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Effect of freezing on the level of contaminants in uncontrolled hazardous waste sites Part 1: Literature review

I.K. Iskandar

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used to cover hazardous waste sites. Ground freezing can be used beneficially to 1) de-water and consolidate hazardous waste materials, particularly slurry-type wastes; 2) serve as an alternative to slurry walls, trenches, etc., to separate contaminated areas; and 3) immobilize the contaminants, particularly if time is a critical factor. All hardware needed for artificial ground freezing is available, and in the United States there are at least three commercial firms equipped to do such work. In addition to the versatility of this technique, little or no environmental impact is associated with its use. A hypothetical example using artificial ground freezing is presented to illustrate both the data needed to analyze a situation and the costs involved.

PREFACE

This report was prepared by Dr. I.K. Iskandar, Chief, Geochemical Research Branch, Research Division, U.S. Army Cold Regions Research and Engineering Laboratory. This project was jointly funded by the U.S. Environmental Protection Agency through the Interagency Agreement DW930180-01-0 and CRREL through program 6.11.02A; DA Project 4A161102AT24.

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CONVERSION FACTORS: U.S. CUSTOMARY TO METRIC (SI) UNITS OF MEASUREMENTS

These conversion factors include all the significant digits given in the conversion tables in the ASTM Metric Practice Guide (E 380), which has been approved for use by the Department of Defense. Converted values should be rounded to have the same precision as the original (see E 380).

Multiply	By	To obtain
inch	25.4	millimetre
foot	0.3048	metre
acre	4046.873	square metre
cubic foot	0.02831685	cubic metre
cubic yard	0.7645549	cubic metre
bar	100,000.0	pascal
ton	907.1847	kilogram
inch per hour	0.0004234	metre per second
pound per foot hour	0.0004133789	pascal second
degrees Fahrenheit	$t_{\circ C} = (t_{\circ F} - 32) / 1.8$	degrees Celsius
pound (force) per square inch	0.006894757	megapascal

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EFFECT OF FREEZING ON THE LEVEL OF CONTAMINANTS IN UNCONTROLLED HAZARDOUS
WASTE SITES - PART 1: LITERATURE REVIEW

I.K. Iskandar

INTRODUCTION

Nature of wastes at uncontrolled hazardous waste sites

One of the major environmental problems currently facing the U.S. is treatment and disposal of hazardous wastes. The EPA (1982) estimates that in the U.S. 65 million tons are now generated and that this is increasing rapidly. In addition to hazardous waste, municipal, mining and agriculture wastes are also generated and must be disposed of properly. The following is an estimate of the types and amounts of wastes produced annually (EPA 1982): 1) 36 million tons of municipal waste disposed of in 18,500 sites, 2) 5.5 million tons of municipal wastewater sludge (EPA thinks that this number will double within this decade because of the higher treatment level given to wastewater), 3) more than 132 million tons of flue gas cleaning sludges, and 4) billions of tons of agricultural and mining wastes.

Although people are concerned about the proper treatment and disposal of hazardous wastes produced now and in the future, they are even more concerned about past disposal practices. A large number of uncontrolled hazardous waste sites have been discovered (EPA 1982), and this is only a fraction of the number of existing sites. Many of the chemicals disposed of on land have been shown to cause cancer, birth defects and other types of harm to humans, animals and plants. Although these sites have been abandoned, the damage to the environment and to humans has either already occurred or can be expected in the future. When remedial action has been taken in these cases, it is mainly for preventing further transport of contaminants. The cost of cleaning up contaminated soil and water is usually very high and this has often led to temporary solutions.

The release of contaminants into the environment following accidental spills is also a major concern. In spite of our awareness of the danger, spills will continue to happen, although perhaps with less frequency than in the past.

Table 1. Types of hazardous waste facilities where cleanup is being attempted (after Neely et al. 1981).

Facility type	Status		Total
	Active	Inactive	
Landfill	16	37	53
Dump	0	27	27
Drum storage	11	25	36
Surface impoundment	18	37	55
Injection well	1	3	4
Incinerator	1	5	6
Spill	0	23	23
Total	47	157	204

The physical properties of hazardous waste cover the spectrum from solids and liquids to gases. The chemical properties also vary enormously. The waste can be almost any type of chemical or a mixture of chemicals and soil materials. Because of this, there is no standard method or procedure for cleanup. In 1980, an engineering consulting firm (Neely et al. 1981) surveyed 204 disposal facilities being rehabilitated (Table 1). Their study evaluated the effectiveness of attempts to clean up ground and surface water at nine sites, and they found that the cleanup was ineffective at four of them. Only in a portion of all 204 uncontrolled sites has an attempt been made at cleaning them up, and the action was effective at only one site. The report cited the high cost of remedial action. The consulting engineers felt that the main reason for the dismal cleanup record was the lack of innovative and effective techniques for cleanup.

In summary, treatment and disposal of both hazardous and nonhazardous wastes is a major problem currently facing the engineering community. Conventional methods are not adequate for ensuring environmental safety and they are expensive. One possible innovative technique is artificial and natural freezing. This has been used for many years for other purposes and should be tested for hazardous waste.

Ground freezing technology

Ground freezing is not a new technology. It has been used with shaft sinking for over 100 years, and, in recent years, is being increasingly used in civil engineering, mainly because of the following advantages:

1. It can be used, in principle, for all types of soils where water is present.

2. It is a very flexible construction method that can meet many boundary conditions and requirements.

3. It causes very few or no environmental problems. However, very careful site investigations, engineering work and construction are required for success.

During ground freezing, the temperature of the soil water is lowered below the freezing point. The freezing point of soil solutions is not at 0°C as for pure water. Dissolved ions in the soil solution lower the freezing point; this is called freezing point depression. Many of the early studies of freezing and frozen soils were done by Bouyoucos and McCool (1915, 1916, 1928). They compiled data on the freezing point depression of 58 soils and used these data to measure solute concentration in soil solutions. Page and Iskandar (1978) used an empirical relationship between pore water salinity and the freezing point in subsea permafrost sediments to predict bounded and unbounded frozen sediments in the Beaufort Sea. This empirical equation can be used to predict the freezing temperature of soils in uncontrolled hazardous waste sites.

When the soil temperature is lowered to the freezing point, ice forms, leading to an important change in the soil properties. The strength of the soil is substantially increased, while at the same time, the soil becomes impermeable. The potential use of ground freezing in cleaning up hazardous waste is based on these two important points. The increase in soil strength upon freezing means that a frozen zone of soil can be formed around or underneath a hazardous waste site or between a hazardous waste site and an uncontaminated environment, without injecting grout or adding materials such as concrete, slurry walls or steel sheet pile walls. Also, the frozen zone of soil becomes impermeable to contaminants and water.

To determine if ground freezing can be used effectively at hazardous waste sites, an understanding of the processes that occur in freezing and frozen soils is reviewed. Also, a general overview of some projects where ground freezing was used and a cost analysis of artificial freezing are provided.

Objectives

The objectives of this report are to review the literature on:

1. The effects of freezing on water, wastewater and sludges, and on chemicals in the soil -- both the effects that are harmful to existing

hazardous waste sites and those that could be used to control the spread of hazardous wastes.

2. Case histories of artificial ground freezing in civil engineering projects.

3. The economic aspects of ground freezing as a cleanup alternative and the availability of hardware for such a job.

In addition, two example cases of using ground freezing to contain hazardous waste spills are given.

WATER, WASTEWATER AND WATER TREATMENT SLUDGE

Freezing of saline water, wastewater and sludge has long been studied. Desalination of seawater by freezing, a process where the water is separated from dissolved salts, was studied by Heller (1939), Geller (1962), Szekely (1964), and Stinson (1976). In these studies the authors used natural freezing to form ice of high purity, leaving behind a concentrated salt solution (brine). The ice was then washed out of the salt and melted to produce fresh water. Although this purpose is different from our interest in using natural and artificial freezing to immobilize contaminants in uncontrolled hazardous waste sites, the same principles, and perhaps the same mechanisms of contaminant rejection upon freezing, can be applied for treatment of contaminated leachate, industrial wastewater and lagoon water.

A preliminary design for renovating municipal wastewater by freezing was adopted in a project supervised by the U.S. Public Health Service (Barduhn 1963). The freezing of wastewater and thawing of ice reduced the salt content of water to acceptable levels. Although the cost of freezing was high, it was less expensive than conventional desalination methods. Inorganic materials can also be removed from wastewater effluents by freezing the feed water slowly. Although its purpose is demineralization, the process is also capable of removing organic materials from water. However, separating the organics from the ice crystals can be difficult.

Several reports are available regarding treating by natural and artificial freezing the sludge left over from water purification. Treatment of water for municipal use frequently includes adding alum [$Al_2(SO_4)_3$] as a coagulant, resulting in the presence of $Al(OH)_3$ in the sludge. Bishop and Fulton (1968) and Bishop (1971) suggested using two lagoons for settling

and thickening water plant sludge, lagoons that would be large enough and shallow enough to allow for complete freezing during the winter. They recommended decanting and returning the supernatant to the treatment plant during the fall to decrease the depth of sludge and allow for complete freezing. They predicted, but did not show, that several seasons' worth of solids could be accommodated and that a front-end loader could be used to remove the dry solids in the summer. In their experiments freezing increased the total solid concentration from 3.5 to 17.5%. The authors did not present a cost analysis but predicted that the cost was likely to be less than that of a filtration plant.

Sludge treatment by freezing in a New York Community Water Treatment Plant was described by Fulton (1970). This system used an existing headrace of an adjacent abandoned power plant to dewater the sludge, instead of constructing lagoons as Bishop and Fulton (1968) suggested. The headrace was divided into two sections. The consolidated solids in one section were pumped to the other and allowed to freeze during the winter. Fulton observed that freezing changed the "jelly-like" consistency of the $\text{Al}(\text{OH})_3$ suspension into a granular form.

More recently, Nekoosa Paper Inc. (1981) described a system at Nekoosa and Port Edwards, Wisconsin, that was constructed in 1980 and found to be effective in dewatering both sludge from a water treatment plant and sludge from a paper mill. The system is being monitored, and the factors affecting the degree of dewatering are being studied, including snow cover (and removal), sludge volume, solids concentration and thickness of the ice cover in the lagoons. The company claims that the sludge was thickened from 3 to 20% and that the leachate was not contaminated.

The use of artificial freezing for treatment of water in Yorkshire, England, was described by Ben and Doe (1969). In this system, installed in 1961 and 1963, the continuous flow of sludge was collected in special steel tanks to overcome the stress produced by ice formation. The cost of this system was very high, and the authors concluded that unless natural freezing is used, the process will not be cost effective.

In her study, Bitterli (1981) collected leachate from a simulated solid waste container and spiked subsamples with heavy metals such as Cd, Zn and Fe. She froze the leachates in a freezer at -12°F overnight, allowed them to thaw, centrifuged them at 5000 rpm, filtered them through

0.45- μm filters, and determined the amounts of metals in solution. She found that the effect of freezing and thawing was similar to that of drying and wetting: decreasing the amount of constituent available to migrate through the soil. She explained that pH increases by wetting-drying or freezing-thawing, and as a result metals precipitate. She added that the increase in pH occurred during the first freeze-thaw cycle and that there was very little increase with additional cycles. In comparing the effectiveness of drying-wetting with freezing-thawing on ion mobility, Bitterli concluded that drying-wetting has more effect on increasing precipitation.

SEWAGE SLUDGE

Sewage sludge is a solid waste, and in a practical sense it can be considered one type of hazardous waste, especially if it contains high concentrations of metals or organic chemicals. Because of its huge volume, municipal sewage sludge dewatering has long been a problem facing sanitary engineers. Over the past 30 years, some interest has been shown in using freezing and thawing of sludge as conditioning before dewatering. Although the beneficial use of freezing sludge has been demonstrated, few systems are in actual operation.

As early as the 1920's, Babbitt and Schlenz (1929) accidentally discovered that upon freezing and thawing the moisture content in a digested sludge sample dropped substantially, and that the sludge developed a spongy appearance, having little cohesion and no odor. The sludge was separated easily from the sand beds. In the late 1940's, Clements et al. (1950) conducted several experiments on freezing primary, digested and activated sludges. They also studied the effects of chemical additives on the process. Garinger (1972) summarized the conclusions drawn from this study.

The settlement of all types of sewage sludge is promoted by freezing. It is accelerated by adding chemicals, but the percentage settlement at the end of an hour is approximately the same whether chemicals are added or not. The chemicals used were chlorinated ferrous sulfate, chlorine gas or alum, and the doses were up to 1000 mg/L. Filtration after freezing with chemicals substantially accelerated the settlement. The best results were obtained by the use of Alum, where dry solids production reached 350 lb/ft³ per hr. Complete freezing is essential, but it must be fairly slow; flash freezing is ineffective. The method of thawing is immaterial, as long as

it is not associated with vigorous agitation. The supernatant liquids, on settlement, are not much worse than ordinary sewage.

Coakley (1955) conducted independent studies on sludge freezing in England, and his conclusions were in general agreement with those of Clements et al. (1950).

Until 1962, a system in Winnipeg (Babbis 1962) was probably the only one in operation where sludge was routinely freeze-dried in lagoons. The procedure at that site involved applying a 10-in. layer of digested sludge, allowing it to stand for 2 weeks, then decanting the surface liquid and allowing the solids to freeze. When the lagoon is filled, the sludge is removed with a heavy scraper. Andrews (1967) mentioned that frozen sludge in Winnipeg is removed from beds and placed on concrete pads, where it rapidly dewateres in spring. He suggested that the effect of freezing is to overcome the attraction of the colloidal particles for water, permitting the water to drain rapidly upon thawing.

Burd (1968), studying sludge handling and disposal, stated that sludge frozen in the winter by nature and later allowed to thaw in sand, drying beds or lagoons had good dewatering and soil-conditioning properties. He suggested provisions for water drainage for rapid stabilization and dewatering, and mentioned that artificial freezing aids in sludge dewatering. He concluded, however, that it will never be economical except in very isolated cases. The high operating costs encountered were partially caused by the need for dewatering equipment to handle high production rates.

The Federal Water Quality Administration (Balakrishnan et al. 1970) also reported on conditioning sludge by freezing. Freezing disrupts the cell walls retaining the internal moisture in sludge and allows the water to be released and drained, but slow and complete freezing was reported to be necessary for good dewatering results. About 8.2×10^{-4} kWh was required to lower the temperature of 1 lb of sludge from 60° to 32°F, and an additional 4.16×10^{-2} kWh was then required to freeze this pound of sludge. To be feasible, artificial freezing must result in a marked improvement in sludge properties.

Cheng and Updegraff (1970) experimented with a film-freezing principle in which freezing time and the temperature driving force were greatly reduced over those used before. They also believed that additives such as alum are necessary for efficient dewatering. The artificial freezing process was controlled via the external heat exchange and the mixing rate.

The rate of production of solids was similar to that obtained by Clements et al. (1950).

In 1975 an experimental program was begun at the Wastewater Technology Centre, Burlington, Canada, to develop engineering design criteria for natural freeze-thaw dewatering of sewage sludge. Rush and Stickney (1979) summarized the laboratory findings of the first 4 years of study and presented preliminary design criteria. In this system, four types of sewage sludge were tested. These were conventional activated, extended aeration waste activated, anaerobically digested and aerobically digested sludge. The study concluded that freezing caused a remarkable improvement in the dewatering of all the sewage sludge types tested. The filtrate water quality after freezing and thawing was three to six times poorer than that of the raw sewage. However, on a mass balance basis, the filtrate water returned to the treatment plant would only increase the hydraulic and organic load by less than 0.8 and 5.0% respectively. With some modification, this procedure can be used in natural freezing of some hazardous waste sites.

More recently, Chamberlain and Blouin (1977) reviewed the literature on dewatering of sludge or dredged material by freezing and thawing. They concluded that freeze-thaw increased the density of the material because of aggregation and restructuring of soil particles, densification of aggregates and segregation of ice. Chamberlain and Blouin investigated the effects of freeze-thaw cycles on material consolidation, permeability and bulk density. They found that natural freezing could be beneficial for dewatering dredged material.

The literature contains substantial evidence that freezing and thawing of moist solid waste can be used to improve physical properties and, so, also for the treatment of uncontrolled hazardous waste sites. Freezing and thawing is particularly useful for reducing the volume of contaminated sludge, sediments, soils or waters. This enables us to handle the waste with mechanical devices for further treatment or ultimate disposal in a properly designed facility. Freezing can concentrate solids in sludge to up to 20%.

Natural freezing saves substantial amounts of energy and money. However, it can only be used in the northern states. During emergencies in warmer months and in warmer climates, artificial freezing must be used.

PROCESSES IN GROUND FREEZING AND THEIR EFFECTS

Water movement

In the simplest description, soils consist of three phases: solids, water or solutes, and gases. The solid portion of the soil consists of clay minerals, amorphous materials and organic residues. Anderson and Morgenstern (1973) summarized the many effects of freezing on soil physical and chemical properties that are important for engineering. The phenomenon of frost heave is probably the most widely recognized effect. Frost heave results from the growth of ice lenses in soil during freezing, causing an expansion of the soil mass and a subsequent rise of the soil surface above the freezing zone. Ice lens growth requires water movement to the lenses from the unfrozen soil beneath.

Frost heave should be considered in uncontrolled hazardous waste sites where frost penetration reaches the depth where waste materials have been buried. For example, soil-covered asbestos, or even buried barrels containing chemicals, could slowly move upward as a result of frost heaving. If these sites were properly designed the soil cover would be deep enough so that frost could not reach the covered hazardous waste.

Prior to a discussion of water and ion movement in freezing and frozen soils, a brief review of the state of knowledge about water in frozen soils is essential.

It is well known (Anderson and Tice 1971) that in freezing and frozen soils, water exists in three forms: vapor, liquid and ice, in variable amounts. The liquid-phase water in frozen soils is believed to be mobile, even at temperature as low as -14°F (Anderson and Morgenstern 1973). The amount of liquid water in a frozen soil is called its unfrozen water content (Anderson and Tice 1971). The unfrozen water content of frozen soils depends on, among other things, soil type (specific surface area), temperature, and the amounts and types of dissolved solids. It decreases with decreasing temperature and generally increases with finer soil texture (high surface area) and with increasing concentration of dissolved solids. The total surface area of the soil consists of external surface area (area around the soil particles) plus internal surface area (surfaces between layers of expandable clay minerals such as montmorillonite). Anderson and Morgenstern (1973) concluded that temperature is the most important determinant of the amount of unfrozen water in frozen soils.

In addition to temperature and dissolved salts, the overburden pressure and pore geometry are additional factors that affect the amounts of unfrozen water in frozen soils and consequently affect ion migration.

Because soil water freezes slowly, pure ice crystals form, excluding the ions present and concentrating them in the remaining soil solution. The temperature and concentration of this solution affect the chemical reactions and the forms of ions in solution. In nature, soil freezing starts at the surface, and the freezing front moves downwards. Soil water, however, moves in the opposite direction of the freezing front, going from the subsurface layers toward the ice lenses and causing ice growth. The reason for this is that as water freezes in the surface, the tension on the surrounding soil particles increases (soils become dryer); therefore, water redistribution occurs and water moves from the higher to the lower water content zone. This is the same thing that happens when evaporation dries a soil.

Much of the research regarding the infiltration and permeability properties of frozen or partially frozen soils has originated in the Soviet Union. Shalobanov (1903) was among the first to investigate this. He showed that up to 80% of the fall and winter precipitation was absorbed by the soil and that a considerable part of this water penetrated even under a snow cover. Infiltration and permeability rates of frozen soil depend on the soil porosity and moisture content at the time of freezing and on the rate of freezing. Stepanov (1957) showed that the permeability of frozen soils having a high porosity was appreciably higher than when they were unfrozen, even at temperatures as low as 23° to 19°F. He cited an experiment where a frozen soil having a 20% moisture content absorbed water close to saturation. The key point here is whether or not the soil pores are filled with ice. If the soil's pores are filled with ice because it was saturated before freezing, the permeability of the frozen soil will be very low. Permeability increases as the frozen water content decreases.

In general, there is a paucity of quantitative evidence to illustrate the infiltration capacity and permeability of frozen soils. Mosiyenko (1957, 1958) and Stoeckeler and Weitzman (1960) found that the infiltrating rate of water in frozen "concrete" loamy sand soil (many small ice crystals, very densely arranged), which was less than 0.75 in./hr, increased to about 2 in./hr under granular, "honeycomb" (loose porous struc-

ture) frost, and increased further to 4 in./hr and 13 in./hr in partially frozen and unfrozen soils, respectively.

As mentioned above, moisture moves in both vapor and liquid phases through porous media. However, there is considerable controversy over the rate and extent of water movement of each form. Crawford (1951), Hutcheon (1955) and Cary (1966) all give complete reviews of this topic. Jemikis (1960) suggested that the electric double-layer theory shows that the electric charge on the surface of colloidal soil particles and the electrolytic action of the surrounding moisture films are significant in the process of soil moisture translocation upon freezing. Nersesova (1962) indicated that the migration of water in freezing soils increased as the temperature decreased. He explained that this was attributable to a change in the surface energy of the soil's solid phase. The surface energy depends on the kind and valence of exchangeable cations. Nersesova claimed that in freezing soil, the presence of polyvalent cations and H ions decreased the rate of water movement substantially, while monovalent ions tended to accelerate the migration of water and the formation of ice.

Hoekstra (1966) showed that the water film around particles can be in equilibrium with ice and still be quite mobile in frozen soil. He found that these moisture films were continuous throughout the frozen soil matrix, supporting the theory that water moves in frozen soil under the force of electrical gradients. He also presented data to show that moisture flow under thermal gradients in frozen and unfrozen soil are of the same order of magnitude. Liquid-flow values in frozen soil reported by Hoekstra agree well with those of both liquid and vapor flow in unfrozen soil reported by Cary (1966). In both studies, initial soil moisture contents were near field capacity. Ferguson et al. (1964) reported considerable upward movement of water during the winter in Montana. They found that soil water held at tensions of 28.4 to 71.1 lb/in.² moved to the frozen soil surface against its own moisture content gradient and contributed to overwinter evaporation losses from bare plots.

Recently, Chamberlain and Blouin (1977) examined the effect of freeze-thaw cycles on the permeability of dredged materials containing water in the range of 60 to 90% of their liquid limit. They found that the permeability of fine materials increased two-fold upon one cycle of freezing and thawing. In the meantime the water content of the wet materials was decreased by about 20%.

The above findings can be applied to uncontrolled hazardous waste sites as follows.

1. If ground freezing is to be used in treating hazardous waste lagoons and saturated sediments, soils and sludges, freezing from the surface (by natural freezing or installation of freezing probes) will cause water to move upwards, opposite of the freezing direction.

2. As the water in the soil solution freezes, more concentrated solutes will be present in soil solution.

3. Water will travel even in frozen soils and sediments because of the presence of unfrozen water films around the particles.

4. The temperature of freezing is most likely to be much lower in hazardous waste sites than in normal soils because of the high concentrations of chemicals.

Ion migration

The migration of chemicals through unfrozen soils has been extensively investigated in the fields of soil chemistry and physics. The results of these studies have contributed to the solution of many environmental problems. Several mathematical models have been developed to predict the fate of chemicals in unfrozen soils and are in various stages of testing (Dutt and Tanji 1962, Frissel et al. 1970, Dutt et al. 1972, Reiniger et al. 1972, Bolt 1979, Iskandar 1981, Bolt and Bruggenwert 1982). These models vary in complexity and purpose but can generally be classified as management-oriented or research-oriented models.

The management-oriented models are simpler and are aids for proper management. However, very little information can be gained on the fate of constituents in soils using these models. The research-oriented models, on the other hand, are more complex and require more knowledge of soil characteristics and the interactions between ions in solution and soil particles under specific environmental conditions.

Research-oriented models can predict ion movement at hazardous waste sites, although the state of the art does not allow accurate prediction of the fate of chemicals in space and time with the degree of accuracy required. However, valuable qualitative and semiquantitative information on ion transport in soils can be obtained from these models. Also, from sensitivity analyses of model parameters, research can be directed toward useful areas. Iskandar et al. (1981) emphasized the need for field evalua-

tion and validation of these models. They also suggested the use of stochastic approaches, along with analytic or numeric modeling, to overcome site heterogeneity and preferential transport of chemicals in soils. Nevertheless, there exist several mathematical models for predicting ion movement in saturated and unsaturated soils to groundwater. The accuracy of results obtained depends largely on the accuracy of the input data.

Information on the migration of chemicals in freezing and frozen soils is not readily available. One reason for this lack is the difficulty in measuring both the rate of ion diffusion and the factors affecting ion exchange and diffusion in soils. Most researchers agree that a large fraction of the total diffusive flux of ions must go through the free liquid phase in the soil pores.

In addition to ion transport by diffusion, which is important in frozen soils and unfrozen soils where the water content is low enough to allow water and chemical transport by vapor convection action, ions can move during mass transport in freezing and thawing soils. The rate and extent of ion movement in freezing soils by this mechanism is far more important in uncontrolled hazardous waste sites than in unpolluted soils. The magnitude and direction of ion migration in this case will depend upon the concentration gradient of ions in soils, the rate of freezing, the water content, and the type of ions in the soil solution.

Ions usually move from higher concentration sites to lower concentrations sites in the soil solution to reach an equilibrium. However, the soil system is complicated by the presence of active ion exchange and by soil solutions where ions can be present in different forms, including precipitates. In addition, the presence of more than one ion in the soil solution and on the exchange sites and the differences in their ability to replace or compete with each other makes it difficult to predict the fate of chemicals in soils. Several mathematical models with various simplifying assumptions have been developed to predict the rate and extent of ion transport in unfrozen and freezing soils (Van Genuchten and Wierenga 1976, Bolt 1979, Harmsen 1979, Mattigod and Sposito 1979, Harmsen and Bolt 1982).

As the water in the soil solution freezes, pure ice crystals form, excluding the ions present and concentrating the remaining soil solution either in pockets or in the thin liquid films around the particles. The temperature and concentration of this solution could affect chemical reactions, such as the formation of precipitates. If a soil freezes from

the surface downwards, as in nature, the water will move upwards towards the freezing front so that ice lenses form (Beskow 1935). It seems likely that some ions will also move upward along with the water. However, as the temperature goes down and the ice forms, salt again precipitates. There is no firm theory in the literature regarding the location where ions accumulate in the soil upon freezing. Nevertheless, researchers agree that the rate of ion movement in freezing and frozen soils depends on ion type, soil characteristics, and rate and direction of freezing. When the soil freezing is rapid, pockets of concentrated solution may be trapped within the frozen soils. Moore (1972) noted little movement of nitrate in saturated sands during rapid freezing. He attributed this to brine pocket entrapment during freezing.

We know that brine pockets form during the freezing of sea ice. If a temperature gradient exists across the brine pockets, a concentration gradient also exists. This concentration gradient will serve as a driving force for the movement of the brine pockets through the ice. In general, however, salt movement in frozen soils by this mechanism is minimal.

Effects on some soil physical properties

Alternate freezing and thawing cycles have been shown to change soil physical properties, which may or may not be desirable when we are dealing with hazardous waste sites. The dramatic phenomenon known as frost heaving (mentioned earlier) is generally caused by the formation of ice lenses in soils during freezing. The three conditions required for frost heave are subfreezing temperatures, water and a frost-susceptible soil. Millions of dollars are spent annually to repair damage to roads, buildings and bridges caused mainly by frost heaving. In uncontrolled hazardous waste sites, frost heaving can cause upward movement of materials and can damage clay liners used for capping. Until recently, explanations of the mechanism of frost heave were based on the work of Beskow (1935), who believed that frost heaving is caused by water migration to growing ice lenses. In the past two decades, researchers have promoted three fundamental explanations for ice segregation and frost heave: capillary theory, secondary heaving theory and segregation freezing theory. Chamberlain (1981) reviewed those theories and concluded that although they disagree about the mechanism of frost heave, they are in general agreement on the factors affecting frost heave.

Substantial effort has been focused on developing methods for measuring the frost susceptibility of soil as a means of predicting frost heaving. Chamberlain (1981) reviewed available methods and discussed the different soil classification indices based on grain size characteristics. Although these findings are not directly applicable to hazardous waste materials, the procedures and apparatus used for measuring the frost susceptibility of soils might be applied to hazardous waste materials; this, however, must be evaluated. Also, the information gathered on frost heave can be useful in predicting the longevity of a hazardous waste site where soils susceptible to frost heave, such as clay liners and fine-textured soils, were used to cover it.

In addition to frost heave, freezing and thawing may affect soil aggregation, soil structure (Bisal and Nielsen 1964, Bisal et al. 1967) and soil permeability. The influence of freeze-thaw cycles is usually expressed by the change in water-stable soil aggregates or the change in the percentage of erodible particles (usually defined as < 0.04 -in. diameter). Slow soil freezing will cause large ice crystals to form (water expands by about 9% on freezing), with the result that pores are enlarged. Water moves from around the clay particles to the ice, causing local dehydration. The combination of ice pressure and dehydration causes aggregation; the process that has been used beneficially in sludge dewatering. It also results in the formation of friable sludge materials that can be handled easier. Based on this process, ground freezing can be used for treating contaminated sediments in lagoons to dewater the sediments and to change their physical properties to a form that can be handled easier. However, prior to approval of such a project, the fate of contaminants in the sediments during freezing and after thawing must be assessed.

Rapid freezing, on the other hand, can be done in artificial freezing projects using liquid nitrogen, causing the formation of small ice crystals that tend to break down soil aggregates. The effects of freeze-thaw cycles on soil aggregates depend not only on the rate of freezing but also on factors such as the water content before freezing, the conditions of freezing as well as thawing, the soil texture, the water conditions of the soil after thawing, and the chemistry of the soil solution.

Hinman and Bisal (1968) studied the effect of initial water content on soil aggregates upon freezing. They found that neither freezing nor thawing had any appreciable effect on the dry aggregate size when the initial

water content was low. However, as the initial water content was increased, the effect of freeze-thaw cycles on the aggregate size and content increased. The freeze-thaw cycles increased aggregation if the original soils consisted of fine particles (clay), containing water equivalents to 1.4 lb/in.². If the soil texture was coarse, the aggregate size upon freezing and thawing was decreased. Changing soil aggregation and structure affects soil erodibility and soil permeability. An increase in soil aggregation is usually associated with an increase in soil permeability. This means that more water from rainfall will penetrate the soil, and less water will be available for runoff and erosion.

Chamberlain and Gow (1978) also reported on the effect of freezing and thawing on soil permeability and soil structure. They found that freezing and thawing of fine-texture soils reduced the void ratio and increased soil permeability. They attributed the increase in permeability to the formation of polygonal shrinkage cracks or to the reduction of the volume of fines in the pores of the coarse fraction, or both.

Benoit (1973) studied the effect of freeze-thaw cycles on the hydraulic conductivity of aggregate soils. He found a relationship between soil water content and the ratio of final to initial hydraulic conductivity. He noted that the ratio of final to initial hydraulic conductivity increased seven times when the soil water content was 20% (about 0.5 bar), while it decreased ten times where water content was 70% (saturated, about 0 bar).

The conclusions from the previous studies imply that freezing and thawing of fine-textured soils, such as clay liners used for capping hazardous waste sites, may increase their permeability because of the increase in soil aggregates and the formation of vertical cracks. If this happens, rainwater will penetrate through the liners, reaching the waste and dissolving chemicals that may reach groundwater. This process is most likely in the northern portions of the U.S., which have natural freezing for several months during each year. Also, the effect of frost heaving on the integrity of clay liners used for capping must be assessed. Upward movement of large particles of waste materials such as asbestos or barrels as a result of frost heaving should be studied.

CASE HISTORIES OF ARTIFICIAL GROUND FREEZING

As discussed earlier, frozen ground exhibits very high strength, very low deformation and compressibility, and very low permeability. Because of

these characteristics, civil engineers have used ground freezing in construction projects. Artificial freezing was first used in 1862 in Swansea, England, to support a mine shaft. In 1883 Poetsch patented the method of ground freezing with cooling pipes, which, with some modification, is still in use. According to Braun and Nash (1982) the use of ground freezing in mining has advantages over conventional methods (dewatering, grouting, slurry walls, caissons) because:

1. It is less sensitive to advance geological prediction.
2. It provides several temporary functions such as support of an excavation, groundwater control and structural underpinning.
3. It is adaptable to practically any size, shape or depth.
4. It keeps excavation unobstructed as no bracing or sheathing are usually required.
5. It does not disturb groundwater quantity or quality.
6. It is environmentally acceptable, as no chemicals must be added, and there is less disturbance to the site.

Up to 1978, more than 200 deep mine shafts have been driven by artificial soil freezing (Sadovsky and Dorman 1980).

In addition to its use in mining, ground freezing has been used for open excavations and construction of deep, unsupported construction trenches. For example, it was used during the construction of subways in Moscow (Dorman 1971) and in Zurich, Switzerland (Aerni and Mettier 1980). According to Dorman (1971), the use of ground freezing in the Moscow Subway Project saved 700 tons of metals and 17,000 ft³ of timber, and the project was completed 11 to 12 months ahead of schedule.

In North America, artificial freezing has also been used since 1888 (Braun and Nash 1982). In 1959 its use was necessary to enlarge a twin railroad tunnel under Montreal, Canada (Low 1960). Construction problems were caused by the presence of a plastic layer of clay in the soil and the location of the tunnel under two large buildings and service pipelines. In Sao Paulo, Brazil, a 26-story building was saved from continuous settlement (0.08 to 0.24 in./day) by having 162 cooling probes driven under its foundation. The frozen soils around the probes acted as pile foundations and stopped the settlement (Careaga et al. 1975) temporarily until a permanent foundation was constructed.

Liquid nitrogen (LN₂ at -321°F) was used in artificial freezing for the first time in Argenteuil, France, in 1964 when a collector sewage pipe

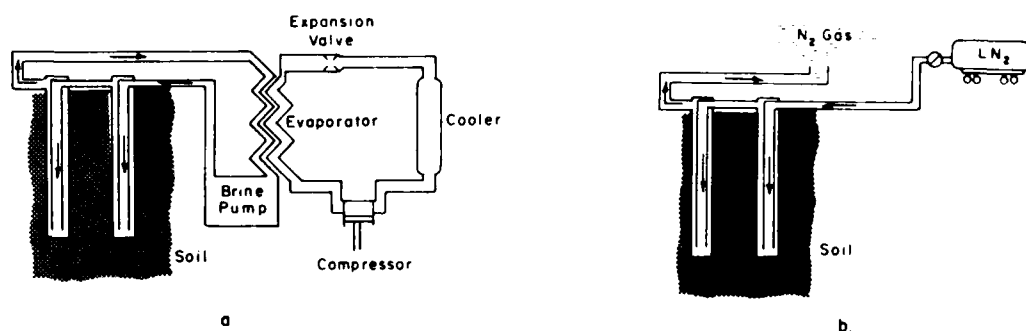


Figure 1. Freezing probes and techniques — with brine (a) and LN₂ (b) (after Jessberger 1980).

in a tunnel was broken and sewage seeped into a nearby stream. The influx was stopped by circulating LN₂ in 25 freezing probes. Later, a concrete wall was constructed between the polluted area and the stream.

Valk (1979) discussed the differences between using brine and LN₂ for cooling in artificial ground freezing. LN₂ can produce a much lower temperature in a very short period. Therefore, LN₂ should be used in emergencies. Also, the fast freezing of soil by LN₂ will not result in transport of chemicals, as the soil water (with contaminants) will freeze in situ. Brine freezing, on the other hand, has the advantage of freezing the soil walls in a more regular shape, allowing water to move (dewatering); the temperature can get as low as -67°F. The cost of LN₂ is higher than brine. Figure 1 shows a schematic representation of the two freezing methods.

The use of LN₂ for ground freezing has increased substantially the number of artificial freezing projects during the last two decades. Also, artificial ground freezing is now used for underground storage of natural gas and bridge supports in permafrost areas. Many case histories have been cited in the proceedings of the three symposia on ground freezing held in West Germany in 1978, in Norway 1980 and at CRREL in 1982.

ECONOMIC ANALYSIS

The economics of ground freezing as a means of hazardous waste containment has been discussed by Sullivan et al. (1984). Briefly, the cost analyses are based on existing construction practices and proven freezing technologies. Prior to the economic evaluation, the technical feasibility of the containment alternatives must be studied, based on the nature and

extent of contamination (EPA 1982). Selection of the containment mode requires site-specific information about geology, soil and groundwater.

The required knowledge of the characteristics of the pollutant include its quantity, toxicity, solubility and other chemical properties. Environmental information about precipitation and temperature is also required. The surface and subsurface characteristics play a major role in the transport of the pollutant and affect the feasibility of cleanup alternatives. Soil texture, permeability and moisture content are primary soil parameters, and the key groundwater variables are the depth to groundwater, the depth to bedrock, and the direction and rate of flow. These site-specific properties are critical to the selection of a treatment process.

The ground freezing containment option uses the above information, and it requires no additional or unique knowledge about the site. Thermal conductivity, specific heat capacity and latent heat can be estimated from the site values of soil texture, permeability and moisture content. For example, Lunardini (1981) provides thermal property values for an extensive range of soils with various moisture contents. Consequently, the costs of the site-specific investigation to collect data are unchanged from the EPA estimate of \$20,000-\$80,000 for conventional cleanup (EPA 1982).

Artificial ground freezing is usually done using the Poetsch method (ASHRAE 1982). Essentially, vertical drill holes with standard steel casings are uniformly spaced along the desired freeze line. Common bore diameters range from 3 National Pipe Standard (NPS) to 6 NPS. Standard black pipes, one-half the bore diameter, are inserted into each drill hole, forming concentric cylinders. A header or manifold system provides coolant to the interior pipe, with the return line being the outer casing. The manifold is insulated and is connected to the freeze pipes with insulated pipes to reduce thermal losses. A self-contained refrigeration system pumps coolant around the freeze loop. Temperature-measuring instrumentation is appropriately placed for monitoring the freeze-front progress.

The equipment that must be brought to the site includes the drilling rig, refrigeration unit, piping and perhaps some form of excavation equipment. Ground freezing, as opposed to a slurry wall construction, need only disturb the environment sufficiently to permit the drill rig access to the freeze hole locations. Removal of large trees is not required, provided their root systems do not allow water to flow through the frozen wall.

Table 2. Unit costs for ground freezing equipment and supplies.

	Daily output	Total costs (\$)			
Mobilization (Means 1982)		100/unit			
Dozer, drill rig, refrigeration unit					
Over 100 miles add		1/mile per unit			
Clear wooded lot (trees less than 10- in. dia.) (Means 1982)	0.7 acre	2450/acre			
Grub stumps and remove	1.5 acre	1100/acre			
Dozer, medium duty clearing	3,000 yd ²	0.31/yd ²			
Header pipe system (Means 1982)		Months 1	2	3	
70 gal./min., 3 in.		1.40	0.85	0.65	
150 gal./min., 4 in.		Rent per linear foot	1.60	0.90	0.70
400 gal./min., 6 in.		2.50	1.00	0.75	
Well hole drilling (King and Moselle 1984)	100 vertical linear ft				
4 in. i.d. steel casing		9/linear ft			
5 in. i.d. steel casing		12/linear ft			
6 in. i.d. steel casing		15/linear ft			
Drive shoe		75 ea.			
Black steel pipe* (King and Moselle 1984)	2 in. dia.	0.22/linear ft per mo.			
	3 in. dia.	0.36/linear ft per mo.			
Self-contained refrigeration units**					
7 ton refrigeration		150/day			
110 ton refrigeration		2000/week			
Liquid N ₂ †		0.85/1000 ft ³			
Oil 147.4x10 ³ Btu/gal. (Babcock and Wilcox 1979)		0.80/gal			

(All prices include parts, labor, operating and profit for subcontractor unless otherwise noted).

* 5.20/linear ft 2 in. pipe - rent at 2 year writeoff 5.20/24 = 0.22/linear ft per month.
\$8.66/linear ft 3 in. pipe - rent at 2 year writeoff 8.66/24 = 0.36/linear ft per month.

** Personal communication with the GeoFreeze Subsurface Construction Co., 22 February 1984.

† Personal communication with the Merriam Graves Corp., 23 February 1984.

Consequently, clearing and grubbing expenses are minimal. Table 2 lists unit costs for most of the equipment required for ground freezing.

The time constraint for the frozen wall plays a primary role in the cost estimate. Mechanical refrigeration units rated at 5 to 110 tons are readily available.* These units will provide the manifold system with reuseable coolant at -4°F . Expendable CO_2 and liquid N_2 are available in large quantities when a rapid freeze front is required. Expendable coolants are a reasonable expense for situations with severe time constraints.

Sanger and Sayles (1979) provide a sound methodology for thermal computations in frozen ground. Their energy requirements and freeze time estimates are somewhat more conservative than those predicted using finite element simulations and field measurements (Frivik 1980, Frivik and Thorbergesen 1980). However, for economic analysis their predictions are applicable. Sanger and Sayles predict the expenditure of energy based on reasonable assumptions of the heat-governing equation.

For waste containment the function of the frozen ground is not primarily structural. Therefore, the frozen thickness need not be the standard 3 ft used for an impermeable slurry wall (EPA 1982). Actually, the 3-ft design thickness of the slurry wall is a limitation of the trench excavation equipment and not a function of structural support. Initially, we might expect an economic advantage for a thin-wall construction using many cylinders of small radii. However, the final cost analysis shows large-radii cylinders as the most economical because of the reduced number of drill holes required. In addition to the economic gains, the large wall thickness design has a greater resistance to permeability. After the frozen wall is formed, a reduced refrigeration load maintains the wall.

EXAMPLE CASES

The EPA uses a hypothetical example where a 10-acre hazardous waste site is located 150 miles from the drilling and refrigeration contractors. Remedial Action at Waste Disposal Sites (EPA 1982) recommends a slurry wall 1000 ft long and 3 ft wide to be placed down to bedrock on the up-gradient side of the site that is to be protected. The bedrock averages 40 ft deep. Table 3 summarizes EPA slurry wall estimates vs ground freezing for

* Personal communication with Geofreeze Subsurface Construction Co., 22 February 1984.

Table 3. Slurry wall (EPA 1982) and frozen ground construction estimates.

	Unit costs*	Total costs
<u>Slurry wall</u>		
Testing - geotechnical, hydrologic and lab filter cake permeability	N.A.	\$20,000-\$80,000
Equipment mobilization - hydraulic backhoe, bulldozer, slurry mixed, etc.	N.A.	\$20,000-\$80,000
Slurry trenching, excavation mixing and backfilling	\$45-\$70/yd ²	\$200,000-\$310,000
Maintenance	-	-
Overall	N.A.	\$240,000-\$470,000
Average		\$355,000
<u>Artificial ground freezing†</u>		
Testing - geotechnical, hydrologic, and lab filter cake permeability	N.A.	\$20,000-\$80,000
*Equipment mobilization, clear, 4-in. drill casing	\$22.5/yd ²	\$100,000
*Rent - refrigeration, 4-in. header 2-in. pipes, manpower	\$ 5.3/yd ²	\$25,000
*Energy consumption	\$10/yd ²	\$45,000
Maintenance	0.34/yd ² per day	\$3000/day
Extra melt buffer because of latent heat		25 days
Overall**	Maintenance +	\$190,000-\$250,000
Average	Maintenance +	\$220,000

* See Table 2 for unit costs.

** 15 days with 232 drill holes.

† Containment time is less than 70 days.

saturated coarse quartz sand initially at 45°F. It can be seen from Table 3 that artificial ground freezing is an acceptable solution provided the containment time requirement is small (less than 70 days). Thereafter, the daily maintenance costs make the ground freezing alternative unattractive as a permanent containment method.

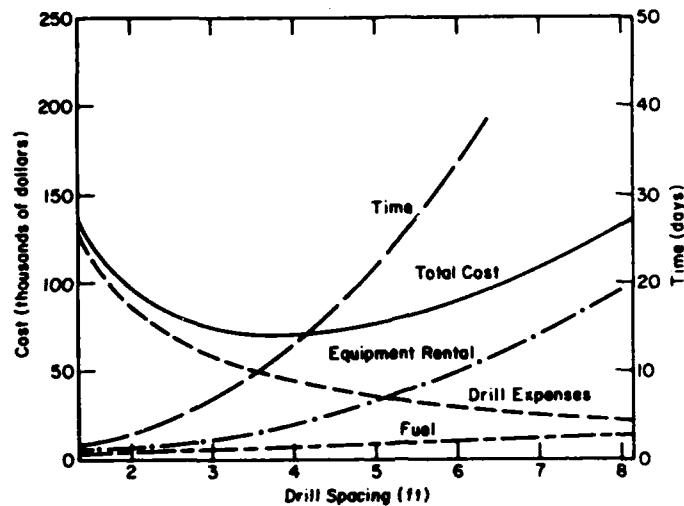


Figure 2. Sample economic overview.

Examining Figure 2, which is an example economic overview, we can see that as the drill spacing becomes tighter the fuel costs, equipment rental and time for wall completion are reduced. A tight drill spacing yields small frozen soil-column radii. This reduces the overall energy requirement and permits use of less expensive refrigeration equipment. The drawback of the close drill spacing is the expense associated with the drilling itself. The lineal footage of piping, a drive shoe for each well drilled and the labor charge per vertical foot drilled overwhelm all other economic parameters.

This example shows artificial ground freezing to be a cost-effective means for constructing an impermeable barrier. However, the example is somewhat unrealistic. A waste site in a rural area will score low on the National Priority List (NPL) as compared to that in an urban environment. Slurry wall estimates in urban environments increase 50% over that presented in Table 3 (Spooner et al. 1982). Ground freezing, on the other hand, is ideally suited to this situation. Urban refrigeration units can tap directly into the power lines, eliminating the need for diesel generators. These mechanical refrigeration units or the expendable coolant systems minimize public disturbance by being noiseless and pollution-free. Frozen soil columns are readily created under buildings and around obstacles without the need of major excavation or construction. Finally, after treatment of the confined area the frozen wall returns to its natural state.

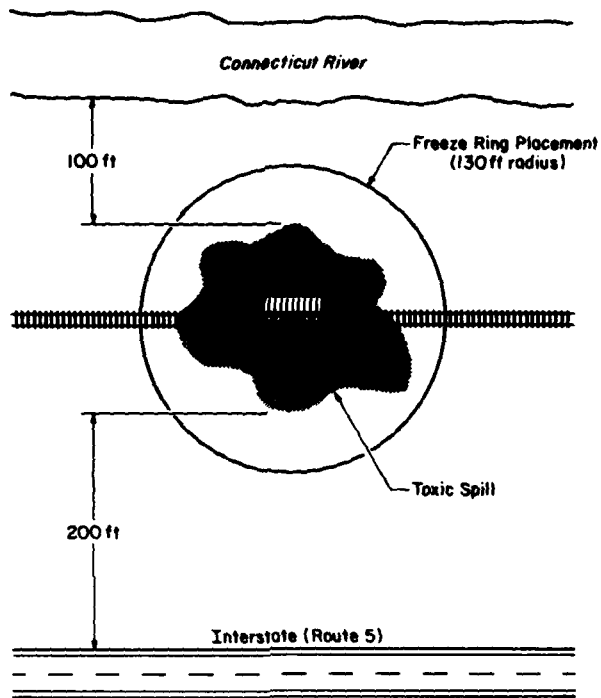


Figure 3. Chemical car pollutes area.

As a second example, consider the situation where a derailed chemical car disperses a toxic substance over an area surrounding the track (Fig. 3). Surrounding towns impose a 30-day containment constraint on the chemical company. A week of preliminary testing is required to define the hazardous spill and obtain general site test results. Initial drill samples estimate the required barrier depth at 15 ft. Under the assumption that the pollutant diffuses horizontally 1 ft/day, the frozen wall is planned at a radius of 130 ft. This information was used to generate the economic overview presented in Figure 4. The optimum cost design calls for

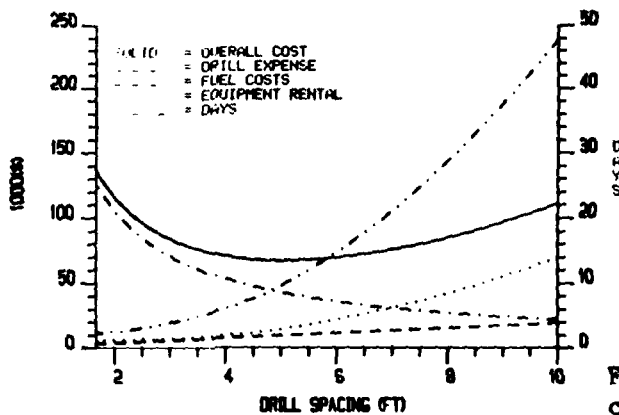
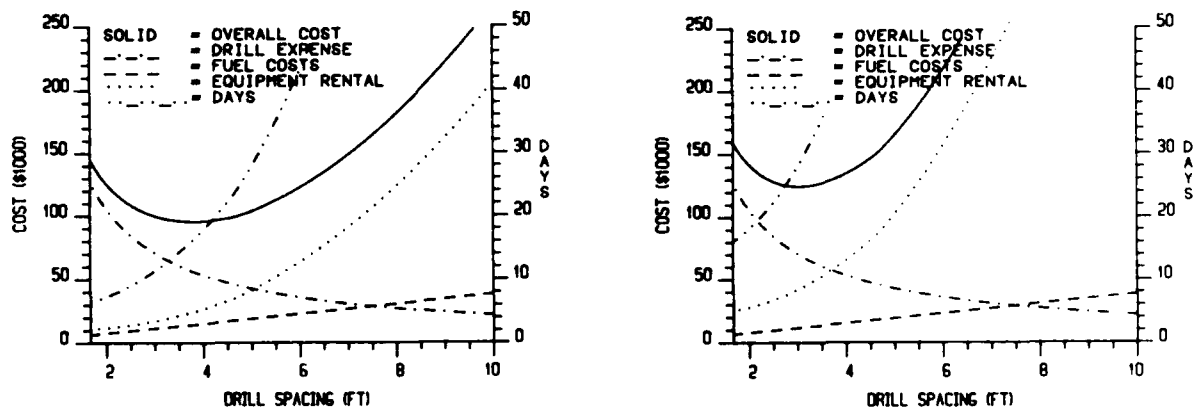
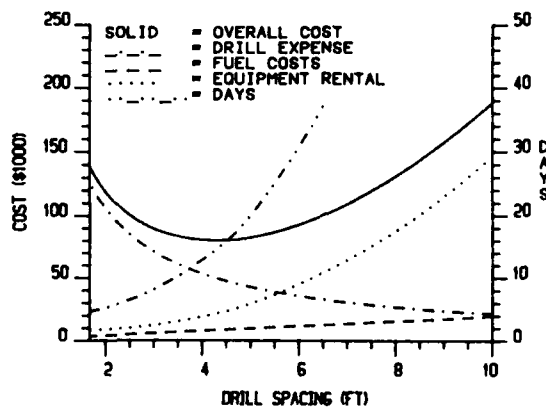


Figure 4. Economic overview for train car spill.



a. Coarse grain, high moisture soils. b. Fine grain, high moisture soils.



c. Fine grain, low moisture soils.

Figure 5. Economic overview for various soils.

a 4.5-ft drill spacing. However, by the time the perimeter is drilled and the header system installed, there would be insufficient time remaining to freeze the soil before the 30-day limit is reached. Reducing the drill spacing to 4-ft saves 3 days in freezing time with a small increase in overall costs.

The thermal properties used in the examples are those determined for saturated quartz sand (O'Neill 1983). The following cases show the dependence of costs and time on thermal parameters. Using data from Lunardini (1981) for saturated soils, we examined the full range of soil texture and moisture content effects. Figure 5 shows the economic overview of each soil system. Increasing the soil moisture content increases the time required to establish a frozen wall. For these high-moisture soils, mechanical refrigeration would fail the 30-day time constraint. However, an expendable LN₂ system with a 3-ft drill spacing can establish an imper-

meable barrier within 10 days. This compares to the 29-day refrigeration time for a mechanical system under the same conditions. Once the LN₂ system establishes the barrier, a mechanical refrigeration unit can maintain it during the waste treatment process.

In summary, ground freezing as a means of hazardous waste containment is cost-effective. The system is limited to temporary treatment sites because of the maintenance expense. The ground-freezing alternative becomes highly attractive in cold environments where slurry wall construction becomes difficult and in residential areas where low noise and minimal environmental disturbances are a high priority.

CONCLUSIONS

1. Freezing and thawing can be harmful to hazardous waste sites. Freezing and thawing of fine-textured soils, such as clay liners used for capping hazardous waste sites, may increase their permeability because of the increase in soil aggregates and the formation of vertical cracks. If this happens, rainwater will penetrate through the liners, reaching the waste and dissolving chemicals that may then reach groundwater. This is most likely in the northern portions of the U.S., where there is natural freezing and thawing for several months. Also, the effect of frost heaving on the integrity of clay liners used for capping must be assessed. Frost heaving can be a big problem. There is evidence in the literature to show that buried chemical wastes can move upward as a result of repeated frost effects if these materials are within the frost depth.

2. The literature shows that ground freezing can be used to immobilize contaminants in soils so that they don't make their way into water supplies. This process can be used temporarily for immediate action and can be used to consolidate soils, sediment and sludge to facilitate transportation and removal. Rapid freezing is preferred for immobilizing contaminants and preventing ion transport during the freezing process. Slow freezing, on the other hand, is more appropriate for dewatering and consolidation.

3. The use of natural and artificial freezing for desalination of seawater and for purification of wastewater has been extensively studied since the 1930's. Lagoons as well as treatment plants have been used. Sludge dewatering and preconditioning treatment by freezing and thawing,

and sometimes by adding chemicals such as alum or iron compounds as stabilizers to speed the process, have also been and are still being investigated. Currently, a few systems such as that in Winnipeg, Manitoba, are in operation. The cost of natural freezing was found to compare well with other methods of sludge treatment.

4. Ground freezing has also been used for over 100 years in tunnel construction and mining. In addition, it is used for bridge supports in permafrost areas and for storing natural gas under the ground.

5. Ground freezing as a means of hazardous waste containment is cost-effective. However, it is limited to temporary treatment because of the maintenance expense. The ground freezing alternative becomes highly attractive in cold environments where slurry wall construction is difficult and in residential areas where low noise and minimal environmental disturbances are a high priority. The literature indicates that all hardware needed for artificial ground freezing is available. In the United States there are three commercial firms equipped to do such work.

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