


Rosa Margesin
Editor

SOIL BIOLOGY

Permafrost Soils

 Springer

Chapter 18

Migration of Petroleum in Permafrost-Affected Regions

David L. Barnes(✉) and Evgeny Chuvilin

18.1 Introduction

An extensive amount of effort has been undertaken by many over the last three or so decades to better understand the movement of crude oil and petroleum products through terrestrial environments. This effort is based on a desire to better characterize and remediate environments that have been impacted by releases of these substances. The presence of ice in Arctic and Antarctic soils, the influence seasonal freeze and thaw cycling has on fluid movement, and the typically shallow active layers found in these environments all impact the movement of fluids in these soils in a manner not found in temperate soils (soils that do not experience deep freezing). How the unique Arctic and Antarctic conditions affect the movement of petroleum-related substances in these environments will be discussed in this chapter.

Understanding the mobility of contaminants in these environments becomes relevant when one considers the high cost of conducting site investigations and cleanup activities at locations in the Arctic and Antarctic that are often remote. In addition, uncertainty as to how cleanup activities may possibly enhance mobility of contaminants and degradation of the ecosystem by disturbing the fragile thermal balance is of concern in any cleanup activity in the Arctic and Antarctic. At the extreme, Snape et al. (2001) discussed the directives of the Antarctic Madrid Protocol (International Council of Scientific Unions 1993) to clean up past and present waste disposal sites. At many of these contaminated sites contaminated material that cannot be treated onsite will have to be removed from the continent, an expensive process. Onsite treatment will require the shipment of treatment equipment and materials to the research stations, again an expensive process. Thus, it becomes evident why understanding the mobility of contaminants becomes important, as even a small reduction in the material to be treated or shipped will result in economic benefit.

David L. Barnes
University of Alaska Fairbanks, Department of Civil and Environmental Engineering,
Water and Environmental Research Center
e-mail: ffdlb@uaf.edu

18.2 Background

Human activities in the Arctic and Antarctic have resulted in releases of a suite of compounds that are harmful to human and environmental health including crude oil and petroleum products. In this chapter the terms *crude oil* and *petroleum products* refer to the actual liquids, and the term *petroleum hydrocarbons* refers to compounds such as benzene that make up the liquids. Most releases that occur in the Arctic and Antarctic are insignificant in volume; however, several larger terrestrial releases have taken place. Arguably, the largest release to have occurred in the Arctic took place north of the city of Usinsk, Russia (65°N), in the Kolva River Basin (Vilchek and Tishkov 1997; AMAP 1998). By one estimate 103,000–126,000 tonnes of crude oil (other experts estimate the release to be as high as 318,000 tonnes) was released over a 2–3 month period from multiple leaks in a pipeline system that continued to pump oil even though the pipeline was leaking (Vilchek and Tishkov 1997).

Recent relatively large releases of crude oil have occurred on the Trans Alaska pipeline. In 2001, a hole was shot in the pipeline near the village of Livengood, Alaska (65°N), resulting in the release of approximately 265,000 l (Spiess 2001). Corrosion of a crude-oil transit line at Prudhoe Bay, Alaska, caused between 761,000 l and 1,010,000 l of crude oil to be released to the tundra (JIC 2006). Relatively smaller releases associated with fuel storage and transportation for industrial activities, predominately mining, and for communities in the Arctic occur with more frequency than the larger more notable releases. Such releases are a result of vehicle accidents, such as a fuel tanker truck roll-over at a large zinc and lead mine in northwest Alaska (68°N) resulting in the release of approximately 10,000 l to tundra (ADEC 2004a), or mishaps related to fuel storage, such as an approximate 9,500 l release that occurred in the village of Point Hope, Alaska (68°N), due to overfilling of a storage tank (ADEC 2004b). Rike et al. (2003) described the presence of petroleum hydrocarbons in soil at a site near the village of Longyearbyen, on Spitzbergen Island in the Svalbard archipelago (78°N), most likely resulting from small releases of petroleum over time at a fire extinction training site.

Relatively smaller releases of petroleum products have occurred in the Antarctic as well. A majority of these releases are due to poor waste management practices at research stations. In the hope that burial in frozen ground would contain waste, most waste from research stations were disposed of in dumps with little to no engineered containment systems (Snape et al. 2002). In addition, minimal attention was given to petroleum spills, owing to the belief that the frozen environment would contain the compounds (Snape et al. 2002). Investigations illustrated that containment of contaminants in this manner is not feasible. Contaminants are mobile in this environment during thawing and thawed periods much in the same manner as in more temperate environments. In addition, cryoturbation and erosion uncovers buried contaminants, exposing them to transport processes both in ground water (suprapermafrost) and surface water (Snape et al. 2001).

To understand movement of petroleum and petroleum hydrocarbons through freezing and frozen soils in the Arctic and Antarctic, an understanding of the

fundamental principles of immiscible fluid (in this case petroleum) movement through unfrozen soil is required. Several authors have presented thorough descriptions of the movement of immiscible fluid, commonly known as non-aqueous phase liquids (NAPL), through unsaturated soils (Mercer and Cohen 1990; Wilson et al. 1990; Poulsen and Kueper 1992). Petroleum is considered a light non-aqueous phase liquid (LNAPL), as the specific gravity of the fluid is less than unity. The remainder of the discussion will focus on petroleum.

Released at or near the ground-surface, petroleum will move downward through unsaturated soil toward the water table. Due to the immiscibility, the fluid migrates as a distinct liquid, separate from the air and water present in the unsaturated soil. Water and petroleum are held in the pore space of partially saturated soils by capillary forces. As petroleum migrates downward, air and possibly some water are displaced from the pore space. Once in soil pore space, individual petroleum compounds will dissolve into soil water according to the specific solubility of each compound and its mole fraction. Solubility of these compounds is low, since most petroleum hydrocarbons are non-polar. Sorption of petroleum hydrocarbons onto natural organic matter in the soil results from the non-polar nature of these compounds. The high volatility of relatively low molecular weight petroleum hydrocarbons dissolved in soil water results in partitioning of a fraction of these compounds into the gas phase. The mixture of gaseous petroleum hydrocarbons and air becomes soil gas in the pore space.

Infiltrating petroleum follows a path through unsaturated soil that is dictated by the properties of the soil encountered; primarily, permeability and pore structure. Results from field studies performed by Poulsen and Kueper (1992) illustrated how small variations in permeability result in extreme heterogeneous distribution of NAPL and some lateral migration, which is also a result of capillary forces.

Capillary forces immobilize a fraction of petroleum in the pore space as the main body of the liquid moves downward through porous medium. Results from a visualization study conducted by Wilson et al. (1990) showed that immobilized NAPL was mostly contained in pore throats and in thin films between soil water and soil gas. Soil water was also contained in pore throats that were bypassed by infiltrating NAPL, and soil gas filled the larger pore bodies.

Infiltrating petroleum that reaches the capillary fringe, sometimes referred to as the nearly saturated zone, will spread laterally as a result of the relatively high water saturations in this zone. For spill volumes that generate sufficient head to displace the water in the capillary fringe water, petroleum that migrates further downward to the water table may displace water from saturated pores and cause depression of the water table. As the water table rises and falls seasonally some petroleum is immobilized or entrapped in the capillary fringe and possibly below the water table during high water level conditions. This immobilized petroleum consists of small pockets (or ganglia) of liquid disconnected from the main body of organic liquid (Wilson et al. 1990). A dissolved phase plume results in the saturated zone below the water table, from petroleum contained above and below the water surface.

18.3 Migration of Petroleum in the Active Layer

The migration of petroleum through soil that comprises the active layer (the zone above permafrost zone that experiences season freezing and thawing) is a function of the season in which the petroleum is released. Migration of released petroleum during periods when the active layer is unfrozen or thawing will be influenced by high soil-water contents in poorly drained soils, and by the shallow nature of the active layer in many permafrost regions. During periods when the active layer is frozen, migration of released petroleum will be greatly influenced by the presence of ice in the soil. Freezing and thawing cycles will impact the distribution of petroleum in the subsurface, independent of the season the petroleum was released.

18.3.1 *Petroleum Releases to Unfrozen Active Layers*

In permafrost-affected regions the thickness of the active layer will be minimal — centimeters to a few meters, depending upon local conditions. The active layer begins to thaw during the spring snowmelt and continues to thicken until reaching maximum thickness in late August or September (Hinzman et al. 2005). As the active layer thaws a layer of water-saturated soil develops, which may be as thick as the entire thawed thickness. Thus, the downward flow of petroleum will be impeded due to low relative permeability to petroleum as a consequence of high soil-water saturation. With downward flow impeded, an increased flow takes place through the near surface layer of partially decayed vegetation that is typically present in many arctic ecosystems or bare ground where vegetation is not predominant. Results from field studies conducted by Mackay et al. (1974a, b, 1975) as well as Johnson et al. (1980), in which petroleum was released to unfrozen soil underlain by permafrost, illustrate how high water contents in poorly drained soils impede downward migration of released petroleum. This flow pattern leads to relatively large aerial distributions of petroleum, tempered by entrapment of the petroleum onto organic matter present in the uppermost layer of soil. However, even under these conditions petroleum does move downward through underlying mineral soil. In areas of large accumulations of petroleum, soil water will be displaced and petroleum will progress into lower mineral soils. Furthermore, over time, the petroleum may migrate deeper into the soil horizon as the active layer freezes and thaws.

In contrast, a study conducted by Mackay et al. (1975) where petroleum was released to unfrozen unsaturated (relatively low soil water contents) soils in a tundra environment resulted in infiltration of petroleum to the top of the frost line or to the water table where present. The petroleum then flowed downgradient (down slope) through a relatively thin horizontal layer of very permeable soils directly above the frost line. Using fundamental principles, the theoretical distribution of petroleum in active layer soils can be investigated.

The thin nature of the active layer and the saturated soil contained within will influence the distribution of petroleum throughout the active layer, and may allow for petroleum to be distributed as a free-phase liquid throughout the entire saturated zone. Recognizing the complex nature of characterizing the water-saturated zone contained in the active layer, due in part to the constant change taking place as thawing and refreezing occurs, the fundamental characteristics of how petroleum may distribute following a release can still be examined. Farr et al. (1990) described the distribution of free-phase LNAPL, such as petroleum, in porous media under hydrostatics considering a deep ground-water aquifer, and developed the mathematical relationships for LNAPL saturation as a function of depth from ground surface. These relationships can be re-derived to take into account the thin saturated zone typically found in a thawed or thawing active layer. As in Farr et al. (1990), total liquid saturation (water and petroleum; S_T) as a function of capillary pressure between air and petroleum (P_c^{ao}) is as follows:

$$S_T = S_w + S_o = (1 - S_r) \left(\frac{P_c^{ao}}{P_d^{ao}} \right)^{-\lambda} + S_r, \quad (1)$$

where S_w is water saturation, S_o is the petroleum saturation, S_r is residual saturation (assumed to be the same for both liquids), λ is the pore size distribution coefficient, and P_d^{ao} is the displacement pressure between air and petroleum. Similarly water saturation (S_w) as a function of capillary pressure between petroleum and water (P_c^{ow}) can be described as follows:

$$S_w = (1 - S_r) \left(\frac{P_c^{ow}}{P_d^{ow}} \right)^{-\lambda} + S_r, \quad (2)$$

where P_d^{ow} is displacement pressure between petroleum and water. Capillary pressures as a function of elevation from the frozen soil layer (z) between each fluid are as follows:

$$P_c^{ao} = \rho_o g (z - T_o), \quad (3)$$

$$P_c^{ow} = \rho_w g (z - b) + \rho_o g (T_o - z). \quad (4)$$

In (3) and (4) ρ_o is density of the released petroleum, ρ_w is water density, g is the gravitational constant, b is the thickness of the saturated zone prior to the petroleum release, and T_o is the thickness of petroleum that would be found in a monitoring screened through the entire saturated thickness. For these calculations, an assumption is made that the thickness of the water-saturated zone stays constant.

To investigate the influence the water-saturated thickness has on petroleum saturation, consider the fluid properties and soil properties for a sandy loam shown in

Table 18.1. Assume that the thickness of petroleum that would be measured in a monitoring well installed in the impacted area is 0.7 m for this example. Again acknowledging our assumptions of hydrostatic conditions and no hysteresis, (1)–(4) can be used to estimate petroleum saturation as a function of depth for different values of saturated zone thickness prior to the petroleum release. Results from these calculations are shown in Fig. 18.1.

Table 18.1 Soil and fluid properties for the example provided in the text (soil properties are from Rawls et al. 1982)

Soil property	Value
• Porosity (Φ)	0.437
• Residual saturation (S_r)	0.08
• Pore size distribution (λ)	0.553
• Air-petroleum displacement pressure (P_d^{ao})	$758 \text{ kg m}^{-1} \text{ s}^2$
• Petroleum-water displacement pressure (P_d^{ow})	$330 \text{ kg m}^{-1} \text{ s}^2$
Fluid property	
• Water density (ρ_w)	$1,000 \text{ kg m}^{-3}$
• Petroleum density (ρ_o)	740 kg m^{-3}

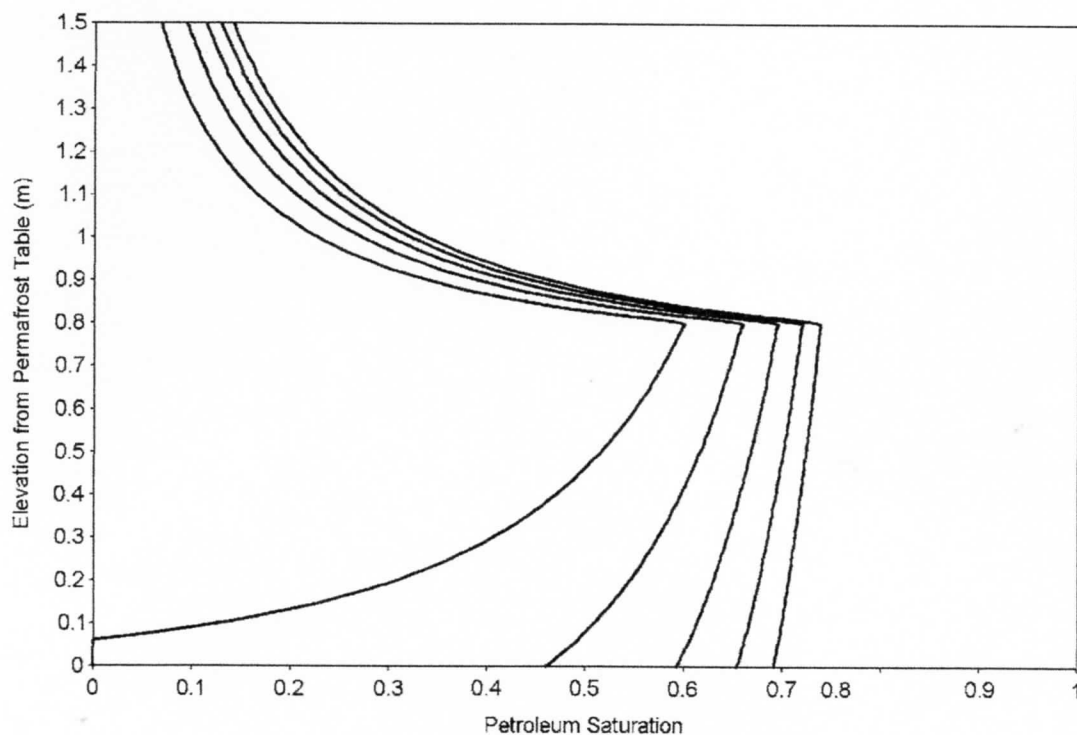


Fig. 18.1 Petroleum saturation with elevation from the top of permafrost. The petroleum saturation curve on the far left corresponds to a thick water-saturated zone where the top of the frozen layer does not interfere with the migration of the petroleum. Curves to the right of the bounding curve on the far left correspond to water saturated zone thickness prior to release of the petroleum to the active layer of 0.4, 0.3, 0.2, and 0.1 m respectively

The first notable result in Fig. 18.1 is the increase in the maximum value for petroleum saturation as the thickness of the saturated zone prior to the release of petroleum decreases. As shown in Fig. 18.1, the petroleum saturations between the top of the frozen soil layer and the elevation at which the maximum value of saturation is reached also increase, as the thickness of the saturated zone prior to release decreases. In addition, at the relatively thinner water-saturated thickness, the saturation of petroleum near the surface of the soil is greater in comparison to saturation values calculated for relatively deeper water saturation zone thicknesses.

While fluctuating water surface elevations, freeze and thaw cycling, and soil heterogeneity will most likely greatly affect the distribution of petroleum in the soil, these results indicate that petroleum release in active layers with shallow saturated zone thicknesses results in comparably greater initial mobility and, thus, a potentially wider lateral distribution of petroleum. This conclusion can be drawn due to the direct correlation between a fluid saturation and the relative permeability of porous media to that fluid. Petroleum as free product will also be distributed throughout the saturated soil thickness, leading to a widespread dissolved phase plume emanating from the source and subsequently little dilution of the dissolved phase plume. In addition, the volume of petroleum contained in a subsurface with a shallow saturated zone will most likely be greater than what would be predicted from models developed by Farr et al. (1990), Lenhard and Parker (1990), and Charbeneau et al. (1999).

18.3.2 Petroleum Releases to Frozen Active Layers

Migration of petroleum resulting from releases to frozen soils is significantly impacted by ice contained in the soil. At the minimum, ice present as pore ice will act as a solid, changing the pore geometry and thus the capillarity and permeability of the soil. In the extreme, the ground surface will be nearly impermeable, and downward migration will be minimal for the most part. Under these conditions surface flow will dominate, resulting in rapid and extensive spread of contamination upon release, though the higher viscosity at cold temperatures will inhibit lateral movement. In contrast to a release of petroleum to an unfrozen active layer, the increased exposure of the petroleum to the surface elements leads to greater losses of petroleum hydrocarbons by physical weathering (evaporation and photochemical oxidation).

Mackay et al. (1975) and Johnson et al. (1980) both conducted releases of crude oil to frozen ground in mature black spruce forests containing permafrost. Soils at both study sites were predominantly fine grain (silt). Results from sampling events shortly after each release in both studies indicated that overall there was minimal infiltration of the crude oil past the surface moss layer. Mackay et al. (1975) did document that infiltration of the crude oil did occur at spring thaw.

A laboratory study conducted by Barnes and Wolfe (2008) illustrates how pore ice in coarse soil impacts the movement of petroleum as the fluid infiltrates frozen

soil. Coarse soils are used extensively in the Arctic for foundations supporting infrastructure necessary for oil production as well as other activities, and are naturally present in Arctic and Antarctic terrain. In this study, petroleum was released to partially water-saturated sand that was frozen to -5°C . Two-dimensional petroleum flow through the frozen sand was approximated by packing moist sand between two vertical sheets of clear Plexiglas secured to a rigid frame and then freezing the entire unit. Once frozen, a volume of colored refined petroleum (JP 2) at a temperature of -5°C was introduced into the column and the progression of the petroleum was documented with time-lapse photography. Results from this study indicate that ice content far less than saturation can greatly affect the movement of petroleum, due to dead-end-pores created by ice forming in relatively smaller pore spaces, thus blocking flow paths. In addition, the formation of preferential flow paths results in deeper penetration of petroleum and unpredictable migration patterns. At the extreme, petroleum infiltration may be limited to the near surface soils due to high ice contents, as others have shown in field tests (Mackay et al. 1975; Johnson et al. 1980; Chuvilin 2001a).

Investigation of petroleum migration in frozen coarse soils in soil flumes can be taken one step further by investigating the infiltration of petroleum into a frozen heterogeneous coarse grain soil (Barnes and Adhikari, unpublished data). For this investigation, a layered soil was created in a soil flume with a layer of fine grain sand (1.3 cm thick) interbedded between coarse grain sand layers. The soil was then thoroughly wetted by introducing water to the top of the flume at timed intervals and allowing the water to drain through the sand layers. The flume was covered (to reduce evaporation) and allowed to drain for a sufficiently long enough time for gravity drainage to end. At this point, water in the pore space is held in the pore space by capillary forces at some residual level. The flume was then insulated on the sides and the bottom and placed in a cold room at -5°C to induce top-down freezing. Once frozen, colored JP2 chilled to -5°C was introduced to the top of the soil layer, and migration of the petroleum through the soil was tracked using time-lapse photography. The test was repeated in layered soil that was prepared in exactly the same manner but left unfrozen. Results from these tests are shown in Fig. 18.2.

The impact the fine grain sand layer has on the movement of petroleum through the frozen soil in comparison to the unfrozen soil is clearly evident in the images shown in Fig. 18.2. The fine sand layer in the frozen soil acts as a barrier to further downward petroleum migration. This result is due to the development of a capillary break between the fine grain sand and the underlying coarse grain sand. As water infiltrates and drains through a layered unsaturated soil, capillary breaks develop at the interface between relatively fine grain soil and underlying coarser grain soil, due to the comparably low relative permeability to water in the coarse grain soil in relation to the overlying fine grain soil. Low relative permeability in these cases is brought about by the comparably lower soil water content in this soil, owing to the larger pore dimensions and thus lower capillary forces in this layer. Once a capillary break develops, the low relative permeability in the underlying coarse soil restricts drainage of water out of the overlying fine grain soil, resulting in high water saturation in the fine grain soil. If a sufficient water saturation exists in the

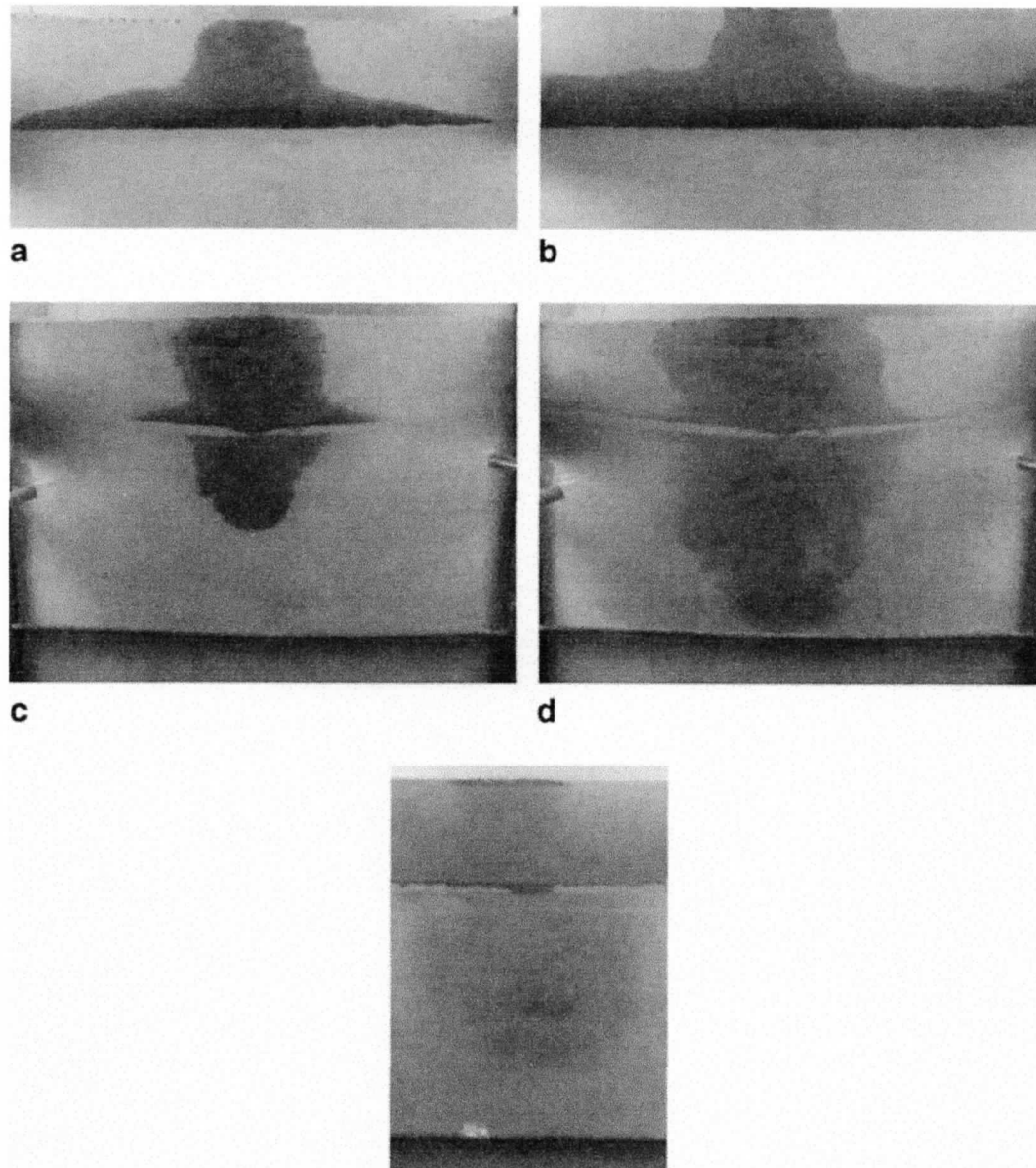


Fig. 18.2 Migration of petroleum through frozen (images **a** and **b**) and unfrozen (images **c** and **d**) layered soil. Images **a** and **c** were taken 1 h after releasing the petroleum to the soil. Images **b** and **d** were taken 1 day after releasing the petroleum to the soil. Image **e** was taken after the frozen soil from images **a** and **b** was thawed from the top down

fine grain soil prior to freezing (at least 91.7%), the pore space will be filled with ice once frozen, creating a barrier to infiltration of any liquids such as inadvertently spilled petroleum. During top-down thawing of the frozen sand in this investigation, the petroleum drained and redistributed as the thawing front advanced downward (Fig. 18.2). Some interference with the sides of the flumes was encountered, so the image has been trimmed to show the central portion of the redistributed plume. One will also note the extensive distribution of petroleum throughout the entire thickness of coarse sand above the thin layer of fine sand, which most likely developed through capillary movement of the petroleum.

In layered soil, the development of capillary breaks in frozen soil (ice-rich capillary breaks) as shown in Fig. 18.2 results in substantial increase in lateral petroleum movement upon the release of petroleum, in comparison to unfrozen soils and in comparison to frozen non-layered (homogeneous) soil (Barnes and Wolfe 2008). Others have noted the preferential lateral movement of petroleum in frozen soil Mackay et al. (1975). Damian Gore personal communications) the development of ice-rich capillary breaks may in part be the reason for these occurrences.

Ice has a substantial impact on petroleum distribution in frozen soils. Present in soil pores, ice impacts flow paths taken by infiltrating petroleum, resulting in extensive lateral distribution and possibly deeper penetration into the subsurface as petroleum seeks preferential paths with relatively low ice contents. In layered frozen soils, complex distributions of petroleum will develop as infiltrating petroleum encounters soil layers saturated with pore ice. Segregated ice (ice lenses) formed in fine grain soils will also impact the flow paths taken by infiltrating petroleum by creating impermeable barriers to flow. During thawing, petroleum released to a frozen soil will redistribute as the properties of the porous media change and water is added through thawing ice contained in the soil and from infiltration from thawing snow and ice on the ground surface. In fine soils containing segregated ice, petroleum movement may be enhanced as the ice melts and petroleum flows through the relatively higher permeable soils where the segregated ice existed.

18.3.3 Influence of Freezing and Thawing Cycles on Petroleum Distribution

As is known, the freezing–thawing processes are attended by structure-forming processes which result in changes in soil properties, which in turns influence petroleum redistribution in the soil and its transformation, fractionating and formation of organic-mineral composition. Results from the experimental investigations of Chuvilin et al. (2001a, b) showed cryogenic expulsion of petroleum from freezing to thawing zone in several different freezing soils (Fig. 18.3). Barnes et al. (2004) showed with a mass balance that the primary mode of downward petroleum migration in a freezing soil is through ice formation in the pore space, resulting in displacement of petroleum out of the pore as the void is filled with ice. The resulting crystallization pressure is usually enough for petroleum displacement, due to non-polar nature of the liquid leading to only slight connectivity with mineral particles.

Petroleum distribution in the pore space, composition of the petroleum, initial content in soils, and freezing speed all influence the efficiency of cryogenic expulsion; for example, in sandy soils the amount of petroleum expulsion into underlying unfrozen soil is more than in clay soils. A coefficient of oil expulsion can be used to quantify the efficiency of cryogenic expulsion. This coefficient is equal to the ratio of displaced petroleum to the initial petroleum content. The experimental developed relation of the coefficient of oil expulsion from freezing rate is shown in Fig. 18.4.

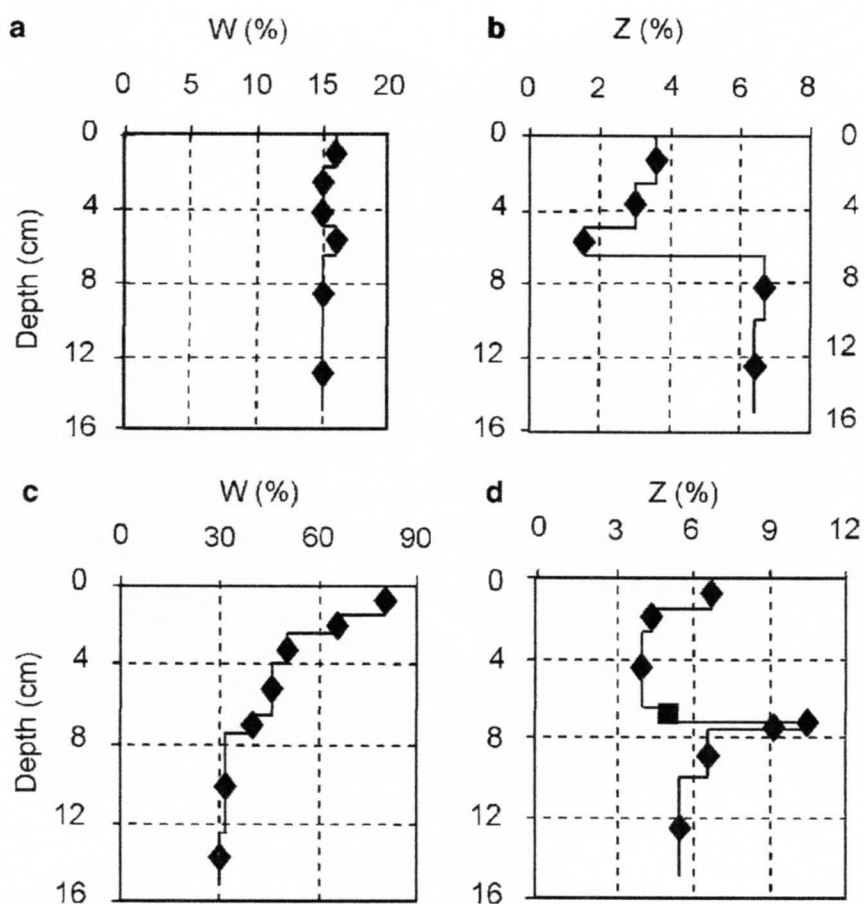


Fig. 18.3 Pattern of the water content (w) and petroleum content (Z) with height of soil freezing at -7°C . **a, b** Sand (initial water and petroleum content 16% and 5% respectively). **c, d** Clay (initial water and petroleum content 43% and 5.4% respectively)

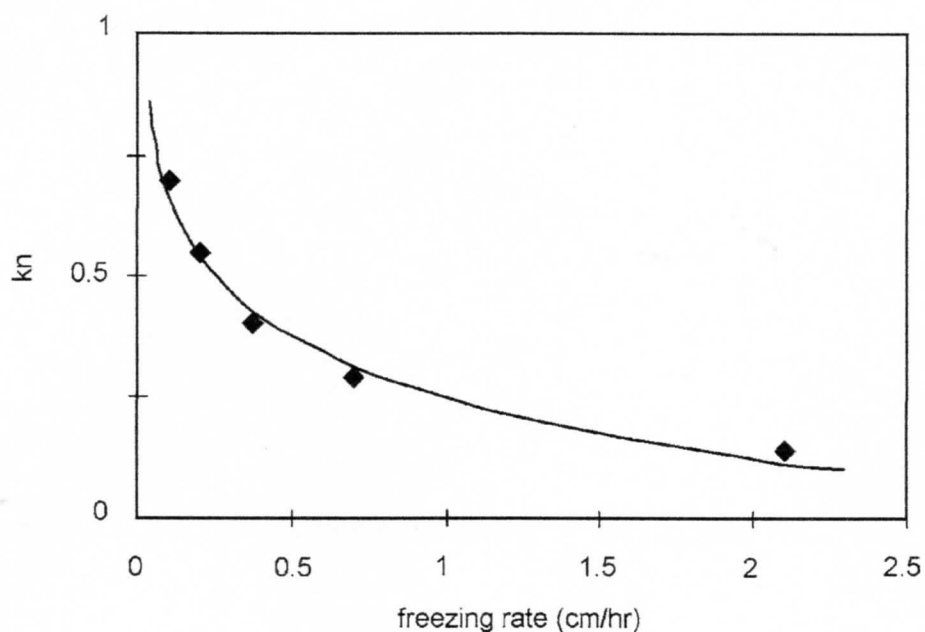


Fig. 18.4 Influence of the freezing rate on the coefficient of oil expulsion (K_n) in sand samples (initial water and oil content 16% and 5%, respectively)

The displacement of petroleum from the frozen soil to the unfrozen soil in the freezing soil sample shown in Fig. 18.4 was determined to be 70% from initial petroleum content under the favorable conditions of the test. In part, we can assume that the cryogenic expulsion is related to the petroleum “cryogenic metamorphization” — the separation of the more mobile petroleum hydrocarbon components from the petroleum. These hydrocarbons then migrate ahead of the freezing front. This process is poorly studied. One can suppose that naphthenes will be more mobile. Naphthenes are saturated hydrocarbons which don’t display the associative properties under temperature reduction. In nature, the cryogenic expulsion may be the significant factor contributing to the petroleum’s mobile formations and further dissipation. This process could have predominant influence in the active layer drained soils, where petroleum hydrocarbons partition into infiltrating water and migrate downward further into the soil horizon.

Laboratory studies of microstructure of freezing oil polluted sediments by White and Williams (1999) and White and Coutard (1999) have shown that their microstructure in frozen soil containing petroleum differs from frozen soil without petroleum under the same conditions. Soil structure change with addition of petroleum depends on the petroleum concentration in the soil. Relatively small concentrations of petroleum (below 200 ppm) promote the aggregation of particles and an increase in sediment porosity, resulting in an increase in hydraulic conductivity. Relatively high content of petroleum, on the contrary, prevents soil particle adhesion, resulting in sediment consolidation and an associated decrease in porosity and hydraulic conductivity. A four-fold increase in hydraulic conductivity (2.9×10^{-4} – 9.8×10^{-4} cm s⁻¹) relative to uncontaminated material was observed where petroleum hydrocarbon concentrations were 50 and 200 ppm TPH (total petroleum hydrocarbons), in a silt subjected to four freeze–thaw cycles. When TPH values approached 1,000 ppm, hydraulic conductivity decreased from 2.9×10^{-4} cm s⁻¹ (uncontaminated silt) to between 5.3×10^{-5} and 8.5×10^{-5} cm s⁻¹.

Grechischev et al. (2001a, b) investigated the influence of petroleum on the formation of segregated ice in fine grain soils. These researchers found that formation of ice lenses depends on composition and properties of the petroleum (crude oil in these studies) contained in the soil. Crude oil with relatively high hardening temperature (above 0°C) was found to reduce the ice segregation and cryogenic heaving of sediments. The influence of low-temperature crude oils is the opposite. Samples containing crude oil were characterized by the magnitude of the resulting cryogenic heaving. For crude oil with low hardening temperature (about –20°C), the value of ice segregation and cryogenic heaving was measured to be almost two times larger than for soils containing no crude oil (Grechischev et al. 2001a, b).

Recently, Haghighi and Ghoshai (2007) have used X-ray computed tomography (CT) to image petroleum (gasoline in this study) in freezing and thawing soils. The use of non-invasive imaging techniques allowed visualization and quantification of petroleum mobilization and displacement, and changes in petroleum blob morphology (volume, specific surface area and fractal dimension) in soil during freezing and thawing conditions. These researchers observed significant mobilization of petroleum from middle sections of the column towards the column end during

freezing. Petroleum volumes changed by up to 150% in certain regions of the column. Porosity distribution in the column changed with freezing, but porosity changes were reversible on thawing. The mean volume of the petroleum blobs increased significantly after freeze-thaw at the two column ends where petroleum migrated, and the blobs over the entire column became more spherical in shape with freeze-thaw. This research confirms redistribution of petroleum and its complicated transformation at freezing and thawing.

18.4 Migration of Petroleum into Permafrost

Petroleum hydrocarbons have been measured at depths of meters in permafrost (Biggar et al. 1998; McCarthy et al. 2004) even though petroleum migration into permafrost should typically be minimal, due to high pore-ice saturations in the upper few meters of these frozen soils. Presence of petroleum hydrocarbons in both these cases was attributed to free-phase petroleum movement through interconnected air voids in the frozen soil. These air voids may result from unsaturated compacted soil, fissures resulting from thermal contraction, or naturally occurring air voids in granular material (such as beach deposits) due to natural processes.

Frozen fine soils can contain unfrozen water at the soil surface boundary. Lacking pathways for petroleum to flow advectively into ice-rich permafrost, a possible transport mechanism is diffusion of petroleum hydrocarbons through the unfrozen water content. Aqueous phase diffusion is a relatively slow transport process in comparison to advection. The contribution this transport mechanism makes to moving contaminants into permafrost soils is most likely minimal. The role of diffusion in the movement of petroleum hydrocarbons into permafrost can be shown with a simple example. Consider the following solution to Fick's Second Law, with a constant concentration of a dissolved petroleum hydrocarbon at the top of a deep layer of permafrost that does not contain the petroleum hydrocarbon initially.

$$C_w(z, t) = C_{w,0} \operatorname{erfc}\left(\frac{z}{\sqrt{4\alpha t}}\right). \quad (5)$$

In (5), $C_w(z, t)$ is the dissolved phase concentration of the petroleum hydrocarbon in the unfrozen soil-water as a function of time and space, $C_{w,0}$ is the dissolved phase petroleum hydrocarbon concentration in the soil-water at the top of the permafrost, z is the depth into the permafrost from the top of permafrost, α is the effective diffusion coefficient divided by the retardation coefficient, and erfc is the complementary error function.

For this example, assume that a release of petroleum has occurred in a permafrost region and that the water-saturated zone in the active layer above the permafrost contains benzene at a concentration that is equivalent to the product of the

compounds solubility and its mass fraction in the released petroleum. The retardation coefficient for benzene in this scenario is 8.55. The temperature of the permafrost is -3°C and the soil is comprised of Fairbanks Silt. From Tice et al. (1976) the unfrozen volumetric water content can be estimated to be 0.062. Assuming the soil to be ice-saturated in the region just below the top of permafrost, the porosity available for diffusion is equivalent to the volumetric unfrozen water content. The resulting effective diffusion coefficient is $2.6 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$. With these reasonable assumptions and considering diffusion as the only transport mechanism, after 10 years the concentration of benzene at a depth of 0.1 m into the permafrost from the top of the permafrost is only 2.2% of the initial concentration. Hence, under the conditions of this example, which are reasonable, movement of petroleum hydrocarbons into permafrost by diffusion is much too slow to be of concern.

18.5 Conclusion

Through laboratory and field studies we are beginning to gain a better understanding of how petroleum migrates through Arctic and Antarctic terrestrial environments. The presence of ice in soils found in these environments greatly influences petroleum migration at the time of release and during subsequent freezing and thawing cycles. Possibly the most predominant effect ice contained in the pore space has on the migration of released petroleum is the formation of preferential pathways, resulting in wider lateral petroleum distributions than would be expected in soils not impacted by extreme cold temperatures. Moreover, freeze and thaw cycles tend to increase the downward migration of petroleum and influence the distribution of disconnected petroleum blobs.

In addition to ice influencing petroleum migration and distribution, the typically shallow nature of the active layer and the resulting thin layer of suprapermafrost ground water impacts the vertical distribution of petroleum in the subsurface. In temperate climates with thick saturated zones, petroleum (as a free phase liquid) does not penetrate past the top few tens of centimeters of saturated soil. Given the thin nature of the saturated zone above permafrost, petroleum will distribute throughout the entire suprapermafrost saturated zone, resulting in dissolved phase plumes distributed throughout the entire depth of the saturated zone, and minimal dilution of the dissolved phase plume by uncontaminated ground water.

An understanding of these processes is necessary as petroleum-impacted areas of the Arctic and Antarctic are cleaned up over the next several decades. More study is needed, however. One of the main topics that require further attention is the validation of what is being measured in laboratory studies and described in theoretical studies against what is occurring in the field. Mackay et al. (1974a, b, 1975) as well as Johnson et al. (1980) provide well-described results from controlled field studies; however, these studies took place over 30 years ago. Laboratory and theoretical studies have focused our attention on influences that these past researchers may have not been aware of and thus not looked for during their studies. Additional

field studies will greatly improve our understanding of petroleum migration in these environments and improve our response methods.

References

- ADEC (2004a) Situation report Nana-Lynden Red Dog truck rollover. Alaska Department of Environmental Conservation, Division of Spill Prevention and Response, Prevention and Emergency Response Program, 12 August 2004
- ADEC (2004b) Situation report Point Hope Day Tank overfill. Alaska Department of Environmental Conservation, Division of Spill Prevention and Response, Prevention and Emergency Response Program, 17 March 2004
- AMAP (1998) AMAP assessment report: Arctic pollution issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, p 669
- Barnes DL, Wolfe SM (2008) Influence of ice on the infiltration of petroleum into frozen coarse grain soil. *Petrol Sci Technol* 26:856–867
- Barnes DL, Wolfe SM, Filler DM (2004) Equilibrium distribution of petroleum hydrocarbons in freezing ground. *Polar Rec* 40:245–251
- Biggar KW, Haidar S, Nahir M, Jarrett PM (1998) Site investigation of fuel spill migration into permafrost. *J Cold Reg Engin* 12:84–104
- Charbeneau RJ, Johns RT, Lake LW, McAdams MJ (1999) Free-product recovery of petroleum hydrocarbon liquids. American Petroleum Institute, Health and Environmental Sciences Department, Publication no 4682
- Chuvilin EM, Naletova NS, Miklyaeva EC, Kozlova EV, Instanes A (2001a) Factors affecting spreadability and transportation of oil in regions of frozen ground. *Polar Rec* 37:229–238
- Chuvilin EM, Miklyaeva ES, Kozlova EV, Instanes A (2001b) Experimental study of freezing soils contaminated by oil. In: *Proceedings of the 7th International Symposium on Thermal Engineering and Sciences for Cold Regions*, July 12–14, Seoul, Korea, pp 145–149
- Farr AM, Houghtalen RJ, McWhorter DB (1990) Volume estimation of light nonaqueous phase liquids in porous media. *Ground Water* 28:48–56
- Grechishev SG, Instanes A, Sheshin JB, Pavlov AV, Grechishcheva OV (2001a) Laboratory investigation of the freezing point of oil-polluted soils. *Cold Reg Sci Technol* 32:83–189
- Grechishev SG, Instanes A, Sheshin JB, Pavlov AV, Grechishcheva OV (2001b) Laboratory studies of the oil-contaminated fine-grained soils freezing and their negative temperature fabric model. *Cryosphere Earth* N2:48–53
- Haghighi SK, Ghoshai S (2007) Freeze–thaw induced mobilization of gasoline in soils. In: *Proceedings of the 5th Biennial Workshop on Assessment and Remediation of Contaminated Sites in Arctic and Cold Climates*, Edmonton, Canada, pp 178–187
- Hinzman LD, Kane DL, Ming-Ko W (2005) Permafrost hydrology. In: Anderson MG (ed) *Encyclopedia of hydrological sciences*, vol 4. Wiley, West Sussex, England, pp 2679–2693
- International Council of Scientific Unions (1993) Protocol on environmental protection to the Antarctic treaty, SCAR Bulletin 110, July 1993. *Polar Rec* 29:256–275
- JIC (2006) Media update 6: Unified command arrives at spill volume estimate. Unified Command Joint Information Center, Alaska Department of Environmental Conservation, Juneau, Alaska
- Johnson LA, Sparrow EB, Jenkins TF, Collins CM, Davenport CV, McFadden TT (1980) The fate and effect of crude oil spilled on subarctic permafrost terrain in interior Alaska. Environmental Protection Agency, Corvallis Environmental Research Laboratory, Office of Research and Development, EPA-600/3–80–040, 127 pp
- Lenhard RJ, Parker JC (1990) Estimation of free hydrocarbon volume from fluid levels in monitoring wells. *Ground Water* 28:57–67

- Mackay D, Charles ME, Phillips CR (1974a) The physical aspects of crude oil spills on northern terrain. Northern Pipelines, Task Force on Northern Oil Development, Environmental – Social Committee, Report 74–25:145
- Mackay D, Charles ME, Phillips CR (1974b) The physical aspects of crude oil spills on northern terrain (second report). Northern Pipelines, Task Force on Northern Oil Development, Environmental–Social Committee, Report No 73–42:213
- Mackay D, Charles ME, Phillips CR (1975) The physical aspects of crude oil spills on northern terrain (final report). Arctic Land Use Research Program, Northern Natural Resources and Environmental Branch, Department of Indian Affairs and Northern Development, INA Publication No QS 8060–00–EE–A1:172
- McCarthy K, Walker L, Vigoren L (2004) Subsurface fate of spilled petroleum hydrocarbons in continuous permafrost. *Cold Reg Sci Technol* 38:43–54
- Mercer JW, Cohen RM (1990) A review of immiscible fluids in the subsurface: Properties, models, characterization and remediation. *J Contam Hydrol* 6:107–163
- Poulsen MM, Kueper BH (1992) A field experiment to study the behavior of tetrachloroethylene in unsaturated porous media. *Environ Sci Technol* 26:889–895
- Rawls WJ, Brakensiek DL, Saxton KE (1982) Estimation of soil water properties. *Transact ASAE* 24:1316–1320
- Rike AG, Haugen KB, Borresen M, Engene B, Kolstad P (2003) In situ biodegradation of petroleum hydrocarbons in frozen arctic soils. *Cold Reg Sci Technol* 37:97–120
- Snape I, Riddle MJ, Stark JS, Cole CM, King CK, Duquesne S, Gore D (2001) Management and remediation of contaminated sites at Casey Station, Antarctica. *Polar Rec* 37:199–214
- Snape I, Gore D, Cole CM, Riddle M (2002) Contaminant dispersal and mitigation at Casey Station: An example of how applied geoscience research can reduce environmental risk in Antarctica. *R Soc N Z Bull* 35:641–648
- Spiess, B (2001) Bullet pierces pipeline. *Anchorage Daily News*, 5 October 2001
- Tice AR, Anderson DM, Banin A (1976) The predictions of unfrozen water content in frozen soils from liquid limit determinations. US Army Cold Regions Research and Engineering Laboratory Report CRREL 76–78
- Vilchek GE, Tishkov AA (1997) Usinsk oil spill – environmental catastrophe or routine event? In: Crawford RMM (ed) *Disturbance and recovery in Arctic lands*. Kluwer, Dordrecht, pp 411–420
- White TL, Coutard JP (1999) Modification of silt microstructure by hydrocarbon contamination in freezing ground. *Polar Rec* 35:34–41
- White TL, Williams PJ (1999) The influence of soil microstructure on hydraulic properties of hydrocarbon-contaminated freezing ground. *Polar Rec* 35:25–32
- Wilson JL, Conrad SH, Mason WR, Peplinski W, Hagan E (1990) Laboratory investigation of residual liquid organics from spills, leaks, and the disposal of hazardous wastes. Robert S. Kerr Environmental Research Laboratory Office of Research and Development, US EPA, EPA/600/6–90/004:267