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Specific composition, microstructure, and grass vegetation support natural attenuation in aged tar contaminated soil

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The development of effective remediation strategies for soils contaminated by aged non-aqueous phase liquids like tars requires detailed investigation of composition, microstructure and microbial communities. We studied an aged tar spill with an overgrowing grass vegetation at a former manufactured gas plant site in Germany. The soil contained 10-120 g kg^{-1} petroleum hydrocarbons, up to 26 g kg^{-1} potentially toxic metals, and up to 100 mg kg^{-1} polycyclic aromatic hydrocarbons. Although these substances are considered toxic and recalcitrant, the microbial biomass was up to twice as much in contaminated layers than in uncontaminated layers of the control soil. We assume the high content of vital elements, such as C (up to 500 g kg^{-1}), S (5 g kg^{-1}), P (4.8 g kg^{-1}), Fe (65 g kg^{-1}), and N in plant residues, compensates possible toxicity.

Investigation of the 2D soil microstructure on thin sections with digital light and scanning electron microscopy showed increased total porosity (2-3 times more than in control) and the share of coarse wide pores (> 50 µm, root channels and large cracks) in contaminated layers. Within the root channels aerobic conditions persist, with free inflow of soil solution and supply of root exudates.

Tar dominated particles between the coarse pores had small isolated pores, and the average distance to the next pore within the particles (assessed by Euclidian distance) was about 3 times higher than for the control soil. This highlights anaerobic conditions within the pores, where tar borne compounds are the source of nutrition and energy.

FTIR microspectroscopy showed oxidized tar on root coatings and near some isolated pores. Natural attenuation of the contaminant proceeds both under aerobic and anaerobic conditions.

Positive matrix factorization analysis of EDX spectra allowed us to map the spatial distribution of different components (quartz, feldspars, secondary minerals, metal-rich particles, tar and the embedding resin). We found presumably authigenic Fe minerals within small isolated pores and along root channels. Based on XANES spectroscopy and the difference between total Fe and Fe in Fe oxides (Fe_{DCB}), they contained Fe^{2+} and Fe^{3+} in different proportions, which suggests Fe reduction to be an accompanying process during tar attenuation.

The 16S rRNA analysis showed similar microbial communities on the rooted rim of the spill and the control soil. The community in the centre of the spill was less diverse and remarkably different. The contaminated profiles contained specific functional groups of bacteria (e.g. Fe-reducing Geobacteraceae or N-fixing Rhizobiales). Microfluidic droplet cultivation facilitated abundant microbial growth from tar layers under both aerobic and anaerobic conditions.

We conclude that aged tar is used as a substrate by the microbial communities, especially in the presence of grass vegetation. Natural attenuation of tar occurs in hotspots under either oxic (root channels and large connected voids) and anoxic (small isolated pores) conditions and is coupled with reduction of Fe.