Sampling and Analysis of Rare Gas Isotopes for In Situ Delineation of Buried Transuranic and Tritium Waste

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ABSTRACT

Improved understanding of the type and location of buried waste is needed at several U. S. Department of Energy sites to make remedial decisions and for planning excavation and retrieval activities. Soil-gas sampling for the rare gases helium and xenon can be used to define the locations of tritium and transuranic waste. Soil-gas sampling is a minimally invasive technique, in that direct penetration of the waste is not required and no hazardous material is brought to the surface. Sampling options include shallow hand or jackhammer driven sampling points and deeper Geoprobe® or cone penetrometer direct-push methods.

Short-lived radioxenon isotopes are continuously produced by spontaneous fission of Pu-240 in wastes. Large volume soil-gas samples provide an extremely sensitive way to measure radioactive xenon in the subsurface and characterize the presence of transuranic waste. The analysis employs a modified Automated Radioxenon Sampling and Analysis (ARSA) system, developed for Comprehensive Test Ban Treaty verification. Proof-of-principle measurements at a Hanford Site liquid waste disposal facility showed Xe-133 at levels approximately 16,000 times the detection limit and lower levels of Xe-135.

Stable xenon isotopes are produced in nuclear fuel during reactor operations and to a lesser extent by spontaneous fission in transuranic waste. The isotopic ratios are determined by rare-gas mass spectrometry and can be used to distinguish reactor xenon from spontaneous fission and natural background. Samples collected outside a Hanford burial ground indicated the burial ground contains irradiated fuel consistent with reactors operated on the Hanford Site. Although considerable information can be extracted from the stable isotope ratios, the method is less sensitive than the radioxenon measurements due to the presence of background atmospheric stable xenon.

He-3, the stable radioactive decay product of H-3, is highly mobile in the subsurface. Anomalous He-3:He-4 isotope ratios have been detected outside a Hanford burial ground at levels that suggest the presence of tritium target material within the disposal caissons.

INTRODUCTION

A number of U.S. Department of Energy sites face the challenge of environmental restoration following nuclear missions dating back to the 1940s or 1950s. Waste disposal activities during past operations often were poorly documented and did not meet current standards for safe disposal. Understanding the distribution of legacy waste in the subsurface is needed to make remedial decisions and to guide cleanup activities.

This paper describes a soil-gas sampling method for characterizing transuranic and tritium bearing waste. The method is based on isotopic analysis of the rare gases xenon and helium. Fission produces short-lived and stable xenon isotopes that are distinct from background atmospheric xenon; beta decay of tritium (H-3) produces He-3, altering the He-3:He-4 isotopic ratio in the soil gas.

Measuring isotopes of these rare gases is attractive because they are nearly inert, minimally interactive with the sediments, and are highly mobile in the subsurface. Thus, they can be detected at some distance from the source material. That distance will be a function of the release rate and of subsurface gas transport conditions. The gas sampling does not need to involve direct penetration of the waste or removal of highly radioactive materials, which simplifies radiological control measures. The isotopic measurements are sensitive and precise.

Three separate isotopic measurements will be discussed:

- **Short-lived radioactive xenon isotopes.** These isotopes are produced in situ through spontaneous fission. Several transuranic isotopes have significant spontaneous fission decay modes. Of these, Pu-240 is generally the most important in the waste of interest. The short half-lives mean that any initial reactor-fission products can be assumed to be decayed away in legacy waste. The short half-life also means that atmospheric background contribution generally can be considered insignificant.

- **Stable xenon isotopes.** Stable xenon isotopes are also produced through spontaneous fission. In addition stable xenon may be released from unprocessed spent fuel. We assume that plutonium production and other separation/purification processes will remove xenon from the product stream.

- **Stable helium isotopes.** H-3 decays to He-3 with a half-life of 12.32 years. Thus an excess of He-3 over the atmospheric abundance is indicative of H-3 decay. Tritiated moisture released into the vadose zone mixes with meteoritic moisture and migrates downward to mix with groundwater. Concurrent with release of H-3 to the vadose zone, He-3 begins to build up in the vadose zone and groundwater at the rate of H-3 decay. The He-3 then diffuses away from its vadose or groundwater source and migrates toward the surface. He-3 is a good surrogate tracer for H-3 in the vadose zone and groundwater because of its nonreactive behavior.

THEORY

Xenon Isotopes

Xenon is a high-yield fission product in nuclear reactors as well as being present in the atmosphere at approximately 87 ppb by volume. There are nine stable isotopes of xenon with mass range from 124 to 136 and six radioactive isotopes with half-lives greater than 1 hour. Of the radioactive isotopes, only Xe-133 with a half-life of 5.243 days and Xe-135 with a half-life of 9.10 hours are of interest here since they are the only radioisotopes with significant fission yields. Stable and radioactive isotopes of interest are summarized in Table I.

Table I. Natural Abundance and, Half-Life, and Thermal Neutron Fission Yield of Xenon Isotopes with Mass Between 124 and 136. From [1].

Isotope	Natural Abundance (atom %)	Half life (from ground state)	U-235 fission yield (%)	Pu-239 fission yield (%)
Xe-124	0.10		blocked	blocked
Xe-125		17.1 hr	blocked	blocked
Xe-126	0.09		blocked	blocked
Xe-127		36.4 d	blocked	blocked
Xe-128	1.91		blocked	blocked
Xe-129	26.4		blocked[*]	blocked[*]
Xe-130	4.1		blocked	blocked
Xe-131	22.2		2.89	3.86
Xe-132	26.9		4.31	5.41
Xe-133		5.243 d	6.70	7.02
Xe-134	10.4		7.87	7.68
Xe-135		9.10 hr	6.54	7.6
Xe-136	8.9		6.32	7.1
[*] Fission production of Xe-129 is effectively blocked by long-lived I-129.				

Thermal neutron fission in reactors will be a mixture of U-235 and Pu-239 fission, with the Pu-239 contribution increasing with higher burn-up in the reactor. The differences in fission yields are not large in the xenon isotopes of interest as shown in Table I.

Xenon is also a product of spontaneous fission. Even mass-number isotopes of plutonium, americium, curium and californium dominate spontaneous fission. Of these, Pu-240 is produced to the greatest extent in plutonium production and power reactors. Weapons-grade plutonium contains less than 7 weight % Pu-240, fuel-grade plutonium 7-19 weight % Pu-240, and power reactor-grade plutonium contains > 19 weight % Pu-240[2]. In practice, the Pu-240 content of weapons grade plutonium is ~ 6 atom %. For the rest of this discussion, percentages will refer to atom % unless specified otherwise.

A recent critical review of spontaneous fission half-lives gives a Pu-240 spontaneous fission half-life of 1.14E11 years [3]. The fission yield for xenon isotopes from spontaneous fission of Pu-240 is not readily obtained from literature tabulations. Pu-240 spontaneous fission yields are not included in the ENDF/B-VI data review[4]. It is a reasonable assumption that the spontaneous fission yield curve for Pu-240 will be similar to the thermal neutron fission yield for Pu-239 (see Table I). In this case, spontaneous fission of Pu-240 is analogous to neutron capture by Pu-239 followed by a fission event.

At secular equilibrium there will be 8.85e-15 atoms Xe-133 and 6.92e-16 atoms Xe-135 present per atom Pu-240 [5]. Xenon in soil gas at secular equilibrium with 100 nCi/g total plutonium (the lower limit defining transuranic waste) will be 15,000 mBq/standard cubic meter air for Xe-133 and 16,000 mBq/standard cubic meter air for Xe-135, assuming all the xenon is released to the gas phase as a bounding condition.

In this paper, stable xenon isotope ratios are reported using δ notation where:

$$\delta \text{Xe-134} = \left[\frac{\left(\dfrac{\text{Xe-134}}{\text{Xe-129}}\right)_{\text{sample}} - \left(\dfrac{\text{Xe-134}}{\text{Xe-129}}\right)_{\text{air}}}{\left(\dfrac{\text{Xe-134}}{\text{Xe-129}}\right)_{\text{air}}} \right] \qquad \text{(Eq. 1)}$$

The choice of Xe-129 as a reference isotope is somewhat unconventional compared to geochemical and cosmochemical studies but has an advantage here because the long half-life of I-129 effectively blocks the production of Xe-129, thus the Xe-129 present is from the atmospheric background.

Helium Isotopes

The decay of H-3 to He-3 with a half-life of 12.32 years produces local variation in the helium isotopic ratio. He-3 makes up only 0.000137 % of atmospheric helium with the bulk being He-4. Thus, even small amounts of He-3 production can perturb the He-3:He-4 ratio. By convention, helium isotope ratios are normalized to the atmospheric ratio (i.e. He-3:He-4 in air is defined as 1).

It should be noted that alpha decay has the potential to decrease the He-3:He-4 ratio through He-4 production. However, due to the larger He-4 background, commensurately greater He-4 production would be needed to perturb the isotopic ratio.

ANALYTICAL

Stable helium and xenon isotopes are analyzed by rare gas mass spectrometry. Helium isotope ratios were analyzed at the University of Rochester on a VG 5400 Rare Gas Mass Spectrometer fitted with a Faraday cup and a Johnson electron multiplier. Additional details on the analytical method are given in [6].

Stable xenon isotope ratios were analyzed at Lawrence Berkeley National Laboratory on an all-metal high resolution magnetic sector mass spectrometer. The samples were purified by passing over a series of cold traps and metal alloy getters to remove other air components. The residual rare gas fraction was sorbed onto activated carbon at 77 K and sequentially desorbed into the mass spectrometer [5].

Radioactive xenon isotopes were analyzed on an ARSA system. The ARSA was designed for continuous monitoring of radioactive xenon in the atmosphere for comprehensive test ban treaty verification. The short-lived radioxenon isotopes must be analyzed promptly (within approximately a few days to a week from production, depending on the isotope and the concentration at time of sample collection). The ARSA system uses a series of chillers and sorbants to separate and concentrate xenon from air [7]. The separation increases sensitivity by removing the major air components to reduce the volume and also by removing radioactive radon that can interfere with the counting. The removal of radon is important because radon background is about 10 mBq/m^3 in the atmosphere and certainly greater in soil-gas. A schematic

of the system is shown in Fig. 1. The system is designed for a flow rate of ~7 m^3/hr and an 8-hour sampling cycle. Make-up nitrogen containing no radioactive xenon is used after sample introduction for the duration of the cycle. Xenon isotopes are quantified in a gas cell scintillation counter using beta-gamma coincidence counting.
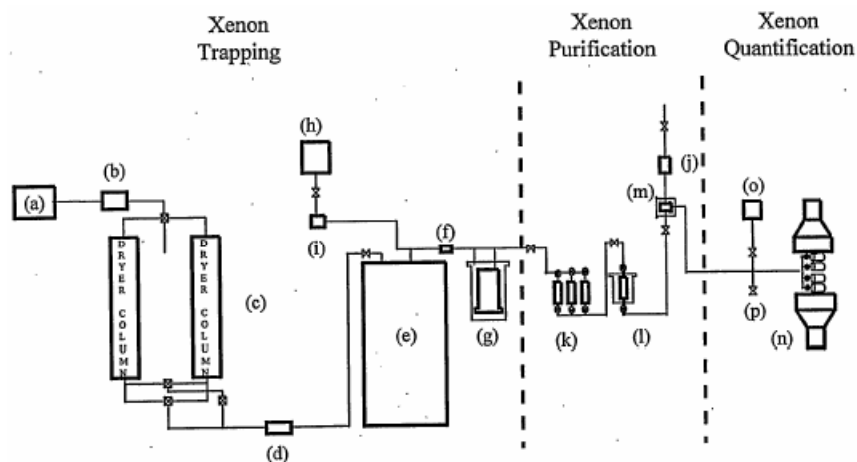

Fig. 1. Schematic illustration of Automated Radioxenon Sampling and Analysis (ARSA) system

For simplicity the components needed for 100% duty cycle are not shown. The system consists of (a) a piston compressor pump (80 to 100 psig, 7 m3/h), (b) heat exchanger, (c) dual air drying/CO2 removal columns filled with 13X molecular sieve and Al2O3, (d) mass-flow controller for process air, (e) cryogenic air-chiller with output temperature -125° C, (f) initial radon "pre-trap," (g) cooled "main" charcoal trap for xenon trapping, (h) nitrogen bottle or generator, (i-j) mass flow controllers for nitrogen flow, (k) ascarite-based CO2-removal traps, (l) cooled 5A-molecular sieve radon removal trap, (m) cooled, final small volume charcoal trap for xenon transfer into counting system, (n) NaI(Tl)-scintillating gas cell based beta-gamma coincidence spectrometer, (o) quadrupole residual gas analyzer, (p) path to archive bottles. From [8].

SAMPLING

Sampling for rare gas isotopes is similar to conventional active soil-gas sampling for volatile organic compounds. Temporary or permanent sampling points may be installed in the vadose zone, depending on project objectives. Sampling points may be installed using direct-push methods (e.g. cone-penetrometer, Geoprobe®) or by hand. Work thus far has used Geoprobe® points and existing open-bottom vadose well casing.

This paper will discuss two sample locations at the U.S. Department of Energy's Hanford Site in south-central Washington State. The 618-11 Burial Ground was originally investigated for helium isotopes to define the source and extent of a groundwater tritium plume [6]. Selected soil-gas points were then used for proof-of-principle sampling for stable xenon isotopes.

Additional xenon sampling was performed at the 216-Z-1A Tile Field liquid waste disposal facility using existing vadose zone wells within the tile field.

Stable Helium and Xenon Isotope Sampling at the 618-11 Burial Ground

Between 1962 and 1967, the 618-11 Burial Ground received low- to high-activity dry waste, fission products, plutonium, and other transuranic constituents in a variety of waste forms from research facilities. There is no clear record of tritium-bearing waste disposed to the facility, but the operation of the burial ground coincided with a lithium aluminate tritium production target development project on the Hanford Site. In 1999, nearly 2,000,000 pCi/L of tritium were detected in a water sample from a groundwater monitoring well immediately downgradient of the burial ground, and the concentration rose to over 8,000,000 pCi/L by 2000 when the soil-gas sampling took place.

At the 618-11 Burial Ground, sampling points for soil gas were installed using a truck mounted Geoprobe® Model 5400 rig and a 3.2 centimeter diameter hollow push-rod with a detachable steel tip. Target depth of installation of the screened interval was 6.1 meters (below ground surface (bgs), but actually ranged from 4.4 to 6.3 meters bgs. The sampling points consisted of a ~20-centimeter-long, fine-mesh, stainless steel screen connected to the surface with a 0.25 centimeter inside diameter polyethylene tube. Each sampling location was allowed to equilibrate for at least 24 hours and up to several weeks before soil-gas samples were collected.

The sampling vessels for helium and stable xenon isotopes were 50-milliliter stainless steel cylinders with fit with a high vacuum needle valve and a ~0.635 cm Swagelok™ fitting. Each vessel was evacuated to less than 5 torr before sampling. The sampling configurations used to collect He-3 samples can be seen in Fig. 1. A Kurz Instruments[TM] mass flow meter was placed in the flow stream between the polyethylene riser tube and the bottom of the rotometer. The flow was adjusted to 1 liter per minute. The soil-gas sampling point was allowed to purge for a minimum of 15 minutes. The sampling cylinder was pressurized, 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.

Seventy soil-gas monitoring points were installed at the periphery of the burial ground and in transects across the suspected groundwater tritium plume downgradient from the facility. Of these, three locations near the burial ground were sampled for xenon isotopic analysis.
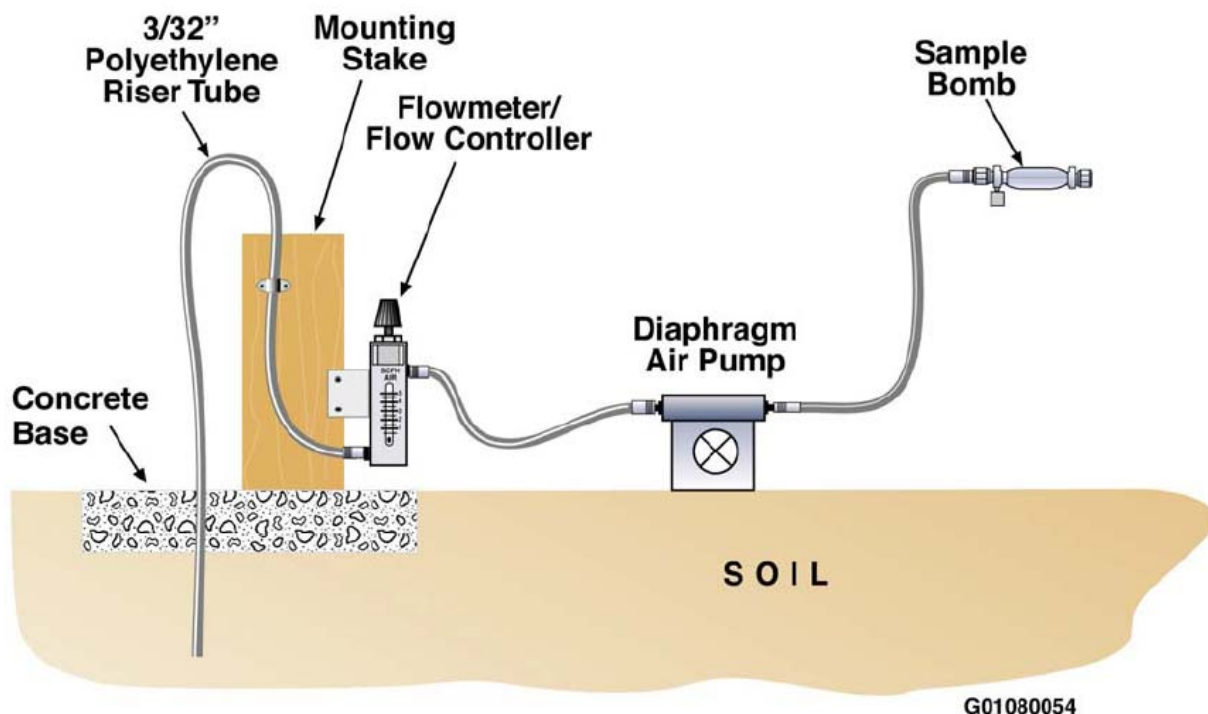
Fig. 2. Sampling system for stable helium and xenon isotopes

Radioactive and Stable Xenon Isotope Sampling at the 216-Z-1A Tile Field

Sampling for the radioactive xenon isotopes requires a large volume of gas. For this reason, the small-diameter sampling points described above were not suitable for radioactive xenon sampling. A high-pressure compressor was used to pressurize ~44-liter gas cylinders to 2,000 psi. A SCUBA type portable compressors was used. Sample coordination with the analysts was important due to the short half-lives of the isotopes of interest.

Radioactive xenon sample collection was performed at the 216-Z-1A Tile Field. This site received liquid waste containing ~57 kilograms of plutonium from the final stages of plutonium processing. Samples were collected from two existing open bottom, ~15-centimeter diameter boreholes in the tile field and composited for analysis. Stable xenon isotope samples were also collected from these two locations but were analyzed as two separate samples.

RESULTS

Results of Stable Helium Isotope Sampling at the 618-11 Burial Ground

Previous work demonstrated that helium isotope anomalies could be measured in soil gas above groundwater contaminated with H-3 at considerable distance from source areas [9]. The study at the 618-11 Burial Ground was designed to delineate the groundwater contamination. Fig. 3 shows the results of the helium sampling in the immediate vicinity of the burial ground. The

highest He-3:He-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 near the northeast corner of the burial ground. In order to determine the likely source of He-3 (vadose zone source or groundwater), a groundwater grab sample was collected at the location of the highest He-3:He-4 ratio. The groundwater grab sample from that location contained only 6,510 pCi/L of H-3 compared to approximately 8,000,000 pCi/L at well 699-13-3A. This result strongly suggested that the highest He-3 concentrations midway along the north side of the burial ground were from a vadose source of tritium located within the burial ground rather than from H-3-contaminated groundwater. A series of caissons located along the north side of the burial ground are a likely source. The second maximum may be from a vadose zone source of H-3 or a groundwater source. The points along the south and west sides of the burial ground approach the He-3:He-4 normalized atmospheric ratio of 1.0. Elevated ratios persist east of the burial ground for hundreds of meters and are attributed to the groundwater H-3 plume.
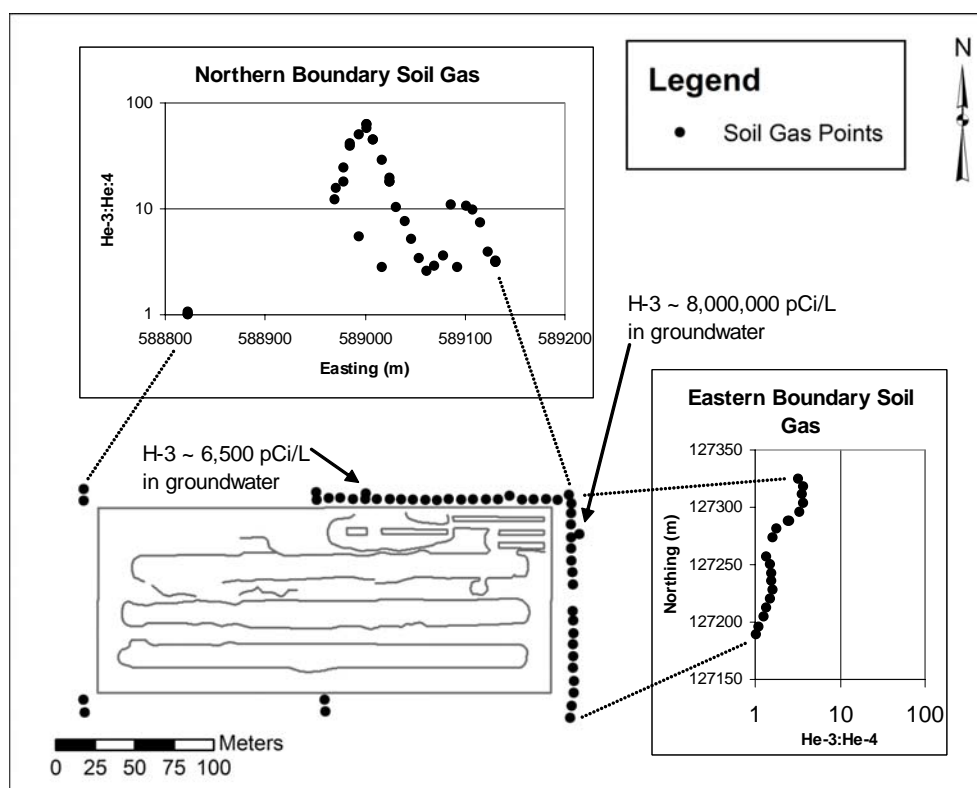

Fig. 3. He-3:He-4 ratio results from soil-gas samples from around 618-11 Burial Ground and immediately downgradient

Results of Radioactive Xenon Isotope Sampling at the 216-Z-1A Tile Field

The proof-of-principle sampling of radioactive xenon isotopes at the 216-Z-1A Tile Field successfully demonstrated the ability to detect and quantify the production of xenon from spontaneous fission in the subsurface. This location was chosen for the sampling because it

contained a large quantity of plutonium waste.   Carry-over of fission xenon from the initial reactor thermal neutron fission is highly unlikely given the multiple dissolution and purification processes prior to the plutonium finishing that generated this waste. The finishing operations that generated the waste discharged to the 216-Z-1A Tile Field were carried out in a different facility from the initial fuel dissolution and separations.

The composite sample contained 16,400 mBq of Xe-133 per standard cubic meter of air and 1,811 mBq of Xe-135 per standard cubic meter of air. The instrument detection limit was approximately 0.58 mBq/standard cubic meter of Xe-133 and 3 mBq/standard cubic meter of Xe-135. The concentrations above are not decay-corrected to the time of sampling because the analysis was carried out on the same day as sampling. Decay correction would be desirable when comparing multiple samples with varying holding times.

Results of Stable Xenon Isotope Sampling at the 618-11 Burial Ground and the 216-Z-1A Tile Field

Three of the five stable xenon isotope samples showed isotopic ratios altered from atmospheric abundance. Fig. 4 shows the δXe-136 vs. δXe-134 for the samples of interest compared to that of Pu-239 thermal neutron fission and U-235 thermal neutron fission. Atmospheric xenon plots at the origin of the figure. The two samples most enriched in Xe-134 and Xe-136 plot far above the fission ratios – i.e., with greater Xe-136 than would be produced by fission. This is interpreted as being due to production of excess Xe-136 by neutron capture on Xe-135. Xe-135 has a very large neutron capture cross section, 2.6e06 barns, and the large shift from fission xenon indicates production in the high neutron flux environment of a fission reactor. Also shown are the isotopic ratios of average Hanford irradiated fuel, from ORIGEN modeling [10]. The agreement between the samples and the model fuel strongly indicates xenon diffusion from irradiated fuel through the soil out of the burial ground. The samples closer to the origin show too great an atmospheric component to distinguish spontaneous fission from reactor exposure. As expected, the 216-Z-1A Tile Field sample is consistent with spontaneous fission, although the error bars are too large to rule out irradiated fuel without a priori process knowledge.
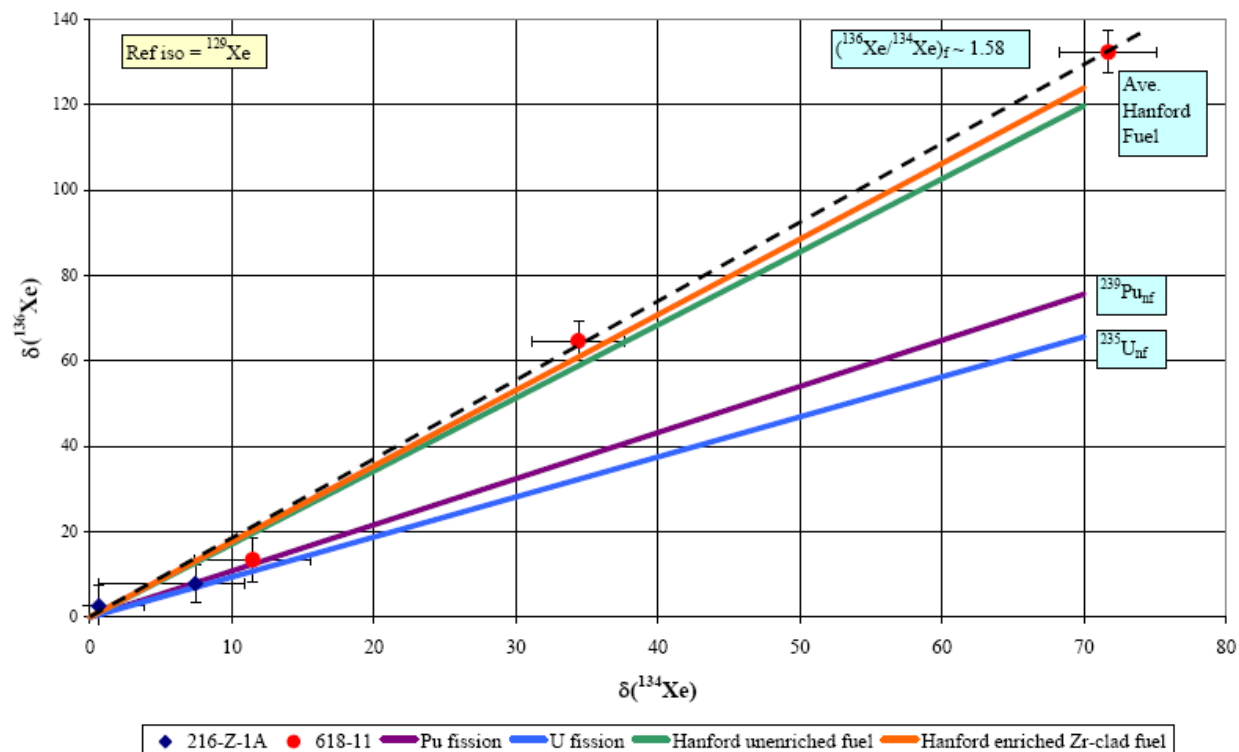
Fig. 4. δ-Xe-136 vs. δ-Xe-134 for soil-gas samples from the Hanford Site. The reference isotope is xenon-129.

DISCUSSION

Sampling for tritium and xenon isotopes can be accomplished using well established methods. Sample points may be either temporary or permanent. Minimally-invasive direct push methods such as Geoprobe® or cone penetrometer are particularly attractive because they return no potentially contaminated cuttings to the surface. The sampling for radioactive xenon isotopes requires large diameter sampling points due to the gas volume needed. Cone penetrometer rods up to 5-centimeter diameter are available – sufficient for sampling.

Analytical through-put is a major issue that may affect sample design. In particular, the radioactive xenon sampling must be coordinated with the analysis time due to the relatively short half-lives of the isotopes of interest. This suggests that the method would currently best be applied to targeted locations or random designs, rather than to gridded sample designs that would require large numbers of samples. Analytical through-put could be enhanced greatly through designing a system specifically for this application rather than using the modified ARSA system.

The helium isotopic method for locating H-3 contamination is well developed as demonstrated by the investigation at the 618-11 Burial Ground. Although that investigation did not interrogate the subsurface within the burial ground, the data from outside the fence line show distinct variations that are interpreted as relating to the proximity of sources.

Application of the xenon-isotopic techniques is more developmental but should be able to provide significant information on waste sources prior to more invasive sampling or excavation.

Container integrity is an issue since the gas must be released from the waste to the vadose zone. However, container breaches are common in buried transuranic waste. Preliminary modeling (not presented) indicates that the xenon released will diffuse outward and into the cap sufficiently rapidly to be successfully sampled. The samples integrate over greater areas that any direct solid waste or soil sampling, improving the ability to characterize heterogeneous wastes.

Radioactive xenon isotopic analyses have the advantage of extremely low background levels allowing for exceptional sensitivity through processing very large samples. However, the short half lives, particularly for Xe-135, and the low spontaneous fission rate means that the sensitivity is needed. It is expected that Xe-133 will be the most useful for overall characterization, with Xe-135 providing additional information near the highest contamination sources.

The stable xenon isotopes with a signature consistent with Hanford fuel at the 618-11 Burial Ground were unexpected and demonstrated the potential for that technique. The atmospheric background xenon, although small, limits the ability to distinguish contaminants to higher levels than the radioactive isotopes. The smaller sample size and longer holding times for stable xenon analyses are advantages. The data from the 216-Z-1A Tile Field illustrate the greater sensitivity of the radioactive isotope analyses. Thus, a combination of both techniques is probably warranted in many situations.

Further work is needed to quantify the relationship between the rare gas concentrations in soil gas to the amount of tritium or transuranic waste in the subsurface. The ability to quantify the concentration of plutonium or tritium in the waste from the soil-gas measurements is limited by the unknowns in the release rates, transport parameters, and relationship of sample volume to waste volume. However, similar issues arise in other soil-gas techniques such as measurements of volatile organic compounds. Although some estimates can be made by making assumptions about release rates and transport parameters, it is probably best to use the isotopic measurements for semi-quantitative mapping of areas of high interest until site-specific parameters are obtained. This does not negate the usefulness of the techniques, given that other alternatives are limited and costly.

CONCLUSION

Soil-gas sampling for helium and xenon isotopes provides a unique capability for in situ characterization of buried tritium and transuranic waste. The sampling method is minimally invasive and interrogates a larger volume than direct sampling or downhole geophysical logging.

Initial deployment of the technique has demonstrated the probable presence of tritium bearing waste and spent fuel at the 618-11 Burial Ground and shown the ability to detect the short-lived products of spontaneous fission at the 216-Z-1A Tile Field.

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