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Determination of depleted uranium using a high-resolution gamma-ray spectrometer and its applications in soil and sediments

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Abstract

A high-resolution gamma-ray spectrometer based on a hyper-pure germanium detector has been used to determine the amounts of depleted uranium in ground features subjected to military operations during the Gulf War of 1991 and in beach sediment samples collected from the northern side of the Arabian Gulf. The determination of $^{235}U/^{238}U$ was evaluated using spiked samples with a series of depleted uranium solutions. According to this method, the levels of depleted uranium were found to exceed 6.5% of the total natural uranium required to achieve reasonable levels for detection. Soil results indicated that the average of the total radioactivity of ^{238}U is 50.59 Bq/kg, with approximately 41.41% of this being represented by depleted uranium. For on-site and off-site individuals in an area of 10000 m^2, the RESRAD computer code was applied to calculate the annual radiological dose, which determined a level of 0.0031 mSv of total U; the code was also used to estimate the cancer risk, the level of which was determined to be 4.75×10^{-6} and 1.9×10^{-6} due to the total U and DU, respectively.

Keywords: Depleted uranium; Gamma spectrometer; Radiological dose; Risk; Sediment; Soil

1. Introduction

Depleted uranium (DU), a by-product of the uranium enrichment process, has been applied as armour-piercing ammunition in international military conflicts. DU is used in this way because of its high density, hardness, and pyrophoric properties [1,2]. The testing and use of such munitions has led to the release of DU into the environment at several locations around the world [3].

Natural uranium exists as three isotopes: ^{234}U, ^{235}U and ^{238}U. Natural uranium is considered to be one of the most important radioactive elements in the environment because it represents the major source of environmental natural radiation that all forms of life are exposed to. Natural uranium exists in soil, air, and water as well as in materials of natural origin. The most abundant isotope is ^{238}U (99.28%), which is known as the parent of a long radioactive decay chain that includes 16 radioactive elements.

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Table 1
Comparison between DU and natural uranium.

Parameter	DU	Natural U
Atom% ^{238}U ($t_{1/2} = 4.47 \times 10^9$ yr)	99.79	99.28
Atom% ^{235}U ($t_{1/2} = 7.04 \times 10^8$ yr)	0.20	0.72
Atom% ^{234}U ($t_{1/2} = 2.45 \times 10^5$ yr)	0.001	0.0054
^{235}U/^{238}U activity ratio	0.013	0.046
^{235}U/^{238}U atom ratio	0.002	0.0072
^{234}U/^{238}U activity ratio	0.18	0.8–1.2
^{234}U/^{238}U atom ratio	0.00001	0.000056

In nuclear fuel production, natural uranium is isotopically enriched and increases the concentration of the fissile isotope ^{235}U, usually to 3–5 atom%. The residue from this enrichment process is DU and a decreased proportion of both ^{234}U and ^{235}U, and hence, the proportion of ^{238}U increases, as illustrated in Table 1 [4,5].

Large amounts of DU are produced annually from the enrichment process of nuclear fuel. The disposal of such wastes has been a problem facing the countries that use these industries for many years. Currently, one of the solutions is introducing DU into industrial applications.

The issue of the possible presence of DU in the environment has recently attracted considerable public interest. Utilizing DU in non-fission nuclear weapons results in the addition of ^{238}U to the natural uranium in the environment. DU weapons are regarded as conventional weapons. The following amounts of ammunition containing DU were used in three recent conflicts: up to 321 tons in the Gulf War (2 August 1990–28 February 1991); approximately 3 tons in Bosnia–Herzegovina, 1995; and approximately 10 tons in Kosovo, 1999 [6–8].

DU is also involved in many civilian applications. DU is used as a cladding material in fast-breeder reactors, where its interactions with neutrons produce additional reactor fuel in the form of ^{239}Pu. Moreover, DU has been used as a fluorescent additive in dental porcelain crowns (recently discontinued) and is used as a shield for X-ray radiation in hospitals. Furthermore, DU is used as a counterweight for rudders and flaps in commercial aircraft and forklifts as well as in the keels of sailing yachts [9].

Monitoring and assessing radioactive materials in the environment are very important for protecting the general public against ionizing radiation due to the radiotoxicities and the chemo-toxicities of uranium, as well as to protect environmental resources, such as fresh water, agricultural soil, foods, and resort areas, against ionizing radiation. Monitoring and assessing radioactive materials are also important for studying the environmental factors affecting the mobility of such materials in different environmental pathways [10]. Studies of environmental radioactivity have been conducted to investigate the weathering effects on the redistribution of uranium in the environment [11]. Such studies are also very important to investigate the radiological minerals of black sands [12] and to evaluate the radiation exposure due to different sources of natural radiation, such as agricultural fertilizers [13].

Comprehensive programs of radiation measurements and assessments have been established in different countries to assess the DU and general background radiation in the environment. In addition, several reports and articles have been published involving the characterization of the nature of uranium and DU munitions.

Several techniques have been published that involve determining the amount of DU in the environment and describing procedures to identify the uranium isotopic composition in solid uranium compounds [14,15]. An inductively coupled plasma-mass spectrometer (ICP-MS) was used to determine the isotopic composition based on ashing and chemical decomposition with mineral concentrated acids [16]. In addition, thermal ionization mass spectrometry (TIMS) was used for the quantification of DU [17]. Additionally, an alpha spectrometer was used to measure ^{234}U/^{238}U ratio in the soil of a UK testing firing range [3].

Low-level high-resolution gamma-ray spectroscopy is a very convenient technique for determining the activity of uranium in environmental samples in which the activity of ^{238}U exceeds 1 Bq/kg [18]. Determining the amount of DU using this method in soil is based on assuming an equilibrium of ^{238}U up to ^{226}Ra [19]. In addition, processing of uranium ore started approximately 200 years ago; therefore, ^{238}U daughters (except ^{234}Th and ^{234m}Pa) do not exist in DU [20]. However, such conditions of equilibrium with ^{226}Ra might no longer be sustainable due to the natural processes and technological activities in the environment, particularly in the surface soil. Additionally, the equilibrium of ^{226}Ra is disturbed due to the leaching of radium or uranium from the soil by ground water.

This work aims to develop an experimental method for evaluating the gamma-ray spectrometry results to quantify DU by measuring two isotopes ^{238}U and ^{235}U as well as to investigate the DU that is present in the sediment and in soil samples collected from the northern area of the Arabian/Persian Gulf.

2. Experimental work

The measurements were performed using an extended energy range (24 keV–3 MeV), reverse electrode,