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RESEARCH ARTICLE

Diesel spills under stilted buildings in Canadian Arctic villages: what is the best remediation method?

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Abstract

In remote communities in the Canadian Arctic, petroleum hydrocarbons supply most household energy needs. Their transportation and use frequently incurs small volume spills in populated areas. The remediation method that is currently used when such spills affect the soil under northern villages' stilted buildings is expensive and not well suited to local conditions. Here, we review local constraints and environmental considerations and select the best remediation technology for this context: in situ chemical oxidation, involving sodium persulfate (SPS) alkali activated with calcium peroxide (CP). Activated SPS presents a good reactivity and amenability to compounds found in diesel. Its high persistence allows a gradual contaminant degradation, regulating heat release from exothermic reactions associated with the oxidative reactions. CP provides suitable alkali activation, acts itself as an oxidant and provides O_2 into the subsurface, which may favour a final smoothing bioremediation step. The SPS properties and the contaminant amenability mean that diesel is removed relatively efficiently, while the subsurface temperature increase is limited, thus preserving the residual permafrost. The solid form of the chemicals offers safe and economic transportation and operation, along with versatility regarding the preparation and distribution of the oxidizing solution into the subsurface. Finally, the oxidation by-products resulting from this method are not considered to be environmentally problematic in the context of the application, and they can be partly confined during the treatment.

Keywords
In situ chemical oxidation, ISCO, Nunavik, sodium persulfate, permafrost, hydrocarbon contamination

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Abbreviations
CAD: Canadian dollars
CP: calcium peroxide
ISCO: In Situ Chemical Oxidation
KMHB: Kativik Municipal Housing Bureau
KRG: Kativik Regional Government
OM: organic matter
PAHs: polycyclic aromatic hydrocarbons
PHCs: petroleum hydrocarbons
SPS: sodium persulfate

To access the supplementary material, please visit the article landing page

Introduction

At the global scale, there are a considerable number of sites where ground or groundwater is affected by organic contamination, especially by Hydrophobic Organic Compounds. Contaminated sites represent an environmental debt and a potential threat to public health, and they are a major issue for municipalities and land developers in all industrialized and developing countries, as well as for the various ministries and responsible agencies (Laperche et al. 2004; Lehr 2004; Xiong et al. 2016). In Canada, Smith & Vaughan (2017) specified that managing contaminated sites under federal jurisdiction represented an average annual cost of about 283 million CAD for the years 2005–06 and 2014–15. In 2013, the total remaining liability for remediating contaminated sites reported in the public account was 4.9 billion CAD (Story & Yalkin 2014). In the circumpolar Arctic region, the contaminated sites issue is well known and has been declared a serious problem by the governments of the USA, Canada and Russia (Filler et al. 2006). Locally, the rapid economic development around mining and petroleum exploration and exploitation as well as military and industrial activities has led to an increase in the number of inhabitants and in energy demand and consumption (Poland et al. 2003; Camenzuli & Freidman 2015). In those remote regions, the most convenient source of energy is diesel fuel, which is used as heating oil in most housing units and fuel for vehicles and municipal electricity power plants. The lack of financial resources, logistical options and expertise,

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together with difficult climatic conditions, lead to a vast number of small- to medium-sized diesel spills (Chuvilin et al. 2000; Yang et al. 2009; Camenzuli & Freidman 2015; Taillard & Baïlon-Poujol 2020) that occur on a regular basis in the heart of inhabited areas.

The cold region context implies specific limitations regarding environmental impacts, contaminant propagation and natural attenuation. Freezing temperatures during most of the year make ecosystem recovery slower than in warmer regions, and any damage or alterations will take longer to recover (Chuvilin et al. 2000; Poland et al. 2003; Filler et al. 2006; Filler et al. 2008; Camenzuli & Freidman 2015). When diesel spills affect the raft soil located under stilted buildings, the current remediation strategy consists in moving the building, excavating the contaminated raft soil and transporting it to the southern part of the country in order to be treated or landfilled. This management option implies high operational costs, the relocation of inhabitants and the production of large amount of greenhouse gas, and it requires an excavation step that can equal or exceed the damage caused by the initial spill (Filler et al. 2006).

Considering the context of the 14 Canadian circumpolar Northern villages in Nunavik, the northern part of the province of Québec, this study reviews different factors associated with the climate, the presence of permafrost and surficial materials. The reality of a Northern village in terms of logistics, expertise, infrastructure and accessibility are also important parameters to consider. Based on those factors, this study aims at selecting the most suitable and efficient remediation method to be implemented at a commercial scale in Nunavik in order to remediate diesel-contaminated soil located under stilted buildings.

Arctic region

Climate overview and limitations

Although polar zone climates can vary considerably depending on their latitude and geographical position, the Arctic climate is commonly divided in two different zones: a maritime zone reflecting the ice-cap climate and a continental tundra climate. The maritime zone covers the Pacific and Atlantic Ocean coasts (Alaska, Canada, Iceland, and northern Russia and Scandinavia) and has characteristically relatively high snowfall and rainfall (60 cm/year to 125 cm/year in water height equivalent), no extreme cold temperature and an annual winter temperature average ranging −10 to −30 °C (NSIDC 2020). The continental zone, covering the northern parts of Canada, Siberia and Alaska, has light snowfall and extremely cold winters, exemplified by mean temperatures between −29 and −34 °C in North America (NSIDC 2020). In both zones, the thawing season (air temperature >0 °C) is short and is the only period of the year suitable for construction work and contaminated site remediation operations. During winter, cold temperatures, heavy wind, snow and a short daylight period make every operation very difficult and costly or just impossible (Poland et al. 2003; Filler et al. 2008). In the Canadian Arctic, most of the local communities are in coastal areas, in the maritime zone. For instance, Inukjuak (58°27.3546′N; 78°7.0894′W) and Puvirnituq (60°2.1369′N; 77°16.5745′W) are two settlements north of the 55th parallel, in the Subarctic region of Nunavik, in the province of Québec, Canada. In these Northern villages, the duration of the thawing season varies between 125 and 142 days (Charron 2015), the average annual temperature does not exceed −5 °C, the average winter temperature is about −16 °C and the mean summer temperature can reach more than 7 °C.

The cold can impact the contaminant availability and migration into the subsurface by affecting PHC multiphase flow and partition in water, soil and air. Amongst other factors, the evaporation rate of light organic compounds decreases, overall PHC viscosity increases and water solubility decreases. The migration of pollutants deeper into the ground is slowed down, and its adsorption onto solid surfaces is reduced (Chuvilin et al. 2001; Balks et al. 2002; Filler et al. 2006). Finally, remediation options are very limited where there is permafrost.

Permafrost

Permafrost is soil frozen for at least two consecutive years and is found at high latitudes and altitudes. It is distinct from glaciers and ice sheets as it is not due to an accumulation of compacted snow but is formed when the average annual air temperature remains below zero for a sufficient amount of time. Permafrost covers between 12 and 18% of the exposed land in the Northern Hemisphere (IPA 2020) and can reach a depth of more than 600 m. So-called continuous permafrost (100% frozen ground) is formed when the ground temperature is lower than −6 °C, whereas in the temperature range of −6 to −1 °C, permafrost is discontinuous (50 to 90% frozen ground) or sporadic (50% frozen ground or less) (Ritter et al. 2002; Anderson & Anderson 2010). Continuous permafrost is an effective aquitard, acting as an impermeable layer in the porous medium (Torrance 2016). During the summer period, when temperature rises above 0 °C, only the top layer of soil thaws to a depth generally ranging from 0.5 m to 3 m. This zone is called the active layer, and it is a key factor for Arctic hydrology since it holds all the flowing groundwater that is essential for plants and animals and is actively participating in geomorphic activity (Brown et al. 2000).

Over recent decades, rising temperatures and other aspects of climate change have had a marked impact on the Arctic and Antarctic (McBean et al. 2005; Anisimov et al. 2007). The thawing permafrost in the Canadian and Russian Arctic is considered problematic as it releases greenhouse gases, such as methane, that have been trapped in the subsurface for thousands of years (Shakhova et al. 2015; Anthony et al. 2016). It also affects such constructions as airport runways and buildings, since the increasing depth of the active layer leads to soil movements due to annual freeze and thaw cycles (Allard & Lemay 2012; L'Hérault et al. 2014; Aubé-Michaud et al. 2017; IPA 2020). Siliciano et al. (2008) have also pointed out that the presence of PHCs can modify the active layer properties since they can influence the water content in frozen soil, impacting ice seams, lenses, pods and layer formation. The complexity of soil movements and the presence of continuous, discontinuous or sporadic permafrost make it very complex to simulate and to predict PHC migration (Bjella et al. 2018; Wagner & Barker 2019).

Permafrost preservation is essential to limit greenhouse gas emissions and to preserve the current infrastructure. It is an important factor to consider when planning new development projects or implementing new remediation technologies that can significantly affect the subsurface.

Surficial materials

In the inland regions of the Arctic, till is the predominant surficial material, along with glaciofluvial deposits, complex colluvial sand and glaciolacustrine sand. The thin layer of topsoil supports tundra vegetation, including shrubs, sedges, grasses, mosses and lichens. In coastal areas, apart from glaciers, surficial materials are mainly composed of marine and lacustrine mud and sand as well as glaciomarine coarse grain material (Fulton 1995; Dredge et al. 1999; Harrison et al. 2011; Aubé-Michaud et al. 2017). In the Canadian Arctic, the village of Inukjuak, on the shore of Hudson Bay (Québec), lies on a terrazzo base gneissic of Archean age (Harrison et al. 2011; Aubé-Michaud et al. 2017). The active layer consists of fine-grained deposits (<2 m) made of coastal and pre-coastal sediments (greyish-brown sand with gravel and 4 to 30% silt) resting on the bedrock or on a layer of sand and thick gravel. It can contain a lot of segregating ice: between 30 and 100% (L'Hérault et al. 2014; Carbonneau et al. 2015).

Northern communities, buildings and infrastructure

The circumpolar Arctic region has about 4 million inhabitants, many of whom live in small settlements. Most are in Siberia, northern Europe, Alaska and Iceland, and the great majority are non-indigenous. This contrasts with Canada and Greenland, with a total Northern population of 130 000 and 58 000 inhabitants, respectively, a majority of whom are indigenous (Haavisto et al. 2016; NSIDC 2020). Over recent decades, Arctic and Subarctic populations in northern Europe and North America have been growing fast in response to economic development based on mineral and petroleum exploration and exploitation (Filler et al. 2008; Camenzuli & Freidman 2015). Canadian Arctic and Subarctic regions have shown population rises ranging 9 to 41% since 2001 due to higher fertility and the younger average age of local inhabitants (Haavisto et al. 2016; NSIDC 2020).

In Nunavik, 90% of the population is housed through the KMHB, which is responsible for acquiring, administering and maintaining 3900 residential buildings (Taillard & Baïlon-Poujol 2020). Housing units are all built according to standards dictated by a construction guide produced by the Government of Québec (2017). Those standards have been created based on a worldwide expertise in order to implement concepts and principles of sustainable development in a cold region context.

The buildings stand on metallic, wooden or concrete structures ranging 0.3 m to 1 m high. These structures lie on a layer of coarse sand and gravel, ranging 0.3 m to several metres in thickness, called raft foundation, which satisfies specific geotechnical requirements (Government of Québec 2017; Fig. 1). The raft foundation is made of material extracted from local pits located around the villages (Table 1). This specific building foundation arrangement leaves a free volume of cold air circulating below the building to prevent the heat produced by the houses from accelerating the thawing of the frozen surficial raft soil and the permafrost below.

Each housing unit has its own diesel storage unit and its own boiler room, producing heat and hot water. Diesel tanks have an average capacity ranging 1000 L to 2500 L and are elevated to a height allowing the fuel to flow from the tank to the boiler by gravity. Each tank stands on a support, which is attached to the building, built as an independent infrastructure fixed to the building, or just set in place close to it. Thus, large volumes of diesel are permanently stored in small tanks spread all over the Northern villages. Limited budgets and resources lead to a lack of maintenance and eventually to the collapse of tank structure, pipe failure and leakage, or boiler leakage due to the presence of rust and material degradation. The limited resources available are insufficient for the proper training of people in charge of refuelling diesel trucks and tanks and those in charge of detecting leakage. Accidents are frequent (Taillard & Baïlon-Poujol 2020).

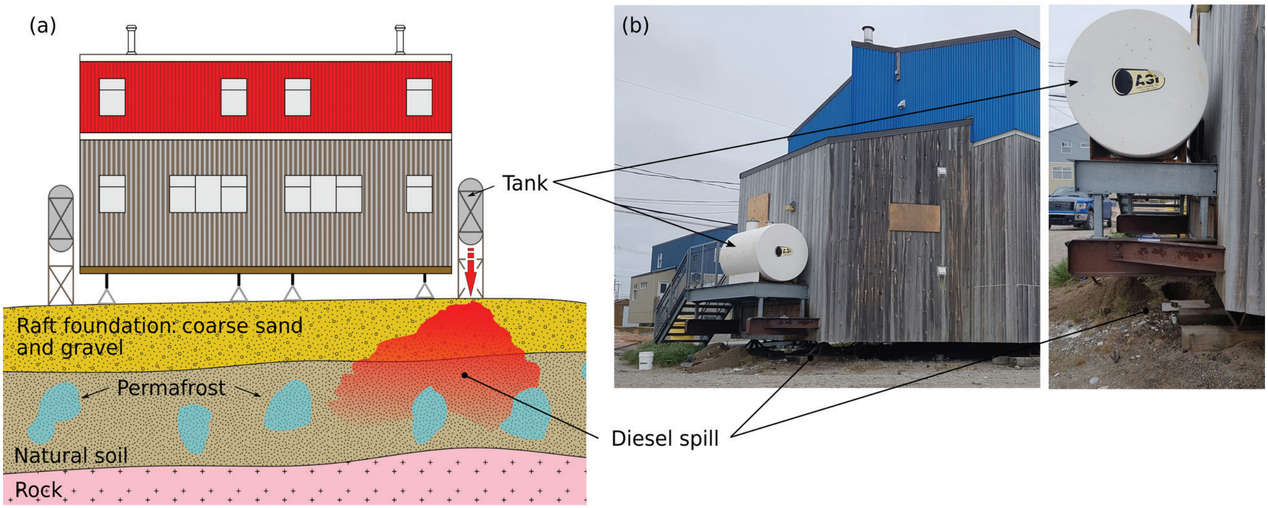

Fig. 1 (a) Conceptual figure of a diesel spill under a stilted building; and (b) photographs of a diesel spill under a stilted building (Puvirnituq, QC, Canada).

Table 1 Raft material parameters.

Parameter	Value	Implication
Origin	Local pits, no stones >0.30 m diameter	Homogeneous reworked material
Geochemistry	Quartz sand and plagiosilicates	Water wet soil
Particle size	Fine to coarse sand, gravels	High hydraulic conductivity, no fine particles
Organic matter	As low as possible to ensure proper drainage and to avoid soil movement	Neither vegetal cover nor organic matter in porous media

The volumes of reported PHC spills in the 14 Northern villages in Nunavik (with a total of about 13 000 inhabitants) varied between 4930 L spread over 10 spills in 2016 to 37 071 L spread over 37 spills in 2015 (Table 2). Diesel was involved in more than 90% of the cases. When a diesel spill occurs, it generally spreads around and below the building, contaminating the raft foundation material and, depending on the depth of the raft, the natural soil beneath.

According to Taillard & Baïlon-Poujol (2020) and the KMHB (pers. comm. 2018), a diesel spill under a building requires the following steps: (1) declaration of the spill to the authorities; (2) relocation of the residents to an available housing unit; (3) moving of the building; (4) excavation of the contaminated soil; (5) backfilling with clean material; (6) moving the building back in position.

In the vast majority of the registered incidents, diesel concentration ranges between 2500 mg/kg and 15 000 mg/kg total PHCs (KMHB pers. comm. 2018). In most cases, the excavated contaminated soil is transported by boat to authorized management or land-filling facilities located south of the province. This management option is well known, and it does not need high expertise or specialized equipment. Nonetheless, transporting the contaminated soil over long distances entails greenhouse gas emissions and prohibitive costs; the environmental consequences of excavation, which can equal or exceed the damage caused by the initial spill, must also be recognized (Filler et al. 2006). The intervention delay can reach several years given the high number of spills, the short operational period, the lack of resources and manpower, the scarcity of clean raft soil and the scarcity of free units in which to temporarily house tenants during operations. During the maturation period induced by the intervention delay, light and volatile compounds found in diesel will gradually evaporate and be degraded by microorganisms. Consequently, heavy and recalcitrant hydrocarbons (longer carbon chains, PAHs with higher number of aromatic cycles) become more concentrated in the remaining mature diesel. Once remediation steps are scheduled, it is necessary to call on companies from the south who provide cranes, crane operators and engineers to safely move the building. The overall costs can exceed 500 000 CAD for a 2000 L diesel spill affecting the raft soil of a housing unit of an average size (KMHB, pers. comm. 2018).

Remediation technologies overview

Available remediation methods for inorganic or mixed organic and inorganic contaminants are commonly divided in three categories, corresponding to three

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Table 2 All registered PHC and diesel spills in the 14 Northern villages in Nunavik in the period 2014 to 2017 (KRG, pers. comm. 2018).

Contaminant	2014		2015		2016		2017	
	Vol. (L)	Number	Vol. (L)	Number	Vol. (L)	Number	Vol. (L)	Number
PHCs	11 524	17	37 071	37	4930	10	6668	22
Diesel	10 604	12	37 002	34	4505	5	6651	16
% of PHCs spills involving diesel	92		100		91		100	

different approaches regarding contaminant management: immobilization, extraction or degradation. Amongst those remediation's options, some can be applied in situ: no excavation step is needed, and the technology is delivered directly into the ground. Others require ex situ operation: the soil is excavated and either moved to an existing treatment facility or treated on site via a mobile unit (USEPA 1993; Lehr 2004; Dermont et al. 2008; De Sousa 2008; USEPA 2006a; Lim et al. 2016; Khalid et al. 2017; FRTR 2020).

Online tools provide a screening matrix that helps to select the appropriate remediation option (FRTR 2020). They give useful information regarding state of development of the technology, cost, performance, availability and relative effectiveness for specific contaminants. Nonetheless, in the context of remote, cold regions, most of the recommendations are incomplete since remediation technologies have to face numerous additional limitations, as mentioned earlier.

Bioremediation techniques

In recent years, the most investigated and tested technologies for implementation in cold regions have been biological processes, alone or assisted. These options take advantage of the activity of specific bacteria to remediate PHCs and can involve in situ bioremediation and on site landfarming or biopiles. Bioremediation technologies are sensitive to parameters such as temperature, availability of microorganisms and the disposability, concentrations and toxicity of targeted contaminants (USEPA 2006b; Siegrist et al. 2011). Thus, the degradation of PHCs to an acceptable level might take several years (Filler et al. 2006) and may not be efficient for aromatic compounds (Van Hamme et al. 2003).

When implemented in situ, bioremediation technologies have shown some promising results in cold regions if applied with bio-stimulation or bio-augmentation steps (Filler et al. 2008; Rayu et al. 2012). Landfarming and biopile technologies are also based on microbial activity to degrade contaminants, although they require an excavation step and the construction of an on site operating deck, which might constitute a considerable logistical or environmental problem. Furthermore, excavating the soil in remote areas outside Northern communities can lead to landscape and vegetation destruction, and permafrost thawing, generating soil shrinkage and groundwater flow modification (Filler et al. 2006; Camenzuli & Freidman 2015).

Camenzuli & Freidman (2015) have identified phytoremediation as an appropriate solution in cold regions. This method can be defined as a plant-assisted bioremediation as enzymes generated by the plant's roots mobilize or degrade contaminants (Lim et al. 2016). Whereas this technology is simple to implement and has low operating costs, it requires long-term monitoring, ranging two to 20 years (Lewis et al. 2013). This can be problematic, considering that a quicker remediation occurs, lessening the economic and the environmental impacts resulting from the spill (Yang et al. 2009). Moreover, removal efficiency depends on various parameters affecting the growing mechanisms of plants, which are not favourable in high latitudes and cold regions.

Biological options might be appropriate to treat large volumes of contaminated soil in remote sites alongside natural resource exploration settlements, for example. In such conditions, remediation delays are not a health issue for inhabitants, and treated soil can be set back in place, limiting its transportation.

Physico-chemical methods

In situ physico-chemical options based on mass transfer to collect PHC contaminants (multiphase extraction, pump and treat, soil flushing) generally require the use of large volumes of solution or gas because of low PHC solubility. Surfactants can be added into an aqueous solution to increase PHC solubility, but the volume of solution needed is still considerable, and it would accelerate permafrost thaw as heat is transferred from the solution to the ground. There are, moreover, environmental risks with using large quantities of surfactants. Finally, these technologies generally require expertise, adapted injection wells, complex drilling operations and the presence of a mobile or fixed facility near the contaminated site hosting the equipment involved (USEPA 2006a).

On site physico-chemical options are poorly suited to remote regions since they require the displacement of the housing unit, the excavation of the contaminated soil and the transportation of a complex and costly mobile facility on site. Their only advantage over the land-filling option is that the treated soil can be set back in place at the end of the operations. Remediation technologies involving heat production and transfer, in situ and on site, are definitely not recommended in a cold region context due to their assumed impact on permafrost, the high expertise and knowledge necessary to implement and operate the technology as well as the high levels of risks for the working team and the environment.

ISCO

ISCO does not require excavating the contaminated soil, and it is efficient on sorbed and dissolved contaminants that are found in groundwater, saturated soil and unsaturated soil. Treatment can be relatively fast in comparison to bioremediation options, and operation costs are reasonable in comparison to physico-chemical treatment. Contaminants are degraded during the treatment, generating no disposal expense. Depending on the logistics available and the chemical selected, various distribution methods can be considered, such as adapted injection wells (pressure injection), direct push wells or surface percolation. Once implemented, it requires relatively low expertise and logistics to operate, depending on the distribution method selected (USEPA 2006b; Siegrist et al. 2011).

Although ISCO can be very efficient, the removal rate and remediation times are very dependent on the capacity to ensure a proper contact between oxidant and contaminant. The presence of natural OM, non-aqueous phase liquid, multiple layers or heterogeneous soil, low hydraulic conductivity or the presence of subsurface infrastructure (such as buried lines) represent significant issues. The context of a small diesel spill affecting raft soil appears particularly suitable to ISCO since the raft is made of reworked and homogeneous soil composed mostly of fine and coarse silicates and quartz sand with high hydraulic conductivity and low OM content (Table 1). Low spill volumes, combined with an average diesel concentration of less than 15 000 mg/kg total PHCs, do not favour the presence of free non-aqueous phase liquid phase when a spill occurs.

Efforts must be made to identify a suitable ISCO process. Indeed, depending on the reagents involved, oxidation by-products might present a risk for the local environment or might modify the underground characteristics by clogging, for instance. In addition, depending on the oxidant and contaminant type and importance, ISCO can significantly increase subsurface temperatures (USEPA 2006a; Siegrist et al. 2011; FRTR 2020), which is a major issue considering the projected context of application.

ISCO process selection

ISCO involves different types of oxidants and can be implemented on site by means of various distribution methods (Table 3). Regarding the contaminants' amenability to the oxidants, permanganate shows good oxidation performance for benzene, toluene, ethylbenzene, xylene and PAHs, and moderate performance for PHCs. Fenton's reagents as well as activated persulfate, ozone and peroxone present good to very good performance regarding the contaminants mentioned above (USEPA 2006b).

Oxidant selection

Considering oxidant parameters (Table 3), local limitations, consequences and impacts (Table 4), the use of hydrogen peroxide or ozone as oxidants is not recommended. These two species have low to very low persistence, and their use could require a large amount of injection wells, depending on the spill size, to ensure a proper contact between reagents. This is an important issue considering the limited logistical means and the necessity to operate under stilted buildings. Unactivated ozone is reactive (E_0 = 2.1 V), and as for hydrogen peroxide, when ozone is activated, the highly reactive hydroxyl radical (E_0 = 2.8 V) is involved in the oxidative process. Low persistence together with high reactivity implies a rapid and violent reaction with contaminants, releasing significant amounts of heat into the subsurface during treatment. Hydrogen peroxide is only available under liquid form, and its concentration cannot exceed 50% v/v to ensure safe transportation and operation, which increases its transportation costs. Ozone has the advantage that it can be synthesized on site, eliminating transportation costs, and it can be adapted to treat unsaturated contaminated soil. However, its on site production requires expertise, and any equipment failure can lead to severe skin, eye and respiratory injuries to operators and inhabitants.

Permanganate and persulfate can be purchased in a 100% solid salt form, which facilitates and reduces expenses related to transportation and storage (Lee et al. 2020). One of the advantages of permanganate over persulfate is its higher persistence, which can extend to more than three months, whereas the persistence of persulfate ranges from several days to several weeks, depending on

Table 3 Synthesis of oxidant parameters regarding reaction leading to the creation of reactive species, their potential and persistence (Watts & Teel 2006; USEPA 2006b; Siegrist et al. 2011; Ranc 2017).

Oxidant	Reactive species	Potential E0(V)	Activator	Advantages	Limitations
Permanganate	$MnO_4^- + 2H_2O + 3e^- \rightarrow MnO_{2(s)} + 4OH^-$	1.7 (permanganate ion)	None	High persistence, chemical reaction well known and documented, solid form, low cost	Low reactivity, MnO_2 production and clogging phenomena, non-suitable for aliphatic hydrocarbons, high NOD[a]
Hydrogen peroxide	$H_2O_2 + 2H^+ + 2e^- \rightarrow 2H_2O$	1.8 (hydrogen peroxide)	Fe^{2+}, Fe^{3+}, Fe^0, 0_3, heat, none	Multi-activation, high multi-radical reactivity, relatively low cost, O_2 production, well documented, no by-products, low cost	Very exothermic reaction, very low persistence, liquid form only, dangerous to manipulate and to operate, targeted injection necessary, vapours creation
	$2OH^{\cdot} + 2H^+ + 2e^- \rightarrow 2H_2O$	2.8 (hydroxyl radical)			
	$HO_2^{\cdot} + 3H^+ + 3e^- \rightarrow 2H_2O$	1.7 (perhydroxylradical)			
	$O_2^{\cdot -} + 4H^+ + 2e^- \rightarrow 2H_2O$	−2.4 (superoxide radical)			
	$HO_2^- + H_2O + 2e^- \rightarrow 3OH^-$	−0.88 (hyperoxide anion)			
Ozone and peroxone	$0_3 + 2H^+ + 2e^- \rightarrow H_2O + 0_2$	2.1 (ozone)	None/H_2O_2	On site production, multiphase reactions (efficient in unsaturated zones), highly reactive, O_2 production, no by-products	Low persistence, gas form, dangerous to synthesize and to operate, low solubility in water, specific injection wells necessary, relatively high operating costs
	$20_3 + 3H_2O_2 \rightarrow 40_2 + 2OH^{\cdot} + 2H_2O$	2.8 (hydroxyl radical)			
Persulfate	$S_2O_8^{2-} + 2e^- \rightarrow 2SO_4^{2-}$	2.1 (persulfate)	Fe^{2+}. H_2O_2, heat, alkaline (NaOH, CaO, CaO_2)	Multiple activation method, high persistence, good reactivity, solid form, SO_4^{2-} as an electron acceptor for a polishing step	Poorly documented, SO_4^{2-} production, depending on the activation method, clogging can occur, medium to high operating costs
	$SO_4^{\cdot -} + e^- \rightarrow SO_4^{2-}$	2.6 (sulphate radical)			

[a] Natural oxidant demand.

Table 4 Limitations associated with Arctic settlements, consequences for remediation operations and impact on oxidant/activator selection (Poland et al. 2003; Filler et al. 2006; Camenzuli & Freidman 2015; Taillard & Bailon-Poujol 2020).

Limitation	Consequences	Impact on oxidant/activator selection
Remote region	Limited and costly transportation options	Solid form recommended
	Expertise, local logistic and technical assistance very limited	Complex drilling operations and specific equipment not recommended
	Limited data regarding contaminant concentration and exact location	High persistence oxidant recommended, targeted injections is an important issue
	Fragile environment	By-products formation has to be considered
Cold climate region	Four months of operation; freezing cycle favours contaminants dispersion	Treatment has to be achieved in one or two years (four- to eight-month operation)
	Preservation of the permafrost	Heat activation is not relevant, limited temperature elevation is required
Northern communities: stilted building built on raft foundation	Conventional drilling operation not possible without moving the building	Alternative distribution method recommended
	Spills occur in unsaturated zone	Oxidant must be able to perform in the unsaturated zone
	Treatment plant is located nearby habitation units	Process implementation must be adapted to ensure inhabitants safety during operations

the oxidative conditions and the type of contaminant (Liang et al. 2004; Crimi & Taylor 2007; Sra et al. 2010). The good to high persistence of both compounds suggests that reactive species are formed and consumed gradually, limiting spontaneous heat production during the oxidation process. Permanganate does not need to be activated but has a low reactivity (E_0 = 1.7 V) in comparison to persulfate ions (E_0 = 2.1 V) and sulphate radical (E_0 = 2.6 V). This limitation can lead to significant remediation delays, affecting economic performance. The by-product of permanganate, manganese dioxide, is an issue since it modifies the porous medium properties by clogging soil pores and decreasing soil permeability.

Once persulfate is activated, the sulphate radical is nearly as reactive as the hydroxyl radical (E_0 = 2.6 V and E_0 2.8 V, respectively), and it has a good amenability regarding various PHC contaminants and chemical species, including mono-aromatic and poly-aromatic compounds (USEPA 2006b). SPS has high solubility in water (about 545 g/L at 20 °C), and its by-products generated during the oxidative reaction are sulphate ions and sodium ions. Sulphate and sodium ions are very soluble and can reach significant concentrations in ground water or in nearby lakes and, thus, may contribute to modifying and altering the fragile local environment. In the context of this study, communities are located on shore, and precipitation can leach sodium and sulphate ions to the nearby seawater, which already contains high concentrations of both components. This suggests a very low to nonexistent impact on the local environment regarding by-products. Finally, carbonates and bicarbonate can act as scavengers of free radicals and can also act as metal-complexing agents when metal activation is considered (Siegrist et al. 2011).

Although the use of persulfate salts such as SPS appears to be the most suitable oxidant in the context of diesel spills occurring under stilted buildings in northern Arctic communities, persulfate ions have to be activated by means of an appropriate method to increase its reactivity and ensure reasonable treatment delays.

Activation method selection

Persulfate can be activated using different methods (Table 3), and each has limitations and advantages (Table 5). When considering PHC contaminants, heat activation has the advantage of being generated on site, to accelerate contaminant desorption and increase contaminant solubility for better contact between oxidant and contaminant. However, it requires a high level of expertise and high operating costs to safely produce heat and distribute it into the subsurface. The heat activation method is not recommended where there is permafrost.

Persulfate activation can be performed by different metal ions (Ag^+, Cu^{2+}, Fe^{2+}, and Fe^{3+}) that release a single electron, initiating oxidant free radical formation by cleaving the peroxygen bond, which is a process similar to that involved in the Fenton reagents reaction (Kolthoff et al. 1951; House 1962). The Fe^{2+} ion participates in the overall oxidation process, acting itself as an oxidant, and is the most common and the most documented activation method regarding persulfate (USEPA 2006b; Siegrist et al. 2011). When the free ion form of Fe^{2+} is used, the pH of the medium has to be kept under three in order to maintain the reagent in solution during the oxidation process. As persulfate activation reactions lead to protons creation, the pH could decrease drastically during the process and would have to be adjusted to ensure the safety of

Table 5 Usual activation methods regarding persulfate ions, advantages and limitations (Block et al. 2004; USEPA 2006b; Crimi & Taylor 2007; Siegrist et al. 2011; Watts 2011; Zhao et al. 2013).

Activator	Advantages and limitations
Heat	Acceleration of the activation process and desorption rate of the contaminant. Very aggressive oxidizing conditions. Poorly understood reactions at high temperature and possibility of reaction inhibition. High energy costs, temperature elevation of the subsurface.
Dissolved Fe^{2+}	Most common and best documented activation method. Fe^{2+} and Fe^{3+} can be involved in the oxidizing reactions. Acidic pH <3.5 is essential to maintain activator in solution, environmental impact. Unwanted reactions between the activator and the oxidant can occur.
Chelated Fe^{2+}	Effective at neutral pH, thanks to ethylenediaminetetraacetic acid (EDTA) or citric acid mainly. Efficiency depends on the persulfate/Fe^{2+}/chelating agent ratio and distribution. Persulfate can react directly with the chelating agent. High costs and environmental impacts of chelation agents.
Hydrogen peroxide	Effective but aggressive and non-specific multi-radical oxidation. Poorly understood reaction mechanisms. Delicate and often repeated handling and injection. Significant increase in temperature can occur when reacting directly with contaminants.
Alkaline (NaOH, KOH, $Ca(OH)_2$, CaO_2, CaO)	Multi-radical oxidation but poorly understood chemistry. Requires a basic pH (>11) at all times, environmental impact. Possible alkaline hydrolysis of contaminants. Solid form and relative low cost.

operators and equipment. Fe^{2+} is only available in a liquid concentrated form, which increases transportation costs and handling risks. Once remediation is completed, the oxidant is depleted, and while the soil is buffering, pH rises slowly to neutral, and the reduced activator precipitates under a solid ferric hydroxide form ($Fe(OH)_3$) that can contribute to the clogging of porous media.

Chelating agents have been widely studied in ISCO iron activation processes since they can improve Fe^{2+} and Fe^{3+} availability in the aqueous solution by maintaining the iron ions in solution at neutral pH for a significant longer period of time (Block et al. 2004; Liang et al. 2004; Crimi & Taylor 2007). The most common chelates used are citric acid, ethylenediaminetetraacetic acid, 1-hydroxyethane-1, 1-diphosphonic acid and sodium triphosphate (Siegrist et al. 2011). They can all form organic-iron ligand complexes that stabilize the ions in solution. Although the use of chelating agents can avoid pH adjustment operations during treatment, the molar ratios between reagents (chelates, activator and oxidant) have to be adjusted with precision to control the activation process (Liang et al. 2004; Crimi & Taylor 2007; Liang et al. 2008). The use of chelating agents can increase the overall costs of operation since they are relatively expensive, and they are available only in liquid form, increasing their transportation costs. Finally, even if citric acid is biodegradable and organic chelates can be oxidized during the process (Rastogi et al. 2009), once remediation is done, the remaining organic and inorganic chelates can have an impact on the environment.

Persulfate activation using hydrogen peroxide leads to multi-radical production since hydrogen peroxide will also take part in the remediation process by oxidizing the targeted contaminant (Supplementary material, section 1). Radicals formed will then be able to activate persulfate ion, improving its reactivity (Block et al. 2004; Watts & Teel 2005). Watts & Teel (2005) and Cronk & Cartwright (2006) have shown that the temperature elevation associated with the use of hydrogen peroxide might contribute to the persulfate activation process. However, since hydrogen peroxide is unstable, the heat is produced and dissipated faster than the persulfate activation rate, and considerable volumes of activator must be added on a regular basis to ensure a proper activation temperature. The high transportation costs due to its liquid form, the operational complexity to ensure a proper persulfate–activator contact, and the significant temperature elevation associated with its use make hydrogen peroxide activation unsuitable in the context of this study.

Persulfate alkali activation is one of the most recently developed methods, and the chemical reactions involved are complex and poorly understood. Watts (2011) described the reduction of persulfate by hydroxide anion, leading to the creation of sulphate anions, superoxide anions and hydroxyl radicals, following Eqn. 1. Table 6 presents the equations involved in the persulfate alkali activation reactions and in the CP hydration reactions.

Furnam et al. (2010) propose that the hydrolysis of persulfate anions under basic activation conditions leads to the production of hydroperoxide anions, which reduces another persulfate ion, leading to the production of sulphate anions, sulphate radical and superoxide radical, following Eqn. 2.

Corbin (2008) assumes that the sulphate radical might be converted to a hydroxyl radical when reacting with hydroxyl anions, following Eqn. 3. It is assumed that the propagation reaction and reactions between hydroxyl radicals lead to the production of hydrogen peroxide, following Eqn. 4. Hydrogen peroxide may decompose to form hydroxyl radicals and superoxide anions,

Table 6 Persulfate alkali activation and calcium peroxide hydration chemical reactions and references.

Eqn.	Chemical reaction	Reference
1	$2S_2O_8^{2-} + 5OH^- \rightarrow 4SO_4^{2-} + O_2^{-\cdot} + OH^{\cdot} + 2H_2O$	Watts 2011
2	$S_2O_8^{2-} + HO_2^- \rightarrow SO_4^{2-} + SO_4^{-\cdot} + H^+ + O_2^{-\cdot}$	Furnam et al. 2010
3	$SO_4^{-\cdot} + OH^- \rightarrow SO_4^{2-} + OH^{\cdot}$	Corbin 2008
4	$OH^{\cdot} + OH^{\cdot} \rightarrow H_2O_2$	Corbin 2008
5	$H_2O_2 + HO_2^{-\cdot} \rightarrow O_2^- + OH^{\cdot} + H_2O$	Corbin 2008
6	$H_2O_2 + OH^{\cdot} \rightarrow HO_2^- + H_2O$	Corbin 2008
7	$SO_4^{-\cdot} + H_2O \rightarrow SO_4^{2-} + OH^{\cdot} + H^+$	Hayon et al. 1972
8	$CaO_2 + 2H_2O \rightarrow H_2O_2 + Ca(OH)_2$	Northup & Cassidy 2008; Wang et al. 2016
9	$CaO_2 + H_2O \rightarrow 0.5O_2 + Ca(OH)_2$	Heitkamp 1997; Wang et al. 2016; Lu et al. 2017
10	$Ca(OH)_2 \rightarrow Ca^{2+} + 2OH^-$	Cassidy & Irvine 1999

following Eqn. 5. Hydrogen peroxide might also react with the hydroxyl anion to form a hydroperoxide anion, following Eqn. 6.

Finally, Hayon et al. (1972) refer to the reaction of sulphate radicals with water to form hydroxyl radicals, as described in Eqn. 7.

Alkali pH has the advantage of giving diesel and soil the same surface charge and may thereby facilitate the dissolution of sorbed contaminants (Kuhlman & Greenfield 1999). But pH elevation and its environmental impact can still be a problem. In addition, Sra (2010) and Sra et al. (2010) warn about the possible buffering effect of carbonates on hydroxides.

The most common reagents used to elevate the pH to values ranging between 10 and 13 are mainly sodium hydroxide (NaOH) and potassium hydroxide (KOH) (Corbin 2008; Furnam et al. 2010; Siegrist et al. 2011; Liang and Guo 2012; Lominchar et al. 2018). However, NaOH and KOH have high water solubility (about 1100 g/L at 25 °C), and hydration reactions are rapid and very exothermic, which is an issue when porous medium temperature elevation is not desired.

Most recently, CP has been used as an oxidant in water or soil treatment processes (Siegrist et al. 2011; Lu et al. 2017; Lapointe et al. 2020), as an activator in modified Fenton Chemistry (Northup & Cassidy 2008) or as an oxidant together with persulfate activated with chelated iron (Wu et al. 2017). CP (CaO_2) is almost insoluble in water, and it has the advantage of releasing H_2O_2 gradually, controlled by the rate of its dissolution (Northup & Cassidy 2008; Wang et al. 2016), following Eqn. 8. Bogan et al. (2003) even suggest that CP is a more effective source of H_2O_2 for ISCO than its diluted liquid form. Hydrogen peroxide can participate in both persulfate and CP activation by cleaving the peroxide bond. It can also participate in the contaminant oxidation, depending on the H_2O_2 concentration and on the free radicals generated. The low dissolution rate of CP suggests that limited H_2O_2 concentration does not entail a significant temperature increase. CP will also release O_2 while hydrating, following Eqn. 9, supplying O_2 to microbes and plants (Heitkamp 1997; Wang et al. 2016; Lu et al. 2017).

$Ca(OH)_2$ has poor solubility in water (1.7 g/L at 20 °C). Its gradual dissolution, following Eqn. 10, liberates hydroxide ions and creates alkali conditions by drastically increasing the pH (Cassidy & Irvine 1999) without a significant temperature increase.

CP is available under a solid powder form together with $Ca(OH)_2$ (75% CP and 25% $Ca(OH)_2$) and is very stable, which facilitates its transportation and manipulation on site (Lu et al. 2017). The fact that it is insoluble in water requires different distribution methods to ensure a proper contact with the oxidant (direct push technology, trench deposits, mixed with soil, etc.). By-products generated will be $Ca(OH)_2$, $CaSO_4$ and Na_2SO_4 depending on temperature, pH and reagent concentrations. $Ca(OH)_2$ and $CaSO_4$ both have a poor solubility and can be confined in controlled locations depending on the CaO_2 distribution method. In contrast, Na_2SO_4 has high solubility but it is found in large quantities in nearby seawater and does not represent an environmental problem in the context of application of this study.

Considering the elements pointed out in this section, the alkali activation method with CP appears to be the most suitable for activating persulfate salts in northern Arctic communities. It is stable and available under a solid form, reducing transportation and storage issues and costs. It can be safely operated on site, and its very low solubility allows versatile distribution methods. It has the capacity to increase the pH to desired values and itself acts as an oxidant, generating H_2O_2 during the dissolution reaction. It is an effective source of 0_2, which can be a great advantage when train technologies are considered. A final smoothing bioremediation step can be considered and would be assisted with O_2 liberation due to remaining CP dissolution over time.

The use of CP as an alkali activator in a persulfate oxidation context is a very recent technology, and field trials have to be performed to: (1) document the temperature increase associated with oxidation reactions involving persulfate and CP; (2) verify that the pH of the oxidizing solution can be easily adjusted and maintained between 10 and 13 during the operation; and (3) identify and quantify the by-products generated during the oxidation process.

Conclusion

The absence of global electricity grids in Arctic and Subarctic regions makes diesel the main source of energy for everyday household needs in small communities. Every stilted housing unit has its own heater and diesel tank, and the lack of maintenance and expertise leads to frequent diesel spills on the raft soil under the buildings. The management option that is used now is based on soil transportation and landfilling and is not well-suited to Far Northern conditions. It is also very expensive, and local authorities are looking for innovative remediation alternatives.

In reviewing and comparing remediation options, ISCO involving activated SPS was identified as the most suitable technology. SPS can be purchased in solid form, which simplifies its transportation and manipulation on site. Once activated, SPS offers good reactivity and amenability to the aliphatic, mono-aromatic, and poly-aromatic compounds found in diesel. Its high persistence allows for different distribution methods and ensures progressive contaminant degradation, assuming a low temperature increase associated with the oxidation reactions. We consider persulfate alkali activation using CP the most convenient and efficient activation method for SPS. CP is also available in solid form, and its hydrated form, $Ca(OH)_{2(s)}$, has a low solubility and will gradually release hydroxide anions in solution, providing the alkali conditions needed to activate the SPS. The CP hydration reaction also releases hydrogen peroxide, which contributes to SPS activation and which can directly oxidize diesel. CP is also an effective source of O_2, which is an advantage when a final polishing bioremediation step is needed. By-products generated by the SPS activation chemical reactions are mainly composed of calcium sulphate, hydrated lime and sodium sulphate. The first two compounds are almost insoluble and can be confined during the operation, avoiding clogging effects in porous media. Sodium sulphate is highly soluble and can be leached by natural precipitation into nearby seawater, which already contains such compounds in large concentrations.

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