CHARACTERIZING TRITIUM WASTE USING HELIUM RATIOS

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ABSTRACT

When routine sampling revealed greatly elevated tritium levels (3.14 x 10^5 Bq/L [8.5-million pCi/liter]) in the groundwater near a solid waste landfill at the Hanford Site, an innovative technique was used to assess the extent of the plume. Helium-3/helium-4 ratios, relative to ambient air-in-soil gas samples, were used to identify the tritium source and initially delineate the extent of the groundwater tritium plume. This approach is a modification of a technique developed in the late 1960s to age-date deep ocean water as part of the GEOSECS ocean monitoring program. Poreda, *et al.* (1) and Schlosser, *et al.* (2) applied this modified technique to shallow aquifers. A study was also conducted to demonstrate the concept of using helium-3 as a tool to locate vadose zone sources of tritium and tracking groundwater tritium plumes at Hanford (3).

Seventy sampling points were installed around the perimeter and along four transects downgradient of the burial ground. Soil gas samples were collected, analyzed for helium isotopes, and helium-3/helium-4 ratios were calculated for these 70 points. The helium ratios indicated a vadose zone source of tritium along the northern edge of the burial ground that is likely the source of tritium in the groundwater. The helium ratios also indicate the groundwater plume is traveling east-northeast from the burial ground and that no up-gradient tritium sources are affecting the burial ground. Based on the helium ratio results, six downgradient groundwater sampling locations were identified to verify the tritium plume extent and groundwater tritium concentrations. The tritium results from the initial groundwater samples confirmed that elevated helium ratios were indicative of tritium contamination in the local groundwater.

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 burial ground. Using this soil gas sampling approach, the project team was able to identify areas where elevated tritium existed in groundwater without going to the time and expense of conducting conventional groundwater characterization sampling. The savings from this characterization approach were $1.4 million.

INTRODUCTION

Routine groundwater sampling revealed greatly elevated tritium levels (3.14 x 10^5 Bq/L [8.5-million pCi/liter]) in a monitoring well near solid waste landfill 618-11 at the Hanford Site. This burial ground consists of several waste storage caissons, 50 vertical pipe units, and three trenches that received low- to high-activity wastes, fission products, plutonium, and other transuranic constituents in a variety of dry waste forms. In addition, some elements of the buried inventory are chemically reactive wastes that could ignite under the right conditions (e.g., in air).

The 618-11 Burial Ground covers an area of 3.5 x 10^4 m^2 (8.6 acres) and operated from 1962 through 1967. 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. Based on historical process information, the tritium remaining in each target could range from 7.55 x 10^9 to 9.44 x 10^{10} Bq (0.204 to 2.55 curies). The condensable tritium form is believed to be what mainly remains within the lithium aluminate target material after processing. This tritium form can react with vadose zone soil moisture and can be carried downward with meteoritic water to groundwater.

The scope of this study was to verify that burial ground 618-11 was the local groundwater tritium plume source and to define the extent of the plume. The study included analyzing soil gas samples for helium isotopes and using helium-3/helium-4 isotope ratios to initially delineate the extent of the plume. Using this initial tritium plume delineation as a guide, only six boreholes were required to collect groundwater samples that verified the helium ratio results and the initially delineated plume footprint. Four of these six boreholes were converted to monitoring wells to track future tritium plume movement.

METHODS

Soil Gas Sample Collection and Analysis

Soil-gas sampling points were installed using a truck-mounted Geoprobe® Model 5400 system equipped with a probe 3.2 centimeters (1.25 inches) in diameter with a detachable steel tip. The target depth of installation of the screen interval was 6.1 meters (20 feet) below ground surface. Approximately 250 milliliters of 20- to 40-mesh, washed silica sand was added around the sampling point and bentonite pellets were added above the sand to the surface of the hole. The bentonite was hydrated with 240 milliliters water several feet above the screened interval. To complete the 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 before soil gas samples were collected. Seventy soil gas sampling points were installed around the burial ground perimeter (with emphasis along the north and east boundaries) and along four transects ranging from 122 meters (400 feet) to just over 945 meters (3,100 feet) to the east of the 618-11 Burial Ground (Figure 1).

Pressurized samples were collected for helium analysis from each location (Figure 2). The sampling vessels were 50-milliliter stainless steel cylinders. Each vessel was evacuated to less than 667 Pa (5 torr) before sampling. Prior to sample collection, the soil gas sampling point was allowed to purge at 1 liter (0.035 $feet^3$) per minute for a minimum of 15 minutes. At the end of the purging period, 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 (1 for every 20 samples) and a background air sample was collected each sampling day.

Helium isotope ratios and concentrations 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. 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 (±0.2%) and variation in the ratio measurement in air (±0.2%) and give a total error of ±0.3% at 2σ for the reported helium isotope value. Helium concentrations were derived by comparing total samples to a standard of known size. The value, as measured by peak height comparison, was accurate to ±1% (2σ).

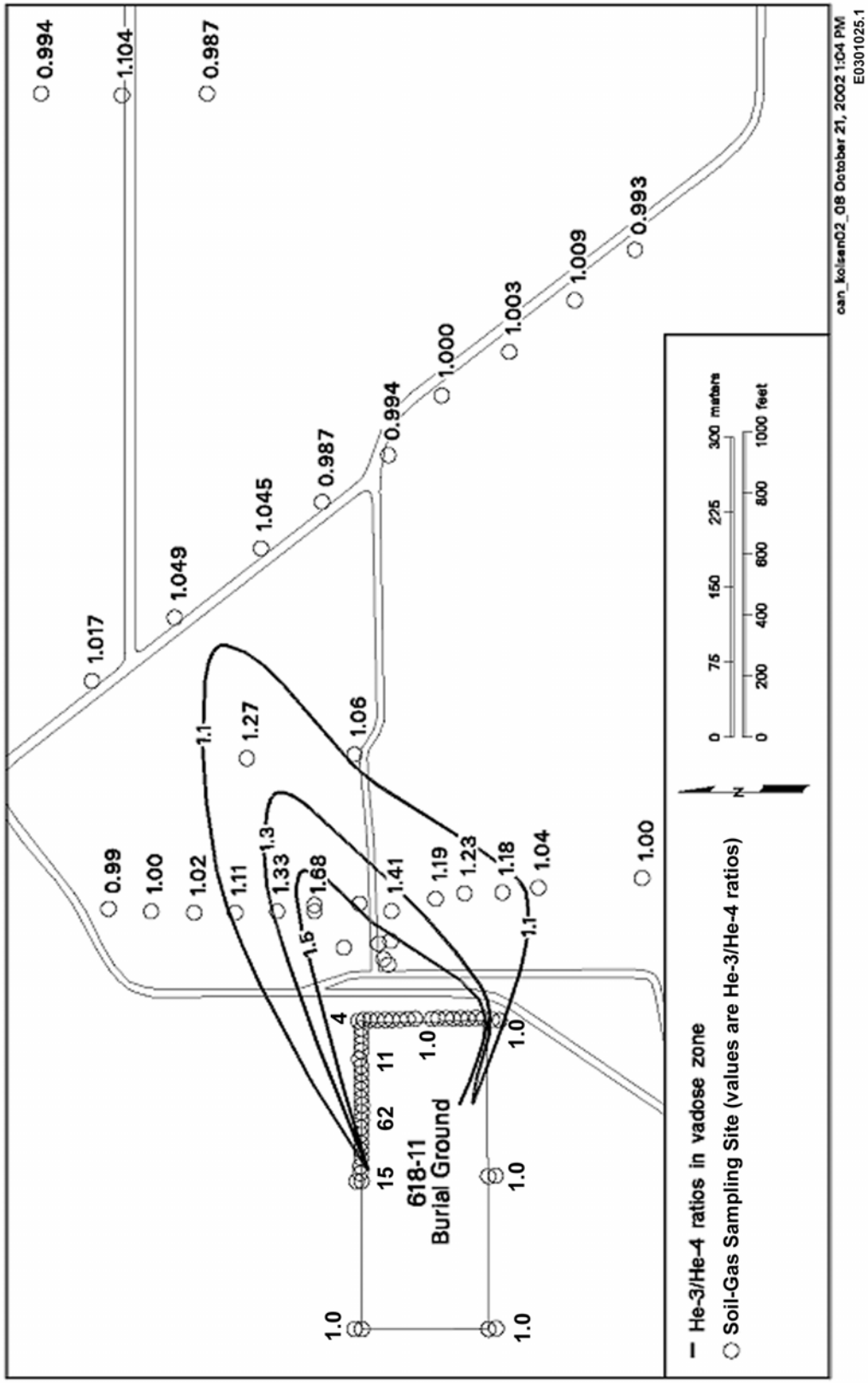

Figure 1. Soil Gas Sample Locations and Helium Ratio Results

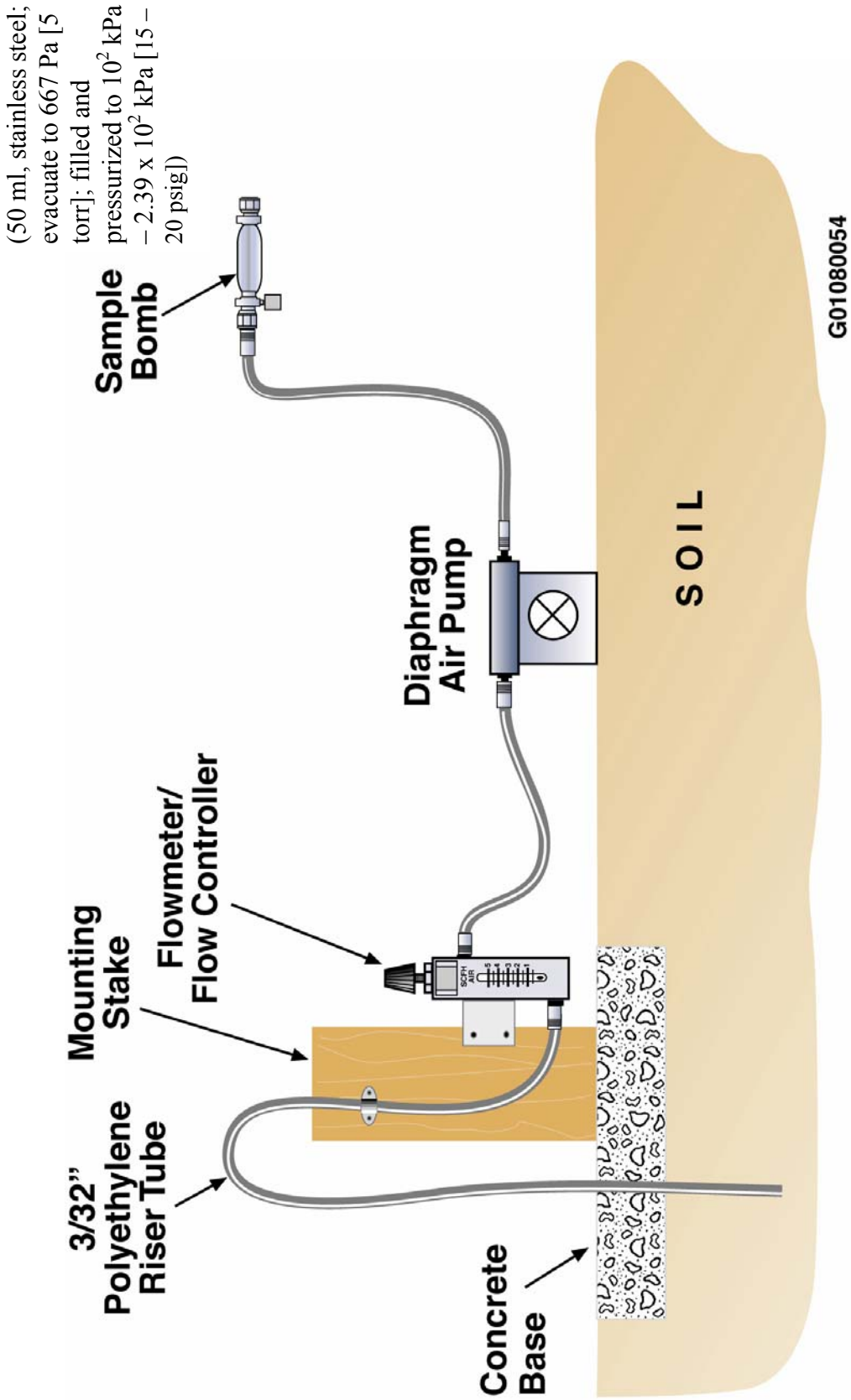

Figure 2. Sampling Schematic Diagram

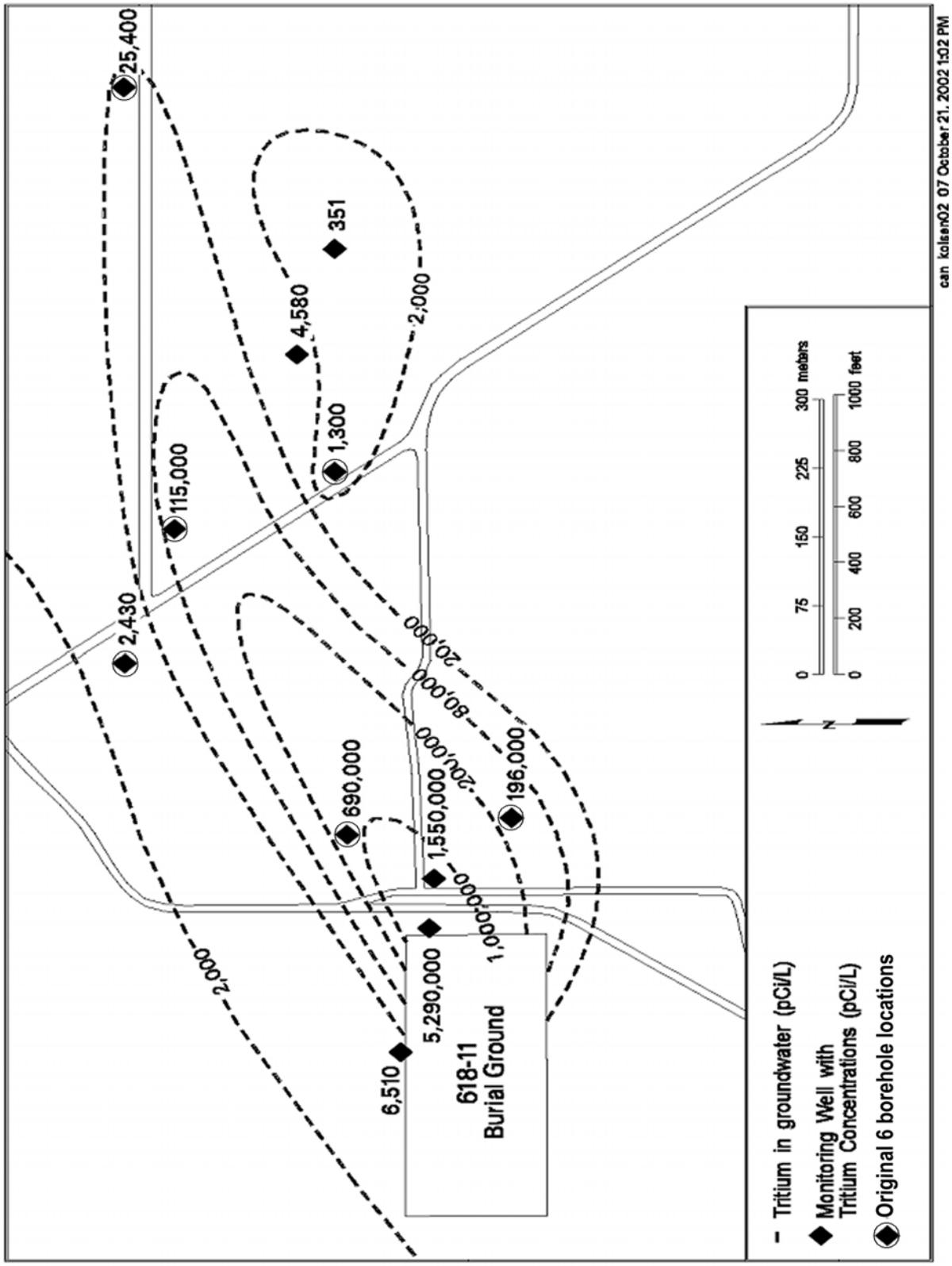

Figure 3. Groundwater Tritium Results

Groundwater Samples

Based on the soil gas helium ratio results, six downgradient borehole locations were identified to verify the tritium plume extent and groundwater tritium concentrations (Figure 3). The boreholes were drilled using a cable-tool drill rig and groundwater samples were collected from each site. Once groundwater was reached, drilling continued to about 0.9 meter (3 feet) into the aquifer, then the drill casing was back-pulled to allow water to come into the hole for sampling. Water was not added during drilling, so the water entering into the borehole was considered to be representative of groundwater. All the grab samples were collected with a bailer and many of the samples were turbid. Since the main analyte of concern for this investigation is tritium, the turbidity of the samples did not affect the quality of the analytical results. The samples were analyzed for tritium, gross alpha, gross beta, technetium-99, field parameters, inductively coupled plasma (ICP) metals, anions and alkalinity.

RESULTS AND DISCUSSION

Soil Gas

Soil gas samples were collected, analyzed for helium isotopes, and helium-3/helium-4 ratios were calculated for the 70 sampling locations. The highest helium ratios (normalized to ambient air) were located along the north edge of the burial ground. Helium ratios ranged from 1.0 to 62 around the burial ground. The helium ratios along the four transects downgradient of the burial ground ranged from 0.988 to 1.68.

The results show extremely high helium ratios (greater than 61) along the northern side of the burial ground (Figure 1). However, a groundwater grab sample collected from a borehole (C3264) at the location of the highest helium ratio, had a tritium concentration of only 241 Bq/L (6,510 pCi/L). This demonstrated that the helium ratio at that location is associated with a vadose source within the burial ground. A secondary high near the northeastern corner is likely associated with the highest tritium concentration in the groundwater plume. This high helium ratio extends along the east side of the burial ground, past monitoring well 699-13-3A. The helium ratios returned to background values (1.00) along the western and southern sides of the burial ground, suggesting there are no other sources of tritium up-gradient or south of the burial ground.

The results of the helium soil-gas transects are also shown in Figure 1. The fourteen readings in Transect 1 located about 122 meters (400 feet) from the burial ground, continue to show higher helium ratios off the northeast corner of the burial ground and slightly elevated helium-3/helium-4 ratios off the eastern side. Background helium-3/helium-4 ratios are observed at both ends of Transect 1. Transect 2's two readings, about 259 meters (850 feet) from the burial ground, continued to track the hypothetical centerline of the plume from the burial ground. Transect 3's nine readings, about 518 meters (1,700 feet) from the burial ground, again verify the presence of the tritium plume, however, the helium-3/helium-4 ratios suggest the plume has narrowed to 204 meters (670 feet) because only two points indicate the presence of tritium in the groundwater. Transect 4's four readings, located about 945 meters (3,100 feet) from the burial ground, had only one sample point that indicated tritium in the groundwater. Although the helium-3/helium-4 ratio (1.104) from the soil gas point at Transect 4 looks relatively high, it is dependent on the distance to groundwater and the time the plume has been in this area. The distance to groundwater along Transect 4 is about 6 meters (20 feet) less than the distance at the other transects. Based on the helium-3/helium-4 ratio results, the width of the plume at Transect 4 is about 88 meters (290 feet).

Groundwater

The tritium results from the initial groundwater samples (Figure 3) confirmed that elevated helium ratios were indicative of tritium contamination in the local groundwater.

Tritium Plume Extent

The groundwater results indicate that high levels of tritium do not appear to be widespread, but do appear to be moving in an east-northeast direction (Figure 3). The tritium plume originating from within the 618-11 Burial Ground has migrated about 975 meters (3,198 feet). The plume appears to follow a rather narrow path. The geology indicates that the water table occurs primarily in the Ringold Formation downgradient of the 618-11 Burial Ground, where groundwater flow occurs on the order of 0.3 meter/day (1 feet/day).

CONCLUSIONS

The outcome of this characterization effort established a baseline for the horizontal extent of the 618-11 Burial Ground tritium plume. The horizontal extent is relatively narrow and limited. Four new monitoring wells were installed as part of this effort. These new wells (plus existing well 699-13-3A) will be used to track the movement of the tritium plume with time, observe any trends in concentration over time, help determine the natural attenuation of tritium in this area, and address concerns on the impact of the 618-11 Burial Ground on groundwater.

The measurement of helium isotopes in soil gas provided a rapid and cost-effective technique to initially define the shape and extent of tritium contamination from the burial ground. Using this approach, the project team was able to identify areas where elevated tritium existed in groundwater without going to the time and expense of conducting conventional groundwater characterization sampling. The savings from this characterization approach were $1.4 million.

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