

Processes influencing PAH (polycyclic aromatic hydrocarbons) distribution in freezing soils

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Due to limited resources the Arctic's reservoirs of oil, gas and minerals are becoming increasingly interesting. The exploration of these resources takes place to a limited extent already. So the vulnerable environment and the permafrost-affected soils in particular are threatened by contamination due to exploration activities and accidents during storage and transportation under difficult conditions and over long distances.

Permafrost-affected soils with their unique and specific properties like active layer, frozen fringe, several freeze-thaw cycles throughout the year, specific water balances and limited microbial activity, interact with contaminants in specific ways, leading to typical distribution patterns in the soil.

Therefore the fate of organic contaminants in permafrost-affected soils and the effect of changing properties due to climate change on the behavior of contaminants are research questions of high relevance. With respect to oil and gas exploration this study deals with Polycyclic Aromatic Hydrocarbons (PAH).

The objectives are (i) to investigate the distribution of PAH in different soil materials influenced by a one-directional freezing process (ii) to differentiate direct and indirect effects of the freezing process in soil on the PAH distribution and (iii) to analyse the impact of the presence of petroleum hydrocarbons on the effects of freezing on PAH distribution.

The freezing process causes heat and water movement in the soil. Based on these fluxes it is hypothesized that zones of higher PAH concentrations in the soil develop, even if the contaminated areas are not frozen. At the freezing front the phase transition of water to ice influences the behavior of the contaminants directly due to physical forces. This is especially directed to non aqueous phase liquids (NAPLs) in the soil system, like petroleum hydrocarbons. Therefore it is hypothesized that the forming ice replaces the NAPLs and a zone of higher contaminant concentration right in front of the freezing front will form.

Laboratory column experiments with ^{14}C labeled PAH, different soil materials and different freezing scenarios were conducted, using four 10 cm high columns. The soil material was contaminated with ^{14}C - Anthracene and ^{14}C - Pyrene, respectively. In one set of experiments the soil material was additionally contaminated with a North Sea crude oil. The experiments lasted 7 days and consisted of a single freezing process. Subsequently the columns were sampled, taking 0.3 cm to 2.0 cm thick slices. The samples were prepared for analysis in three parallels.

Results for the first set of experiments show a strong increase in water content at the freezing front. The high water fluxes did not influence the PAH distribution in the different soil materials, due to the low water solubility of the PAHs. However in the presence of petroleum hydrocarbons, the freezing process had an impact on the distribution of PAH in the soil. The concentration of the PAH decreased right before the freezing front and increased just below it. This reveals that the PAH were excluded from the soil pores together with the NAPLs during ice formation at the freezing front. Differences in the results of sandy (Su2) and silty (Ut2) material will be discussed.