Distribution of Natural Radionuclides at the Nevada National Security Site

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Abstract

According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) publications, contributions of terrestrial gamma doses are mainly from the presence of ^{40}K, and of ^{238}U and ^{232}Th together with their progeny in various rocks and soils. A survey of soil distributions of radionuclides ^{40}K, ^{238}U, and ^{232}Th was performed at the Nevada National Security Site (NNSS) using in situ gamma-ray spectrometry with a High-Purity Germanium (HPGe) detector. The average activity concentrations of ^{40}K, ^{238}U, and ^{232}Th in natural soils at the NNSS are 867 Bq kg^{-1} (range from 150 ± 8 to 1297 ± 56 Bq kg^{-1}), 50 Bq kg^{-1} (range from 29 ± 3 to 74 ± 8 Bq kg^{-1}), and 56 Bq kg^{-1} (range from 11 ± 2 to 96 ± 10 Bq kg^{-1}) respectively. The concentration at each location is significantly associated with its geological lithology. The terrestrial gamma dose rates around the NNSS were estimated from 26 to 144 nSv h^{-1} with mean value of 93 nSv h^{-1}. Our results provide useful information about the natural background radiation and radiological effects of naturally occurring radionuclides at the NNSS.

Keywords: environmental assessment; naturally occurring radionuclides; soil; in situ gamma-ray-spectrometry

1. Introduction

Gamma radiation from the primordial radionuclides ^{40}K, the ^{238}U series (^{238}U to ^{206}Pb) and ^{232}Th series (^{232}Th to ^{209}Pb) is the main source of external background irradiation of the human body in the environment. Therefore, monitoring the distribution of natural radionuclides is an important component of human health protection (IAEA 2004; Al-Khawlany et al. 2018; Kasoga et al. 2015). The natural background radiation level can also be used as a reference for future remedial actions of environmental contamination (Medeiros and Yoshimura 2005; Khan et al. 2019). Furthermore, studies on radionuclide distribution can provide important information on geological formation, rock/soil type for environment management (IAEA 2003; Deblonde et al. 2020).

Gamma-ray spectrometry measurements can be performed to identify and quantify radionuclides in complex radiation fields. High-Purity Germanium (HPGe) detectors have resolution suitable for radionuclide specific analysis of soil surface measurements in the field (Reinhardt and Herrmann 2019). In situ gamma-ray spectrometry with HPGe detectors is widely used for monitoring of natural radionuclides and man-made radionuclides in the environment (Hassan et al. 2018; IAEA 1998, 2017). Compared to soil sampling, in situ gamma-ray spectrometry is a faster, more cost effective, and less destructive method suitable for surveying large areas.

The United States (US) Department of Energy (DOE), National Nuclear Security Administration Nevada Field Office (NNSA/NFO) operates the NNSS (formerly the Nevada Test Site) in south-central Nevada. Historical nuclear weapons testing and related activities have resulted in areas contaminated with man-made radionuclides. In 1981, the US-DOE began the Radionuclide Inventory and Distribution Program (RIDP) to assess the distribution of radionuclides in surface soil at the NNSS. The RIDP surveyed large portions of the NNSS focusing on areas around aboveground nuclear tests, underground tests where significant amounts of surface radioactivity exist, and near nuclear-rocket test facilities. Estimated inventories and distributions were reported for 14 man-made radionuclides. The results were published in a series of five reports between 1983 and 1989 (McArthur and Kordas 1983, 1985; McArthur and Mead 1987, 1988, 1989). After three years of further data analysis, the distribution of radionuclides in the topsoil of the NNSS was summarized in a RIDP report (McArthur 1991). None of these reports included data on distributions of natural radionuclides. In this work, we studied the activity distributions of the radionuclides ^{40}K, ^{238}U, and ^{232}Th on the NNSS using in situ gamma-ray spectrometry. This study provides the first basic data on natural radionuclides in the NNSS environment.

2. Materials and Methods

2.1. Survey Area

The NNSS is located in Nye County, Nevada. The site's southeast corner is about 105 km northwest of the major population center, Las Vegas, Nevada. The NNSS covers about 3,523 km^2 and is 46 to 56 km east to west and 64 to 88 km north to south. Elevations at the NNSS range from 0.9 to 2 km. The NNSS is characterized by desert valley and great basin mountain topography, with

climate, flora, and fauna typical of the southwest deserts. The northwestern part of the site contains tertiary volcanic and granitic rocks (Orkild 1983; Orkild et al. 1969; Slate et al. 1999). The Yucca Flat (east to northeast NNSS), Frenchman Flat (east to southeast NNSS), Jackass Flats (southwest NNSS) mostly consist of quaternary and tertiary alluvial sediments (Orkild 1983; Slate et al. 1999). The southeast corner of the NNSS consists primarily of young (Holocene and latest Pleistocene) alluvium from sedimentary rocks (Orkild 1983; Slate et al. 1999).

Measurements were made at 101 locations across the NNSS (Fig. 1). Those locations include areas with elevated radiation levels from historical nuclear weapons testing, current and past radioactive waste management sites (RWMS), and/or current operations involving radioactive materials or radiation-generating devices. Fifty-one measurements are in locations with negligible anthropogenic radioactivity.

2.2. In Situ *Gamma-Ray Spectrometry*

Measurements were performed using an HPGe detector (model: GX5520, Canberra) with preamplifier, shaping or spectroscopy amplifier, high-voltage bias supply, multichannel analyzer (MCA), laptop computer, and tripod for supporting the detector. Energy calibration was performed using standard gamma sources ^{241}Am (59.5 keV), ^{137}Cs (661.7 keV), and ^{60}Co (1173 and 1333 keV) (North American Scientific Inc., North Hollywood, CA). The detector bias voltage was 4,000 V and the energy resolution was 1.1 keV at 662 keV and 1.5 keV at 1333 keV. For the efficiency calibration and determination of radionuclides the ISOCS (In Situ Object Counting Software), LabSOCS (Laboratory Sourceless Calibration Software) and Genie 2000 v.3 software packages were used. For in situ gamma-ray spectrometry measurements, the

detector was positioned one meter above the ground and the spectra were acquired during a period of 1,800 seconds.

2.3. Soil Sampling

Grab samples of soil were collected from each of four locations in the southern portion of Yucca Flat. The topsoil from each point was sampled in vertical profile of four increments to a total sampling depth of 15 cm. Each sample was oven-dried, homogenized and sieved through a mesh with holes of diameter of 2.0 mm. The homogenized samples were weighed and placed in polyethylene bottles with diameter of 7.0 cm and height of 3.0 cm. Each soil sample containing approximately 125 to 150 grams was sealed for at least 30 days to allow ^{214}Pb and ^{214}Bi to reach equilibrium with ^{222}Rn (Myrick 1983). Each sample was counted with the HPGe detector for 28,800 seconds in a low background chamber shielded with 6-inch-thick steel.

2.4. Radionuclides Analysis and Dose Estimates

The gamma-ray spectra from in situ measurements were analyzed by ISOCS Genie 2000 software using circular plane geometry of 20 m in diameter assuming a homogeneous distribution of the natural radionuclides in topsoils. The activity concentration for ^{40}K was estimated using gamma line at 1460.8 keV (gamma yield of 10.7%). The activity concentrations of ^{238}U and ^{232}Th were calculated from their progeny photo-peaks. The activity concentration of ^{238}U was calculated from two gamma lines of ^{214}Pb (351.9 keV, gamma yield of 35.6%) and ^{214}Bi (609.3 keV, gamma yield of 45.5%). The activity concentration of ^{232}Th was calculated using two gamma lines of ^{212}Pb (383.6 keV, gamma yield of 43.6%) and ^{228}Ac (911.2 keV,

gamma yield of 25.8%). The minimum detectable activities for ^{40}K, ^{214}Bi, ^{214}Pb, ^{212}Pb and ^{228}Ac were 6, 3, 2, 3, and 3 Bq kg^{-1}, respectively.

The absorbed gamma dose rate (D, in nGy h^{-1}) in air at 1 m above the ground surface was estimated using the following equation from effective dose coefficients for adults summarized in UNSCEAR (2000):

$$D = 0.462A_U + 0.604A_{Th} + 0.0417A_K \quad (1)$$

where A_U, A_{Th}, and A_K are the activity concentration of ^{238}U, ^{232}Th, and ^{40}K in Bq kg^{-1}, respectively.

3. Results and Discussion

3.1. Activity distribution of ^{40}K, ^{238}U, and ^{232}Th and Lithology

Activity concentrations of the natural radionuclides were studied using in situ gamma-ray spectrometry measurements with an HPGe detector. Table 1 lists the activity concentrations of ^{40}K, ^{232}Th, and ^{238}U measured from the 101 NNSS locations.

The activity concentration of ^{40}K ranged from 150 ± 8 to 1297 ± 56 Bq kg^{-1} with an average of 867 Bq kg^{-1}. The activity concentration of ^{238}U ranged from 29 ± 3 to 74 ± 8 Bq kg^{-1} with an average of 50 Bq kg^{-1}. The activity concentration of ^{232}Th ranged from 11 ± 2 to 96 ± 10 Bq kg^{-1} with an average of 56 Bq kg^{-1}. The average activity concentration of each ^{40}K, ^{238}U, and ^{232}Th are higher than the world population-weighted average concentrations of 420, 33, and 45 Bq kg^{-1} (UNSCEAR 2000), respectively.

The distribution of ^{40}K, ^{238}U, and ^{232}Th in the Earth's crust is dependent upon lithology (Hassan et al. 2018; UNSCEAR 2008). As shown in Table 2, the NNSS regional average activity concentrations of ^{40}K, ^{238}U, and ^{232}Th were 1119, 57, and 80 Bq kg^{-1} in Pahute Mesa and Rainer

Mesa (northwest NNSS); 913, 47, and 56 Bq kg^{-1} in Jackass Flats (southwest NNSS); 772, 49, and 51 Bq kg^{-1} in Yucca Flat (northeast NNSS); 630, 44, and 37 Bq kg^{-1} in Frenchmen Flat (southeast NNSS), and 292, 41, and 18 Bq kg^{-1} in Mercury, respectively. The higher levels of activity concentrations of the primordial radionuclides were found in the Mesa areas which consist of tertiary volcanic and igneous rocks (Orkild 1983; Orkild et al. 1969; Slate et al. 1999). The lower concentrations of those natural radionuclides were detected in the areas of geological origin of quaternary basalt and tertiary alluvial sediments in Yucca Flat and Frenchman Flat. Tertiary volcanic and igneous rocks tend to be enriched in potassium, thorium, and uranium, compared to the basalt or alluvial sediments found in the easter portions of the NNSS. Our results agree with the previously published data (Khan et al. 2019; Hassan et al. 2018; Wang et al. 2005).

3.2. In Situ *Measurements vs Soil Sampling*

A homogeneous assumption is used to calculate the activity concentrations of the natural radionuclides from in situ gamma-ray spectrometry measurements. To demonstrate the reliability of the results obtained from in situ gamma-ray spectrometry, it is necessary to study the homogeneity of the activity concentration distributions of ^{40}K, ^{238}U, and ^{232}Th in topsoil. The soil samples were collected within 250 m in radius from four in situ measurement locations (all in the same soil type unit). Samples were collected at four depths (0.0 to 2.5, 2.5 to 5.0, 5.0 to 10.0, and 10.0 to 15.0 cm) at each location and analyzed in the laboratory. Although the radionuclide concentrations slightly varied in the samples from different locations, the vertical distribution of ^{40}K, ^{238}U, and ^{232}Th is obviously homogeneous in all soil samples studied (Table 3). The activity concentrations of ^{40}K, ^{238}U, and ^{232}Th determined by soil samples were compared to those from

the four in situ gamma measurement locations. Table 4 compares the results of the activity concentrations from in situ gamma measurements with those from soil samples. The average activity concentrations of ^{40}K, ^{238}U, and ^{232}Th measured by in situ spectrometry from those four locations are 1054, 53, and 61 Bq kg^{-1}, respectively. The average activity concentrations of ^{40}K, ^{238}U, and ^{232}Th from the soil samples are 1162, 41, and 59 Bq kg^{-1}, respectively. The ratio between the results from the in situ and the laboratory measurements were 0.91, 1.29, and 1.03 for ^{40}K, ^{238}U, and ^{232}Th, respectively (Table 4). For radionuclides ^{40}K and ^{232}Th, our results demonstrate a good agreement between the activity concentrations obtained from both methodologies (Table 4). The differences in the activity concentrations of ^{40}K and ^{232}Th between those two methodologies are less than 10%. The activity concentration of ^{238}U was determined from its daughters ^{214}Bi and ^{214}Pb. The higher levels of ^{238}U activity measured by in situ gamma-ray spectrometry method may be due to overestimate of radon progenies. Similar results were reported by Baeza et al. (2016), Dzaluk et al. (2018), and Hassan et al. (2018).

4. Conclusions

Activity concentrations of the natural radionuclides in soil were measured from more than one hundred locations across the NNSS using in situ gamma-ray spectrometry. The natural radionuclide distribution in the NNSS depends on the rock and soil types. The increased level of activity concentration is significantly associated with tertiary volcanic and igneous rocks. Homogeneous distribution of the radionuclides ^{40}K, ^{238}U, and ^{232}Th in the topsoil were observed from the results of all soil samples studied in this work. No significant difference was detected between the results obtained from the soil samples and in situ gamma-ray spectrometry measurements except for the activity concentrations of radionuclide ^{238}U. Our study

demonstrated that in situ gamma-ray spectrometry measurement with an HPGe detector is very useful to study and survey the gamma-emitting natural radionuclides for large areas at the ground level of the soil. Although the mean value of the absorbed gamma dose rate (93 nGy h^{-1}) all over the NNSS exceeded the world average of 60 nGy h^{-1}, the average absorbed gamma dose rate in Mercury (42 nGy h^{-1}) is significantly lower than the world average.

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List of Figure Captions:

Figure 1. NNSS map and *in-situ* gamma spectrometry measurement locations.