Development of a New Technique for the Use of Dissolved Helium as an Environmental Groundwater Tracer

Problem and Research Objectives

Tracer tests are an important method for determining the flow characteristics and patterns of subsurface water (such as groundwater aquifers) and surface water bodies (such as streams and the ocean). In such tests, a constituent is added to the water. The constituent is either non-native to that water or high enough in concentration to be distinguished from the native component in that water. Many of the commonly used tracers have one or more limitations, such as being toxic, being esthetically objectionable, being difficult to be differentiated from the naturally occurring fraction, requiring complex laboratory analysis for their detection and quantification, requiring a high concentration, and reacting with the aquifer matrix. These limitations are not common to helium, however. Thus, if a field method can be developed to detect and quantify the concentration of dissolved helium in surface or groundwater in real time, then helium has great potential for use in certain tracer applications, including its use near drinking water sources or in environmentally sensitive areas such as wetlands and fish farms, its use in areas where esthetics are a concern (e.g., recreational beaches), and its use in conjunction with investigations of the diffusion characteristics of groundwater aquifers.

Helium was successfully tested as a reliable and economical tracer (S.K. Gupta, L.S. Lau, P.S. Moravcik, and A.I. El-Kadi, 1991, Injected Helium: A New Hydrological Tracer, Special Report 06.01.90, Water Resources Research Center, University of Hawaii at Manoa, 94 pp.; L.S. Lau, and P.S. Moravcik, 1994, Ground-water tracing with injected helium, *Ground Water* 32(1):96–102). Its behavior in open tanks was identical to that of fluorescein dye. Although the system was simple and specific for helium, the small surface area and fragility of the thin quartz window limited its sensitivity and reliability. Moreover, the technique was only suitable for the analysis of discrete samples.

The objective of this project is to develop and demonstrate a new analytical system that substantially improves the precision and utility of the helium tracer for routine use in surface water and groundwater. The work proposed encompasses developing and calibrating the system, and testing the helium tracer in the laboratory against a commonly used tracer.

Methodology

Instrumentation

The new system consists of a gas extraction system and a separate helium analyzer (Figure 1). The design allows on-site, continuous, real-time monitoring in a completely automated structure. The results of the analysis are automatically recorded on a lap-top computer.

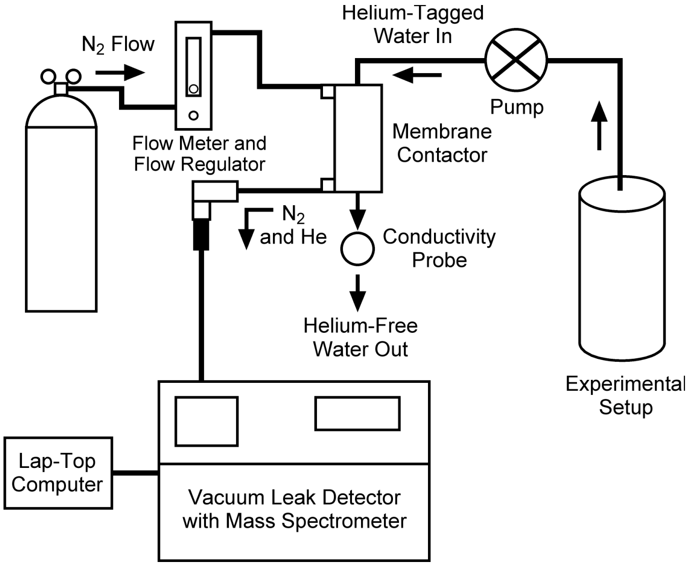

Figure 1. Schematic of the helium tracer system

Our system is built around a permeable membrane contactor, which is used commercially to extract dissolved gases from liquids. This contactor is a shell-and-tube device containing thousands of microporous hydrophobic polypropylene hollow fibers woven into a fabric array. To extract dissolved helium, a steady flow of helium-tagged water through the wet side of the contactor is maintained. A constant flow of clean, dry nitrogen through the gas side of the contactor provides a helium concentration gradient and results in the diffusion of helium from water to the gas side of the membrane contactor. Our design includes the use of the 6.35 cm × 20.32 cm Extra-Flow Membrane from Liqui-Cel (Charlotte, NC). Nitrogen gas is moved inside the hollow fibers at a specified pressure. The rate of helium extraction depends on the partial pressure difference and the flow rate of water. Through calibration, a trial-and-error approach is used to estimate a certain combination of the flow rate and pressure. For example, at 13.8 N/s^2 the optimal flow rate is 10 cm^3/s.

A vacuum leak detector probe is placed in the discharge path from the gas side of the membrane contactor. The leak detector has a helium-based mass spectrometer which generates an electrical signal that is proportional to the helium concentration in the gas discharge from the membrane contactor. This electrical signal is displayed as a leak rate.

A Veeco MS-40 Portable Leak Detector manufactured by Vacuum Instrument Corporation (Plainview, NY) was used in our design. The MS-40 detector is equipped with software that allows communication with a computer to automatically record output signals. The test port of the MS-40 detector is connected to the gas outlet of the extraction system by polytetrafluoroethylene tubing, which is permeable in regard to the small helium molecules. To prevent water vapor from entering the mass spectrometer's high vacuum chamber, a 33.02 cm × 2.54 cm PVC cylinder filled with Drierite desiccants (0.64-cm granules) is placed in the flow line between the extraction system outlet port and the mass spectrometer's inlet port.

Instrument Calibration

The instrument was calibrated to convert signals from the helium detector to helium concentrations. Solutions of known concentrations were prepared based on the solubility of helium in equilibrium with atmospheric air at sea level (i.e., 5.0×10^{-5} mg/l) and the maximum solubility of helium at standard atmospheric pressure (i.e., 8 mg/l). Figure 2 shows the relationship between the concentration based on the helium signal recorded by the mass spectrometer and the known helium concentrations in the five helium solutions. A near-linear relationship exists between the two helium concentrations with a slope of the fitting line of 0.95. A potential error in results is likely due to the inability of the contactor membrane to extract all the helium gas from the solution.

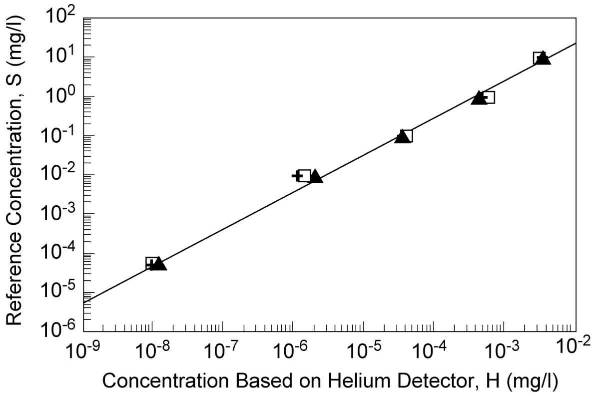

Figure 2. Instrument calibration curve using five different helium solutions. Three replicas (shown by different symbols) were tested for each solution. The solid line represents the least square best fit.

Principal Findings and Significance

Pipe Experiments

Water was pumped from a reservoir through a 6.1-m-long PVC pipe with a 5.08 cm diameter at a constant flow rate of 18.33 cm^3/s. Two 3.05-m pipes were connected by two 90-degree elbows to fit the experimental setup into the laboratory. An He/NaCl solution was injected into the pipes through a 0.64-cm inlet at the same constant flow rate of 18.33 cm^3/s for 2 minutes. After the tracer injection, pumping from the water reservoir resumed for the remainder of the experiment. The 0.64-cm PVC pipe outlet was connected to the helium extraction and detection unit by 0.64-cm flexible PVC tubing. At the outlet of this unit the conductivity was measured by a probe. The relative He and NaCl concentrations were continuously recorded throughout the experiments.

The helium concentration results are compared against NaCl results in Figure 3 for one experiment. The success of the method is evident by the instantaneous response of the detection system to changes in solute concentration. Excellent match can be seen regarding the breakthrough curve, the first arrival, the slope of the curve, and the value of the relative peak concentration. However, the location of the peak in Figure 3 is slightly ahead for helium, and fluctuations are evident in the elution of helium. The resolution of the helium detector is a limiting factor in the accuracy of the readings, as is evident by the horizontal segments in the elution curve. The results were sensitive to a number of factors, including the helium saturation level in the injected water. Signals from the detector fluctuate if changes in their values occur over an order of magnitude in response to concentration variations.

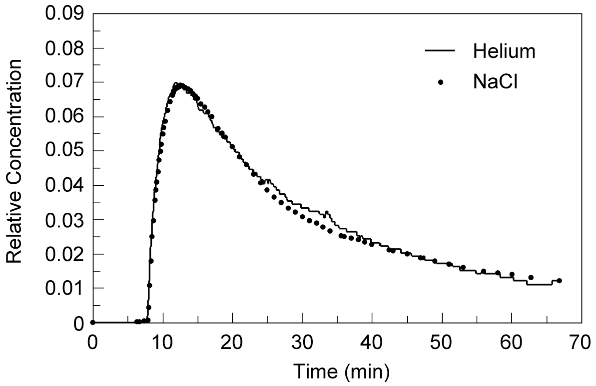

Figure 3. Concentration results for pipe experiment with NaCl and helium

Open-Tank Experiments

The experiments involved tracing a plume of dissolved helium and saltwater in an open tank (wedge shape of 244 cm length × 60 cm width (at the widest part) × 25 cm depth). NaCl dissolved in 3 liters of a saturated helium solution was injected at the wider end of the tank in about 15 seconds. At the opposite end, water was continuously pumped out of the tank and into the helium extraction/detection system at a specified rate. In some experiments, solute-free tap water was continuously injected into the tank at a specified rate.

The results showed that, compared to the helium curve, the first arrival and peak of the NaCl breakthrough curve were delayed by about 10 seconds. This can be explained by the different methods used to measure the two solutes. The conductivity probe is mounted in the line discharging the helium-free water from the membrane contactor (Figure 1). In this case, it seems it takes that many seconds for a water molecule to travel from the contactor membrane to the conductivity probe. Shifting the time axis by 10 seconds for helium showed that the two solutes are almost identical in terms of the breakthrough curve and the value of the peak (Figure 4).

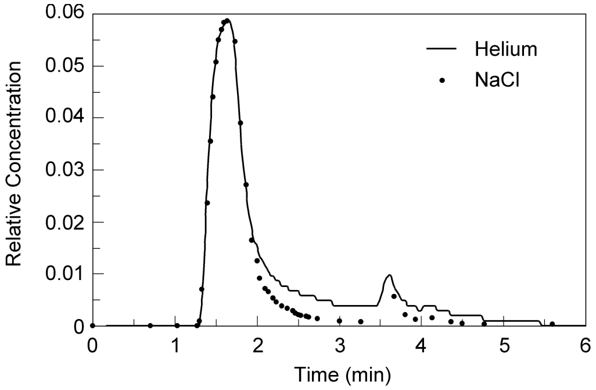

Figure 4. Concentration results for open-tank experiment with NaCl and helium

Another experiment included the introduction of two helium injections to test the ability of the detection system to respond to multiple-injection occurrences. The results are shown in Figure 5, where good overall match can be seen between relative concentrations of the two solutes, after shifting the time axis by 10 seconds for helium. However, loss of helium to the atmosphere is evident after about 6 minutes.

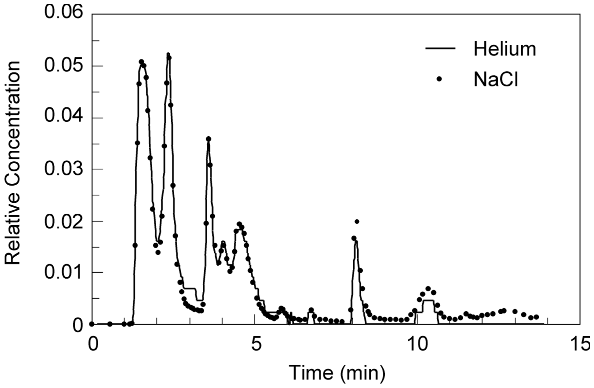

Figure 5. Concentration results for open-tank experiment O-3 with NaCl and helium. The experiment included two helium injection episodes.