

Helium-3/Helium-4 Ratios in Soil Gas as an Indicator of Tritium Contamination Near the 618-11 Burial Ground, Hanford Site, Washington

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Abstract

Pacific Northwest National Laboratory sampled and analyzed soil gas for helium-3 and helium-4 concentrations from the vicinity of the 618-11 burial ground. Helium-3, the first daughter product from tritium decay, was used to investigate the source and extent of tritium contamination in the area

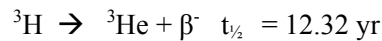
Seventy soil-gas sampling points were installed around the perimeter of the 618-11 burial ground, approximately 122 meters downgradient of well 699-13-3A, and in four transects downgradient of the burial ground to a maximum distance of 946 meters. Soil-gas samples were collected, analyzed for helium isotopes, and helium-3/helium-4 ratios were calculated from these 70 points. Helium-3/helium-4 ratios determined from the sampling points showed significant enrichments compared to ambient air helium-3/helium-4 ratios. The highest helium-3/helium-4 ratios were located along the north perimeter of the burial ground. Helium-3/ helium-4 ratios (normalized to their abundances in ambient air) ranged from 1.0 to 62 around the burial ground. The helium-3/helium-4 ratios from the 4 transects downgradient of the burial ground ranged from 0.988 to 1.68.

The helium-3/helium-4 ratios from around the burial ground suggest there is a vadose zone source of tritium along the north side of the burial ground. This vadose zone source is likely the source of tritium in the groundwater. The helium-3/helium-4 ratios also suggest the groundwater plume is traveling east-northeast from the burial ground, and the highest groundwater tritium value may be to the north of well 699-13-3A. Finally, there appears to be no significant upgradient sources of tritium affecting the burial ground since all the upgradient helium-3/helium-4 ratios equal the air background level of 1.0. Based on the helium-3/helium-4 results from the soil-gas survey, six downgradient sampling locations were

identified to verify the tritium groundwater plume locations and tritium groundwater concentrations. The results of the measurement of helium isotopes in soil gas provided a rapid and cost-effective technique to define the shape and extent of tritium contamination from the 618-11 burial ground.

Introduction

This study measured helium-3/helium-4 ratios relative to ambient air in soil-gas samples to detect and delineate groundwater tritium plumes originating from the 618-11 burial ground. This approach is a modification of a successful technique developed in the late 1960s used to age-date deep ocean water as part of the GEOSECS ocean monitoring program and applied to shallow aquifers in the late 1980s by Poreda et al. (1988) and Schlosser et al. (1988). An earlier study was conducted in 1999 on the Hanford Site to demonstrate the proof-of-concept for using helium-3 as a tool to locate vadose zone sources of tritium and tracking groundwater tritium plumes (Olsen et al. 1999). All these studies were based on the presence of tritium, which decays to a stable, inert isotope, helium-3, i.e.:



When soil moisture reacts with tritium waste, the result is tritiated moisture that is released into the vadose zone. The moisture migrates downward to mix with groundwater, with meteoric recharge providing the primary driving force. Concurrent with tritium's release to the vadose zone, its daughter isotope, helium-3, begins to build up in the vadose zone and groundwater at the rate of tritium decay. The helium-3 then diffuses away from its vadose or groundwater source and migrates toward the surface. Because helium-3 is nonreactive in the subsurface environment it is a good surrogate tracer for tritium in the vadose zone and groundwater.

This helium soil-gas survey was part of a study designed to help define the tritium plume originating from the 618-11 burial ground. This study began in January 2000 after elevated tritium was detected in well 699-13-3A, immediately east of the burial ground.

Historical Background

The 618-11 burial ground covers an area of 3.5 hectares and is located adjacent to an operating commercial reactor facility. The site consists of 3 trenches, 2 to 5 large-diameter caissons, and 50 vertical pipe storage units. The trenches are 274 meters long by 15 meters wide; vertical pipe units are five 208 liter drums welded together end-to-end and are approximately 4.6 meters long by 56 centimeters in diameter. The caissons are metal pipe 2.4 meters in diameter, 3 meters long, buried vertically 4.6 meters below grade, connected to the surface by an offset pipe 91 centimeters in diameter pipe connected to a dome-type cap. All vertical pipe units and caissons were capped with concrete and covered with dirt (Demiter and Greenhalgh 1997).

The burial ground received low to high-activity dry waste, fission products, plutonium, and other transuranic constituents in a variety of waste forms from research operations (DOE 2000). The burial ground operated from 1962 through 1967 (DOE 2000). The operation of this burial ground coincided

with the development of the lithium aluminate tritium target project on the Hanford Site. Circumstantial evidence suggests that tritium targets, as irradiated lithium aluminate, may have been disposed to the burial ground during its operational lifetime.

Driven by the strategic need for tritium production by our weapons program, it was recognized that co-production of plutonium and tritium during operations of the reactor would be technically, economically, and strategically attractive. Thus, General Electric Company initiated a coproduct program for N Reactor early in 1963. Several fuel-target models were tested in the reactor and testing culminated in October 1965 with the selection of the Mark II design used for full reactor demonstrations. After irradiation, tritium must efficiently be removed from the targets. Development of a tritium extraction process for ceramic targets was jointly assigned to Battelle, Pacific Northwest Laboratory and Savannah River Laboratories. Initial research in the development of a process to recover tritium from irradiated lithium aluminate was done with samples weighing less than one gram. Production-size pellets (70 grams [2.5 ounces]) became available on February 7, 1966 (Johnson et al. 1976). These pellets were used in a series of experiments designed to determine the characteristics of the tritium extraction. Results of this study estimated that each target contained approximately 51 curies of tritium and that 35% of the tritium was recovered in a non-condensable form at -196°C with the remainder recovered in a condensable form as T_2O or HTO. Furthermore, residual tritium remaining with the lithium aluminate target after processing ranged from <0.4 to 5% (Yunker 1976). Based on the aforementioned information, the tritium remaining in each target could range from 0.204 curies to 2.55 curies. The condensable form is believed to be what mainly remains within the lithium aluminate target material after processing. This form is also an environmental labile form, which when released to the environment reacts with soil moisture in the vadose zone and can be carried downward with meteoritic water to groundwater.

Methods

Seventy points to sample soil gas were installed during the course of this study. Forty-three sampling points were installed in August 2000 around the 618-11 burial ground and within 122 meters to the east (downgradient) of the burial ground. Twenty-seven sampling points were installed in April to May 2001 in four transects to the east of the burial ground. Sampling points ranging from 305 meters to the north, 518.5 meters to the south, and 945.5 meters to the east of groundwater monitoring well 699-13-3A. The logic for locating the soil-gas points in four transects is presented in the discussion section of this report.

Sample Point Installation

Sampling points for soil gas were installed using a truck mounted GeoprobeTM Model 5400 system equipped with a probe 3.2 centimeters in diameter with a detachable steel tip. Target depth of installation of the screen interval was 6.1 meters below ground surface (bgs), but actually ranged from 4.4 to 6.3 meters bgs. When the tip achieved its desired depth, a 20.3 centimeter long, fine-mesh, stainless steel sampling point connected to the surface with a polyethylene tube 6.35 millimeters outside diameter by 2.38 millimeters inside diameter that was strung down the center of the push rod. The rod assembly was withdrawn 15.2 centimeters to release the steel tip and allow the sampling point to extend into the void space just below the push rod. Approximately 250 milliliters of 20 to 40 mesh washed silica sand was added around the sampling point through the center of the push rod. The push rod was slowly withdrawn

and bentonite pellets were added through the center of the push rod. The bentonite was hydrated with 240 milliliters water several feet above the screened interval. Bentonite pellets were added to the surface of the hole. To complete those sampling points, a cement cap was poured around the sampling tube at the ground surface. Each sampling location was allowed to equilibrate for at least 24 hours and up to several weeks before soil-gas samples were collected.

Sample Collection

All soil-gas samples were collected with the aid of a Thomas Model 107CA14 flexible diaphragm pump. Power to the pumps was supplied with a portable generator.

Pressurized samples were collected for helium analysis from each sampling location. The sampling vessels were 50 milliliter stainless steel cylinders with one end sealed with a pipe plug and the other end with a high vacuum needle valve with a 1/4-inch Swagelok™ fitting. Each vessel was evacuated to less than 5 torr before sampling. The sampling configurations used to collect helium-3 samples can be seen in Figure 1. After a short equilibration period (minutes), a Kurz Instruments™ mass flow meter was placed in the flow stream between the polyethylene raiser tube and the bottom of the rotometer. The initial flow was adjusted to 1 liter per minute. The soil-gas sampling point was allowed to purge at 1 liter per minute for a minimum of 15 minutes. At the end of the purging period, a hose was connected to the pressure side of the pump, and the sampling cylinder was pressurized to the maximum pressure of the pump, allowed to vent to atmospheric pressure twice without removing the sampling tube from the sample cylinder, then allowed to pressurize to the maximum pressure of the pump. Random duplicates were collected at 1 for every 20 samples, and an air background sample was included for each sampling day. When there were discontinuities in sampling (e.g., a day or weeks) a previously sampled soil-gas location was resampled.

Groundwater grab samples were taken at six locations suggested by the helium-3/helium-4 ratio results. Six boreholes to groundwater were drilled using a cable-tool drill rig. Once groundwater was reached, drilling continued to approximately 1 meter into the aquifer and then the drill casing was back-pulled to allow water to enter the hole for sampling. Water was not added during drilling, so the water entering the borehole was considered representative of groundwater at that location. All the grab samples were collected with a bailer. Four of these six boreholes were later converted to permanent monitoring wells.

Sample Analysis

After collection, soil-gas samples were sent to the University of Rochester for helium isotopes (helium-3 and helium-4) analysis. Upon receipt of the samples, a 0.5 milliliters aliquot of soil gas was processed through a high vacuum line constructed of stainless steel and Corning-1724 glass to minimize helium diffusion. After removal of water vapor and carbon dioxide at -90°C and -195°C respectively, the amount of non-condensable gas (helium, neon, argon, oxygen, nitrogen, and methane) was measured using a calibrated volume and a capacitance manometer. Gas ratios (argon, nitrogen, methane) were analyzed on a Dycor Quadrupole mass spectrometer fitted with a variable leak valve. The results are combined with the capacitance manometer measurement to obtain gas concentrations ($\pm 2\%$). Prior to helium isotope analyses, nitrogen and oxygen were removed by reaction with zirconium/aluminum alloy (SAES ST707),

argon and neon were adsorbed on activated charcoal at 77°K and at 40°K, respectively. SAES-ST-101 Getters (one in the inlet line and 2 in the mass spectrometer) reduced the HD⁺ background to ~1,000 ions/sec.

Tritium in groundwater was measured using standard liquid scintillation beta counting. An aliquot of groundwater is made alkaline with sodium hydroxide and distilled. After distillation, the distillate and a scintillation cocktail is mixed into a scintillation vial. The solution is placed into a refrigerated liquid-scintillation counter and counted. Detection limits range from 300 pCi/L to 400 pCi/L tritium.

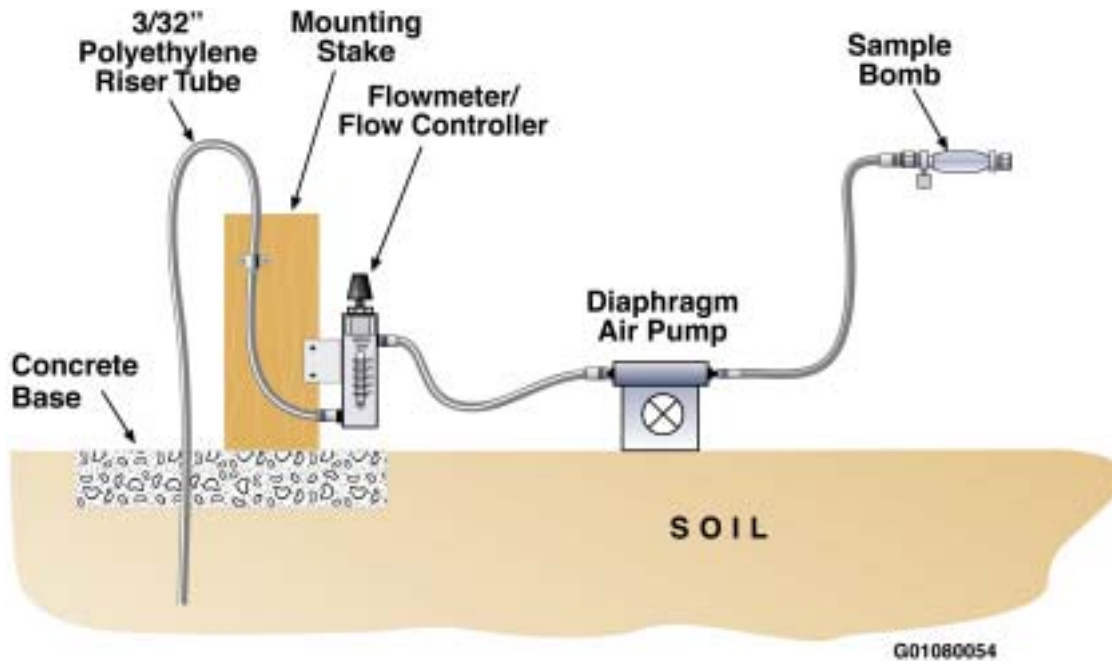


Figure 1. Schematic Diagram of the Soil Gas Sampling System Used for Collection of Helium Soil-Gas Samples

Helium isotope ratios and concentrations in soil gas were analyzed on a VG 5400 rare gas mass spectrometer fitted with a Faraday cup (resolution of 200) and a Johnston electron multiplier (resolution of 600) for sequential analyses of the helium-4 (F-cup) and helium-3 (multiplier) beams. On the axial collector (resolution of 600), $^3\text{He}^+$ was completely separated from HD^+ with a baseline separation of <2% of the HD^+ peak. The contribution of HD^+ to the helium-3 peak was <0.1 ion/sec at 1,000 ions/sec of HD^+ . For 2.0 milliliters of helium with an air ratio sensitivity of 2×10^{-4} Amps/torr, the helium-3 signal averaged 2,500 ions/sec with a background signal of ~15 cps, due to either scattered helium-4 ions or the formation of helium-4 ions at lower voltage potentials within the source of the mass spectrometer. All helium-3/ helium-4 ratios were reported relative to the atmospheric ratio (R_A), using air helium as the absolute standard. Errors in the helium-3/helium-4 ratios result from the precision of the sample measurement ($\pm 0.2\%$) and variation in the ratio measurement in air ($\pm 0.2\%$) and give a total error of $\pm 0.3\%$ at 2σ for the reported helium isotope value. Helium concentrations were derived from comparison

of the total sample to a standard of known size. The value, as measured by peak height comparison, was accurate to $\pm 1\%$ (2σ).

Results and Discussion

Sampling points for soil gas were installed and sampled in two time periods (DOE 2001). Installation of the first set of sampling points occurred in August and September 2000 around the 618-11 burial ground and within 122 meters of the burial ground in a downgradient direction from groundwater monitoring well 699-13-3A. The second set of sampling points were installed in April to May 2001 and focused on locations downgradient and cross-gradient of the groundwater flow direction beneath 618-11 burial ground. The second set of sampling points were arranged in four transects ranging 305 meters to the north, 518.5 meters to the south, and 945.5 meters to the east of groundwater monitoring well 699-13-3A located on the east side of the 618-11 burial ground.

Burial Ground Results

Figure 2 displays the results from soil-gas samples collected from the perimeter of the burial ground and immediately downgradient of well 699-13-3A. Helium-3/helium-4 ratios ranged from 1.00 to 62.5 in this region. The highest helium-3/helium-4 ratios were located on the north side of the burial ground where two distinct maxima were observed. One maximum (62.5) was located near the middle of the north side fence line. The second maximum (10.93) was located at the northeast corner of the burial ground. The highest helium-3/helium-4 ratio along the north side of the burial ground was believed to be either from a tritium source in the vadose zone or tritium contaminated groundwater. In order to determine the likely source of helium-3 (vadose zone source or groundwater), a groundwater grab sample was collected at the location of the highest helium-3/helium-4 ratio. This sample was collected using cable tool drilling techniques. The groundwater sample from that location contained 6,510 pCi/l of tritium. After the sample was collected, the hole was abandoned. This result strongly suggested that the highest helium-3 concentrations midway along the north side of the burial ground was from a vadose source of tritium located within the burial ground rather than from tritium-contaminated groundwater. Thus, the likely source of the tritium is a series of caissons located along the north side of the burial ground.

The second helium-3/helium-4 maximum along the north side of the burial ground is located at the northeast corner. The source of the elevated helium-3/helium-4 ratios could also be from a vadose zone source of tritium or a tritium groundwater plume. However, it is believed that the source of the elevated helium-3/helium-4 ratios is from the tritium groundwater plume that affects well 699-13-3A. The tritium concentration in groundwater samples from that well measured from 1.86 to 8.38 million pCi/L (Figure 3). The direction of groundwater flow is believed to be in a generally easterly direction. It appears that the highest groundwater tritium concentration is slightly to the north of well 699-13-3A, corresponding to the highest helium-3/helium-4 ratios along the northeast corner of the burial ground, though this has not been confirmed by groundwater sampling. If you compare the helium-3/helium-4 ratio at the

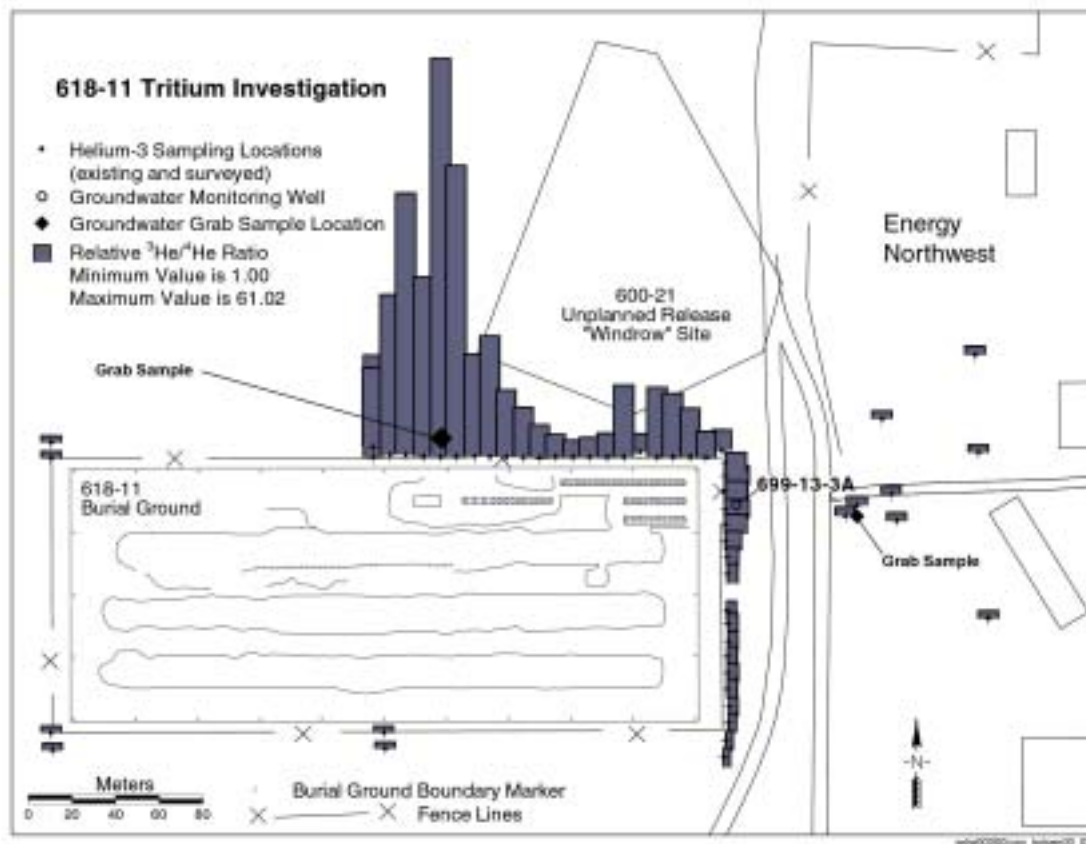


Figure 2. Helium-3/Helium-4 Ratio Results from Soil Gas Samples from Around 618-11 Burial Ground and Immediately Downgradient of Well 699-13-3A

soil-gas point nearest the well (3.73) to the highest helium-3/helium-4 ratio at the northeast corner of the burial ground (10.93), one would estimate a tritium concentrations in groundwater below that point at 30.3 million pCi/L.

The helium-3/helium-4 ratios on the east side of the burial ground are highest at the northeast corner, adjacent to well 699-13-3A, and decrease to 1.0 at the south side of the burial ground (Figure 4). This maximum helium-3/helium-4 ratio (3.73) is believed to be from the tritium groundwater plume affecting well 699-13-3A and follows the discussion presented above for the north side of the burial ground. There appears to be a second smaller maximum (1.63) located 45.7 meters north of the southeast corner of the burial ground. The second maximum may be a vadose zone source of tritium or a groundwater tritium source. The remaining points along the south and west sides of the burial ground are approaching the ambient air helium-3/helium-4 ratio of 1.0.

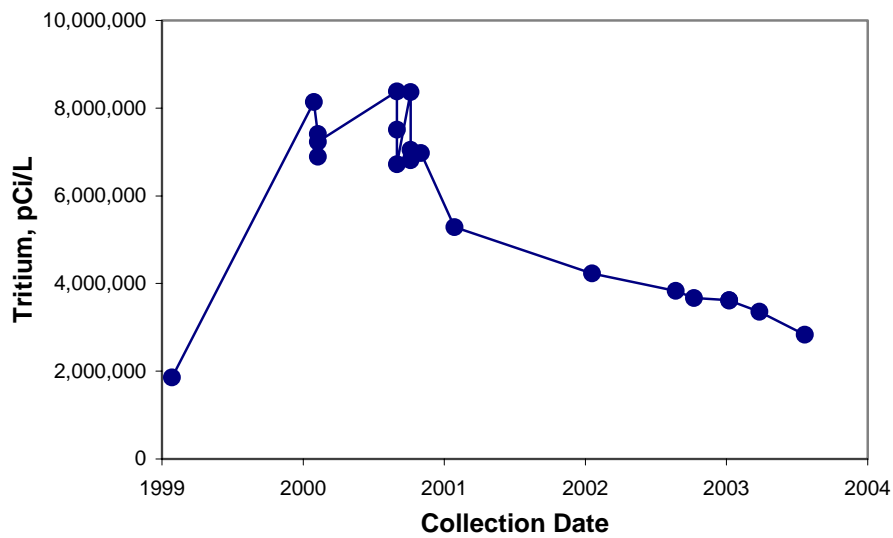


Figure 3. Tritium Concentrations from Well 699-13-3A

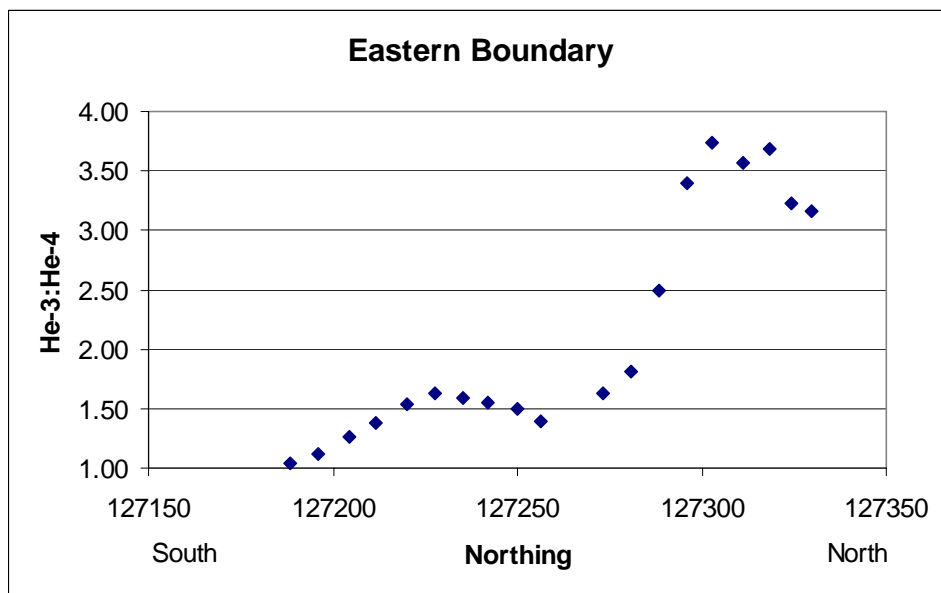


Figure 4. Helium-3/Helium-4 Ratio Results from Soil-Gas Samples from the East Side of the 618-11 Burial Ground

Downgradient Results

Helium-3/helium-4 ratios were determined in soil-gas sampling points installed within 122 meters east of the 618-11 burial ground in August and September 2000 and the four transects downgradient from

the burial ground in April to May 2001. Soil-gas sampling points in the four transects ranged from 305 meters to the north, 518.5 meters to the south, and 945.5 meters to the east of groundwater monitoring well 699-13-3A.

The helium-3/helium-4 ratios in the first round of sampling from soil gas locations within 122 meters of the burial ground ranged from 1.12 to 1.38. Based on these results, a groundwater grab sample was collected near the cluster of soil-gas sampling points approximately 45.7 meters east of well 699-13-3A (see Figure 2). The tritium concentration in the groundwater from this grab sample location was measured at 1.5 million pCi/L.

Based on the helium-3/helium-4 results from the perimeter of burial ground and within a radius of 122 meters of well 699-13-3A in the downgradient direction and the two groundwater grab samples, a tritium groundwater plume appears to be heading east-northeast from the burial ground. Based on this assumption, 27 additional soil-gas sampling points were installed in four transects further downgradient of well 699-13-3A (Figure 5). To assure there were no additional tritium sources in the area of the 618-11 burial ground and, with some certainty, that helium-3/helium-4 ratios returned to background levels (1.00) Transect 1 and 3 spanned 793 meters and 732 meters, respectively. Transect 2 consisted of two soil-gas sampling points. Transect 2 was limited because of industrial facilities and construction activities in the immediate area. Transect 4 was located 945.5 meters from well 699-13-3A and spanned 213.5 meters across the potential path of the tritium plume.

The helium-3/helium-4 ratios in Transect 1 ranged from 0.99 to 1.68. Background helium-3/helium-4 ratios were obtained at the ends of each transect and the maximum helium-3/helium-4 ratios were near the imaginary centerline of the tritium plume generated from earlier data. The graph of data from Transect 1 shows that the helium-3/helium-4 signal has an asymmetrical shape with high ratios extending farther south of the maximum point (Figure 6). This asymmetry appears to be an extension of the secondary maximum seen along the east boundary of the burial ground (see Figure 4).

The helium-3/helium-4 ratios in Transect 2, which included only two sampling points, ranged from 1.06 to 1.27. The highest ratio was closest to the hypothesized centerline of the plume.

The helium-3/helium-4 ratios in Transect 3 ranged from 0.987 to 1.049. Clearly elevated helium-3/helium-4 ratios were only seen in two sampling points. Background helium-3/helium-4 ratios were obtained at the ends of each transect, and the maximum helium-3/helium-4 ratios were near the imaginary centerline of the tritium plume generated from the earlier data.

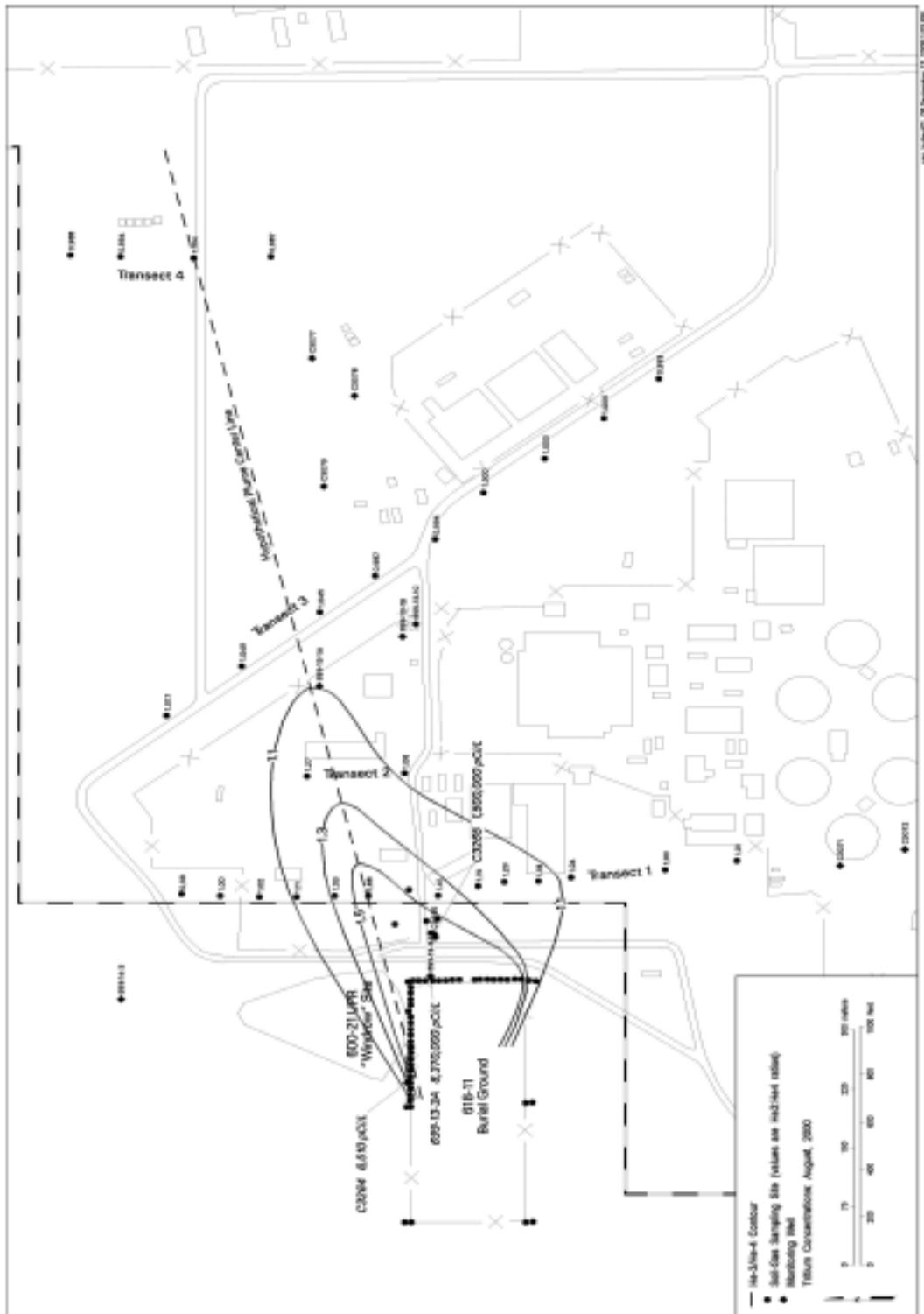


Figure 5. Helium-3/Helium-4 Ratio Results and Isopleths from Soil-Gas Samples Downgradient of the 618-11 Burial Ground

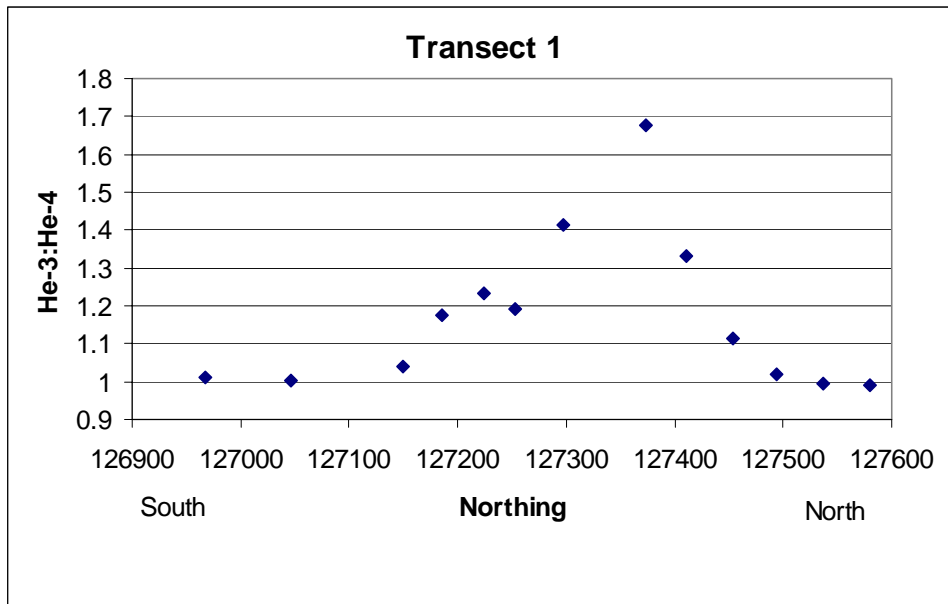


Figure 6. Helium-3/Helium-4 Ratio Results from Soil-Gas Samples from Transect 1

The helium-3/helium-4 ratios in Transect 4 ranged from 0.987 to 1.104. Background helium-3/helium-4 ratios were obtained at the ends of each transect and elevated helium-3/helium-4 ratios were observed in only one sampling point. Transect 4 is topographically lower than the other transects so the sample points are closer to the water table. This may explain why the maximum helium-3/helium-4 ratio seen in Transect 4 is greater than the maximum seen closer to the burial ground, in Transect 3.

Groundwater Results

Using the results from the soil-gas survey, six new wells were installed and sampled. The results of groundwater sampling indicated the highest level of tritium was found at well 699-13-3A directly adjacent and downgradient of the burial ground. Tritium concentrations at well 699-13-3A ranged from 1.86 million to 8 million pCi/L (see Figure 3) and represented a localized plume originating from the burial ground. Figure 7 is a plume map generated using tritium groundwater concentrations from 12 different locations within the study area. The plume appears to be moving along a narrow path in an east-northeast direction, as indicated by the soil gas. The tritium groundwater plume migrated 950 meters downgradient of the burial ground, and the spatial distribution of tritium in groundwater is consistent with the pattern revealed by the soil-gas survey results.

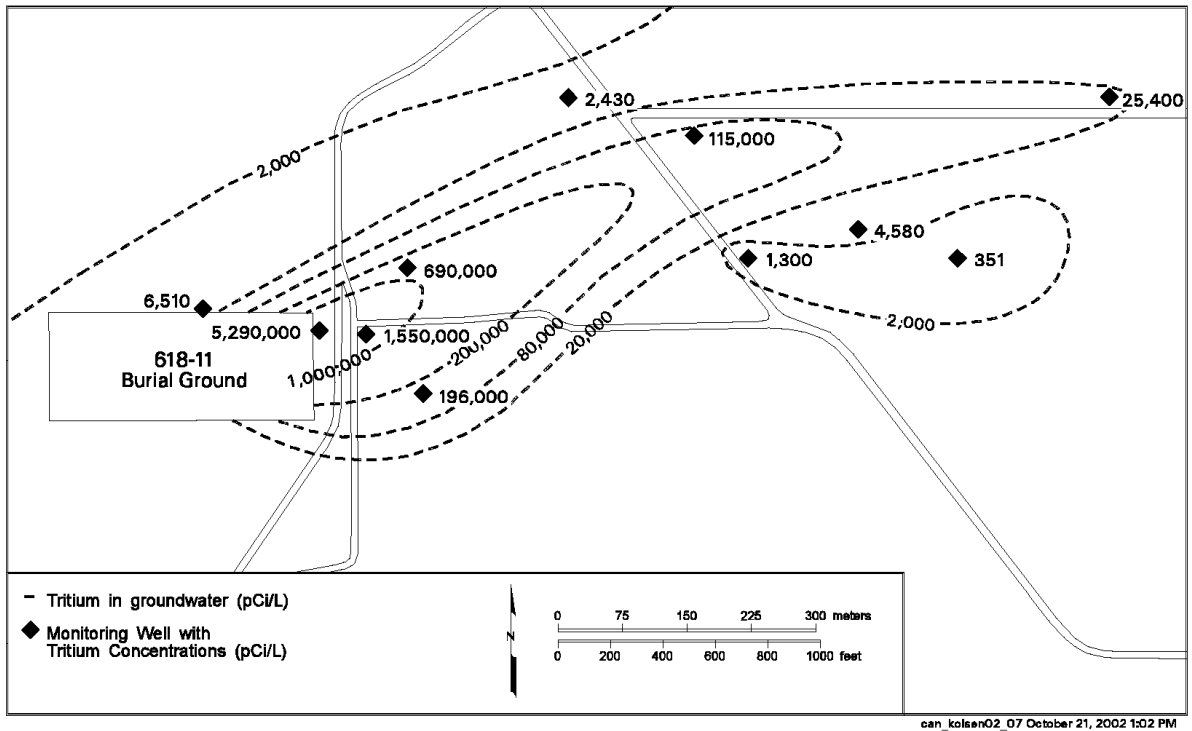


Figure 7. Tritium Groundwater Concentrations and Tritium Isopleths Results from 618-11 Burial Ground.

Conclusions

The helium soil-gas measurements provided a rapid and cost-effective technique to define the probable shape and extent of tritium contamination from the 618-11 burial ground. Based on the helium-3/helium-4 ratio results from around the burial ground and the groundwater grab samples from the highest helium-3/helium-4 ratio on the north side, the main vadose zone source of the tritium is likely from within the burial ground near the mid point of the north side. The second helium-3/helium-4 ratio maximum located at the northeast corner of the burial ground is probably from the groundwater tritium plume affecting well 699-13-3A.

Based on the helium-3/helium-4 ratio data immediately downgradient of burial ground 618-11 and the downgradient transects, a groundwater tritium plume appears to be traveling toward the east-northeast direction. The northeastward direction of plume movement was not predicted prior to the investigation by initial hydrologic and geologic interpretations that predicted flow directly to the east or to the southeast. The tritium groundwater plume has reached the farthest transect for the burial ground 950 meters downgradient of the burial ground. The width of the tritium plume is estimated to be 305 meters at Transect 1, 204.3 meters at Transect 3, and 88.5 meters at Transect 4. The helium-3/helium-4 ratio results also suggest there are no other tritium sources either upgradient of the burial ground or cross gradient of the burial ground. Based on the helium-3/helium-4 results from the soil-gas survey, six downgradient groundwater sampling locations were identified to verify the tritium groundwater plume locations and tritium groundwater concentrations.

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