Factors affecting spreadability and transportation of oil in regions of frozen ground

E.M. Chuvilin, N.S. Naletova, E.C. Miklyaeva, E.V. Kozlova
Department of Geocryology, Faculty of Geology, Moscow State University, Vorobjovy Gory, Moscow 119899, Russia
A. Instanes
Norwegian Geotechnical Institute, PO Box 3930, Ullevaal Stadion, N-0806 Oslo, Norway

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ABSTRACT. The physical behaviour of oil interacting with soils subjected to seasonal frost or permafrost was investigated. An experimental programme was carried out to investigate the transportation and spreading of oil on a frozen surface, and transportation and accumulation of oil into freezing or frozen soils. The results show that spreading of oil at the surface at air temperatures below freezing depends on oil composition, soil temperature, and the type of mineral surface. It was observed that an ice surface has the least spreading and the greatest wetting angle of the surfaces studied. The oil penetration into frozen soils depends on soil and oil composition and temperature conditions. It was observed, as expected, that oil accumulation in frozen soils decreases with increasing ice content in the pores. However, penetration of oil components is observed even in completely ice-saturated soils. Freezing of oil-saturated soils causes a redistribution of the oil components. In sandy soils, the oil concentrates in a thawed zone in front of the freezing front; in clay soils, the oil can accumulate in the frozen zone under certain temperature conditions. A summary of the influence of various factors affecting oil behaviour in frozen and freezing soils is presented based on the experimental data and published data from other authors.

Contents

Introduction

Research into the behaviour of oil pollutants in permafrost and areas of deep seasonal frost has increased during the last decade due to the industrial development of northern regions and oil and gas exploration and production in the Arctic. Of special concern to developers is the possible negative influence of hydrocarbon pollution of the natural environment and ecology (vegetation, wildlife, permafrost, surface and ground water, and atmosphere) (Arctic Monitoring and Assessment Programme 1997). Pollution of the ground surface can result in changes of the surface energy balance, and as a consequence lead to the thawing of permafrost and the onset of other cryogenic processes (Collins and others 1993). If oil and oil components penetrate into the ground, this may lead to changes of the physical, mechanical, and engineering properties of the soil. This may have negative effects on the stability and reliability of engineering structures.

Most of the research on hydrocarbon pollution of soils has been conducted outside permafrost areas, in locations with positive mean annual soil temperatures. The main focus of these studies has been the negative influence of hydrocarbon spills on biota, soil properties, and surface and ground water. Research concerned with hydrocarbon pollution in permafrost areas has previously focused on surface pollution; the study of penetration into permafrost has been limited to the active layer. Until recently, frozen soils were considered a practically impermeable barrier to both mineral and organic pollutants.

Recently, experimental results have confirmed that hydrocarbon pollutants can penetrate into frozen soils. The pore volume in frozen soils may not be ice-saturated, and micro-cracks existing in ice-saturated frozen soils may explain hydrocarbon transportation and spreading in frozen soils. The presence of a pore volume not filled by ice may cause the oil to move into frozen soil not only under action of gravitational forces, but also under action of surface forces of the mineral skeleton (Yershov and others 1997; Chuvilin and others 1999). Ice-saturated soils are not an absolute impermeable barrier for oil penetration. This has been confirmed by laboratory experiments (Chuvilin and others, in press) and field studies (Biggar and others 1998). It has been shown that even though the presence of ice lenses and pore ice is an important obstacle for transport and spreading of oil pollution in frozen soils, the migration and pollutant flow is not completely interrupted. The transportation of pollutants in ice-saturated soils is most probably caused by the presence of unfrozen water films on the surface of mineral particles and ice. The unfrozen water can be a medium for transportation of water-soluble components of oil (Biggar 1995; Chuvilin 1999).

The spreading of hydrocarbon pollution on the ground surface is dependent on the time of the year at which the spill occurs. In the winter, oil spreads mainly on the surface of snow cover and/or frozen soil; a winter spill may, therefore, cover a larger surface area than a summer

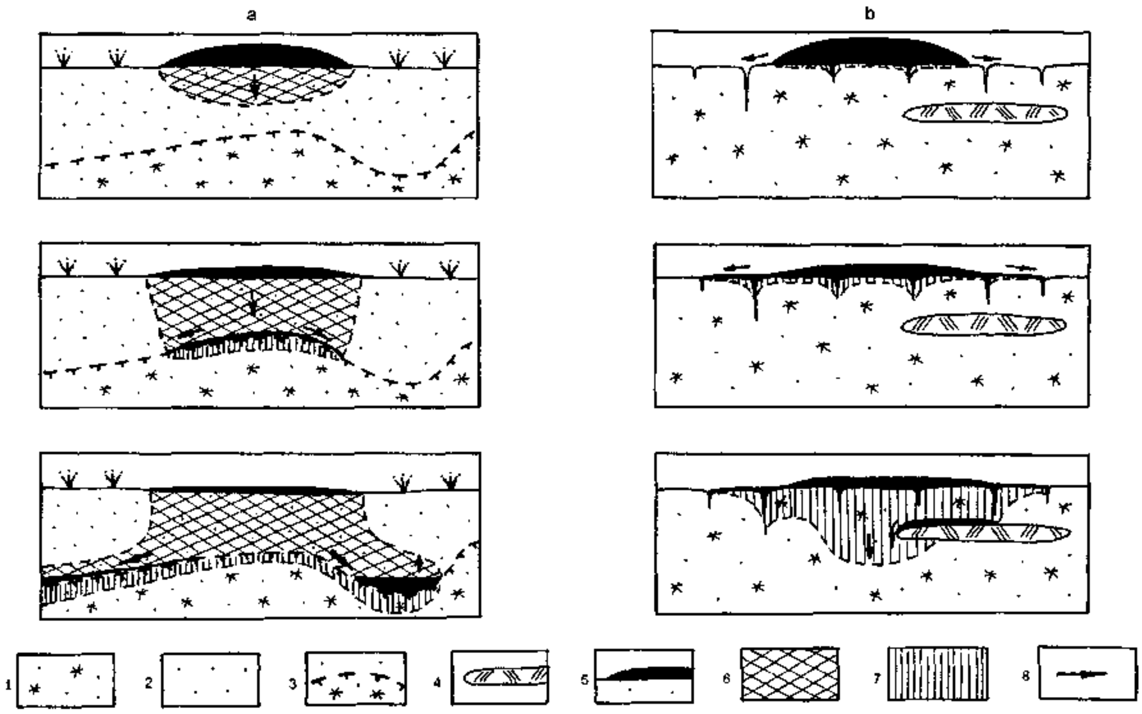

Fig. 1. The scheme of distribution of oil pollution within the permafrost area at a spill. a: on a surface of seasonal-thawed soils in summer time; b: on a surface of frozen soils in winter time. 1 = frozen soils; 2 = unfrozen soils; 3 = border of thawed and frozen soils; 4 = lens of underground ice; 5 = oil spill; 6 = pollution of thawed soils by oil; 7 = pollution of frozen soils by oil; 8 = direction of oil migration.

spill (Biggar 1995). In the summer, the lateral distribution of an oil spill is reduced due to detention of the oil by the vegetative cover. Infiltration of precipitation may increase penetration of oil into thawed soils (Solntseva 1998). The hydrocarbon pollution may spread laterally when it penetrates into the ground and reaches a surface of frozen ice-saturated soils (on the top of the permafrost layer). The hydrocarbon pollution may accumulate in zones with increased thawing depth and closed taliks have been observed.

Figure 1 illustrates the transportation and spreading of hydrocarbon pollution in areas of permafrost during summer and winter conditions, with different time intervals. At a summer spill (Fig. 1a), the oil on a surface of a thawed soil spreads and penetrates in a seasonal thawed layer, reaching the top of the frozen soil. The oil concentrates on a frozen surface and begins to spread, accumulating in local downturns and through talik zones. At that point, the oil can penetrate into the permafrost (Collins and others 1993). By contrast, during a winter spill, when the seasonal thawed layer is absent, the oil only spreads on the surface of the frozen soil (Fig. 1b). No soaking of lower layers occurs because the seasonal thawed layer is absent during the cold-weather months. Only separate components of the spills can penetrate.

The data available today regarding the interaction of oil with permafrost soils do not give a complete picture of the influence of various factors affecting the transportation and accumulation of hydrocarbon pollutants in frozen and freezing soils. Therefore, an experimental research programme was initiated in order to study the physical mechanisms that control the migration of oil in frozen soils.

Description of the laboratory investigation

Soils and oils used in the laboratory investigation

The laboratory investigation was carried out on three different soils. The grain-size distribution curves are presented in Figure 2 and the mineralogy of the soils is given in Table 1. Four different oils were used in the laboratory investigations, and some of their key parameters are presented in Table 2.

Methodology for laboratory investigations

Two suites of laboratory tests were carried out: an investigation of oil spreading on a frozen surface, and an investigation of oil transportation and accumulation in frozen and freezing soils.

In the first suite of tests, the characteristics and velocities of spreading of the four different oils on the ice-saturated soils and ice surface were studied. Thus the features of physical–chemical interaction between hydrocarbons and frozen soils (wettability of mineral surfaces by oil) were investigated.

In order to study oil spreading on different surfaces, the experiments were carried out at three different temperatures (−1, −7, and −20°C). A drop of cooled oil (approximately 1 ml in volume) was applied to a flat, horizontal surface of the samples (diameter 20 cm). The initial contact diameter of the drop was measured, and then the changes of this diameter as a function of time. The duration of each exper-

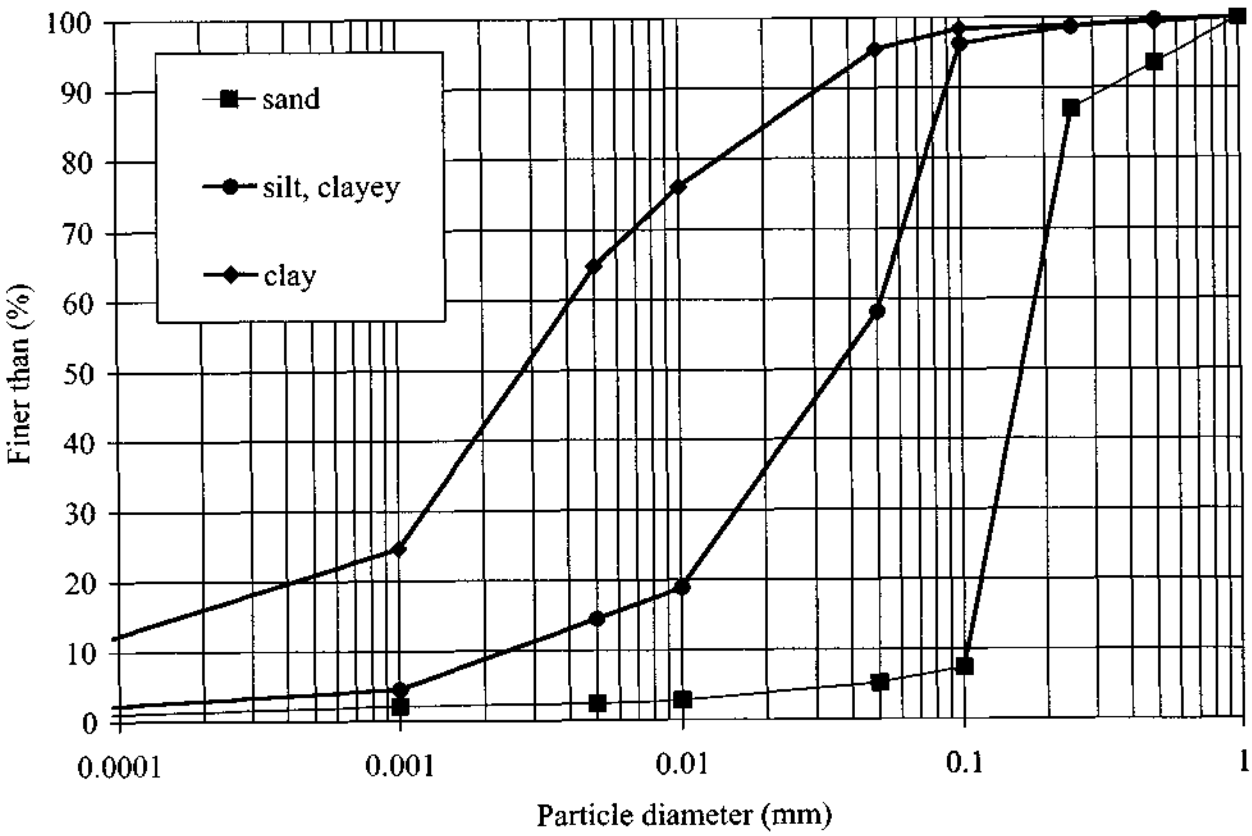

Fig. 2. Grain-size distribution of test soils.

Table 1. Mineral content of soils used in the laboratory investigation.

Soil type	Mineral type
Sand	quartz >90%
Silt, clayey	particles >0.01 mm: microcline + albite 45%, quartz 38%; particles <0.01 mm: illite 55%, kaolinite + clorite 27%, montmorillonite 18%
Clay	kaolinite 92%, quartz 6%, muscovit 2%

iment was approximately 30 days. The calculated parameter 'oil spreadability' was introduced for a description of the process of oil spreading. This parameter, S_f, was defined as the ratio between the present radius of the drop of oil to its initial radius:

$S_f = (R_R - R^0_d) / R^0_d$, where
R_R = radius of the spreading = $R_f + R^0_d$
R_f = radius of 'oil fringe'
R^0_d = radius of initial drop.

Note that the 'oil fringe' is defined as the external zone of oil drop, which differs from the basic volume of oil by colour, thickness, and consistency. Its development is connected with oil fraction processes (that is, the allocation of oil-separate mobility fractions).

The wetting of various mineral surfaces by oil was studied by measuring the boundary angles of wettability on the different surfaces, including the soils described in Figure 2 and Table 1, and on pure ice.

In the second suite of tests, the permeability of frozen and freezing soils to various types of oil was studied. The experiments with frozen soils were conducted on artificially prepared frozen soil samples. The degree of saturation was varied from 45% to approximately 100%. The experiments were carried out at two isothermal temperature conditions (–1.5 and –7°C). The frozen soil samples were placed in pallets on sponges saturated by oil and covered by a lid. The frozen soil sample was in contact with the oil on the bottom end only, in order to remove the influence of gravitational forces.

The experiments with freezing soils were carried out by one-dimensional freezing of artificially prepared soil samples from the top downwards. At first the soils were mixed with distilled water and oil. Then the silt and clay samples were compacted in steps up to 200 kPa prior to freezing. The duration of each test was approximately 24 hours. Temperature change in the soil sample during the test was measured with thermocouples.

Before the start of each test and at the end of the experiments, water content, oil content, and density were measured in different layers in each sample. The oil was extracted using chloroform.

Results

Investigation of oil spreading on a frozen surface

The experimental results show that oil spreading on mineral surfaces depends on a number of factors such as:

Table 2. Description of oils used in the laboratory investigation.

Oil	Description	Hardening temperature	Density at +20°C	Surface tension
1	paraffin and naphthene-paraffin	+5°C	$d_4^{20} = 0.8660$	–
2	paraffin and naphthene-paraffin	+2°C	$d_4^{20} = 0.8692$	10.32 mN m^{-1}
3	paraffin and naphthene-paraffin	–11°C	$d_4^{20} = 0.9200$	31.03 mN m^{-1}
4	naphthene and naphthene-aromatic	–23°C	$d_4^{20} = 0.8220$	–

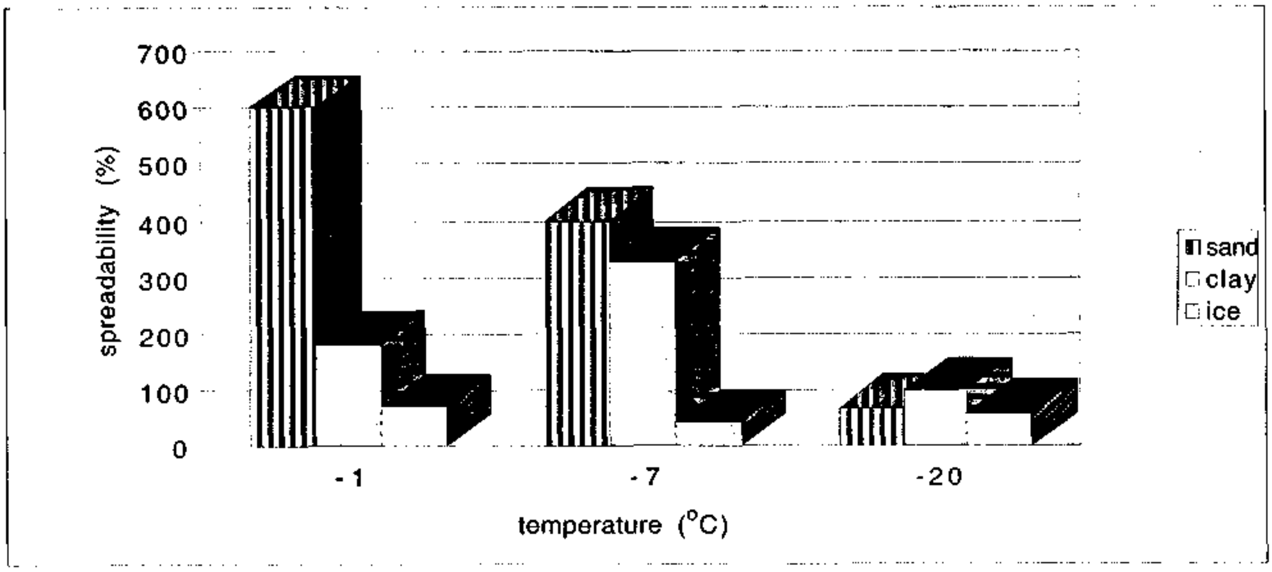

Fig. 3. Dependence of the spreadability of oil (2) on mineral surface and temperature.

1. type and character of the mineral surface (mineral structure, morphological features of elements composing the surface, and porosity),
2. temperature conditions, and
3. oil structure and properties.

The physical behaviour of the spreading of surface oil shows a distinct change at the transition from positive to negative (sub-zero) temperatures. Tests on thawed water-saturated soils (T = +20°C) show that the oil quickly spreads in a thin film on all types of mineral surfaces tested. At sub-zero temperatures the oil forms a drop on the frozen mineral surface, the size of which increases with time. In addition, change of colour and consistency of the area adjacent to the drop was observed. This is probably due to the movement of fractions in the oil with high mobility or water solubility.

The influence of the mineral surface on oil spreadability can be observed by comparing samples of sand, clay, and ice (Fig. 3). Minimum spreadability was observed for the ice surface. High oil spreadability on sand at negative temperature is connected to the mineral structure and also the surface roughness, which provides large wettability.

With decreasing temperature from –1 to –20°C, oil spreadability is reduced. The experiments show that the variation in spreadability on the different surfaces is also reduced. Thus, the influence of a mineral surface on oil-spreading processes is reduced at low or negative temperatures, and spreadability will be governed by oil structure and its properties.

The influence of oil structure on spreadability on a mineral surface is dependent on the physical properties and characteristics of the oil, the most important parameter being the hardening temperature. It was observed that a decrease of the temperature at which oil hardens resulted in increased spreadability (Fig. 4). Maximal spreadability on frozen sand (about 700%) corresponded to the oil with the lowest hardening temperature of –23°C (oil 4), and the minimum (200%) to the oil with the highest hardening temperature of +5°C (oil 1). However, the spreadability on a pure ice surface is not affected to the same degree by the temperature at which oil hardens.

Determination of boundary angles of wetting

The experiments performed in order to define oil boundary angles of wetting on various mineral surfaces showed that at a positive temperature, T = +20°C, the majority of oils that were studied (oil 2, 3, and 4) wetted the mineral surfaces completely (oil spreads in a film, the angle does not exceed 5°C). Oil 1 wets the mineral surfaces to a lesser degree. For example, it forms an angle of ~10° on the surface of quartz.

At a negative temperature, T = –7°C, wetting by oil is more dependent on oil type and the characteristics of the mineral surface. The oil with the lowest temperature of hardening (oil 4; –23°C) wets the surfaces of quartz and kaolinite completely (boundary angle <5°). The oil with a temperature of hardening of –11°C (oil 3) completely wets

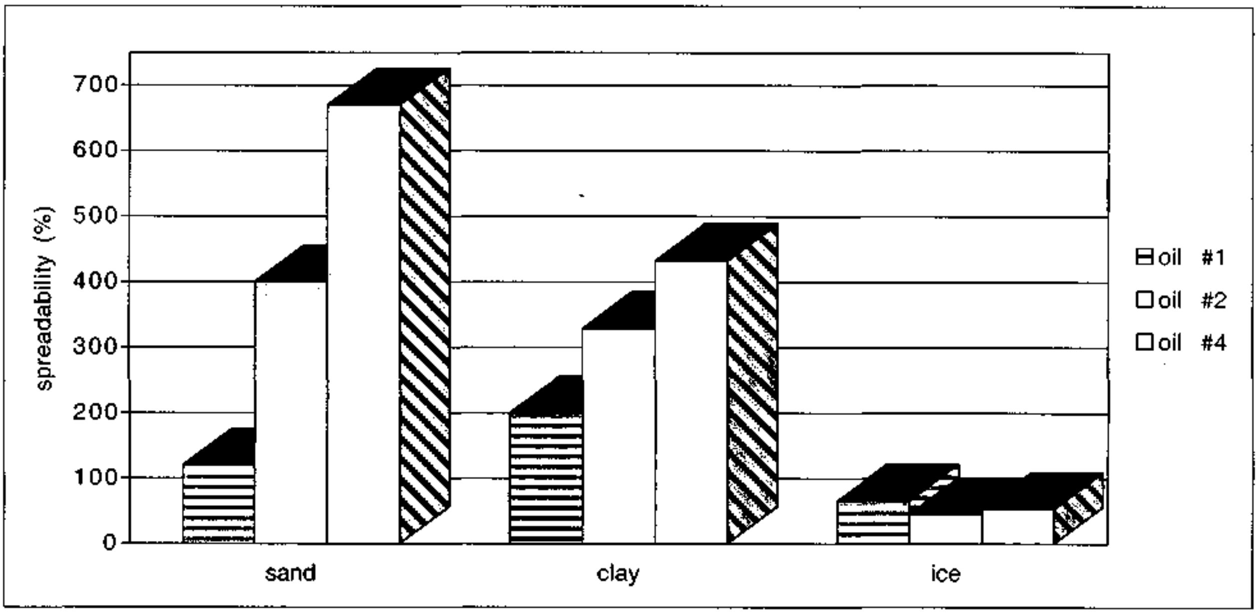

Fig. 4. Dependence of the spreadability of the oils on hardening temperature.

the surface of quartz but can form an angle up to 10° on kaolinite. On an ice surface both of these oils form angles up to 20–30°. The oils with high temperatures of hardening (oil 1, +5°C; oil 2, +2°C) form angles on all investigated mineral surfaces. Oil 1 had angles close to 20° on kaolinite, quartz, and ice. Oil 2 formed angles from 70 to 100° and had a very low wettability.

With the oils used in these experiments, the ice surface was characterised by lower wettability than the mineral surfaces.

Oil penetration into frozen soils

The experiments on interaction of soil samples with oil have shown that frozen soils can also be permeable to oil. However, accumulation of oil is, as expected, much less in frozen soils than in thawed soils. For example, in kaolinite clay samples with an initial humidity of w = 30%, the

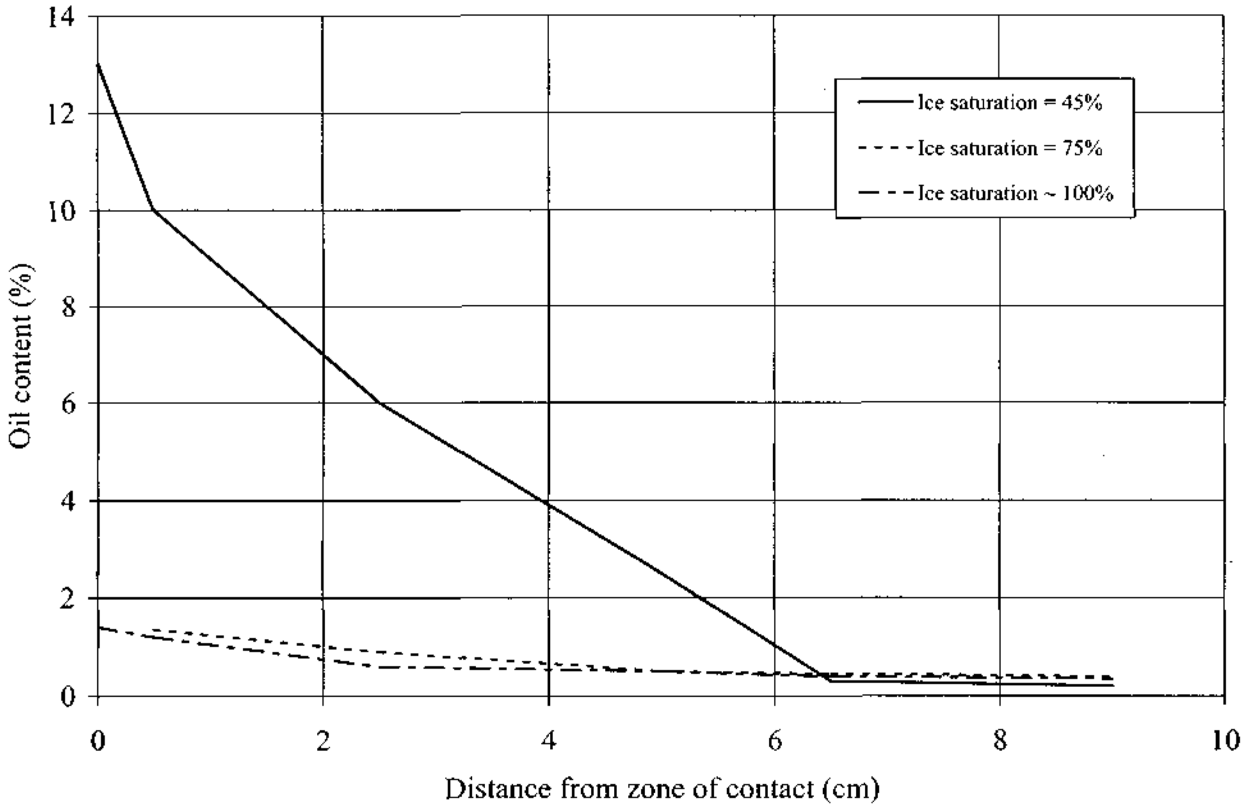

Fig. 5. Accumulation of the oil (3) by the height of frozen samples with different ice saturation (sand, T = –7°C, t = 7 days).

Table 3. Values of density-migration flows for various soils in contact with oil.

Soil	Water content (%)	Ice saturation (%)	Oil	Temperature (°C)	Time (days)	Jn (g/cm² • s)
sand	11	45	oil 3	−7	7	87.8×10^{-8}
sand	17	75	oil 3	−7	7	17.7×10^{-8}
sand	20	~100	oil 3	−7	7	15.6×10^{-8}
sand	20	~100	oil 4	−1.5	14	8.5×10^{-8}
sand	21	~100	oil 4	−7	14	2.3×10^{-8}
silt	21	~100	oil 4	−7	14	6.5×10^{-8}
clay	35	~100	oil 4	−7	14	10.4×10^{-8}
clay	34	~100	oil 4	−7	60	4.2×10^{-8}
sand	21	~100	oil 2	−7	60	1.4×10^{-8}

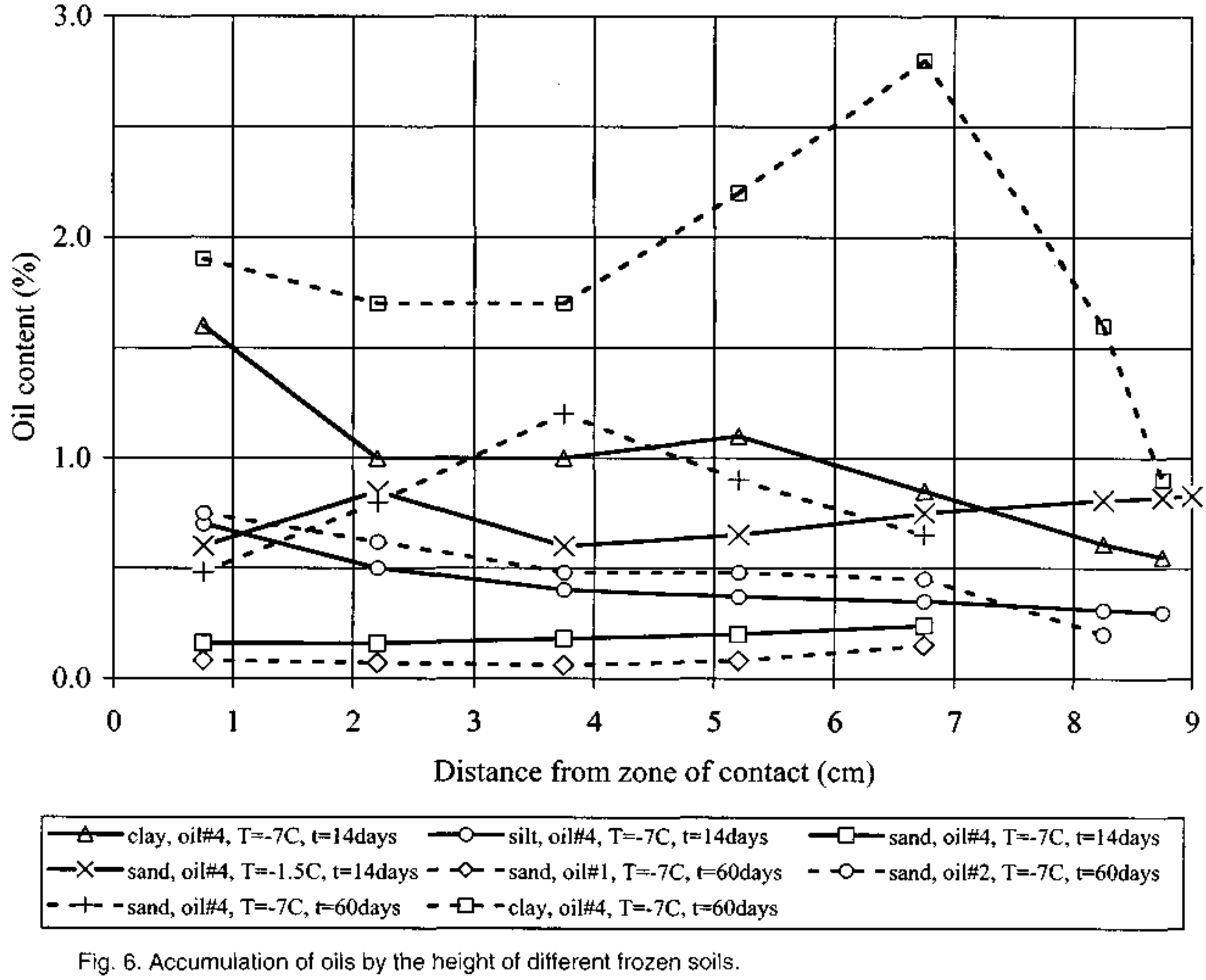

Fig. 6. Accumulation of oils by the height of different frozen soils.

effective diffusion coefficient was 2.5×10^{-3} cm² s⁻¹ at temperature T = +20°C, decreasing to 4.5×10^{-5} $cm^2 s^{-1}$ at temperature T = −6°C (Chuvilin and others 1999). The reduction in the effective diffusion coefficient is caused by an increase of the oil viscosity at decreasing temperature and increased ice content and hence decrease of the unfrozen water content in the soil samples.

The oil transportation and spreading in frozen soils is mainly dependent on the ice saturation. Figure 5 shows the oil content (oil 3) versus the distance from contact for a sand at temperature T = −7°C for three different values of ice saturation. From the figure, it can be observed that the oil content decreases with increasing ice saturation. However, an increase of ice saturation from 75% to ~100% does not result in a significant decrease of oil penetration into the sample. This can be explained by a 'threshold ice saturation,' where the ice in the pores closes the migration routes. Table 3 presents the values of density-migration flow from the experiments.

Figure 6 shows oil content versus distance from the zone of contact for ice-saturated soils. The following trends can be observed from the figure:

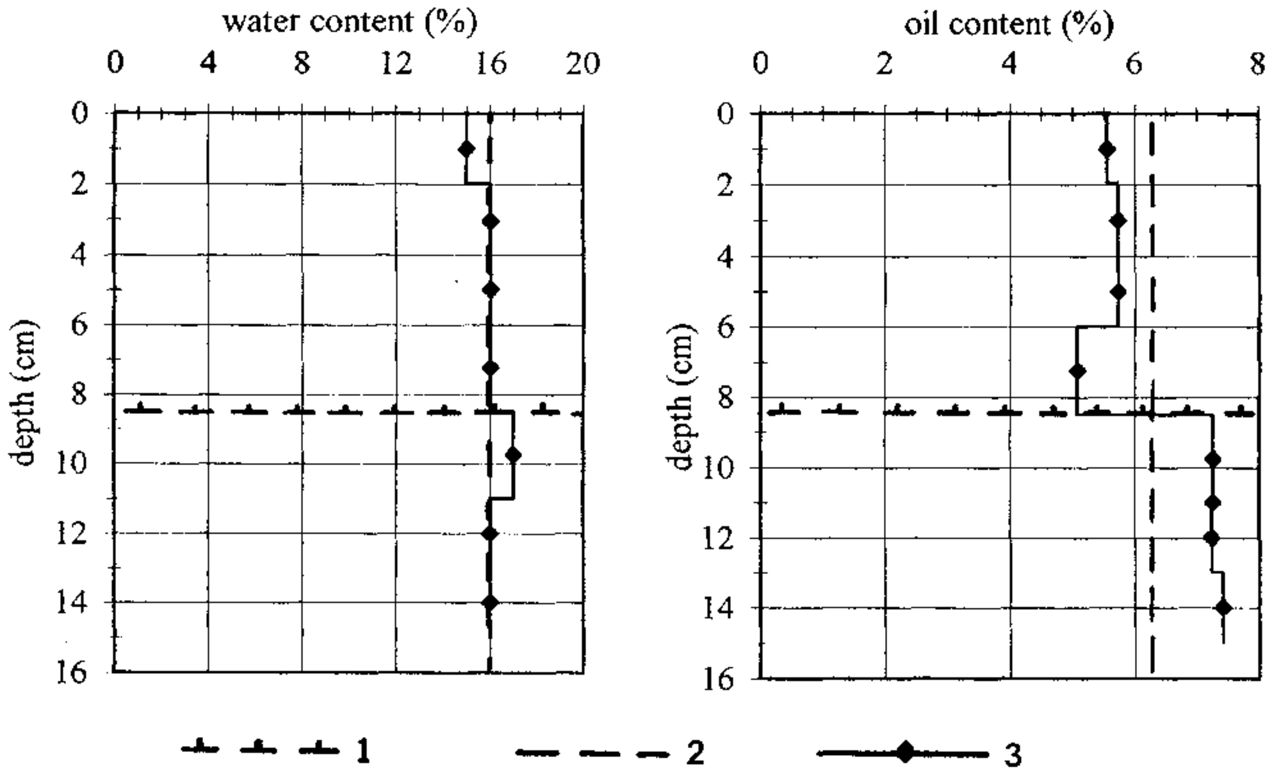

Fig. 7. Pattern of the water and oil (2) content with height of sand freezing at −7°C. 1 = freezing front at the end of the experiment (touches directly on the frozen zone); 2 = initial patterns of the water and oil content; 3 = final patterns of the water and oil content.

1. oil concentration in the soil increases with increasing fines content in the soil,
2. oil concentration in the soil increases with increasing temperature,
3. oil concentration in the soil increases with time,
4. oil concentration in the soil increases as the temperature for hardening of oil decreases.

The higher permeability of ice-saturated clay in comparison with sand is probably associated with higher unfrozen water contents, which allow volatile and soluble oil components to penetrate into soil. As shown above, temperature conditions, oil structure, and time elapsed from oil interaction with the frozen soils will influence the accumulation of oil in frozen soils.

The permeability of frozen soils by oil decreases with decreasing temperature. This is probably associated with decreasing unfrozen water content. The decrease in temperature changes the oil properties and causes an increase in oil viscosity.

Oil accumulation in frozen soil at temperatures below the hardening temperature of the oil can be explained by an assumption that separate mobile components (in particular, aromatic cyclic hydrocarbon), and heavy high-molecular compounds penetrate into the soil. The oil pollution distribution can vary along the depth of the sample. This means that the horizon of the maximal accumulation can be displaced from the contact zone between the oil and the sample. This indicates that transportation and spreading of hydrocarbon pollution in frozen soils involves complex physical processes and that the oil does not penetrate in one well-defined front, but rather as separate fractions with different speeds of penetration.

Freezing of oil-saturated samples

Soil samples polluted with oil were subjected to one-dimensional freezing. It was observed that oil was rejected and expelled at the freezing front, and that in this manner oil can be transported into the thawed soil by an advancing freezing front.

Three different soils were used in the experiments: sand, silt, and clay. The results are presented in Figures 7 and 8.

The sand was saturated with water (water content 16%) and oil (oil content 6.2%). It was observed that oil was expelled from the freezing sand and pushed into the lower unfrozen layer. It was further observed that the expulsion efficiency increased with decreasing temperature gradient. The oil distribution in the sample was not uniform after freezing. The oil contents have become much lower in the frozen zone and there was oil accumulation in the thawed zone. The lowest oil concentration was found in the bottom part of the frozen zone, which is characterised by the lowest freezing speeds.

Freezing of silt is associated with moisture migration from the thawed to the frozen zone. The migration of water can carry oil contained in pore space at a high density of migration flow, in the same manner as salt ions during freezing of saline clay soils (Chuvilin 1999). However, oil migration through moisture flow occurs less intensively

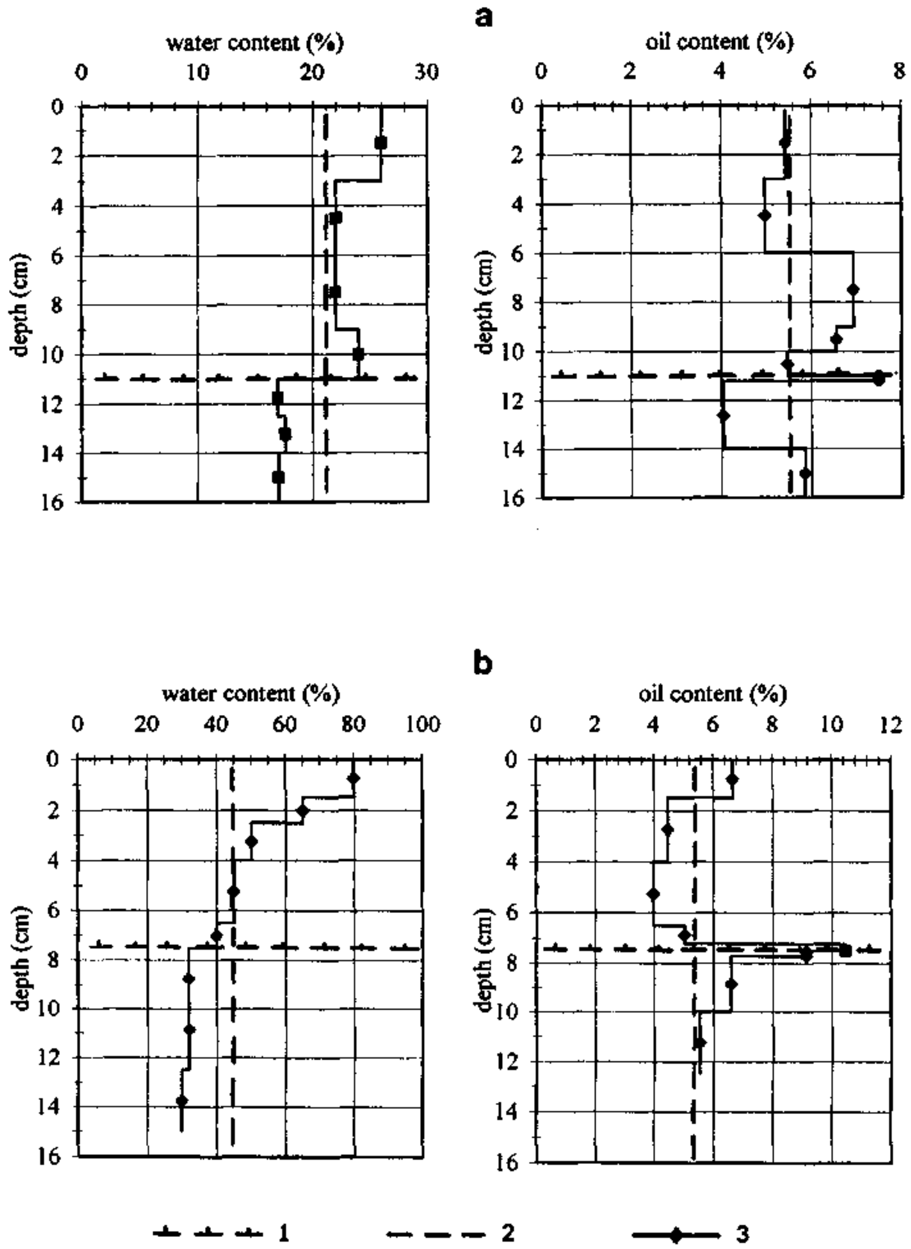

Fig. 8. Pattern of the water content and oil (2) content, with height of soil freezing at −7°C. a = silt; b = clay. 1 = freezing front at the end of the experiment (touches directly on the frozen zone); 2 = initial patterns of the water and oil content; 3 = final patterns of the water and oil content.

than salt, because of the hydrophobic properties of oil. The silt was saturated with water (water content of 21.5%) and oil (oil content 10%). It was observed that the oil content in the frozen zone increased during freezing. For the clay sample it was observed that the oil content increased in the beginning of the freezing, when there was an intensive moisture accumulation in the frozen zone. Oil expulsion occurred in the bottom part of the freezing clay with a reduction of migration flow and a reduction of freezing speed, which resulted in an increased oil content in a thawed zone on the border with the frozen zone.

It is expected that the oil redistribution will increase with decreasing oil hardening. However, detailed studies of oil-redistribution processes in freezing soils, particularly fine-grained soils, require additional experiments.

Conclusions

Based on the experimental results presented in this paper and the data from the literature review, the various factors affecting oil interacting with frozen and freezing soils are summarised in Table 4. The table presents the three basic processes occurring in regions of frozen soils:

1. oil spreading on the surface,
2. oil penetrating into the frozen soils, and
3. oil redistribution during freezing.

The first group of factors includes the soil and oil characteristics; the second group includes the surrounding (temperature) conditions determining phase structure and phase transitions of the soil moisture. In which direction each factor affects the process is indicated with arrows. Downward direction of the arrow indicated that the process is decreasing, and upward direction indicates an increase. The absence of an arrow indicates that the given factor does not influence the process or was not investigated. The table can be used qualitatively to estimate the role of basic factors affecting spreadability and transportation of oil in regions of frozen ground.

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Table 4. The influence of various factors on oil interacting with frozen and freezing soils.

	Factor	Direction of change	The processes: In frozen soils: *Spreading of oil on a surface*	The processes: In frozen soils: Penetration of oil into soils	The processes: In freezing soils: Redistribution of oil at freezing
The characteristics of soils and oils	Character of a surface of the frozen substratum	From sand to clay to ice	⇓	–	–
The characteristics of soils and oils	Dispersion	Increase of the dispersion	⇓	predominantly ⇑	Expulsion into thawed zone ⇓ Accumulation in the frozen zone ⇑
The characteristics of soils and oils	Degree of ice saturation	Increased ice saturation	⇑	⇓	–
The characteristics of soils and oils	Oil content	Increased initial content of oil on surface	–	–	⇑
The characteristics of soils and oils	Type of oil (based on hardening temperature)	Decrease of hardening temperature of oil	On soil surface ⇑ On ice surface – influence is poor	⇑	⇑
External factors	Temperature of the surroundings (isothermal conditions)	Decrease of surrounding temperature	On soil surface ⇓ On ice surface – influence is poor	⇓	–
External factors	Freezing velocity (one-dimensional freezing)	Increase of freezing velocity	–	–	⇓

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