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Comprehensive Approach for Monitoring and Remediating Petroleum-Derived Contaminants in the Arctic

Case Study of the Former NARL Site near Utqiagvik, Alaska (Formerly Barrow)

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Simplified schematic showing the layout and major point source locations of petroleum hydrocarbons at the former NARL complex in Utqiagvik, Alaska

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Abstract

The Arctic region of Alaska has a history of petroleum contamination from repetitive fuel spills and the overuse of petrochemicals. Notably, the presence of the former Naval Arctic Research Laboratory (NARL) outside the city of Utqiagvik, Alaska (formerly known as Barrow), resulted in the contamination of local soils and groundwater with petroleum-derived hydrocarbons. Since the NARL closure in 1987, the U.S. Navy (primarily) has implemented many environmental investigations, remediation, monitoring, and containment strategies. However, the soil and subsurface soil unique to the Arctic complicates traditional remediation techniques as a result of the harsh environment and underdeveloped infrastructure of the remote site. Bioremediation and stimulating the existing microbial community represent attractive methods of decontamination because they are nontoxic and relatively easy to implement. The results from this study offer a comprehensive approach for characterizing petroleum-derived contamination specific to Arctic regions by coupling nondestructive geophysical tools with in situ hydro-biogeochemical methods. The overall goals of this project were to investigate the surface and subsurface soil properties at the former NARL site for the Naval Facilities Engineering Command Northwest, monitor the distribution of hydrocarbons, characterize petroleum-derived hydrocarbons, and test various bio- and phytoremediation scenarios both in the laboratory and as a field study.

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Preface

This study was conducted for the Naval Facilities Engineering Command Northwest (NAVFAC NW) under Project #N6247315MPT0010. The technical monitor was Ms. Kendra Leibman.

The work was performed by the Force Projection and Sustainment Branch (CEERD-RRH), the Biogeochemical Sciences Branch (CEERD-RRN), and the Engineering Resources Branch (CEERD-RRE) of the Research and Engineering Division (CEERD-RR), U.S. Army Engineer Research and Development Center, Cold Regions Research and Engineering Laboratory (ERDC-CRREL). At the time of publication, Dr. Harley Cudney was Acting Chief, CEERD-RRH; Dr. Justin Berman was Chief, CEERD-RRN; Mr. Jared Oren was Chief, CEERD-RRE; and Dr. Mark Moran, CEERD-RZT, was the ERDC-CRREL Technical Director. The Deputy Director of ERDC-CRREL was Mr. David B. Ringelberg, and the ERDC-CRREL Director was Dr. Joseph L. Corriveau.

COL Ivan P. Beckman was the Commander of ERDC, and Dr. David W. Pittman was the Director.

Acronyms and Abbreviations

ADEC	Alaska Department of Environmental Conservation
AKRO	Alaska Research Office
ARL	Arctic Research Laboratory
ASE	Accelerated Solvent Extraction
ATSDR	Agency for Toxic Substances and Disease Registry
BFTF	Bulk Fuel Tank Farm
CCR	Capacitively Coupled Resistivity
CRREL	Cold Regions Research and Engineering Laboratory
DRO	Diesel Range Organics
EPH	Extractable Petroleum Hydrocarbon
ERDC	U.S. Army Engineer Research and Development Center
ERT	Electrical Resistivity Tomography
GC-FID	Gas Chromatography–Flame Ionization Detection
GC-MSD	Gas Chromatography–Mass Spectrometric Detection
GC-PID	Gas Chromatography–Photo Ionization Detection
GPR	Ground-Penetrating Radar
GPS	Global Positioning System
GRO	Gasoline Range Organics
HAVE	Hot Air Vapor Extraction
IR	Infrared Spectroscopy
LIF	Laser-Induced Fluorescence
LLE	Liquid-Liquid Extraction

LOI	Loss on Ignition
LTM	Long-Term Monitoring
NARL	Naval Arctic Research Laboratory
NAVFAC	Naval Facilities Engineering Command
NSF	National Science Foundation
NW	Northwest
PAH	Polycyclic Aromatic Hydrocarbon
Pb	Lead
PCB	Polychlorinated Biphenyls
PHC	Petroleum Hydrocarbon
RME	Reasonable Maximum Exposure
RRO	Residual Range Organics
SFE	Supercritical Fluid Extraction
SPE	Solid Phase Extraction
SWE	Snow Water Equivalence
UIC	Ukpeaġvik Iñupiat Corporation
USACE	U.S. Army Corps of Engineers
USCS	Unified Soil Classification System
USN	U.S. Navy
UVOST	Ultra-Violet Optical Screening Tool
VOC	Volatile Organic Compounds
VPH	Volatile Petroleum Hydrocarbon

1 Introduction

1.1 Background

1.1.1 Petroleum-derived contamination

Petroleum-derived products account for some of the most widely used chemical substances in modern society and are used for a variety of applications, including fuel for cars, aircrafts, and ships, etc.; generating heat and electricity; and producing synthetic fibers, lubricants, solvents, plastics, paints, fertilizers, and synthetic rubbers (Agency for Toxic Substances and Disease Registry [ATSDR] 1999; Speight 2014). In general, the primary uses of petroleum are as a fuel source and as a raw material (petrochemical) for the synthesis of organic compounds (Eneh 2011). Chemically, petroleum is a complex mixture consisting of various widely occurring hydrocarbon compounds, often along with minor amounts of nitrogen, oxygen, sulfur, and metals. The chemical composition of petroleum hydrocarbons (PHCs) varies widely from simple carbon chains (paraffins, such as methane), to ring-shaped hydrocarbons (aromatics, such as benzene). While the early history of petroleum dates back to roughly 4000 BC, modern usage is typically attributed to the refining and subsequent commercialization of petroleum in the late 1850s and early 1860s. Widespread petroleum use over the last approximately 150 years has caused significant concern for the anthropogenic input of PHCs to the environment (Speight 2014).

Studies have documented the release of petroleum-based contaminants into the environment by a variety of sources, including industrial activities (Cheremisinoff and Rosenfeld 2009), military activities (Naval Facilities Engineering Command Northwest [NAVFAC NW] 2001), and international communities (National Round Table on the Environment and the Economy 1997; European Environmental Agency 2011; Zhang et al. 2015). Unintended release of petroleum and petroleum-derived products in the environment is often accidental in the form of a major spill or can also be a result of industrial, commercial, urban, or domestic leaks; underground storage leakage; storm water runoff; and waste effluents. In Alaska alone, an estimated 41 million liters of oil was spilled by *Exxon Valdez* in the coastal waters and another estimated 1.386 million liters of fuel were spilled at the U.S. Naval Arctic Research Laboratory (NARL) when the *Air-strip* was operational in Utqiagvik (ATSDR 1999; NAVFAC NW 2013b).

Petroleum contamination as a result of large spills is a major health concern and can harm wildlife, decimate land and ocean habitats, degrade agriculturally available lands, and impact air pollution. Most petroleum-based contaminants are considered toxic, carcinogenic, and difficult to remediate given the diffuse and persistent nature of hydrocarbons and the wide variability of compounds that can be located at any given site (Braddock et al. 1997; ATSDR 1999). The degree to which PHCs are toxic is highly dependent on their polarity and subsequent solubility in water (World Health Organization 2005). For humans, negative long-term effects of PHC exposure include prenatal toxicity, reproductive complications, lung cancer, and leukemia (International Agency for Research on Cancer 1989).

Petroleum-derived contamination on land generally leaches into the subsurface and can migrate into surface water, groundwater, and local marine environments. Petroleum hydrocarbons vary widely in their chemical properties, dependent on the speciation, which affects their relative water solubility, toxicity, volatility, and biodegradability (Park and Park 2010). For example, aromatic hydrocarbons, in comparison to alkanes and unsaturated aliphatic hydrocarbons, tend to be relatively more toxic and more resistant to microbial degradation (Ahmed et al. 2015; Adeniji et al. 2017). In addition, each environmental location represents a unique system (soil type, presence of organics and microbes, pH, colloids, etc.) for contamination that can affect overall mobility and bioavailability. Once PHCs are released into the environment, they can transform and biodegrade in a process called fractionation; therefore, only representative chemical signatures often remain (Fan et al. 2011). Hydrocarbon fractions (chemical signatures) can be used to identify parent sources of petroleum contamination and link PHC migration plumes to point-source locations. Petroleum hydrocarbons are reactive, readily transform and biodegrade, and bio-accumulate over time; and their fate is largely controlled by microbiological, physical, and chemical processes that occur in subsurface environments (Cozzarelli et al. 1994). In addition, PHCs can partition between phases, resulting in often wide dispersion throughout the environment (Nadim et al. 2000). As a result, detailed understanding of not only the amount spilled but also the speciation of the PHC, subsurface distribution, and environmental fate of where the hydrocarbons will ultimately end up at a contaminated site is necessary before planning risk assessments and designing site-specific remediation strategies.

Determining the (1) quantity, (2) type of PHC, and (3) speciation (or major fractions present) of PHCs is essential for monitoring and assessing petroleum-contaminated sites and planning remediation strategies. These three factors can be complicated to investigate and time consuming since the analytical procedures to quantify and characterize PHCs tend to be detailed and difficult to compare across methods due to various extracting solvents and calibration standards (Adeniji et al. 2017). Therefore, measuring PHC distribution in situ is an attractive alternative to laboratory-based methods. Direct chemical sensing of PHCs using laser-induced fluorescence (LIF) coupled to an Ultra-Violet Optical Screening Tool (UVOST) offers the unique capability of PHC data acquisition in real time and can also detect residual and free-phase PHCs in both unsaturated and saturated zones. While implementing LIF-UVOST does not yield information on PHC speciation, it remains a powerful tool for determining overall PHC distribution and has the added potential of providing insight into the overall fate of PHC at a given site when coupled with hydrological groundwater modeling methods and tracer-dye studies. For PHC speciation (and fractionation) analyses, typically some form of extraction procedure is employed, such as liquid-liquid extraction (LLE), solid phase extraction (SPE), accelerated solvent extraction (ASE), and supercritical fluid extraction (SFE), depending on the type of suspected PHC present in the samples. Typical instrumentation for PHC fraction analyses includes infrared (IR) spectroscopy, gas chromatography–flame ionization detection (GC-FID), gas chromatography–mass spectrometric detection (GC-MSD), and Fourier transform ion cyclotron resonance mass spectrometry (Marshall and Podgers 2004; Wang et al. 2006). The techniques involving gas chromatography and mass spectrometry are specifically useful due to the added information the technique can provide concerning the PHCs structure and type, which is integral for deciding site-specific remediation strategies (Fan et al. 2011; Adeniji et al. 2017).

Remediating point-source locations and surrounding ecosystems contaminated by PHCs has been the subject of numerous studies, including the use of activated carbon (Ayotamuno et al. 2006), ozone (Masten and Davies 1997), iron minerals (Kong et al. 1998), chemical oxidation (Yen et al. 2011; Yoo et al. 2017), sewage sludge (Asadollahi et al. 2016), and bioremediation (Das and Chandran 2011). Bioremediation of PHCs by microbes is an attractive technique for site cleanup because it is nondestructive to the surrounding environment when compared to other more invasive, traditional remediation techniques like dredging, excavation, or incineration.

Microbial bioremediation has been shown to be promising for the biodegradation of PHCs by completely breaking down the hydrocarbon contaminants into other less-toxic substances like carbon dioxide, water, and inorganic compounds (Atlas 1981; Das and Chandran 2011).

The present case study focuses on the site-specific petroleum contamination located at the former NARL site in Utqiagvik, Alaska, and highlights the unique implementation of a suite of technologies, both in situ and non-destructive, to monitor and remediate PHCs in the local soil and groundwater. Using this comprehensive approach—coupling both remote, noninvasive geophysical tools with in situ hydro-biogeochemical methods—yields not only site-specific information on the overall distribution of PHCs but also provides a necessary understanding of the overall fractionation of PHCs. Characterization of PHC fractionation is vital for deciding which remediation techniques might yield the highest success rates. Overall, the work in this study presents a comprehensive approach for assessing petroleum-derived contaminated sites in Alaska and provides a methodology applicable to other similar sites located in Arctic regions.

1.1.2 Arctic surface and subsurface soil systems

The Arctic presents a unique environment for studying contaminant fate and transport, mobility, and speciation and for designing remediation strategies. Unlike more temperate climates, the Arctic experiences harsh winters with minimal sunlight and short, sun-intense summers, which cause the soil-thermal regime to freeze and thaw multiple times every year. However, there is a portion of the soil column in the Arctic that never thaws, called permafrost. Permafrost is defined as soil that has consistently remained frozen for at least two years (Jansson and Taş 2014). Coupled moisture and thermal states for soil freezing and thawing translate to water and ice coexisting below 0°C (Williams 1964; Anderson et al. 1973). Permafrost regions with high relative salinity (like Utqiagvik, given the proximity to the coast) tend to exhibit poorly bonded soil particles due to the presence of liquid water as a result of the freezing-point depression. The behavior exhibited is similar to thawed soil conditions despite the ground temperature being below 0°C (Andersland and Ladanyi 2004). Above the permafrost layer and below the ground surface exists an “active layer” that participates in annual freeze–thaw cycles, and the depth of the active layer is dependent on a variety of factors, including ambient air temperature, soil condition, and snow depth.

Arctic hydrological processes are highly seasonal and are therefore dependent on the annual freeze–thaw cycle. Spring (warmer temperatures and longer times between sunrise and sunset) initiates the annual freshet, or snowmelt, which produces some of the highest volumes of water to surface waters and marine environments measured annually (Hinzman et al. 1991). Summer in the Arctic is controlled by alternating warm and sunny days with rain and storms that can contribute significantly to groundwater flow regimes and surface water depths. Late fall and early winter correspond to the annual time where the active layer is at its deepest yearly extent. Also during this time of year, the surface of the soil is frozen. Consequently, pore waters and baseflow can produce nutrient and elemental pulses (for some elements) to surface waters during this time of the year (similar to fluxes observed during spring freshet) as a result of redox gradients present in the subsurface regime at the permafrost–active-layer boundary (Barker et al. 2014, Pokrovsky et al. 2016). Various interfaces that are present within the subsurface regime are unique to Arctic environments and can influence chemical processes that control contaminant transport, including precipitation, adsorption/desorption, complexation, and microbial interactions. Interfaces typically present in Arctic environments include taliks, pingos, ice wedges, and ice lenses, among others (Shur and Jorgenson 2007). Additionally, the presence of permafrost acts as a physical gradient in which the various ice layers, or lack thereof, can affect overall migration patterns (Pokrovsky et al. 2016). As a result, seasonality and the extent to which the soil is thawed or frozen has the potential to significantly contribute to the fate and transport of contaminants in the Arctic.

Understanding how contaminants partition and migrate in Arctic ecosystems requires detailed knowledge of the soil thermal regime and the location of the permafrost and active-layer depths. For example, snow coverage has been shown to insulate soil layers (particularly as a function of depth) and to contribute to an increased soil temperature for the subsurface (Guymon and Luthin 1974). Not only does the insulating effect of snow coverage impact hydrological and soil processes but it also has substantial implications for soil biological activity and ecological respiration. The activity of the microbial community present in soil tends to have an increased functionality at higher temperatures, measurable to an extent. Walworth et al. (2001) found that the microbial community in soil near the former NARL complex responded positively to temperature increases from 5°C to 20°C, but the upper extent of positive response was around

40°C. Therefore, bioremediation efforts will be affected by increased soil temperature and the extent to which soil freezes as a function of depth (Tate 2017).

Bioremediation efforts have the potential to experience a higher degree of success given more snow coverage, where a higher soil temperatures is experienced, further necessitating the need for accurate and detailed surface and subsurface characterization. Conversely, spring snowmelt has been shown to release from organic matter and vegetation leachate that can penetrate permafrost surface soils (Buckeridge et al. 2016). The penetration of this nutrient-rich leachate is one of the major factors that drives terrestrial microbial activity; therefore, remediation activities may be most successful in areas that have high leachate pools in the spring to summer but that also experience large snow accumulations in the winter given the extent to which snow accumulation insulates the soil thermal regime. Therefore, it is essential to understand, constrain, and characterize all seasonal chemical, hydrological, and geophysical parameters to most effectively plan remediation strategies for PHC and any other potential contaminants present in Arctic regions.

Field and laboratory efforts in the Arctic are novel, and resulting data have considerably broad implications with respect to the impact of climate warming on Arctic and subarctic ecosystems (Douglas et al. 2014). As such, there exists a need for comparably more-powerful instrumentation that is also noninvasive and easy to use in extreme climates. The Arctic environment can be harsh, often limiting the use of advanced technologies and restricting the types of methods that can be used (Cury et al. 2015). The case study presented in this work highlights the implementation of ground-penetrating radar (GPR), electrical resistivity tomography (ERT), and capacitively coupled resistivity (CCR) as highly effective and robust geophysical tools for the characterization of the soil and subsurface soil in Arctic Alaska. While GPR, ERT, and CCR techniques are all noninvasive and highly efficient, they also have the capabilities of mapping frozen versus unfrozen soil and also determining the extent to which highly resistive substances distribute in the subsurface. Among other capabilities, GPR can specifically estimate thaw-layer thickness and ice-wedge characteristics, which is extremely important considering the variability of soil properties in the Arctic (Dafflon et al. 2017). Since the electrical resistance of water in the ground is relatively low compared to the electrical resistance of ice in the ground, which is several orders of magnitude higher, ERT is a

highly effective method for mapping frozen versus unfrozen soil and water compartments in surface and subsurface soil systems. Similarly, CCR also uses resistivity as a geophysical tool but uses the earth as one conductor of a parallel plate capacitor and sends a continuous current sine wave through the dipole, which polarizes the surrounding earth material. The receiver then measures the induced polarization, and the resistivity can be calculated. While GPR, ERT, and CCR are not new techniques, the instrumentation and end-user interface technologies have substantially advanced in recent years, making them more attractive alternatives to destructive geophysical characterization tools, such as creating boreholes and digging soil pits to expose soil layers. GPR, ERT, and CCR techniques coupled with soil thermal-regime modelling and ground truthing using frost-probing techniques are necessary tools for characterizing soil systems in the Arctic.

1.1.3 Bioremediation in the Arctic

Microorganisms present in soil and aquatic systems naturally possess the ability to break down hydrocarbons comprising complex petroleum-derived contaminants into relatively harmless end products (carbon dioxide, water, and simpler cellular constituents) and in the process use the hydrocarbons as a source of energy (Atlas 1981; Naseri et al. 2014). This natural process of biodegradation in addition to many other natural processes that occur when contaminants enter into the environment (oxidation, reduction, precipitation, sorption, complexation, etc.) all contribute to a process called weathering. Chemical, physical, and biological weathering transforms specific contaminants and ultimately controls fate and transport at a given site. Landscape-scale remediation efforts must take into account weathering reactions, weathering rates, and key mechanistic pathways before determining potentially successful remediation scenarios.

Implementing remediation techniques in the Arctic is costly, time consuming, and technologically complex due to the harsh nature of the environment, underdeveloped infrastructure, and difficulties of transportation (Reed and Ronhovde 1971). Microorganisms that are naturally present in soils have the ability to breakdown complex hydrocarbons, but the processes that initiate the breakdown of PHC compounds are relatively slow (Cheremisinoff and Rosenfeld 2009). Therefore, bioremediation by artificial methods seeks to maximize the specific organisms and environmental conditions that favor PHC degradation to enhance positive results at contaminated sites.

Arctic soils have lower biological populations compared to other soils (Robinson and Wookey 1997). Therefore, understanding site-specific microbial communities before implementing remediation strategies is essential. Conditions in the Arctic will influence the effectiveness of bioremediation techniques. In particular, the harsh climate (freezing soil temperatures), annual freeze–thaw cycles, snow coverage, low soil nutrient levels, and lack of sunlight will play a dramatic role in the overall microbial stimulation (Atlas 1981; World Wildlife Fund 2007). Factors that influence the rate of biodegradation, including type of bioremediation (biostimulation, bioaugmentation, biopiling, composting, metabolic engineering, etc.), soil temperature, soil nutrient level, soil humidity, oxygen level, PHC saturation, soil pH, and soil salinity, can be modified *in situ* to increase decontamination at PHC-contaminated sites in the Arctic (Samanta et al. 2002; Naseri et al. 2014). For example, lower soil temperature inhibits the rate of metabolic activity, and the microbial community must be adapted to cold climates in order to utilize hydrocarbons. Successful microbial additions to soils in the Arctic primarily focus on bioaugmentation (addition of cold-adapted soil microbes to already existing communities) along with collecting a detailed understanding of site-specific Arctic soil conditions and PHC environments.

Many studies have focused on bioremediation for managing and decontaminating areas saturated with PHCs (Eweis et al. 1998), but there remains a limited understanding of bioremediation in the Arctic (Mohn et al. 2001). Braddock et al. (1997) showed that the removal of various hydrocarbons was possible in this region by using mesocosms that were incubated on-site and stimulated by nutrient additions to the soil. A separate bioremediation study done by Mohn et al. (2001) in Cambridge Bay and Komakuk Beach of Yukon, Canada, demonstrated a reduction in total petroleum hydrocarbons from 196 to below 10 mg/kg and 2109 to 195 mg/kg, respectively, after one year of treatment using inoculated biopiles. Garneau et al. (2016) highlighted the effects of dissolved organic matter in stimulating hydrocarbon degradation and also showed microbial diversity decreases with increasing hydrocarbon exposure (a result also supported by Cury et al. [2015]), which is particularly important considering contaminated sites often have convoluted chemical histories with repeat PHC spills (Naseri et al. 2014), like the former NARL complex.

1.2 Objectives

The major objective of the outlined case study is to describe the comprehensive efforts done by the U.S. Army Cold Regions Research and Engineering Laboratory (CRREL) team in response to a project request by NAVFAC NW and to provide an outlined methodology for future PHC monitoring programs in cold regions. The comprehensive studies presented spanned a total of 2 years of field work and assessed the surface and subsurface soil features (permafrost, soil salinity, etc.) at the former NARL site; quantified the current state of PHC distribution, including any previously installed remediation techniques (subsurface containment berm); and tested different bioremediation methods for decreasing the amount of PHCs in soils.

1.3 Approach

The CRREL team used nondestructive geophysical tools to characterize both the surface and subsurface soil systems and combined those surveys with in situ hydro-biogeochemical methods to monitor the transport and distribution of PHC contamination at the former NARL site. Specifically, the current case study determined baseline characteristics of the primary petroleum constituents, determined PHC concentrations in a variety of soil samples collected from the NARL site, quantified the soil thermal regime properties of the soil, and outlined the groundwater flow paths that dominated at the different sites. The study also investigated the physical and microbial soil attributes of the PHC-contaminated site in Utqiagvik, Alaska. This included soil pH, soil carbon fraction, respiration rate, water content, soil moisture, soil temperature, and soil DNA concentrations. Finally, a bench-scale laboratory experiment and paralleled pilot-scale field study were implemented to study different bioremediation options. Altogether, this data will be archived and revisited when planning further bioremediation strategies in the future.

2 History

2.1 Former Naval Arctic Research Laboratory (NARL) case study

The Arctic Research Laboratory (ARL) under contract with the Office of Naval Research was preliminarily established on 13 August 1946 by Vice-Admiral Harold G. Bowen, USN* (Ret.), Chief of Naval Research; and research officially began in 1947. The intent of Vice-Admiral Bowen was to build an establishment where “civilian scientists from universities, research institutions, and government departments could conduct basic studies on Arctic problems” (Shelesnyak 1948). In initial scope and design, it was to be one of the foremost research facilities for Arctic achievements and to contribute substantially to the field of Arctic research (Reed and Ronhovde 1971). In the mid-1900s, it was the only full-time U.S. laboratory devoted to basic research in the Arctic (Britton 1964).

The location of Point Barrow, Alaska, was specifically chosen because it was the most northerly U.S. settlement in North America, and it exemplified a typical coastal region in the Arctic. In addition, already in existence on-site was the Naval Petroleum Research Station No. 4 (“Pet 4”), which was a 90,000 km² plot of land set aside by President Harding in 1923 for oil exploration. At that time, the oil exploration camp contained camp-style huts parallel to the Chukchi Sea and an Airstrip between North Salt Lagoon and Imikpuk Lake (NAVFAC NW 2010). In actuality, the existence of “Pet 4” at Point Barrow made the early operation of ARL possible (Reed and Ronhovde 1971). In February 1948, Dr. Laurence Irving was appointed the first scientific director of ARL; and the facility focused on research topics close to the interests of the U.S. Navy (Shelesnyak 1948). At the height of ARL research activity, the site housed approximately 190 buildings and received more research grant applications than it could support (NAVFAC NW 2010).

Between 1950 and 1960, ARL’s name was changed to the Naval Arctic Research Laboratory (NARL); and it was also during this time that the U.S. Navy’s interest in the Arctic began to decline. Three events likely contributed to decline of NARL’s robust research activity: (1) the discovery of oil began pushing science away from Point Barrow to Prudhoe Bay; (2) the National Science Foundation (NSF) was created and was mandated by

* U.S. Navy

Congress to focus on research in the Arctic; and (3) the large, bureaucratic projects under NSF taxed the infrastructure and facilities at NARL (NAVFAC NW 2010). Finally, in 1980, NARL officially closed; and the facility was decommissioned in 1981. The majority of NARL property (except the Airstrip and antenna field site) was transferred to the Ukpeaġvik Iñu-piat Corporation (UIC) in 1986. It is now referred to as UIC-NARL and is used as a facility support center for local activities (Hart Crowser 2000a; 2000b; McCarthy et al. 2003; NAVFAC NW 2010).

Over the active life of NARL, approximately 1.4 million liters of fuel were spilled with the majority of spills reported at the Airstrip. These spills were documented in a total of four separate events (Integrated Concepts and Research Corporation 2003). In August 1976, an underground pipe failed and discharged an estimated 182,000 L of gasoline. In December 1978, an estimated 95,000 and 1 million liters of JP-5 fuel were spilled in two separate events. The fourth documented spill occurred in 1986 and involved JP-5 fuel again, but the quantity remains unknown (Baldwin et al. 2000). Currently, the complex includes three main sites of interest for the U.S. Navy's continual monitoring programs: the Airstrip, the Powerhouse, and the former Bulk Fuel Tank Farm (BFTF). Figure 1 shows a map of the area.

Figure 1. Map of the former NARL complex showing its broad location within Alaska and major lakes and bodies of water. Locations of the village of Utqiaġvik (formerly known as Barrow), Airstrip site, Powerhouse site, former Bulk Fuel Tank Farm site, and Imikpuk Lake are noted.

The Airstrip site, adjacent to the shoreline of the Arctic Ocean (about 250 m inland), includes a 1520 m runway, a large hangar, and an apron area and covers approximately 500,000 m². The Airstrip site is also where the containment berm and fuel recovery trench was placed to stop migration of free PHC product from entering Imikpuk Lake. The Powerhouse site is approximately 1 km southwest of the Airstrip site and includes the former Powerhouse building, an inactive water pump house, and an equipment storage area. The Powerhouse site lies approximately 300 m inland from the Arctic Ocean shoreline. The former BFTF site lies between the North Salt Lagoon and a melt water pond on the west and east, respectively (Figure 1), and covers approximately 20,200 m². In operation, the BFTF site held six aboveground fuel storage tanks, a pump house, and piping for fuel for the NARL complex (Wagner et al. 2016b).

2.2 Monitoring and remediation efforts

In 1978, there were 530,000 L of usable gasoline recovered following a spill in December of that year. In 1996, 207,000 L of free product (fuel) were also recovered. In September 1996, Arctic GeoScience, Inc., under contract with Foster Wheeler Environmental Corporation, performed a geophysical investigation of the Airstrip Fuel Site at NARL to determine the thickness and depth of a gravel fill, to locate potential fuel spills, and to determine the extent of any located fuel spills. The primary method of analysis was electromagnetic ground conductivity and GPR. The results from this investigation yielded two types of areas based on historical data and field measurements: (1) areas assumed to contain no contamination and (2) soil highly saturated with hydrocarbons. In addition, the crew focused on locating a known, abandoned, underground fuel line and was supposed to determine if there was any potential contamination associated with that fuel line. However, the pipeline location was overlain by a metallic mat; and any contamination could not be delineated during this first monitoring effort, necessitating further monitoring and characterization of the site (Arctic GeoScience 1996).

Later in 1996, a fuel recovery trench was placed along the eastern shoreline of Imikpuk Lake, and a 460 m long containment berm was constructed below the surface of the ground with the purpose of creating an ice wall barrier to limit the transport of petroleum-derived contaminants migrating into Imikpuk Lake. Foster Wheeler Environmental Corporation implemented the subsurface barrier (containment berm) to prevent existing groundwater contamination from migrating into the adjacent Imikpuk

Lake, which was the source of local drinking water (Baldwin et al. 2000). The design of the berm uses two barriers to control groundwater movement: (1) an installed rigid insulation near the ground surface to create a permafrost mound and (2) an installed vertical geomembrane curtain wall within the center of the permafrost mound. The containment berm in conjunction with the recovery trench limited the movement of contaminated groundwater into Imikpuk Lake. Contaminated groundwater that was collected by the recovery trench was pumped to a treatment plant and discharged into an existing sewage lagoon for final disposal. During the first year of operation of the containment berm and recovery trench, approximately 2.8 million liters of suspected contaminated groundwater were treated and discharged into the sewage lagoon, and approximately 204,000 L of free product were recovered from the collection system at the treatment plant (Baldwin et al. 2000; Bjella 2015).

In 1997, approximately 78,000 L of fuel were recovered. In July–August of that same year, engineers from Linder Construction, Inc., contracted by NAVFAC NW, excavated, cleaned, and disposed of approximately 1300 m of abandoned fuel pipeline at NARL. The pipeline originated at the Airstrip, along the northwest shore of the North Salt Lagoon and Imikpuk Lake before ending at the Power Plant (at the Powerhouse site), west of Imikpuk Lake. The fuel pipeline was 76 mm thick and had to be cut using a gasoline-powered cut-off saw. The excavation work was difficult given that residual fuel and petroleum-derived contamination was present in many locations along the route of the abandoned pipeline. Also during this historical remediation effort, contracted environmental engineers performed a field investigation for soil contamination along the route of the abandoned pipeline. During the soil study along the pipeline, the crew found significant soil staining and visible contamination in the excavated soil. The “grossly” contaminated soil was stockpiled next to the excavation trenches.

During this time, the crew also found broken joints and elbows in the pipeline, which were determined to be the major cause for fuel leaks, and also found that the majority of the pipeline was below the groundwater table. Finally, another concern that arose from this excavation was the presence of deteriorated and leaking drums that were stored near the power plant in an area adjacent to the pipeline location. In the area surrounding the drums, the crew found significant surface staining of the soil. The final directive from the U.S. Navy during the summer 1997 pipeline excavation was to remove the pipeline by direct excavation, and any contaminated soil

was returned directly to the excavation. During this excavation, benzene, toluene, ethylbenzene, xylene, gasoline range organics (GRO), diesel range organics (DRO), and residual range organics (RRO) were detected along various points in the pipeline route. The highest concentrations of benzene detected were 4.94 mg/kg, and the highest levels of DRO and RRO were reported at 19,600 mg/kg and 2120 mg/kg, respectively. The crew noted that in the sections of high DRO and RRO, the abandoned fuel pipeline was visibly intact, indicating there was likely another source for these petroleum-derived contaminants present in the surface and subsurface. Other sources found during this excavation could be attributed to the leaking drums or significant vehicle and equipment operations and storage in these areas (Linder 1998).

In 1998, a total of 340 L of fuel was recovered and collected. Also, in October 1998, NAVFAC NW, contracted with Bristol Environmental and Engineering Services Corporation to remove abandoned drums, tanks, transformers, and debris at NARL. For this excavation project, the crew focused on the Old Waste Disposal Area and the Airstrip. The Old Waste Disposal Area is located along the northern shore of Middle Salt Lagoon, and the Airstrip is bordered by Imikpuk Lake and the North Salt Lagoon. The majority of this work provided the necessary investigation of the contents of the drums and other debris, and removal was completed by shipping the containers off-site to appropriate waste storage and disposal facilities in December 1998. Contents of select drums were analyzed and contained GRO, DRO, and RRO at concentrations of 0.388 mg/L, 7.65 mg/L, and 0.727 mg/L, respectively. No evidence of metals, reactive cyanide, reactive sulfide, or polychlorinated biphenyls (PCB) were detected in the drums. Filter separator tanks were also located on-site, sampled, and determined to contain lead (Pb), but the sample amount consumed the entire liquid product in the tanks; therefore, the tanks were assumed to be clean and were removed, dismantled, and taken to Borough Landfill. However, some workers suspected that metal contamination (at least Pb) could be problematic on-site given the nature of the filter separator tanks.

Also during the October 1998 excavation effort, the Bristol Environmental and Engineering Services Corporation crew sampled and analyzed the soil for petroleum-derived contamination and found the presence of GRO (ranging from below detection to 74 mg/kg), DRO (ranging from 570 mg/kg to 13,200 mg/kg), RRO (ranging from below detection to 1350

mg/kg), xylene (ranging from 0.0554 mg/kg to 2.16 mg/kg), and Pb (ranging from 5.08 mg/kg to 624 mg/kg). However, all soil samples analyzed were below the detection limit of the analytical method for benzene, ethylbenzene, and toluene. When the on-site transformers were sampled, Bristol Environmental and Engineering Services Corporation determined that two of the thirteen transformers contained detectable PCB. Previous sampling of three different transformers on-site also showed elevated concentrations of PCB. Therefore, of the overall 16 transformers on-site, five contained PCB. Concentrations of PCB were determined to range from 1 mg/kg to 24 mg/kg, and one of the transformers containing PCB was located on the roadside shore of Imikpuk Lake. However, during inspection before removal, the crew noted that all of the transformers were sealed and that there was no evidence of leaking or spills near any of the transformers.

Overall, the October 1998 excavation effort by Bristol Environmental and Engineering Services Corporation removed four aboveground storage tanks with a total volume of 1000 L of liquid, sixteen transformers, four buildings, and 122 × 122 m of surface debris. Later, in August 1999, Bristol Environmental and Engineering Services Corporation returned and removed 188 m of purged pipe that had been abandoned in the area, more surface debris, and a tank that was located in the Middle Salt Lagoon. They removed the tank by rowing an inflatable raft to the tank and tying a rope around it for towing to shore. Unfortunately, the Lagoon tank had drained by the time it reached the shore. The tank was crushed and taken to the borough landfill, and the contents were never determined. One final analysis effort of the soil in the area surrounding Middle Salt Lagoon was to dig soil pits and to test the soil for total petroleum hydrocarbons in August 1999. The results from this extensive sampling showed that there were total halogens ranging from 104 mg/kg to 191 mg/kg and total petroleum hydrocarbons ranging from below detection of the analytical method to 2260 mg/kg for one sample (Bristol Environmental and Engineering Services Corporation 1999).

In 2000, oil was recovered during two separate recovery collections (1300 L and 900 L of oil), and Foster Wheeler Environmental Corporation extended the subsurface containment berm 67 m to provide increased protection from contaminated groundwater migrating into Imikpuk Lake (NAVFAC NW 2001). In September 2000, 135 m³ of soil contaminated with polycyclic aromatic hydrocarbons (PAHs) were removed. Then in August 2002, 1734 m³ of GRO- and DRO-contaminated soil were removed

and treated on-site using Hot Air Vapor Extraction (HAVE) technology by AGVIQ Environmental Services, Inc. (AGVIQ 2004). Additionally, after the soil extraction in August 2002, the crew placed a 0.3 m thick soil cap over the contaminated sediments in the South Depression.

In June 2003, Bristol Environmental and Engineering Services Corporation investigated soil located beneath a portion of the Old Power Plant building at the former NARL site and determined that it was contaminated with petroleum. Specifically, the analyses indicated that RRO were present in soil in two of the seven locations sampled; and concentrations in these two samples were above 22,000 mg/kg. The results also indicated that RRO concentrations ranged from 22,000 to 32,000 mg/kg in two of the seven locations sampled. The crew did, however, determine that concentrations of volatile organic compounds (VOCs), PAHs, PCB, and metals were all below the detection limit of the analytical method in all seven locations. In August 2003, the previously reported contaminated soil was remediated by Bristol Environmental and Engineering Services Corporation. Soil remediation included removing the petroleum contaminated soil to a depth of 0.5 m below ground surface (where permafrost was encountered) and an area of approximately 2.4×2.4 m for each separate location. The crew noted that the soil was “visually” and “obviously” contaminated, and the soil was excavated and turned over to the U.S. Navy for disposal. The soil excavation pits were loaded with clean, imported backfill by the crew (Bristol Environmental and Engineering Services Corporation 2003).

The U.S. Navy has conducted long-term monitoring (LTM) at the former NARL site for many years with particular focus on the Airstrip, Powerhouse, and former BFTF. The first U.S. Navy LTM project at the three sites was performed in 2003. The U.S. Navy has published multiple monitoring plans over the years, which identify the media to be sampled, the frequency and locations of sampling, the analytes to be sampled, and the criteria for the sampling program (NAVFAC NW 2013a). The first comprehensive monitoring plan was published in 2004. In addition, the U.S. Navy annually monitors the site and produces annual monitoring reports that typically document the current contaminant levels in the soils, sediments, and active-zone water (i.e., groundwater) and surface water; evaluates the progress towards achieving cleanup levels; assesses whether natural attenuation is occurring; and reports ongoing protections in place at Imikpuk Lake. Finally, the U.S. Navy produced two Five-Year Reviews, in 2008 and

2013 that present cumulative and comprehensive results and recommendations for the three target locations at the former NARL site (NAVFAC NW 2013a).

The U.S. Navy has stated that there may be permafrost and localized pockets of free product (“hot spots”) in the soil that were not previously removed that may be contributing to increasing groundwater concentration at the Airstrip, Powerhouse, and BFTF. With this statement came proposed actions necessary to protect human health and the environment. These actions included performing permafrost depth studies at the three sites of interest (Airstrip, Powerhouse, and BFTF), enhancing the LTM projects, reevaluating the soil cleanup levels, assessing the feasibility of implementing additional clean-up actions (particularly at the Airstrip and Powerhouse sites), and treating additional soil located at the former BFTF site (NAVFAC NW 2013a).

A variety of additional research-based studies have included the former NARL site (Braddock and McCarthy 1996; McCarthy and Solin 1995; Braddock et al. 1997; Baldwin et al. 2000; Walworth et al. 2001; McCarthy et al. 2003). The results and comprehensive approach presented in this case study highlight the work done by CRREL, part of the U.S. Army Corps of Engineers (USACE) Engineer Research and Development Center (ERDC), from 2014 to 2017 and include five basic components: (1) permafrost and active-layer characterization, (2) soil thermal-regime quantification, (3) groundwater modeling, (4) PHC characterization, and (5) bioremediation of petroleum-derived contamination.

2.3 Primary contaminants of concern

The primary PHC contaminants that are of concern at the former NARL site include GRO, DRO, RRO, PAHs, and VOCs (further outlined in Table 1). Specifically, GRO and DRO refer to hydrocarbons with boiling-point ranges corresponding to those of gasoline (C_6 to C_{10-12}) and diesel fuel (C_{8-12} to C_{24-26}), respectively. Since boiling point is dependent on molecular weight and chemical structure, the boiling-point range for GRO is approximately 60°C to 170°C while DRO range from approximately 170°C to 430°C . Jet fuel, like what makes up a large portion of the spills at the former NARL site, are light petroleum distillates (often composed of GRO, DRO, and RRO) that are used in jet engines. Typical hydrocarbon chain lengths for jet fuel vary from C_4 to C_{16} and the hydrocarbon structures con-

sist primarily of straight and branched alkanes and cycloalkanes. The major jet fuels used in military aircraft are referred to as JP-4, JP-5, JP-6, JP-7, and JP-8 (ATSDR 1999). The carbon content of RRO is larger than that of GRO and DRO. Typical carbon range for RRO is between 25 and 36 carbon molecules (Geosphere 2006).

Table 1. Descriptions and chemical characteristics of primary contaminants of concern at the former NARL complex.

Name	Acronym	Carbons (#)	Structure	BP (°C)
Gasoline range organics	GRO	6–12	Straight alkanes, branched alkanes, cycloalkanes	60–170
Diesel range organics	DRO	8–26		170–430
Residual range organics	RRO	25–36		400+
Volatile organic compound	VOC	<4		50–100
Polycyclic aromatic hydrocarbon	PAH	12–36	Multiple aromatic rings bonded in a linear or angular cluster	variable

Aromatic hydrocarbons tend to produce smoke when burned and are only reported to account for approximately 20%–25% of jet fuel (ATSDR 1999). In the history of NARL, PAHs were found in some of the soil and liquid samples during remediation and excavation projects. PAHs are considered toxic, carcinogenic, and mutagenic and can persist in the environment for long periods of time. PAHs are hydrocarbons like GRO, DRO, and RRO; but they are composed of multiple aromatic rings, typically two or more benzene rings bonded in a linear or angular cluster. PAHs have high melting and boiling points, low vapor pressure, and very low aqueous solubility, translating to very high persistence in groundwater (Abdel-Shafy and Mansour 2016).

The VOCs typically contain hydrocarbons with four or fewer carbon molecules in each chain. Smaller carbon chains that exist in VOCs have a higher toxicity for acute inhalation exposure because they are more easily volatilized (ATSDR 1999). This is particularly important for fresh petroleum spills with high surface area because the susceptibility for human and animal exposure is extremely high. Since the type of hydrocarbon (alkanes, cycloalkanes, alkenes, and aromatics) and size (number of carbons) will dictate the analytical method used for characterization, it is important to understand not only how many carbons are involved in bonding but also the bonding structure. Furthermore, it is essential to understand the size, distribution, and structure of the PHC (in the case of NARL, GRO, DRO, RRO, PAHs, and VRO) because the potential health (route of exposure)

and environmental impacts (mobility and transport) are dictated by concentration and speciation, among other factors. It is important to note that environmental factors such as evaporation, biodegradation, and co-contamination can complicate detailed analyses and characterization of the specific petroleum-derived contaminants of concern; therefore, detailed analyses must be planned and implemented carefully to ensure adequate monitoring and effective site-specific remediation.

2.4 Landscape of the surrounding area

Point Barrow lies at the northern tip of Alaska and is the northernmost point of U.S. territory. The NARL site is approximately 10 km southwest of Point Barrow and 6 km northeast of the town of Utqiagvik. The former NARL complex covers approximately 14 km² and contains a 1.5 km Airstrip. The Powerhouse site is located southwest of the Airstrip site, and the BFTF is located at the northernmost end of the former NARL complex and northeast of the Airstrip and Powerhouse sites. The facility is bordered by the Chukchi Sea to the west (Figure 1). Bjella (2012) provides further details.

2.4.1 Climate

The climate at the former NARL site has been described as “cold, windy, and generally inhospitable” and is often a limiting factor on operating equipment for research activities. In the summer, precipitation can be characterized as numerous light showers or heavy mists; in the winters, the majority of precipitation is snowfall (Reed and Ronhovde 1971).

2.4.2 Vegetation

The former NARL site lies in the Arctic coastal plain region. While there is little vegetation located directly on-site due to the presence of a man-made gravel pad, the primary vegetation is tundra. The surrounding area is blanketed with sphagnum mosses, grasses, sedges, and lichens. The northern limit of the spruce tree is within the Brooks Range; therefore, there are little to no trees in this area besides sparse, small willows, small alders, and cottonwood varieties (Reed and Ronhovde 1971).

2.4.3 Geology

The former NARL site (and the town of Utqiagvik) lies within the Arctic Coastal Plain; and the primary geologic composition is marine and non-marine deposits, consisting of unconsolidated silts and sands with some

clays and gravels (Black 1964) also with shale, mudstone, and sandstone (Payne et al. 1951). In contrast, the surrounding area adjacent to the former NARL site is underlain by fine-grained, organic-rich soils with high soil moisture, compared to areas with high gravel and sand content (McCarthy and Solin 1995; Wagner et al. 2016a). The area is in the continuous permafrost zone of Alaska, and permafrost has been estimated in places to be as much as 400 m thick or more (Reed and Ronhovde 1971). The land surface is dominated by buildings and gravel roads, and the annual thaw depth was estimated by McCarthy et al. (1994) to range from 1.0 to 1.5 m. Annual thaw depths in the surrounding areas (Imikpuk Lake) are less (~0.5 m) due to increased vegetation acting as an insulator and the high moisture content of the soils.

The dominant ground features associated with areas in the Arctic zone include permafrost, the seasonally thawed active layer, matrix ice, segregated ice, wedge ice, and taliks. The permafrost in this area was formed epigenetically and is composed of soil and ice. The active layer is the portion of the ground that undergoes an annual freeze–thaw cycle and the extent to which the active-layer thaws is dependent on a variety of factors, including snow coverage, extent of surface and shallow subsurface organic layers, soil conditions, and local impacts of ground engineering. The boundary between the bottom of the active layer and the top of the permafrost table is referred to as the transition zone and often includes segregation ice (Shur et al. 2005). Matrix ice can form when interstitial water freezes within sediment and fractured rock, typically occurring in fine-grained soils more often due to their containing a higher portion of water at saturation. Finally, ice wedges can be several meters in circumference and are formed as a result of surface water infiltrating surface cracks in the ground and propagating downward for many years. The formation and propagation of the dominant features of permafrost soils in Arctic coastal regions can often be viewed from the ground surface due to their exhibiting polygonal ground features (Bjella 2015; Bjella and Gelvin 2016).

2.4.4 Hydrology

The area surrounding the former NARL site features distinct ocean shorelines, sand spits, lagoons, and sea ice. The area can be characterized by low topographic relief, intricate water tracks, numerous meandering streams, and thousands of lakes and swamps that are frozen in the winters and swampy in the summers (Black and Barksdale 1949; Hussey and Michelson 1966; Black 1969). The Beaufort Sea and the Chukchi Sea lie to the

northwest and northeast of Point Barrow, respectively. In the summer, there is often standing water, and the land is swampy, wet, and poorly drained (Ugolini et al. 1982). The region has three defining surface-water drainage basins: the northwestern part drains to the Arctic Ocean, the southwestern part drains to Imikpuk Lake, and the eastern part drains to North Salt Lagoon (Braddock and McCarthy 1996).

Hydrologic processes at the former NARL complex are largely controlled by the short annual thaw season and presence of near-surface permafrost, which can restrict groundwater movement in the shallow subsurface and to the extent of the permafrost depth. In addition, the abundant presence of ice-wedge polygons affects surface water flow to outlying regions. Thaw season (early/mid-June to August/September) causes increases in runoff for short periods of time and translates to shallow and transient groundwater tables (McCarthy et al. 1994; McCarthy and Solin 1995). The groundwater flow systems in the surrounding area have been described as complex due to variations in local land-surface topography. McCarthy et al. (1994) estimated the groundwater flux from the Airstrip site to Imikpuk Lake to be approximately 173 m³ per year, which accounts for less than 0.1% of the estimated annual inflow to Imikpuk Lake (300,000 m³/year), indicating groundwater flow in the subsurface is likely minimal and restricted. Annually, the majority of the area experiences freezing conditions, and streamflow is minimal or does not occur at all. Therefore, hydrological processes are primarily controlled by accumulation of snow, sublimation from the snowpack, and redistribution of soil moisture (McCarthy et al. 1994; Braddock and McCarthy 1996).

2.4.5 Chemistry and biochemistry

The groundwater in Utqiagvik has a high suspended solids content as a result of the abundance of organics, fine-grained soils, sands, and gravels in the area. Solutes and suspended solids have been shown to accumulate at the boundary between the permafrost and active layer as a result of differences in redox conditions and physical gradients between the active layer and the ice-rich surface of the permafrost table (McCarthy and Solin 1995; Kimbrough et al. 1999; Barker et al. 2014). The abundance of dissolved minerals and solutes can depress the freezing point of water and, in conjunction with the presence of fine-grained soils that often already contain unfrozen water, can cause taliks to form in the subsurface (Osterkamp and Burn 2002). The presence of pure-phase hydrocarbons in soil pores can also cause chemical taliks to form in addition to causing an abundance of

discrete fluid-filled pockets in soil pores characterized by variable freezing-point gradients (McCarthy and Solin 1995; McCarthy et al. 2003).

Ice-rich permafrost often present at the boundary between the permafrost and active layer provides a physical and redox gradient that has been shown to increase contaminant transport and groundwater flow beyond what is typically predicted by migration models using diffusive transport alone (up to four orders of magnitude) due to the presence of ice lenses and perched veins greatly increasing lateral hydraulic conductivity (Dyke 2001). This process (increased hydraulic conductivity as a result of freeze-thaw cycles in the active layer) is particularly accelerated when a density or elevation gradient is present. This has important implications for contaminant transport at the former NARL site where there is a clear and visible gradient between the natural environment and the man-made additions, such as the Airstrip and gravel pads.

Chemically, the surface and subsurface soil layers are composed of naturally occurring minerals, precipitates, and organic matter; and detailed investigation of the overall composition contributes to understanding contaminant fate and transport and the ability to remediate a given site. The presence of nitrates, ferric iron, and sulfates at the former NARL site serves as electron acceptors for microbial growth. Additionally, the presence of nitrates provides a secondary source of nitrogen for the microbial community. Braddock and McCarthy (1996) observed iron staining at groundwater discharge areas, suggesting biodegradation using iron as an electron acceptor was naturally occurring. Braddock and McCarthy (1996) also reported an active microbial community (divided into three groups: crude oil emulsifiers, gasoline degraders, and total heterotrophs) in soils adjacent to Imikpuk Lake (adjacent to NARL, Figure 1) but found that microbial processes were likely limited by the short annual thaw season. Higher concentrations of microorganisms were found in water draining contaminated sites than in control sites with no contamination. This finding was supported by the observation of higher concentrations of ferrous iron and sulfide in the contaminated wells in comparison to the control wells, which strongly supports the notion that in situ microbial processes are actively occurring. Braddock and McCarthy (1996) reported that total heterotrophs ranged from 10^6 to 10^7 cells/g in soil, biological population that could demulsify crude oil ranged from 10^5 to 10^7 cells/g soil, and the gasoline degrader populations were estimated at 10^4 to 10^5 cells/g soil.

2.5 Impact to local community

Imikpuk Lake (Figure 1) was previously used as one of the primary drinking-water sources for the UIC-NARL facility (McCarthy et al. 1994). The UIC-NARL facility currently obtains drinking water from the Isatkuak Reservoir, the City of Utqiagvik's primary drinking-water source. Currently, local hunters use the water and ice from Imikpuk Lake for drinking and cooking water on hunting trips. The proximity of Imikpuk Lake to the former NARL site and the fact that the Airstrip is a large part of the lake's drainage basin leaves it vulnerable to potential contamination as a result of surface runoff and groundwater leaching from the repeat fuel spills to the surrounding area. As a result, baseline human health and ecological risk assessments have been conducted at two sites at the former NARL complex: the Powerhouse and the Airstrip. The two site-specific studies were conducted by Hart Crowser and prepared for the Department of the Navy, Engineering Field Activity, Northwest, Naval Facilities Engineering Command (now NAVFAC NW), in an effort to support the transfer of the NARL property to UIC. The main goals of the reports were to determine the potential adverse health effects as a result of residual exposure to PHC contamination and to determine the necessary remedial actions for the sites.

The Hart Crowser human health risk assessments found the lifetime cancer risks for potential future residential use at the Airstrip are above the Alaska Department of Environmental Conservation (ADEC) cancer risk management level, assuming reasonable maximum exposures (RME). Assuming RME for a recreational/subsistence user at the Airstrip site, the estimated cancer risks were equal to the cancer risk management level set by ADEC. The risk assessment study done for the Powerhouse Site estimated that an unacceptable cancer risk from residual contamination is unlikely. However, the study concluded that for the residential and construction-worker scenarios, the risk of cancer exceeds the ADEC hazard indices; but for the industrial worker exposure and subsistence hunter scenarios, the cancer risk is below the management threshold set by ADEC (Hart Crowser 2000a, 2000b). As a result of the risk assessment study, remedial actions were conducted at the Airstrip and Powerhouse sites, including soil excavation and treatment and monitoring of current contaminant levels in soils, sediments, active zone water, and surface water.

3 Methods

Figure 2 outlines the timeline of the individual activities, sampling campaigns, and field efforts carried out by the CRREL team for NAVFAC NW for the comprehensive assessment, monitoring, and remediation of PHCs at the former NARL.

Figure 2. Infographic timeline highlighting the major tasks completed by the CRREL team for the overall assessment and monitoring of the former NARL site in Alaska.

3.1 Nondestructive geophysical tools for Arctic surface and subsurface soil characterization

3.1.1 Ground-penetrating radar

The dual presence of ice and water in soil systems containing permafrost can affect overall contaminant transport and impact the efficacy of remediation techniques. Permafrost soil systems are often heterogeneous in terms of frozen versus thawed soils, interstitial water content, redox environment, and locale of segregation ice and ice wedges, which creates a wide array of chemical gradients. Furthermore, Arctic soils are delicate ecosystems that not only require care when sampling but are also often remote and as a result can present difficulties when characterizing the distribution of water and ice in the subsurface soil. Therefore, the implementation of noninvasive permafrost mapping techniques is ideal for Arctic regions because they offer a nondestructive means of mapping the subsurface. GPR transmits high-frequency radio waves on the order of 10 MHz to 4 GHz into the subsurface and records the reflections of the transmitted waves (Bjella et al. 2015; Arcone and Bjella 2016). Based on the differences

in the velocities of the reflected radio waves, visible electrical phase changes (waveforms) can be distinguished, which are due to differing dielectric permittivity from one substance to another. The physical difference between water and ice provides a detectable dielectric contrast, making this technique especially suited for permafrost soils. The GPR technique is particularly useful when distinguishing segregation ice at the bottom of the active layer in comparison to water that can often pool on the top of the permafrost table (Bjella 2015). The ability to distinguish between segregation ice and pooled water is important for analysis of soils at the former NARL complex because previous monitoring reports for the site noted the distinct presence of pore water and PHC fluid-filled pockets in the subsurface (McCarthy et al. 2003; NAVFAC NW 2013b).

This study highlights GPR results and the efforts to characterize the subsurface soil system and to determine whether the containment berm that was installed to prevent free PHC product from migrating into Imikpuk Lake was effective. To characterize permafrost site parameters, to determine the efficacy of the previously installed containment berm, and to characterize the soil and subsurface soil systems at the former NARL complex, the CRREL Alaska Research Office performed GPR surveys orthogonal to the berm every 15 m for the entire length of the containment berm in 2014. Additionally, the CRREL team completed two longitudinal GPR surveys in opposite directions directly over the axis of the berm for the full length. The CRREL team completed additional surveys across the Airstrip and Powerhouse sites in 2015. All surveys were conducted with either the 400 or 200 MHz center frequency antenna, but the 400 MHz antenna generally produced better visual resolution of the areas of interest. Additionally, a high-density polyethylene sled packed with plastic Bubble Wrap was used so the antenna would slide along the surface of the ground in a smooth, consistent manner. Calibration of the GPR was accomplished by frost-probing support, which provided a known depth to an object visible in the GPR images. The dielectric constant was adjusted based on the depth of the object in the image to the correct scale, which calibrated the travel time of the radar signal. The GPR data was processed and normalized in both the horizontal and vertical dimensions using Radan 7 software manufactured by GSSI Inc. (Nashua, NH), and the gain was adjusted across the depth of the survey. The data was stacked and compressed into shorter horizontal frames, and the transmitted reflections were filtered.

Depth profiles were corrected using the Kirchoff method of hyperbola migration (GSSI, Inc.). Mapping transect locations was accomplished with a hand-held Global Positioning System (GPS) (Bjella 2015).

3.1.2 Electrical resistivity tomography

Constraining frozen versus unfrozen soil and water compartments in surface and subsurface soil systems is an integral first step for monitoring and assessing contaminated sites in the Arctic and characterizing soil systems. Frozen material in the earth is resistive to electric current flow, and resistivity has been shown to change between frozen and thawed ground and ice-rich to ice-poor terrain (Wagner et al. 2016a). For this study, electrodes that transmit current were used to induce current into the subsurface at the former NARL complex; and the resultant voltage was read using the Advanced Geosciences, Inc., Super Sting R-8 ERT system (Austin, TX). The apparent resistivity over a range of depths was calculated by averaging algorithms, and the data was processed using the RES2D software package (Geotomosoft Solutions, Malaysia).

3.1.3 Capacitively coupled resistivity

To measure the resistivity of the subsurface material and to map the distribution of permafrost features at the former NARL complex, this study also employed CCR using the Geometrics OhmMapper TR5 (San Jose, CA). Dipole cable lengths were primarily 5 m with a separation length of 5 m. The RES2D software was used to process the collected data, and the repetitive transects were similar to the survey described in section 3.1.1. This report does not explicitly show the results from the CRREL team's CCR study at the NARL site, but these can be found in the Wagner et al. (2016a) report.

3.1.4 Thermal analysis and modeling

Interactions between the environment and climate in the Arctic are often complex and require detailed monitoring to understand the soil thermal regime in relation to the changing seasons. For example, snow has been shown to provide a thermal barrier insulating the soil, which causes decoupling between the air temperatures and ground temperatures. Understanding the thermal properties of the former NARL complex was an integral step in assessing where treatment locations for PHC remediation might be most effective given the environmental conditions.

Soil temperature was monitored at eight locations within the Airstrip and Powerhouse sites using TMC20-HD sensors coupled to 1 in. PVC pipe inserted into the ground using a Geoprobe 6610DT. The aboveground cable was protected from disturbance by using a flexible aluminum conduit, and the logger housing consisted of a Pelican Case enclosure. Meteorological measurements were collected from September 2015 to April 2016 using a 3 m tall station outfitted with a Campbell Scientific CR1000 logger that recorded air temperature, wind velocity and direction, relative humidity, and snow depth every 15 minutes. Snow coverage at the former NARL complex was further investigated using a snow fence, which was installed in September 2015 and consisted of plastic fencing, wood cross-beams, and metal fence posts (approximately 1.4 m tall). The CRREL-installed snow fence was placed adjacent to the meteorological station at the Airstrip site. There was an existing snow fence (40 m long and 2 m tall) installed at an unknown time at the Powerhouse site (30 m west of Imikpuk Lake) that was also monitored during this study. Additional snow depths and snow density measurements were collected using a mapping-grade GPS-enabled Magnaprobe (procedural method further outlined in Sturm 2009) in-line with a Campbell Science CR-800 Datalogger coupled with manual snow water equivalence (SWE) measurements. The SWE measurements were accomplished by collecting a column of snow with a known height and placing the snow column into plastic bags with Federal Sampler snow tubes and subsequently massing the melted water.

Modeling of the thermal soil regime at the former NARL complex (including snow depth thickness, snow coverage, snow rate accumulations, and increased air temperature scenarios) was performed at seven of the eight thermistor sites by using a two-dimensional finite element thermal model (SVFlux module of Soilvision, Saskatoon, Canada). Model simulations were varied for sensitivity by changing snow-depth thickness, snow accumulation rates, snow arrival, and air temperature increases.

3.1.5 LIF-UVOST

In situ and nondestructive determination of PHC distribution can be achieved by laser-induced fluorescence (LIF), which is a powerful tool for monitoring and assessing a contaminated site. The LIF instrumentation uses a narrow wavelength of electromagnetic radiation that is focused into fiber optics, allowing for a direct and powerful energy emission. Coupling LIF with the Ultra-Violet Optical Screening Tool (UVOST) allows for the

direct emission of ultraviolet light into a substrate. PHCs that contain aromatic chains are highly fluorescent, and any in the substrate will absorb the ultraviolet light and become excited. Once excited, the PHCs will relax back to a lower energy state and release energy in the process. The energy is measured with a detector and is used to determine (in real time) the distribution of various PHCs present at a contaminated site. LIF-UVOST has the capability to differentiate between PHC types based on their unique spectral pattern. This tool has the potential both to be noninvasive to the soil system structure at a site and to yield powerful results in real time, which are necessary to limit cost and response time of remediation strategies for larger spills.

For this study, the LIF-UVOST (Dakota Technologies, Fargo, ND) survey was completed by GeoTek Alaska Inc. (Anchorage, AK) at the former NARL complex, including the Airstrip, Powerhouse, and BFTF. UVOST probing was accomplished using a grid pattern, and data was modeled using Esri ArcGIS Desktop Version 10.2.2 with inverse distance weighting, further described in Wagner et al. (2016b). Additional surveys were conducted on each of the UVOST probe sites using a Trimble R8 survey-grade GPS. The data was processed within the WGS 84 UTM Zone 4N coordinate system, and the vertical control datum NAVD88 was used to establish ground elevations.

3.2 In situ hydro-biogeochemical methods for monitoring contaminant transport and constraining site characteristics

3.2.1 Frost-probing

Determining the phase distribution of water in soil is important for a variety of reasons, including engineering issues, construction concerns, agricultural purposes, and contaminant fate and transport. Frost-probing has been used by numerous studies and provides support to GPR for the in situ determination of the depth of the active layer (Hayhoe et al. 1986; McFadden and Bennett 1991). Frost-probing was carried out at the former NARL complex as an in situ ground-truthing method to support the results obtained by GPR. A 1–2 cm diameter pointed, graduated, steel rod with a handle was forced into the ground to the point that the rod would not penetrate further with additional force, and the soil depth was marked as the top of the permafrost table. Frost-probing also verified the existence of plywood installed during the containment wall construction. Probing

was performed in August 2014 and 2015 near the end of the thawing season when the active-layer thickness was likely at its deepest yearly extent. Mapping transect locations was accomplished with a hand-held GPS.

3.2.2 Drilling boreholes

Assessing permafrost conditions manually is often a necessary addition to remote noninvasive techniques like GPR and ERT to ground-truth the results. For this study, CRREL subcontracted drilling services with GeoTek Alaska, Inc. (Anchorage, AK), to drill and sample five exploratory borings distributed between the Airstrip and Powerhouse sites. The boreholes were drilled using a track-mounted Geoprobe 6620DT drill rig equipped with the Geoprobe 5.7 cm Macro-Core MC5 continuous soil sampling system and a 10.8 cm internal diameter continuous flight hollow stem auger. The collected soil samples were sealed in airtight containers and were transported to the CRREL Alaska Research Office laboratory and classified using the Unified Soil Classification System. Soil samples were analyzed for soil salinity by placing 50 g dry soil into a 200 mL glass beaker, adding 200 mL distilled water, stirring the slurry for 2 minutes, allowing slurry to settle for 1 hour, and analyzing with a Hach H170G conductivity/salinity meter (Hach Company, Loveland, CO).

3.2.3 Hydrological groundwater modeling

Groundwater elevation levels were determined using existing monitoring wells installed by Sealaska Environmental Services, LLC, which is the Navy's long-term monitoring contractor in Utqiagvik, Alaska. Additionally, CRREL installed nine new monitoring wells to track water elevation. A total of 45 groundwater monitoring wells were included in the manual water elevation portion of this study with 27 wells located at the Airstrip site and 18 wells located at the Powerhouse site. For the installation of the wells drilled by CRREL, a hydraulic earth drill (Little Beaver HYD-TB11H, Livingston, TX) was used; and each new well was surveyed using a Trimble R8 survey-grade GPS with reported positional accuracy of less than 2 cm. Data was processed using WGS 84 UTM Zone 4N, and the water elevations were based on the NAVD88 vertical datum (Wagner et al. 2016b). A total of eight groundwater-level loggers (Solinst, Georgetown, Ontario, Canada) were used for continuous water-level measurements. Each continuous logger was corrected to the barometric pressure by using a barometric logger also placed in a well on-site.

Hydrological modeling was performed for steady-state groundwater migration for areas at the former NARL complex, including the Airstrip and Powerhouse sites, by using the finite element SVFlux module (SVOOffice module of Soilvision, Saskatoon, Canada) and a grid size of 2 m and 1 m. Surface and top of the permafrost table elevations were set as boundaries for the model and were estimated previously (Wagner et al. 2016a).

3.2.4 Tracer-dye studies

A variety of studies have used tracer dyes to determine hydrological flow paths and to establish the likelihood of various migration patterns for contaminant plume concentrations. In the current study, four tracer dyes were introduced at the Airstrip site, and three tracer dyes were introduced at the Powerhouse site. The tracer dyes were deployed in monitoring wells that had been previously installed by Sealaska Environmental Services and nine monitoring wells that were installed by CRREL in June 2016. The dyes used were Rhodamine WT, Eosine, Fluorescein, and Sulforhodamine B. The Sulforhodamine B dye was not used at the Powerhouse site, but all four dyes were deployed at the Airstrip site. The Rhodamine dye was the only dye already a liquid and was flushed down with 19 L of filtered water whereas the other three solid dyes were dissolved in 15 L of filtered water and thoroughly mixed in a carboy and flushed down with an additional 6–19 L of filtered water. Each of the dyes was deployed in the monitoring wells by using a large plastic funnel.

Carbon sample packs were placed in the wells at various times to sample the dye concentration. Additionally, 15 mL of groundwater from the monitoring wells were collected during each sampling event by using a disposable bailer dedicated to each individual well, and background samples were collected from the wells before tracer dyes were introduced. For instance, when there was limited or no water in the wells, there were no samples collected. Ozark Underground Laboratory analyzed all samples.

3.3 Bioremediation

3.3.1 Soil properties and existing microbial activity

Soil samples were collected in August 2015 from the Airstrip and Powerhouse sites at the former NARL complex to investigate soil properties (gravimetric water content, pH, percent carbon, and percent moisture) and the extent of microbial activity as a function of location (upgradient

and downgradient of groundwater flow direction at the site). Soil was collected by first using a gas-powered hydraulic earth drill (Little Beaver HYD-TB11H, Livingston, TX) to drill a hole directly above where the sample was to be collected, and then a hand-operated auger with removable plastic sleeves was used to actually sample the soil. Soil samples were placed on ice and stored at -4°C until analysis.

Gravimetric water content of the soils collected from the Airstrip and Powerhouse sites was analyzed by massing the soil sample at room temperature and subsequently massing the same sample again after heating in an oven at 105°C for 24 hours and allowing it to cool back to room temperature.

The soil pH was measured by allowing 10 g of air-dried soil to equilibrate with 10 mL deionized water (10 minutes) and measuring pH using a probe (Hanna Instruments, Woonsocket, RI). The soil pH in a dilute salt solution was also measured to stabilize the pH readings for comparison during other sampling events. For this, 0.1 mL of 1 M calcium chloride solution was added to the equilibrated soil slurry, mixed, and allowed to equilibrate (10 minutes), and pH was measured again.

The carbon content in the soil samples was measured using the loss on ignition (LOI) method as described in Wright et al. (2008). The soil samples were placed in a muffle furnace at 360°C and heated for 2 hours and massed. The mass was again measured once the temperature dropped below 150°C .

Gas chromatography coupled to a flame ionization detector (GC-FID) was used to determine the extent of biological activity in 10 g of soil from the various sample sites at the former NARL complex by measuring the production of headspace carbon dioxide over a measured period of time. Soil samples were placed in glass vials, sealed using a silicone-lined septum and aluminum crimp cap, and incubated for 48 hours at 7°C .

The soils were analyzed for moisture content by massing 10 g of collected soil, were dried for 2 hours at 110°C , and were massed again once cooled to room temperature.

3.3.2 Determination of petroleum hydrocarbon fractions

The quantification and characterization of the PHC fractions present in the soils at the former NARL complex were carried out using a gas chromatograph equipped with a photo-ionization detector (GC-PID) and GC-FID in series. Soil samples were collected on-site in the same procedure outlined in section 3.3.1. Stainless steel tools cleaned with methanol were used to sample the soil. Soils were separated into samples for volatile petroleum hydrocarbon (VPH) analysis and extractable petroleum hydrocarbon (EPH) analysis. For VPH analysis, 5 g of soil were stored in a sterile 40 mL amber glass vial pre-preserved with purge-and-trap-grade methanol, whereas for EPH analysis, 25 g of soil were stored in a sterile 0.1 L amber glass jar and sealed. Samples for VPH and EPH were both covered with parafilm as a final step and stored on ice prior to being shipped to Fremont Analytical, Inc. (Seattle, WA), for analysis.

Fractions of VPHs detected in soil samples were analyzed using the purge-and-trap method and were compared to a standard containing known aromatic and aliphatic hydrocarbons by using the differences in the GC-PID and GC-FID detector responses. Concentrations of PHCs were subsequently divided into different aromatic hydrocarbon fractions within the C₈ to C₁₀, C₁₀ to C₁₂, and C₁₂ to C₁₃ ranges and aliphatic hydrocarbons within the C₅ to C₆, C₆ to C₈, C₈ to C₁₀, and C₁₀ to C₁₂ ranges, further outlined in Barbato et al. (2017b).

Fractions of EPHs were extracted into methylene chloride, dried over sodium sulfate, solvent exchanged into pentane and methylene chloride, and concentrated before separating into aliphatic and aromatic fractions using silica gel (modified from Environmental Protection Agency method 3630). A final volume of 1 mL extract was produced for both aromatic and aliphatic fractions, and both extracts were analyzed separately using GC-FID and compared to the aliphatic and aromatic standard. Concentrations of PHCs were subsequently divided into different aromatic hydrocarbon fractions within the C₈ to C₁₀, C₁₀ to C₁₂, C₁₂ to C₁₆, C₁₆ to C₂₁, and C₂₁ to C₃₄ ranges and aliphatic hydrocarbons within the C₈ to C₁₀, C₁₀ to C₁₂, C₁₂ to C₁₆, C₁₆ to C₂₁, and C₂₁ to C₃₄ ranges, further outlined in Barbato et al. (2017b).

4 Results and Discussion

4.1 Breach in subsurface containment berm and implications for PHC migration

The subsurface containment berm (finished in 1996 by Foster Wheeler Environmental Corporation) was a major installation designed to create a raised permafrost table that would effectively prevent free PHC product from migrating into Imikpuk Lake. As such, investigating the efficacy and design robustness of the subsurface containment berm was an important initial step for the CRREL team to establish site characteristics and to assess long-term PHC fate and transport. Probing the active layer by means of noninvasive geophysical tools to evaluate if the active-layer thickness was effectively decreasing and the permafrost feature was rising proved to be a successful methodology for this study, and the CRREL team was able to determine whether the containment berm was functioning as designed.

The GPR results obtained by the CRREL team showed the artificial containment berm was effectively raising the permafrost table along the survey transects explored but also showed that there was a detectable gap in the barrier of the berm. In design, the containment berm consists of two major features: (1) the permafrost ridge created by installing insulation near the ground surface and (2) a vertical geomembrane curtain wall that was embedded into the center of the permafrost ridge (Baldwin et al. 2000). Figure 3 is a schematic of the overall projected design of the subsurface containment berm. CRREL performed a GPR survey using transects that ran orthogonal to the berm every 15 m for the entire length of the berm for a total of 36 orthogonal transects. CRREL additionally completed two longitudinal surveys in opposite directions directly over the axis of the berm for the full length. Based on the GPR results, the containment berm was determined to be effectively raising the permafrost as a result of the design structure and likely has a significant impact on limiting the migration of PHCs into Imikpuk Lake. Figure 4 presents the major results from the GPR analysis and shows that the permafrost table is raised in either direction of the containment berm structure (Bjella 2015). Additionally, frost probing was used to verify the GPR reflections, and probing correlated well with the imaging (Bjella 2015).

Figure 3. Schematic showing the overall containment berm design that was installed by NAVFAC at the former NARL complex. Major features outlined in the figure were to create a raised permafrost feature to provide a barrier for subsurface flow of free PHC product and contaminated groundwater to Imikpuk Lake. Location of Imikpuk Lake is identified. Fig. adapted from Baldwin et al. (2000). Not to scale.

Figure 4. A GPR image showing that the permafrost table has been effectively raised on either side of the containment berm installation. The shown transect is north of Dew Line Road, and the right y-axis stops short of the shoreline of Imikpuk Lake. The image has been corrected for surface elevation changes, and frost probe depths are shown as *red dots*. The *thin, dashed yellow line* traces the inferred frost depth and the *thin, dashed red line* represents the thaw depth based on the frost probing. The *red arrow* is the insulation/plywood layer for the containment structure, and the *yellow arrow* indicates the location of the recovery trench. The *short, vertical white lines* mark 10 m intervals. The image was taken and adapted from Bjella (2015).

The most notable finding as a consequence of CRREL's GPR surveys was a breach in the subsurface containment berm, which likely has significant

impact on the overall efficacy of the berm at limiting free PHC product from reaching Imikpuk Lake. The long GPR transects above the axis of the containment berm showed the insulation/plywood layer was intact for the entire length. However, GPR showed that there was one area with a break in the containment berm structure (Figure 5), which was confirmed by frost-probing. The area where GPR found no insulation or plywood was also topographically depressed compared to the adjacent ground. The breach the CRREL team found in the subsurface containment berm likely plays a role in the overall migration of PHC to the recovery trench.

Figure 5. A GPR image showing the break in the subsurface containment berm starting at approximately 46 m and ending at approximately 55 m. The *green* and *red arrows* show transect locations, and frost probing (*red dot*) was conducted in the break area at a depth of 1.2 m. The *short, vertical white lines* mark 10 m intervals. The image was taken and adapted from Bjella (2015).

The results from CRREL's GPR and frost-probe efforts show that the containment berm is effectively raising the permafrost table but that there is a definite breach in the extent of that artificial barrier.

The containment berm (installed in 1996) should be continuously monitored for possible design failures or malfunction after decades of use. The permafrost and subsurface investigation done by the CRREL team (Wagner et al. 2016a) showed locally altered permafrost and ground thermal-regime conditions adjacent to Imikpuk lake, likely attributable to topographic and thermal changes to near-surface soils as a result of ponded water and thaw lake development, migration, and degradation. Additionally, distinct saline zones (areas containing higher or lower salt content) were found when soil cores from the Airstrip were investigated (results outlined in Table 2 for the Airstrip site). While the variability in the salt content in the soil cores is not drastic, variations in salt content could still play a role, particularly considering that the variations in salt content in the groundwater samples were orders of magnitude different (NAVFAC NW 2013a).

Table 2. Soil core analysis collected from the Airstrip site showing distinct saline zones and depths with substantially higher soil moisture content. Three cores were collected. The Unified Soil Classification System (USCS) was used to classify soil types. Data was taken and adapted from Wagner et al. (2016a).

Boring #	Sample Depth (m)	Classification	Moisture (%)	Salinity (ppt)
AS-B1	1.5–1.7	SM	38.7	0.2
AS-B1	2.2–2.3	SM	39.8	0.5
AS-B1	2.0–2.2	SM	145.6	---
AS-B1	3.4–3.6	SM	23.9	0.1
AS-B1	3.9–4.1	SM	22.2	1.2
AS-B1	4.9–5.1	SM	29.6	2.1
AS-B1	5.4–5.6	SM	31.2	0.3
AS-B2	1.8–2.2	GP	27.4	0
AS-B2	3.3–3.7	SP	19.5	0.1
AS-B2	2.8–3.2	SP	15	0.2
AS-B3	1.8–2.2	SP	96.4	---
AS-B3	3.1–3.5	SP	26.3	0.1

Similar soil salinity results were shown for the soil cores collected from the Powerhouse site (data shown in Table 3), indicating that soil salinity is variable as a function of soil depth. The presence of saline zones (Bjella and Gelvin 2016) in the subsurface (borehole soil analyses) could potentially result in the formation of thermal gradients because salinity tends to depress the soil freezing point, resulting in poorly bonded soil particles and behavior similar to thawed soil conditions (Andersland and Ladanyi 2004). Any further degradation to the containment berm could have an impact on overall PHC transport and subsequent runoff into Imikpuk Lake.

Table 3. Soil core analysis collected from the Powerhouse site showing distinct saline zones and depths with substantially higher soil moisture content. Two cores were collected. The Unified Soil Classification System (USCS) was used to classify soil types. Data was taken and adapted from Wagner et al. (2016a).

Boring #	Sample Depth (m)	Classification	Moisture (%)	Salinity (ppt)
PH-B1	0.6–0.8	SP	7.7	0
PH-B1	1.4–1.5	SP	11.6	0
PH-B1	1.5–2	SP	13.8	0.1
PH-B1	2.0–2.3	OL	106.2	0.6
PH-B1	3.2–3.5	SM	17.6	0.5
PH-B2	0.8–1.5	GP	18.4	0
PH-B2	1.8–2.1	SM	35.1	0.3
PH-B2	2.1–2.3	SM	68.5	1.6
PH-B2	2.4–2.6	SM	151.7	0.5
PH-B2	2.6–3.0	SM	28.7	0.5

4.2 Contaminant transport and mobility in the subsurface

The distribution of PHCs at the former NARL site was initially investigated by Sealaska Environmental Services and was subcontracted to Hammer Environmental to perform the LIF-UVOST survey in the summer of 2012, and CRREL investigated additional locations (subcontracted to GeoTek Alaska, Inc.) in summer 2015 to obtain site coverage in more detail. Specific investigation areas were divided into the Airstrip and Powerhouse sites. Major results can be found in Wagner et al. (2016b). The Airstrip site contained three major hotspot PHC locations primarily located on the western part of the Airstrip landing apron. From the Airstrip apron, concentrations of PHCs were found to primarily dissipate as a function of distance. There were multiple locations with detections of PHCs, including adjacent to Imikpuk Lake. Concentrations of PHCs were found to vary with depth at the Airstrip site.

The Powerhouse site contained relatively high concentrations of hotspot locations for PHC distribution. There were several areas in close proximity to the western embankment of Imikpuk Lake that contained relatively high concentrations of PHCs. Farther west from Imikpuk Lake, concentrations of PHCs decreased substantially before increasing closer to Stevenson Road and the city of Utqiagvik. In contrast to the Airstrip site, the depth of PHCs at the Powerhouse site tended to be closer to the surface to some extent, primarily above 1 m soil depth.

While timing of the petroleum spill and type of PHC that was spilled plays a role in the overall distribution of PHCs at both the Airstrip site and the Powerhouse site, there is the added influence of groundwater flow paths that can dictate distribution of PHCs at a given site. The CRREL team did hydrological modeling of major flow paths at the former NARL site after extensive manual and automatic water elevation surveys were completed using monitoring wells (Wagner et al. 2016b). A hydrological model was set up to estimate the groundwater levels at the Airstrip and Powerhouse sites for each day of manual measurements. At the Airstrip site, groundwater elevations decrease from north to south in the direction of Imikpuk Lake and towards the south depression to the east (Figure 1). At the Powerhouse site, groundwater elevations decrease from the Powerhouse building to the east in the direction of Imikpuk Lake. In general, the hydrological modeling effort determined that groundwater flow paths naturally flow towards Imikpuk Lake at both the Airstrip and Powerhouse sites, which has implications for the overall fate of PHCs at the former NARL complex.

The inevitability of groundwater containing PHCs reaching Imikpuk Lake was also supported by the findings in the tracer dye studies, which saw Imikpuk Lake visually containing tracer dye after introduction into a well at the Airstrip site (Wagner et al. 2018b).

4.3 Soil thermal regime and the effects of snow coverage and increasing air temperatures

Snow coverage is heterogeneous at the former NARL complex with depths ranging from zero to a couple of meters. Even small amounts of snow coverage provide thermal protection for tundra plants in the harsh winters and in the process shields the active layer from emitting as much heat as with zero snow coverage. Consequently, the thermal regime of the active layer and upper regions of the permafrost table are effectively decoupled from air temperatures in the Arctic. The results from the CRREL study show that the rate of heat transfer from the subsurface soil decreases as the amount of snow depth at the surface increases, to an extent. Wagner et al. (2018a) provide a detailed report of the results.

The snow coverage at the Airstrip and Powerhouse sites at the former NARL complex was widely variable, which according to the model simulations of soil temperature will affect thermal shielding of the subsurface layers. The results from this study suggest that investigating snow coverage has the potential to be used as a proxy for understanding soil temperatures at depth and that snow management may be an additional factor when deciding the best sites for bioremediation implementation.

4.4 Characterization and accumulation of PHCs at the former NARL complex

Determining major hydrocarbon constituents in soil and groundwater was an essential part of the CRREL team's study to understand the relative mobility and bioavailability of the petroleum contamination at the former NARL site. Specifically, understanding site-specific hydrocarbon fractionation is necessary to develop targeted remediation strategies. The chemical properties of hydrocarbons significantly affect behavior in the environment. The degree to which hydrocarbons can dissolve in water depends on whether the bonding structure is primarily straight chains (aliphatic) or rings (aromatic) and the number of carbons that contribute to bonding in the overall structure. Larger chains tend to be significantly less soluble

than smaller chains and can persist to a greater extent in contaminated areas. Therefore, a major portion of the effort by the CRREL team was (1) to classify the PHC pollutants at the former NARL complex into aliphatic or aromatic fractions and (2) to determine the numerical range of carbon chains. Organic contaminants like PHCs degrade into different fractions and partition in the environment into different phases, which is necessary information to determine overall mobility and bioavailability and to plan remediation strategies. To determine partitioning coefficients and the extent to which PHCs distribute in the environment, the relative PHC fractions must be determined in addition to soil parameters and associations between the in situ environment and the PHCs in soil. Barbato et al. (2017a) provides a detailed account of the baseline assessment of petroleum contamination.

Soil type and environmental characteristics (pH, soil moisture, and percent organic matter) play a role in the ability for PHCs to accumulate, become mobilized, and persist in Arctic regions. In general, pH at the former NARL complex was inversely proportional to soil moisture and percent organic matter in the soils collected from the Airstrip and Powerhouse sites. In general, soil samples collected from the upgradient sites contained higher amounts of both aromatic and aliphatic hydrocarbons than the downgradient sites, which was expected since the contamination sources of petroleum occurred within and adjacent to the upgradient areas at both the Airstrip and Powerhouse sites. The distribution of hydrocarbons was determined to be highly heterogeneous even within duplicate samples, indicating local source hotspots within the sample areas. The Powerhouse site exhibited higher variability between duplicate samples than the Airstrip site (Barbato et al. 2017a).

In general, increased environmental persistence of aromatic hydrocarbons has been shown to be directly related to size and angularity (Kanaly and Harayama 2000; Chikere et al. 2011). Additionally, high molecular weight aromatics have been shown to be considerably recalcitrant and exhibit slow rates of biodegradation (Atlas and Bartha 1998; Stroud et al. 2007). It is likely that the presence of organic matter increases the dissipation of PHCs at the former NARL complex, but the higher molecular weight and larger carbon chain fractions tend to be recalcitrant. The degree of persistence has yet to be determined, and further monitoring is needed to determine overall fate of PHCs.

The baseline results from the PHC characterization by the CRREL team hold promise for future work with bioremediation at the former NARL complex. Braddock and McCarthy (1996) previously documented the active microbial population at the NARL complex and determined that in situ biodegradation is naturally occurring. The results from Barbato et al. (2017a), in conjunction with the study done by Braddock and McCarthy (1996), suggest that bioremediation and specifically accelerating the effects of the natural microbial population (nutrient additions) would likely be effective methods for PHC degradation.

4.5 Bioremediation and factors affecting efficacy

Environmental constraints specific to Arctic environments (low temperatures, presence of permafrost, and low precipitation) present unique challenges for contaminant removal. In this study, the CRREL team investigated the bioremediation potential of PHCs at the former NARL complex to determine innovative remediation technology targeted for Arctic regions and necessary to reduce contamination (Barbato et al. 2017a). Highly contaminated areas at the former NARL complex sites (Airstrip and Powerhouse) were identified based on the 2012 and 2015 LIF-UVOST surveys (see section 4.2) as targets for high-resolution chemical analyses of the aliphatic and aromatic hydrocarbons. Furthermore, biological attributes such as soil respiration were tested to determine the activity of the soil microflora despite long-term exposure to the PHCs. The overall goal of this study was to determine the baseline conditions at the sites to down-select treatment technologies that would be successful at enhancing the biodegradation of residual PHCs to inform remediation efforts. Barbato et al. (2017b) provides a detailed report of the results.

5 Summary

Petroleum-derived contamination is an environmental concern particularly with respect to hydrocarbon partitioning to organic matter, soil layers, and groundwater. Potential migration of PHCs into local drinking-water sources necessitates a rapid and efficient response to limit subsurface movement off-site. Constraining and remediating PHC contamination in Arctic soil systems presents additional difficulties due to the harsh, often remote environments; minimal infrastructure; and logistical support that the Arctic provides. The case study presented in this report highlights the substantial and comprehensive effort that took place over four years to assess, monitor, and develop remediation strategies for PHC contamination at the former NARL complex. Particularly for sites with a history of repetitive spills like the former NARL complex, the need for a comprehensive approach is significant. By coupling (1) nondestructive geophysical tools for Arctic surface and subsurface soil characterization with (2) in situ hydro-biogeochemical methods for monitoring contaminant transport and constraining site characteristics, the CRREL team was able to give a detailed site assessment to a level that was lacking in previous studies on the former NARL complex. The approach documented by the CRREL team can be used in other areas with similar contamination issues and has particular application for Arctic regions.

The former NARL complex just outside of Utqiagvik, Alaska, has a history of land-based fuel spills. Particularly for sites with a history of repetitive spills and a variety of remediation efforts conducted over many years, the need for a first assessment of the site is paramount. By using noninvasive GPR and ERT techniques coupled with validation methods of frost-probing and boreholes, this study was able to determine the extent of active-layer thaw, the presence of ice wedges and polygonal permafrost features, distinct saline zones in the subsurface soil layers, and the presence of significant segregation ice at the permafrost–active-layer boundary (Shur et al. 2005). The presence of segregation ice, ice wedges, and distinct saline zones in the subsurface soil layers likely affects overall transport of PHCs with respect to the subsurface containment berm. Extensive analysis using noninvasive GPR probing of the subsurface detected a breach in the containment berm that was previously installed to limit the movement of groundwater to Imikpuk Lake by creating a barrier consisting of a raised permafrost table. Limiting PHC transport to Imikpuk Lake via groundwater is integral for the community of Utqiagvik. Therefore, a breach in the

containment berm is a significant finding that likely affects overall transport of PHCs toward Imikpuk Lake.

The transport of PHCs to Imikpuk Lake is further supported by soil characterization of PHC fractions near the lake, which showed the presence of heavy aromatics. The downgradient areas (closer to the lake) at the former NARL complex containing significantly less aliphatic and aromatic hydrocarbons than upgradient areas and the presence of heavy aromatics near Imikpuk lake is concerning. The comprehensive study done by CRREL determined that the distribution and transport of PHC at the former NARL complex is likely widespread and complex.

Characterizing the hydrocarbon fractionation and quantifying the distribution of PHCs at the former NARL complex is necessary for planning bioremediation strategies. By combining fractionation characterization and distribution with a snow coverage and soil thermal-regime investigation, the CRREL team was able to more accurately assess the potential success of implemented bioremediation efforts since snow coverage and soil temperature play a significant role in the overall rate of PHC biodegradation. Snow coverage was found to occur heterogeneously with depths ranging from zero to a couple meters, which was determined to play a substantial role in thermal protection for the subsurface soil regime and limiting the rate of heat transfer. Snow coverage, in general, was directly correlated with soil temperature; and the snow coverage between the Airstrip and Powerhouse sites at the former NARL complex was widely variable, in comparison. The results from this study suggest that investigating snow coverage in areas within the Arctic has potential to be used as a proxy for understanding soil temperatures at depth and that snow management should be included as an additional factor when deciding the best sites for bioremediation implementation.

Hydrological flow paths and movement of groundwater at the former NARL complex showed groundwater elevations at the Airstrip site decrease from north to south in the direction of Imikpuk Lake. Additionally, at the Powerhouse site, groundwater elevations decrease from the Powerhouse building to the east in the direction of Imikpuk Lake. In general, it was determined that groundwater flow paths are naturally flowing towards Imikpuk Lake at both the Airstrip and Powerhouse sites, which has implications for the overall fate of PHCs at the former NARL complex. The inevitability of groundwater containing PHCs reaching Imikpuk Lake was also

supported by implanting tracer dyes and monitoring migration, which yielded Imikpuk Lake visually containing tracer dye after introduction into a well at the Airstrip site. Future predictions for PHC migration and transport at the former NARL site suggest further migration and diffusion of the PHCs given the velocities of the tracer dyes and the results from the UVOST study, indicating concentrations of PHCs were present throughout the soil column and relatively widespread throughout the sites with notable hotspot locations. The results from the hydrological study suggest infiltration to the groundwater is a major factor contributing to the overall transport of PHCs at the former NARL site.

The laboratory baseline study of the former NARL complex yielded novel results pertaining to microbe-facilitated bioremediation and the genes present (Barbato et al. 2017a). The laboratory results showed that respiration rates, biological activity, and hydrocarbon degradation genes were present in the soils collected from the Airstrip and Powerhouse sites. The next phase of the study is to assess bioremediation potential in the laboratory by using plants and nutrients as stimulants of microbial degradation and then to scale it to the field.

6 Recommendation

The comprehensive approach tested by the CRREL team for this study proved to be integral in assessing and monitoring PHC contamination at the former NARL site just outside of Utqiagvik, Alaska. Coupling (1) non-destructive geophysical tools for Arctic surface and subsurface soil characterization with (2) in situ hydro-biogeochemical methods for monitoring contaminant transport and constraining site characteristics gives a detailed assessment of contaminated sites more so than other previous studies. Figure 6 presents an overall, simplified schematic for the steps this study found to be particularly useful for trying to assess, monitor, and remediate a petroleum-derived contaminated site in Alaska.

Particularly for sites with a history of repetitive spills and a variety of remediation efforts conducted over many years, the need for a first assessment of the site is paramount. The step-wise methodology developed by the CRREL team (Figure 6) stages the noninvasive geophysical methods as the first step of the site assessment to minimize disturbance. Determining the seasonal thaw depth and permafrost features (ice wedges, segregation ice, etc.) is integral for an overall assessment of the subsurface. Additionally, assessing soil characteristics, such as distinct regions with varying salt content and presence of soil gradients, is important for contaminant fate and transport. For this study, assessing the surface and subsurface soil layers was accomplished using GPR, ERT, CCR, and frost-probing and by drilling boreholes. Of the methods used, GPR, frost-probing, and the borehole sampling provided the most useful data for understanding subsurface soil layers, particularly the depth of the permafrost and the breach in the containment berm (Bjella 2015; Wagner et al. 2016a). Field-scale analysis by ERT, CCR, and GPR can be accomplished during one summer field season.

The next step in the methodology developed by the CRREL team is to assess the hydrological flow paths and investigate groundwater movement on-site, which can be accomplished during the same summer field season as the noninvasive geophysical work. However, collecting a minimum of two years of groundwater elevation data is ideal for Arctic regions. Estimating surface and subsurface flow paths with respect to PHC distribution and migration is a necessary step in trying to understand transport and the potential for PHC migration off-site or to local bodies of water. Installing monitoring wells or using existing monitoring wells to install water elevation loggers and manually measure groundwater depth can be coupled

with tracer studies to estimate groundwater flow and to establish directional migration as a function of velocity. Estimating groundwater flow paths should be accomplished before any major bioremediation work is attempted or any large-scale soil sampling due to the likelihood of disturbing the subsurface. Additionally, understanding hydrological transport is a necessary component when trying to determine PHC distribution in various environmental compartments (e.g. soils, interstitial waters, colloids, groundwater, pore water, etc.).

Figure 6. Schematic showing the overall methodology for monitoring, assessing, and remediating petroleum-derived contaminated in Arctic regions.

A significant portion of the effort by the CRREL team to characterize the former NARL site was to constrain the soil thermal regime in relation to snow coverage, timing of snow accumulation, and increasing air temperatures. The timing of the soil thermal-regime analysis needs to be carefully planned. Soil temperature thermistors (also potentially equipped with soil moisture dataloggers to support hydrological results) and snow fences need to be installed in summer, but snow coverage and SWE surveys require multiple winter field excursions. Ideally, there would be three field deployments for assessing snow coverage and mapping distribution of snow while also ensuring soil temperature (and moisture) loggers are functioning properly. Harsh conditions in the Arctic and the abundance of animals that are partial to chewing electrical wires cause data logging to be often time-consuming and difficult.

To minimize overall site disturbance, soil core samples for PHC fraction characterization and biogeochemical analyses can be collected when the soil temperature loggers are installed. Key biogeochemical measurements for soil and pore water characterization are pH, soil organic matter, soil moisture, soil type, major phase assemblage mineralogy, soil respiration rates, major fauna, and oxidation-reduction potential. Oxidation-reduction-potential measurements can be made using an oxidation-reduction-potential probe in situ or coupled with additional redox measurements: iron (II)/(III) liquid speciation using ultraviolet-visible spectroscopy, graphite furnace atomic absorption spectrometry, or liquid chromatography-inductively coupled plasma-mass spectrometry (Bangthanh et al. 1999). The soil analyses need to be analyzed rapidly with a few hours, particularly for pH and redox; and for the other analyses, soil and pore waters need to be stored appropriately (see section 3). Characterization of PHCs can be divided into VPHs and EPHs and analyzed by a variety of methods, similar to what was reported in section 3.3.2.

Once the major fractions of the petroleum-derived contaminants are characterized, necessary steps can be taken for developing bioremediation strategies. Investigating the natural microbial community that exists on-site can and should be accomplished prior to laboratory-based development of remediation techniques. Additionally, site assessment of indicators of biodegradation naturally occurring can be accomplished during any one of the initial site assessments, including the noninvasive geophysical analysis (step 1, Figure 6). These assessments can include (1) inspecting for iron staining in sediments and soils and in any location where water

can pool, (2) analyzing a small amount of sample or pore water for nitrogen content, and (3) using DNA extraction techniques and quantitative polymerase chain reaction, which was particularly useful to the CRREL team's site assessment. Finally, bioremediation on the bench-level scale can be attempted, modified, and refined before deployment on-site. Nitrogen concentrations and water content need to be monitored and adjusted as needed within the scope of the experiment.

Additional work recommended for overall site assessment at petroleum-derived contaminated sites includes extensive sampling of pore water, groundwater, and local surface waters. The current CRREL effort did not include any significant water sampling programs because the U.S. Navy's monitoring programs included water sampling. Therefore, additional water sampling programs were deemed unnecessary. However, this study notes that site monitoring of other bodies of water within and adjacent to the former NARL complex besides Imikpuk Lake are a likely, necessary addition to any future monitoring efforts. Overall, the methodology implemented by the CRREL team to assess, monitor, and remediate the former NARL complex was successful; but further work on accelerating the degradation of PHC by microbes is needed. The CRREL-developed methodology presented in this case study can assist in future site evaluations of petroleum-derived contaminated sites in cold regions.

7 Conclusion

The results presented in this work of the NARL case study highlight the overall effectiveness of the work headed by the CRREL team, which used two distinct and separate methodologies to ensure adequate and comprehensive monitoring of PHCs in Arctic regions. First, Arctic environments necessitate nondestructive geophysical tools to characterize both the surface and subsurface soil systems. Relatively more invasive techniques, such as soil-pit excavation and boreholes, disturb the pristine environment surrounding permafrost and are not necessary given the powerful remote-sensing capabilities of GPR and ERT. Secondly, investigating contaminant fate and mobility requires the use of in situ hydro-biogeochemical methods to constrain site characteristics, monitor transport and distribution of contaminants, determine fractionation of target chemical elements, and develop key mechanistic transformation pathways that are site specific. The implementation of both distinct methodologies is essential for controlling PHC-contaminated sites in the Arctic and other similar cold regions.

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14. ABSTRACT The Arctic region of Alaska has a history of petroleum contamination from repetitive fuel spills and the overuse of petrochemicals. Notably, the presence of the former Naval Arctic Research Laboratory (NARL) outside the city of Utqiagvik, Alaska (formerly known as Barrow), resulted in the contamination of local soils and groundwater with petroleum-derived hydrocarbons. Since the NARL closure in 1987, the U.S. Navy (primarily) has implemented many environmental investigations, remediation, monitoring, and containment strategies. However, the soil and subsurface soil unique to the Arctic complicates traditional remediation techniques as a result of the harsh environment and underdeveloped infrastructure of the remote site. Bioremediation and stimulating the existing microbial community represent attractive methods of decontamination because they are nontoxic and relatively easy to implement. The results from this study offer a comprehensive approach for characterizing petroleum-derived contamination specific to Arctic regions by coupling nondestructive geophysical tools with in situ hydro-biogeochemical methods. The overall goals of this project were to investigate the surface and subsurface soil properties at the former NARL site for the Naval Facilities Engineering Command Northwest, monitor the distribution of hydrocarbons, characterize petroleum-derived hydrocarbons, and test various bio- and phytoremediation scenarios both in the laboratory and as a field study.					
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