The challenge of interpreting environmental tracer concentrations in fractured rock and carbonate aquifers

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Atmospheric environmental tracers are constituents and dissolved gases (e.g., chlorofluorocarbons, isotopes of hydrogen, helium, carbon, and oxygen) that are entrained in precipitation and recharged to groundwater. Often the time-varying atmospheric concentrations or processes affecting their evolution in groundwater are used with simplified conceptual models of groundwater flow to estimate residence times, which can then be used to infer velocity, recharge, and properties affecting chemical transport. For example, translating concentrations to residence time (measured in years) is often conducted using concepts of plug flow, binary mixtures of groundwater, and simple models of recharge and flow (Cook and Herczeg 2000). Successful interpretations of environmental tracer concentrations have been widely reported for unconsolidated, porous-media aquifers, and residence times have been introduced as calibration targets using particle-tracking methods in regional flow models (Sanford et al. 2004).

To a lesser extent, interpretations of environmental-tracer concentrations have been applied to fractured-rock aquifers and carbonate formations that have been subject to karstification (Cook and Simmons 2000; Long and Putnam 2006). Such aquifers offer a degree of geologic complexity that exceeds that of porous-media aquifers. Consequently, methods of interpreting environmental-tracer concentrations commonly applied in porous media may not be applicable to fractured rock and carbonate aquifers (herein collectively referred to as fractured rock).

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The purpose of this article is to highlight those processes that affect environmental tracers in fractured rock, identify deficiencies in applying interpretive methods commonly used in porous media, and suggest alternative approaches for interpreting environmental-tracer concentrations to better understand the regional hydrogeology of fractured-rock aquifers.

Challenges in fractured rock

In fractured rock, significant challenges in interpreting environmental tracer concentrations arise because of geologic complexities that result in abrupt spatial changes in hydraulic properties. Fractures, joints, conduits, and vugs are the principal pathways for flow and have hydraulic properties that vary over orders of magnitude. The complex connectivity of these features yields highly convoluted flow paths over distances from meters to kilometers, where velocities can also vary over orders of magnitude.

Complexities of interpretation also arise because of the topology of individual permeable features. Flow within an individual fracture yields preferential flow paths and areas subject to slow advection or stagnant water. The volume of fractures occupied by preferential flow paths can be significantly less than the total fracture volume. In addition, fractured rock is characterized by a primary porosity of the rock matrix. Crystalline rocks are reported to have matrix porosity as large as 3%, and some sedimentary rocks are reported to have larger matrix porosity. The rock matrix is not significant in contributing to groundwater flow; however, it offers a fluid-filled void space in contact with fractures where constituents can diffuse from fractures to the matrix, or vice versa, depending on the direction of the chemical gradient (Grisak and Pickens 1980).

These hydrogeologic complexities are not always consistent with methods of interpreting environmental-tracer concentrations that have been successfully applied in porous media. For example, a plug-flow interpretation assumes the concentration in a parcel of water is unchanged as it is advected. Thus, residence times are estimated by comparing concentrations from groundwater samples to records of concentrations in recharge. Interpretations using plug flow, binary mixtures of ground-

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water, and idealized spatial distributions of recharge and flow are based on simplified aquifer conditions. Nevertheless, such interpretations have been applied to fractured rock. The following sections outline complexities that arise in interpreting environmental-tracer concentrations in fractured rock using conceptually simple interpretations of the flow regime. Cook et al. (2006) give a thorough discussion of the physical, chemical, and biological interferences that affect many environmental tracers.

Diffusion, advection, dispersion

The diffusive exchange to or from permeable features alters the concentration of environmental tracers. Consequently, interpretations of residence times based on advection, or even advection and dispersion, will be flawed. Diffusion of environmental tracers into the rock matrix can lead to underestimating the groundwater velocity by orders of magnitude (Neretnieks 1981). The effect of chemical diffusion, however, will depend on the velocity and properties of the rock matrix.

Natural variability in geologic materials leads to variability in the velocity. The common assumption in the modeling of chemical transport is that the mean velocity controls chemical advection, and the variability about the mean velocity is incorporated into dispersion. The effect of dispersion on the interpretation of environmental tracer concentrations has been quantified in hypothetical situations and field settings (Ekwurzel et al. 1994; Reilly et al. 1994). In a coastal plain aquifer, Reilly et al. (1994) showed a negligible effect of dispersion on the concentrations of environmental tracers in comparison to plug-flow interpretations over flow paths of several kilometers. In their investigation, collecting samples from short screened intervals of monitoring wells reduced the mixing of groundwater from multiple flow paths and the effect of dispersion; however, the complexity of the geologic setting will also play a role in the magnitude of dispersion. In fractured rock, velocities are expected to vary over many orders of magnitude, and variability is anticipated to be more extreme than that encountered in porous media. Therefore, the influence of dispersion on environmental tracer concentrations garnered from experiences in porous media will not necessarily be representative of conditions in fractured rock.

In addition, fractured rock is characterized by flow regimes where a few preferential flow paths can control the majority of groundwater flow. In contrast to porous media, the void volume of the most permeable features constitutes only a small percentage of the total void volume of the aquifer. The large variability in the velocity and the retention of a large percentage of the groundwater in low-permeability features provides the capacity for mixing of groundwater with dramatically different velocities. Interpretations of environmental-tracer concentrations that are based on plug flow or a binary mixture of velocities are unlikely to represent the tracer concentration that has migrated through convoluted flow paths associated with multiple fractures.

The variability of the velocity that is anticipated in fractured rock may prompt one to adopt an advection-dispersion model such as the direct simulation of residence times (see, for example, Goode 1996). Recent investigations have raised concerns regarding the applicability of the advection-dispersion equation in geologic settings that are characterized by velocity spanning many orders of magnitude (Shapiro 2001; Shapiro et al. 2008). Fractured rock is characterized by void space where the representative length dimension of the void space exceeds its width or height. This is unlike the void space of porous media, where the characteristic length and width are of similar dimensions, offering a tortuous flow path that restricts the spatial persistence of the velocity at the pore scale. The tortuous flow path in porous media exposes a chemical constituent to a wide range of pore-scale velocities over a short distance, which is a condition for applying the advection-dispersion equation with a constant dispersion coefficient. In contrast, preferential flow paths in fractured rock have void dimensions that will result in spatial persistence in the velocity, and the conditions for the application of the advection-dispersion equation may not be met over tens of meters or more.

Groundwater-flow models

Although the complexity of flow in fractured rock is acknowledged, regional models over dimensions of hundreds of meters to kilometers can be developed. These models are based on a water balance, where spatially distributed bulk properties are estimated from recharge, discharge, and measurements of hydraulic head. Only those geologic features that are hydraulically significant over regional dimensions are incorporated into the model. Bulk properties consider the aggregated effect of fractures, but do not account for individual preferential flow paths. Thus, models that are based on a water balance are unlikely to capture the range and complexity of the velocity for the purpose of interpreting environmental-tracer concentrations. The uncertain location of preferential flow paths and the computational expense of simulating large numbers of fractures preclude the construction of regional models that explicitly consider realistic numbers of fractures.

Even if one accepts deficiencies in a flow model based on a water balance, there are additional concerns that arise in using residence times interpreted from environmental-tracer concentrations in refining the flow model. Using particle-tracking methods, porosity can be adjusted to obtain favorable comparisons between modeled advective residence times and residence times estimated from simplified interpretations of environmental-tracer concentrations. The comparison between computed residence times from a flow model and the interpreted residence times using environmental tracers constitutes a comparison between two models. Comparing results from two models that are based on different assumptions may not necessarily lead to a better understanding of the hydrogeology. It is possible that physically unreasonable

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estimates of aquifer properties may be achieved in this calibration procedure, because processes affecting the evolution of the tracers are not captured in either model.

Interpreting environmental-tracer concentrations in fractured rock

In geologic settings where velocity can vary over orders of magnitude and diffusion is significant for chemical constituents, the application of methods of interpreting environmental-tracer concentrations in understanding hydrogeology must move away from inferring residence times, and instead move toward the use of the underlying concentration data as a calibration target. The interpretation of environmental-tracer concentrations should be regarded similarly to controlled tracer experiments, where the tracer concentration is directly simulated. This approach, while alleviating uncertainties associated with residence times, is fraught with its own complexities; modeling of chemical transport in complex geologic settings cannot be achieved without some degree of conceptual complexity.

Because regional flow models do not represent intricacies of the velocity and classical theories of dispersion need to be revisited in complex geologic settings, new mathematical formulations have been proposed to capture the effect of velocity variability, dispersion, and diffusion (Berkowitz et al. 2001; Cvetkovic and Haggerty 2002). These models attempt to capture the aquifer complexity, but do not necessarily rely on the underlying aquifer properties. Instead, the velocity is characterized by a probability density function, which may require the use of a chemical-transport experiment in identifying its parameters.

Although stochastic methods of incorporating the extreme variability of the velocity are promising for interpretations of environmental-tracer concentrations, their implementation in conjunction with geologic complexities needs to be realized. Modeling concentrations of environmental tracers must be cognizant of complexities of the regional-flow regime by incorporating the spatial complexity of recharge. In addition, regional geologic features that constrain the flow regime also need to be incorporated, either deterministically or with some degree of uncertainty. Thus, the basic geologic framework that is instrumental in developing a water balance should not be discarded in attempting to characterize the complex velocity.

Other opportunities in the direct simulation of environmental tracers may also be realized with computationally efficient algorithms that incorporate large numbers of discrete features over regional dimensions. These models need to incorporate flow and chemical transport in both permeable features and the rock matrix. Geologically sound conceptual models that characterize the complex topology of permeable features need to serve as the underpinnings for such models. Invariably, there will be tradeoffs between the computational efficiency and the detail incorporated in characterizing permeable features and the rock matrix.

Other evidence of chemical transport under ambient flow conditions can constrain interpretations of environmental-tracer concentrations. There is experience in implementing controlled tracer tests in carbonate aquifers over distances consistent with the interpretation of environmental-tracer concentrations. Fluorescent dyes have been used to trace the water from recharge locations to points of groundwater discharge. Although many of these controlled tests are conducted qualitatively to map connections between recharge and discharge locations, quantitative tests have also been conducted where the mass of the injected tracer and its time-varying concentration at one or more points of groundwater discharge are monitored (Field and Pinsky 2000).

The resultant signal from quantitative tracer tests in carbonate aquifers is observed over days to months, where there may be an initial rapid breakthrough of the tracer and a peak concentration (in hours to days), followed by an elongated declining limb of the breakthrough curve over weeks to months. Eventually, the concentration of the tracer falls below detection limits. Even though the residence times of environmental tracers are measured in years, there is not necessarily an inconsistency between the interpretation of the environmental-tracer concentrations and the results of controlled tracer tests. The controlled tracer test accentuates the fastest components of the velocity. The dilution experienced from recharge to discharge locations in the controlled test indicates contributions from other recharge locations and less permeable fractures. The fact that there is an elongated declining limb in breakthrough curves of controlled tracer tests, indicates that the tracer is migrating through flow paths having slower velocity than the most permeable features. Because the concentration of the tracer in the controlled experiment eventually falls below detection limits, the controlled test is unable to resolve the entire spectrum of velocities in the flow regime. The longevity associated with environmental tracers in aquifers offers an opportunity to resolve the spectrum of velocities when used in concert with the results of controlled tests.

In many fractured-rock aquifers, it becomes difficult to conduct controlled tracer experiments under ambient flow conditions. Unlike many carbonate aquifers, rates of groundwater flow in other geologic settings may be too slow to monitor tracer arrival at points of groundwater discharge. In addition, there may not be focused areas of groundwater discharge, making it difficult to design monitoring locations. Complex fracture connectivity makes it difficult to design borehole locations to capture injected tracers under ambient conditions, as tracers may bypass monitoring boreholes.

Controlled tracer experiments have been successfully conducted in most types of fractured rock under hydraulically stressed conditions, where tracers are injected in boreholes and recovered by pumping at other boreholes. Such tests are usually conducted over relatively short distances (tens of meters) and can be interpreted to

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estimate the effective fracture porosity. The porosity can then be applied to estimate the ambient velocity using the hydraulic conductivity and the ambient hydraulic gradient. In general, however, there can be errors in estimates of the hydraulic conductivity and the hydraulic gradient, which may lead to unreliable estimates of the ambient velocity.

In instances where controlled tracer tests under ambient conditions cannot be conducted, point measurements of the ambient velocity may provide insight into the range of the groundwater velocity affecting chemical migration. Point-dilution and single-hole push-pull tests involve the injection of a tracer into a single borehole and the subsequent monitoring of the concentration in that borehole. Interpretations of such tests provide estimates of the velocity in the vicinity of the borehole; however, they need to be conducted using equipment to isolate individual or closely spaced fractures so an accurate picture of the velocity is obtained in individual permeable features. A number of single-well tracer tests are needed in permeable features of varying hydraulic conductivity, as well as at various depths and over an area that is representative of the regional flow. Data sets comprising the distribution of the velocity would need to be integrated with geologic conceptual models from which one can infer the spatial persistence of the velocity over regional dimensions and chemical retention characteristics that arise due to diffusion (Frampton and Cvetkovic 2007).

Conclusions

Interpretations of environmental-tracer concentrations offer opportunities to characterize flow and transport in aquifers over time frames and distances that cannot be realized using conventional hydrologic testing. While there has been success in interpreting environmental tracers in porous media, interpretations in fractured rock have faced many challenges. Residence times of years to tens of years are frequently interpreted from environmental tracers in fractured rock using simplified interpretive methods. In many fractured-rock aquifers, controlled tracer tests indicate that groundwater velocities are capable of transporting constituents significant distances in hours, days, and months. This perceived inconsistency between the interpretation of environmental-tracer concentrations and controlled tracer tests is a result of the heterogeneous nature of fractured rock and the inability of simple interpretations to capture the velocity variability encountered in complex geologic settings.

The use of environmental tracers in characterizing fractured rock over regional dimensions needs to move away from simple interpretations of residence times. The residence time inferred from the concentrations of environmental tracers using plug flow and binary mixing models are inconsistent with the complexities of fractured rock. Concentrations of environmental tracers should be interpreted similarly to controlled tracer tests, where processes affecting the tracers are incorporated into the interpretation. Methods that accurately capture the extreme variability in the velocity need to be introduced into these interpretations; however, flow models based on a regional water balance are unlikely to capture the variability in the groundwater velocity. Other data sources that constrain the hydrogeologic conceptual model and chemical transport over regional dimensions should be introduced into interpretations of environmental-tracer concentrations. These data could include controlled tracer tests such as quantitative dye tracing commonly conducted in karst formations, and experiments that explicitly characterize the range of the groundwater velocity through point measurements.

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