

In Situ Measurement of Electroosmotic Fluxes and Conductivity Using Single Well-Bore Tracer Tests

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***In Situ* Measurement of Electroosmotic Fluxes and Conductivity Using Single Well Bore Tracer Tests**

Walt W. McNab, Jr.¹ and Roberto Ruiz¹

Abstract

Electroosmosis (EO), the movement of water through porous media in response to an electric field, offers a means for extracting contaminated groundwater from fine-grained sediments such as clays that are not easily amenable to conventional pump-and-treat approaches. The EO-induced water flux is proportional to the voltage gradient in a manner analogous to the flux dependence on the hydraulic gradient under Darcy's law. The proportionality constant, the soil electroosmotic conductivity or k_{eo} , is most easily measured in soil cores using bench-top tests where flow is one-dimensional and interfering effects attributable to Darcy's law can be directly accounted for. In contrast, quantification of EO fluxes and k_{eo} in the field under deployment conditions can be difficult because electrodes are placed in groundwater wells that may be screened across a heterogeneous mixture of lithologies. As a result, EO-induced water fluxes constitute an approximate radial flow system that is superimposed upon a Darcy flow regime through permeable pathways that may or may not be coupled with hydraulic head differences created by the EO-induced water fluxes. A single well comparative tracer test, which indirectly measures EO fluxes by comparing well bore tracer dilution rates between background and EO-induced water fluxes, may provide a means for routinely quantifying the efficacy of EO systems in such settings. EO fluxes measured in field tests through this technique at a groundwater contamination site were used to estimate a mean k_{eo} value through a semi-analytic line source model of the electric field. The resulting estimate agrees well with values reported in the literature as well as values obtained by bench-top tests conducted on a soil core collected in the test area.

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Introduction

Efforts to remediate contaminated aquifers by conventional pump-and-treat approaches are often frustrated by the presence of fine-grained, low-permeability materials harboring high concentrations of contaminants. Because such low-permeability materials (i.e., silts and clays) do not yield contaminants effectively by hydraulic pumping, transport of contaminants into more permeable portions of an aquifer is diffusion-limited, potentially requiring decades or even centuries for purging. This creates a model within which pump-and-treat functions only to maintain hydraulic control over a contaminant plume, still failing to rapidly remediate fine-grained contaminant source areas.

Electroosmosis (EO) provides a means to expedite the removal of contaminants from fine-grained materials (silts and clays) and is thus a viable complement to pump-and-treat approaches for a more complete approach to aquifer remediation. Briefly, EO entails the movement of pore water under the influence of an electric field. This movement is a result of the coulomb attraction of the diffusive double layer (the cloud of water molecules and positively-charged ions that forms over the negatively-charged surfaces of clay minerals) toward the negatively charged electrode (i.e., cathode). Viscous drag tends to pull the remaining pore water in the same direction. EO was first explored as a means for de-watering clays for purposes of soil stabilization (Casagrande, 1952). In recent years, the potential of EO for removing contaminants from fine-grained sediments has been explored in both laboratory studies and field scale demonstrations (Hamed et al., 1991; Bruell et al., 1992; Segall and Bruell, 1992; Acar and Alshawabkeh, 1993; Lageman, 1993; Probststein and Hicks, 1993; Shapiro et al., 1993). Recently, the Lasagna Project at the Paducah Gaseous Diffusion Plant in Kentucky entailed utilizing EO to transport TCE to activated carbon or iron filing treatment zones within the electric field, resulting in an estimated removal efficiency of 98% in 3 pore volumes (Ho et al., 1995, Athmer et al., 1997). Shultz (1997) explored the variables affecting the economics of EO technology for soil remediation and developed techniques for assessing costs per unit mass of soil treated, given treatment system size, electrode spacing, soil electrical properties, time, and initial capital costs. He

concluded, for example, that the costs associated with EO compare favorably to those of excavation and *ex situ* treatment in shallow soils, given a sufficient time to accomplish the cleanup.

The velocity of water associated with EO, q , is proportional to the voltage gradient, $\frac{d\phi}{dx}$, in a manner analogous to Darcy's law

$$q = \frac{-k_{eo}}{n} \frac{d\phi}{dx} \quad (1)$$

where k_{eo} is the electroosmotic conductivity and n the porosity. In reality, the EO conductivity is not truly a constant property of the material because it depends on a number of factors (e.g., ϕ -potential) that may vary with pH and other variables. However, k_{eo} is nonetheless useful for engineering design purposes; values range between 1×10^{-9} to $10 \times 10^{-9} \text{ m}^2 \text{ sec}^{-1} \text{ V}^{-1}$ (Mitchell, 1993) for typical soils. Given these values, EO water fluxes generated under reasonable voltage gradients (e.g., 1 V/cm) can exceed those induced by realistic hydraulic head gradients under Darcy's law by one or two orders-of-magnitude in low permeability materials (i.e., hydraulic conductivities less than 1×10^{-10} to $1 \times 10^{-9} \text{ m/s}$). However, the resulting hydraulic head difference between the cathode and anode produces a hydraulic gradient that acts to oppose EO-induced fluxes. In relatively homogeneous silt or clay bodies, this effect is unimportant because the low hydraulic conductivities do not provide a pathway for any return flow of water. In addition, to facilitate removal of contaminants and to prevent de-watering of the region surrounding the anode, water is extracted from the cathode well and injected into the anode well. Nevertheless, in heterogeneous sediments where relatively high permeability sand lenses or stringers may lie embedded within a matrix of otherwise low permeability material, the opportunity for flow short-circuiting exists (Figure 1A). Coupling a low-flow pump-and-treat system with an EO system may be essential in this scenario to offset the effect of flow-short-circuiting so that EO fluxes and Darcy fluxes are oriented in the same direction (Figure 1B).

Regardless of whether or not supplemental pumping is used to support an EO-based groundwater extraction system, the presence of contiguous permeable lenses within the electrode array can make the quantification of EO

fluxes very difficult. Because of the problem of Darcian return flow against the prevailing EO flux, direct measurement of EO flux is not possible. Similarly, methods for quantifying travel time between anode and cathode wells, using a tracer species for example, cannot distinguish EO and Darcian flux components of the groundwater velocity. In this study, we have attempted to overcome these issues by using single well tracer tests to indirectly measure EO fluxes by comparative analysis in a saturated water-bearing zone at Lawrence Livermore National Laboratory (LLNL) in California. The ultimate goal of the study was to utilize the EO flux data, together with estimates of soil conductivity, the applied electric current, and the electrode geometry to quantify the mean soil k_{eo} .

Methods

Test Area Hydrogeology and Equipment

The LLNL site, located some 35 miles (56 km) east of San Francisco, is located in a sedimentary basin underlain by Tertiary and Quaternary unconsolidated alluvial sediments derived from the surrounding California Coast Ranges. The subsurface environment is highly heterogeneous across the site, with multiple hydrostratigraphic units having been identified (Blake et al., 1995) that exhibit a spectrum of lithologies ranging from clays to gravels. The EO test area, a site previously contaminated by fuel hydrocarbons from former underground storage tanks in the 1950s through the 1970s, has been the subject of extensive prior remediation and investigation efforts since the early 1990s (Happel et al., 1996). During these investigations, a zone of predominantly fine-grained sediments located between 110 and 120 feet (34 to 37 m) below ground surface harboring residual fuel hydrocarbon contamination was identified. Average depth-to-water in the test area is approximately 95 ft (29 m). For purposes of evaluating EO, a total of four groundwater wells were installed in the test area and screened across this zone, with the wells aligned approximately in a 10-ft by 10-ft (3.0-m by 3.0-m) grid. Lithologic and geophysical logs, similar among each of the wells, suggest that these wells are screened primarily across silts and clays, although some sandy intervals, with thicknesses on the order of a few inches or centimeters, are also present. An aquifer test conducted by pumping on one of the wells, with the other three wells in the array used as monitor wells, indicated a high

degree of hydraulic communication between the wells, with a relatively high mean hydraulic conductivity (approximately 2×10^{-4} to 2×10^{-3} m/s). However, separate bench-top hydraulic conductivity measurements on soil cores collected from one of the wells indicated very low hydraulic conductivities in the silty/clayey materials (approximately 1×10^{-10} m/s, Cherepy et al., 1999). Hence, the relatively high hydraulic conductivities suggested by the pumping tests are presumably reflective only of the sandy intervals.

The four wells in the test area were each equipped with 10-ft (3.0-m) electrodes. In the two large (6 in. or 15.2 cm) diameter wells, these consisted of graphite rods, 3 in. (7.6 cm) in diameter. In the two small diameter (4 in. or 10.2 cm) wells, carbon steel rods, 2 in. (5.1 cm) in diameter, were installed. Both the soil electrical conductivity test as well as the single well bore EO flux tests (described below) required only one anode-cathode pair to be active; power was supplied in each case by a 100 V, 10 amp power supply.

In the vicinity of the electrodes, electrolysis reactions of water can produce high pH and low pH at the cathode and anode, respectively. The resulting pH gradient in the soil can alter surface properties of the clay mineralogy and hence the k_{eo} , reducing the efficacy of EO over time if steps are not taken to control pH. Given that the tracer tests entailed relatively short periods of time, pH control was not attempted in the phase of the study described in this paper.

Soil Electrical Conductivity Measurements

Bulk soil electrical conductivity, σ_s , a required parameter for the calculation of k_{eo} from the EO flux data, was measured using a four-electrode array. A voltage difference of 50 V was applied across two of the electrodes on one side of the grid while the passive electrodes were used to monitor the voltage potential distribution. The advantage of measuring the voltage difference between the passive electrodes, as opposed to the active ones, is that voltage drops associated with surface chemistry effects, as well as those associated with the well bore water and the PVC well casing, could be avoided. Because two of the electrodes consisted of unlike materials (carbon steel and

graphite), a DC offset associated with oxidation-reduction reactions of approximately 0.683 V was subtracted from the data to yield the correct voltage difference.

Based on the observed passive electrode voltage differences and the electrode geometry, a semi-analytical model of the potential distribution was used to calculate the soil bulk electrical conductivity, assuming a uniform, constant value for σ_s in an infinite three-dimensional domain. The semi-analytical model utilizes a point source solution to the steady-state potential field problem, integrated in the vertical direction to simulate a line source (i.e., an electrode), to predict the potential (i.e., voltage), ϕ , as a function of position with respect to the line source:

$$\phi(x, y, z) = \sum_{j=1}^N \frac{1}{L} \int_{ze_j}^{ze_j+L} \frac{I_j}{4\pi\sigma_s} \frac{1}{r_e} - \frac{1}{\sqrt{(x-xe_j)^2 + (y-ye_j)^2 + (z-\zeta)^2}} d\zeta \quad (2)$$

where L is the electrode length, x_{e_j} , y_{e_j} , and z_{e_j} , the coordinates for the location of the bottom of each electrode, and r_e the electrode radius. Superposition, indicated by the summation, allows for multiple electrodes, N , with the sign on the current flow through each j th electrode, I_j , used to distinguish anodes and cathodes. Thus, Equation 2 accepts current flow through individual electrodes as input and can be used to calculate the voltage difference between electrodes.

Utilizing MathCad 8.0 (MathSoft, Inc.) as a computational platform, the value of σ_s in Equation 2 was adjusted until the predicted and observed voltage differences across the two passive electrodes were in approximate agreement at 0.59 V, with approximately 5.4 amps of current flowing through the entire circuit. The resulting estimate for σ_s , approximately 0.13 siemens/m (S/m), is well within the typical range reported for soils of 0.01 to 1 S/m (Mitchell, 1993). As a comparison, σ_s was also measured in the laboratory from a single soil core collected from one of the soil borings and was estimated to range between approximately 0.06 to 0.09 S/m (Cherepy et al., 1999).

The modeled voltage difference between the two active electrodes, approximately 23 V, is considerably less than the actual applied voltage of 50 V. This apparent voltage loss may be attributable to a number of factors, including the resistance offered by the slotted PVC well casing, the water within the well bore, sand pack, and various surface chemistry effects. While it is possible to model this voltage loss using individual resistivity estimates and an appropriate numerical method, such a calculation is outside the scope of the present paper.

Measurement of Electroosmotic Flux

EO flux measurements were conducted using a series of single well comparative tracer tests. These tests entailed placing a tracer within the well bore and measuring its rate of removal over time in response to mixing and dilution processes; differences in the tracer concentration history between tests with the DC current both on and off would be indicative of the electroosmotic flux. Single well bore tests have been used previously to estimate groundwater velocities in the vicinity of the test well (Drost et al., 1968; Grisak et al., 1977), although focusing effects can complicate the interpretation. However, in this particular application, it is only the difference between the two test cases (current on or off) that is of interest. As long as the conditions of the tests remain otherwise the same, focusing and other dispersive effects are not directly relevant.

To minimize potential interference from ion electromigration, adsorption or chemical transformation processes, water with a different oxygen-18 signature from the native groundwater was selected as the tracer. The isotopic fractionation of oxygen in a water sample is given as a comparison between the oxygen-18/oxygen-16 ratios of the sample to that of standard mean ocean water (SMOW) and is referred to as ^{18}O . A negative value of ^{18}O is indicative of water which is depleted in the heavy oxygen-18 isotope relative to the SMOW, while a positive value indicates enrichment. Evaporation and condensation processes tend to fractionate the lighter oxygen-16 water molecules from those of the heavier oxygen-18; water vapor generated by evaporation at colder temperatures tends to have strongly negative ^{18}O values, with less negative values at higher temperatures (Faure, 1986). For this reason, differences in ^{18}O between native groundwater in the test area, recharged locally, and local tap water, originating as snowmelt from the

Sierra Nevada some 100 miles (160 km) away, can be exploited to yield an effective tracer. Specifically, the ^{18}O values associated with native test area groundwater, -8.1, and tap water originating from the Hetch Hetchy Reservoir in Yosemite National Park, -13.5, provided a broad dynamic range. All oxygen-18 analyses were conducted using standard mass spectroscopy methods.

A mechanical circulation system was devised to pump water from the well bore up to the surface and back again into the well during (Figure 2). This system allowed simultaneous injection of the tracer water at the base of the water column, at approximately 1 gal/min (3.9 L/min), while native groundwater was extracted at an equivalent rate from the top of the column, thus minimizing differences in hydraulic head between the well bore and the surrounding aquifer. This configuration also served to keep the tracer well mixed within the well bore throughout the course of the test and provided a convenient means of periodically obtaining groundwater samples for oxygen-18 analysis.

Two tests were conducted, both using a single well as the monitored electrode well while the well on the opposite corner of the grid housed the electrode of opposite charge. Each test involved a different polarity orientation, with the test well electrode serving as the anode for the first test and the cathode for the second. Two trials were conducted for each test so that background tracer dilution rates were measured first (without an applied electric field), followed by a trial with a 50 V difference placed across the electrodes. Each trial lasted for approximately three days with residual tracer extracted from the vicinity of the well bore between tests by pumping out approximately 200 gallons (800 L, or roughly 6 well volumes). The measured ^{18}O values from the four trials over time are shown on Figure 3A; conversions of these data into the fraction of the tracer water present in the well bore are shown on Figure 3B. The well bore tracer fraction data clearly indicate that a significant portion of the tracer, approximately half, is lost from the well bore at the start of each of the trials. Mechanical mixing across the well screen as a result of the recirculating pumping action may be responsible for much of the loss. Some of this loss may also result from density differences between the tracer water and native groundwater. Groundwater in the test area remains at

elevated temperature (35 to 40 C) several years after thermal treatment approaches (steam injection, electrical resistive heating) as a consequence of relatively slow rates of groundwater movement and the thermal insulating properties of the sediments. The tracer water, on the other hand, was held at the surface in a fiberglass bubble, exposed to temperatures only on the order of 25 to 30 C. Presumably, the colder and thus denser tracer water, injected at the base of the water column, would tend to flow out of the well screen into the surrounding sand pack as well as into the sand fingers of the formation. Regardless of the cause, the tracer loss holds important implications for the interpretation of the tracer test results. When used as an anode, water will be generally drawn away from the vicinity of the well bore by EO, which by itself exerts no immediate effect on the observed tracer concentration. However, the resulting decline in the hydraulic head within the well bore will cause water to flow back into the well by Darcy's law through the most permeable units, the sand fingers (Figure 4A). The presence of tracer in these sand fingers implies the return of some of the tracer mass to the well bore, thus reducing the apparent rate of tracer loss in the well compared to the scenario when no electrical current is applied. In the case of a cathode configuration in the well bore, water will be drawn toward the well from the surrounding formation by EO, displacing tracer-laden water in the sand pack back into the well bore (Figure 4B). The resulting increase in hydraulic head within the well will tend to drive the well bore water back out into the formation through the sand fingers, although this effect will not influence the tracer concentration in the well bore itself. Thus, with either electrode polarity, the tracer concentration within the well bore would be expected to decline more slowly when the electric current is applied than when it is not.

Analysis

The tracer data from the four trials, normalized to the respective initial tracer concentrations at the beginning of the monitoring period, are shown on Figure 5. The changes in well bore tracer fraction over time reflect several mechanisms, including groundwater advection (focused into the well by the relatively high permeability of the sand pack), along with dispersive mixing across the well screen, sand pack, and surrounding formation (enhanced by the

re-circulating pumping action). Nevertheless, for measuring the EO flux, it is only the differences between the curves for the cases when electric current is applied, and when it is not, that are of interest.

Differences in well bore tracer fraction over time between the background and applied current trails for both test pairs are shown on Figures 6A and 6B. For both tests, mismatches in sampling times between the trails were addressed using linear interpolation between the sampling events when the electric field was not present. For both tests, linear regression indicates a significant linear trend, with a slope of 0.2%/hr, corresponding to approximately 1.6 gal/day (6 L/day) given the 33 gallons (125 L) of water within the well bore. In contrast, a similar analysis of the differences between the two background trials indicates no relationship (Figure 7).

The estimated EO flux, taken with the electrode geometry, electric current, and the estimated k_{eo} , can be used to estimate a mean value for k_{eo} . Because the EO-induced groundwater velocity is, as an engineering approximation, proportional to the voltage gradient (Equation 1), the semi-analytical model used to calculate the voltage potential distribution can easily be extended to calculate the local EO-induced groundwater velocity field. That is, at any point in the model domain, the local groundwater velocity components due strictly to EO are given by,

$$v = \begin{pmatrix} -\frac{k_{eo}}{n} \frac{\phi}{x} \\ -\frac{k_{eo}}{n} \frac{\phi}{y} \\ -\frac{k_{eo}}{n} \frac{\phi}{z} \end{pmatrix} \quad (3)$$

where n is the material porosity and the voltage gradient components are calculated by numerical differentiation of Equation 2 (assuming k_{eo} is not directionally dependent). With this relationship available for quantifying the EO-induced flow field, the calculated summation of the EO fluxes across a mathematical cylindrical surface surrounding an electrode provides an approximation of the flux of water, Q , to the well (neglecting contributions across the top and the base of the cylinder),

$$Q = L \int_0^{2\pi} n v_n (xe + \rho \cos(\theta), ye + \rho \sin(\theta), ze + \frac{L}{2} \rho) d\theta \quad (4)$$

where v_n is the component of the EO-induced groundwater velocity vector normal to the cylinder at the midpoint of its length (L), ρ is the radius of the cylinder, and θ is the variable of integration. Again using MathCad 8.0 (MathSoft, Inc.) for computation, the value of k_{eo} in Equation 3 was adjusted until the predicted and observed values of Q (viz. Equation 4) were in approximate agreement at 1.6 gal/day (6 L/day). The resulting estimate for k_{eo} , roughly $2.3 \times 10^{-9} \text{ m}^2 \text{ sec}^{-1} \text{ V}^{-1}$, is within the typical range reported for soils (1×10^{-9} to $10 \times 10^{-9} \text{ m}^2 \text{ sec}^{-1} \text{ V}^{-1}$, Mitchell, 1993). For comparison, k_{eo} was estimated in the laboratory by applying a voltage gradient across a single soil core from one of the soil borings at the site and measuring the resulting water flux. The k_{eo} was found to range between approximately 0.7 to $1.2 \times 10^{-9} \text{ m}^2 \text{ sec}^{-1} \text{ V}^{-1}$ (Cherepy et al., 1999).

The agreement between the inferred value of k_{eo} , the range reported in the literature and the results of the laboratory soil core measurements provide a level of confidence that the approach used in this study—a comparative single well tracer test and relatively simple semi-analytic models for data interpretation—is valid. As such, this approach may be useful for assessing the efficacy of EO-based remediation systems operating in heterogeneous geologic materials and may also provide a useful screening model for the design of such systems.

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Biographical Sketches

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Figure 5. Tracer data normalized to the respective initial tracer concentrations for each trial.

Figure 6. Differences in normalized well bore tracer fraction between the background test (no electric current) and a voltage difference of 50 V, for (A) anode configuration (top) and cathode configuration (bottom). Heavy lines indicate best-fit linear regression; light lines show 95% prediction bands.

Figure 7. Differences in normalized well bore tracer fraction between the two background tests. The lack of a discernible trend with time (i.e., negligible differences in well bore fluxes) contrasts with the significant temporal trends in the differences in the well bore tracer fraction when an electric field was applied and when it was not, respectively (Figure 6). The difference in the plots (i.e., Figure 6 versus Figure 7) is suggestive of a response in the pattern of water movement through the well bore to the application of an electric field (i.e., electroosmosis).

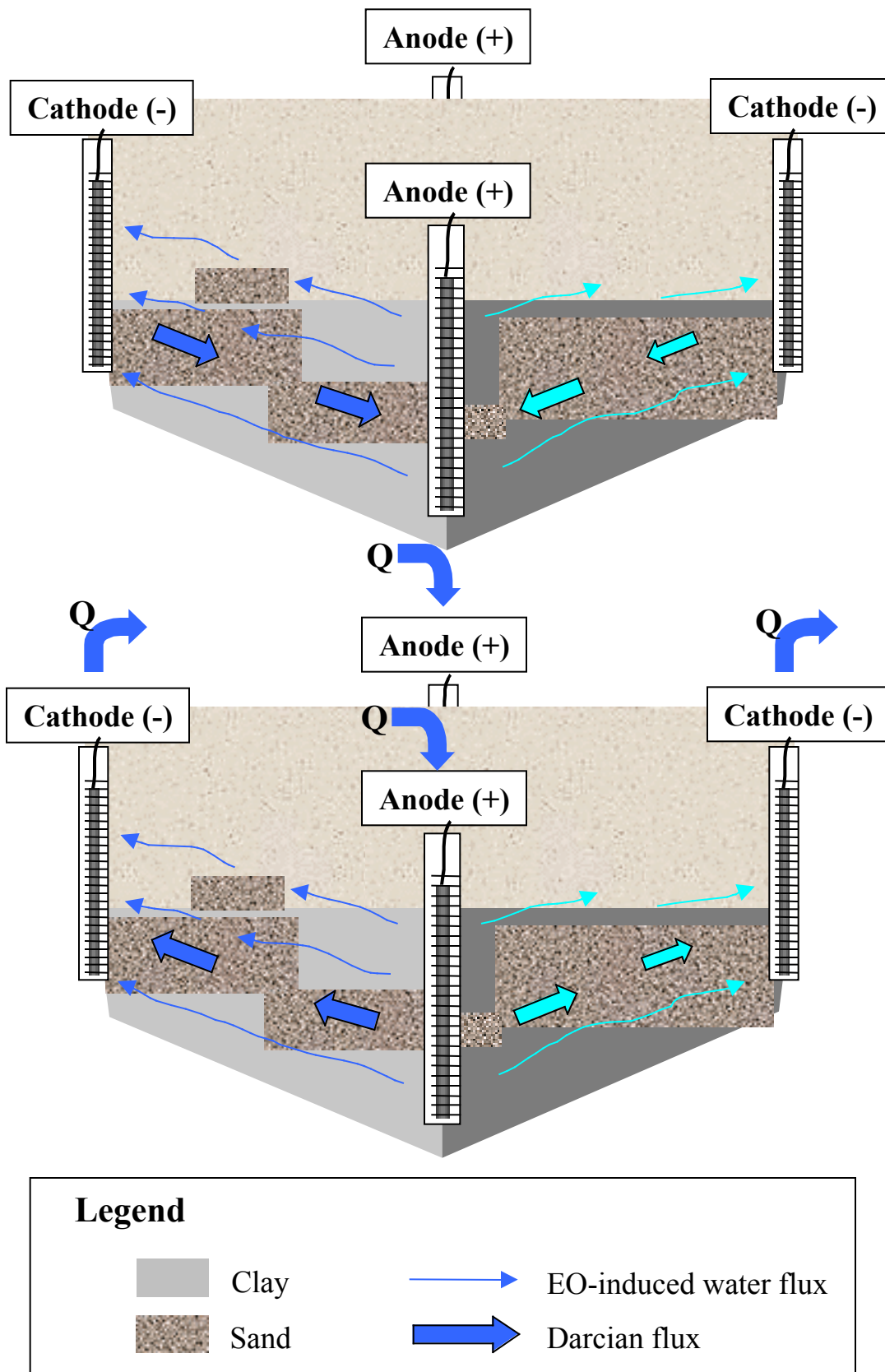


Figure 1A & 1B

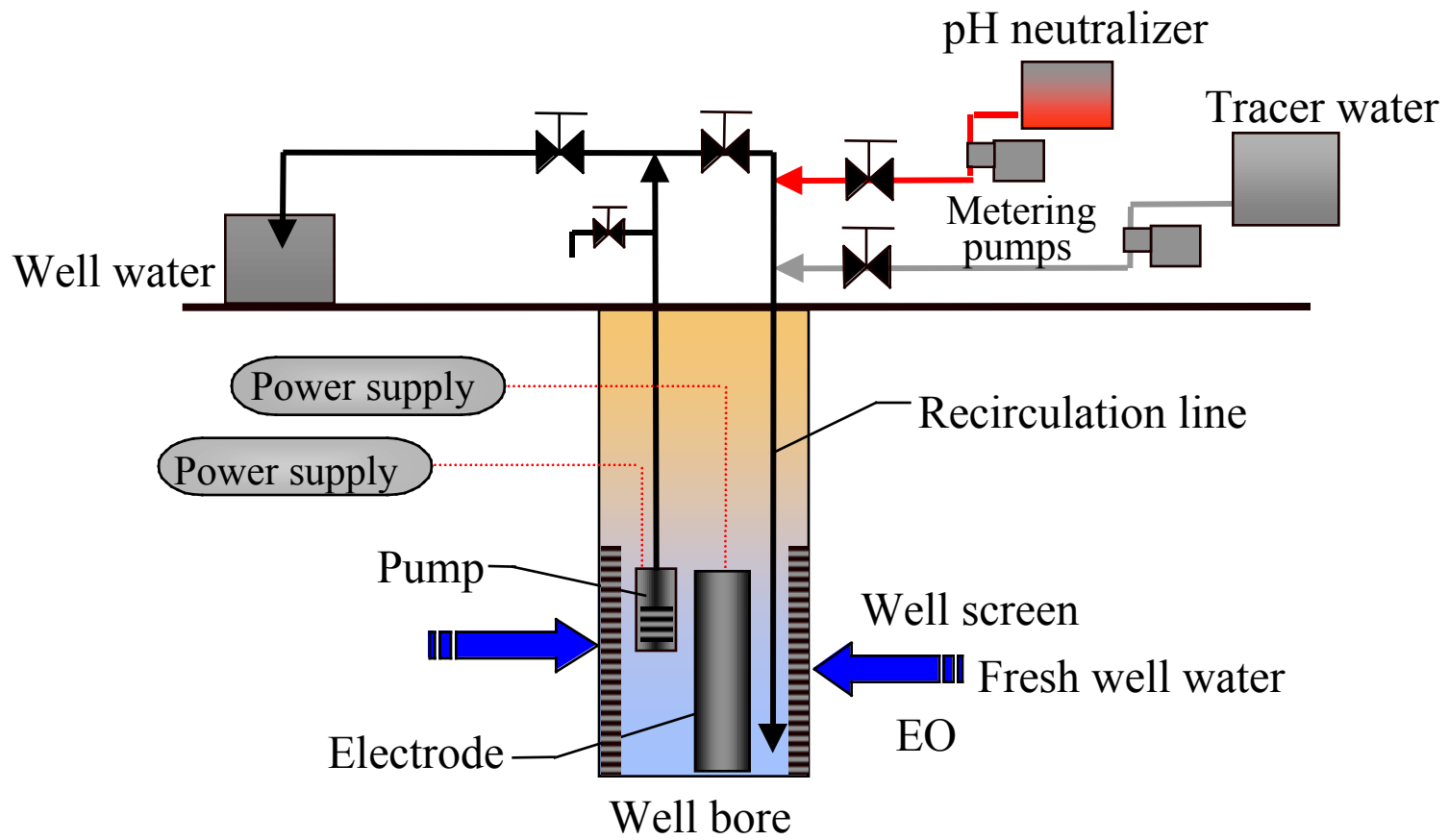


Figure 2

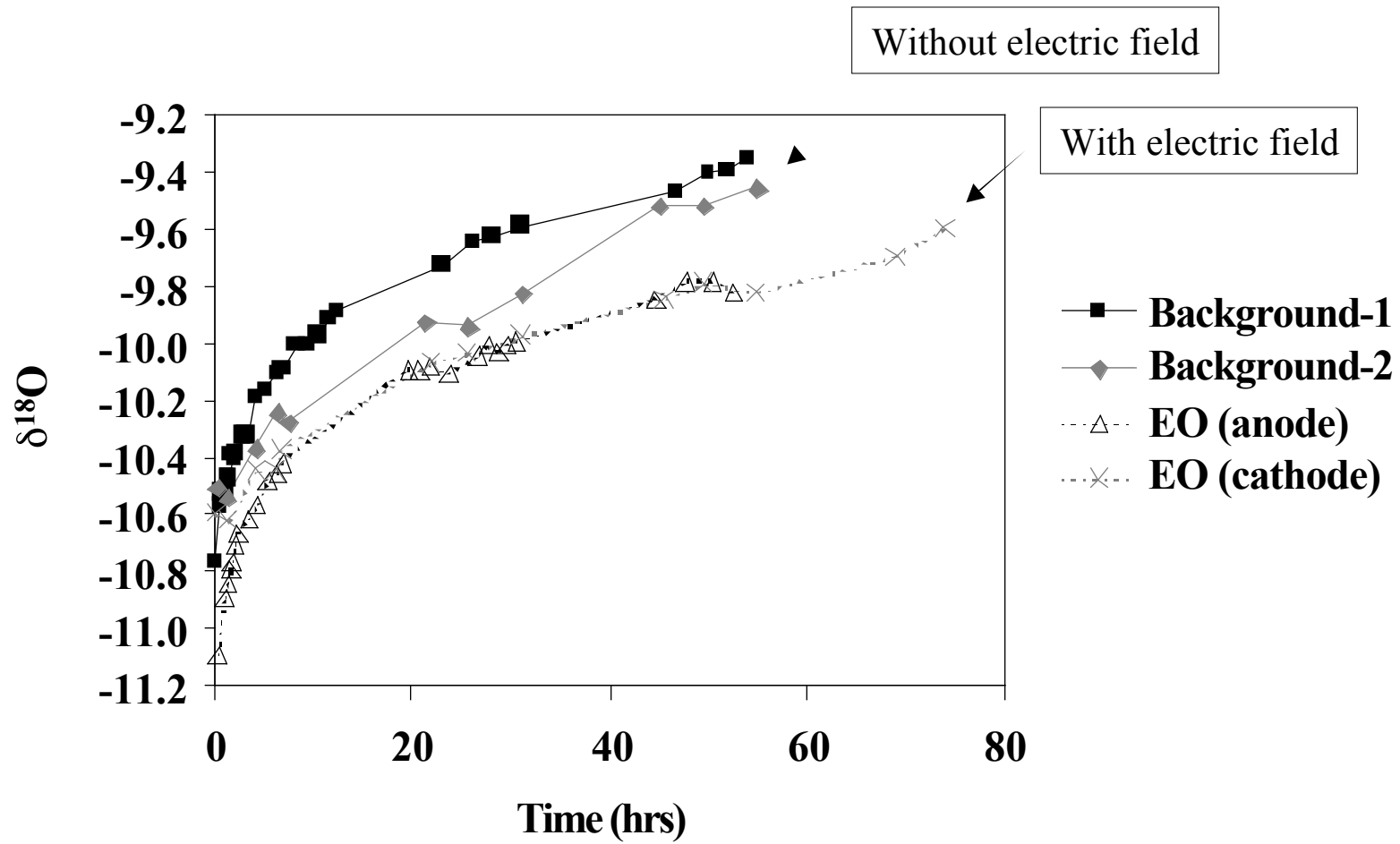


Figure 3-A

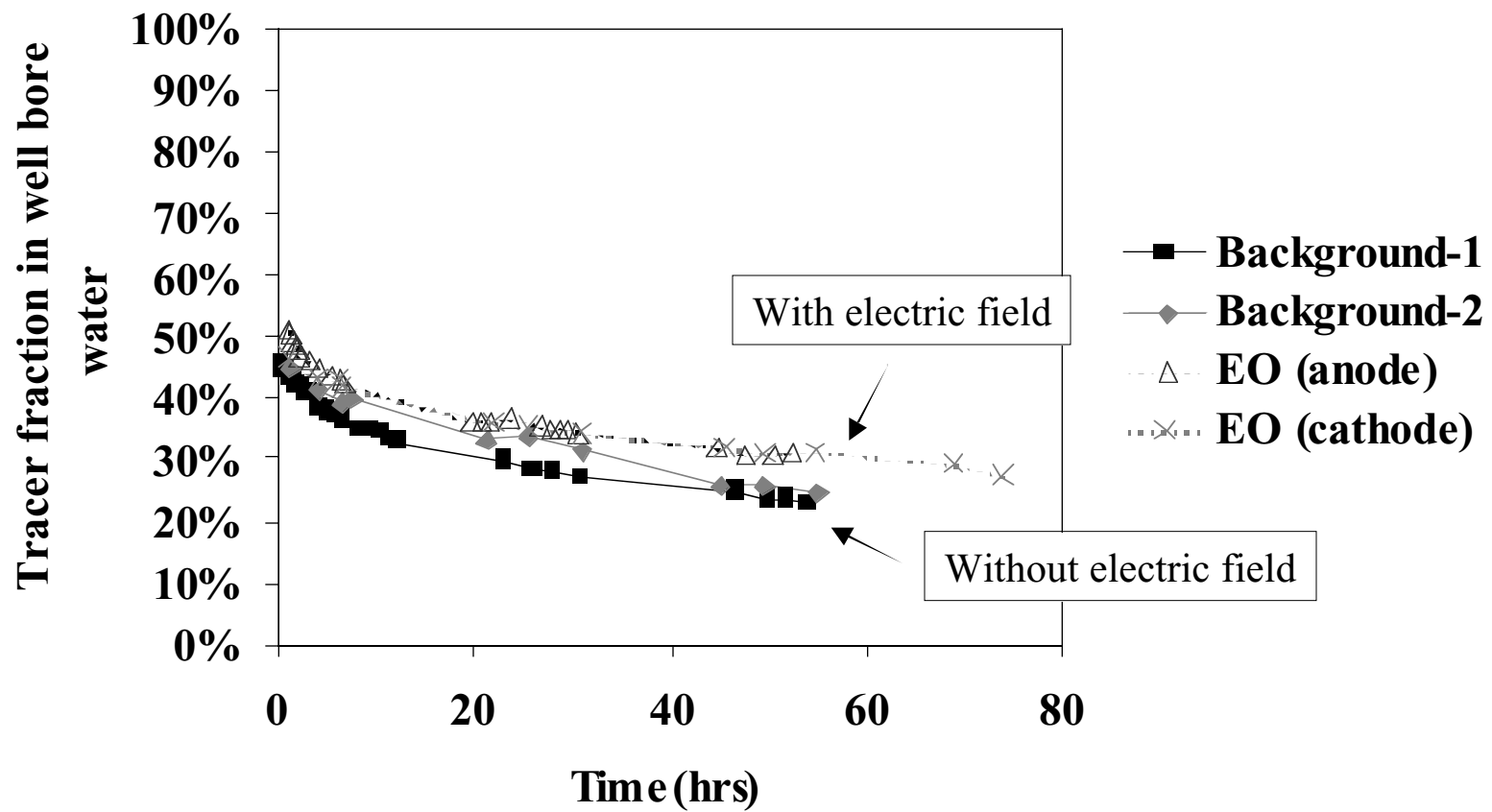


Figure 3-B

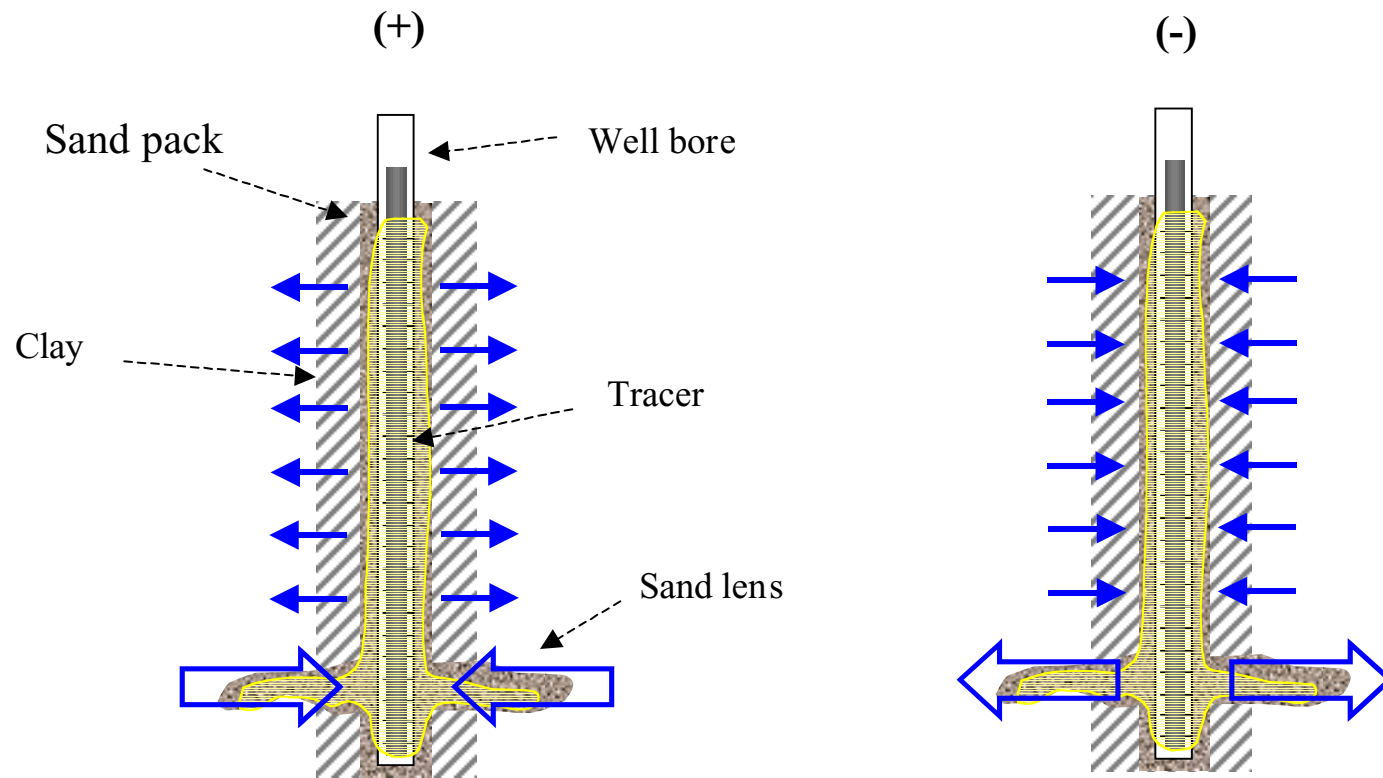


Figure 4A and 4B

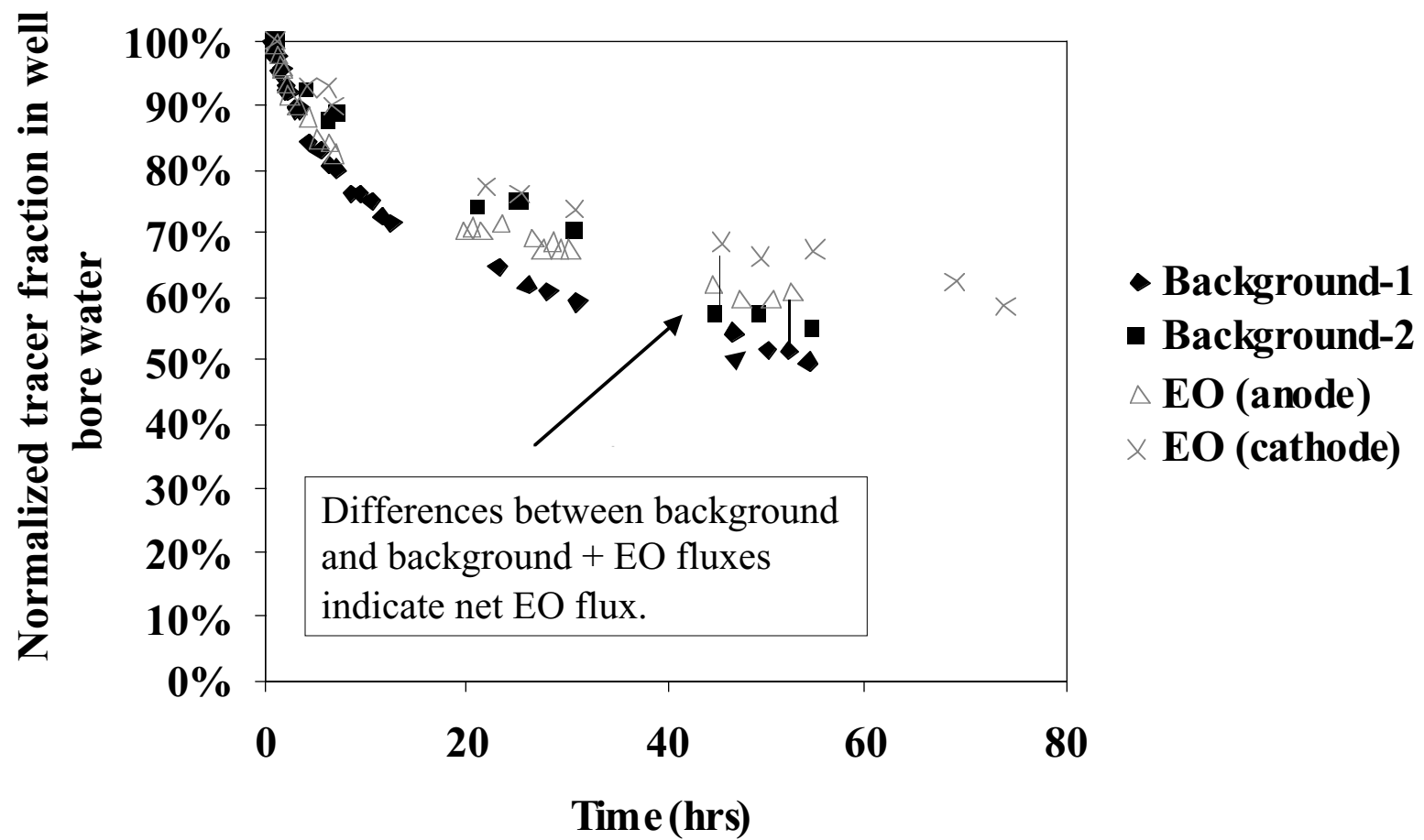
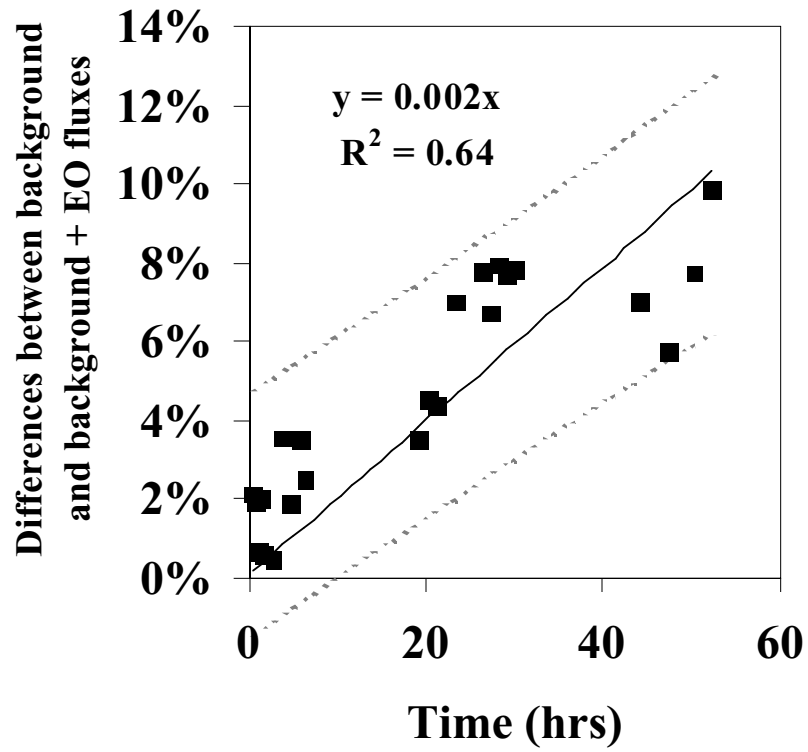
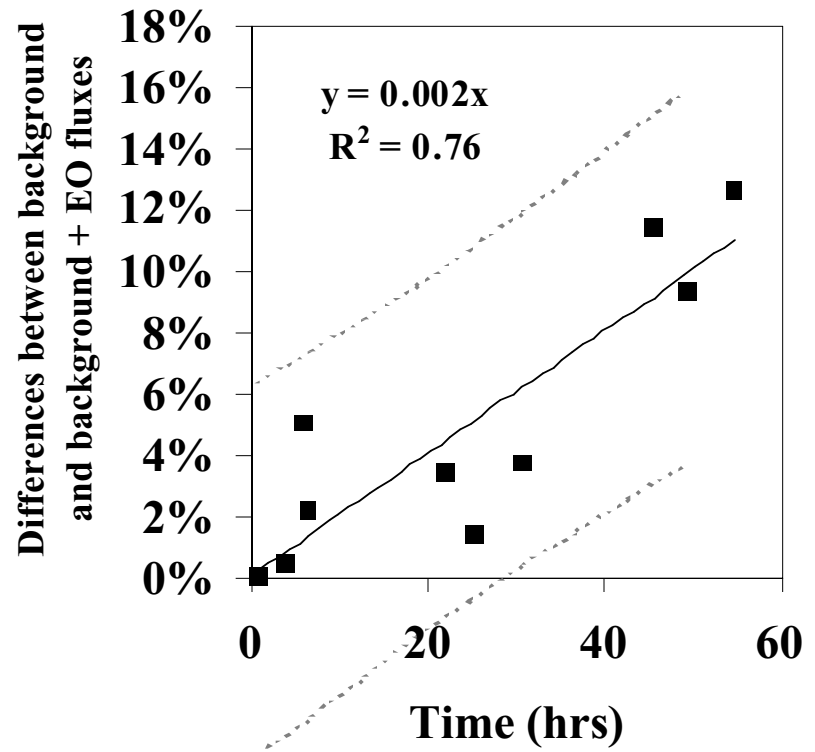


Figure 5

Test #1 (anode)



Test #2 (cathode)



Figures 6-A and 6-B

Background differences - no trend

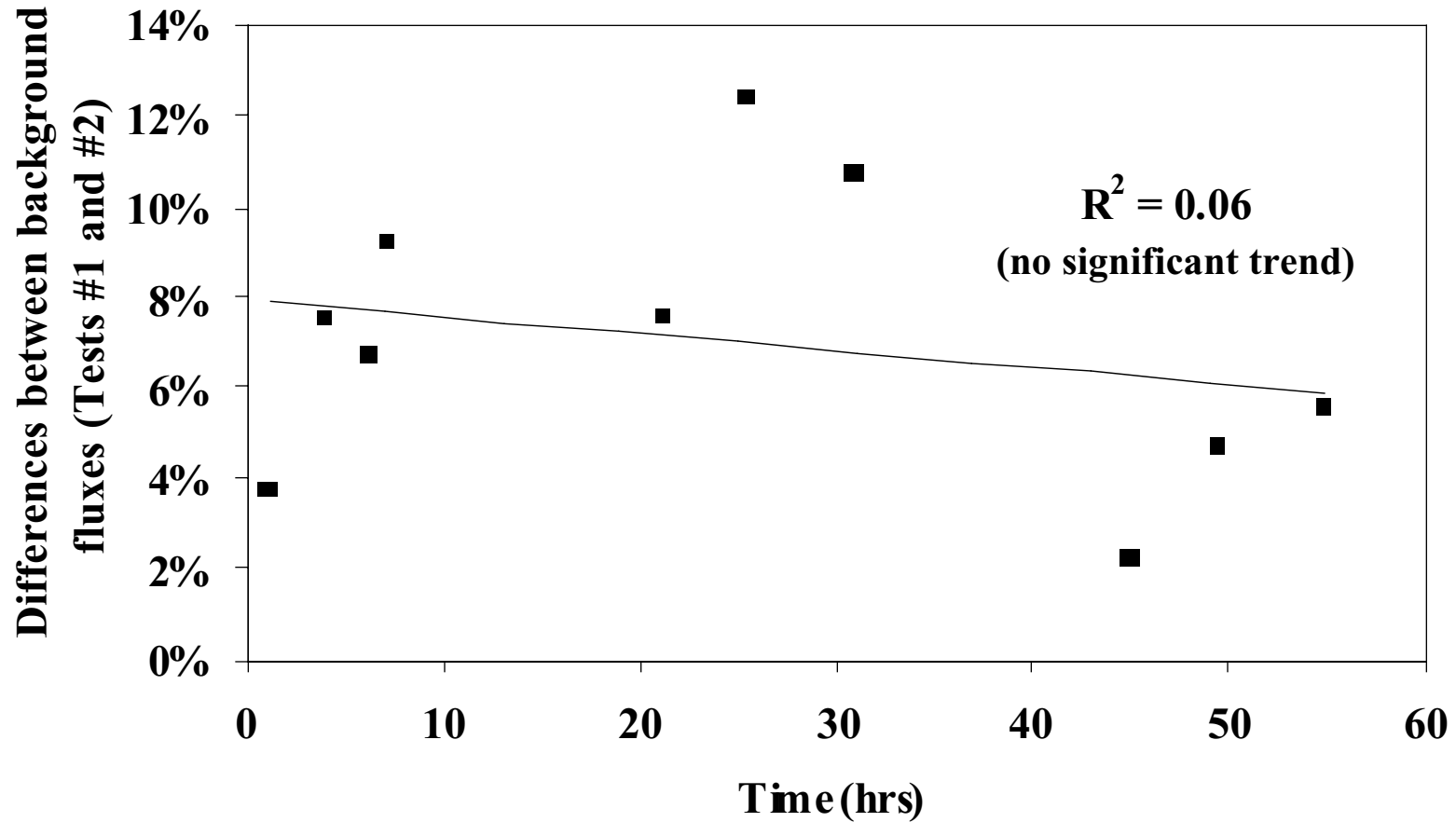


Figure 7