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Evaluation of naturally occurring radioactivity across the State of Kuwait using high-resolution gamma-ray spectrometry

A.D. Bajoga [a,b,d,*], N. Alazemi [a,c], H. Shams [a,c], P.H. Regan [a,d], D.A. Bradley [a,e]

[a] Department of Physics, University of Surrey, Guildford GU2 7XH, UK
[b] Department of Physics, Gombe State University, Gombe, Nigeria
[c] Environmental Radiation Protection Laboratory, Qadesiyah, PO Box 16087, Kuwait
[d] National Physical Laboratory, Teddington, Middlesex TW11 0LW, UK
[e] Department of Physics, University of Malaya, 50603 Kuala Lumpur, Malaysia

HIGHLIGHTS

- 90 independent soil samples obtained across State of Kuwait were analysed using high-resolution gamma-ray spectrometry.
- Mean activity concentrations determined for ^{232}Th, ^{238}U and ^{40}K.
- In current work no firm evidence established for presence of depleted uranium.
- Consistent with natural uranium, the mean isotopic activity ratio for ^{235}U/^{238}U across the samples was 0.045 ± 0.003.

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ABSTRACT

A study of natural radioactivity from 90 different soil samples from the state of Kuwait has been carried out to ascertain the NORM concentration values across the country. The calculated activity concentrations were determined from: (i) the decays of the ^{226}Ra, ^{214}Pb and ^{214}Bi members of the $4n+2$ decay chain headed by ^{238}U and; (ii) the ^{228}Ac, ^{212}Pb and ^{208}Tl members of the $4n$ chain headed by ^{232}Th. The study also included evaluations for the ^{235}U decay chain with the 186 keV doublet transition used together with the measured $4n+2$ activity concentration values to determine the ^{235}U/^{238}U isotopic ratios for each sample. The values for the arithmetic mean activity concentrations for 90 separate locations across Kuwait as determined in the current work were 17.2, 14.1, and 368 Bq/kg, with standard deviations of 5.2, 3.7 and 90 Bq/kg for the ^{238}U, ^{232}Th and ^{40}K activity concentrations respectively. Measured isotope ratios for ^{235}U/^{238}U give an arithmetic mean value for all of the samples of 0.045 ± 0.003, consistent with that expected for natural uranium. These results indicate no evidence for a radiologically significant dispersion of additional depleted uranium across the entire State of Kuwait from the 1991 Gulf War.

1. Introduction

Natural radioactivity arises from both cosmic and terrestrial sources. Radiation from naturally occurring materials which include the radioisotopes ^{238}U, ^{235}U and ^{232}Th and ^{40}K represent particularly important natural sources. These radionuclides are primordial and their very long radiological half-lives mean that they are ubiquitously distributed in all components of the wider surface environment including in foods, soil, air, water, vegetation, building materials etc. (Wilson, 1994). The radioactive decay series headed by ^{238}U, ^{235}U and ^{232}Th transmute into a range of radioactive progeny, finally decaying into a stable isotope of lead, namely ^{206}Pb, ^{207}Pb, and ^{208}Pb, respectively.

The earth's crust constitutes the major source of naturally occurring radioactive materials in the environment but the distribution of these radionuclides depends on petrological factors and the degree of mineralisation in the local geology. In a natural ecosystem, the distribution of ^{238}U, ^{235}U, ^{232}Th and their daughter products are governed by many factors such as chemical properties of the nuclides, physical features of the ecosystem, physiological and ecological attributes of the biota (Wallbrink et al., 1994; Al-Sulaiti, 2010; Boyle, 2013; Tesec-int.org). These factors determine the pathways along which some of these elements traverse a terrestrial ecosystem. For example, the weathering of bedrock releases U, Th and K to the surrounding soil, which then

* Corresponding author at: Department of Physics, University of Surrey, Guildford GU2 7XH, UK.
E-mail address: a.bajoga@surrey.ac.uk (A.D. Bajoga).

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subsequently allows transfer of the U and Th progenies within that medium. From the soil, potassium, radium, a small amount of uranium and a trace amount of thorium are taken up by plants, potentially finding their way into vegetation, groundwater and eventually into the food chain (Eisenbud and Gesell, 1997). The plants naturally utilise ^{40}K in the same manner as it uses the essential element of potassium. Radium is also taken up by plants since it is chemically similar to calcium, which is an essential element. In contrast to these, the uptake of U and Th by plants is usually small or negligible since these radionuclides normally are relatively insoluble (Dreicer et al., 1984). In general, the biological effect on the human body is through inhalation and ingestion and directly through external exposure.

The most significant component of total dose received by humans arises from environmental doses produced by the natural radiation. About 85% of the total terrestrial flux of gamma radiation is from the natural environment, from ^{238}U, ^{232}Th and ^{40}K sources, the majority of which is from radon exposure (UNSCEAR, 2000). Anthropogenic activities can also elevate the level of radiation dose received by the human population. In particular, the use of depleted uranium has been a source of concern by the government and other stakeholders. To our knowledge, no detailed studies were conducted to ascertain the levels of activity concentration across the State of Kuwait prior to the 1991 Gulf War. Following the war, however, various studies have been published (Saad and Al-Azmi, 2002; Bou-Rabee and Bem, 1996; Bou-Rabee, 1997; Al-Azmi et al., 1999; IAEA, 2003; Bem and Bou-rabe, 2004). The current work aims to give a comprehensive evaluation of the uranium isotope ratios and related activity concentrations across the State of Kuwait. The aim is to determine the nature and presence (if any) of depleted uranium in the environment due to Gulf War-related military activities, which have been reported to play a significant role in the radioactive contamination of the surface soil in Kuwait (Jallad, 2015). We have measured the activity concentrations and isotopic activity ratio for ^{238}U and ^{235}U as a function of the terrestrial distribution across Kuwait, using high-resolution low-background gamma-ray spectroscopy. A preliminary report on this work has been presented previously in (Bajoga et al., 2015).

1.1. Study area

Kuwait has a land mass area of 17,820 km^2 on longitude 46^0–48^0 and latitude 28^0–31^0 (UN2010, 2010). It is situated in the north-eastern part of the Arabian Peninsula and consists of a number of offshore islands such as the Bubiyan, Warbah and Failaka. It is a uniform flat-to-gently-undulating desert region. The surface soil is predominately sandy, poor in organic matter and low in water retention capacity, with oilfields and military activities occupying 7% and 4% of the total land areas, respectively (Omar et al., 2009). According to (Bou-Rabee, 1996), not much difference in natural radioactivity is expected from this soil type. The geological topography of Kuwait is relatively uniform with smooth sand-sheets having a somewhat consistent concentrations of radionuclides of natural origin (Saad and Al-Azmi, 2002; Jallad, 2013). For the current study, individual samples were collected during the period 2012–2013 from 90 separate locations across Kuwait and then subsequently transferred to the University of Surrey Environmental Radioactivity Laboratory for preparation and detailed gamma-ray spectrometric measurement.

1.2. Depleted uranium sources

Depleted Uranium (DU) is a by-product of uranium enrichment and has civil and military applications. Discriminating between natural and depleted uranium is essential for the proper management of nuclear fuel materials (Shoji et al., 2001). DU is a chemically toxic metal, potentially hazardous and in the course of nuclear forensics analysis it has been trafficked as an illicit radioactive material (Thompson et al., 2006).

The possibility of uranium contamination in 1991 Gulf War has been a source of concern due to the use of uranium-tipped anti-tank shells during the military campaign, coupled with uranium discharges from the burning oil wells. About 300 t of DU were dropped and fired in Kuwait and southern Iraq, covering an area of about 20,000 km^2 (U.S. AEPI, 1995a, 1995b), but only 10% of the projectiles hit their intended target while the remaining ones are still buried at various depths in the ground across the territory. The post-war estimates indicates that approximately 250 t of DU in the form of small metallic rods “penetrators” was deposited in surface soil (up to a few metres depth) due to inaccurate shots, in addition to ~10 t of additional DU aerosols released from the burning tanks (Bem and Bou-Rabee, 2004). In particular, the emission from these wells caused an unknown but rather small part of the estimated 6.7 GBq uranium activity (550 kg of pure uranium), to be uniformly deposited on Kuwait soil (Fisenne, 1993). The study by (Bem and Bou-Rabee 2004), reports that exposure of depleted uranium to humans is possible but not limited to the following ways: external exposure to the radiation emitted from the uranium isotopes, the inhalation of depleted uranium particles in the form of particulates, and/or ingestion of contaminated water or food which may adversely affect the function of the kidney, liver, lungs and heart (Bleise et al., 2003; Zavodska et al., 2008; Katz, 2014).

2. Materials and methods

2.1. Sample collection, preparations and measurements

The sampling plan was formed using a grid pattern, with a point-to-point width of 10 km (Fig. 1). If for some reason there is an obstacle (e.g. buildings, agricultural facilities) and a sample could not be collected at the exact grid position, then the nearest location with soil was taken as the sampling point and noted (IAEA, 2004). The removal and collection of the soil was done with a steel shovel and each sample was taken at a depth between 5 and 25 cm covering around 15 cm diameter on the surface, with each sample weighing 1.5–2 kg. Samples were then placed in plastic bags, numbered and the relevant information noted (eg. GPS location, date and time of sampling, sample number for id). After the sample collection was completed, the samples were taken to the Environmental Radioactivity Laboratory at the University of Surrey for further processing and preparation. The sampling grid was conducted using geographic information system software, Quantum Geographic Information System or QGIS [QGIS]. All samples were transferred to the Environmental Radioactivity Laboratory at University of Surrey where they were sieved to remove any unwanted impurities such as twigs, vegetation and glass, using a 2 mm sieve. The sieved samples were then dried in a drying oven to remove any excess moisture present in the soil followed by additional sieving using a 500 μm sieve to produce uniform and homogeneous samples of the soil. This grain size was chosen to optimise full volume filling of the Marinelli beakers. The prepared samples were then transferred into an active volume of a 550 ml Marinelli beaker, labelled and sealed air-tight with PVC tape to avoid escape of radon gas. These were then weighed and kept for at least 30 days (~7 half-lives) to ensure that the decays of ^{226}Ra and its ^{222}Rn daughter were in a state of radioactive secular equilibrium. All samples were measured using high-resolution gamma-ray spectrometry in a low-background set up, consisting of a co-axial and passively shielded HPGe detection

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