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INFLUENCE OF FREEZING ON ACCUMULATION AND REDISTRIBUTION OF HYDROCARBON IN SOILS

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ABSTRACT

Laboratory investigations are an important tool in the study of spreading, transportation and accumulation of hydrocarbon pollution in permafrost and seasonal frozen soils. The present work has focused on the laboratory behaviour of hydrocarbon contamination in freezing soils.

The results from one-dimensional freezing tests show that freezing of the soil can cause oil accumulation and concentration in the thawed zone in front of the freezing front or accumulation of hydrocarbon pollution in the frozen zone of freezing soil.

The first case is characteristic for freezing of coarse, frictional (sandy) soils and fine soils without ice segregation. The second case is characteristic for fine, cohesive (clayey) soils with substantial moisture migration in the frozen zone. In this case the oil accumulation in the frozen zone occur through advection.

This means that hydrocarbon pollution of initially non-polluted soils can occur under open system conditions. For example, the presence of oil polluted ground water beneath the zone of seasonal frost, can cause pollution of the soil subjected to freezing.

Spreading, transportation and accumulation of hydrocarbon pollution in freezing soils will depend on many factors, the most important being the physical properties of soil and oil, and freezing conditions

INTRODUCTION

Production and transportation of oil in permafrost regions often results in extensive oil pollution of the active layer. Penetration of hydrocarbon pollution in the active layer is often considered to only occur during the summer, when the active layer is thawed (Solntseva et al. 1980; Guseva 1995; Solntseva 1998; Collins et al. 1993).

It is well known that the freezing process may influence transportation and redistribution of substances in freezing fine soils (Marion 1995; Fundamentals of Geocryology, 1995).

The influence of the freezing processes on accumulation and redistribution of oil pollution in the active layer has not been studied in detail in Russia, but some data on diesel fuel is available from Neufeld and Biggar (1996) and crude oil, Chuvilin et al. (1998). In addition, some researchers have investigated the change in composition, structure and properties of freezing soils polluted by hydrocarbons (White et al. 1999; Sheshin et al. 1992). However, additional research is required to gain a better understanding of the physical processes involved in accumulation and redistribution of hydrocarbon pollution in freezing soils.

In this article the results from experimental laboratory tests on accumulation and redistribution of oil pollution in freezing soils are presented. Two base cases are considered:

1. Oil redistribution in polluted soils subjected to one-dimensional freezing and
2. Oil accumulation in "clean" soils subjected to one-dimensional freezing connected to an external moisture and oil flow.

The mechanism of oil redistribution is analysed and the influence of the soil structure and oil concentration on the characteristics of the mass-transfer processes isinvestigated.

METHODOLOGY

The experiments were carried out on remoulded samples of sand (quartz), sandy loam (polymineral) and kaolinite clay. The experiments were carried out on three soil types (see Table 1) polluted with two types of oil (see Table 2) at different concentrations.

The experiments were carried under two different boundary freezing conditions:

i) *Closed system freezing (undrained)*, without external moisture and oil flow to and from the sample during freezing. The soil samples were mixed with oil before freezing started.

ii) *Open system freezing (drained)*, where moisture and oil was allowed to flow to (and from) the sample during freezing. The initially "clean" soil specimen was subjected to an external (open system) migration flow of moisture containing oil during freezing. The bottom part of the freezing sample was in contact with sand saturated with water and oil. This was meant to simulate the natural condition when an aquifer beneath the freezing front is polluted by oil.

Table 1 Characteristics of soils

Soil	Density of the solid particles, ρ_s (g/cm^3)	Plastic limit w_p (%)	Liquid limit w_L (%)	Particle composition (%)		
				2-0.05 mm	0.05-0.002 mm	<0.002 mm
Sand	2.65	-	-	94.8	3,0	2.2
Sandy Loam	2.68	22.2	26.2	41.8	52.2	6.0
Clay	2.66	34	45.8	4.5	62.9	32.6

Table 2 Characteristics of oils

Oil	Hardening temperature (°C)	Density, d_4^{20}	Temperature of boiling (°C)	Fraction composition (%)
#1	-11°C	0.9200	58	58 to 100^{0}C: 3,6 100 to 200 °C: 13,1
#2	+2°C	0.8692	60	122 to 150^{0}C: 2,6 150 to 200^{0}C: 17,1

In both cases one-dimensional freezing from the top downwards was applied to the sample. The temperature boundary conditions during one-dimensional freezing were constant temperatures of:

-7°C or -15°C (top of sample) and +6°C (bottom of sample). The duration of each test was approximately 24 hours.

The temperature changes in the soil specimen were monitored by thermocouples. The test was concluded when a stationary temperature distribution in the sample was reached (based on data from the thermocouples).

The experiments were carried out on saturated remoulded samples with dimensions 5 cm x 5 cm x 15 cm.

The samples were prepared and saturated with distilled water and oil (for the first series of experiments) and distilled water (for the second series of experiments) The clay samples were consolidated up to 200 kPa.

After each test was completed, the soil specimen was analysed and density, water content (W) and oil concentration (Sn) at different levels were determined. Oil concentration could be determined based on oil extraction from the soil using chloroform.

RESULTS

The experimental results showed two modes of behaviour: i) Oil was expelled from the freezing front in the oil polluted soil during freezing and accumulated in the thawed zone. ii) Oil polluted water migrated from the thawed zone to the freezing front (and further into the frozen zone).

The first mode of behaviour was characteristic for oil polluted sandy soils, where migration of moisture in the frozen zone is limited. This behaviour was also observed for oil polluted fine soil without ice segregation.

The physical explanation for expulsion during freezing is connected to hydrostatic water pressure in the soil pore space and the kinetics of ice crystal growth. Ice crystals growing in the pore space of the soil can either expel oil or lock-in oil, depending on the speed of crystallisation.

As expected, the expulsion rate increases with decreasing penetration velocity of the freezing front.

Figure 1a shows the results from closed system freezing of a specimen of oil polluted sandy soil. It can be observed from the figure that the oil concentration after freezing is substantial higher in the thawed zone compared to the frozen zone. This is due to the relative high permeability of the sand and that moisture migration does not occur in the frozen zone. The oil expulsion can be observed visually on the sample, see Figure 2. The bottom part of the frozen zone (area 2, Figure 2) has a lighter colour, due to oil expulsion and reflects the lower oil concentration.

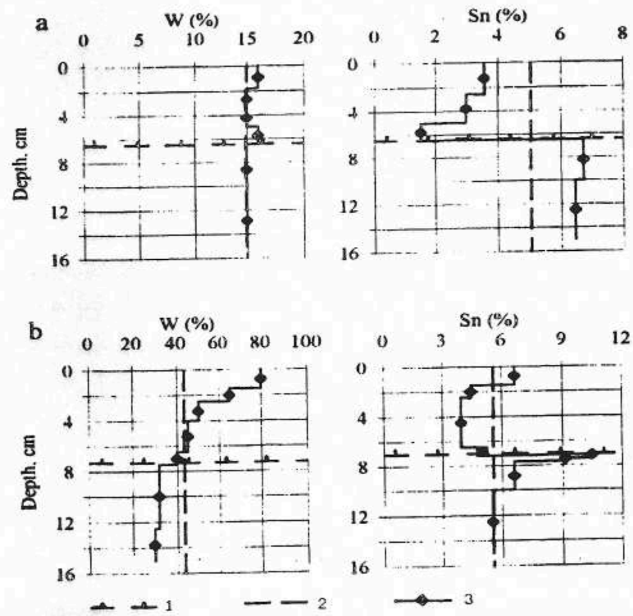

Figure 1 *Closed system freezing. Water content (W) and oil#1 content (Sn) in soil sample after freezing for a) Sand and b) Clay, 1: Freezing front at the end of the experiment; 2: Initial water and oil content. 3: Final water and oil content.*

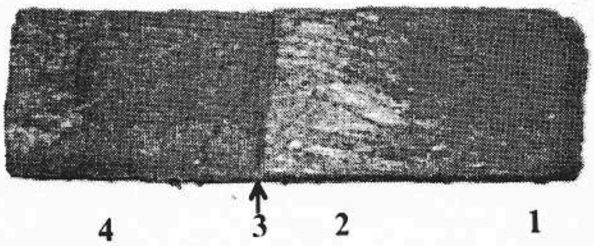

Figure 2 *Photograph of oil polluted (oil #1, initial oil content 5 %) sand sample after freezing: 1: Upper part of frozen zone; 2: Lower part of frozen zone; 3: Freezing front; 4: Thawed zone.*

It should be noted that in sandy soils, the oil accumulation observed in the thawed zone is relatively uniform. In clayey soils the maximum oil concentration is found in the thawed zone just beneath the freezing front (see Figure 1).

The increase of oil çoncentration in the frozen zone of freezing soils, is caused by intensive moisture migration from the thawed zone to the freezing front (and into the frozen zone), see Figure 1b.

If the migration flow is high, the migrating moisture can transport oil through advection from the soil pore space in a similar manner to what occurs with salt ions in freezing saline clayey soils (Chuvilin 1999).

Intensive mass-transfer from the thawed zone to the freezing front of soils during freezing can result in pollution of initially non-polluted soil.

Figure 3 shows the results from open system freezing of a sandy loam (clayey soil). It can be observed from the figure, that also in this case there is an increased oil concentration in the thawed zone just beneath the freezing front. It is believed that this phenomenon is caused by change in freezing parameters during the process.

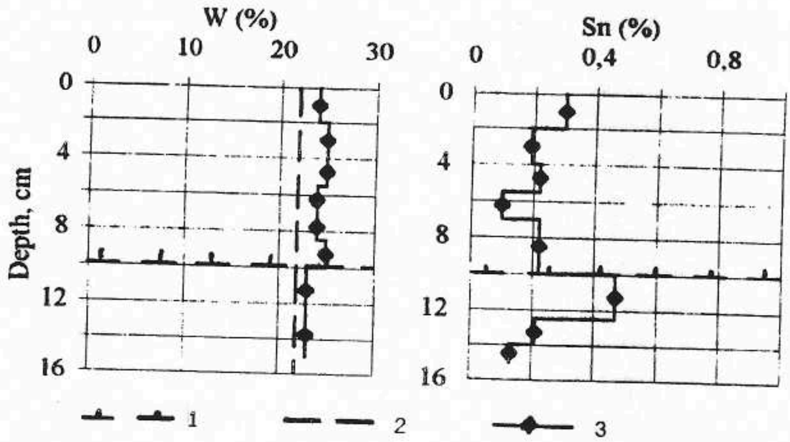

Figure 3 *Open system freezing. Water content (W) and oil#2 content (Sn) in sample of sandy loam after freezing. , Initial Sn=0%, Initial W=22%. 1: Freezing front at the end of the experiment; 2: Initial water and oil content. 3: Final water and oil content.*

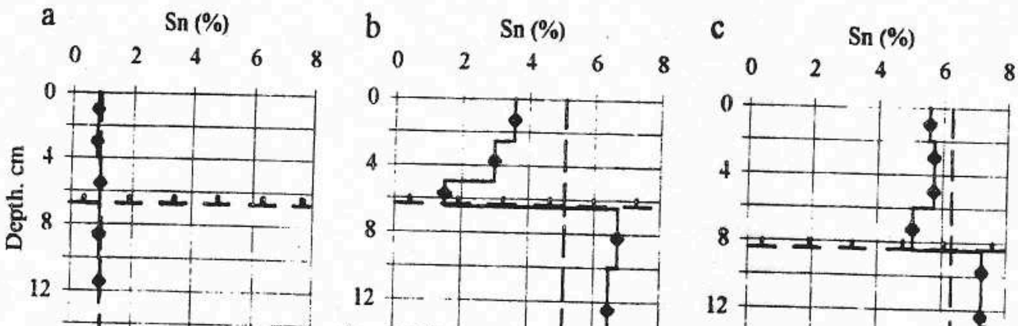

Figure 4 *Closed system freezing. Water content (W) and oil content (Sn) in sample of sand after freezing;*
a: oil #1, Initial Sn=0,88%, Initial W=20%
b: oil#1, Initial Sn=5%, Initial W=16%
c: oil #2, Initial Sn=4,9%, Initial W=16%;
1: Freezing front at the end of the experiment; 2: Initial water and oil content. 3: Final water and oil content.

Reduced frost penetration velocity will reduce the driving forces of moisture migration. This means that there can be a change in the direction of oil transportation and flow during freezing: Initial water/oil migration to the freezing front followed by oil expulsion at lower thermal gradients.

The oil redistribution in freezing polluted soil is closely connected to the initial oil concentration and structure (properties) of the hydrocarbons. The experiments showed that the redistribution is sharply reduced with decreasing initial oil concentration (see Figure 4a).

Apparently, at low oil concentrations, the oil is immobilized in the soil pores and is kept in place by growing ice crystals during freezing.

At the phase transition temperature, the viscosity of the oil increases with decreasing hardening temperature.

The redistribution of the oil pollution, therefore, increases with decreasing hardening temperature of the oil (see Figure 4b,c).

SUMMARY AND CONCLUSIONS

The experimental laboratory tests presented in this paper have shown that significant oil pollution redistribution take place during freezing of soils. The experimental results showed two modes of behaviour: i) Oil was expelled from the freezing front in the oil polluted soil during freezing and accumulated in the thawed zone. ii) Oil polluted water migrated from the thawed zone to the freezing front (and further into the frozen zone).

The first mode of behaviour was characteristic for oil polluted sandy soils, where migration of moisture in the frozen zone is limited. This behaviour was also observed for oil polluted fine soil without ice segregation. The physical explanation of expulsion during freezing is connected to the hydrostatic water pressure in soil pore space and the kinetic of ice crystal growth. Ice crystals growing in the pore space can either expel oil or lock-in oil, depending on the speed of crystallisation. As expected, the expulsion rate increases with decreasing penetration velocity of the freezing front.

Freezing of oil polluted soils may, therefore, be a method for reducing the oil concentration in the soil.

It has also been shown that oil pollution may migrate from the thawed zone to the freezing front. This means that hydrocarbon pollution of initially non-polluted soils can occur under open system conditions. For example, the presence of oil polluted ground water beneath the zone of seasonal frost, can cause pollution of the soil subjected to freezing.

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