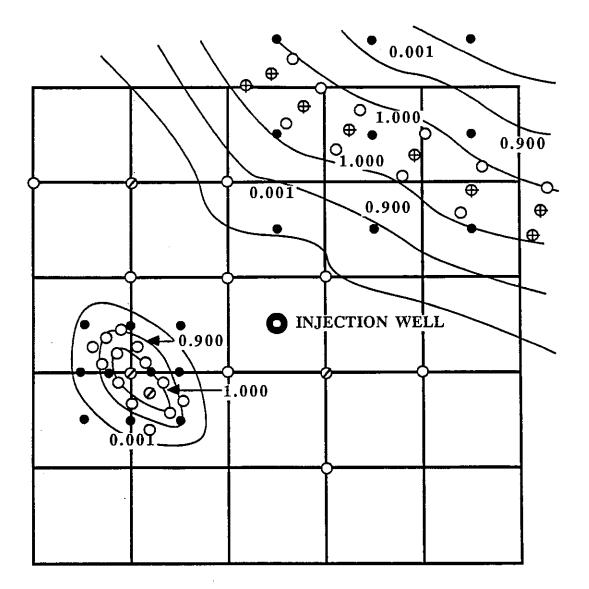
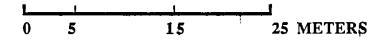


Figure 4. Results of Soil-Gas Survey No. 1 (SGS-1).



RELATIVE ISOCONCENTRATION CONTOURS (C/Co) CONTOUR INTERVAL : VARIABLE

- OFF-GRID SAMPLING LOCATION
- O BOREHOLE (ALL UNITS PRESENT)
- **⊕** BOREHOLE (GRAY AND RED SANDY SILTS ABSENT)
- **Ø** BOREHOLE (LOG OR STUMP)



次

Figure 5. Results of Soil-gas Survey No. 2 (SGS-2) and locations of boreholes used to confirm the the presence or absence of preferentially permeable pathways that connect the unsaturated portion of the gravel aquifer with the red sandy-silt unit.

University of Idaho (Agreement Number C0278001).

<u>References</u>

- Balfour, W. D., et. al., 1986, Measurement of Volatile Organic Emissions from Subsurface Contaminants, Radian Corporation (In-house Report), 20 p.
- Barrows, L. J., 1987, Field Calibration Tests of the Kerfoot Soil-Gas Survey Technique at Robins and Tinker Air Force Bases, U. S. EPA (TS-AMD-97580), 295 p.
- Bristow, Q, and Jonasson, R. I., 1972, Vapor Sensing for Mineral Exploration, Canadian Mineral Journal, 93, No. 5, pp. 39-44.

Chavez, J. E., 1986, The Relationship of Sulfur Gases Desorbed from Surface Soils to Massive Sulfide and Elemental Sulfur Mineralization at Depth, M.S. Thesis University of Idaho, 117 p.

- Dames and Moore 1990. Bunker Hill Site RI/FS Task 3.0: Final Hydrogeologic Assessment. U. S. EPA Pre-Issuance Copy. 263 p.
- Devitt, D. A., et. al., 1987, Soil Gas Sensing for Detection and Mapping of Volatile Organics, U. S. EPA Pre-Issuance Copy, 266 p.
- Farwell, S. O., et. al., 1984, Determination of Soil Adsorbed Sulfur-containing Gases for Geochemical Exploration: Preliminary Results, Abstract Presented at the 187th National ACS Meeting, St. Louis, Missouri.
- Hinkle, M. E., 1978, Collection and Analysis of Soil Gases Emanating From Buried Sulfide Mineralization, Johnson Camp Area, Cochise County, Arizona, Journal of Geochemical Exploration, 9, pp. 209 - 226.
- Kerfoot, H. B. and Barrows, L. J., 1987, Soil-Gas Measurement for the Detection

of Subsurface Organic Contamination, U. S. EPA Pre-Issuance Copy, 50 p.

- Kirschner, F. E., Jr., 1992. Delineation of Preferential Pathways of Contaminant Transport Within The Unsaturated Zone Using Soil-Gas. pp 3 - 44. <u>In</u> Evaluation of Waste and Water Contamination for Closure of a Hard Rock Mine Complex. December 31. 287 p. U.S. Bureau of Mines Cooperative Agreement No. C027801.
- Kirschner, F. E., Jr., 1991. Hydrogeological Evaluation of an In-situ Leach Cell in Abandoned Mine and Mill Wastes at the Kellogg, Idaho Superfund Site Using Tracers. Ph. D. Dissertation. University of Idaho. 262 p.
- Kirschner, F. E., Jr. and Williams, R. E. 1990. Ground Surface Delineation of Fractures Over Mined-Out Openings Using Carbon Dioxide Emissions, Groundwater, Vol. 8, No. 4, pp. 576 -582.
- Kirschner, F. E., Jr., 1989, Ground-Surface Delineation of Fractures that Appear to be Connected to Underling Mined-Out Openings Using a Naturally Occurring Gaseous Tracer, M.S. Thesis, University of Idaho, 103 p.
- Kirschner, F. E., Jr., 1989. Testing the Feasibility of In-Situ Leaching Heavy Metals from the Smelterville Flats Portion of the Kellogg Superfund Site. pp 3 - 44. In Evaluation of Waste and Water Contamination for Closure of a Hard Rock Mine Complex. October 30. 287 p. U.S. Bureau of Mines Cooperative Agreement No. C027801.
- Kunkel, D. in progress. M. S. Thesis, University of Idaho.
- La Brecque, et. al., 1985, Hydrocarbon Plume Detection at Stovepipe Wells, California, U.S EPA (TS-AMD-85402) 373 p.

- Marrin, D. L., 1987, Soil Gas Sampling Strategies: Deep vs Shallow Aquifers, NWWA Conference Entitled: "First National Outdoor Action Conference on Aquifer Restoration, Ground Water Monitoring and Geophysical Methods, pp. 437 - 454.
- Marrin, D. L. and Thompson G. M., 1984, Final project Report: Investigation of Volatile Contaminants in the Unsaturated Zone Above TCE Polluted Groundwater (Hughes Terminal -Phoenix Arizona), U. S. EPA (Proj. ID CR811018-01-0), 70 p.
- Norton, M. A., 1980. Hydrogeology and Potential Reclamation Procedures for an Uncontrolled Mine Waste Deposition Site, Kellogg, Idaho. M.S. Thesis. University of Idaho. 230 p.
- Philip, R. P. and Crisp, P. T., 1982, Surface Geochemical Methods Used For Oil and Gas Prospecting - A Review, Journal of Geochemical Exploration, 17, pp. 1 - 34.

http://dx.doi.org/10.1016/0375-6742(82)90017-6 Ralston, D. R. and Kunkel, D. 1989. Characterization of the Hydrostratigraphy of the Valley Alluvium and Tailings Near Smelterville Flats. pp. 45 - 77. <u>In</u> Evaluation of Waste and Water Contamination for Closure of a Hard Rock Mine Complex. June 30. 310 p. U.S. Bureau of Mines Cooperative Agreement No. C027801.

- Reisinger, H. J., et. al., 1987, Factors Affecting the Utility of Soil Vapor Assessment Data, NWWA Conference Entitled: "First National Outdoor Action Conference on Aquifer Restoration, Ground Water Monitoring and Geophysical Methods, pp. 425 - 435.
- Taylor, et. al., 1982, Sulfur Gases Produced by the Decomposition of Sulfide Minerals: Application to Geochemical Exploration, Journal of Geochemical, 17, pp. 165 - 185.

http://dx.doi.ora/10.1016/0375-6742(82)90001-2

Thompson, G. M. and Marrin, D. L., 1987, Soil Gas Contaminant Investigations: A Dynamic Approach, Ground Water Monitoring Review, Summer 1987, pp. 88 - 93.

 Voorhees, K. J., et. al., 1984, Analysis of Groundwater Contamination by a New Surface Static Trapping/Mass Spectrometry Technique, Analytical Chemistry, 56, pp. 2602 - 2604.
http://dx.doi.org/10.1021/ac00277a077 -2 **v** •