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A Comparison of Contaminant Migration from Waste Disposal Sites Located on Fractured, Porous Clay and Fractured, Porous Shale

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RACT

Contamination has occurred in both non-indurated and bedrock systems wherein the groundwater flows almost exclusively through a network of connected, open fractures. The matrix surrounding the fractures often possesses porosity which allows contaminant diffusion into the matrix. If the diffusion rates are fast, partition equilibrium between the fractures and the porous matrix will be present. Retardation effects may then be predicted by considering the system to be an equivalent porous medium (EPM). The rapidity with which fracture/immobile-matrix equilibrium is established will be determined in part by the: fracture aperture (2b); interfracture spacing (2B); porosity in the immobile matrix (θ_{im}); and the matrix diffusion coefficient (D'). Two systems which are characterized by very different values of the above parameters have been studied by our laboratories. At Alkali Lake, Oregon, the EPM approach describes contaminant transport well. At Bayview Park, Ontario, the EPM approach is not appropriate. Several features of the two sites are compared to illustrate the different natures of these two sites. These features include: 1) natural characteristics of the groundwater systems; 2) contaminant distributions; 3) observed retardation factors; and 4) computed fracture/immobile-matrix diffusion times.

RODUCTION

The method used for the prediction of contaminant migration pathways and rates will depend heavily upon the hydrogeological nature of the groundwater system under investigation. The vast majority of the information that currently exists pertains to non-indurated porous media where water and solutes move exclusively through pore channels between soil grains (Bear, 1972; Fried, 1975; Freeze and Cherry, 1979). In many instances, however, contamination has also occurred in systems wherein the groundwater flows almost exclusively through a permeable network of connected, open fractures (e.g., joints and bedding plane openings). Long flow paths can exist.

Both non-indurated as well as bedrock systems may be fractured. In both cases, the matrix material surrounding the fractures often possesses some porosity. Such porosity allows for the diffusive transport of contaminants from the fractures into the matrix due to the concentration gradient between the fracture and the matrix. This transport will take place both for compounds which sorb directly to the matrix material, as well as for non-sorbing compounds. The latter will diffuse into the matrix due to the presence of pore water in the matrix (Feenstra *et al.*, 1984; Johnson *et al.*, 1985).

The permeabilities of fractured systems are such that groundwater can move at rates from a few meters per year to hundreds of meters per year. When the interfracture spacings are small in comparison to the scale of the system being studied, and when there is some degree of interconnection between the fractures, good results can often be obtained in modeling the groundwater flow in such a system as an equivalent porous medium (EPM): Darcy's Law, pumping drawdown tests, and Hvorslev's interpretation of slug tests have all been applied successfully (Gerhart, 1984; Barker and Black, 1983). The modeling of flow in a large, complex cross-sectional system containing various till and ale units, some of them fractured, has been described by Frind and Matanga (1981).

Contaminant transport in such systems, however, is greatly complicated because the fracture systems are typically both complex and spatially variable. Therefore, the relative validity of the EPM approach for modeling contaminant transport is sometimes difficult to establish. Unfortunately, the variable number density, orientation, aperture, and continuity of fractures also make such systems too complex to model exactly. When the EPM approach cannot be used for transport modeling, current efforts are consequently limited to the simulation of transport in idealized systems of fractures. Within these restrictions, models have been developed which consider the effects of advection, dispersion, sorption, and molecular diffusion both for single and multiple fracture systems (Tang *et al*., 1980; Sudicky and Frind, 1984).

The processes of sorption and dispersion in fractured porous systems are more complicated than in simple porous systems because they are influenced by the rates of molecular diffusion into and out of the matrix. Indeed, as mentioned above, even non-sorbing compounds will partition to the matrix. If the rates of diffusion are slow, partition equilibrium between the fractures and the porous matrix will not be present, and retardation effects predicted on the basis of equilibrium partitioning of the solutes to the porous matrix (i.e., the EPM approach) will not be valid. This is entirely analogous to the situation of slow sorption/desorption kinetics in non-indurated porous media (e.g., as caused by slow diffusion within individual grains) when equilibrium between the sorbing grains and the intergranular groundwater is not established rapidly (e.g., Schwarzenbach and Westall, 1981).

The rapidity with which fracture/matrix equilibrium is established will be determined by the values of a number of hydrogeological parameters (Table I). They include: fracture aperture (2b); interfracture spacing (2B); porosity of the immobile matrix region (θ_{im}); and matrix diffusion coefficient (D′). Our laboratories have been involved in the study of two fractured porous systems which are characterized by very different values of the above parameters. In one case (Alkali Lake, Oregon), it may be argued that the EPM approach will work well in describing contaminant transport. In the other case (Bayview Park, Ontario) the EPM approach is not appropriate.

TWO WASTE DISPOSAL SITES ON FRACTURED POROUS MEDIA - A COMPARISON

The Alkali Lake, Oregon system has been studied by Pankow et al. (1984b), Johnson et al. (1985), and Johnson (1985). It is composed of a fractured porous soil. The soil components include alluvium, lake sediments, and eolian deposits. The depth to groundwater is in the range 0.6 to 1.0 m. A portion of the groundwater has been severely contaminated with a variety of chlorophenolic compounds from an inactive chemical waste disposal site. In the zone of contamination, the values of 2b and 2B are both small, and the values of θ_{im} and D′ are large (Table I). Such features increase the likelihood that the EPM approach will be valid.

The Bayview Park Landfill in Burlington, Ontario (also inactive) has been studied by Cherry et al. (1985), Hewetson and Cherry (1985), and Hewetson et al. (1985). It has caused groundwater contamination in the underlying fractured porous bedrock. The bedrock is red Paleozoic shale of the Queenston Formation, and it is essentially continuous to depths of over 100 m. The depth to groundwater is in the range 10 to 45 m. In the zone of contamination, the shale is characterized by large values of 2B, and small θ_{im} and D′ values (Table I). Such features will tend to preclude the application the EPM approach.

Alkali Lake Specific Conductance vs. Bayview Park Tritium.

Specific conductance distributions obtained at Alkali Lake in April 1983 are presented in Figure 1a. The disposal site itself does not contribute to the naturally-high salinity of the groundwater. To the east and hydraulically upgradient of the site are freshwater springs. The effect of these springs is to create a tongue of low conductance water which flows approximately co-axially through the site (Pankow et al., 1984). The high degree of regularity in the isopleths provides evidence for a reasonably uniform flow pattern through the site and into the downgradient region of this water table aquifer. This uniformity is a necessary but not sufficient condition for the argument that this system behaves as an EPM with respect to groundwater flow and solute (e.g., specific conductance) transport.

Tritium (^{3}H) is a constituent which may be used as an indicator of the input of post-1952 precipitation to a groundwater system (Freeze and Cherry, 1979). After 1952, tritium levels in precipitation have been continuously above the pre-1952 level. Water that recharged the groundwater zone prior to 1952 was originally characterized by 10-20 tritium units (TU) and now, as a result of radioactive decay, is below 3 TU. In typical porous medium systems, the input of post-1952 precipitation has generally lead to regular concentration isopleths since surface water infiltration and groundwater flow there are typically not restricted to narrow zones (e.g., Egboka et al., 1983). At Bayview Park, on the other hand, where the flow occurs in fractures in bedrock, the tritium patterns are highly erratic. Figure 1b presents tritium data for this site for the period 1983-1984 along with concurrent hydraulic head isopleths (Hewetson and Cherry, 1985). The B-B′ transect is perpendicular to the A-A′ transect at piezometer nest OW13. Water from level

1 of the multi-level WAT3 device (Cherry *et al.*, 1985) was found to contain 22 TU. At nearly the same level in the aquifer, the tritium content was only 4 TU at WAT4 but 45 TU at WAT5. Further downgradient at level 3 of OW12, the tritium content was 46 TU. At a shallower level at OW12, (where one might expect similar concentrations due to infiltration), the level was only 3 TU. Other erratic variations such as those found in monitoring devices OW13, OW16, and OW17 along transect B-B′ are also present. Such behavior indicates that 1) groundwater transport is occurring in restricted zones, and 2) equilibrium between the fractures and the matrix and from fracture to fracture is not present in this system at the meter scale.

<u>Alkali Lake Contaminant Distributions vs. Bayview Park Contaminant Distributions</u>.

The patterns exhibited by the concentration distributions of eight chlorophenolic contaminants present at Alkali Lake (Johnson *et al.*, 1985) reflect the same type of regularity found there for the specific conductance. Data for 2,6-dichlorophenol obtained in April 1984 are presented in Figure 2a. This compound has been shown to be non-sorbing in the Alkali Lake groundwater system (pH = 10) (Johnson *et al.* 1985). As was evidenced by the specific conductance isopleths (Figure 1a), groundwater flow occurs nearly coaxially through the site. As expected then, the concentration isopleths for 2,6-dichlorophenol indicate an approximately symmetrical zone of contamination coaxial with the site.

In contrast to this regularity, the organic contaminant distributions at Bayview Park contain numerous irregularities (Hewetson *et al.*, 1985). Concentration data obtained by Pankow *et al.* (1984a, 1985) in 1983 and 1984 for 1,1,1-trichloroethane (1,1,1-T) are presented in Figure 2b. As an example of the irregularities, at level 3 of piezometer nest OW1, the 1,1,1-T value was µg/l, and only 1.8 at approximately the same level at nearby WAT3. Additional irregularities become evident when the organic contaminant data are compared to the tritium data. Since tritium input to the system began in 1953, and since the landfill was operated between 1961 and 1974, one would expect tritium everywhere that compounds such as 1,1,1-T were found. That this is not the case may be seen by comparing the relatively high levels of 1,1,1-T at level 3 of nest OW1, and levels of 2 and 3 of nest OW13 with the low tritium levels at the same monitoring points. One possible explanation for this behavior would be to invoke spillage of 1,1,1-T at the quarry which preceded both 1) the landfill, and 2) the input of tritium to the system. However, if such spillage had occurred, it remains difficult to understand how a presumably limited number of isolated spillage points could have lead to such widespread 1,1,1-T contamination. Another possible explanation is that tritium has a larger effective diffusion coefficient in the shale matrix than does 1,1,1-T, and therefore that tritium is more retarded by the matrix diffusion effect than is 1,1,1-T (Hewetson *et al.*, 1985).

Alkali Lake Retardation vs. Bayview Park Retardation.

For the transport of a non-sorbing (ns) tracer or contaminant in a given fractured porous system which is behaving as an EPM, the retardation of such contaminants relative to the *actual groundwater velocity* in the fractures (v_f) may be calculated. When the fractures through which groundwater movement is occurring are totally void, that retardation factor will be given by (Johnson *et al.*, 1985)

$$R_{ns} = 1 + \theta_{im} B/b \qquad (1)$$

where the various parameters are as defined above. The EPM velocity of a non-sorbing solute is then given as

$$v_{EPM} = v_f/R_{ns} \qquad (2)$$

At Alkali Lake, $R_{ns} \approx 14$. The fracture groundwater velocity there has been calculated to be ~1 m/day (Johnson, 1985). Therefore, at Alkali Lake, v_{EPM} = ~0.07 m/day. Since the major input of contaminants to the groundwater occurred in November 1976, ~2700 days (7.3 years) had elapsed by the time the Figure 2a data were obtained. Based on the contaminant transport distances reported in Johnson et al. (1985), an approximate "contamination front" velocity (V_{cf}) of ~0.09 m/day may be obtained. A tracer velocity (v_{tr}) value of 0.06 m/day was observed in tests at the site (Johnson, 1985). These values agree remarkably closely with the 0.07 m/day estimated for v_{EPM}.

At Bayview Park, by Equation 1 and based on the range of parameter values observed at the site by Hewetson and Cherry (1985), R_{ns} may be calculated to range between 50 and 5,000. With v_f values at the site in the range 0.1 - 1.0 m/day, the range of v_{EPM} values possible is 2×10^{-4} - 2×10^{-3} m/day. The value of v_{cf} at the site may be approximated as done above for Alkali Lake. The landfill was opened 23 years prior to the aquisition of the data presented in Figure 2b. Since 1,1,1-T was found as far as 900 m downgradient of the landfill, discounting the possibility of excessively large dispersion coefficients, a minimum non-sorbing v_{cf} value of 0.1 m/day may be calculated (1,1,1-T may be considered essentially non-sorbing in this f_{oc} = 0.001 system.) Such a v_{cf} value can only be explained in terms of non-equilibrium (or "non-EPM") solute transport through the system's fractures.

Alkali Lake Diffusion Times vs. Bayview Park Diffusion Times.

As noted above, the EPM approach will be valid if diffusion equilibrium can be reached rapidly between the fractures and the matrix. On the basis of the foregoing statements and data, it may be expected that relative to v_f, the

...es required to achieve fracture/matrix equilibrium are short at Alkali Lake, and long at Bayview Park.

Figures 3a (Alkali Lake) and 3b (Bayview Park) present the results of calculations in which a non-sorbing species is diffusing from two parallel fractures into a slab of matrix material between the fractures. The mathematical solution used here was analytical in nature (Crank, 1975). The concentration in the fractures C_0 was assumed to stay constant along the length of the fractures. (An alternate approach would have been to allow the fracture concentration to drop due to mass loss to the matrix. The trends in the results would have been the same.) The data are presented in a normalized manner as C_{im}/C_0, where C_{im} is the concentration of the diffusing species in the immobile matrix pore water. The parameter values used for Alkali Lake were 2B = 0.3 cm, and D′ = 0.1 cm^2/day. The 2B value was obtained by the examination of core samples taken at the site, and the D′ value was the best fit value obtained from push-pull tracer tests there (Johnson, 1985). The values used for Bayview Park were: 2B = 10 cm; and D′ = 0.0032 cm^2/day. As at Alkali Lake, the 2B value was obtained by examination of core samples (Hewetson and Cherry, 1985). The D′ value was obtained as the average of experimental D′ results for chloride (non-sorbing) for three samples of shale at 22^{o}C (Cherry, 1985), corrected to the mean annual temperature of the Bayview Park groundwater system (12^{o}C). (The small value of θ_{im} at Bayview Park accounts for the relatively low D′ value there.) The Figure 3 results for both sites are independent of 2b since the fracture concentration was assumed to stay constant at C_0.

The time period required to achieve C_{im}/C_0 = 0.5 at the center of the matrix slab is very large (~5000 days, or 13.7 years) in the case of the Bayview Park system. In contrast, at Alkali Lake, C_{im}/C_0 = 0.8 is obtained at the center of the matrix slab in only 4 hours. These equilibration times may e compared with fracture groundwater velocities which are nearly the same at the two sites: at Alkali Lake, $v_f \approx 1$ m/day, and at Bayview Park, $v_f \approx 0.35$ m/day. Thus, the possibility of the achievement of equilibrium between the matrix and the flowing groundwater is much greater at Alkali Lake than at Bayview Park. When taken in conjunction with a good deal of fracture connectivity at Alkali Lake, it is understandable why the EPM approach appears to work at Alkali Lake both with respect to concentration isopleth regularity and solute retardation. Conversely, the erratic nature of the data obtained at Bayview Park and the long transport distances there may be understood in terms of long equilibration times, and in all likelihood, a system of fractures which is not nearly as well connected as at Alkali Lake.

Although the diffusion equilibration times are long in the shale at Bayview Park, the length of time that leachate has been entering the shale is sufficiently long that a considerable fraction of the mass of contaminants may be expected to have entered the shale matrix between the fractures. Indeed, since the matrix porosity there is approximately 5%, a large voids volume is available for the storage of contaminants.

CONCLUSIONS

The suitability of a given geological formation to serve as a site for the disposal of wastes must be considered in terms of the possible presence of fractures. It is very probable that the original disposal of wastes at both Alkali Lake and Bayview Park would have received more careful consideration had the fracture systems at these two sites been appreciated earlier. Now that contamination does exist at these and other sites located on fractured media, however, there is a growing need to 1) understand the processes governing the transport of contaminants in such systems, and 2) develop propriate models to describe such transport. In cases such as Alkali Lake, the EPM approach provides a good description of the transport of contaminants. At Bayview Park, the EPM approach is not applicable.

Through the examination of the values of parameters such as $2B$, θ_{im}, D', and v_f, together with calculations of the type presented in Figure 3, an initial evaluation of the applicability of the EPM approach may be obtained. It is important to note, however, that the mere applicability of the EPM approach in a given system does not guarantee that transport modeling in that system will be straightforward. Indeed, problems such as incomplete input data regarding heterogeneities and anisotropies in the hydraulic conductivity and in the transport characteristics of the fractures can greatly frustrate attempts at transport simulations. Nevertheless, regardless of such ancillary complications, if a system is behaving as an EPM, then the realization of that fact will be an important step towards the understanding of solute transport in that system.

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REFERENCES

Barker, J. A., and J. H. Black, 1983, "Slug Tests in Fissured Aquifers", Water Resources Research, 19, 1558-1564.

Bear, J., 1972, Dynamics of Fluids in Porous Media, American Elsevier, N.Y.

Cherry, J. A., P. E. Johnson, R. J. Blackport, J. P. Hewetson and R. D. Blair, 1985, "Development and Applications of a Multilevel Device for Groundwater Monitoring in Fractured Rock", in press The Canadian Geotechnical Journal.

Cherry, J. A., 1985, unpublished results, University of Waterloo.

Crank, J., 1975, The Mathematics of Diffusion, Second Edition, Clarendon Press, Oxford, 414 pp.

Egboka, B. C. E., J. A. Cherry, R. N. Farvolden, and E. O. Frind, 1983, "Migration of Contaminants in Groundwater at a Landfill: a Case Study, 3. Tritium as an Indicator of Dispersion and Recharge", J. Hydrology, 63, 51-79.

Feenstra, S., J. A. Cherry, E. A. Sudicky, and Z. Haq, 1984, "Matrix Diffusion Effects on Contaminant Migration from an Injection Well in Fractured Sandstone", Ground Water, 22, 307-316.

Freeze, R. A., and J. A. Cherry, 1979, Groundwater, Prentice-Hall, Englewood Cliffs, N.J.

Fried, J. J., 1975, Groundwater Pollution, Elsevier, Amsterdam.

Frind, E. O., and G. Matanga, 1981, "Ground Water Flow and Salt Transport in Irrigated Land in the Bow River District, Alberta: A Simulation Study", Report Prepared for the Alberta Agriculture, University of Waterloo Institute for Groundwater Research.

Gerhart, J. M., 1984, "A Model of Regional Ground-Water Flow in Secondary-Permeability Terrane", Ground Water, 22, 168-175.

Hewetson, J. P., and J. A. Cherry, 1985, "An Investigation of the Groundwater Zone in Fractured Shale at a Landfill. 1. Groundwater Flow and Tritium Occurence", submitted J. Hydrology.

Hewetson, J. P., J. A. Cherry, and J. F. Pankow, 1985, "An Investigation of the Groundwater Zone in Fractured Shale at a Landfill. 2. Contaminant Distribution and Migration", submitted, J. Hydrology.

Johnson, R. L., S. M. Brillante, L. M. Isabelle, J. E. Houck, and J. F. Pankow, 1985, "Migration of Chlorophenolic Compounds Stored at the Chemical Waste Disposal Site at Alkali Lake, Oregon. 2. Contaminant Distributions, Transport, and Retardation", Ground Water, 23, in press.

Johnson, R. L., 1985, "The Groundwater Transport of Chlorophenolics through a Highly Fractured Soil at Alkali Lake, Oregon", Ph.D. thesis, Oregon Graduate Center, Beaverton, Oregon.

ankow, J. F., L. M. Isabelle, J. P. Hewetson, and J.A. Cherry, 1984a, "A Syringe and Cartridge Method for Down-hole Sampling of Trace Organics in Ground Water", Ground Water, 22, 330-339.

Pankow, J. F., R. L. Johnson, J. E. Houck, S. M. Brillante, and W. Jerry Bryan, 1984b, "Migration of Chlorophenolic Compounds Stored at the Chemical Waste Disposal Site at Alkali Lake, Oregon. 1. Site Description and Ground Water Flow", Ground Water, 22, 593-601.

Pankow, J. F., L. M. Isabelle, J. P. Hewetson, and J.A. Cherry, 1985, "A Tube and Cartridge Method for Down-hole Sampling for Trace Organics in Groundwater", in press, Ground Water.

Schwarzenbach, R. P., and J. Westall, 1981, "Transport of Nonpolar Organic Compounds from Surface Water to Groundwater. Laboratory Sorption Studies", Environ. Sci. Technol., 15, 1360-1367.

Sudicky, E. A., and E. O. Frind, 1984, "Contaminant Transport in Fractured Porous Media. Analytical Solutions for a System of Parallel Fractures", Water Resources Research, 18, 1634-1642.

Tang, D. H., E. O. Frind, and E. A. Sudicky, 1980, "Contaminant Transport in Fractured Porous Media, 1. Analytical Solution for a Single Fracture", Water Resources Research, 17, 555-564.

ABLE I. Properties of the Alkali Lake and Bayview Park Groundwater Systems.

PARAMETER	SYMBOL	ALKALI LAKE[a]	BAYVIEW PARK[b]
fracture aperture	2b	150 um	1 - 50 um
interfracture spacing	2B	0.3 cm	5 - 35 cm
matrix porosity	θ_{im}	0.65	0.05
matrix diffusion coefficient	D′	0.1 cm^2/day	0.002 cm^2/day
matrix retardation factor	R_{ns}	14.	50 - 5000[c]
calculated fracture groundwater velocity	v_f	1 m/day	0.1 - 1.0 m/day[c]
EPM groundwater velocity	v_{EPM}	0.07 m/day	2.0×10^{-4} - 2.0 x 10-3 m/dayc
contaminant front velocity	v_{cf}	0.09 m/day	0.1 m/day
tracer velocity	v_{tr}	0.06 m/day	NA[d]
fraction organic carbon (matrix)	f_{oc}	0.018	0.001
hydraulic gradient	dh/dl	0.001	0.05

[a] Johnson (1985) and Pankow _et al._ (1984).

[b] Hewetson _et al._ (1985) unless otherwise noted.

[c] Calculated based on data from Hewetson _et al._ (1985).

[d] NA = not available.

LIST OF FIGURES