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Microbial processes influencing the transport, fate and groundwater impacts of fuel ethanol releases

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Fuel releases that impact groundwater are a common occurrence, and the growing use of ethanol as a transportation biofuel is increasing the likelihood of encountering ethanol in such releases. Microorganisms play a critical role in the fate of ethanol-blended fuel releases, often determining their region of influence and potential impacts. This review summarizes current understanding on the biogeochemical footprint of such releases and the factors that influence their natural attenuation. Implications for site investigation, risk assessment and remediation strategies are also addressed along with research priorities.

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Introduction

The use of renewable transportation fuels (biofuels) is rapidly growing to alleviate dependence on imported oil and enhance energy security, as well as to mitigate air pollution and greenhouse gas emissions by fossil fuel combustion [1^{••},2,3[•]]. Currently, the major commercialized biofuel products include ethanol and biodiesel. Ethanol holds a much larger global market share than biodiesel (23 483 vs. 5510 million gallons/year) [4].

Incidental and accidental fuel releases that impact groundwater are a common occurrence and the likelihood of encountering biofuels (mainly ethanol) in such releases is increasing. Thus, it is important to understand how such releases behave and affect groundwater geochemistry, and how indigenous microorganisms respond and affect their migration, fate, and overall impact. This information is critical to optimize

site characterization, risk assessment and remediation practices when dealing with releases of current and future biofuel blends.

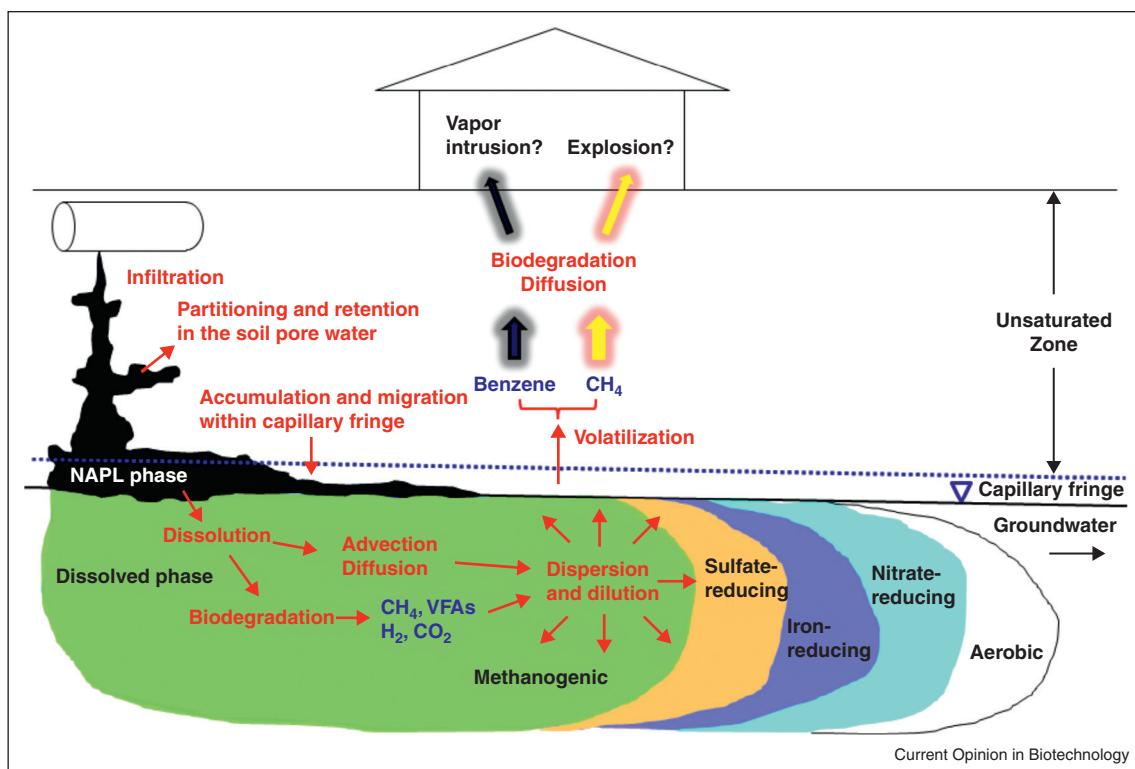
Physical behavior of ethanol-blended fuel releases

When an ethanol-blended fuel release occurs, it infiltrates as a non-aqueous phase liquid (NAPL) through the unsaturated zone to the water table and forms a floating NAPL pool at the water table when a sufficient volume is spilled (Figure 1). Ethanol will partition into pore water throughout the unsaturated zone [5–7] and will tend to accumulate at the water table interface and the capillary fringe owing to its buoyancy [6,8–11]. For high content ethanol fuels (e.g. E95, which has 95% ethanol and 5% gasoline by volume), the fuel will probably migrate through this interface, initially as a water miscible phase, and then separate into two phases as the fuel becomes diluted, precipitating a new NAPL phase along its path [9,10,12]. Pore water containing high ethanol concentrations will also be enriched in hydrocarbons owing to their enhanced solubility in the presence of ethanol (cosolvent effect) [13–16]. Thus, different domains of microbial activity are likely to develop: a region of anaerobic activity in the core of a contaminant plume in the saturated zone (where the biochemical oxygen demand [BOD] exerted by the release exceeds the available dissolved oxygen) with aerobic degradation occurring at the fringes of the plume; a second region of high anaerobic activity in the capillary zone (except in cases when ethanol concentrations are sufficiently high to be toxic to microbial processes); and a third region in the unsaturated zone where aerobic degradation of methane (emanating from the anaerobic fermentation of ethanol in the capillary zone) is predominant (Figure 1).

Biodegradation of ethanol-blended fuel

Direct exposure to ethanol in drinking water has minimal adverse impacts on human health, but ethanol may increase the exposure potential of toxic fuel constituents (i.e. benzene, toluene, ethylbenzene and xylenes [BTEX]) by hindering their biodegradation and increasing their region of influence [1^{••}]. Because ethanol generally biodegrades faster than BTEX, the latter tend to form larger and more persistent plumes than ethanol. Therefore, substrate interactions during ethanol and BTEX degradation and their effect on plume dynamics (range and longevity) have received considerable attention [17–25,26[•]].

Figure 1



Fate, transport, and potential impacts of ethanol-blended fuel releases.

During transport in groundwater, ethanol and BTEX can undergo a series of biotransformations which can be performed by a variety of microorganisms in aerobic or anaerobic environments [1^{••}]. The relatively high concentration of ethanol found in recently-impacted groundwater exerts a high BOD that rapidly consumes the available dissolved oxygen and other terminal electron acceptors in the vicinity of the source zone, which results in the development of strongly anaerobic, fermentative methanogenic conditions (Figure 1). Nevertheless, aerobic microbial activity might be important for the natural attenuation of the leading edge of the plume.

Under aerobic conditions, BTEX are activated by oxygenases to form catechol or structurally related compounds, which subsequently undergo ring fission to byproducts such as acetyl-CoA, acetaldehyde and pyruvic acid that enter central metabolic pathways such as Krebs' cycle (for final mineralization to CO₂) or glycolysis [27]. Ethanol can also be aerobically metabolized to the pivotal intermediate acetyl-CoA via acetaldehyde and acetate [1^{••}].

Under anaerobic conditions, BTEX are initially transformed via different pathways (*fumarate addition*, *O₂-independent hydroxylation*, and *carboxylation*) to a common aromatic intermediate, benzyl-CoA, which subsequently undergoes ring reduction followed by hydrolytic cleavage

[28^{••}]. Further anaerobic transformations in anaerobic (methanogenic) food webs eventually produce acetate, which is finally mineralized by acetoclastic methanogens to produce CH₄ and CO₂. BTEX fermentation also generates H₂, which is consumed by different commensal anaerobes, including hydrogenotrophic methanogens. Ethanol is similarly transformed to acetate and H₂, which are subsequently metabolized by methanogens to produce CH₄ and CO₂ [1^{••}]. Depending on the available electron acceptors, sulfate reducers, iron reducers, and denitrifiers could also participate in the anaerobic degradation of ethanol-blended fuel, and spatially distinctive redox zones could form in plume (Figure 1).

How will ethanol affect BTEX biodegradation?

The major impact from ethanol may be related to its inhibitory effect on BTEX biodegradation (Table 1), which (depending on the release scenario) may increase the likelihood of BTEX to reach receptors (longer plumes) as well as the potential duration of exposure (more persistent plumes).

Benzene, which is the most toxic compound of the BTEX and often drives the need for cleanup action, is relatively resistant to degradation under anaerobic conditions [29], while ethanol and its degradation byproducts (e.g. volatile fatty acids [VFAs]) are easier to degrade under both

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