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SOIL VAPOR EXTRACTION ENHANCED BY OSCILLATORY FLOW

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ABSTRACT

The rate of contaminant removal by soil vapor extraction becomes low when diffusion limits the transfer of vapor from the soil to the channels in the soil where air preferentially flows. This paper suggests that adding an oscillatory component to the pressure and velocity of the subsurface air may increase the transport to the channels of flow, and thereby increase the rate of extraction, when the diffusion limit occurs. Three physical mechanisms by which oscillatory flow may increase the transport are described. Algebraic expressions and numerical estimates are given for the penetration of oscillatory pressure into various soils. Exploratory experiments with a laboratory soil column indicate enhanced extraction when an oscillatory component is superimposed upon nonequilibrium steady flow.

NOMENCLATURE

- Channel radius а
- Ďo Molecular diffusion
- coefficient in air
- С Periodic capacitance $\tilde{\mathbf{F}}^{\mathbf{a}}$
- See Table I
- k Permeability
- L Length of channel
- P₀ P Average pressure
- Amplitude of oscilla'.ory 1
- pressure (may be complex)
- Pg Amplitude of oscillatory
- pressure at x = 0v
- Superficial air velocity (a)

- v Volume of a pore
- v See Table I ve
 - See Table I
- , m Distance
- Z Complex impedance

Greek

- δ Penetration depth
- -γ Ratio of specific
 - heats of air
- Porosity of soil n
- µ Viscosity of air
 - Angular frequency

BACKGROUND

Soil vapor extraction (in various forms also known as "soil venting," "vacuum extraction" and "air stripping") is widely used in the remediation of soils contaminated with volatile organic compounds [Hutzler et al. 1989]. The concentration of the contaminant in the extracted airstream (and consequently, the rate of extraction) may be high when extraction is commenced, but the concentration usually decreases with time for a variety of reasons [Rainwater et al. 1988; EPA 1989]. The concentration in dissolved or adsorbed phases decreases as the contaminant is removed, causing the equilibrium partial pressure of the contaminant in the soil and the extraction rate to decrease according to Raoult's law and Henry's law [DiGiulio et al. 1990; Sleep and Sykes 1989; Stephanatos 1990]. If a contaminant, such as gasoline, is composed of several constituents with different solubilities or vapor pressures, the contaminant will gradually fractionate as extraction proceeds, thereby decreasing the equilibrium concentration in the soil gas [Baehr et al. 1989; Thornton and Wootan 1982].

In some cases, temporary cessation of extraction causes an increased concentration of contaminant ("restart spike") in the extrar ed gas when extraction recommences, indicating that the soil gas is not in equilibrium with the soil during extraction (Payne and Lisiecki 1988). This and other indications of nonequilibrium have led several authors to speculate that flow may occur through channels in the soil somewhat removed from locations of contamination, and that the extraction rate eventually becomes limited by a diffusion process [Thornton and Wootan 1982; Johnson et al. 1987; Johnson and Sterrett 1988; Trowbridge and Malot 1990; DiGiulio et al. 1990; Rainwater et al. 1989]. In response to the diffusion limitation, various researchers have proposed intermittent cessation of extraction, or so-called "pulsed flow" [Keely et al. 1987; Rainwater et al. 1989]. In this process, extraction is simply stopped to allow time for the contaminant to diffuse to the channels where the air flow occurs. This saves equipment operating time, but would not be expected to reduce the calendar time required for remediation because it does not increase the rate of transport between the locations of contamination and the flow channels.

In contrast with pulsed flow, we suggest avoiding the diffusion limitation by superimposing an oscillatory component upon the steady flow of air that is forced through the soil during vapor extraction. By the term "oscillatory flow," we imply that the subsurface air pressure is cycled with a period much less than the time required for the entire volume of soil gas to reach equilibrium with the contaminant. When the extracted gas is not in equilibrium with the soil, we expect that oscillatory flow will increase the concentration in the extracted gas. If the amplitude of the oscillatory component of the pressure gradient is greater than the steady component, the area-average flow velocity (often called the "superficial velocity") will reverse for a portion of each cycle. However, because oscillatory flow may increase the soil ventilation by three separate mechanisms, enhanced extraction might not require flow reversal. In addition to increasing the concentration of contaminants in nonequilibrium extracted gas, oscillatory flow is expected to increase oxygenation of the soil, thereby further accelerating the bioremediation induced by the venting process [Thornton and Wootan 1982; Connor 1988; Hinchee et al. 1989; DiGiulio et al. 1990].

HISTORY OF PERIODIC FLOW IN SOILS

Fukuda [1955] derived an expression for the penetration of a periodic pressure into the soil and noted agreement with archival experimental data. He reasoned incorrectly that the periodic flow would influence moisture transport to a soil depth of only the few millimeters in which the pore volume is displaced by atmospheric air. Farrell et al. [1966] extended the calculation of the penetration of periodic pressures to include the two-dimensional effects of a pressure wave at the soil surface and predicted greater subsurface air motion than predicted by Fukuda. Hanks and Woodruff [1958] measured increased moisture transport through thin layers of soil and mulches caused by wind-induced fluctuations, and they speculated that diffusion was supplanted by some other process. Scotter et al. [1967] directly measured the frequencydependence of the dispersion coefficient during onedimensional flow in soils, noting that the coefficient increased dramatically both with frequency and with the tidal displacement (that is, volume amplitude) of oscillatory flow. These historical investigations indicated that oscillatory flow would enhance transport in soil, but they lacked a clear description of the responsible physical mechanisms.

MECHANISMS OF VENTILATION BY OSCILLATORY FLOW

1. Longitudinal Dispersion. Oscillatory flow in a small channel has the same effect as increasing the rate of diffusion along the channel. In the channel, the axially transported vapor is carried by an oscillating central slug of air [Uchida 1956], which alternately receives and delivers the vapor radially from and to the boundary layer during each cycle. Axial transport along the channel proceeds in analogy with the transport of water by men in bucket brigade, in which the fixed line of men is analogous to the boundary layer, the swinging arms are analogous to the oscillatory slug flow, and the buckets of water are analogous to the transported vapor. Slutsky et al. [1980] presented dramatic evidence of this transport mechanism when they removed CO₂ from the alveoli of a dog's lungs with oscillatory flow of tidal volume much less than the dead volume of the conducting airways. Oscillatory flow is currently used as a medical substitute for natural breathing in humans [Frantz 1985]. Joshi et al. [1983] experimentally verified the theory of oscillatory transport in channels as presented by Watson [1983]. The data of Scotter et al. [1967] for oscillatory transport in soils qualitatively agree with the longitudinal dispersion theory for channels as formulated by Kurzweg and co-workers [1984, 1987].

The rate of longitudinal dispersion in channels depends on the channel radius, the frequency, and the tidal displacement In a cylindrical channel of 1-mm radius with a 5-mm peak-co-peak displacement of gas along the channel, transport equal to twice the static diffusion rate requires that the frequency be approximately 2.4 Hz. As shown in the discussion of penetration below, significant penetration of flow at this frequency occurs only in soils of high permeability.

2. <u>Compressive</u> <u>Advection</u>. Because of the compressibility of air, an oscillatory component of pressure will force air into and out of all gas-filled pore volumes during each cycle, thereby increasing transport of vapors. For example, compressive advection caused by changes in barometric pressure increases the radon flux [Clements and Wilkening 1974].

3. Network Flow Induced by Frequency-Dependent Impedance. The magnitude and phase of an oscillatory pressure in the soil will vary with location and will thereby induce flow along paths that remain stagnant under conditions of steady flow. This is analogous to an R-C electrical network in which an alternating voltage will establish currents between nodes that would be equipotentials with a dc voltage.

Examples. Figure 1 illustrates the transport mechanisms of oscillatory flow. In Fig. 1, A and B represent the main channels for air flow; C represents a smaller network channel between points y and y'; and D represents a pore with a single connecting channel, E. The shaded areas represent locations of liquid, aqueous, or adsorbed phases of contaminant. Under conditions of steady flow, it is assumed that points y and y' are at the same pressure so that no flow occurs in the network channel C. Because pore D has only one opening, no steady flow occurs in it. Steady flow can directly remove contamination from the surfaces of the main



Fig. 1. Conceptual diagram of porosity in the soil.

flow channels, A and B. However, the contaminant is removed from C and D only by diffusion, and the concentrations in C and D will be close to the equilibrium value. Because the effects of steady flow and oscillatory longitudinal dispersion are additive [Watson 1983], oscillatory flow will tend to distribute the concentration more evenly along A and B. This will probably be a small effect, because the transport along A and B is usually dominated by the steady advection. However, by Mechanism (2), an oscillatory component of pressure will cause a periodic flow into and out of D. Even if the tidal volume of this flow is less than the dead volume of the connecting channel E, Mechanism (1) will increase the rate of transport along E.

If the tidal volume into and out of pore D is larger than the volume of E, then D will be partially ventilated during each cycle with air of lower concentration from A. The ratio of the contaminant fluence provided by this oscillatory advection to the fluence provided by diffusion is

fluence ratio =
$$\frac{\omega}{\Pi a^2} \frac{\frac{2P_1}{P_0}}{\frac{D_0}{V}}$$
 (1)

If $P_1/P_0 = 0.05$, $V = 1 \text{ cm}^3$, L = 1 cm, a = 0.1 mm, and $\omega = 0.01/s$ (10 minute period), then the fluence ratio would be

fluence ratio =
$$\frac{0.5 \text{ cm}^2/\text{s}}{D_0}$$
.

For benzene, D is approximately $0.09 \text{ cm}^2/\text{s}$ [Bruell and Hoag 1986]. Therefore, advection caused by such slow pressure cycles might increase the extraction rate from the hypothetical pore by a factor of five, as compared with the extraction rate provided by steady flow and diffusion.

For the example of Fig. 1, it is assumed that the resistive impedance between point y and the pressure source is the same as the resistive impedance between y' and the pressure source; therefore, channel C remains stagnant during steady flow. However, the complex flow impedance between roint y and the pressure source will not, in general, equal the complex impedance between y' and the pressure source because the impedance along any particular path depends upon the frequency and the distribution of both porosity and permeability along that path. Therefore, by Mechanism (3), an oscillatory component of applied pressure can induce an oscillatory flow in a connecting channel that would remain stagnant at zero frequency. If, by chance, the magnitudes and phases of the pressures at y and y' were equal at one particular frequency, they would not be equal at some other frequency.

Enhanced Oxygenation: In situ biodegradation is emerging as a very promising technology for remediation of soils con-

taminated with fuels. By the three mechanisms described above, oscillatory flow will increase the distribution of atmospheric oxygen throughout the soil, and is thereby expected to enhance both bioremediation and in situ decomposition of hydrocarbons.

PENETRATION OF OSCILLATORY FRESSURE

Other authors [Fukuda 1955; Hanks and Woodruff 1958; Farrell et al. 1966] derive the expression for the isothermal penetration of a pressure wave into the soil. In the small amplitude limit, the differential equation for pressure in a porous medium is analogous to the heat conduction equation for which solutions are known for many boundary conditions [Carslaw and Jaeger 1959]. The quantity $n/\gamma P_0$ is analogous to heat capacity per unit volume, and k/μ is analogous to thermal conductivity. For generality, we present here the results for an adiabatic pressure wave; the isothermal result is obtained simply by setting gamma equal to one. Although the flow geometry near an extraction well is often cylindrical, for clarity of illustration we present predictions for a plane pressure wave.

If the oscillatory pressure amplitude, P_1 , is much less than the average pressure, P_0 , a plane wave propagates from a source at x = 0 into a semi-infinite medium according to

$$P_1(x,t) = P_s e^{-X/\delta} \cos(\omega t - x/\delta), \qquad (2)$$

in which δ is the exponential penetration depth. In analogy with the lumped parameters for an electrical circuit, Eq.(3) represents the soil as a complex impedance, for which pressure is the driving element and oscillatory superficial velocity (volume flow rate per cross-sectional area) is the current.

 $P_{g} = v \cdot Z \quad . \tag{3}$

Table I lists algebraic expressions for quantities that may be of use in selecting the frequency and pressure amplitude. Note that the medium is capacitive, so the phase of the oscillatory superficial velocity always leads the phase of pressure by 45°. The periodic capacity, C_a , represents the volume of air that enters and leaves a unit cross-sectional area of soil during each cycle, per unit peak-to-peak pressure change. Qualitatively, one may think of the air as being injected into and withdrawn from a volume of soil with unit cross section and affected length δ , as shown in Fig. 2. R_v is the ratio of the air volume that is periodically injected to the pore volume within the affected length of soil. It is proportional to the air change rate in the soil. F is the ideal mechanical power required per unit crosssectional area (i.e., the mechanical energy flux) at the pressure source. In Table I, V_e is the ratio of air volume injected and removed to the ideal mechanical energy expended at the pressure source. For steady flow, the corresponding volume/energy ratio is $1/\Delta P$, in which ΔP is the steady pressure difference across any length of soil.

 V_m (the product of R, with frequency and penetration depth) is a figure of merit, which is expected to be related to the effectiveness of treatment. This product is presumably related to the rate of overall remediation, as represented by the number of air changes in the



Fig. 2. Diagram of air volume injected into soil.

soil per time and the volume of soil being ventilated. For Mechanism (2), the contaminant removal rate is expected to be roughly proportional to this figure of merit. V is analogous to a the superficial velocity of air in steady flow. The ratio of V to the superficial velocity at a steady pressure gradient of P_s/δ is 0.11/n, which is in the range 0.2 to 0.5 for most soils.

Table II presents estimates of δ , F, and V_ for typical values of the parameters. Preferential flow channels may permit deeper penetration than that predicted by Table II. However, unless there are numerous fractures in the soil, the penetration depth will permit ventilation of a clay layer that is no more than a few centimeters thick. Oscillatory pressure with a period of one hour to one day will penetrate silty soils. In coarse sand, a frequency of a few Hertz may be utilized. Table II shows that the predicted mechanical power for oscillatory flow is generally of no economic consequence. The figure of merit, V_m , increases with frequency; therefore, if well spacing were of no consequence, the more rapid remediation would occur with higher frequency and injection wells would be spaced at smaller intervals to compensate for the smaller penetration depth. In this simple analysis, the economic optimum will be the point at which the cost of decreased well spacing becomes larger than the economic value of faster remediation. The economic optimum is a function of the figure of merit.

EXPERIMENTAL INVESTIGATION

For exploratory experiments, we constructed a vertical soil column in a 1-m long pipe with a 20-cm inside diameter, as shown in Fig. 3. A 75-cm length of soil was held in the

<u></u>	<u>Duantity</u>	Expression
ა	penetration depth	ν ² κ ⁴ ² 0 η
3	complex impedance	$\sqrt{\frac{\omega}{\mu}} \frac{\pi}{\gamma r_0}$
-a	Ratio of air volume displaced each cycle per unit area to P.	$\sqrt{\frac{1}{\omega} \frac{k}{\mu} \frac{7}{\gamma P_0}}$
3.7	air volume displaced each cycle pore volume within length δ	12 Pg P0
7	Techanical power Dross sectional area	P ² V 2μγPg
^v e	Mechanical energy	<u>λΥ2 - 2.90</u> πPs Ps
v _m	air volume displaced per time x pore volume within depth δ	$\delta \qquad \frac{P_{q}}{\gamma e_{0}} \frac{1}{\pi} \sqrt{\omega \frac{k}{\mu} \frac{\gamma e_{c}}{\eta}}$

TABLE I EXPRESSIONS FOR PERIODIC FLOW IN SEMI-INFINITE POROUS MEDIA

Soil Gravel Sand Silt Clav						Clay
		k(cm²)	:0-3	12-7	12-10	-14
erica	ω(===)	δ(m)	7137	71.37	2.257	0.26
14 h	7.27x10-5	F (W/m²)	0.389	3. 89×10⁻³	1.23×10-4	1.23×1077
		V _m (m/h)	4.205	0. 042	1.33×10 ⁻³	1.33×1077
		δ(m)	1920	19.2	0.609	6.09×10-3
1.7 h	0. 001	F(W/m ²)	1.44	0.014	4.56x10-4	4.56x10-6
		۷ _m (m/h)	15.6	0.156	4.93x10 ⁻³	4.93x10-5
		ð(m)	192	1.92	0.0609	6.09x10-4
2.8 5	0.1	F (₩/m²)	14.4	0.144	4.56×10-3	4.56x10-5
		V _m (m/h)	156	1.56	0.0493	4.93x10-4
		δ(m)	60.8	0.608	0.0192	1.92×10-4
: 28 %	1.0	F(W/m²)	45.6	0.456	0.0144	1.44×10 ⁻⁴
		V _m (m/h)	493	4.93	0.156	1.56x10-
		ð(m)	19.2	0. 192	6.09x10-3	5.0 9x 10 ⁻³
.la .	: 10.0	E(W/m-)	144	1.44	.3456	4.56x10-
		۷ _m (m/h)	1560	15.60	0.493	4,93x30-
		ð(m)	10.9	0.109	J.43x10-3	3.43x10-3
1.2 8	31.4	$F(M/m^2)$	256	2.56	0.0809	8.09x10-
		V _m (m/h)	2760	27.65	0.874	8.74x10-

 $\gamma = 1, \eta = 0.3; P_g = 0.01 \text{ atm}; F_0 = 1 \text{ atm}; \mu = 1.8 \times 10^{-5} \text{ Pa s}$



Fig. 3. Schematic diagram of soil column apparatus.

pipe by screens at each end. The unoccupied volumes at the ends of the soil served to spread the air flow. Ports (not shown) were located vertically at 5-cm intervals for connection to pressure transducers, gas sampling, or air injection. Pressure transducers were installed at five of the ports. Dried compressed air with a dewpoint less than -20°C was fed through regulating and metering valves to the bottom of the column. Air leaving the top of the column passed through a metering valve and then to an integrating flowmeter. A small, passive flow of sampling air could be directed from the inlet, outlet, or middle of the column to a cold mirror dewpoint sensor. Adjustment of the outlet metering valve provided the pressure for the sampling flow.

Initially, oscillatory pressure was provided by oscillation of the inlet metering valve. Subsequently, in some cases we wanted the amplitude of the oscillatory component of flow to exceed the steady flow speed, so as to generate reversed flow during part of the cycle. Consequently, we attached a motor-driven piston and cylinder with an 88 cm³ displacement to a screened pipe lying on a diameter near the middle of the column, as indicated in Fig. 3. This device injected opposing oscillatory flows toward both ends of the column from the middle. The air-filled volumes beyond the screens provided an approximately constant-pressure boundary condition to each half of the column. When the penetration depth was longer than a half-length of the column (as has been the case in all experiments to date) the end volumes permitted more oscillatory flow volume per unit pressure amplitude than would occur with a semi-infinite length of soil. (C, of Table I is calculated for a semi-infinite length.) Conditions that generate large tidal velocity with minimal pressure variation would accent Mechanism (1) rather than Mechanism (2). The

experiments reported here were not deliberately designed to select particular mechanisms, but these considerations indicate how individual mechanisms could be explored in future experiments.

To minimize the carbon content and to assure adequate permeability, we chose sand for the initial experiments. The air permeability of the poorly sorted dry sand, measured in the column, was 1.6×10^{-6} cm². At the maximum frequency of the metering valve apparatus (0.74 Hz), the oscillatory pressure amplitude varied linearly with distance along the column, as would be expected for any value of the infinite-medium penetration depth greater than 0.5 m. For porosity in the range 0.3 to 0.5, the expression in Table I predicts the penetration depth to be in the range 0.75 to 1.0 m. We therefore conclude that the flow was as expected.

For experimental convenience, water was chosen as the surrogate contaminant. Approximately 4 liters of water were poured into the top of the column, and the bottom was allowed to drain for several days, leaving the column at field saturation. Air sampling connections for the dewpoint sensor were connected only at the inlet, middle, and exit of the column, so detailed measurements of the profile of water vapor concentration versus distance along the column could not be made. However, all experience was consistent with the following interpretation. At steady air flow rates typical of soil vapor extraction (superficial velocity approximately 0.1 cm/s), the air reached equilibrium with the water in the sand within a distance of less than 10 cm. This was true even when the sand was nearly dry ard the equilibrium dewpoint of stagnant soil gas was less than 10°C. Thus, as extraction proceeded, the sand in the bottom of the column became very dry, while that a short distance above was still at field saturation, as indicated by a dewpoint equal to the dry bulb temperature. In retrospect, sand was a poor choice for exploratory experiments because it permitted uniform, rather than channeled, flow. For the extracted air to be other than at equilibrium, we had to increase the steady superficial velocity to 0.25 cm/s when only the sand in the top several centimeters of the column still held appreciable moisture. Under these conditions, the equilibrium dewpoint could decrease several degrees centigrade during an hour, causing the dewpoint in the extracted air to decrease with time. When the extracted air was not in equilibrium with the sand, and only under this condition, oscillatory flow caused a marked rive in the dewpoint at the outlet of the column, as shown in Fig. 4. In Fig. 4, the oscillatory flow increased the water vapor concentration in the extracted air by one-third to onehalf the difference between the steady-flow value and the equilibrium value. Only the sand at the exit end of the column contained moisture when nonequilibrium conditions were achieved. Being adjacent to a volume at nearly constant pressure, this sand was exposed to oscillations in air velocity

but to only minor oscillation in pressure. We therefore suggest that the experiment presents stronger evidence for Mechanism (1) than for Mechanisms (2) and (3).



Fig. 4. Outlet dewpoint vs time during intermittent oscillatory flow.

CONCLUSIONS

By three potential mechanisms, oscillatory flow is expected to increase the rate of extraction when diffusional limitations prevent the subsurface airstream from reaching equilibrium with the soil. Wells for imposition of an oscillatory component of pressure could be spaced about two penetration depths apart, which is a few meters in many soils. Although incomplete because of budgetary constraints, the experiments with the sand column indicate that oscillatory flow causes an increased concentration of vapor in the extracted air when the air is not in equilibrium with the sand.

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