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Soil Gas Survey Results Supporting Groundwater Correction Action Plan (GCAP) Development for the Moab Site

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EXECUTIVE SUMMARY

A soil gas survey was performed at the Moab Uranium Mill Tailings Remedial Action (UMTRA) Project Site during the week of November 6, 2023. Soil gas surveys are used to characterize residual subsurface sources of volatile contaminants, such as volatile organic compounds, as well as contaminants that generate a surrogate indicator gas or otherwise influence soil gas composition.

The primary objective of the Moab soil gas survey was to confirm, identify, quantify, and refine secondary contaminant source area locations for uranium and ammonium/ammonia (NH_4^+/NH_3) in the vadose zone and shallow groundwater. The overarching goal was to provide information to assist in developing the technical basis for the Groundwater Compliance Action Plan (GCAP). Specifically, the soil gas data will support the deployment of source control technologies; e.g., where supplementary capping, removal actions, or amendments might be beneficial.

Surrogate gases were used during the survey as indicators of key contaminants of concern (COCs) for the Moab site. The gas-phase surrogate-indicator for uranium and its associated radionuclides was radon (Rn) and the surrogate-indicator gas phase analytes for NH_4^+/NH_3 were NH_3 and nitrous oxide (N_2O). General geochemical indicator soil gases (carbon dioxide [CO_2], methane [CH_4], and humidity) were also measured. Samples were collected using soil probes into Tedlar® bags and all parameters were analyzed on site. Rn was measured using scintillation cells and the other parameters were measured using a photoacoustic spectrometer (PAS). Soil gas samples were collected from 58 locations that represented four different spatial areas (Mill Yard, Tailings, North Off-Pile, and Wellfield/Riverbank). In addition, features associated with former mill operations and other historical features were identified and targeted to support a high degree of granularity in the data interpretation. The collaborators for the field work were the Department of Energy Office of Environmental Management (DOE EM), North Wind, and Savannah River National laboratory (SRNL).

The Moab shallow soil gas survey was effective in identifying residual sources of uranium and ammonium in the subsurface. The soil gas survey identified general-areal differences in residual subsurface sources and specifically identified small residual hot spot sources associated with the containment pond in the Mill Yard area and the purification pond in the tailings area.

- Significant variations were measured in the data for Rn (4 orders of magnitude), N_2O (> 3 orders of magnitude), CO_2 (> 2 orders of magnitude), CH_4 and NH_3 (both about 1.5 orders of magnitude). For example, Rn measurements varied from 12 pCi/L to 143,136 pCi/L.
- The north off-pile former ore storage area, where removal actions and revegetation efforts have been completed, were found to have relatively low “background” Rn levels in the shallow soil gas. These data affirm that the criteria used for the remediation resulted in effective removal of the original source uranium/thorium/radium materials.
- The samples collected along the edge of the contamination area -- “CA Boundary” -- were generally found to have low levels of Rn, ranging from background to a few thousand pCi/L. These soil gas results affirm the current CA delineation and provide and independent confirmation of the survey/screening/measurement processes that have been used for delineating the CA, as well as the overall radiation control policies and processes and management at the Moab site.
- Significant residual source contamination hot spots were identified for both uranium and ammonium associated with containment pond in the replace area and the purification pond in the tailings area. The maximum Rn levels, for example, were 143,136 pCi/L and 102,982 pCi/L for the containment pond and nearby purification pond, respectively. The general biogeochemical soil gas indicators for these features indicated that a significant, sustained, and active subsurface microbial community is present in the hot spot area.

- NH_3 was elevated in the soil gas at a number of sampling locations in the well field and near the riverbank, likely resulting from the flushing of ammonium from source zones and subsequent groundwater transport from the upgradient sources.
- The sample locations in the tailings area were generally elevated for both Rn and NH_3, confirming that the remaining tailings are a bulk source for residual uranium and ammonium. Based on the low levels measured in the shallow soil gas data from the north off-pile former ore storage area, the planned removal and relocation of the tailings over the next several years should be effective in mitigating this bulk residual source.
- While future work to characterize the geochemistry in the hot spot areas may be needed, the soil gas results combined with other existing data can be utilized to develop a compelling conceptual model of the likely geochemical controls. Uranium geochemistry and formation of mineral phases (such as solid phase uramphite or uraninite) limit the solubility and release of uranium to the groundwater and are significant at Moab, particularly for the area near the former containment and purification ponds and possibly significant throughout the entire plume.
- Removal and relocation of the hot spot sources to the extent practicable would be prudent - coincident with the removal of the bulk tailings. For any residual high strength source that cannot be removed or relocated, the GCAP could be structured to include enhanced geochemical controls using a site-specific recipe of amendments.
- The shallow soil conditions (dry silt and fine-grained sand) with some cobbles were difficult to sample using the hand probe and sliding hammer system. Several modifications were made to the sampling equipment over the course of the study. Future soil gas survey work should select equipment and develop sampling procedures that incorporate the lessons learned from this sampling campaign.

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LIST OF ABBREVIATIONS

CA	Contamination Area
CH_4	methane
CO_2	carbon dioxide
COCs	contaminants of concern
DOE	Department of Energy
DOE EM	Department of Energy Office of Environmental Management
DOE-LM	Department of Energy Office of Legacy Management
EM	Environmental Management
GCAP	Groundwater Compliance Action Plan
N_2	nitrogen
NEON	National Ecological Observatory Network
NH_3	ammonia
NNLEMS	Network of National Laboratories for Environmental Management and Stewardship
NO	nitric oxide
N_2O	nitrous oxide
NRC	Nuclear Regulatory Commission
NRPP	National Radon Proficiency Program
NRSB	National Radon Safety Board
PAS	photoacoustic spectrometer
pCi/L	picocuries per liter
ppmv	parts per million volume
RAP	remedial action plan
Rn	radon
^{220}Rn	radon 220
^{222}Rn	radon 222
SRNL	Savannah River National Laboratory
^{232}Th	thorium 232
TD	Technology Development
^{238}U	uranium 238
UMTRA	Uranium Mill Tailings Remedial Action
VOCs	volatile organic compounds

1.0 Introduction

A key groundwater compliance action development data gap for the former mill site in Moab, Utah relates to knowledge about the nature, location, and extent of residual or secondary sources of uranium and/or ammonium in the soil, vadose zone, and/or shallow groundwater. An improved understanding of these sources would provide actionable information and an improved technical basis for decisions related to selecting, locating, and designing future environmental remedies.

A soil gas survey is a relatively low cost, systematic approach for addressing the subject data gaps and supporting development of the Moab UMTRA Project GCAP. The data and mapping from a soil gas survey have the potential to assist in determining if targeted-supplemental GCAP source remediation in the vadose zone and shallow groundwater may be needed, and (if needed) reduce costs associated with such remediation, limit collateral impacts of future treatments, and improve efficiency by refining the target zone footprint. To this end, shallow soil gas samples were collected throughout the Moab site in November 2023 and analyzed for surrogate gas phase indicators of uranium and ammonium. Collected samples with elevated uranium-related gases (radon) indicate potential uranium source areas or residual shallow source materials. Similarly, areas where NH_3 or other nitrogen related gases (N_2O) are detected would provide insights about residual ammonium sources and nitrogen transformation processes. Quantification of the target constituents was performed on-site using alpha counting radon monitors and a photoacoustic spectrometer (PAS).

2.0 Background

The Moab UMTRA Project site is a 475-acre former uranium-ore mill site located in Grand County, Utah bordered by the Colorado River to the southeast. The Moab mill operated from 1956 through 1970 to support national defense programs, and then until 1984 to support commercial reactors. The mill processed an average of 1,400 tons of ore per day using anhydrous ammonia and other chemicals to separate uranium from the ore. Milling operations resulted in the creation of process-related tailings and other mill-related radioactive debris/materials. The tailings were slurried and transported to an unlined pile in the western portion of the property that ultimately occupied about 130 acres of the Moab millsite footprint. When processing operations stopped, the pile contained an estimated 16 million tons of tailings and contaminated material. Interim actions, like temporary covering of the tailings pile, were taken during decommissioning. Relocation of the mill tailings to an engineered disposal cell in Crescent Junction is in progress.

The Department of Energy (DOE) has primary responsibility for cleanup of both soil and groundwater at the Moab site. DOE Office of Environmental Management (EM) in Grand Junction manages the Moab Project, with a primary scope of tailings removal from the Colorado River floodplain and relocation to an engineered disposal cell. Interim groundwater mitigation actions to date include contaminant mass removal/control and protection of Colorado River habitats utilized by endangered fish species.

Environmental management and cleanup at the Moab mill site are governed by the Uranium Mill Tailings Radiation Control Act (UMTRCA). A key Moab Project milestone is the development of a GCAP, which is currently scheduled in FY2027. The GCAP will require concurrence from the U.S. Nuclear Regulatory Commission (NRC). The GCAP will provide the plans and strategies for groundwater remediation, regulatory compliance, and monitoring to support future groundwater management at the site

Ongoing investigations by several National Laboratories and private industry partners are supporting characterization of site conditions in ultimate support of the GCAP development. These investigations are an outgrowth of the technical recommendations for the Moab Project made by collaborators from the DOE Network of National Laboratories for Environmental Management and Stewardship (NNLEMS) (NNLEMS, 2023). One of the recommendations from the NNLEMS collaboration was performing a soil gas survey as a rapid-cost-effective means to refine understanding of residual and secondary sources underlying the former Moab mill site. A soil gas survey was recently completed by the Savannah River National Laboratory (SRNL) in the November 2023.

3.0 Objectives of Soil Gas Survey

The primary objective of the soil gas survey is to collect and analyze shallow soil gas to confirm, identify, quantify, and refine secondary contaminant source locations for uranium and NH_4^+/NH_3 in the vadose zone and shallow groundwater. Soil gas is defined here as the gas phase in subsurface pore spaces, fractures, and openings in soil and rock. A secondary objective is to determine if ammonium/ammonia are undergoing biological transformation and measurable attenuation.

The overarching goal is to provide information to assist in developing the technical basis for a protective, effective, and efficient GCAP. Specifically, the soil gas data will help focus the deployment of source control technologies (e.g., where supplementary capping, removal actions, or amendments might be beneficial).

This report summarizes methods and results from the survey used as a rapid screening tool for identifying residual subsurface contamination.

4.0 Methods

4.1 Overview

Soil gas surveys are commonly applied in characterizing volatile organic compounds (VOCs) such as chlorinated solvents and light hydrocarbons. In these surveys, the pattern of vapor phase concentrations in gas collected from a grid of sample locations is diagnostic of the location of residual subsurface contamination and contaminant sources, as well as geologic features such as preferential flow paths. Like VOCs, some inorganic contaminants, such as uranium and ammonium, have the potential to be characterized using soil gas surveys based on the concentrations of surrogate gases.

The strategy applied in this survey was to collect soil gas samples from approximately 0.6 to 0.9 m depth at representative grid/transect locations across the Moab site. Instruments that provide near-real time results were then used to analyze “surrogate” gas phase constituents that indicate the presence of secondary contaminant sources. The gas-phase surrogate-indicator for uranium and its associated radionuclides was Rn, and the surrogate-indicator gas phase analytes for ammonium were NH_3 and N_2O. General geochemical indicator soil gases (CO_2, CH_4, and humidity) were also measured.

The collaborators for this work were DOE EM, North Wind, and SRNL.

4.2 Sampling Approach

Fifty-eight soil gas samples were collected and analyzed during the week of November 6, 2023 (Table 4-1; Figure 4-1). The samples were positioned to provide information on areas where tailings are still present and where surface remediation and revegetation is complete, as well as former pond areas, former ore storage pile areas and background areas. The Moab site was broadly subdivided into four areas (see annotations in Figure 4-1) to aid in data interpretation. Rn served as the indicator of contamination associated with residual uranium/radium, while N_2O and NH_3 gas served as indicators of residual NH_4^+ contamination.

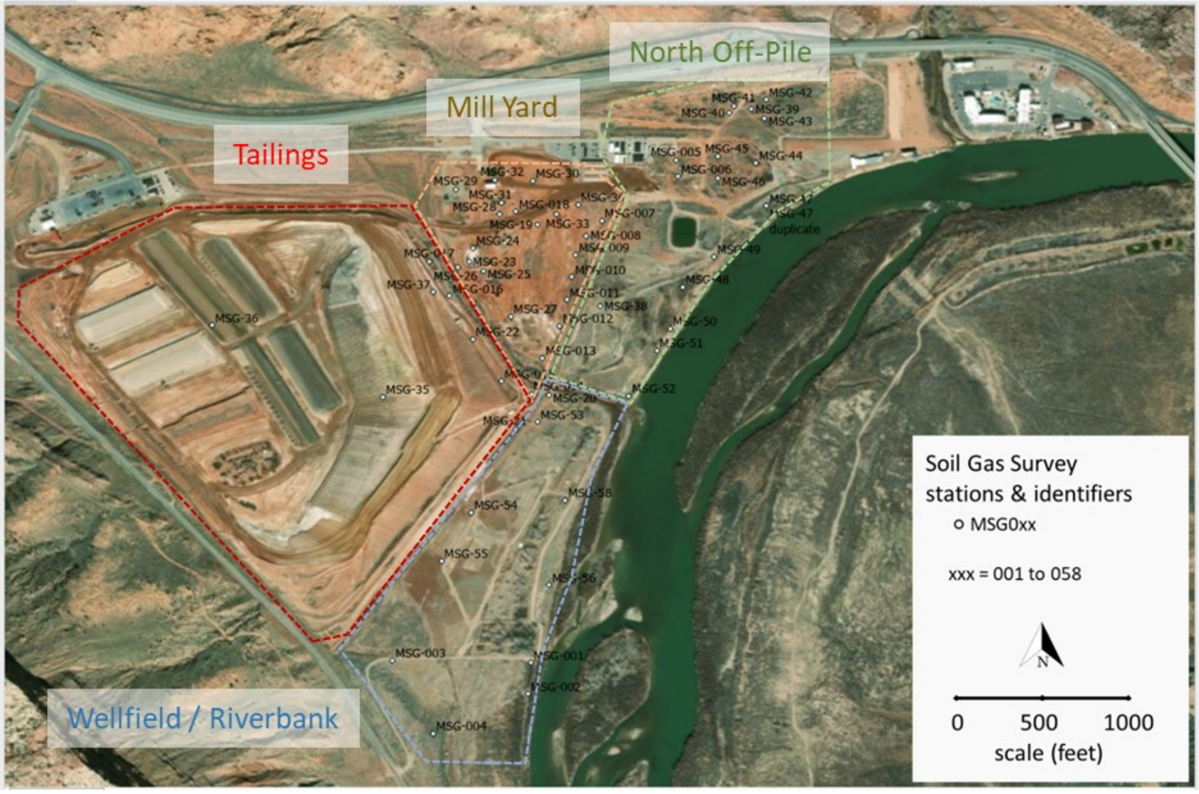

Figure 4-1. Sampling Locations

4.2.1 Sampling Location Selection

The Moab UMTRA Project site is a former uranium mill and consists of 475 acres, of which 195 acres are within the contamination area (CA). Soil gas samples were collected from 58 locations that represented four different spatial areas, the Mill Yard, Tailings, North Offpile, and Wellfield/Riverbank. For the purposes of this investigation, the CA was divided into the Tailings and the Mill Yard areas. The area outside of the CA was divided into the Wellfield/Riverbank (located between the tailings pile and Colorado River), and the North Offpile (between the former mill yard and the Colorado River) areas. The Tailings area is where the tailings are currently being excavated and is a known primary source of uranium and ammonium contamination. The Wellfield area contains the interim action groundwater systems and is adjacent to suitable habitats that may develop. The Mill Yard area is a known source for a uranium plume, and the North Offpile has been remediated and has a less extensive network of monitoring wells.

Sampling locations were based on current monitoring results and historic activities (Figure 4-1). Seven locations included in this investigation (MSG-003, -018, -019, -035, -036, and -038) coincide with locations targeted during a 2023 independent secondary source investigation. Two transects were placed, one along the riverbank and one along the eastern CA boundary. A dense sample pattern was placed in the Mill Yard area as it was expected to uncover actionable point sources. Mill Yard locations were chosen based on a 1975 mill layout diagram (Figure 4-3) overlain onto a 1981 mill aerial photo (Figure 4-2) and more recent satellite imagery (Figure 4-4). Figures included in the *Site Observational Work Plan* (SM Stoller, 2003) identified several pond features around the tailings pile that merited sampling. Former ore storage locations located in the north off pile were also points of interest. Additional locations were placed near active monitoring wells.

All work was performed during one standard work week by a collaborative effort of the Moab team and SRNL team. Monday morning was dedicated to site familiarization, prejob briefings, planning, and initial laboratory setup. The equipment crate arrived on Monday afternoon allowing final setup and mobilization of the lab and field equipment. Soil gas samples were collected and analyzed from late Monday afternoon through Friday morning and the equipment was packed for shipping on Friday afternoon. During the 3.5 days of active field work, 58 locations were sampled and analyzed. The Moab and SRNL teams met regularly throughout the week to assure that the selected sample locations would best address DOE and GCAP development objectives.

Figure 4-2. Aerial View of Mill at former Moab Site (1981)

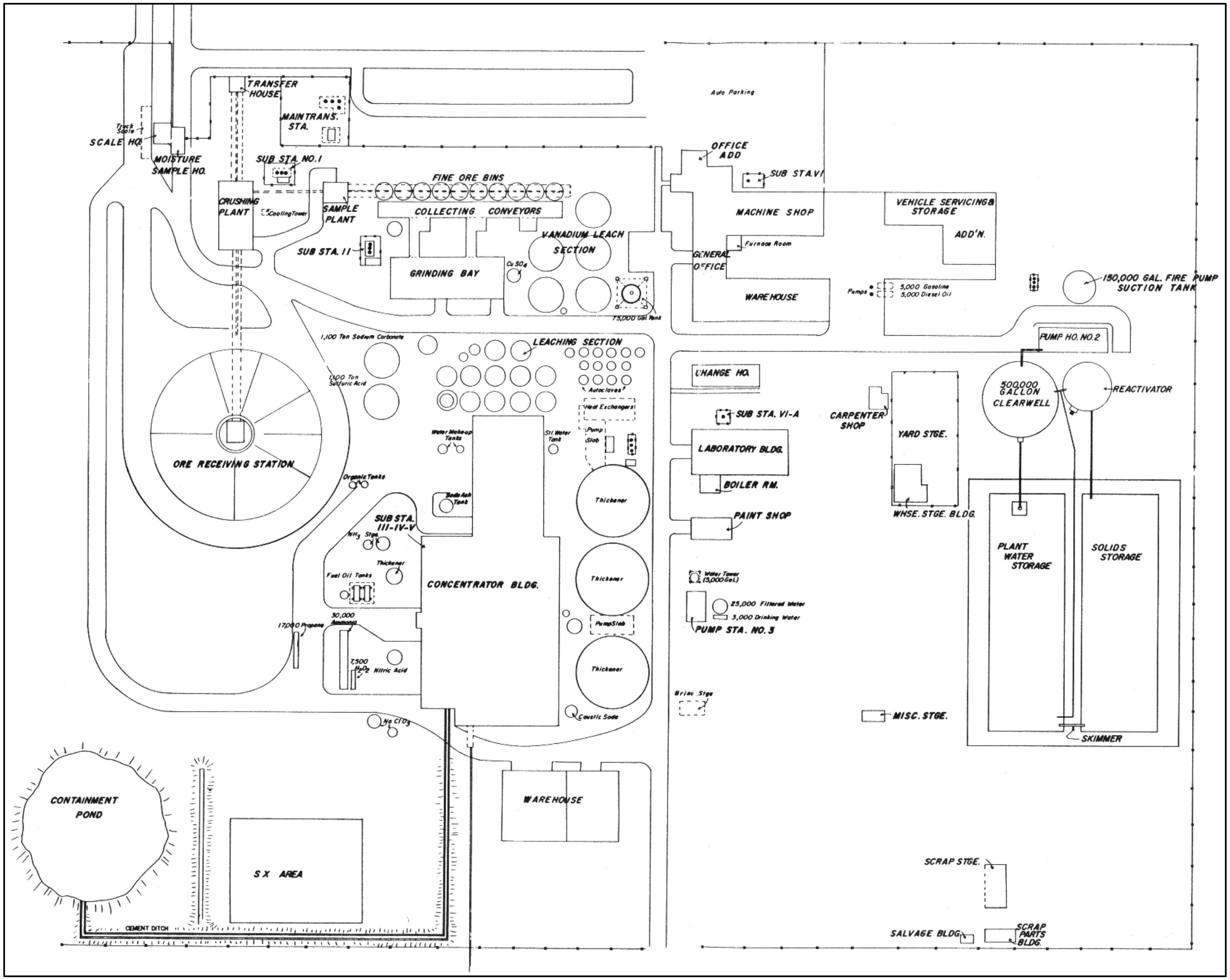

Figure 4-3. Mill Layout Diagram (1975)

Figure 4-4. Mill Layout Diagram Overlaid Onto 2020 Satellite Imagery for Former Moab Site

Table 4-1. Sampling Location Information and Associated Features.

Station	Category	Associated Feature	
		Historical	Modern
MSG-001	Well Field		Riverbank / Well 0492/ Secondary source
MSG-002	Well Field		Riverbank
MSG-003	Well Field		Well TP-23/Secondary Source
MSG-004	Well Field		Well TP-20
MSG-005	North Off-pile	Ore Storage	
MSG-006	North Off-pile	Ore Storage	
MSG-007	Mill Yard		CA/ Well SMI-PZ3
MSG-008	Mill Yard		CA
MSG-009	Mill Yard		CA
MSG-010	Mill Yard		CA
MSG-011	Mill Yard		CA
MSG-012	Mill Yard		CA
MSG-013	Mill Yard		CA Boundary/ Well UPD-18
MSG-014	Tailings	Sump Pond	
MSG-015	Tailings	Pond (BaCl2)	
MSG-016	Tailings	Pond (Purification)	
MSG-017	Tailings	Pond (Purification)	
MSG-018	Mill Yard		Well UPD-20/ Secondary Source
MSG-019	Mill Yard		Well 0411/ Secondary Source
MSG-020	Tailings	Sump Pond	
MSG-021	Tailings	Pond (BaCl2)	
MSG-022	Tailings	Pond (BaCl2)	
MSG-023	Mill Yard	Containment Pond	
MSG-024	Mill Yard	Containment Pond	
MSG-025	Mill Yard	Containment Pond	Well UPD-19
MSG-026	Mill Yard	Containment Pond	
MSG-027	Mill Yard	Pond (Unknown)	
MSG-028	Mill Yard	Ore Receiving	
MSG-029	Mill Yard	Ore Receiving	
MSG-030	Mill Yard	Grinding Bay	
MSG-031	Mill Yard	Ore Receiving	
MSG-032	Mill Yard	Ore Receiving/Crushing Plant	
MSG-033	Mill Yard	Concentrator Building	
MSG-034	Mill Yard	Laboratory Building	UPD-24
MSG-035	Tailings		Secondary source

Station	**Category**	**Associated Feature**	
		Historical	**Modern**
MSG-036	Tailings		Well 0437/ Secondary source
MSG-037	Tailings		
MSG-038	North Off-pile		Secondary source
MSG-039	North Off-pile	Ore Storage	
MSG-040	North Off-pile	Ore Storage	
MSG-041	North Off-pile	Ore Storage	
MSG-042	North Off-pile	Ore Storage	
MSG-043	North Off-pile	Ore Storage	
MSG-044	North Off-pile	Ore Storage	
MSG-045	North Off-pile	Ore Storage	
MSG-046	North Off-pile	Ore Storage	
MSG-047	North Off-pile		Riverbank
MSG-048	North Off-pile		Riverbank
MSG-049	North Off-pile		Riverbank
MSG-050	North Off-pile		Riverbank
MSG-051	North Off-pile		Riverbank
MSG-052	North Off-pile		Riverbank
MSG-053	Well Field		CA / Well MW-3
MSG-054	Well Field		CA
MSG-055	Well Field		CA / Well 0814
MSG-056	Well Field		Riverbank / Well 0403
MSG-057	Well Field		Riverbank / Well 0407
MSG-058	Well Field		Riverbank

4.2.2 Sample Collection Approach

Soil gas collection involved use of a soil gas probe to penetrate the ground to the desired depth (~ 0.5 -0.9 meter), followed by collection of soil gas into Tedlar® bags using either a hand pump or hand-held battery pump. Pilot holes were dug using a solid rod adaptor to the soil probe prior to driving with the sampling rod. A sliding hammer was used to help with driving the probes into the subsurface (Figure 4-5). At each location, two Tedlar® bags (1 L and 3 L) were filled with soil gas for onsite (in nearby laboratory space) measurement with the Rn monitor and PAS, respectively (Figure 4-6).

Figure 4-5. Driving a Soil Gas Probe for Soil Gas Sample Collection

The terrain of the Moab site proved difficult for established sampling techniques and resulted in modifications being made to the methods used during the survey. Very fine-grained dry sand made it difficult to install and to remove the probes using the manual sliding hammer. The fine dry sediments repeatedly clogged sampling equipment, ultimately slowing throughput and causing excessive wear to equipment (e.g., damaging sampling pumps). In addition, some sampling locations were more compact than expected and others contained rocks which also made sample collection difficult in certain areas. In the CA for example, the ground was compacted from vehicle and equipment traffic. To improve sample collection and decrease sample collection time, the team adapted the sampling approach during the weeklong survey. All sites were cleared for underground utilities to assure safety prior to sampling (“digsafe”). The initial and adapted sampling methods for this survey are summarized below:

- Baseline (initial) Method: Samples were collected using a standard reusable stainless steel soil gas probe kit and supporting items such as a sliding hammer and a solid rod with tip for driving holes in resistant material (Items 427.01, 400.99 and accessories, AMS, American Fall ID). Sampling protocols consisted of driving the soil gas sampling probe to depth (0.6 to 0.9 m) using a sliding hammer. For sites where significant resistance was encountered, a solid rod was driven first to make a pilot hole and removed before the soil gas probe driven to depth. The pilot holes filled with the surficial fine, dry powdery material present at the site. The sliding hammer drive adapter was removed from the probe assembly and the central support rod was removed. A tubing adapter was connected to the probe assembly. Using a 1L/min battery powered sampling pump or a manual sampling pump, the probe was evacuated by pulling 1 L of gas. Then, two Tedlar® bags (1 L and 3 L) were collected and labeled for analysis. The sample location was recorded using a multi-network GPS (Garmin Model GPSMAP 65S; accuracy +/- 0.7 m). This baseline method encountered several issues, including difficulty in driving and removing the sample probe (fine dry material "sand-locked" the probe, some very compacted sites, and the presence of some small stones or cobbles). This resulted in significant physical exertion and strain on the sampling crew, as well as resulted in deforming and damaging the sampling equipment. Further, the fine dry silt and fine-grained sand were drawn into the gas stream resulting in damage to sampling pumps and poor sealing of the tube fitting. Based on these issues, an alternative sampling system was purchased and sent to the site with expedited shipping.

- The modified method of sample collection made use of single use barbed adaptors ("implants") that are left in place after installation (Item 211 and accessories, AMS, American Fall ID). Like the baseline method, the barbed metal tips are driven into the subsurface to the desired depth using a sliding hammer and using a pilot hole if needed. However, the metal soil gas tips are left in place and fit with tubing that ultimately extends up to the surface for gas sampling after the drive pipe is withdrawn. Samples can be collected through this tubing after installation, but the implants remain in place after the completion of this survey to allow for future sampling if needed.
- The new system increased sampling efficiency since the extraction of the probe is not needed and since a jack was purchased to help pull out the drive tube. After the first few samples, the team determined that the modified system still generated excessive amounts of fine silt and fine-grained sand in the gas samples. To remedy this, the sample holes in the probe tips were wrapped with a small strip of standard medical gauze pad and secured with a spot of clear tape. This wrap eliminated issues of solids in the samples. There was also still some difficulty in achieving an annular seal by the fine dry fine-grained surficial deposits at Moab (see below).
- Lessons learned: If future soil gas sampling is needed at the Moab site, the modified method was reasonable and robust and is recommended. Possible additional improvements based on the field experience and lessons learned include a) availability of a hammer drill and auger capability for pilot hole installation, and b) possible use of a small quantity of bentonite powder and a water in each sampling location around the tubing followed by a waiting period to assure that there is no annular air inleakage.

All soil gas samples were analyzed by both a radon monitor and PAS. Carbon dioxide (CO_2) measurements (made with the PAS) were used to assess soil gas sample quality during the survey and identify any samples that may have been diluted by surface air during collection. CO_2 serves as a qualitative indicator that soil gas samples collected are representative of the subsurface instead of the surface atmosphere. Low CO_2 concentrations are indicative of soil gas samples collected with poor annular sealing that have been diluted by the surface atmosphere. Elevated stable concentrations indicate that sample collection was successful, and the results are representative of the vadose zone. Any samples with CO_2 concentrations below 1000 ppmv were adjusted on a case-by-case basis to account for sample dilution. The 1000 ppmv threshold was determined based on reference data published by the National Ecological Observatory Network (NEON) for their reference site in Moab, Utah during late fall/ early winter months (NEON, 2024). This NEON reference station is sampled approximately monthly at a shallow (< 1 m) depth, similar to the approach used in this survey.

Figure 4-6. Collection of Soil Gas with Hand-Pump

4.3 Radon Analysis- Residual Uranium and Radium Indicator Gases

4.3.1 Conceptual Basis

The decay chain of uranium includes Rn, a radioactive noble gas. Soil gas Rn surveys and related methods (e.g., mapping bismuth 214 anomalies using walkover or aerial gamma surveys) have documented in the literature for identifying residual radioactive contamination associated with mining/milling activities and mill tailings (Nielson et al., 1981; Karp, 1988; Fuhrman et al., 2021) and for identifying areas of natural uranium mineral deposits in the subsurface (Denham and Looney, 2007). Soil gas surveys are a simple, low-cost, and effective strategy for identifying residual sources in cases where the uranium and associated decay-chain products (notably radium) are present in the subsurface.

Figure 4-7 depicts the decay chain for the most abundant naturally occurring isotopes of uranium and thorium, uranium 238 (^{238}U) and thorium 232 (^{232}Th). For each isotope and decay product, the half-life, type of decay, and progeny (if radioactive) is noted along with other key information. Rn in each decay chain is highlighted (*) in Figure 4-7. The presence of natural uranium and thorium in the earth's crust typically results in the worldwide presence of elevated levels of Rn in soil gas. The amount of Rn present in any location is a function of the amount of uranium and thorium present, as well as other factors such as mineralogy, moisture content, material properties, and meteorology/driving forces.

As presented in Figure 4-7, radon is an indicator of the overall uranium and thorium decay chain since it is formed from the direct decay of radium. Radon requires the presence of the intermediate elements and isotopes that are formed over time, and which are typically in secular equilibrium with the uranium and thorium in mineral ores and tailings.

For uranium mining and milling sites, the presence of ores and tailings (materials with elevated levels of uranium and thorium and associated decay products) results in an increase in shallow soil gas Rn. This signal corresponds for to uranium impacted areas – i.e., areas in/overlying accumulation zones of uranium-related constituents throughout the vadose zone and in the shallow groundwater.

Sampling and analysis of soil gas collected from a grid of locations allows straightforward mapping of Rn anomalies to help identify and provide insights on where residual primary and secondary uranium source materials may be located.

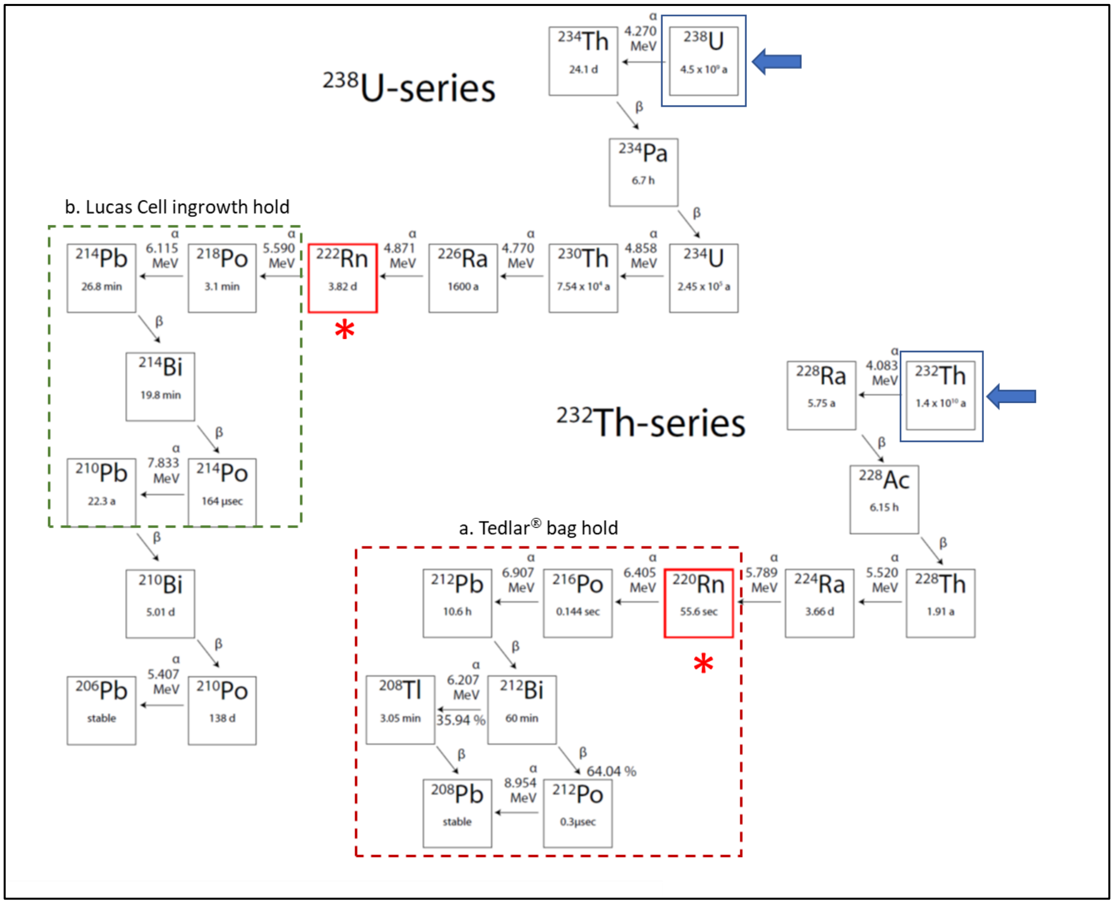

Figure 4-7. Annotated Decay Chain for 238U and 232Th (←) Highlighting Radon (*) and Protocol Development Topics (a/b)

4.3.2 Radon Analysis Paradigm– Synopsis

A method using an RDA-200 monitor for radon 222 (^{222}Rn) measurement in soil gas samples was developed in support of this effort (Looney, 2023)[1]. Soil gas samples were collected in 1 L Tedlar® bags, transferred to EDA Lucas Cells, then counted with the Rn monitor (field deployable alpha scintillation counting). Each Lucas Cell was pre-evacuated and counted to determine the background for the cell before the sample was introduced. The interior of the Lucas cells is coated with zinc sulfide doped with silver (ZnS(Ag)), which emits a light photon when struck by an alpha particle.

The Rn monitor counts photons from the decay of Rn gas and “daughters” in the sample (counting of alpha decays resulting from Rn and progeny in the sample) after a set hold time and then predetermined calibration factors can be used to calculate activities. A hold time in the sample transfer (Tedlar®) bag is incorporated into the ^{222}Rn analysis protocol to allow for complete decay of short-lived ^{220}Rn gas so that it is not introduced into the Lucas Cell and will not contribute to the alpha counts.

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[1] The radon analysis protocols developed for this effort are documented in more detail in Appendix A and in Looney et al. (2023).

After the Tedlar® bag hold time was complete, the sample gas was introduced into a pre-evacuated Lucas Cell after passing through a glass fiber filter and polyester fiber cartridge to remove any solids and remove daughters that were produced by the decay of ^{220}Rn. The sample in the Lucas Cell was then held for a known time to allow controlled ingrowth and equilibration of ^{222}Rn progeny. The count rate was converted to a decay corrected Rn level using predetermined Lucas Cell calibration factors for each type of cell and supplementary information such as sampling time, counting time, background counts, local barometric pressure, loading vacuum preset, and Lucas Cell ingrowth hold time. Prior to the survey, the Lucas Cells were calibrated to known Rn concentrations measured by an instrument that is certified by, and traceable to, the National Radon Proficiency Program (NRPP) and National Radon Safety Board (NRSB).

4.4 Photoacoustic Spectrometer – Residual Ammonium Indicator Gases

4.4.1 Conceptual Basis

The use of soil gas surveys for residual sources of ammonium/NH_3 is based on the known microbial transformation pathways of nitrogen in the environment (Cheng et al., 2022). Figure 4-8 is a diagram that combines the information from the referenced scientific literature, integrating the various known pathways for a range of biogeochemical conditions. The figure summarizes the nitrogen species, sequential transformations in each pathway, and the associated enzymes. Importantly, there are several gaseous species in Figure 4-8. These gases are identified using a gold color and are potential targets for inclusion in soil gas surveys.

NH_3 is a primary source material that has a high vapor pressure. Thus, NH_3 is a potential soil gas analyte that provides a direct measure of residual contamination. However, the ammonium/NH_3 relationship and associated partitioning between the aqueous and gas phases is strongly influenced by pH. The pKa of NH_4^+ is 9.25 and the aqueous NH_4^+ ion is the predominant form of NH_4^+/NH_3 at pH below 9. The presence of NH_3 in the gas phase is influenced by both the presence of residual source material and the chemistry of the soil pore water, with typical pH values limiting the amount of NH_3 in the soil gas. In this scenario, a significant concentration of NH_3 is measured in soil gas would be a compelling qualitative indicator of nearby (or underlying) NH_4^+/NH_3 in the vadose zone or shallow groundwater.

N_2O, nitric oxide (NO), and nitrogen (N_2) are gaseous products of various redox pathways. The measurement of significant N_2O and NO provides confirmatory evidence of both: a) the presence of residual NH_4^+/NH_3 source material, and b) ongoing-active microbial ammonium transformation in the subsurface and measurable attenuation of ammonium. Based on the literature, NH_3 and N_2O were selected as the primary soil gas metrics for ammonium/NH_3, along with bulk gases that provide a general indication of biological activity (CO_2 and CH_4). NO is typically present in low concentration and N_2 is a major component of the atmosphere and baseline soil gas. Therefore, these two nitrogen species were not selected as target analytes.

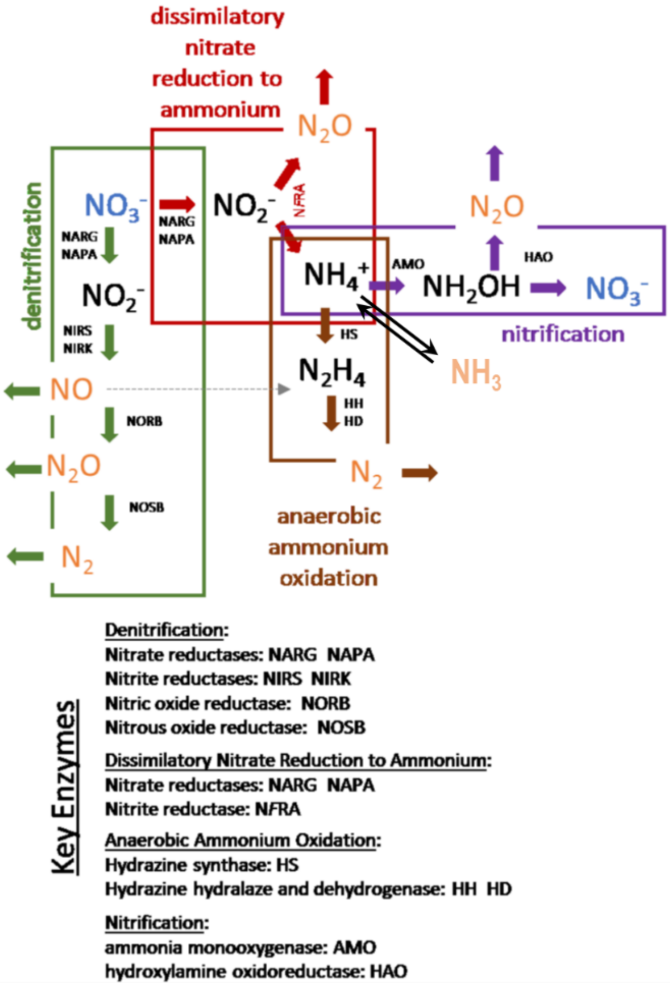

Figure 4-8. Biological Nitrogen Transformation Pathways and Associated Key Enzymes with Gases Shown in Gold (arrows exiting boxes)

4.4.2 Ammonium/Ammonia Indicator Gas Analysis Paradigm–- Synopsis

An INNOVA 1312 Photoacoustic Multi-gas Monitor was used for measuring gas-phase surrogate indicators of ammonium, including NH_3 and N_2O. The PAS was also calibrated to measure general indicator soil gases, including CH_4, CO_2, and water vapor. Soil gas samples are introduced into the instrument for measurement from Tedlar® bags. Specialized optical filters allow for selective detection of different gases within a mixed sample.

Once inside the PAS, the gas is exposed to pulsed light, some of which is absorbed by the gas. The amount of light energy absorbed is proportional to concentration. If the target analyte is present, the absorbed light is then released as heat, which generates pressure changes. Since the light input is pulsed ("chopped"), the pressure changes result in sound waves that are detected by an internal microphone. If the gas contains many chemicals, optical filters are used to resolve the mixture and allow for selective analyte detection.

Volumetrically mixed gas standards were used for calibration of all filters. Calibrations also included corrections for humidity, CO_2 and cross interferences. These corrections were applied for measuring soil gas samples collected in the field. A MESA reference (calibration) gas mixture containing known concentrations of CH_4, CO_2, and N_2O was also used for validating the calibration.

5.0 Results and Discussion

The data were organized into various bins that represent different levels of granularity at the Moab site, specifically: 1) overall data set, 2) area data sets, and 3) feature level data sets. The binned results are presented below followed by a discussion that presents the soil gas data in context with previous studies and with measured groundwater concentration information.

5.1 Overall Data Set

Rn, N_2O, NH_3, CH_4, and CO_2 were measured in soil gas samples collected throughout the Moab site in November 2023. A complete set of these results (coordinates, activity/concentration, and notes) for all sampling locations is included in Appendix C.

Table 5-1 summarizes overall data for each constituent in the survey including the median and quartile values and range (minimum and maximum). Significant variations were measured in the data for Rn (4 orders of magnitude), N_2O (> 3 orders of magnitude), CO_2 (> 2 orders of magnitude), CH_4 and NH_3 (both about 1.5 orders of magnitude). A more detailed look at each analyte at the area and feature level summaries, as well as a context discussion, are provided below.

Table 5-1. Soil Gas Data (2023) Overall Summary Statistics.

	Radon (pCi/L)	**Nitrous Oxide (ppmv)**	**Ammonia (ppmv)**	**Methane (ppmv)**	**Carbon Dioxide (ppmv)**
minimum	12	0	0	1	1,000
1Q	113	0	0	4	1,307
median	199	1	1	5	2,427
3Q	2,020	4	2	10	5,538
maximum	143,136	3,980	23	56	71,285

Sitewide maps have been generated for Rn, NO, NH_3, CH_4, and CO_2 (Figure 5-1, Figure 5-3, Figure 5-5, Figure 5-7, and Figure 5-9, respectively). The color and size of the circles on the maps indicate concentration of the analyte being measured, the darker and the larger the spot, the higher the concentration. Bar charts of the data, sorted from low to high, showing the activity/concentration for each constituent in the dataset are shown in Figure 5-2, Figure 5-4, Figure 5-5, Figure 5-7, and Figure 5-9for Rn, NO, NH_3, CH_4, and CO_2, respectively.

5.1.1 Radon

^{222}Rn was used in this study as an indicator of residual uranium/radium contamination (Figure 5-1). This strategy has been demonstrated in the literature (Mudd, 2008; Fukui, 2007) for mapping localized residual uranium source material. Rn in soil gas is a function of the natural or perturbed geologic conditions (e.g., the presence of rocks containing uranium, thorium, and progeny), moisture content, weather, vadose zone thickness, proximity to cover materials or structures, and other factors. Higher soil gas Rn activity is typically measured in areas that have rocks/deposits with high uranium/thorium/radium content. Because caps and covers limit the efflux of Rn, higher Rn is also typically measured after rain events (generating moderately high moisture content), beneath covers or structures, and in areas where the vadose zone is thicker.

The radon results were binned into three groups. Low activities (< 1,000 pCi/L) are similar to natural background measurements in shallow soil gas around the world (Adepelumi et al., 2005; Kemski et al., 2005; Kemski et al., 2007; King and Minissale, 1994; Gunderson, 1991). Elevated activities (1,000 to 8,000 pCi/L) are typical of the radon levels measured in previous soil gas studies at sites impacted by tailings (Mudd, 2008; Fukui, 2007). Very high activities (> 8,000 to 143,136 pCi/L) indicate a significant quantity of concentrated-localized residual uranium source material in the vadose zone or shallow groundwater. As displayed in Figure 5-2, of the 58 locations sampled, 39 locations (67%) were classified as low, 14 locations (24%) were classified as elevated, and five locations (9%) were classified as very high.

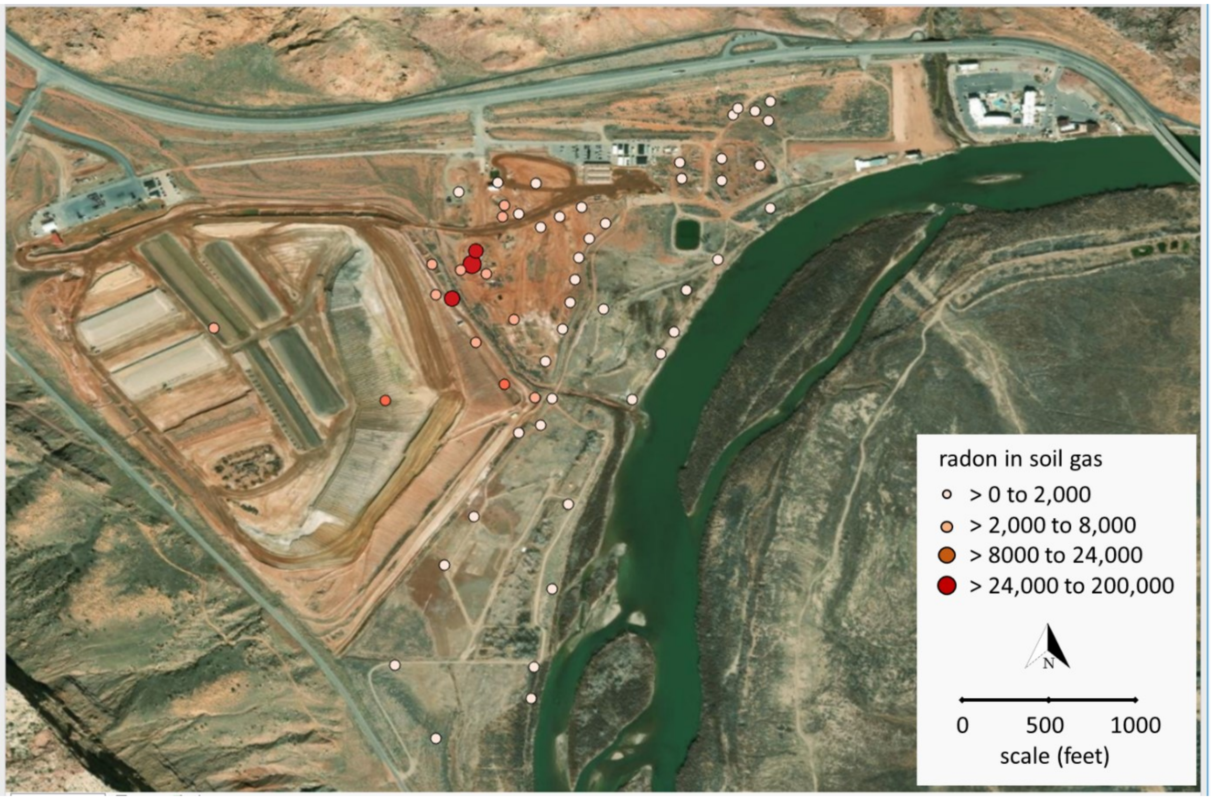

Figure 5-1. Radon Soil Gas Survey Data

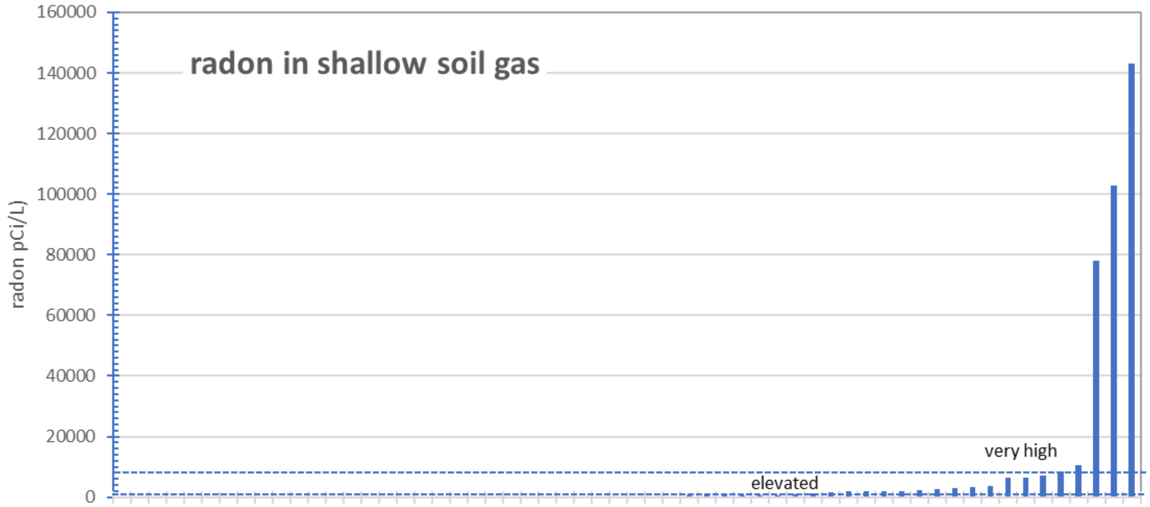

Figure 5-2. Measured Radon Soil Gas Activities – histogram sorted from low to high.

5.1.2 Nitrous Oxide

N_2O is produced by various pathways throughout the nitrogen cycle, including denitrification of nitrate, oxidation of ammonia, and nitrite reduction. Since N_2O is a greenhouse gas, several studies document the concentration of nitrous oxide in soil gas and efflux to the atmosphere from both agricultural and non-agricultural areas. The natural background of nitrous oxide in soil gas is typically < 1 ppmv, ranging up to higher values in agricultural areas that have been impacted by nitrogen fertilizers.

N_2O is a gas that serves as an indirect indicator for residual NH_4^+/ NH_3 inventory/sources in the vadose zone or shallow groundwater. In addition, it is an indicator of NH_4^+/ NH_3 oxidation, biological activity, and nitrogen cycling in the subsurface ecosystem. Significant concentrations of a nitrogen precursor (NH_4^+ and NH_3 for the Moab site) or parent must be present as a source material to produce elevated N_2O concentrations.

The soil gas survey results for N_2O are presented in Figure 5-3 and Figure 5-4. The overall dataset for N_2O was binned into three groups: Low (< 4 ppmv), elevated (4 to 400 ppmv), and very high concentrations (> 400 to 3,980 ppmv). As presented in Figure 5-4, 43 locations (74%) were classified as low, 14 locations (24%) were classified as elevated, and one location (2%) was classified as very high.

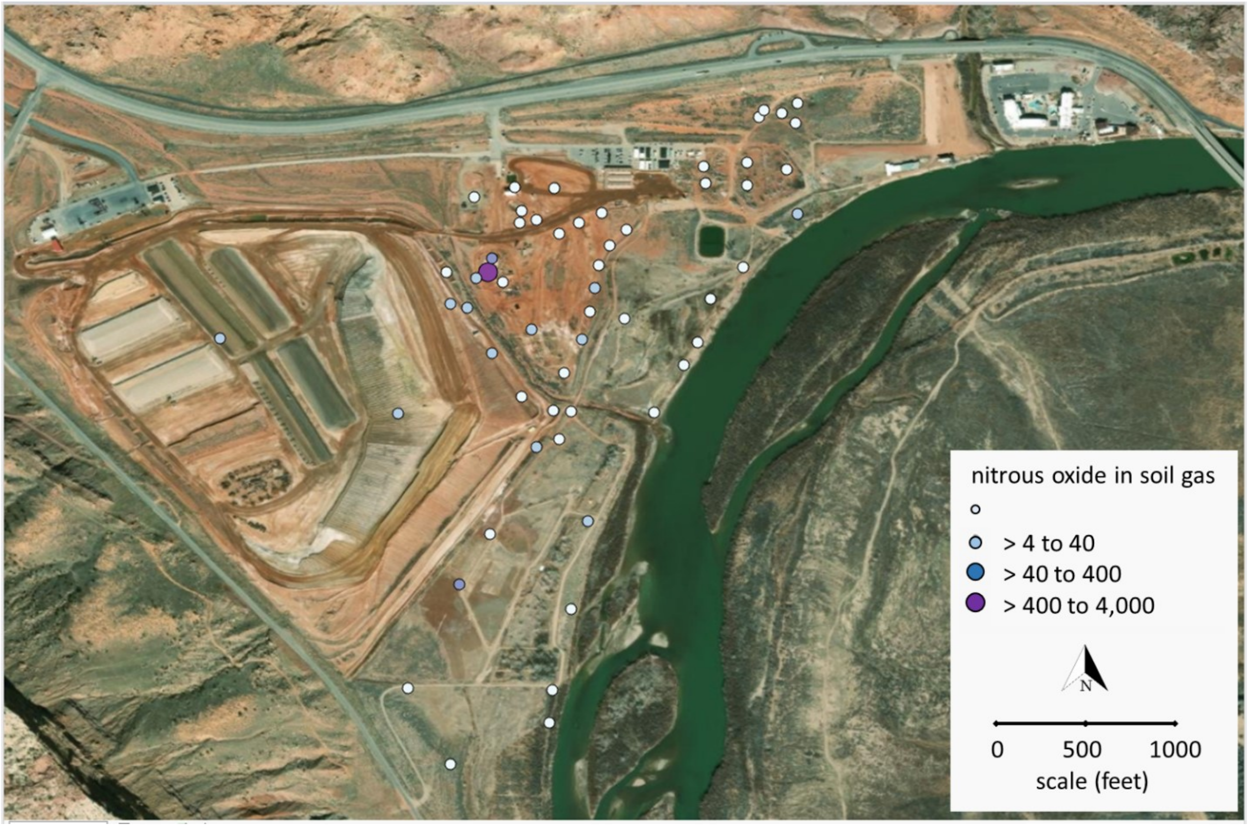

Figure 5-3. Nitrous Oxide Soil Gas Survey Data

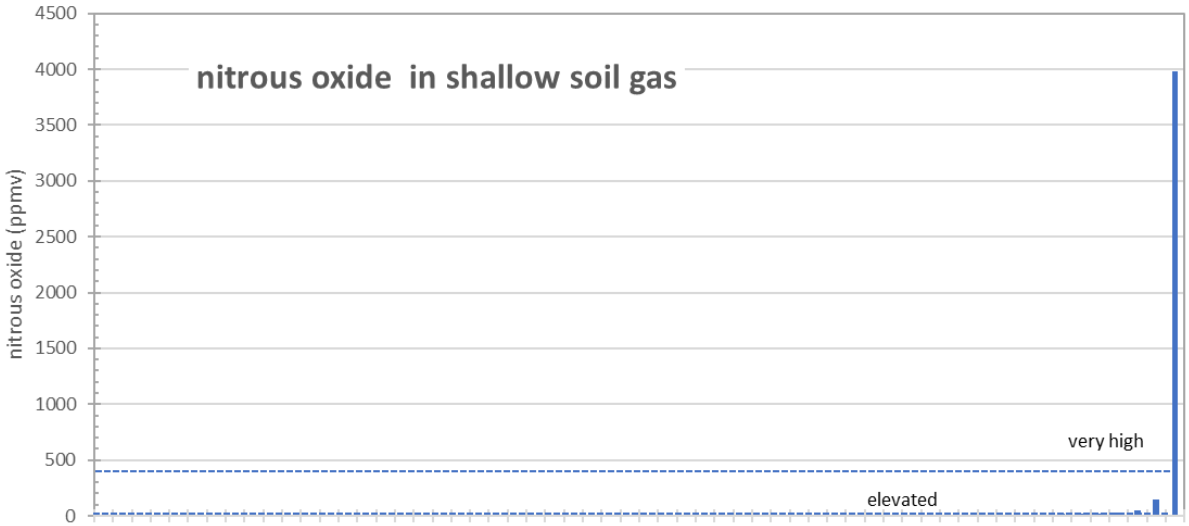

Figure 5-4. Measured Nitrous Oxide Soil Gas Concentrations – histogram sorted from low to high.

5.1.3 Ammonia

NH_3 was used as a direct gas phase indicator of residual NH_4^+/NH_3. The use of NH_3 as a direct indicator of total NH_4^+/NH_3 nitrogen inventory in the soil and vadose zone is not as well documented compared to the use of Rn for uranium. However, the extensive historical NH_3 use in the Moab millsite processes and the ease of field detection were used as a basis for sampling and analyzing NH_3 with the objective to indicate potential areas of significant residual source mass. The fraction of NH_4^+/NH_3 that is present as gaseous NH_3 is strongly pH dependent, so NH_3 gas concentrations may not track total NH_4^+/NH_3 inventory with a high fidelity, particularly at circumneutral pHs (below pH 10) where a significant fraction of the NH_4^+/ NH_3 is in the ionic (NH_4^+ form).

The soil gas survey results for NH_3 are depicted in Figure 5-5 and Figure 5-6. The NH_3 results were divided into low (< 1 ppmv), elevated (1 to 10 ppmv), and very high concentrations (> 10 to 23 ppmv). As presented in Figure 5-6, 40 locations (69%) were classified as low, 13 locations (22%) were classified as elevated, and five locations (9%) were classified as very high.

Figure 5-5. Ammonia Soil Gas Survey Data

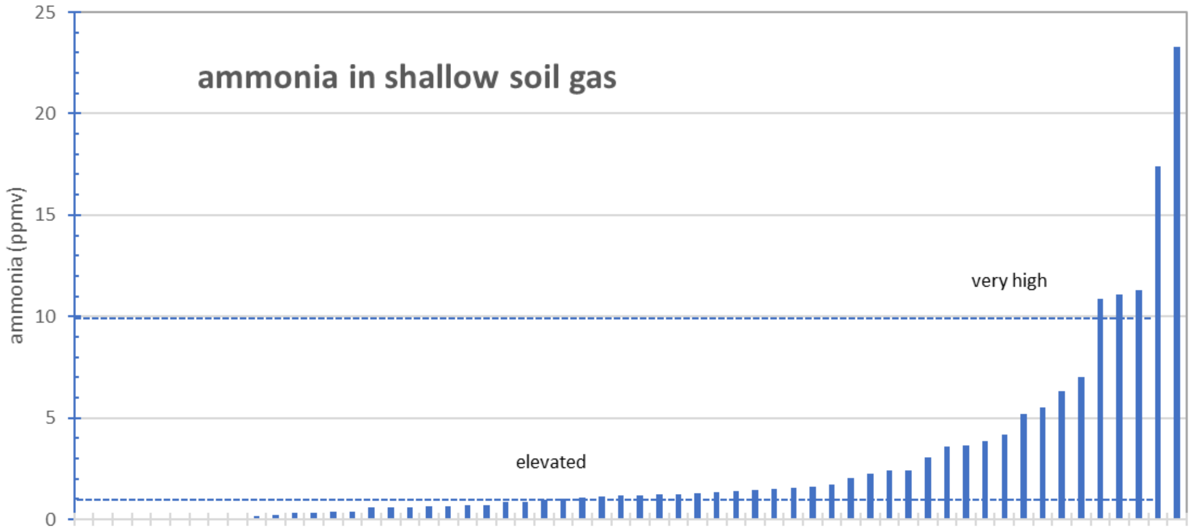

Figure 5-6. Measured Ammonia Soil Gas Concentrations – sorted from low to high.

5.1.4 Methane

CH_4 in soil gas was used as a geochemical indicator of microbial activity. Lower concentrations of CH_4 in soil gas are typically found in areas where microbial activity is low, in aerobic systems where oxygen is present and in oligotrophic systems where carbon/electron donors and nutrients are low. In contrast, elevated CH_4 would be measured at locations where there is a significant and active subsurface microbial community, where there is limited oxygen available so that the system is anaerobic or contains anoxic regions, and in eutrophic or nutrient rich areas.

The soil gas survey results for methane are depicted in Figure 5-7and Figure 5-8. The overall dataset for CH_4 was roughly binned into three groups. Low (< 10 ppmv), elevated (10 to 25 ppmv), and very high concentrations (> 25 to 56 ppmv). As displayed in Figure 5-8, 42 locations (72%) were classified as low, 13 locations (22%) were classified as elevated, and three locations (6%) were classified as very high.

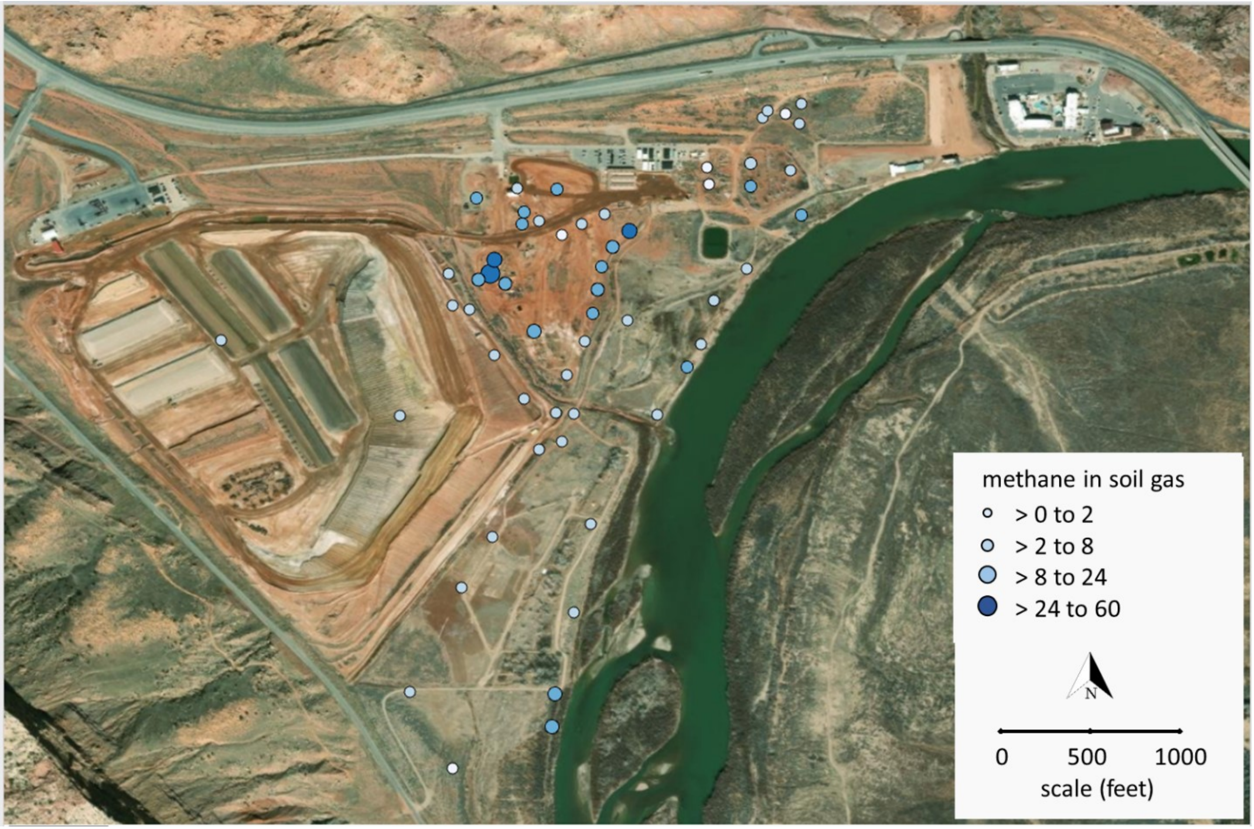

Figure 5-7. Methane Soil Gas Survey Data

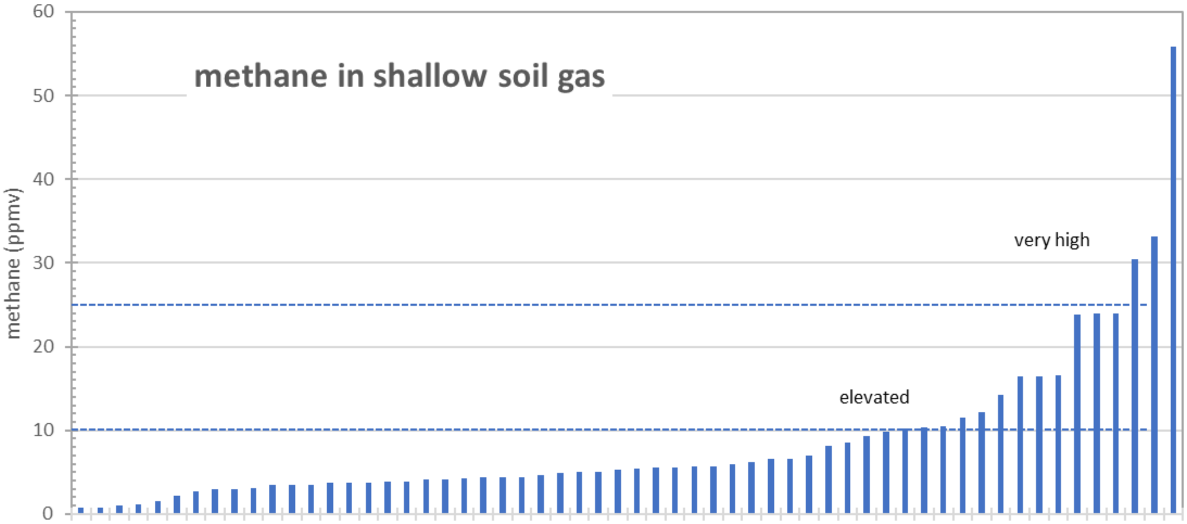

Figure 5-8. Measured Methane Soil Gas Concentrations – sorted from low to high.

5.1.5 Carbon Dioxide

CO_2 is elevated in almost all soil gas samples and was used as a composite indicator of sample integrity and microbial activity. The soil gas CO_2 data are depicted in Figure 5-9 and Figure 5-10. High CO_2 concentrations in a soil gas sample (1000 ppmv or more) increase confidence that the sample is representative of the vadose zone. Conversely, a low CO_2 suggests that there has been dilution of the sample with surface atmosphere due to sample probe annular leakage or a poor tubing connection. In cases where the CO_2 in a soil gas sample was below 1000 ppmv during the survey, we adjusted the data to account for that dilution on a case-by-case basis as noted in Appendix C. Like CH_4, elevated CO_2 would be detected at locations where there is an active subsurface microbial community.

The soil gas survey results for CO_2 are depicted in Figure 5-9 and Figure 5-10. The overall dataset for CO_2 was roughly binned into three groups. Low (< 5,000 ppmv), elevated (5,000 to 20,000 ppmv), and very high concentrations (> 20,000 to 71,285 ppmv). As displayed in Figure 5-10, of the 58 samples collected 42 locations (72%) were classified as low, 14 locations (24%) were classified as elevated, and two locations (4%) were classified as very high.

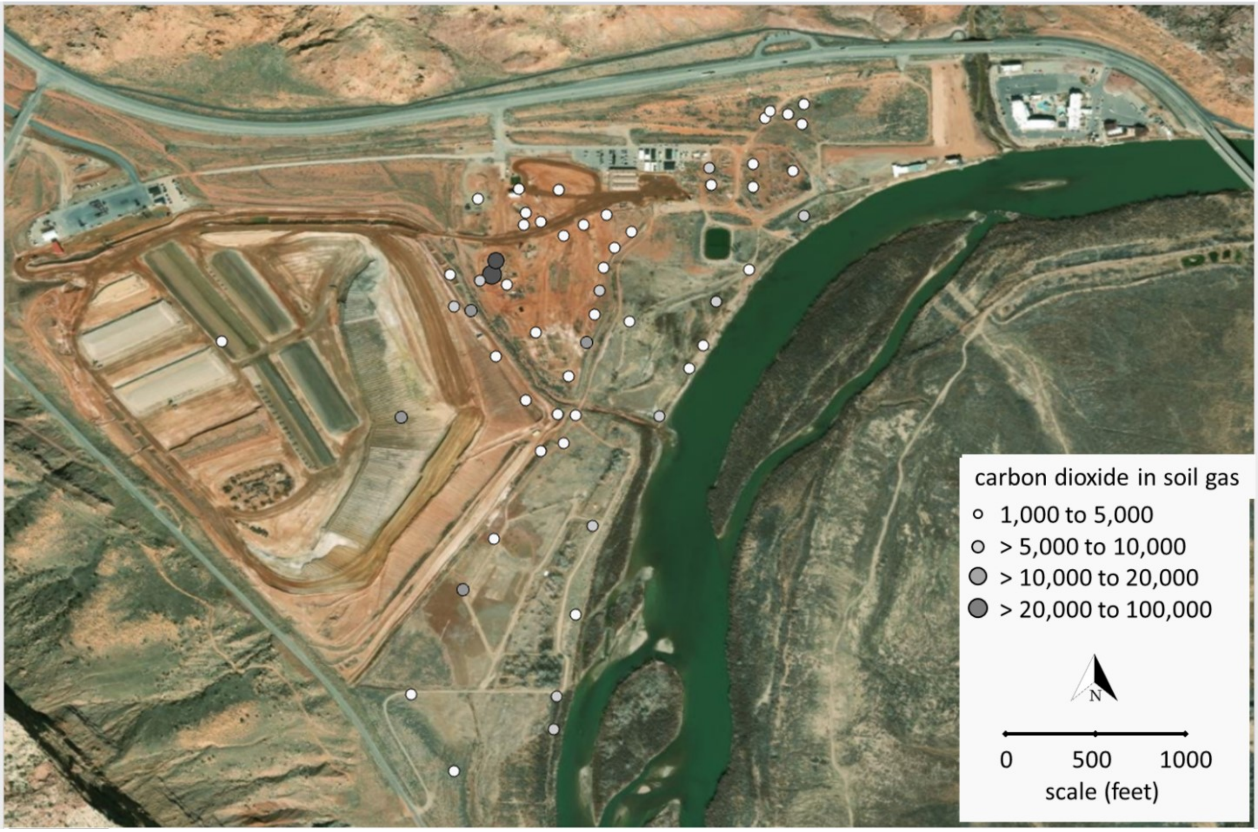

Figure 5-9. Carbon Dioxide Soil Gas Survey Data

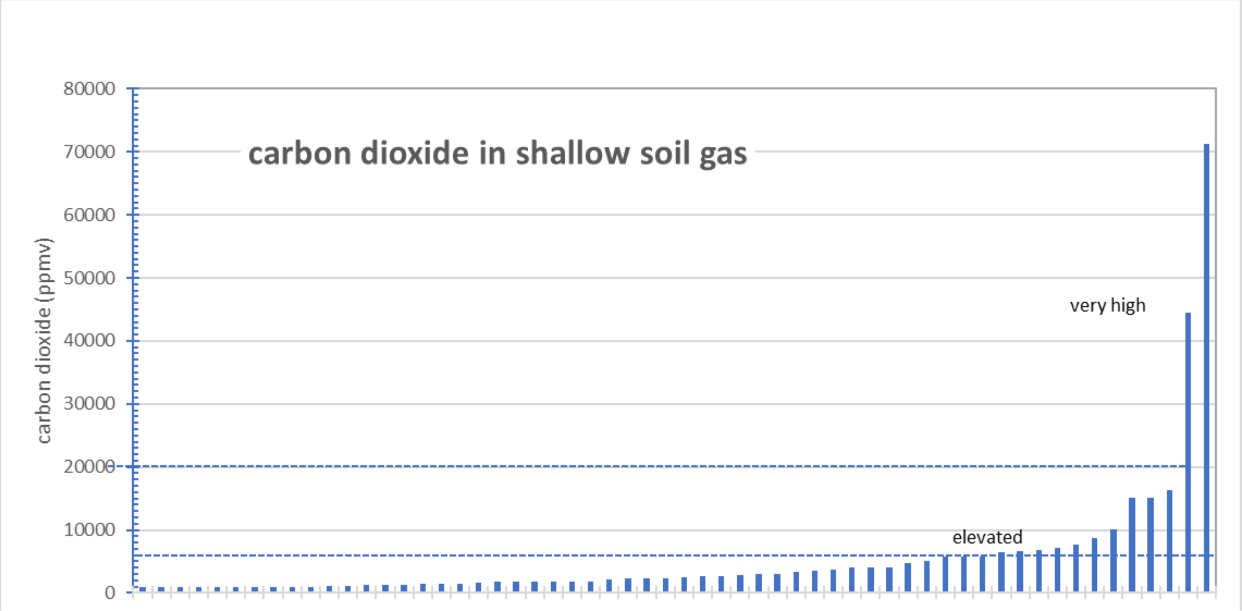

Figure 5-10. Measured Carbon Dioxide Soil Gas Concentrations

5.2 Area Data Summaries

The sampling strategy identified four target areas: Mill Yard, North Off-Pile, Tailings and Wellfield/Riverbank, which represent general zones of potential interest and provide some mesoscale spatial granularity and insights about the different areas. Table 5-2 provides a summary of the Moab soil gas survey by area and Appendix D provides the detailed data for the individual samples collected in each area.

Table 5-2. Soil Gas Data – Area Summary Statistics.

Area	**Radon**				
	Minimum	**1Q**	**median**	**3Q**	**maximum**
Mill Yard	48	183	**878**	2,165	143,136
North Off-Pile	12	58	**79**	118	751
Tailings	470	2,250	**3,290**	7,882	102,982
Wellfield/Riverbank	96	148	**178**	205	413

Area	**Nitrous Oxide**				
	Minimum	**1Q**	**median**	**3Q**	**maximum**
Mill Yard	0	0.0	**0.0**	5.1	3,980
North Off-Pile	0	0.0	**1.5**	2.3	4
Tailings	0	0.4	**5.6**	15.4	27
Wellfield/Riverbank	0	0.8	**1.4**	1.8	48

Area	**Ammonia**				
	Minimum	**1Q**	**median**	**3Q**	**maximum**
Mill Yard	0	1.0	**2.1**	4.2	23
North Off-Pile	0	0.2	**0.6**	1.2	2
Tailings	0	0.9	**1.3**	1.7	5
Wellfield/Riverbank	0	0.7	**1.2**	2.9	11

Area	**Methane**				
	Minimum	**1Q**	**median**	**3Q**	**maximum**
Mill Yard	1	6	**11**	17	56
North Off-Pile	1	4	**4**	6	10
Tailings	2	3	**5**	6	7
Wellfield/Riverbank	1	4	**4**	5	24

Area	**Carbon Dioxide**				
	Minimum	**1Q**	**median**	**3Q**	**maximum**
Mill Yard	1000	1,000	**2,348**	4366	71,285
North Off-Pile	1000	1,198	**1,773**	4062	7,691
Tailings	1433	2,084	**3,571**	5263	16,378
Wellfield/Riverbank	1,000	2,188	**3,641**	6,870	10,067

The following narratives summarize the area concentration results for each target analyte. To supplement the tabulated data, a set of pie charts was prepared to aid in visualizing the data and the differences between the areas. Each pie chart depicts the distribution of sample locations in the dataset. The segments and colors represent the proportion of sample locations that were designated as low (green), elevated (yellow), and very high (orange) using the same criteria described for each analyte in Section 5.1. The designated bins are a tool to distinguish low samples from high samples on an order of magnitude basis. Data in the elevated and very high groupings can result from either anthropogenic impacts (e.g., mill operations, ore storage, or tailings/waste disposal) or from natural processes (e.g., microbial production of carbon dioxide or methane). For the analytes, five pie charts were prepared, one for the overall dataset and one each for the Mill Yard, North Off pile, Tailings and Wellfield/Riverbank.

5.2.1 Radon

Elevated Rn soil gas concentrations were detected throughout areas where tailings materials containing residual uranium/thorium/radium are still present. In addition, samples collected from specific locations in the Mill Yard) were significantly elevated – particularly in some of the former pond areas where hot spots were clustered (Figure 5-11). The Tailings area had the highest general concentrations (median = 3,290 pCi/L) consistent with the presence of tailings awaiting removal and relocation.

The Mill Yard samples also contained elevated Rn concentrations (median = 878 pCi/L) but were generally lower compared to the tailings area samples. This area had the highest Rn concentration detected in the overall dataset (143,136 pCi/L). Relatively low activities (near typical background concentrations for soil gas) were measured throughout the Well Field and North Off-pile areas (outside of the CA). These general patterns are clear in pie charts for Rn (Figure 5-11). The Well Field and North Offpile areas are completely green (low) with no data in the yellow (elevated) or orange (very high) categories. The Mill Yard area is about 52% green, 38% yellow, and 10% orange. The tailings are more consistently elevated with 10% green, 60% yellow, and 30% orange.

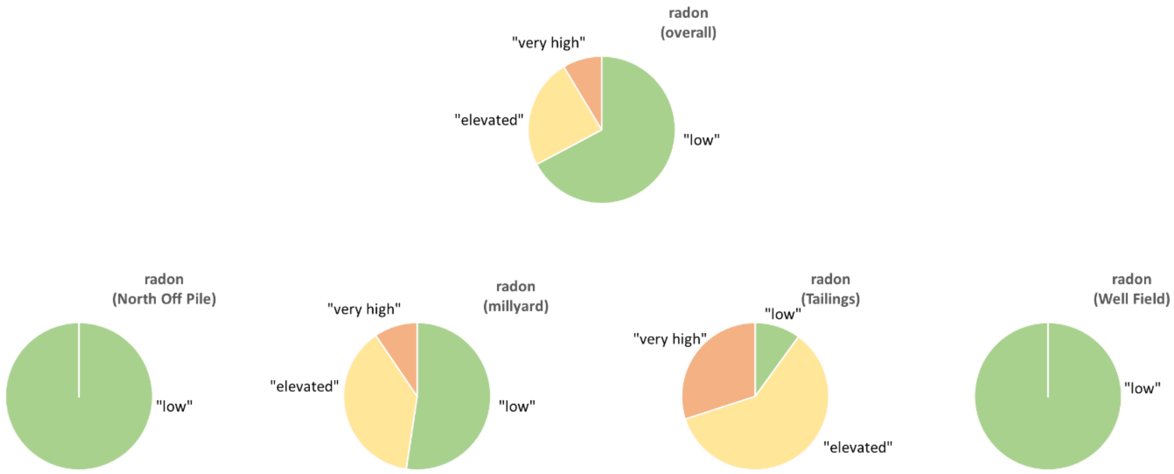

Figure 5-11. Radon Pie Chart Information – Overall and Area-Level Summaries

5.2.2 Nitrous Oxide

At Moab, N_2O concentrations in soil gas were generally low with a few elevated concentrations measured in all areas (Figure 5-12). N_2O is a naturally occurring constituent in the global nitrogen cycle. Elevated and very high concentrations in this study would generally indicate a source of nitrogen, such as NH_4^+/NH_3, and a measurable level of subsurface biological activity that is transforming/removing the nitrogen source. As depicted in Figure 5-12, the concentrations in the North Off-Pile area were generally low (94% green) with one sample (6%) in the yellow-elevated bin. The Mill Yard area is about 71% green, 24% yellow, and 5% orange. No very high N_2O locations were measured in Tailings and Wellfield/Riverbank areas; however, both of these areas had elevated N_2O in a significant number of samples. In the Tailings the nitrous oxide distribution was 40% green and 60% yellow. The N_2O distribution in the Wellfield/Riverbank area was 80% green and 20% yellow. The N_2O data are consistent with the historical sluicing transfer of the tailings into the pile (in NH_4^+ containing water), the general mobility of NH_4^+ moving through the vadose area and shallow groundwater, and the natural presence of a nitrogen cycling microorganisms in subsurface environment.

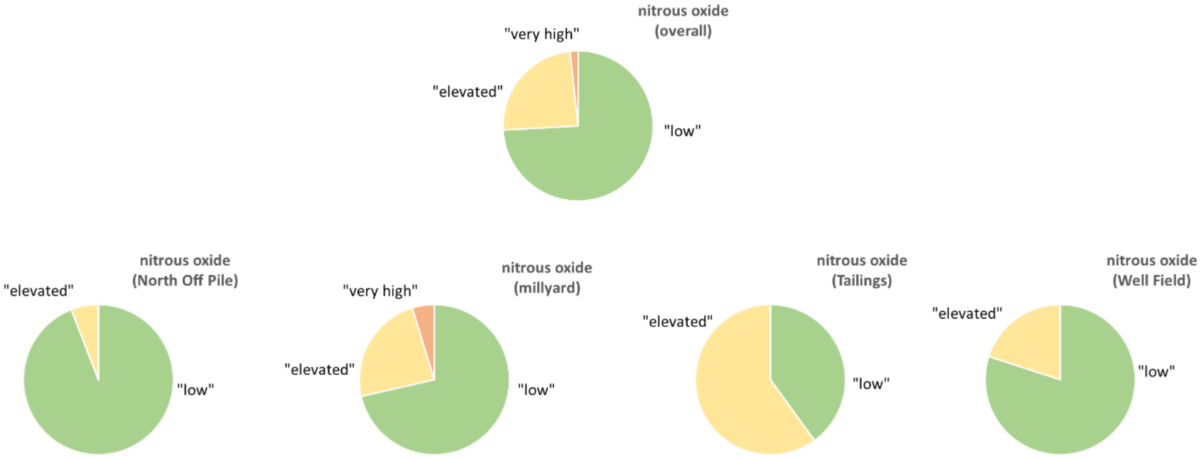

Figure 5-12. Nitrous Oxide Pie Chart Information – Overall and Area-Level Summaries

5.2.2 Ammonia

At Moab, NH_3 concentrations in soil gas ranged widely with elevated levels measured in all areas (Figure 5-13). NH_3 is a naturally occurring constituent in the global nitrogen cycle. Elevated and very high levels in this study would provide a direct (but pH dependent) indication of a residual source of nitrogen in the vadose area or shallow groundwater. As depicted in Figure 5-13, the levels in the North Off-Pile area were generally low (94% green) with one sample (6%) in the yellow-elevated bin. The Mill Yard area was highly variable (about 48% green, 38% yellow, and 14% orange), reflecting the historical diversity of activities in this area. The distribution in the Tailings Area was 80% green and 20% yellow, suggesting some residual NH_4^+ in this area. The NH_3 distribution in the Wellfield/Riverbank area was highly variable with 60% green, 20% yellow, and 20% orange. The NH_3 data are consistent with Mill Yard operations, historical sluicing transfer of the tailings into the pile (in NH_4^+ containing water), and the general mobility of NH_4^+ moving through the vadose area & groundwater. The relatively higher levels in the well field (versus the tailings area) may be indicative of the current flushing status, small changes in pH, or other factors.

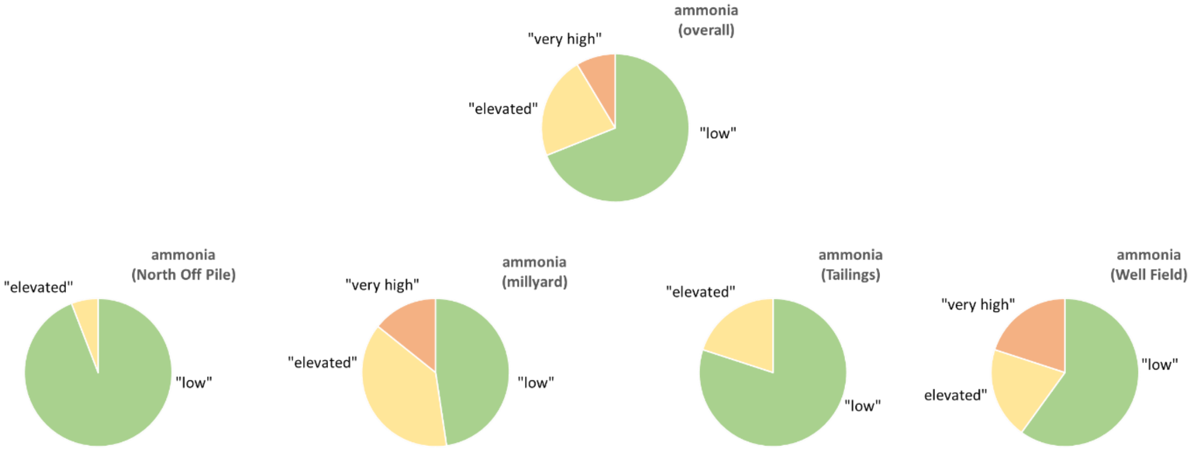

Figure 5-13. Ammonia Pie Chart Information – Overall and Area-Level Summaries

5.2.3 Carbon Dioxide and Methane

CH_4 and CO_2 were both measured as general biogeochemical indicators, specifically indicators of subsurface microbial activity. CH_4 and CO_2 are naturally present in soil gas in many environments. CO_2 is typically elevated in all soil gas samples, while CH_4 levels range more widely. CH_4 is typically low in areas with low microbial activity or areas that are dry or oxidizing, whereas CH_4 increases in areas where there is high microbial activity, high organic carbon, and/or limitations to the transfer of oxygen (e.g., moist areas). As depicted in Figure 5-14 and Figure 5-15, varying levels of these analytes were present in soil gas. Two notable features of the data are: 1) CH_4 is low in the tailings, which is consistent with the thicker vadose zone and suggests more oxidizing conditions and 2) the Mill Yard is the only area with isolated very high levels of both CH_4 and CO_2. More detailed feature level evaluation of the data (below) provides additional information and insights. Significant subsurface biological activity is likely present near the specific sample locations that had very high levels CH_4 and CO_2 in the dataset.

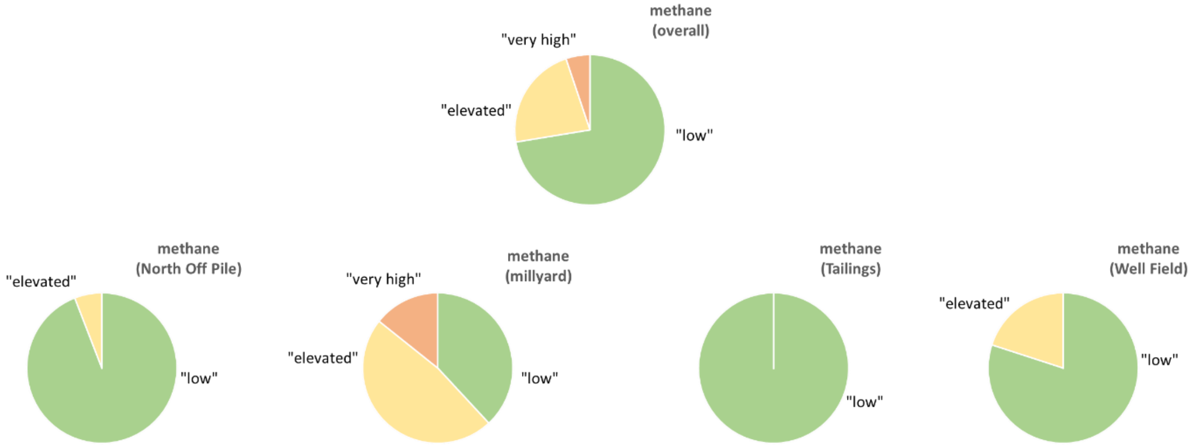

Figure 5-14. Methane Pie Chart Information – Overall and Area-Level Summaries

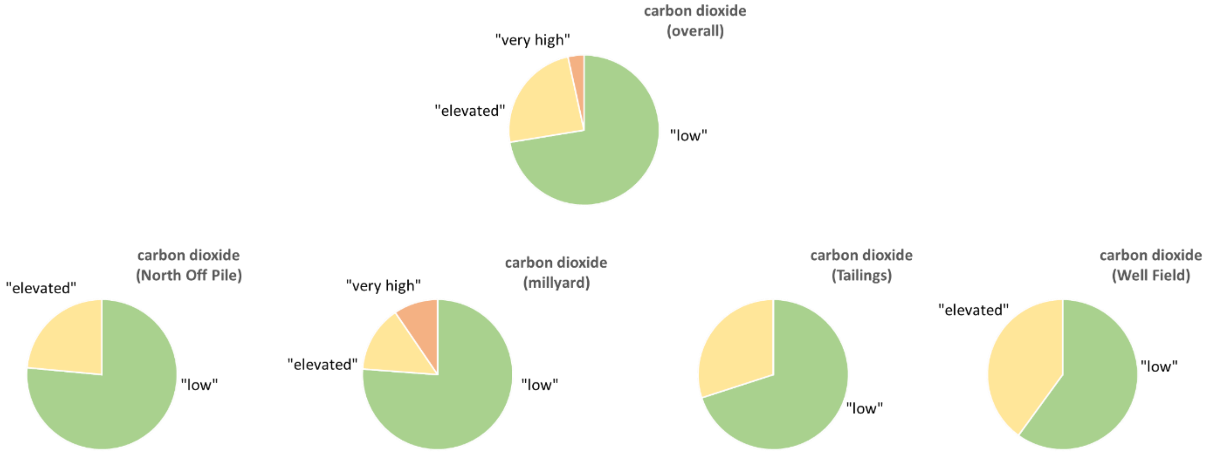

Figure 5-15. Carbon Dioxide Pie Chart Information – Overall and Area-Level Summaries

5.3 Feature Data Summaries

As shown above, increasing the granularity of the data interpretation by organizing the data into areas provided significantly improved insights into environmental management at Moab. To provide further refinements and insights, the soil gas sampling strategy identified several features of potential interest and assigned the soil gas sampling stations to those features.

The eighteen identified features correspond to historical and modern processes/conditions. Historical features in the Mill Yard area included the 1) concentrator building, 2) containment pond, 3) grinding bay, 4) laboratory building, 5) ore receiving, and 6) pond (unknown). Modern features in the Mill Yard area included the 7) CA Boundary, and 8) miscellaneous Mill Yard locations. Historical features in the North Off-pile area included the 9) ore storage area, while modern features in that area included the 10) riverbank area and 11) miscellaneous north off-pile locations. Historical features in the Tailings area included 12) pond (BaCl), 13) Pond (purification), and 14) sump pond, and modern features in the tailings area include 15) miscellaneous tailings locations. There were no historical features in the Well field area, but modern features include the 16) CA boundary, 17) riverbank area, and 18) miscellaneous well field locations.

Error! Reference source not found.provides a summary of the Moab soil gas survey by feature. Appendix E provides the detailed data for the individual samples assigned to each feature. Note that at this level of granularity, the number of samples in each feature is relatively low (e.g., 1 to 10 – typically 2 to 5). Based on the relatively small number of samples in each feature, we based any interpretation of the feature level information on large (order of magnitude) differences. In general, the soil gas data for most features in each area corresponded to the overall area data. Notable and significant information that has the potential to influence environmental management decisions and GCAP development are provided in the following narratives. Specific notable features include the former north off-pile area ore storage area (locations MSG-28 through 32), various samples near the CA Boundary (locations MSG-07 through 13 and MSG-53 through 55), the former Mill Yard area containment pond area (locations MSG-23 through 26), and the tailings area purification pond area (MSG-16 and 17).

The North Off-Pile former "ore storage area" feature sampling locations, where surface remediation and revegetation activities have been completed, were found to have relatively low "background" Rn concentrations in the shallow soil gas. The soil gas at those locations has Rn is one to three orders of magnitude lower than the Rn in the tailings area. These data affirm that the criteria used for surface remediation result in reducing the original source uranium/thorium/radium materials to levels that are not detectable using soil gas methods.

The "CA Boundary" feature locations (as well as those locations outside of the CA) were generally found to have low levels of Rn (background to a few 1000 pCi/L). These soil gas results affirm the current CA delineation and provide an independent confirmation of the survey/screening/measurement processes that have been used for delineating the CA, as well as the overall radiation control policies and processes and management at the Moab site.

The sampling locations associated with the Mill Yard area containment pond and the Tailings area purification pond features yielded the soil gas with the most complex signature of analytes. Compared to all other locations, these former ponds had the highest radon, ammonia, nitrous oxide, methane, and carbon dioxide concentrations and were considered to be elevated to very high. The maximum radon concentrations, for example, were 143,136 pCi/L and 102,982 pCi/L for the containment pond and nearby purification pond, respectively. The soil gas analytes/indicators for NH_4^+/NH_3 were also very high in the soil gas samples, representing these features; particularly in the Mill Yard area containment pond that had nitrous oxide concentrations up to 3980 ppmv and ammonia concentrations as high as 24 ppmv.

The various contaminant related soil gas indicators suggest that the vadose zone beneath the Mill Yard area containment pond and the Tailings area purification pond features contains a significant quantity of residual source mass for both uranium/thorium/radium and NH_4^+. This significant residual source will be considered during the GCAP development. The general biogeochemical soil gas indicators for these features resulted in data that were also orders of magnitude higher than the baseline for the Moab site, indicating that a significant, sustained, and active subsurface microbial community has formed in the vadose zone and shallow groundwater in response to the historical process and wastewater discharges that occurred during mill operations. The presence of clear and consistent hot spots for Rn, N_2O, and NH_3 (e.g., near location MSG-23), suggest that the major residual source areas for uranium/thorium/radium and NH_4^+ are small and that supplemental future actions to address these sources can be small, surgical, and targeted.

5.4 Miscellaneous Feature Level Comments:

NH_3 was elevated in the soil gas at several sampling locations in the well field and near the riverbank, likely resulting from the flushing of NH_4^+ and subsequent transport from the upgradient sources. NH_3 and Rn in soil gas in the area where pilot scale hydroxyapatite injections were recently completed were low (near northeast plume area).

The sample locations in the tailings area were generally elevated for both Rn and NH_3, confirming that the remaining area of tailings is a bulk source for uranium and NH_4^+. Based on the low concentrations measured in the shallow soil gas data from the North Off-Pile former ore storage area, the planned removal and relocation of the tailings over the next several years should be effective in mitigating this bulk residual source.

Table 5-3. Soil Gas Data – Feature Level Summary Statistics.

Zone	Associated Feature		Radon		
	Historical	Modern	median	minimum	maximum
Millyard	Concentrator Building		117	n/a	n/a
	Containment Pond		42,122	2,165	143,136
	Grinding Bay		289	n/a	n/a
	Laboratory Building		172	n/a	n/a
	Ore Receiving		2,574	1,109	7,173
	Pond (Unknown)		2,078	n/a	n/a
		CA Boundary	420	48	2,078
		misc. mill yard	357	143	571
North Off-pile	Ore Storage		89	49	147
		River	93	29	751
		misc. North Off-pile	12	n/a	n/a
Tailings	Pond (BaCl2)		2,097	470	8,417
	Pond (Purification)		54,629	6,275	102,982
	Sump Pond		2,334	1,834	2,834
		misc. tailings	3,745	2,710	10,478
Well Field		CA Boundary	100	96	182
		River	212	145	413
		misc. wellfield	180	174	185

Table 5-3. Soil Gas Data – Feature Level Summary Statistics (continued).

Zone	Associated Feature		Nitrous Oxide		
	Historical	Modern	median	minimum	maximum
Millyard	Concentrator Building		0.0	n/a	n/a
	Containment Pond		76.8	0.3	3,980.0
	Grinding Bay		0.0	n/a	n/a
	Laboratory Building		0.0	n/a	n/a
	Ore Receiving		0.0	0.0	2.5
	Pond (Unknown)		21.6	n/a	n/a
		CA Boundary	0.0	0.0	21.6
		misc. mill yard	0.0	0.0	0.0
North Off-pile	Ore Storage		1.2	0.0	2.5
		River	2.5	1.4	4.4
		misc. North Off-pile	0.0	n/a	n/a
Tailings	Pond (BaCl2)		4.7	1.5	6.5
	Pond (Purification)		9.2	0.0	18.3
	Sump Pond		0.0	0.0	0.0
		misc. tailings	20.8	6.5	26.8
Well Field		CA Boundary	1.3	1.2	48.4
		River	1.6	0.7	4.2
		misc. wellfield	0.0	0.0	0.0

Table 5-3. Soil Gas Data – Feature Level Summary Statistics (continued).

Zone	**Associated Feature**		**Ammonia**		
	Historical	**Modern**	**median**	**minimum**	**maximum**
Millyard	Concentrator Building		0.4	n/a	n/a
	Containment Pond		9.9	1.4	23.3
	Grinding Bay		1.1	n/a	n/a
	Laboratory Building		0.1	n/a	n/a
	Ore Receiving		1.1	0.3	2.1
	Pond (Unknown)		3.7	n/a	n/a
		CA Boundary	4.2	1.0	10.9
		misc. mill yard	0.0	0.0	0.0
North Off-pile	Ore Storage		0.4	0.0	1.6
		River	1.2	0.6	2.4
		misc. North Off-pile	0.0	n/a	n/a
Tailings	Pond (BaCl2)		1.3	1.2	1.5
	Pond (Purification)		3.0	0.9	5.2
	Sump Pond		0.9	0.2	1.7
		misc. tailings	1.2	0.6	5.5
Well Field		CA Boundary	0.9	0.0	3.1
		River	2.3	0.0	11.3
		misc. wellfield	0.7	0.6	0.7

Table 5-3. Soil Gas Data – Feature Level Summary Statistics (continued).

Zone	Associated Feature		Methane		
	Historical	Modern	median	minimum	maximum
Millyard	Concentrator Building		6	n/a	n/a
	Containment Pond		24	17	56
	Grinding Bay		10	n/a	n/a
	Laboratory Building		5	n/a	n/a
	Ore Receiving		9	5	11
	Pond (Unknown)		24	n/a	n/a
		CA Boundary	12	6	33
		misc. mill yard	2	1	3
North Off-pile	Ore Storage		4	1	9
		River	5	4	10
		misc. North Off-pile	4	n/a	n/a
Tailings	Pond (BaCl2)		5	3	7
	Pond (Purification)		3	2	4
	Sump Pond		4	3	4
		misc. tailings	6	5	7
Well Field		CA Boundary	4	3	5
		River	5	3	24
		misc. wellfield	2	1	4

Table 5-3. Soil Gas Data – Feature Level Summary Statistics (continued).

Zone	Associated Feature		Carbon Dioxide		
	Historical	Modern	median	minimum	maximum
Millyard	Concentrator Building		1,000	n/a	n/a
	Containment Pond		25,077	3,066	71,285
	Grinding Bay		1,492	n/a	n/a
	Laboratory Building		1,000	n/a	n/a
	Ore Receiving		2,049	1,037	2,389
	Pond (Unknown)		4,088	n/a	n/a
		CA Boundary	1,689	1,000	15,177
		misc. millyard	1,733	1,000	2,466
North Off-pile	Ore Storage		1,389	1,000	6,694
		River	3,909	1,773	7,691
		misc. North Off-pile	1,000	n/a	n/a
Tailings	Pond (BaCl2)		3,730	1,433	3,960
	Pond (Purification)		8,424	1,748	15,100
	Sump Pond		2,637	1,875	3,398
		misc. tailings	5,698	2,708	16,378
Well Field		CA Boundary	2,606	1,000	10,067
		River	5,749	1,265	8,636
		misc. wellfield	2,209	2,167	2,251

5.5 Context Discussion

Reconciling the shallow soil gas data with the existing mapped groundwater plumes is a final step in interpreting the shallow soil gas data. Figure 5-16 overlays the Rn soil gas results from this study with the mapped 2023 uranium plume. The most significant feature in this overlay is seen in the area where the highest soil gas was measured (i.e., the area with highest residual uranium/thorium/radium in the subsurface). Based on the soil gas data, the containment and purification pond areas have about an order of magnitude more residual source than the bulk tailings area and two to three orders of magnitude higher uranium/thorium/radium compared to the low/background areas; however, the groundwater beneath the former containment and purification ponds is significantly lower in uranium compared to the surrounding groundwater. The overlay map for Rn/uranium (Figure 5-16) can be usefully contrasted with the overlay map for NH_4^+/NH_3 (Figure 5-17). In the NH_3 overlay, the start of the high NH_4^+/NH_3 concentrations in groundwater generally corresponds to the location of the mapped high concentration hot spot pond sources.

The disconnection between the high residual uranium/thorium/radium source material in the subsurface and the uranium concentration in the underlying groundwater is notable, as it represents a clear difference in behavior when contrasted with NH_4^+. The soil gas and associated groundwater plume data provide relatively compelling evidence of biogeochemical controls on contaminant source mass flux in the subsurface, particularly in the vicinity of the most significant identified sources for both uranium and NH_4^+. Further evaluation of the controlling processes and mineral phases may support developing innovative, efficient, and effective geochemical actions to support the GCAP portfolio.

The initial step in developing the technical basis and details of any geochemical corrective measures is examining the available information. Several boreholes have been installed for site characterization and the resulting core samples have been analyzed to support understanding of the nature and location of residual sources and to assess subsurface geochemistry. As shown in Figure 5-18, characterization boreholes were installed in 2001 and 2023. The focus of the studies was to characterize the vadose zone and potential residual vadose zone sources. Therefore, the sample locations focused toward the areas with the highest mapped concentrations for uranium in the underlying groundwater. Geochemical characterization boreholes have not been installed in/near the containment and purification ponds (Figure 5-18) and there is little information to directly assess the geochemical controls that are limiting the release of residual uranium to the groundwater (i.e., limiting the source mass flux). While future work to characterize the geochemistry in these areas may be needed, the soil gas results and existing site data are sufficient to develop a relatively compelling conceptual model of the likely geochemical controls in the area.

Several soil gas survey locations were collected that correspond to the 2023 secondary source investigation borehole locations at the Moab site (performed by Geosyntec and North Wind in support of GCAP development). Soil gas locations near/comparable to the secondary source sampling locations include: MSG-001, 003, 019, 033, 035, and 036. During this survey, uranium in the groundwater below the tailings pile (secondary source location 3) was found to be elevated, which supports the elevated concentrations in soil gas measured in that area. Similarly, groundwater concentrations in the northeast plume area (secondary source location 6) were low where soil gas concentrations were low. Secondary source study boreholes were not drilled near the containment and purification ponds, which were found to be the most significant residual point sources for both uranium and ammonium based on the soil gas results.

Geochemical controls are the most probable basis for the observed disconnect in the behaviors of uranium and NH_4^+. These controls include formation of solid phases that contain and sequester uranium (and/or NH_4^+/NH_3) – such phases can be formed by precipitation or uranium-ammonium-phosphate minerals or by uranium redox reactions (e.g., U(VI) to U(IV) and associated mineral precipitation. Based on the soil gas data each of these geochemical controls could be contributing to the plume behavior at Moab.

5.5.1 Uranium-Ammonium-Phosphate Geochemical Controls

The seed concepts for an updated and actionable conceptual model based on phosphate mineral controls is discernable in the review comments on the secondary source study provided to Geosyntec by Miles Denham (Denham, 2024), a subject matter expert provided by the DOE EM Technology Development Office (DOE EM-TDO) through the DOE EM Interagency Agreement Working Group.

> {excerpt of recommendations – emphasis added by underline} "…{recommended that the Moab team} … Choose the thermodynamic database carefully for the mineral saturation and uranium speciation calculations. The dissociation constants for aqueous uranyl phosphate complexes in the GWB default database (thermo.dat) are inconsistent with other databases, several orders lower than other databases including the NEA database. The low values preclude any uranium(VI) phosphate minerals from approaching saturation. Also, most databases do not contain the mineral uramphite $[(NH_4)_2(UO_2)(PO_4)_2 \cdot 3H_2O]$, a possible phase present in portions of the plume with high ammonium concentrations. It wouldn't significantly affect mobility of ammonium, but its presence may increase flushing times of uranium by allowing a greater mass of uranium to accumulate in soils than by adsorption alone. The first figure in the attachment shows the stability field of uramphite made using the thermo_phreeqc.dat database with uramphite added from the thermo_NEA.dat database. This was made at pH=7 and an activity of PO_4^{-3} = 1E-10, not unreasonable considering the dominant phosphate species at pH = 7 is $H_2PO_4^-$."

Denham provided several other significant comments in his communiqué, but this particular item is prescient when interpreting the observed differences in behavior in uranium and NH_4^+. In fact, Denham specifically noted that the modified geochemical conceptualization would not significantly affect the predicted mobility of NH_4^+ but would predict increased flushing time of uranium (i.e., decreases in source mass flux and slower uranium releases to groundwater). Like other phosphate minerals, the solubility of uramphite is low (log Ksp = -26.5; Gorman-Lewis et al. 2008; Foster and Lee, 2020; Markovic et al. 1988; Stohl and Smith, 1981) and this mineral has been observed to form in lab studies and in the field. Based on the documented Ksp of uramphite and measured concentrations of uranium and NH_4^+ in Moab groundwater, it is likely that uranium geochemical controls, such as formation of solid phase uramphite (or related uranyl phosphate, vanadate, arsenate or silicate minerals – see supplemental information box), are significant at Moab, particularly for the area near the former containment and purification ponds, and possibly significant throughout the entire plume.

Importantly, the scientific literature indicates that an array of phosphate/vanadate/arsenate/silicate minerals may serve as important geochemical controls, regulating uranium solubility and mobility at many sites (Foster et al., 2020; Wang et al, 2022; Martinez et al, 2014). This type of phosphate mineral control is the basis for the various hydroxyapatite-based in situ remediation strategies such as the one recently field tested at Moab. In these remediation strategies, the natural phosphate controls are enhanced by amending the system with reagents that further decrease uranium solubility and/or mobility and to increase robustness and sustainability of the system. Traditionally, the strategy is for the amendment to sequester uranium by precipitating hydroxyapatite family minerals, as well as to increase sorption, providing for some direct precipitation of uranium phosphate minerals and co-precipitation into the various mineral phases that are formed. Much of the literature has not considered the potential presence of NH_4^+. However, at sites where NH_4^+ is present, additional sequestration is possibly associated with direct precipitation of uramphite (uranium-ammonium-phosphate) and related phases, as well as by increased sorption on other phosphate minerals (e.g., hydroxyapatite) and NH_4^+ phases (e.g., struvite - a magnesium-ammonium-phosphate). In fact, previous investigations at the DOE Pacific Northwest National Laboratory identified significant unintentional formation of uramphite in experiments when an NH_4^+ buffer was used to control pH in the laboratory (Wellman et al., 2006). Formation of uramphite was specifically identified in a field study of in situ hydroxyapatite-based remediation (Wang et al., 2022). In addition, several studies highlight the role of microorganisms in facilitating the rapid formation of uramphite, including bacteria (Pan et al., 2015; Zhu et al., 2023; Zhang et al., 2019), fungi (Liang et al., 2015) and yeasts (Liang et al., 2016). In the hot spot areas underlying the former containment pond and purification pond, the soil gas survey also identified that a significant and active microbial community is present in the subsurface. These data provide a consistent and multi-pronged technical basis that supports the potential importance of geochemical controls in general, and the role of uramphite in particular.

5.5.2 Redox Geochemical Controls for Uranium

Redox-based geochemical controls for uranium have been widely studied; for example, in the DOE Office of Science Natural and Accelerated Bioremediation Research (NABIR) Program. These controls rely on the general tendency of reduced uranium (e.g., U(IV)) species to have a lower solubility and mobility compared to oxidized (e.g., U(VI)) species and complexes. Uraninite is an example of a low solubility U(IV) solid mineral phase that can form under mildly to strongly reducing conditions. The methane and carbon dioxide signal in the containment and purification pond areas indicates an active subsurface biological community and the potential for localized zones where U(IV) solid phases may form. One of the key findings of the NABIR program was that redox based stabilization of uranium is reversible. Based on data from the boreholes in the 2023 secondary source zone characterization (Keaton Belli, preliminary briefing/communication), the conditions supporting and potential for formation of U(IV) minerals, such as uraninite, may be present in areas of the vadose zone and groundwater beneath the Moab site.

5.5.3 Summary of Geochemical Context

Based on soil gas data, groundwater data, and preliminary geochemical conceptualization, removal and relocation of the hot spot sources to the extent practicable would be prudent, coincident with the removal of the bulk tailings. For any residual high strength source that cannot be removed or relocated, the GCAP may be structured to include enhanced geochemical controls using a site-specific recipe of amendments (either injected liquid or blended solids) that would further decrease solubility and mobility of uranium and possibly decrease mass flux of NH_4^+. The site-specific recipe might consider addition of phosphate and or magnesium and removal of NH_4^+ as potential modifications to the baseline recipe that was originally developed for the Old Rifle Site and pilot tested at Moab. Development of a site-specific recipe, if needed, should be based on geochemical principles, previous and supplemental field characterization, and laboratory tests.

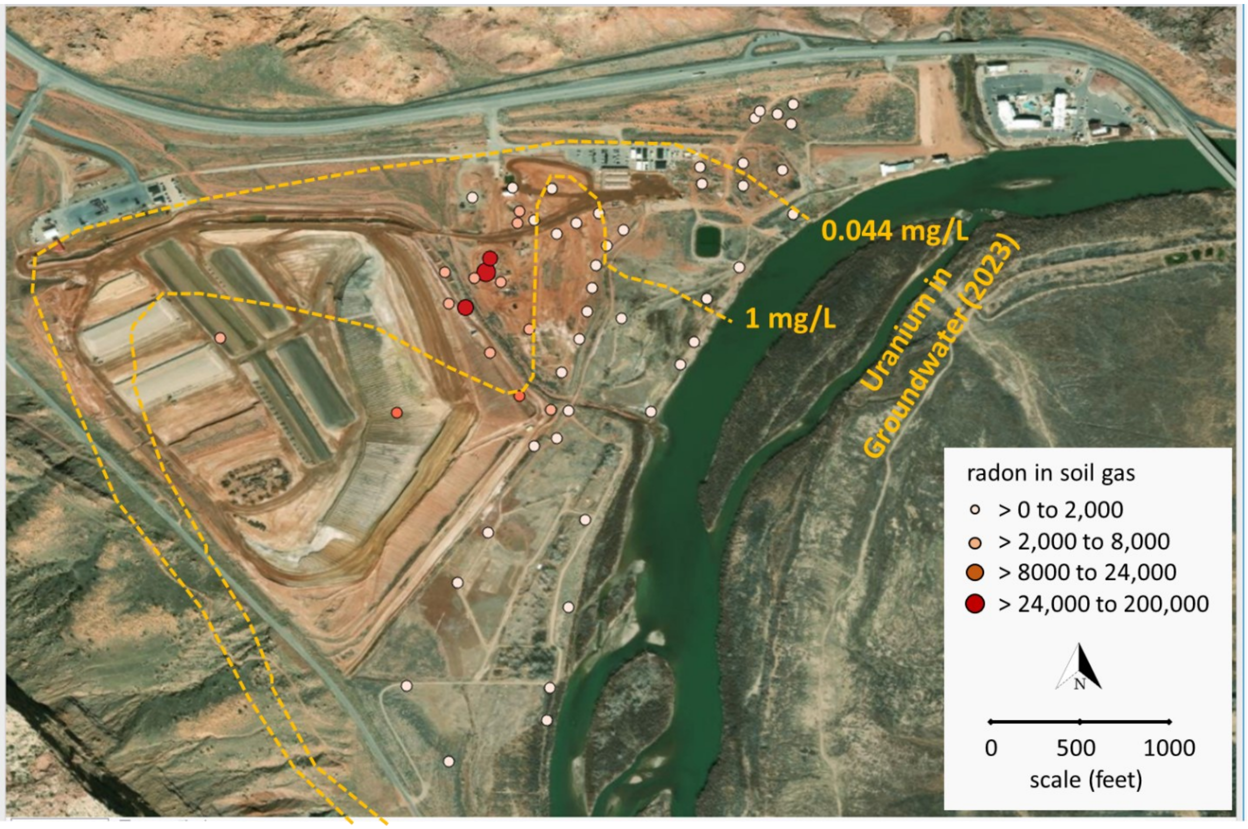

Figure 5-16. Radon Overlay Map of Groundwater Beneath Site Features

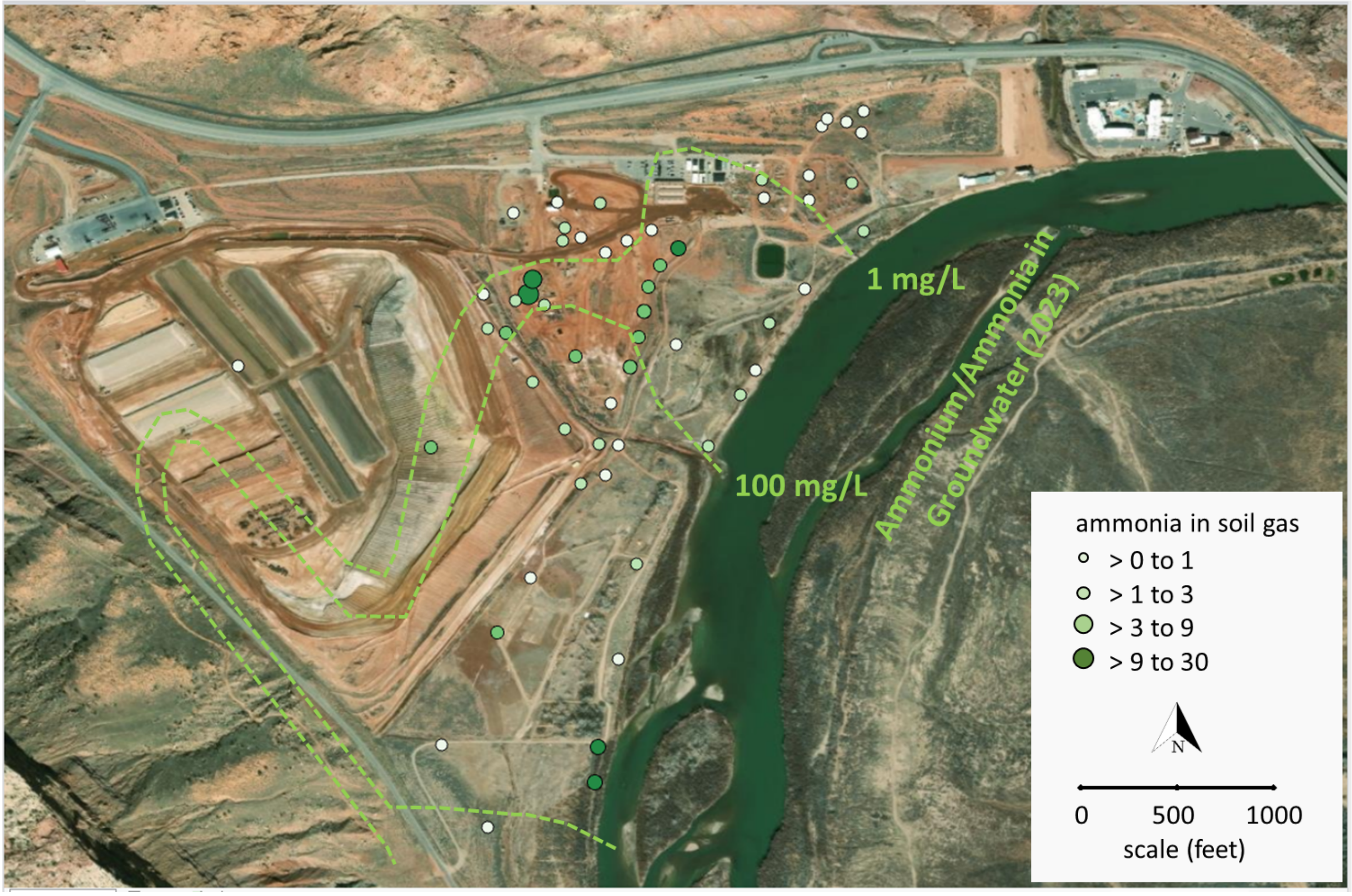

Figure 5-17. Ammonium/Ammonia Overlay Map of Groundwater Beneath Site Features

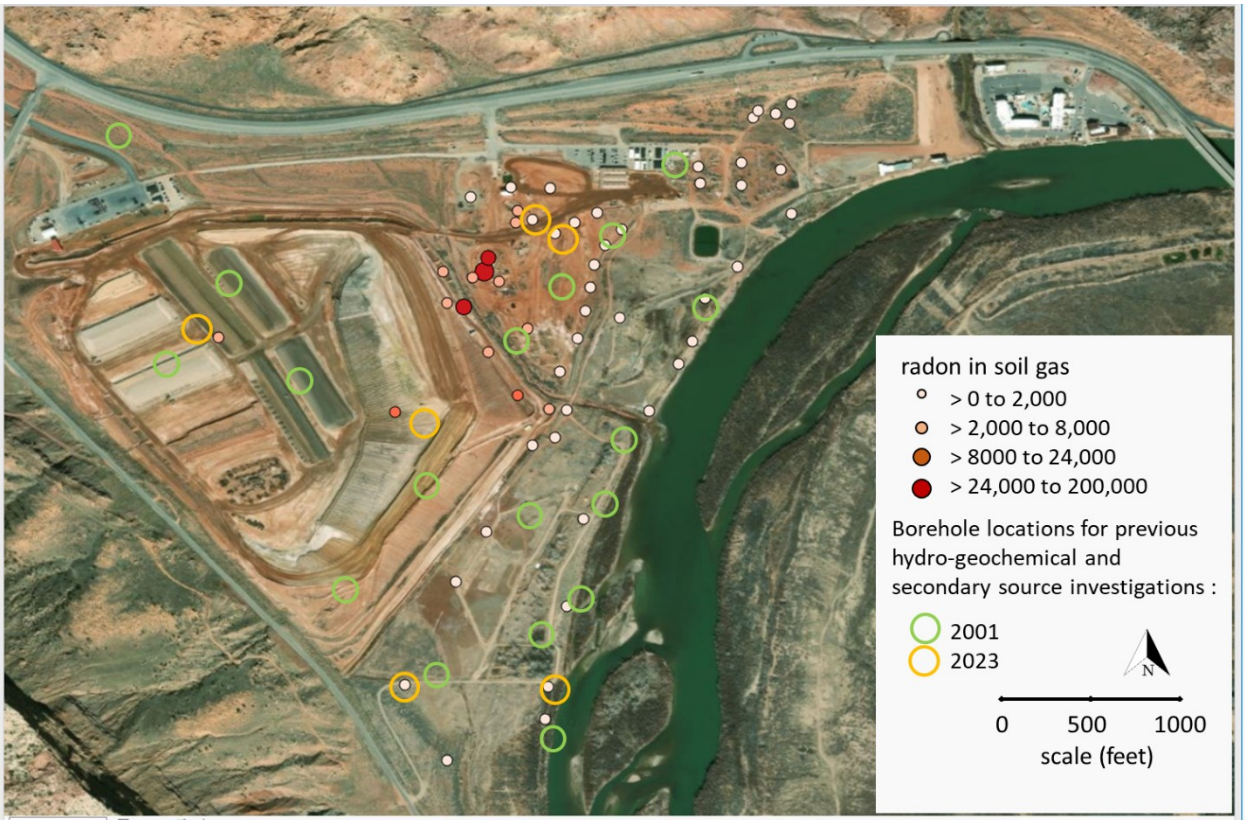

Figure 5-18. Historical Borehole Location Overlay of Radon Soil Gas Survey Map

Supplemental Information
Uranium mineralogy with a focus on Moab conditions

Based on Moab soil gas and groundwater data, potentially significant biogeochemical controls associated with formation of various minerals, such as uramphite, could be considered in updating the site conceptual and numerical models and in developing and optimizing the GCAP. While uramphite was noted as a specific exemplar of a potential solid mineral that may serve as a residual/secondary source phase, several other minerals should also be considered in assessing, understanding, and accounting for uranium biogeochemistry in sequestering uranium and reducing uranium and ammonium mobility in the subsurface. Additional minerals that may be significant include other low-solubility uranium minerals, as well as minerals that do not contain uranium, but which enhance sorption or provide for coprecipitation. A summary of candidate minerals is provided below.

Uranium containing minerals: Uramphite is representative of the general classes of minerals that are known as hydrated phosphates/vanadates/arsenates and silicates. These minerals generally contain four components – 1) One or more companion element(s) or cations, 2) uranium ("uranyl"), 3) an anion (phosphate, vanadate, arsenate, or silicate), and 4) waters of hydration. The mineral phases/names and the characteristics of the minerals are related to the specific combination of components. Uramphites is an ammonium uranyl phosphate phase. Closely related simple uranyl phosphate minerals that have different companion elements, include: autunite (Na), Ankoleite (K), bassetite (Ca), saleeite (Mg), and several others. Notably, uramphite has one of the lowest solubilities of the simple uranyl phosphate minerals, increasing the likelihood of its presence at any site with elevated ammonium. Similar lists of mineral phases are available for vanadates (e.g., carnotite and fritzscheite), arsenates (e.g., kahlerite) and silicates (e.g., uranophane, soddyite and weeksite). For many of these minerals, the dehydrated forms (without the waters of hydration) are designated using the prefix "meta-".

In real world systems, uranium containing minerals may represent a) one of these minerals, or a blend of these phases, b) may occur with related non uranium phosphates, vanadates, arsenates, and/or silicates, and/or c) may occur with varying degrees of hydration in the vadose zone.

Non-uranium minerals: These may significantly influence uranium attenuation, particularly related phosphate minerals that have a low solubility. Notably, the presence of calcium phosphate minerals (e.g., hydroxyapatite and apatite) increase attenuation of uranium. Similarly, at Moab, the formation of magnesium ammonium phosphate (struvite) and related minerals has the potential to contribute to both uranium and ammonium attenuation.

In advancing the conceptual and numerical modeling at Moab, consideration of key uranium and non-uranium minerals may provide an improved technical basis for DOE as the GCAP is structured and developed.

6.0 Conclusions

The Moab shallow soil gas survey was effective in identifying residual sources of uranium and NH_4^+ in the subsurface (vadose zone and shallow groundwater). The survey identified general areal differences in residual subsurface sources and specifically identified small hot spot source zones associated with the containment pond in the Mill Yard area and the purification pond in the tailings area. Key conclusions are listed below:

- The north off-pile former ore storage area, where removal actions and revegetation efforts have been completed, were found to have relatively low "background" Rn levels in the shallow soil gas. These data affirm that the criteria used for the remediation resulted in effective removal of the original source uranium/thorium/radium materials.
- The "CA Boundary" samples were generally found to have low levels of Rn – background to a few thousand pCi/L. These soil gas results affirm the current CA delineation and provide an independent confirmation of the survey/screening/measurement processes that have been used for delineating the CA. Significant residual source contamination hot spots were identified for both uranium and NH_4^+ that are associated with containment pond in the Mill Yard area and the purification pond in the tailings area. The maximum Rn levels, for example, were 143,136 pCi/L and 102,982 pCi/L for the containment pond and nearby purification pond, respectively. The general biogeochemical soil gas indicators for these features indicated that a significant, sustained, and active subsurface microbial community is present in the hot spot area.
- NH_3 was elevated in the soil gas at several sampling locations in the well field and near the riverbank, likely resulting from the flushing of NH_4^+ and subsequent transport from the upgradient sources as well as with interim action injection operations.
- NH_3 and Rn in soil gas in the area where pilot scale hydroxyapatite injections were recently completed were low.
- The sample locations in the Tailings area were generally elevated for both Rn and NH_3, confirming that the remaining area of tailings area is a bulk source for uranium and NH_4^+. Based on the low levels measured in the shallow soil gas data from the North Off-Pile former ore storage area, the planned removal and relocation of the tailings over the next several years should be effective in mitigating this bulk residual source.
- While future work to characterize the geochemistry in the hot spot areas may be needed, the soil gas results and the existing data are sufficient to develop a compelling conceptual model of the likely geochemical controls in the area. Uranium geochemistry and mineral phases (such as solid phase uramphite or uraninite) limit the solubility and release of uranium to the groundwater and are significant at Moab – particularly for the area near the former containment and purification ponds, and possibly significant throughout the entire plume.
- Removal and relocation of the hot spot sources to the extent practicable would be prudent, coincident with the removal of the bulk tailings. For any residual high strength source that cannot be removed or relocated, the GCAP could be structured to include enhanced geochemical controls using a site-specific recipe of amendments.

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Appendix A. Detailed Summary of Radon Methods

Figure A-1 depicts the decay chain for the most abundant naturally occurring isotopes of uranium and thorium, uranium 238 (^{238}U) and thorium 232 (^{232}Th). For each isotope and decay product, the half-life, type of decay, and progeny (if radioactive) is noted along with other key information. Rn in each decay chain is highlighted (*) in Figure A-1.

Various methods have been used through the years to measure Rn in soil gas samples and in associated Rn surface-flux chambers. These methods include alpha scintillation cells (Nielson et al., 1981; Karp, 1988; Tuccimei, et al., 2006), alpha solid-state scintillators, track detectors, and gamma spectroscopy applied to passive or active trap samples (Fuhrman et al., 2021, Tuccimei, et al., 2006; Durridge 2023 a-c). In most cases, the target Rn isotope is the ^{238}U progeny Rn 222 (^{222}Rn) with a half-life of 3.82 days. The ^{232}Th progeny Rn 220 (^{220}Rn) has a significantly shorter half-life (55.6 seconds), which increases the difficulty and cost of representative sample collection and accurate measurement of this isotope. Further, for active sampling methods, the presence of ^{220}Rn and its progeny can contribute alpha decays and complicate the accurate analysis of ^{222}Rn. One strategy that is used to improve the quality of the analysis for ^{222}Rn is to collect samples in an interim container, such as a Tedlar® bag, allow time for the ^{220}Rn to "fully" decay, and then remove ^{220}Rn progeny from the gas sample using filtration and sorption prior to counting; see the annotation in Figure A-1 for "a. Tedlar® bag hold". Previous studies demonstrate that Tedlar® bag hold times ≥ 7 to 10 minutes are sufficient to support high quality ^{222}Rn analysis protocols (Nielson et al., 1981; Karp, 1988; Fuhrman et al., 2021,).

For this study, ^{222}Rn in collected soil gas was analyzed using a scintillation cell (Lucas Cell). A Lucas Cell (Figure A-2) is a sealed canister that has its inside coated with zinc sulfide doped with silver (ZnS(Ag)). This coating emits a light photon when struck by an alpha particle. The top of the cell is equipped with fittings for evacuation and filling of the canister while the bottom of the cell is a clear polymer window to allow photon detection by a photomultiplier tube. The count rate is measured and related to the Rn level in a soil gas (or air) sample. As shown in Figure A-1 (see annotation "b. Lucas Cell ingrowth hold") the ingrowth of ^{222}Rn progeny results in count rates that increase over time when the canister is first loaded. After loading, sufficient ingrowth time for equilibration of Rn and progeny results in a steady state "stabilization" of the count rate. This Lucas Cell ingrowth hold time has ranged widely in past analytical protocols from a few minutes to four hours. Previous work for the Department of Energy Office of Legacy Management (DOE-LM) used a Lucas cell hold time of 60 minutes when sampling soil gas associated tailings and tailings cover systems (Nielson et al., 1981; Karp, 1988). In practice, a relatively long Lucas Cell hold provides the most stable steady-state count rate, and a relatively short hold time minimizes long- term adverse impacts associated with of Rn progeny adhering to surfaces and building up inside the Lucas Cells. Thus, the optimal ingrowth hold time is the shortest time that provides a stable and reliable count with protocols specifying that the Lucas Cell be purged and cleared immediately after counting.

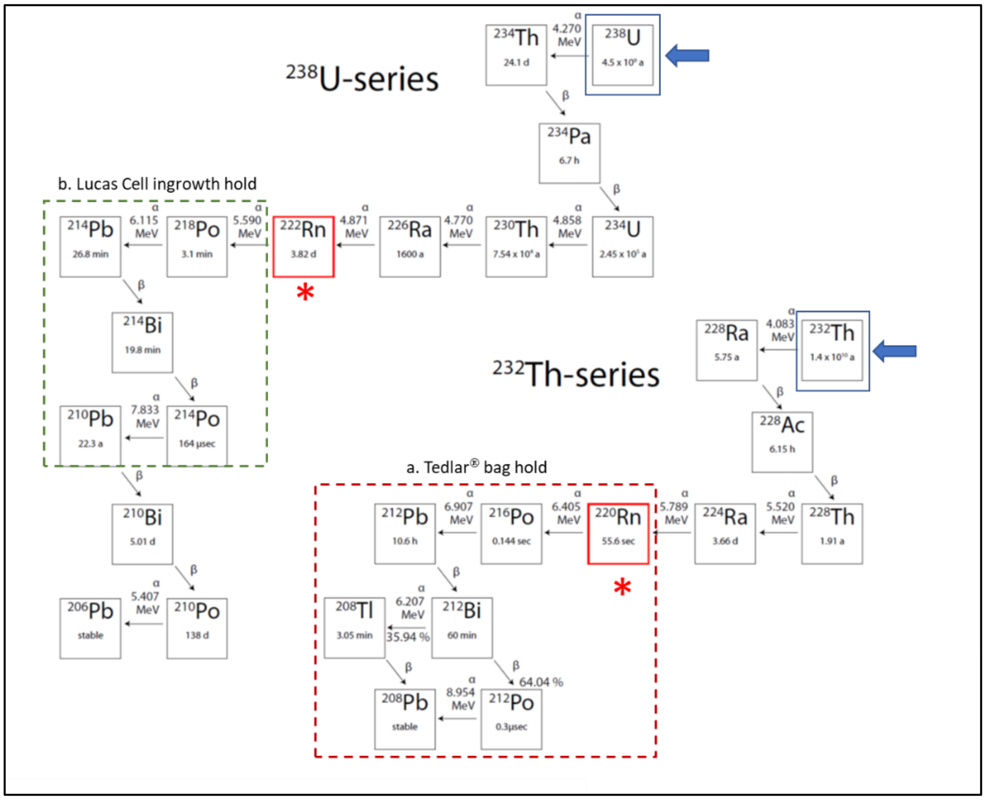


Figure A-1. Annotated Decay Chain for 238U and 232Th (←)Highlighting Radon (*) and Protocol Development Topics (a/b)

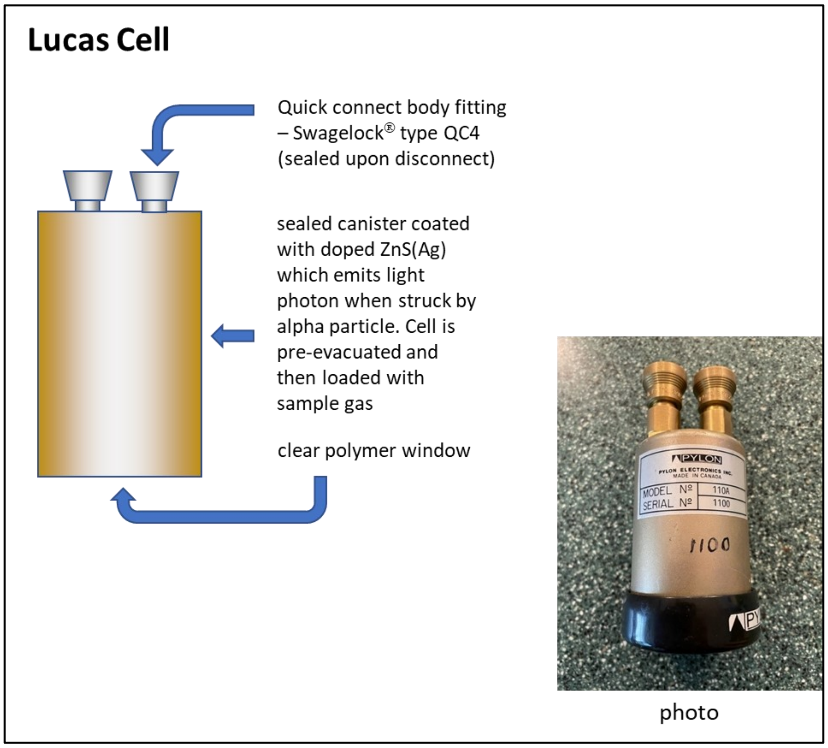
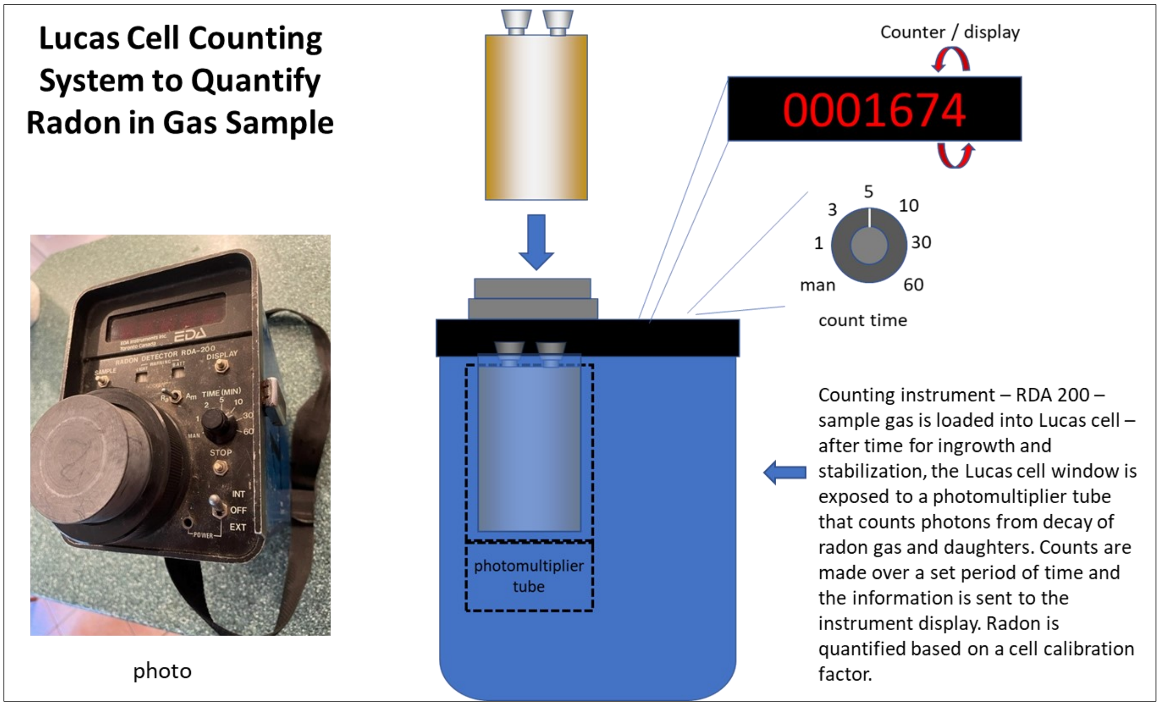


Figure A-2. Lucas Cell Schematic and Example Photograph

Looney (2023) provides detailed documentation for the SRNL soil gas Rn analysis protocol used in this study. The overall objective of the protocol is reliable, accurate, and reproducible analysis of Rn levels in soil gas samples collected in Tedlar® bags. The protocol development addressed several key topics: a) Tedlar® bag hold time, b) Lucas cell loading, ingrowth hold times & appropriate correction factors for short hold times, and c) instrument performance and calibration factors. Note that the instruments used for this work were the RDA-200 (Figure A-3) and the Lucas cells used were Model 110-A or equivalent (Figure A-2) – EDA Instruments / Pylon Electronics, Toronto Ontario Canada).



Counting instrument – RDA 200 – sample gas is loaded into Lucas cell – after time for ingrowth and stabilization, the Lucas cell window is exposed to a photomultiplier tube that counts photons from decay of radon gas and daughters. Counts are made over a set period of time and the information is sent to the instrument display. Radon is quantified based on a cell calibration factor.

Figure A-3. RDA-200 Radon Monitor

Summary of Analysis Sequence

Each Lucas Cell was pre-evacuated to a preset vacuum level (15 inHg vacuum) and counted for 5 minutes to obtain a background count for the cell. This count was recorded.

Each sample was then collected into a Tedlar® bag and the collection time recorded. A Tedlar® bag hold time of at least 10 minutes was used to allow decay of ^{220}Rn.

The Lucas cell evacuation process was repeated, and vacuum level checked just prior to cell loading to assure accuracy. The preset vacuum level is used to determine a loading factor (equal to 1 / fraction of soil gas in cell). For example, if the atmospheric pressure is 29 inHg and the preset vacuum level is 15 inHg, the loading factor is 1.93 (29/15=1.93) which indicates that about half of the gas in the loaded cell is soil gas and half is uncontaminated air remaining in the cell after cell evacuation. In the extreme case where cells are pre-evacuated to a perfect vacuum the loading factor would be 1 (all of the gas in the cell is soil gas) and if the pre-evacuated cell is at atmospheric pressure, then the loading factor would be 0 (no soil gas would be drawn in the cell). In the field, the local atmospheric pressure is used to calculate the loading factor for each sample loading.

After the Tedlar® bag hold, the sample bag was connected to the pre-evacuated Lucas cell using a Lucas cell loading assembly (Figure A-4). The loading assembly includes a filter and polyester fiber trap to remove ^{220}Rn progeny. The loading time was recorded in the spreadsheet to allow for ingrowth corrections. Ingrowth corrections were performed as documented in Looney (2023).

Following counting, the activity of parent ^{222}Rn in the sample pulled from the Tedlar® bag was estimated using the documented calibration factors for the Pylon and EDA Lucas Cells. This value was corrected for decay to the original sample collection time to provide a Rn activity in the original soil gas pulled from the subsurface.

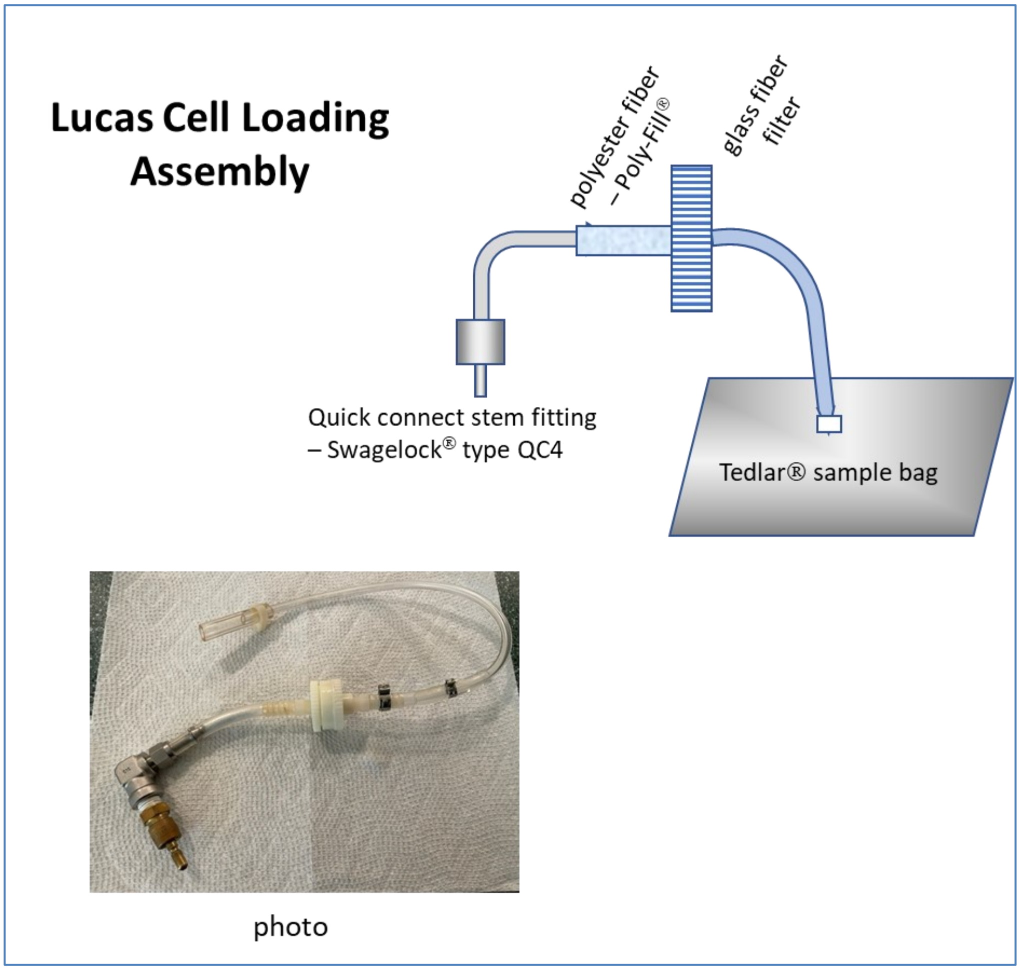


Figure A-4. Lucas Cell Loading Assembly Schematic and Example Photograph

Summary

A straightforward protocol was documented to support ^{222}Rn analysis using Lucas Cells and field deployable alpha counting Rn monitors. The method consists of several steps; including sample collection into Tedlar® bags, a Tedlar® bag hold time to allow decay of short-lived ^{220}Rn, pre-evacuation of a Lucas Cell and loading from the Tedlar® bag through a filter and polyester fiber cartridge, a Lucas Cell hold time to allow controlled ingrowth and equilibration of progeny, determine sample count rate, and converting the count rate to a decay corrected Rn level using predetermined Lucas Cell calibration factors and supplementary information such as sampling time, counting time, background counts, Lucas Cell hold time, and barometric/loading pressures.

The Rn analysis protocol was calibrated and verified based on a Rn monitor with a traceable calibration certified by the NRPP (Airthings Corentium Pro SN 2700014917). At low Rn levels (3 pCi/L), a small dataset resulted in a relative standard deviation (RSD) of 17%. At higher Rn levels (5000 to 7000 pCi/L), the Pylon and EDA Lucas Cells provided equivalent data and provided replicate data that was tightly clustered with an RSD < 1% to 1.4%. Based on the results from all the measurements at low Rn levels (3 pCi/L) and higher Rn levels (5000 to 7000 pCi/L), Looney (2023) estimated that a nominal conservative RSD for the overall method is approximately +/- 20% at low Rn levels (below 100 pCi/L) and +/- 10% at higher Rn levels that are typical of soil gas (above 100 pCi/L).

Appendix B. Detailed Summary of Photoacoustic Spectrometer Methods

Summary

NH_3 is a primary source material that has a high vapor pressure. Thus, NH_3 is a potential soil gas analyte that provides a direct measure of the contamination. However, the ammonium/ NH_3 relationship and associated partitioning between the aqueous and gas phases is strongly influenced by pH. The pKa of ammonium is 9.25 and the aqueous ammonium ion is the predominant form of ammonium/ NH_3 at pH below 9. As a result, the presence of NH_3 in the gas phase is influenced by both the presence of residual source material and the chemistry of the soil pore water, with typical pH values limiting (depressing) the amount of NH_3 in the soil gas. In this scenario, if significant levels of NH_3 are measured in soil gas, the result would be a compelling qualitative indicator of nearby (or underlying) ammonium/ NH_3 in the vadose zone or shallow groundwater.

An INNOVA 1312 Photoacoustic Multi-gas Monitor was used for measuring gas-phase surrogate indicators of ammonium, including NH_3 and N_2O. The photoacoustic spectrometer (PAS) was also calibrated to measure general indicator soil gases, including CH_4, CO_2, and water vapor. Soil gas samples are introduced into the instrument for measurement from Tedlar® bags. Specialized optical filters allow for selective detection of different gases within a mixed sample.

N_2O, nitric oxide (NO), and nitrogen (N_2) are gaseous products of various redox pathways. Thus, the measurement of significant N_2O and NO provides confirmatory evidence of both: a) the presence of residual ammonium/ NH_3 source material, and b) ongoing-active microbial ammonium transformation in the subsurface and measurable attenuation of ammonium. Note that NO is typically present in low concentration and N_2 is a major component of the atmosphere and baseline soil gas; thus, these two nitrogen species were not selected as target analytes.

Once inside the PAS, the gas is exposed to pulsed light, some of which is absorbed by the gas. The amount of light energy absorbed is proportional to its concentration. The absorbed light is then released as heat and passed through a ‘chopper’, which generates changes in pressure. These pressure (sound) waves can then be detected by an internal microphone. If the gas contains many chemicals, optical filters are used to resolve and allow for selective detection of analytes of interest.

MESA reference gas standards were used for calibration of all filters. Calibrations also included corrections for humidity and cross interferences that were applied for measuring soil gas samples collected in the field. A MESA reference (calibration) gas mixture containing known concentrations of CH_4, CO_2, and N_2O was also used for validating the calibration.

Summary of Analysis Sequence

The PAS was started in continuous sampling mode at the beginning of each day and allowed to run for at least one hour before use. This ensured the chamber within the instrument had time to warm up and reach a steady temperature. Temperature variations can impact gas behavior and bias measurements. Prior to any sample measurements, the ‘background’ values of each constituent in lab air were recorded (after the initial warm-up period).

Soil gas samples were collected into 3 L Tedlar® bags and then brought back to the groundwater laboratory for analysis.

Each Tedlar® bag was attached to the inlet of the instrument using flex tubing via the fitting on the bag (Figure B-1 c). The fitting was then rotated one full turn to open the bag and analysis was initiated. The PAS collects approximately 0.25 L of air per measurement (for all analytes of interest). Five measurements were made and the fifth was recorded to ensure no carry over from the previous sample occurred. Very good reproducibility was seen between repeated measurements of the same sample.

a) Front of instrument

b) Back of instrument

c) Connection to Tedlar bag for sample analysis

Figure B-1. Innova 1312 Photoacoustic Spectrometer and Sample Inlet

Appendix C. Overall Data Summary Table.

Station	Latitude (N)	Longitude (W)	Easting	Radon (pCi/L)	Nitrous Oxide (ppmv)	Ammonia (ppmv)	Methane (ppmv)	Carbon Dioxide (ppmv)
MSG-001	38.5936933	109.5920717	-109.5920717	250.00	0.68	11.10	23.80	7230.65
MSG-002	38.5930367	109.5921350	-109.5921350	413.00	1.57	11.30	23.90	8635.90
MSG-003	38.5937417	109.5949733	-109.5949733	174.00	0.00	0.68	3.73	2250.96
MSG-004	38.5922117	109.5941167	-109.5941167	185.00	0.00	0.64	1.06	2166.64
MSG-005	38.6042383	109.5890283	-109.5890283	103.00	0.00	1.60	1.15	6694.10
MSG-006	38.6039017	109.5889817	-109.5889817	130.00	0.00	0.69	1.53	3500.35
MSG-007	38.6029783	109.5905833	-109.5905833	182.93	0.00	10.88	33.18	1000.00
MSG-008	38.6026483	109.5909033	-109.5909033	496.76	0.00	3.83	16.61	1000.00
MSG-009	38.6022550	109.5911233	-109.5911233	878.00	2.99	4.16	11.50	3066.00
MSG-010	38.6017883	109.5912033	-109.5912033	1506.00	5.07	7.01	14.20	6745.20
MSG-011	38.6013117	109.5913167	-109.5913167	48.02	0.00	3.59	12.13	1000.00
MSG-012	38.6007717	109.5914667	-109.5914667	420.00	16.50	6.33	5.65	15176.70
MSG-013	38.6000967	109.5918233	-109.5918233	146.00	0.00	0.97	5.51	1688.86
MSG-014	38.5993300	109.5920417	-109.5920417	2834.00	0.00	1.71	4.38	3398.15
MSG-015	38.5996033	109.5926833	-109.5926833	8417.00	1.52	1.32	3.11	3730.30
MSG-016	38.6013900	109.5937683	-109.5937683	102982.00	18.30	5.17	3.83	15100.05
MSG-017	38.6021117	109.5942000	-109.5942000	6275.00	0.00	0.87	2.24	1747.62
MSG-018	38.6031667	109.5923833	-109.5923833	571.30	0.00	0.00	3.44	1000.00
MSG-19	38.6028983	109.5919333	-109.5919333	143.00	0.00	0.01	0.77	2465.58
MSG-20	38.5993067	109.5916950	-109.5916950	1834.00	0.00	0.16	3.02	1875.37
MSG-21	38.5986017	109.5923817	-109.5923817	470.00	4.65	1.49	5.25	3960.25
MSG-22	38.6004883	109.5932867	-109.5932867	2097.00	6.50	1.17	6.55	1433.36
MSG-23	38.6021133	109.5933700	-109.5933700	143136.00	3980.00	23.30	55.90	71284.50
MSG-24	38.6023967	109.5932833	-109.5932833	77886.00	145.00	17.40	30.50	44457.00
MSG-25	38.6019167	109.5930683	-109.5930683	2165.00	0.26	1.41	16.50	3066.00
MSG-26	38.6020017	109.5936067	-109.5936067	6358.00	8.52	2.43	16.50	5697.65
MSG-27	38.6009717	109.5924883	-109.5924883	2078.00	21.60	3.65	24.00	4088.00
MSG-28	38.6031117	109.5927300	-109.5927300	3301.00	2.54	2.05	10.50	2348.05
MSG-29	38.6036367	109.5936467	-109.5936467	1109.00	0.00	1.02	10.20	1750.18
MSG-30	38.6038167	109.5920283	-109.5920283	289.00	0.00	1.06	9.79	1492.12
MSG-31	38.6033433	109.5926883	-109.5926883	7173.00	0.00	1.22	8.18	2388.93
MSG-32	38.6038250	109.5928183	-109.5928183	1847.00	0.00	0.33	5.08	1037.33
MSG-33	38.6031050	109.5915233	-109.5915233	117.11	0.00	0.40	5.89	1000.00

Station	**Latitude (N)**	**Longitude (W)**	**Easting**	**Radon (pCi/L)**	**Nitrous Oxide (ppmv)**	**Ammonia (ppmv)**	**Methane (ppmv)**	**Carbon Dioxide (ppmv)**
MSG-34	38.6033100	109.5910783	-109.5910783	171.76	0.00	0.07	4.93	1000.00
MSG-35	38.5992733	109.5951700	-109.5951700	10478.00	26.80	5.49	6.58	16377.55
MSG-36	38.6007767	109.5987533	-109.5987533	3745.00	20.80	0.59	5.70	2708.30
MSG-37	38.6014767	109.5941117	-109.5941117	2710.00	6.49	1.23	4.63	5697.65
MSG-38	38.6011867	109.5906217	-109.5906217	12.07	0.00	0.00	4.38	1000.00
MSG-39	38.6053167	109.5874483	-109.5874483	142.00	0.00	0.03	0.78	1055.22
MSG-40	38.6052390	109.5879100	-109.5879100	52.89	0.00	0.30	2.97	1000.00
MSG-41	38.6053750	109.5878167	-109.5878167	70.00	1.52	0.20	4.34	1259.62
MSG-42	38.6055117	109.5871400	-109.5871400	99.00	0.90	0.03	3.54	1198.30
MSG-43	38.6051267	109.5871683	-109.5871683	65.00	1.86	0.40	6.19	1768.06
MSG-44	38.6041867	109.5873483	-109.5873483	147.00	2.46	1.14	3.86	4062.45
MSG-45	38.6043150	109.5881533	-109.5881533	79.00	1.46	0.62	7.02	1517.67
MSG-46	38.6038633	109.5881550	-109.5881550	48.53	1.46	0.00	8.55	1000.00
MSG-47	38.6032867	109.5871267	-109.5871267	751.00	4.38	2.40	10.30	7690.55
MSG-47 dup	38.6032867	109.5871267	-109.5871267	613.00	---	---	---	---
MSG-48	38.6015817	109.5888850	-109.5888850	73.00	2.27	1.31	4.24	5058.90
MSG-49	38.6022133	109.5882267	-109.5882267	112.00	1.76	0.61	5.53	1773.17
MSG-50	38.6007083	109.5891467	-109.5891467	58.00	1.38	0.60	4.13	1811.50
MSG-51	38.6002517	109.5894300	-109.5894300	29.00	2.68	1.16	9.32	2759.40
MSG-52	38.5992983	109.5900083	-109.5900083	118.00	3.37	1.46	3.75	6387.50
MSG-53	38.5987467	109.5919283	-109.5919283	100.00	1.31	0.87	3.76	2606.10
MSG-54	38.5968500	109.5933183	-109.5933183	95.79	1.20	0.00	5.05	1000.00
MSG-55	38.5958283	109.5939450	-109.5939450	182.00	48.40	3.05	3.48	10066.70
MSG-56	38.5953233	109.5916867	-109.5916867	145.00	1.86	0.00	5.42	1264.73
MSG-57	38.5958333	109.5915175	-109.5915175	212.00	1.47	1.56	2.69	4675.65
MSG-58	38.5971083	109.5913483	-109.5913483	157.00	4.15	2.25	4.14	5748.75

Appendix D. Area Level Information -- data for the individual samples assigned to each area.

Station	**Category**	**Radon (pCi/L)**	**Nitrous Oxide (ppmv)**	**Ammonia (ppmv)**	**Methane (ppmv)**	**Carbon Dioxide (ppmv)**
MSG-033	Millyard	117	0.0	0.4	6	1000
MSG-023		143136	3980.0	23.3	56	71285
MSG-024		77886	145.0	17.4	44	44457
MSG-025		2165	0.3	1.4	17	4366
MSG-026		6358	8.5	2.4	17	5698
MSG-043		289	0.0	1.1	10	1492
MSG-034		172	0.0	0.1	5	1000
MSG-028		3431	2.5	2.1	11	2348
MSG-029		1109	0.0	1.0	10	1750
MSG-031		7173	0.0	1.2	8	2389
MSG-032		1847	0.0	0.3	5	1037
MSG-027		2078	21.6	3.7	24	4088
MSG-007		183	0.0	10.9	33	1000
MSG-008		497	0.0	3.8	17	1000
MSG-009		878	3.0	4.2	12	4366
MSG-010		1506	5.1	7.0	14	6745
MSG-011		48	0.0	3.6	12	1000
MSG-012		420	16.5	6.3	6	15177
MSG-013		146	0.0	1.0	6	1689
MSG-018		571	0.0	0.0	3	1000
MSG-019		143	0.0	0.0	1	2466

Station	**Category**	**Radon (pCi/L)**	**Nitrous Oxide (ppmv)**	**Ammonia (ppmv)**	**Methane (ppmv)**	**Carbon Dioxide (ppmv)**
MSG-005	North Off-pile	103	0.0	1.6	1	6694
MSG-006		143	0.0	0.7	2	3500
MSG-039		142	0.0	0.0	1	1055
MSG-040		53	0.0	0.3	3	1000
MSG-041		70	1.5	0.2	4	1260
MSG-042		99	0.9	0.0	4	1198
MSG-043		65	1.9	0.4	6	1768
MSG-044		147	2.5	1.1	4	4062
MSG-045		79	1.5	0.6	7	1518
MSG-046		49	1.5	0.0	9	1000
MSG-047		751	4.4	2.4	10	7691
MSG-048		73	2.3	1.3	4	5059
MSG-049		112	1.8	0.6	6	1773
MSG-050		58	1.4	0.6	4	1812
MSG-051		29	2.7	1.2	9	2759
MSG-052		118	3.4	1.5	4	6388
MSG-038		12	0.0	0.0	4	1000

Station	**Category**	**Radon (pCi/L)**	**Nitrous Oxide (ppmv)**	**Ammonia (ppmv)**	**Methane (ppmv)**	**Carbon Dioxide (ppmv)**
MSG-015	Tailings	8417	1.5	1.3	3	3743
MSG-021		470	4.7	1.5	5	3960
MSG-022		2097	6.5	1.2	7	1433
MSG-016		102982	18.3	5.2	4	15100
MSG-017		6275	0.0	0.9	2	1748
MSG-014		2834	0.0	1.7	4	3398
MSG-020		1834	0.0	0.2	3	1875
MSG-035		10478	26.8	5.5	7	16378
MSG-036		3745	20.8	0.6	6	2708
MSG-037		2710	6.5	1.2	5	5698

Station	**Category**	**Radon (pCi/L)**	**Nitrous Oxide (ppmv)**	**Ammonia (ppmv)**	**Methane (ppmv)**	**Carbon Dioxide (ppmv)**
MSG-053	Well Field	100	1.3	0.9	4	2606
MSG-054		96	1.2	0.0	5	1000
MSG-055		182	48.4	3.1	3	10067
MSG-001		250	0.7	11.1	24	7244
MSG-002		413	1.6	11.3	24	8636
MSG-056		145	1.9	0.0	5	1265
MSG-057		212	1.5	1.6	3	4676
MSG-058		157	4.2	2.3	4	5749
MSG-003		174	0.0	0.7	4	2251
MSG-004		185	0.0	0.6	1	2167

Appendix E. Feature Level Information - Data for the Individual Samples Assigned to each Feature.

Station	Category	Associated Feature		Radon (pCi/L)	Nitrous Oxide (ppmv)	Ammonia (ppmv)	Methane (ppmv)	Carbon Dioxide (ppmv)
		Historical	Modern					
MSG-033	Millyard	Concentrator Building		117	0.0	0.4	6	1000
MSG-023		Containment Pond		143136	3980.0	23.3	56	71285
MSG-024				77886	145.0	17.4	31	44457
MSG-025				2165	0.3	1.4	17	3066
MSG-026				6358	8.5	2.4	17	5698
MSG-030		Grinding Bay		289	0.0	1.1	10	1492
MSG-034		Laboratory Building		172	0.0	0.1	5	1000
MSG-028		Ore Receiving		3301	2.5	2.1	11	2348
MSG-029				1109	0.0	1.0	10	1750
MSG-031				7173	0.0	1.2	8	2389
MSG-032				1847	0.0	0.3	5	1037
MSG-027		Pond (Unknown)		2078	21.6	3.7	24	4088
MSG-007			CA Boundary	183	0.0	10.9	33	1000
MSG-008				497	0.0	3.8	17	1000
MSG-009				878	3.0	4.2	12	3066
MSG-010				1506	5.1	7.0	14	6745
MSG-011				48	0.0	3.6	12	1000
MSG-012				420	16.5	6.3	6	15177
MSG-013				146	0.0	1.0	6	1689
MSG-018			misc millyard	571	0.0	0.0	3	1000
MSG-019				143	0.0	0.0	1	2466

Station	**Category**	**Associated Feature**		**Radon (pCi/L)**	**Nitrous Oxide (ppmv)**	**Ammonia (ppmv)**	**Methane (ppmv)**	**Carbon Dioxide (ppmv)**
		Historical	**Modern**					
MSG-005	North Off-pile	Ore Storage		103	0.0	1.6	1	6694
MSG-006				130	0.0	0.7	2	3500
MSG-039				142	0.0	0.0	1	1055
MSG-040				53	0.0	0.3	3	1000
MSG-041				70	1.5	0.2	4	1260
MSG-042				99	0.9	0.0	4	1198
MSG-043				65	1.9	0.4	6	1768
MSG-044				147	2.5	1.1	4	4062
MSG-045				79	1.5	0.6	7	1518
MSG-046				49	1.5	0.0	9	1000
MSG-047			River	751	4.4	2.4	10	7691
MSG-048				73	2.3	1.3	4	5059
MSG-049				112	1.8	0.6	6	1773
MSG-050				58	1.4	0.6	4	1812
MSG-051				29	2.7	1.2	9	2759
MSG-052				118	3.4	1.5	4	6388
MSG-038			misc. North Off-pile	12	0.0	0.0	4	1000

Station	Category	Associated Feature		Radon (pCi/L)	Nitrous Oxide (ppmv)	Ammonia (ppmv)	Methane (ppmv)	Carbon Dioxide (ppmv)
		Historical	Modern					
MSG-015	Tailings	Pond (BaCl2)		8417	1.5	1.3	3	3730
MSG-021				470	4.7	1.5	5	3960
MSG-022				2097	6.5	1.2	7	1433
MSG-016		Pond (Purification)		102982	18.3	5.2	4	15100
MSG-017				6275	0.0	0.9	2	1748
MSG-014		Sump Pond		2834	0.0	1.7	4	3398
MSG-020				1834	0.0	0.2	3	1875
MSG-035			misc. tailings	10478	26.8	5.5	7	16378
MSG-036				3745	20.8	0.6	6	2708
MSG-037				2710	6.5	1.2	5	5698

Station	Category	Associated Feature		Radon (pCi/L)	Nitrous Oxide (ppmv)	Ammonia (ppmv)	Methane (ppmv)	Carbon Dioxide (ppmv)
		Historical	Modern					
MSG-053	Well Field		CA Boundary	100	1.3	0.9	4	2606
MSG-054				96	1.2	0.0	5	1000
MSG-055				182	48.4	3.1	3	10067
MSG-001			River	250	0.7	11.1	24	7231
MSG-002				413	1.6	11.3	24	8636
MSG-056				145	1.9	0.0	5	1265
MSG-057				212	1.5	1.6	3	4676
MSG-058				157	4.2	2.3	4	5749
MSG-003			misc. wellfield	174	0.0	0.7	4	2251
MSG-004				185	0.0	0.6	1	2167

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