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In situ vadose zone bioremediation

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Contamination of the vadose zone with various pollutants is a world-wide problem, and often technical or economic constraints impose remediation without excavation. *In situ* bioremediation in the vadose zone by bioventing has become a standard remediation technology for light spilled petroleum products. In this review, focus is given on new *in situ* bioremediation strategies in the vadose zone targeting a variety of other pollutants such as perchlorate, nitrate, uranium, chromium, halogenated solvents, explosives and pesticides. The techniques for biostimulation of either oxidative or reductive degradation pathways are presented, and biotransformations to immobile pollutants are discussed in cases of non-degradable pollutants. Furthermore, research on natural attenuation in the vadose zone is presented.

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Early work in vadose zone bioremediation

Engineered *in situ* bioremediation is a widely used technology for depollution of contaminated sites since the mid 1970ies, with numerous advantages compared to purely chemical/physical techniques or excavation. In the vadose zone, early applications targeted petroleum hydrocarbons, and bioventing [1,2] became rapidly a standard technique for remediation of light petroleum products. Bioventing can be implemented either by air injection, or by soil vapor extraction, which needs a vapor treatment system and is somewhat more expensive and effective. Both techniques convey oxygen (air) to indigenous microbial populations and promote degradation of hydrocarbons to carbon dioxide. In 2010, soil venting/bioventing was the most-widely used *in situ* technique in France [3]. There is usually no need for inoculation of exogenous microorganisms, and additional nutrients such as nitrogen or phosphorus are needed only in nutrient-poor geological settings [4], or sometimes in *ex situ* applications of bioremediation [5].

The objective of this review is to present recent developments in vadose zone bioremediation apart from conventional bioventing, and research on natural attenuation. The article presents first the particular characteristics in the vadose zone, and then presents different strategies for depollution.

Particular characteristics of the vadose zone

Low numbers and activities of microbes

The vadose zone is extending from the soil surface to the groundwater table, including the soil zone, the intermediate vadose zone, and the capillary fringe. The name ‘‘vadose zone’’ is considered to be somewhat more logic than the synonymous term ‘‘unsaturated zone’’ [6]. The vadose zone has also been called ‘‘No-man’s land’’ [6], because relatively little research has been performed below the soil horizon. However, the vadose zone is far from being free of life. The numbers of microbial cells generally decrease in the intermediate vadose zone by one to four orders of magnitude, but remain still at fairly high counts (in the order of 10^{-6} to 10^{-8} cells per gram of soil, e.g. [7–10]). In the capillary fringe and the saturated zone, the numbers of bacteria and protozoa can increase again. The catabolic activities of these organisms are lowest in the intermediate vadose zone. However, the catabolic diversity does not change substantially from the intermediate vadose zone to the saturated zone [9,10].

Bioremediation can generally be based on two major strategies: biostimulation or bioaugmentation [11]. The first involves the supply of stimulating agents (nutrients, electron acceptors or donors) to indigenous microorganisms, whereas the second involves the injection of pregrown microbial cultures to enhance degradation. For *in situ* vadose bioremediation, bioaugmentation is very difficult to practice: fine-grained porous media such as sand or silt are very effective filters for microbial cells, and therefore pregrown cultures can hardly be transported to deep vadose zones composed of fine sediments. Therefore, it is not surprising that bioaugmentation has only been applied in few studies of *in situ* bioremediation, in fractured geologic media [12,13••] or in surface soils [14,15] where microbes can be transported in macropores. Elsewhere, biostimulation was applied to stimulate growth of indigenous microbes. In contrast, bioaugmentation is common in *ex situ* applications [5]. Ongoing innovative work is investigating whether microbes can be transported by means of fungal hyphae in fine-grained porous media [16•,17,18].

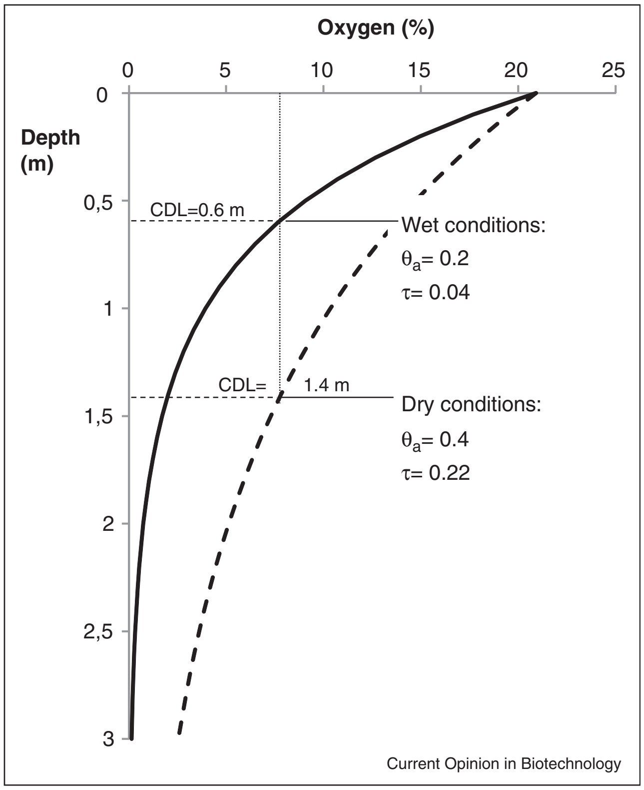
Gas-phase diffusion as dominant transport process

Advective fluxes of fluids in the natural vadose zone are small, consisting in vertical water flow velocities in the order of 1 m/year, and gaseous advective fluxes caused by barometric pumping in the order of cm/day [6]. For gases, the major transport process is gas-phase diffusion. For reactive gaseous compounds, a useful property to scale vadose zone processes is the characteristic diffusion length (CDL) which is defined as (Eqn (1)):

$$\mathrm{CDL} = \sqrt{\frac{\tau D_{m,\mathrm{air}}}{k}} \qquad (1)$$

where CDL is characteristic diffusion length in meters, τ is the tortuosity factor, $D_{m,\mathrm{air}}$ is the molecular diffusion coefficient of the reactive gas in air ($\mathrm{m}^2\ \mathrm{day}^{-1}$), and k is a first-order reaction constant for the reactive gas (day^{-1}). The usefulness of CDL for the characterization of limitations is illustrated in Figure 1 for the diffusion of oxygen into a hydrocarbon-contaminated soil. The CDL represents the length scale across which diffusion tends to homogenize gradients induced by reaction. In homogeneous settings with spatially uniform reaction, a reactive gas will be depleted at depths equaling approximately three times the CDL (Figure 1). In aquifer sediments contaminated by chlorobenzenes studied in laboratory columns, O_2 was depleted within 25 cm [19•]. Note that at very high water saturations, τ approaches zero, and only aqueous-phase diffusion is active. This reduces D_m by about four orders of magnitudes, and thus CDL by two orders, from the m-scale to cm-scale. In water-logged soil, oxygen can be depleted on a cm scale in soil aggregates.

Figure 1



Characteristic diffusion lengths (CDL) of oxygen in a sandy vadose zone where hydrocarbon oxidation consumes O_2 with a rate *k* of 0.2 day^{-1}. The tortuosities τ are calculated for wet and dry conditions with the relation of Moldrup $\tau = \theta_a^{2.5}/\theta_t$ [55], where subscripts *a* and *t* design air-filled and total porosity, respectively.

Strategies employed in vadose zone bioremediation

Table 1 presents an overview of applications in vadose zone bioremediation. The techniques can be grouped into four strategies: firstly, biostimulation of oxidative transformation (mostly mineralization to CO_2). This requires injection of oxygen to the vadose zone; secondly, biostimulation of reductive transformations of contaminants to harmless products. This requires the injection of electron donors and exclusion of oxygen; thirdly, stimulation of biofilms for pollutant immobilization, e.g. promotion of aerobic biofilm growth for immobilization of mobile hexavalent chromium to immobile trivalent chromium; fourthly, rhizosphere technologies: plant roots and associated mycorrhiza and bacteria transform pollutants under ill-defined redox conditions. These strategies permit actually to target an interestingly wide list of pollutants (Table 1).

Selected studies

Hydrocarbon vapor attenuation

The short CDL of oxygen causes frequently insufficient aeration below large buildings, causing pollutant vapor intrusion through fissures into indoor environments. A recent study has shown the successful installation of an aerobic vapor biobarrier below a building [20••]. To this end, horizontal air delivery wells were installed 3 m below a 200 m^2 building foundation and connected to low-delivery air pumps (max. 10 L/min). This resulted in sub-slab aeration and massive reduction of hydrocarbon and methane concentrations. The technique is much more economic than pressurization in buildings and may become a standard technique in hydrocarbon-vapor impacted vadose zones.

Naphthalene

Naphthalene is the most volatile polyaromatic hydrocarbon. The migration of its vapors from creosote-contaminated groundwater to surface soil poses a risk at the former railroad tie operation site at Oneida, Tennessee [21]. Phytoremediation by poplar trees *Populus deltoides* × *nigra* DN34, was originally designed to provide hydraulic control of the dissolved contaminant plume. Detailed field measurements and studies of soil columns from the planted vadose zone above the creosote plume and from unplanted reference locations revealed a marked stimulation of naphthalene vapor attenuation