Acoustically Enhanced Contaminant Recovery in Low Permeability Soils.

Theodore G. Cleveland[1] and Sanjay Garg[1]

Abstract

This communication presents on-going research that is studying the use of acoustic excitation to improve the recovery of contaminants in in-situ soil washing for low-permeability soils. Two series of experiments were conducted to explore the feasibility and suggest mechanisms of acoustically enhanced contaminant recovery from porous media. Based on modeling projections the acoustic treatment can improve overall contaminant removal by 7%, and the primary mechanism of removal is the suspension of the fines in a soil that tend to strongly adsorb contaminants.

Introduction

Contaminated low-permeability soils are difficult to recover contaminants from because the contaminants strongly sorb to the soils, and because preferential pathways can develop so that soil washing fluids cannot come into contact with all the contaminant. Typically low permeability soils contain large fractions of clay or silt. They are often used (after engineering modifications) as liner material for lagoons and landfills. However, when they become contaminated the properties that make them impermeable to water (surface charge and high specific surface), also make them excellent sorbents for the contaminants. After sorbing the compounds, the soils can serve as long term sources of water contamination.

[1] Department of Civil and Environmental Engineering, University of Houston, Houston, TX 77204-4791.

Low permeability inclusions in productive aquifers are also comprised of clay and silt, and behave in a similar fashion when exposed to contamination - thus when a groundwater contamination source is removed, the groundwater equilibrium is altered, and the low permeability inclusions that initially adsorbed the contaminants, then serve as a long term source of contamination.

Current practice is to recover all pumpable contaminants, then use bioremediation, soil washing, or excavation to address the residual (sorbed) contamination. Most successful in-situ washing programs are limited to high permeability soils, yet it is the low permeability inclusions that serve as sources of long term contamination. Thus techniques that specifically address low-permeability soils are of great interest.

Evangelista and Zownir (1989) presented data that indicate that the majority of lead contaminants in typical soils are sorbed to fines that pass through a #200 sieve (75 μm or smaller) and concluded that the majority of the heavy metals they studied (lead, zinc, nickel, cadmium, and copper) are contained in the fines. Farrah and Pickering (1978) demonstrated in earlier work that clays have a tremendous binding energy for heavy metals and make desorbtion of these contaminants difficult.

Stauffer et al. (1989) reported similar findings for organic contaminants. They found through principal component analysis that the sorbtion of organic compounds is most strongly correlated with the organic fraction and the silt clay fraction of the porous medium; remarkably at about the same degree of importance. What their work implies is that the finer grained portions of an aquifer contribute greatly to the sorbtion of organic compounds regardless of the organic fraction of the aquifer. Clearly, any in-situ method which could remove these finer particles would contribute greatly to the efficiency of any contaminant recovery scheme by simultaneously removing contaminants attached to the solid phase, and reducing the residual contaminant source.

This research is studying the use of acoustic excitation to improve the recovery of contaminants in in-situ soil washing for low-permeability soils. The idea behind acoustic excitation is to either stimulate desorbtion of contaminants themselves or to stimulate the suspension of the fines in a soil that tend to strongly adsorb contaminants and then use hydrodynamic gradients (pumping) to recover the contaminants. The remaining soil left behind is thereby less contaminated.

Literature Review

Ultrasonic vibration has been used to separate soil particles and to disperse clays for chemical and physical analysis (Edwards and Bremner, 1967). It is also used to extract humic acids for soils analysis, and to cause cell lysis for biochemical analysis (Ramunni and Palmeri, 1985).

Sonic excitation to enhance and produce chemical reactions is well documented in the sonochemistry literature. Sonic irradiation is thought to enhance reactions by increasing interparticle collisions that fracture particles and increasing reactive surface area. In liquid systems sonic irradiation causes cavitation within the liquid releasing tremendous amounts of energy (Suslick et al., 1989).

Related phenomenon include the acoustic-electric coupling due to compressional waves predicted by Biot (1956). Martner and Sparks (1959) later presented evidence that compressional waves (Biot slow waves) induce relative fluid-solid motion which in turn generates an electro-magnetic wave with the same time variation as the compressional wave. These results were followed by generally unsuccessful attempts to replace geophones with antennas (Broding et al., 1963). The phenomenon is thought to be due to the induction of a streaming potential with the same time variation as the acoustic wave and is being pursued by geophysicists as a prospecting tool to locate gas/water interfaces and to identify the nature and pore content of sedimentary rock.

In 1988 the Superfund Innovative Technology Evaluation (SITE) program funded research into electro-acoustic decontamination of soil (Muralidhara, et al., 1990). The contaminants they studies were the inorganic compounds zinc and cadmium and the organic compound decane. The research encountered analytical difficulties with the decane at results were inconclusive for this compound. However, for the inorganic compounds the researchers concluded that electro-acoustic soil decontamination is technically feasible for inorganic contaminants.

They also concluded that there appears to be an apparent synergistic effect due to the acoustic field, but the magnitude of the effect was not quantified. The report states that the mechanism that augments dewatering when using the combined electro-acoustic drainage is not well understood, but they hypothesized several mechanisms - including the streaming potential explanation of the geophysicists.

The U.S. EPA risk reduction laboratory in Cincinnati conducted research into using sonic vibrations for permeability amendment as an enhanced recovery technique to increase the yield of a recovery well (Murdoch, et al., 1988). They reported a four-fold permeability increase in one laboratory study.

Researchers at Kansas State University and New Jersey Institute of Technology are currently studying vibratory phase separation of immiscible contaminants, and ultrasonic vibration as a way to enhance the flow of water in clayey sands (Lakshmi, 1992; Lakshmi, et al., 1993). The major complications are difficulties with analytical and experimental equipment, lack of suitable exciters to generate high amplitude sonic waves, and internal reflections from the experimental media.

From these works and the geophysical and rock mechanics literature there appears to be some useful connection between acoustic irradiation and the possible mobilization of contaminants.

The remainder of this communication describes two series of experiments conducted to explore the feasibility and suggest mechanisms of acoustically enhanced contaminant recovery from porous media.

Laboratory Experiments

Our experiments were conducted in a 1 meter long, 0.25 meter diameter column schematically depicted in figure 1. The column was filled with a mixture of sand, shale aggregate, and Beaumont clay collected from the University of Houston Coastal Center in LaMarque, Texas. An ultrasonic cell disintegrator was modified to fit into the column and be placed near the media screens shown in figure 1. This configuration was used to mimic proposed placements in the field.

Flow in the column was maintained by small marine bilge pumps, and we monitored total suspended solids (TSS) in the outflow because our literature search suggested that sorbing contaminants will be carried by the fine particles and TSS will be proportional to the amount of fines in the effluent.

The first experiment studied the effect of ultrasonic vibration in a column where flow and acoustic gradient were in the same direction. Figure 2 compares the TSS in the effluent of the column before and then during treatment. The figure clearly indicates that acoustic excitation increases the elusion of suspended solids, however when the exciter is turned off the solids removal returns to the pre-treatment level rapidly. We noticed that after excitation head loss through the column increased dramatically.

We compared the permeabilities before and after excitation and found that the permeabilities were reduced by the acoustic excitation by about 25%. We attribute the decreased permeability partially to auto-filtration of the solids (a filter cake builds up very rapidly after excitation) and partially to compaction produced by the mechanical agitation. We made no attempt to quantify the relative magnitude of these permeability damage mechanisms.

We repeated the experiment, but reversed the hydraulic gradient to explore what effect having the acoustic gradient and hydraulic gradients in opposite directions might have. This second experiment indicated that when the gradient is reversed solids removal can be sustained.

The results of these experiments led us to the following conclusions - acoustic excitation can increase solids removal and when the acoustic and hydraulic gradient are in opposite direction the removal can be sustained. These experiments suggested that for our field experiment to be useful we should place any acoustic exciters in a pumping well to enhance solids removal.

Our third experiment was designed to mimic our proposed field experiment, which will be a modified radial injection-extraction test - similar to a dispersion test used in subsurface hydrodynamic characterization.

In the same size column as before we prepared a medium of coarse sand and coastal center clay. We contaminated the soil in place by pumping dye into the column until breakthrough and maintained this pumping for two days. Then we allowed the column to sit so that some quasi-equilibrium could occur. We then reversed the gradient so that the pumping side of the column now became the effluent side. Figure 3 schematically depicts the contamination portion of the experiment.

We monitored the effluent for total suspended solids and dye concentration. We tried several extraction techniques to quantify the amount of dye removed in the liquid phase and on the solid phase with little success - finally we resorted to ultra-centrifuge to separate the liquids and solids and a quasi-equilibrium isotherm to estimate the amount of dye on the solids.

The adsorbtion of the dye is not instantaneous, but appears to reach a quasi-equilibrium after 24 hours. Figure 4 shows a 24-hour isotherm for the porous medium. The isotherm is non-linear and suggests that two different adsorbtion regimes exist,

however all our samples fell in the lower left hand corner of the isotherm and we assumed linear behavior for our analysis.

The column was operated without excitation for 48 hours until we felt it had reached its asymptotic performance where removal of solids and dye was relatively constant. Then ultrasonic excitation at 200 watts was applied on a schedule of 10 minutes on, 50 minutes off for 8 to 10 hours a day. No excitation or monitoring occured overnight.

Figure 5 shows the total suspended solids in the column effluent during the experiment. As shown on the figure, after 20 hours the suspended solids in the effluent was on the order of 5 mg/l and relatively constant. When excitation is applied suspended solids increases dramatically then reduces to about 20 mg/l. When the exciter is turned off, the concentration drops back to the pre-treatment level in a couple of minutes.

Figure 6 shows the measured dye concentrations in the effluent liquid that was centrifuged to separate the liquid and solid phases. Liquid phase concentration was initially high then decreased rapidly, at 20 hours it increased then declined. The figure shows that acoustic excitation has little net effect on the liquid phase concentration and the fluctuations decrease with time as the medium is swept of dye.

Figure 7 shows the solid phase concentration estimated from the isotherm shown on figure 4. The solid phase concentration decreases rapidly initially but increases with excitation. Figure 8 shows the combined contaminant concentration expressed as mg/l of dye. The figure compares the experiment to the projected no-excitation case based on liquid phase concentrations and suspended solids removed before excitation was started. This figure indicates that the treatment has a measurable improvement over the projected asymptotic performance estimated by modeling. The difference in areas between the two curves, which represents the increased mass removed due to excitation, is 7%.

When the excitation is applied contaminant removal is primarily with the solid phase (as expected). Head loss across the column increased slightly during the pre-treatment portion of the experiment and decreased dramatically afterwards. The permeability was increased by excitation almost 100%, and is attributed to the removal of additional fines suspended by the acoustic excitation.

Conclusions

This communication presented on-going research that is studying the use of acoustic excitation to improve the recovery of contaminants in in-situ soil washing for low-permeability soils. Results from a laboratory experiment are presented supporting the following conclusions: high power ultrasonic excitation can increase solids removal and it can be sustained when the acoustic and hydraulic gradient are in opposite direction; excitation has little effect on liquid phase concentration (at least in our ability to resolve minor changes in concentrations); contaminant removal is facilitated by the solids removal because contaminants are sorbed onto the solids; and based on modeling the acoustic treatment can improve overall contaminant removal by 7%.

The idea holds some promise to either stimulate desorbtion of contaminants themselves or to stimulate the suspension of the fines in a soil that tend to strongly adsorb contaminants and then use hydrodynamic gradients (pumping) to recover the contaminants. The remaining soil left behind is thereby less contaminated.

Future work includes a field experiment conceptually similar to the laboratory experiments to verify that the method can be scaled up. The vibrations will be induced by a high-frequency pin vibrator similar in design to a concrete densification vibrator, but operating at higher energies and vibrational frequencies. The same tracer compounds will be used in a permitted injection/extraction experiment.

References

Biot, M.A., 1956. Theory of propogation of elastic waves in a fluid-saturated porous solid, J. Acoust. Soc. Am., 28, 168.

Broding R.A., Buchanan, S.D., and D.P. Hearn, 1963. Field experiments on the electroseismic effect: IEEE Trans. on Geosci. and Electronics, GE-1(1), 23.

Edwards A.P., and J.M. Bremner, 1967. Dispersion of soil particles by sonic vibration, J. Soil Sci., Vol. 18, No. 1, pp 47-63.

Evangelista, R.A., and Zownir, A.P., 1989. Lead contamination removal by soil washing, In Proceedings of Air and Waste Management Association, Annual Meeting, Anaheim, CA., pp 2-16.

Farrah, H., and W.F. Pickering, 1978. Extraction of heavy metal ions sorbed on clays, Water, Air, and Soil Pollution, Vol. 9, No. 4.

Lakshmi, N.R., 1992. Personal communication.

Lakshmi, N.R., Berliner, S. III., amd K.Y. Lee, 1993. Feasibility of ultrasonic enhancement of flow in clayey sands, to appear in ASCE Journal of Environmental Engineering Division.

Martner, S.T., and N.R. Sparks, 1959. The electroseismic effect: Geophysics, 24, 297.

Ramunni, A.U., and F. Palmeri, 1985. Use of ultrasonic treatment for extraction of humic acid with inorganic reageants from soil, Org. Geochem. Vol 8, No. 4, pp 241-246.

Stauffer, T., MacIntyre, W.G., and D.C. Wickman, 1989. Sorption of nonpolar organic chemicals on low-carbon-content aquifer materials, Environmental Toxicology and Chemistry, Vol. 8, pp 845-852.

Suslick, K.S., Doktycz, S.J., and E.B. Flint, 1989. On the origin of sonoluminescence and sonschemistry, Sonochemistry Symposium of the International Congress of Pacific Basin Societies: Pacifichem 89, Honolulu, HI.

Muralidhara, H.S., Jiris, B.F., Stulen, F.B., Wickramanayke, G.B., Gill, A., and R.E. Hinchee, 1990. Develpment of electro-acoustic soil decontamination (ESD) process for in-situ applications, National Technical Information Service, No. PB 90-204 728/AS.

Murdoch. L.B., Patterson, B., Losonsky, G., and W. Harrar, 1988. Innovative technologies of delivery or recovery: A review of current research and a strategy for maximizing future investigations, U.S. EPA, Risk Reduction Engineering Laboratory, Cincinnati, OH.

Table 1. 24 hour - adsorbtion data for Rhodamine 6G onto coastal center soil :
(20%:80%-beaumont clay:sand)

Initial Liquid Concentration (mg/L)	Equilibrium Liquid Concentration (mg/L)	Equilibrium Solid Phase Concentration (mg-dye/g-soil)
1.0	0.000	0.040
2.5	0.000	0.100
5.0	0.000	0.200
7.5	0.000	0.300
15.0	0.001	0.600
25.0	0.003	1.000
50.0	0.006	2.000
125.0	0.020	4.900
300.0	0.053	11.998
500.0	0.103	19.996
625.0	0.153	24.994
750.0	0.229	29.991
1000.0	0.406	39.984
1250.0	0.580	49.977
1750.0	1.138	69.954
2000.0	5.800	79.768
2500.0	55.536	97.779
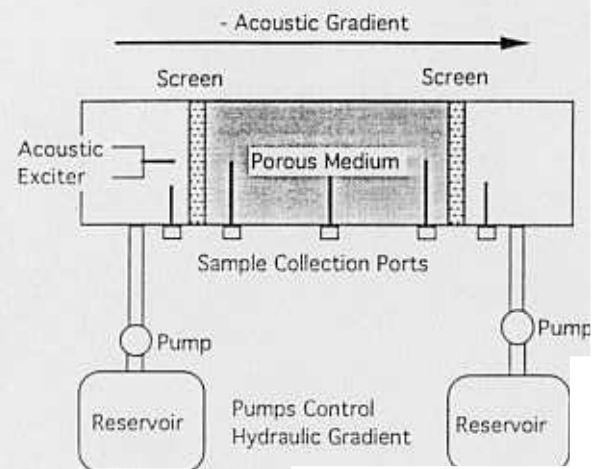
Figure 1.
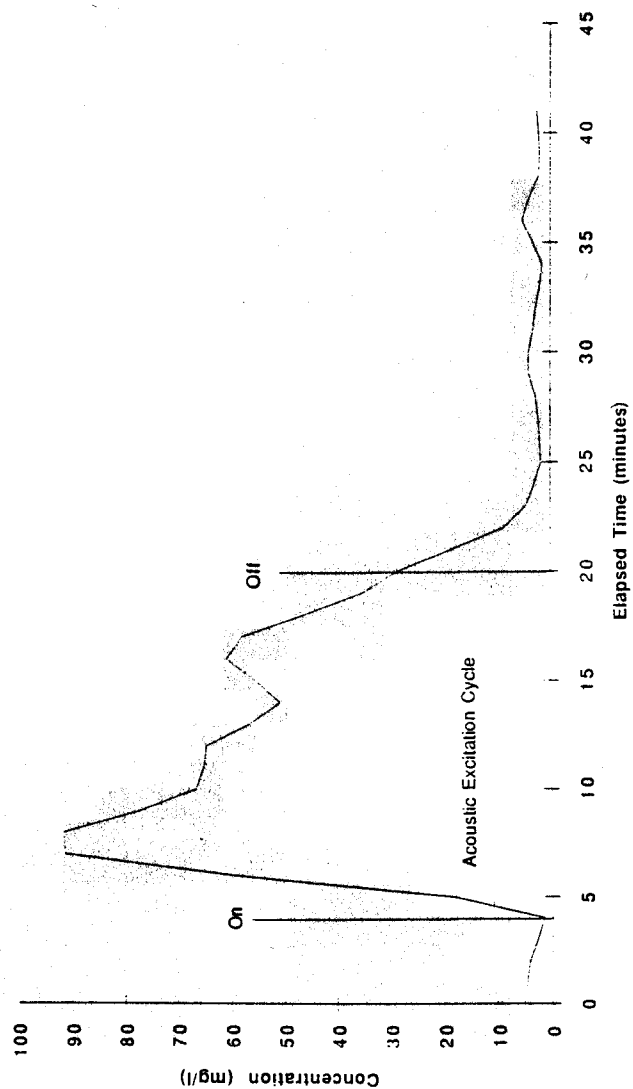
Figure 2
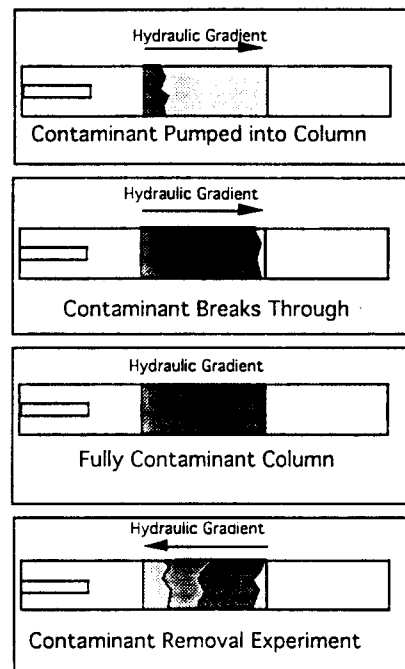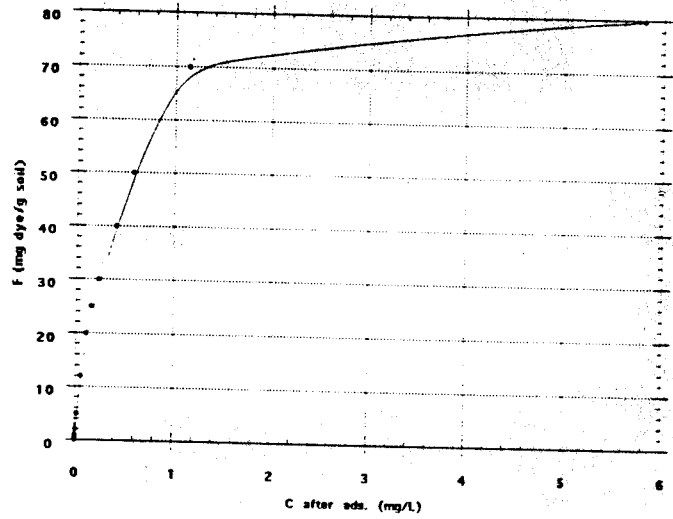
Contaminant Pumped into Column

Contaminant Breaks Through

Fully Contaminant Column

Contaminant Removal Experiment

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

The figure plots Concentration (mg/L) on the vertical axis (0 to 0.45) against Elapsed Time (minutes) on the horizontal axis (0 to 7000). Curves labeled "Treatment Response" and "No-Treatment Response" are shown, with a marker for "Acoustic Excitation Cycles Begin."
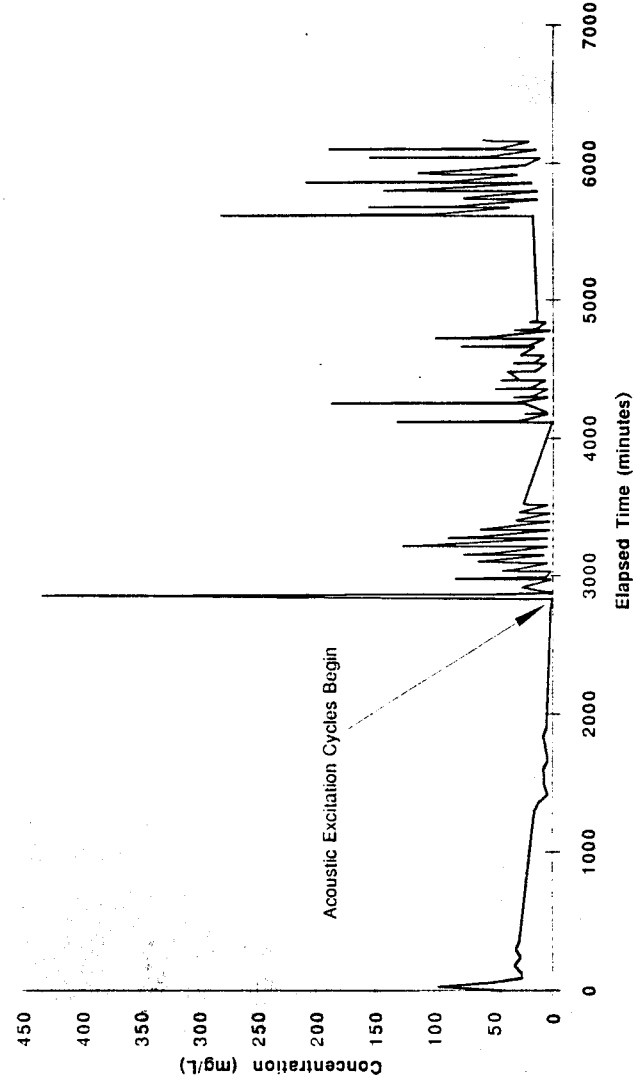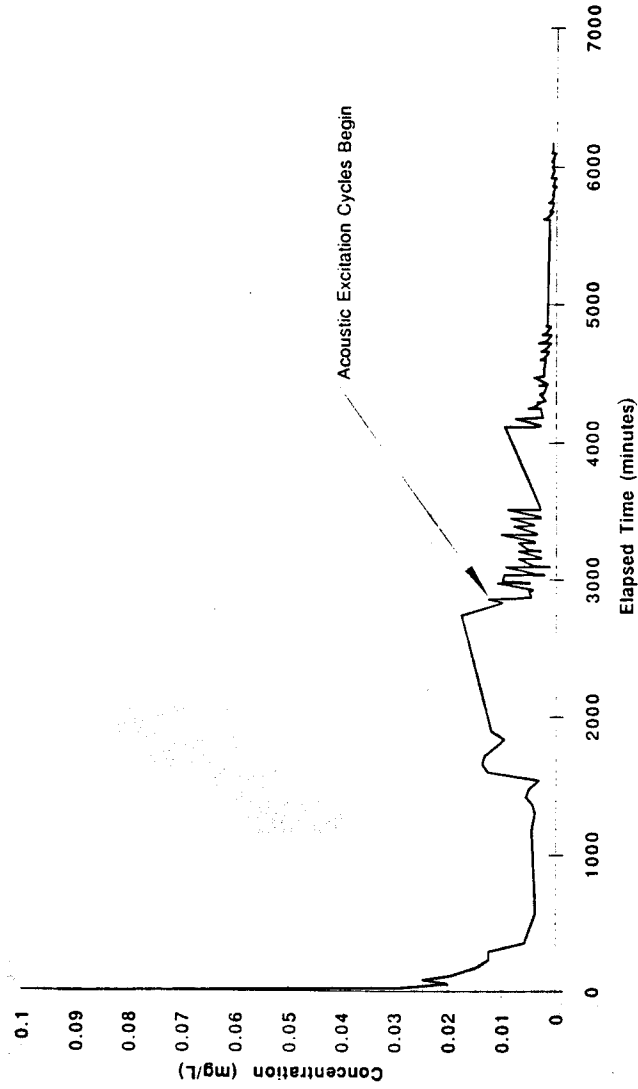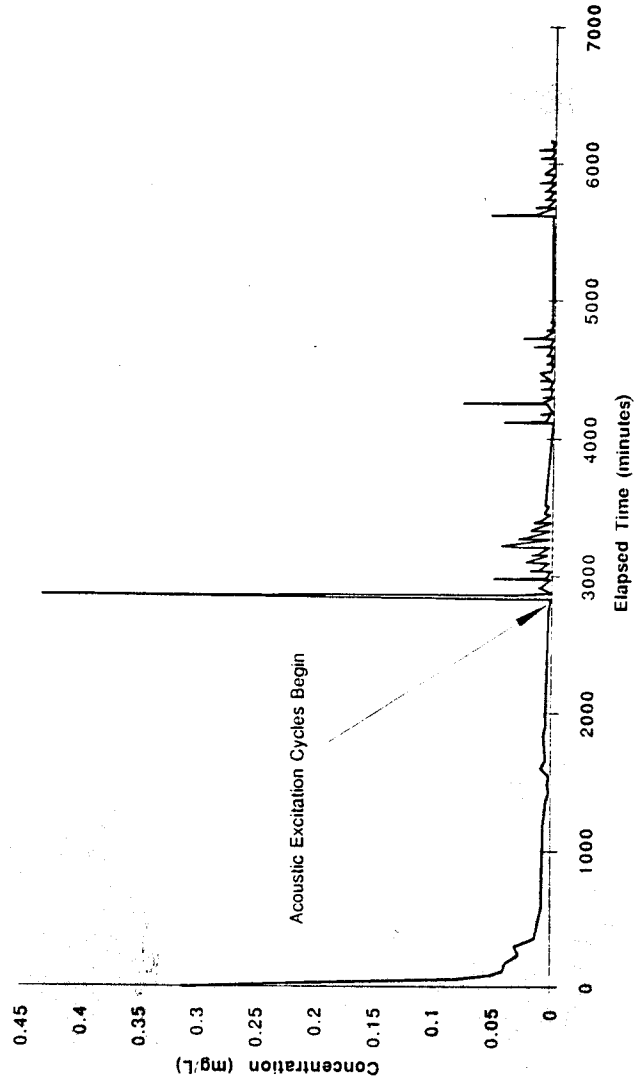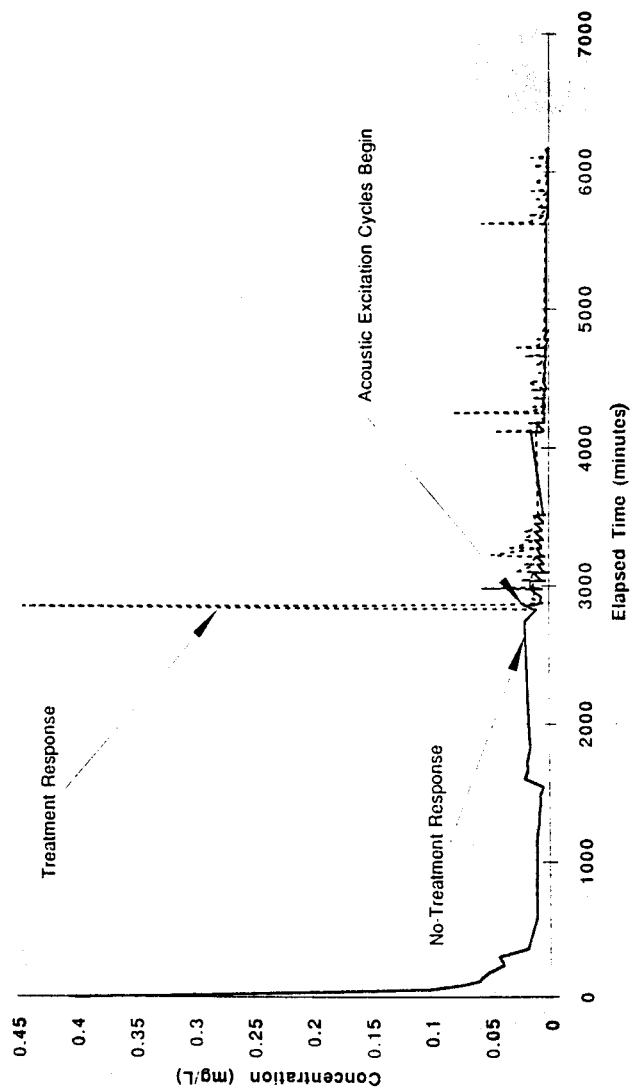
Biographical Sketches

Theodore G. Cleveland (Department of Civil and Environmental Engineering, University of Houston, Houston, TX 77204-4791) received his B.S. in Environmnetal Resources Engineering from Humboldt State University, and his M.S. and Ph.D. degrees form the University of California, Los Angeles. He teaches groundwater hydrology and contaminant hydrology and conducts research in these areas.

Sanjay Garg (Department of Civil and Environmental Engineering, University of Houston, Houston, TX 77204-4791) is a graduate student in Environmental Engineering, prior to coming to the University of Houston, he worked as a civil engineer in India.

— Now at Shell