ESTCP

EXECUTIVE SUMMARY

Remote Monitoring of Natural Source Zone Depletion Using Temperature Data to Support Long-Term Passive Management Strategies

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ESTCP Project ER19-5091

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ACRONYMS AND ABBREVIATIONS

API	American Petroleum Institute
ASTM	American Society for Testing and Materials
CO_2	carbon dioxide
CRCCare	Cooperative Research Center for Contamination Assessment and Remediation of the Environment
DCC	dynamic closed chamber
DoD	Department of Defense
ESTCP	Environmental Security Technology Certification Program
ITRC	Interstate Technology and Regulatory Council
LNAPL	light non-aqueous phase liquid
MN ANGB	Minnesota Air National Guard Base
NSZD	natural source zone depletion
ORP	oxidation-reduction potential
TEAD-S	Tooele Army Depot—South

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1.0 INTRODUCTION

Many Department of Defense (DoD) sites are affected by historical releases of light non-aqueous phase liquids (LNAPL), including fuels, lubricants, and heating oil. Traditionally, costly active treatment technologies (e.g., hydraulic recovery, air sparging, multi-phase extraction, soil vapor extraction, etc.) have been applied as the presumptive remedy for most LNAPL sites. Except for complete excavation, in the project team's experience, none of these in-situ remediation technologies have been able to completely remove all LNAPL. Today, monitoring of natural source zone depletion (NSZD) is gaining broad acceptance as a viable and cost-effective remedy for mature LNAPL releases (e.g., Sale et al. 2018; ITRC 2018; ASTM E3361-22 2022), while ensuring that the goals of protection of human health and the environment are met while progressing towards site cleanup. Key factors supporting NSZD-based remedies include natural LNAPL depletion rates that may exceed what can be achieved with active remedies, greater sustainability, and reduced costs. Similar to a compost pile, the bacterial degradation of LNAPL in the subsurface generates heat. One accepted approach to documenting LNAPL NSZD is real time monitoring of subsurface temperatures and use of the heat generated by NSZD to resolve NSZD rates (e.g., Stockwell 2015; Sale et al. 2015; Warren and Bekins 2015; Karimi Askarani et al. 2018; Karimi Askarani and Sale 2020).

2.0 OBJECTIVES

This project sought to demonstrate that temperature-based quantification of NSZD rates will:

Provide Continuous Measurement of NSZD Rates: Other existing technologies for measuring NSZD rates rely on one-time (or short-term) snapshots of the NSZD rates (on the order of minutes to approximately 2-3 weeks). By continuously recording temperatures within the LNAPL source area, the on-going NSZD processes can be documented and quantified, enhancing the regulatory acceptance of the technology. In addition, continuous monitoring can be used to document seasonal variations in NSZD rates and gain an improved understanding of how changes in site conditions affect NSZD rates.

Permit Monitoring of NSZD Below Paved Surfaces: Many DoD facilities with hydrocarbon LNAPL issues are covered with low permeability surfaces (e.g., parking lots, repair buildings and facilities, tarmacs, etc.). Although current DoD efforts have utilized Carbon Traps to measure NSZD rates (Environmental Security Technology Certification Program [ESTCP] Project ER-201582), Carbon Traps are not suitable for deployment inside buildings or on paved surfaces because Carbon Traps are unable to obtain a representative measurement of carbon dioxide (CO_2) flux when installed on top of or through a paved surface. In contrast, temperature-based monitoring directly measures the heat generated by petroleum biodegradation, and the temperature sensors can be installed through either open ground or paved surfaces (including building foundations).

Reduce Treatment and Monitoring Cost: At many sites, active treatment of LNAPL reaches a point of diminishing returns, especially once LNAPL transmissivity declines as a result of active treatment. For example, Interstate Technology and Regulatory Council (ITRC) (2018) notes that the practical limits of LNAPL recovery are represented by an LNAPL transmissivity of 0.1 to 0.8 ft^2/day, and at lower transmissivities, the majority of LNAPL at a site is in a state of lesser mobile and residual saturation. At this stage, the volume of LNAPL destroyed by NSZD can be one or more orders of magnitude greater (e.g., median site-wide average of 1,020 gallons/acre/year; Kulkarni et al. 2022b) than the volume removed with low efficiency, late-stage existing active systems, such as hydraulic recovery systems, which may only recover a few gallons or tens of gallons per year.

Accurate quantification of NSZD rates supports significant cost savings at these sites by transitioning from the current active technology to a passive technology that verifies LNAPL destruction quantitatively on a continuous basis. In addition, automated uploads and processing of the temperature-based monitoring data allow for low-cost long-term monitoring and can reduce greenhouse gas emissions by reducing on-site visits.

3.0 TECHNOLOGY DESCRIPTION

Based on thermodynamic principles, biological degradation of petroleum in the subsurface generates the same amount of heat per volume of petroleum degraded as combustion of petroleum (for example, combustion of petroleum in an oil furnace used for home heating). At remediation sites, heat from oxidation of hydrocarbons allows use of innovative temperature-based methods to quantify biologically-mediated depletion of LNAPL in the subsurface. The break-through method for the conversion of generated heat to NSZD rates is based on thermodynamics. Methods were originally developed by Colorado State University (Stockwell 2015; Karimi Askarani et al. 2018), with collaborative work by others (e.g., Sweeney and Ririe 2014; Warren and Bekins 2015). Well documented, peer reviewed guidance has been developed for this technology since then (ITRC 2018; ASTM E3361-22 2022). Based on monitoring done at several sites, soil temperatures within LNAPL source areas are commonly observed to be 1˚C to 3˚C above the temperatures at matched background locations. NSZD-related temperature differences can be readily measured with available temperature sensors (e.g., thermocouples with a stated accuracy +/- 0.1˚C, resolution 0.01˚C).

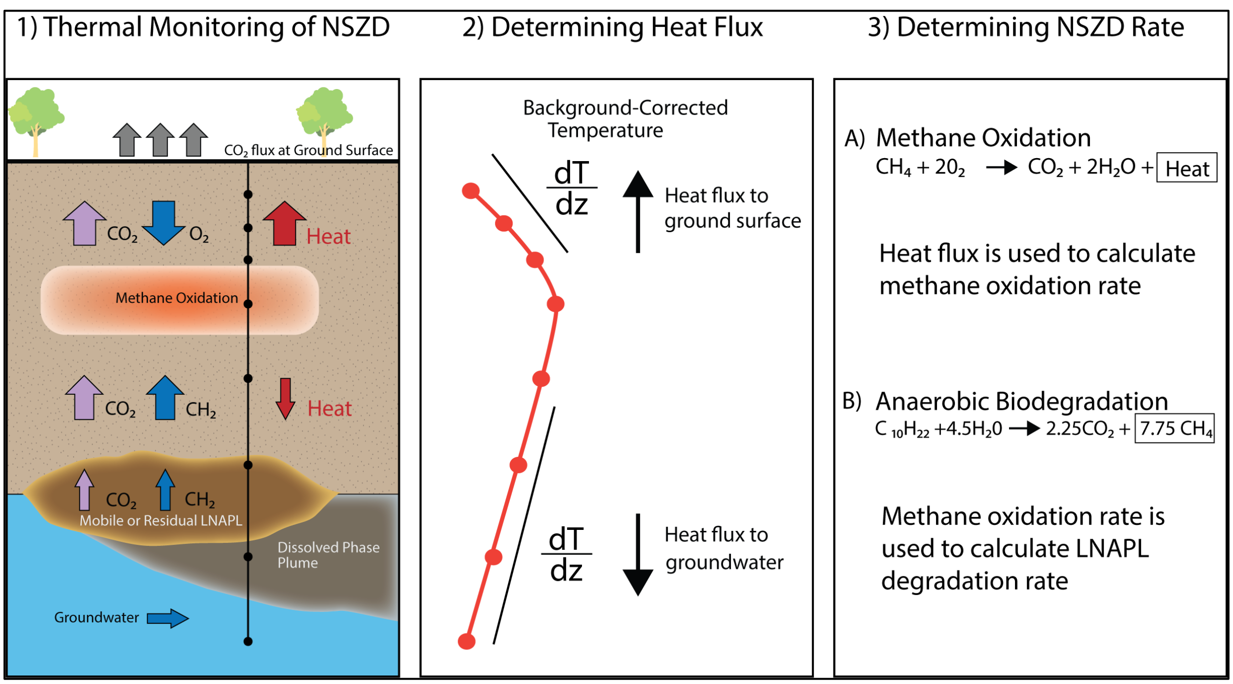

Figure ES-1. Conceptual Model for Temperature-Based Monitoring of NSZD

The application of temperature-based monitoring to determine NSZD rates is illustrated in Figure ES-1. Multiple vertically-spaced temperature sensors (e.g., thermocouples) are installed in the LNAPL impacted area (and at least one non-impacted (i.e., background) location for background-based methods).

A "background-correction" step is required to separate the biogenic heat signal produced by NSZD processes from other sources of heat to the subsurface (e.g., solar heating and cooling, pipelines, etc.). The background-corrected vertical temperature profile is used to determine the upward and downward temperature gradients, which, in turn, are used to calculate the heat flux, which corresponds to the amount of heat being generated from the biodegradation. Based on the amount of heat energy produced from NSZD according to the thermodynamics of petroleum combustion/degradation, the volume of petroleum being degraded per area per unit time (i.e., the NSZD rate) is calculated. Within the United States, NSZD rates are commonly expressed in units of gallons of petroleum degraded per acre per year. Quantification of the NSZD rate allows the mass of petroleum removed by NSZD to be compared to that removed by other remedies.

4.0 PERFORMANCE ASSESSMENT

The demonstration was completed at two sites: Solid Waste Management Unit 13, located at Tooele Army Depot—South (TEAD-S) in Tooele County, Utah and AOC ZZ013, located at the Minnesota Air National Guard Base (MN ANGB), Minneapolis, Minnesota. At each demonstration site, the monitoring locations included: one unpaved non-impacted background location, one paved non-impacted background location, one unpaved source area location, and two paved source area locations. At each of the two demonstration sites, field testing involved: i) installation of temperature monitoring stations; ii) deployment and collection of Carbon Traps; iii) measurement of oxygen, carbon dioxide, and methane soil gas concentrations along a vertical transect; iv) 18 to 24 months of temperature data collection; and v) decommissioning of monitoring locations.

Table ES-1 summarizes the performance objectives and a brief summary of key results, which are explained in more detail below.

Table ES-1. Performance Objectives and Summary of Results

Performance Objective	Success Criteria	Results Summary
1. Collection of a Complete Dataset	Collection of temperature measurements for >95% of planned measurement locations/days and >90% of planned alternative method measurements.	For vertical soil gas profiles of oxygen, carbon dioxide, and methane, and for Carbon Traps, 100% of planned data were collected. For temperature and ORP data, at TEAD-S, data set completeness (>99%) exceeded the performance objective of 95% at each sensor location. At the MN ANGB, data set completeness (72%) was below the performance objective of 95% at the site level, and below 95% at four of five sensor locations.
2. Improved Background Correction for Temperature-Based Monitoring of NSZD	Attainment of more stable background-corrected temperature gradients and NSZD rates compared to 1st generation methods. More accurate short-term (weekly to monthly) NSZD rates compared to 1st generation methods.	Three different temperature-based methods were used to calculate NSZD rates: Method 1 (annual average), Method 2 (background correction), and Method 3 (Single Stick). Although the daily rates at both TEAD-S and the MN ANGB were highly variable and may not be reliable for estimating short-term NSZD rates (daily to weekly), they do provide a reliable estimate of annual NSZD rates.

Table ES-1. Performance Objectives and Summary of Results (Continued)

Performance Objective	Success Criteria	Results Summary
3. Comparison between NSZD rates from temperature-based monitoring and other NSZD methods	Comparability of NSZD rates between temperature and alternative methods. Agreement between methods within a factor of 2X (or evidence that temperature-based monitoring method is more accurate based on, for example, lower spatial and temporal variability).	The background correction method yielded NSZD rates closer to the annual average method than the Single Stick method. However, the agreement in NSZD rates between the individual temperature-based methods and the alternative methods was more variable. Additional field demonstration is needed to determine which temperature-based method yields the most accurate estimates of NSZD rates across a larger number of sites.
4. Demonstration of 2nd Generation Equipment	Lower costs and more detailed vertical temperature dataset compared to 1st generation equipment.	At both sites, the 2nd generation equipment was able to record temperatures at more than twice as many depths as the 1st generation equipment, with less cost per sensor and in total.
5. Documentation of NSZD Below Paved Surfaces	Temperature profile consistent with NSZD. Methane/carbon dioxide/oxygen distribution consistent with NSZD.	The weight of evidence suggests NSZD in paved areas at TEAD-S, but the NSZD rates appear to be lower than those at the unpaved location. At the MN ANGB, each temperature-based and alternative method support the occurrence of NSZD beneath paved surfaces. These results and results published by Smith et al. (2021) provide evidence that temperature-based methods are suitable for quantification of NSZD rates beneath paved surfaces.
6. Compilation of NSZD Rates Across NSZD Monitoring Sites	Documentation of typical range of NSZD rates across sites. Identification of site factors predictive of higher or lower NSZD rates.	At 40 impacted sites, NSZD rates were compiled using the following temperature-based methods: Gradient Method, Carbon Traps, Dynamic Closed Chamber (DCC), and Thermal Monitoring. Site-average NSZD rates ranged from 650 to 152,000 L/ha/yr (70 to 16,250 gallons per acre per year (gal/acre/yr)), with a median value of 9,540 L/ha/yr (1,020 gal/acre/yr). No clear bias was observed between NSZD rate measurement methods, with the difference between any two methods generally within a factor of 2-3x. Although NSZD rates vary across sites, fuel type is not the primary factor explaining observed differences in rates.

4.1 PERFORMANCE OBJECTIVE 1: COLLECTION OF A COMPLETE DATASET

At each monitoring location, the data collection included: i) vertical soil gas profiles of oxygen, carbon dioxide, and methane; ii) Carbon Traps; and iii) temperature monitoring locations that recorded temperature and the oxidation-reduction potential (ORP).

- For oxygen, carbon dioxide, methane, and Carbon Traps, 100% of planned data were collected. However, methane data from the MN ANGB were rejected due to quality assurance problems in the field.
- For temperature and ORP, at TEAD-S, data set completeness (>99%) exceeded the performance objective of 95% completeness for planned measurement days for temperature measurements. At the MN ANGB, the data set completeness (72%) over the performance period was below the performance objective of 95% at the site level, and data set completeness was below 95% at four of five sensor locations. Since the performance objective was not met for temperature measurements at the MN ANGB, linear interpolation was used to estimate missing hourly measurements, and the likely effect of the data interpolation on calculated NSZD rates was assessed to determine whether failure to meet the performance objective meaningfully affected the validity of the NSZD results.

4.2 PERFORMANCE OBJECTIVES 2 & 3: IMPROVED BACKGROUND CORRECTION FOR TEMPERATURE-BASED MONITORING OF NSZD AND COMPARISON BETWEEN NSZD RATES FROM TEMPERATURE-BASED MONITORING AND OTHER NSZD METHODS

To quantify NSZD rates, the increase in soil temperature attributable to petroleum biodegradation must be separated from other factors that affect the soil temperature. The primary driver of other heat sources is solar insolation, which leads to seasonal variations in soil temperature (i.e., summer warming and winter cooling), although other anthropogenic sources such as pipelines may also need to be considered. At the two demonstration sites, no apparent alternate sources of heat existed near the temperature monitoring locations. At TEAD-S, all buildings had been decommissioned, and there were no active facilities or utilities near the temperature monitoring locations. At the MN ANGB, based on underground utility diagrams provided by the facility, one location, background location BG-1, was located approximately 13 feet away from a water line, and it was unlikely that the underground water line impacted the subsurface temperature data at this location. Each of the other temperature monitoring stations were located at least 35 feet from the nearest utility line. For this demonstration, the project team evaluated three methods for "background correction" (i.e., removing non-biodegradation-related heat sources): Annual Average Temperature Method (Method 1), Correction Using Background Location (Method 2), and the Single Stick Method (Method 3) (see Table ES-2).

Table ES-2. Comparison of Net NSZD Rates (gal/acre/yr)

Temperature Monitoring Station Location	Method 1—Annual Temperature Method	Method 2—Correction Using Background Location Method	Method 3—Single Stick Method
TEAD-S			
L-1 (paved)	-90	-140	570
L-2 (paved)	170	180	1,320
L-3 (unpaved)	300	360	800
MN ANGB			
L-1 (unpaved)	630	610	850
L-2 (paved)	2,060	2,060	3,090
L-3 (paved)	850	1,020	2,810

Note: NSZD rates rounded to the nearest 10

Absolute NSZD rates at each of the five locations at each demonstration site are shown in Table ES-3, and net NSZD rates for the three impacted locations are shown in Table ES-2. The background corrected method yielded NSZD rates closer to the annual average method than the Single Stick method. The Single Stick method consistently calculated higher NSZD rates than the other two methods, with larger differences at TEAD-S than the MN ANGB. However, the agreement in NSZD rates between the individual temperature-based methods and the other non-temperature-based alternative methods to estimate NSZD rates was more variable. Additional field demonstration is needed to determine which temperature-based method yields the most accurate estimates of NSZD rates across a larger number of sites.

Table ES-3. Absolute NSZD Rates (gal/acre/yr) for Carbon Trap Method and Qualitative Evidence of NSZD Based on Gas Gradients

Temperature Monitoring Station Location	NSZD Rate by Method 1—Annual Temperature Method (gal/acre/year)	NSZD Rate by Method 2—Background Location Method (gal/acre/year)	NSZD Rate by Method 3—Single Stick Method (gal/acre/year)	NSZD Rate by Carbon Trap Method (gal/acre/year)	Evidence of NSZD Based on Soil Gas Gradients
TEAD-S					
L-1 (paved)	150	-140	370	295	Strong
L-2 (paved)	420	170	1,150	84	Medium
L-3 (unpaved)	640	360	1,080	1,105	Medium
BG-1 (paved)	250	NC	-200	Not Detectable	No Evidence of NSZD
BG-2 (unpaved)	370	NC	270	Not Detectable	No Evidence of NSZD
MN ANGB					
L-1 (unpaved)	1,220	610*	730	72	Weak
L-2 (paved)	2,710	2,060	3,180	1,856	Medium
L-3 (paved)	1,440	1,020	2,910	404	Medium
BG-1 (unpaved)*	570*	NC	-120*	129	Weak
BG-2 (paved)	610	NC	100	37	Weak

Note: * LNAPL observed in monitoring well at this intended background location
NC – net NSZD rate cannot be calculated with Method 2

4.3 PERFORMANCE OBJECTIVE 4: DEMONSTRATION OF 2ND GENERATION EQUIPMENT

At the TEAD-S demonstration site, the 2nd-generation equipment performed as expected (>99% data recovery with no significant data gaps). At the MN ANGB demonstration site, however, the 2nd generation equipment at times failed to record temperature data throughout the demonstration period, resulting in data gaps and an incomplete temperature record. The equipment vendor, S3NSE Technologies, was unable to fully resolve the issues despite numerous attempts over the course of the demonstration. At both sites, the 2nd generation equipment was able to record temperatures at more than twice as many depths as the 1st generation equipment.

Additional temperature data at these two demonstration sites improved the performance of the Method 3 Single Stick method, whereas the impact of using the 2nd generation equipment appeared to be less for the Method 1 annual average and Method 2 background-correction methods. The soil gas analyses from the 2nd generation equipment soil gas sample ports were comparable to the result obtained from adjacent stand-alone soil gas sample points, and the ORP data provided a secondary line-of-evidence for the qualitative evaluation of NSZD at each site. The 2nd generation equipment provides additional sensors (i.e., soil gas, ORP) at less per sensor and total cost than the 1st generation equipment. In summary, if the equipment vendor is able to fully resolve the equipment data reliability issues, the 2nd generation equipment provides an improvement over the 1st generation equipment.

4.4 PERFORMANCE OBJECTIVE 5: DOCUMENTATION OF NSZD BELOW PAVED SURFACES

At TEAD-S, while the soil gas profiles (see Section 5.7.1; Figures 5.5 and 5.6; Section 6.3.2 of the full final report) were consistent with the expected profiles and demonstrate the occurrence of NSZD below paved surfaces, the Method 1 average annual vertical temperature profiles at the paved locations at TEAD-S did not correspond to the average annual vertical temperature profiles expected for NSZD processes (i.e., no clear evidence of elevated soil temperatures in the subsurface at the impacted locations compared to the non-impacted background location). Likewise, the Method 2 background-corrected NSZD results were low (or negative), suggesting little to no NSZD, and the Carbon Trap results indicated low NSZD rates at the paved locations. In contrast, the Method 3 Single Stick method indicated measurable NSZD results notably higher than the rates indicated by the other methods. In summary, the weight of evidence suggests NSZD in paved areas at the first demonstration site, TEAD-S, but the NSZD rates appear to be lower than those at the unpaved location.

At the MN ANGB, in contrast, the two paved locations, L-2 and L-3, did show average annual vertical temperature profiles consistent with NSZD, and the calculated NSZD rates based on temperature-based monitoring methods and Carbon Traps at these locations supported the occurrence of NSZD beneath paved surfaces.

In addition, Smith et al. (2021) demonstrated the use of temperature-based methods beneath paved sites at a retail fuel station in Europe. Thus, the general theory and results from other published studies (e.g., Smith et al. 2021) provided further evidence that temperature-based methods were suitable for quantification of NSZD rates beneath paved surfaces, although additional field verification may be desirable to support the limited data to date.

4.5 PERFORMANCE OBJECTIVE 6: COMPILATION OF NSZD RATES ACROSS NSZD MONITORING SITES

The project demonstration included a data mining component to characterize rates of NSZD measured across a wide range of petroleum-impacted sites. The goals of the data mining study were to: i) characterize the range of site-wide average NSZD rates measured across a wide range of sites, ii) evaluate the impact of fuel type on NSZD rates, iii) evaluate the comparability of different methods to measure NSZD rates, and iv) characterize how NSZD rates vary at individual sites over a time scale of a few months to a few years.

At each site, the following data were compiled: i) general site location; ii) LNAPL fuel type; iii) measurement method, number of locations, and number of measurements per location; and iv) calculated sitewide average NSZD rate and the associated measurement method (i.e., Gradient Method, Carbon Traps, Dynamic Closed Chamber (DCC), or Thermal Monitoring). The resulting dataset showed average sitewide NSZD rates that ranged from 650 to 152,000 liters per hectare per year (L/ha/yr) (70 to 16,250 gallons per acre per year (gal/acre/yr)), with a median value of 9,540 L/ha/yr (1,020 gal/acre/yr).

No clear bias was observed between the four NSZD rate measurement methods. When comparing the different NSZD measurement methods applied to the same sites, the site-average NSZD rates differed by a median factor (i.e., ratio of faster rate to slower rate) of 2.1 times. Despite the variability from measurement methods, seasons, and time-scales, a reasonable estimate of the long-term NSZD rate (e.g., within a factor of 2 or 3) can be achieved at the majority of sites by: i) a single measurement method employed at 3-7 locations per site; and ii) spanning at least two semi-annual (fall and spring) or four seasonal measurements per location.

Additionally, based on a limited dataset of four sites, NSZD rates were typically higher during the summer and fall (when subsurface temperatures are highest) compared to winter and spring (when subsurface temperatures are lowest), which suggests that biodegradation rates are enhanced by low-level increases in temperature. This is discussed in various literature studies (Kulkarni et al. 2022b). As such, increasing the mean annual soil temperature with engineered methods could potentially increase the biodegradation rate at a site. Although NSZD rates vary across sites and over time at an individual site, the fuel type does not appear to be the primary factor explaining the observed differences in NSZD rates.

The findings of this study have recently been published in the journal Water Research. Additional citation information on this paper (Kulkarni et al. 2022b) is available online at: https://www.sciencedirect.com/science/article/abs/pii/S0043135422011150?via%3Dihub.

5.0 COST ASSESSMENT

Table ES-4 shows the estimated costs for installing temperature monitoring stations at a representative field site. It is assumed that five temperature monitoring stations are installed at relatively shallow depth (less than 20-25 feet below ground surface), that installation takes 4 10-hour days, and that the site is local (i.e., no travel expenses, lodging, per diem, etc.).

Comparable costs (2023) for Carbon Traps are approximately $2,500 per Carbon Trap. Assuming similar costs for project planning and preparation, data evaluation and reporting, and field program implementation (excluding the cost of the drilling subcontractor and equipment rental), the total cost for implementing five Carbon Traps for a one-time sampling event is approximately $24,000. Each additional sampling event with Carbon Traps would be expected to cost approximately $13,300 (assuming 2 days of field work for installation and retrieval). Thus, for sites requiring a one-time NSZD rate, Carbon Traps may be more cost effective, but for those sites requiring continuous monitoring over multiple sampling events or long-term monitoring over a period of years to decades, there are cost advantages to utilizing the temperature-based NSZD continuous monitoring technology.

Table ES-4. Estimated Cost for Installation of 5 Temperature Monitoring Stations at 1 Site

Cost Element	Cost Element	Units	Cost Per Unit	Estimated Cost
Project planning and preparation	Senior Project Scientist/Engineer	8 hours	$200	$1,600
	Project Scientist/Engineer	24 hours	$125	$3,000
Hardware procurement	Temperature Monitoring Stations	5 units per site	$8,000	$40,000
Field Program Implementation	Labor hours: Senior Project Scientist/Engineer	4 hours	$200	$800
	Labor hours: Project Scientist/Engineer	40 hours	$125	$5,000
	Drilling Subcontractor	1 per site	$20,000	$20,000
	Supplies	1 per site	$50	$50
	Equipment Rental, Supplies, Shipping	4 days	$220	$880
Data evaluation and reporting	Senior Project Scientist/Engineer	8 hours	$200	$1,600
	Project Scientist/Engineer	16 hours	$125	$2,000
			TOTAL	**$74,930**

6.0 IMPLEMENTATION ISSUES

There are no widespread barriers to the implementation of temperature-based methods. Further guidance on NSZD methods, and the temperature-based (thermal or biogenic heat) methods specifically, is available in various guidance documents, including API (2017), ITRC (2018), Cooperative Research Center for Contamination Assessment and Remediation of the Environment (CRCCare) (2018), and CL:AIRE (2019). In addition, temperature-based methods and their application are described in the recently published ASTM guidance document (E3361-22) on NSZD.

To the project team's knowledge, there are no current regulations or permits that are required to implement the technology as a monitoring technology, although site-specific application and use of the NSZD data should be considered within the larger site conceptual model and in consultation with any applicable State or Federal regulators. While reluctance to implement natural remedies that rely on NSZD remains among some regulators, other regulatory bodies have included the qualitative evaluation of NSZD when considering whether site closure is acceptable even with LNAPL remaining in-place (e.g., VA 2012; WV 2019). The continual advancement of guidance documents such as ITRC (2018) and ASTM E3361-22 (2022) provides the fundamental scientific basis for NSZD and accepted measurement methods. A continuing body of evidence reviewed during this project, including that collected at the two demonstration sites, indicates that NSZD has been measured at all sites in the literature (Kulkarni et al. 2022b; Available online at: https://www.sciencedirect.com/science/article/abs/pii/S0043135422011150?via%3Dihub).

The 2nd generation monitoring sensors and communication equipment can be procured from S3NSE Technologies as newly commercialized, custom-built equipment. Colorado State University Research Foundation currently owns the patent (Sale et al. 2015; US Patent No. 10,094,719) for devices and methods for measuring the thermal flux and estimating the NSZD rate, which GSI Environmental Inc. has exclusively sublicensed.

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