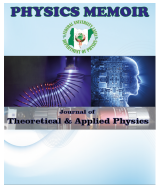

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Review

A Review on Environmental Radioactivity Measurement and its Impact on Human Health

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Abstract

Natural radioactivity in drinking water sources, no doubt poses serious contamination hazards. Assessment of its level in drinking water therefore provides useful information on potential unwanted radiation exposure to humans. To assess the effects of natural radionuclides in the environment, analysis is carried out on samples from the environmental media such as water, soil and species using different radiation detection techniques. This article reviews some of such studies with a view of gathering vital information on the recently used techniques, discusses current situations and give highlights of recent findings.

Keywords: Natural Radioactivity, Radiation Exposure, Radiation Dose, Radon Gas.

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1. Introduction

The origin of radionuclides in domestic water sources can be traceable to the trace deposits of naturally occurring or artificial radioactive material that inevitably exist within the environment. Their presence in form of Naturally Occurring Radioactive Materials (NORM), that is sometimes technically enhanced, resulting to what is generally termed as Technically Enhanced NORM (TENORM), occur mainly in domestic water sources due to contamination as a result of leaching of minerals in the earth crust while the artificial radioactive sources enter the aqueous media mainly through waste disposal practices, spills, and land application of chemicals. These contaminants vary in concentrations in water sources heavily depending on hydrogeological conditions as well as human activities.

Over the years, the behaviour of radionuclides in water, soils and sediments, has been the subject of considerable scientific interest and numerous investigations were carried out as a result. These investigations provide some basic

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understanding of radionuclide distribution and dynamics in lakes and rivers, as well as in their respective catchments, in different hydrogeologic systems and geographic regions. A number of researches conducted confirms that existence and mobility of radionuclides through the surface and the ground water systems is dependent on the physical and chemical properties of the contaminant, and on the rock and sediment characteristics [1-3]. Radionuclides that commonly occur in the aqueous phase were found to be very mobile within the aquatic environment thereby distributing their concentrations. In some cases, radionuclides strongly interact with the particulate matter suspended in water and the bottom sediments and consequently transported via flowing water [4].

The major percentage of drinking water sources comes from the abundant surface water and groundwater resources. Radionuclides of natural origins usually find their way into surface water as a result of leaching of the earth crust. The earth crust contains small amounts of uranium, thorium and radium as well as radioactive isotopes of uranium. Similarly, quite a number of radionuclides find their way into the drinking water sources due to human activities of agriculture, medicine and industry [5]. Uranium is one of the most abundant radionuclides in the surface of the earth, and water constitutes the principal route by which uranium is incorporated into humans, due to its commonly high solubility. In general, a wide range of radionuclides are known to occur in water, these include cesium-137, chromium-51, cobalt-60, iodine-131, iron-59, lead-210, phosphorous-32, plutonium-238, radium-226 radon-222, ruthenium-106, scandium-46, strontium-90, thorium-232, tritium uranium-238, zinc-65, zirconium-96 [5].

2. Human Health Hazard due to Radionuclides Ingestion through Drinking Water

Naturally occurring radionuclides of terrestrial origin (also called primordial radionuclides) exist in different compositions in all environmental media. Radionuclides from the natural decay series of uranium, thorium and potassium (^{238}U, ^{232}Th and ^{40}K), whose half-lives are comparable to the age of the earth, exist in significant quantities virtually everywhere within the earth crust [6]. Their natural occurrence in varying abundances is known to be responsible for the largest radiation exposure to humans [7]. NORM's hazard usually comes as a result of ingestion and inhalation of natural radioactive materials and their decay products found the soil and subsequently in air and water. Various countries proffered recommendations on the protection of the public against radiation exposure above maximum contaminants levels (MCL).

The World Health Organization (WHO) and International Commission on Radiological Protection (ICRP) proposed recommendations on the limitations of domestic exposure above the natural background [8-9]. These recommendations were adopted by many countries through their radiological protection agencies. The United States Environmental Protection Agency, USEPA, the United Kingdom National Radiological Protection Board (NRPB) are among such bodies. The EPA has established drinking water standards for several types of radioactive contaminants; combined radium 226/228 (5 pCi/L); beta emitters (4 mrems); gross alpha standard (15 pCi/L); and uranium (30 μg/L). The essence of all these regulations is to achieve an MCL goal of zero, with the basis that continuous accumulation of NORM in the body leads to ionizing radiation exposure which increases the risk of carcinogenesis.

The world annual natural radioactivity level, including external exposure, consumption of food and water containing natural radionuclides, and inhalation of radon with its daughter products, amounts on average to 2.4 mSv, with a range from 1 to 10 mSv [7]. The United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR) report also indicated that the average worldwide exposure to natural sources in foods and drinking water is 0.29 $mS\,vy^{-1}$ (about 0.17 $mS\,vy^{-1}$ from ^{40}K and about 0.12 $mS\,vy^{-1}$ from Uranium and Thorium) [7]. For prolonged exposure situations in case of all environmental radioactive sources, including natural and human-made sources, the International Commission on Radiological Protection (ICRP) recently recommended a generic intervention level of annual dose of 10 mSv as the level below which intervention is unjustified, taking into account radiological, economic and social factors [10]. The world health organization in its guidelines for drinking water quality, adopted the International commission on radiological protections' recommendation of reference dose level (RDL) of the committed effective dose, to be equal to 0.1mSv from 1 year's consumption of drinking-water (from the possible total radioactive contamination of the annual drinking-water consumption).

^{222}Rn is an innert gas, emanated through the decay of ^{226}Ra, both of which are members of the natural decay series. It has a half-life of 3.28 days. Its progeny forms a series of short-lived nuclides that decay in a matter of hours to a relatively long lived nuclide of ^{210}Po (half-life 22 years). This short half-lives gives ^{222}Rn progenies the ability to attain rapid radioactive equilibrium with ^{222}Rn. ^{222}Rn is present virtually everywhere on earth, but particularly in

the air over land and in buildings. Underground rock containing natural uranium continuously releases radon. ^{222}Rn is readily released from surface water and groundwater normally at much higher concentrations [11].

Radon gas also dissolve and accumulate in water from underground natural radioactive sources. When water containing radon is used for domestic purposes, radon gas escapes from the water into the air, hence this cause an increase in the airborne radon level of the area. The effect of elevated content of radon and its daughters in the atmosphere has been found as increased berration rates in the chromosomes in the peripheral blood of the investigated persons living in houses with increased levels of indoor radon concentrations [12-13]. The observed primodal nuclides in the study area, are known to be the parents of the natural decay series, resulting in a chain of radionuclides.

^{222}Rn is a daughter of special interest from this decay chain. This is for the fact that it is responsible for a large percentage of natural radiation exposure. The potential health hazards associated with ingestion of radionuclides through drinking water respectively have been well established, with many countries adopting the guidelines of activity concentration recommendations of the World Health Organization [8, 14-16]. Radon is extremely volatile and is readily released from water it can also dissolve and accumulate in water until aerated. Radon (^{222}Rn) and its daughter products from natural background has been a source of concern to the public [12-13, 17]. The highest organs dose due to ingested radon goes to the stomach, which receives > 90% of the total effective dose [13].

3. Estimation of Committed Effective Dose

Effective dose is an important concept used as a tool to enable the radiation doses from different radionuclides to be estimated. It is based on the risks of radiation induced health effects and the use of the International Commission on Radiological Protection (ICRP) metabolic model that provides relevant conversion factors to calculate effective doses from the total activity concentrations of radionuclides measured. The dose arising from the intake of 1 Bq (by ingestion) of radioisotope in a particular chemical form can be estimated using a dose conversion factor (DCF). Data for age-related dose conversion factors for ingestion of radionuclides has been published by the International Commission on Radiological Protection (ICRP 1996) [18]. Estimates of the radiation induced health effects associated with intake of radionuclides in the body are proportional to the total dose delivered by the radionuclides while resident in the various organs. Radiation doses ingested are obtained by measuring radionuclide activities in environmental samples (Bq l^{-1}) and multiplying these by the volume of water consumed over a period of time (litre y^{-1}). A dose conversion factor (Sv Bq^{-1}) can then be applied to give an estimate of ingestion dose.

4. Methods of Assessment of Environmental Radioactivity

Different techniques were employed by different studies to assess the level of radionuclides in drinking water. To analyze drinking-water for gross alpha and gross beta activities, the most common approach (ISO standard procedure) is to evaporate a known volume of the sample to dryness and measure the activity of the residue using a Gas Flow Proportional Counter. As alpha radiation is easily absorbed within a thin layer of solid material, the reliability and sensitivity of the method for alpha determination may be reduced in samples with a high total dissolved solid (TDS) content. Counting efficiency of alpha particles by this method is typically moderate and declines with increase in TDS. In a survey conducted to evaluate the gross alpha and gross alpha activity concentrations in the ground water from Katsina, northern Nigeria, Muhammad *et al.*, [19] used a gas flow proportional counter and relatively high efficiency values were reported. Similarly, in the measurement of gross alpha and beta radioactivity concentration in water, soil and sediment of the Bendimahi River and Van Lake in Turkey, Zorer *et al.*, [20] used gas-flow proportional counter and good detection limits were reported. Palomo *et al.*, [21] also used alpha/beta counter of low background multiple detector type to measure the radioactivity in bottled drinking water in Spain using the thin deposit method and good sensitivities were attained.

An alternative method is the use of Liquid Scintillation Counter. The inherent counting efficiency for alpha particles by this method is relatively higher. Since it does not suffer from self absorption problems, this method can equally handle higher levels of dissolved solids. However, the sample size is limited by what can be put into a Liquid Scintillation Counter vial, and so for large samples, some form of sample concentration must be performed. Kleinschmidt [22] reported an achievable gross alpha and gross beta minimum detection limits of 0.05 and 0.08 Bq l^{-1} respectively, in measurement of gross alpha and beta activity analysis of drinking water in Australia using liquid

scintillation assembly. A study by Rusconi *et al.*, [23], also indicated that in emergency situations, a gross activity screening can be carried out without any sample treatment by a single and quick liquid scintillation counting. In this method alpha and beta activities can be measured promptly in many samples with sensitivities of a few Bq/L. Varlam *et al.*, [24] similarly applied direct liquid scintillation counting measure low level environmental tritium reporting a very low detection limit of below 50 Bq l^{-1}.

An all encompassing technique that involves quantitative as well as qualitative radio elemental analysis is the use of gamma spectroscopic methods in measurement of both natural and artificial radionuclides in all environmental samples. This method determines the energy and count rate of gamma rays emitted by radioactive substances. In this method, a detailed analysis of the gamma ray energy spectrum is used to determine the identity and quantity of gamma emitters present in an environmental sample. The equipment uses a gamma detector, normally a thallium doped sodium iodide scintillation counter NaI(Tl) or a high purity germanium detector HPGe, a pulse sorter (multi channel analyzer), and associated amplifiers and data readout devices usually a computer system. A comparison of these detectors was carried out in terms of minimum detectable activity (MDA), efficiency and resolution by Perez-Andujar *et al.*, [25]. The study confirmed that the HPGe detector has better energy resolution while the NaI(Tl) has higher efficiency and can detect lower minimum detectable activity (MDA). In terms of efficiency and MDA, the study favoured the use of NaI(Tl) for low level radioactivity measurements due to low energy gamma ray emission ranging from 35 keV to 2 MeV, but highlighted the superiority of HPGe in terms of radionuclide identification due to its unmatched resolution.

In a study to assess the natural radioactivity in Guarani aquifer groundwater in Brazil, by Bonotto *et al.*, [26], a 2" × 2" NaI(Tl) well-type scintillation detector was employed to measure the activity concentration of ground water samples collected from the area. Tsabaris *et al.*, [27] also monitored the effect of rainfall on the natural and artificial radioactivity in the Aegean Sea using NaI(Tl) underwater floating measuring systems. The study observed some variations with the continuous monitoring by acquisition with a NaI-system at open sea. NaI(Tl), was also employed in measurement of environmental samples of water, soil and coal in a study by Muminov *et al.*, [28].

The study gave an alternative method of activity concentration determination based on decomposition of samples gamma ray spectra into spectral components. The study showed that scintillation detectors can be comparable to large-volume HPGe detectors in the effectiveness of registration and can be successfully used for the studies on environmental samples with low levels of radioactivity. In a study to determine activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K and gamma radiation absorbed dose rate levels in farm soil for the production of different brands of cigarette tobacco smoked in Nigeria, Jabiri *et al.*, [29] uses a lead-shielded 76 mm × 76 mm NaI(TI) detector with a resolution of about 8% at energy of 0.662 MeV (^{137}Cs).

This detector was considered adequate to distinguish the gamma ray energies of interest as reported by the study. Al-Azmi *et al.*, [30] uses small quantities of activated charcoal and a 7.6 cm × 7.6 cm NaI(Tl) well-type detector to investigate the indoor radon level in Kuwait dwellings. The use of the wide energy window proved to allow more efficient collection of counts from the radon progeny which further increases with increasing counting time. Xinwei *et al.*, [31] in a study to determine the natural radioactivity in some common building materials and by-products of coal-fired power plants collected from Baoji, West China, also uses gamma ray spectrometry with an NaI(Tl) detector. In another study by same authors, natural radioactivity measurements in rock samples of Cuihua Mountain National Geological Park, China was performed using NaI(Tl) detector. Similarly, in a survey to determine terrestrial gamma radiation dose rates, the concentration level of U-238 and Th-232 and alpha and beta activities for the surface soil in Ulu Tiram, Malaysia [32], measurements were performed using a NaI(TI) gamma-ray detector with crystal size of 1" × 1". The study indicated the suitability of the small size crystal (1" × 1") for environmental gamma radiation measurements.

High purity germanium semiconductor detector, HPGe, on the other hand was employed in a wide range of applications and studies to determine the radioactivity in environmental samples for its higher resolution option. To measure the radioactivity concentrations in soil and vegetables from the northern Jordan rift valley, Ababneh *et al.*, [33] used a high-resolution high-purity n-type germanium detector having a relative efficiency of 25% and a resolution of 1.9 keV at 1.33 MeV. In another study by Ababneh *et al.*, [34], a coaxial HPGe (High Purity Germanium) detector with an energy resolution of 1.9 keV and a relative efficiency of 25% at 1.33 MeV was used to assess the gamma-emitting radionuclides concentration in sediment cores from the Gulf of Aqaba, Red Sea. Ajayi *et al.*, [14] also in a study to measure the radioactivity in drilled and dug well drinking water of Ogun state southwestern Nigeria, employed the use of a co-axial type, high-purity germanium detector with about 50% relative counting efficiency and

energy resolution of 2.4 keV at 1.33 MeV. In a study to measure the natural radioactivity in bottled drinking water in Pakistan, Fatima *et al.*, [15] used a 4k series-85 Canberra multi-channel analyser model 8503 coupled with a Eurisys Measure coaxial 245 cm^3 high-purity germanium (HPGe) detector. The above are but a few instances that involves gamma detection techniques in the determination of environmental radioactivity level.

Neutron activation analysis (NAA), is another widely acceptable technique in qualitative as well as quantitative multi-elemental analysis of environmental samples. In this method, environmental samples are irradiated with neutrons to activate them. During irradiation, the naturally occurring stable isotopes of most elements are transformed into radioactive isotopes through neutron capture. The activated nucleus then decays according to a characteristic half-life; some nuclides emit β particles only, but most nuclides at the same time emits gamma, with specific energies. The quantity of radioactive nuclides is determined by measuring the intensity of the characteristic gamma-ray lines in the spectra. This technique, offers unmatched sensitivities in terms of accuracy and reliability that are superior to those attainable by other methods, in the order of parts per million or better. Neutron Activation Analysis (NAA) provides a highly efficient quantitative and qualitative technique for the precise determination of trace elements in different types of samples. The instrumentation used to measure gamma rays from irradiated samples in NAA also consists of a semiconductor detector, associated electronics, and a computer-based, multi-channel analyzer. Most NAA laboratories use hyper pure or intrinsic germanium (HPGe) coaxial detectors which operate at liquid nitrogen temperatures (77K) to measure gamma-rays with energies over the range from about 60 keV to 3.0 MeV, thereby analyzing multiple energy spectra.

Abdullah *et al.*, [35] determined the arsenic concentrations in water, soil and arum (vegetables) samples from Bagerhat, Bangladesh using the Neutron Activation Analysis method. The study observed that almost all the water samples were contaminated by a hazardous level of arsenic that exceeding the World Health Organization recommended value of 0.05 mg/L for Bangladesh. In another study to investigate the influence of cancer on selected trace elements among Sudanese patients with confirmed breast cancer, [36] use of instrumental neutron activation analysis was employed to estimate contents of Se, Zn, Fe, Cr, Rb, Cs, Co and Sc in some subjects. The result showed that the presence of Se, Zn and Cr elements from the malignant tissues are significantly higher when compared to the normal tissue. Neutron activation analysis (NAA) was also used in a study to quantify chromium and 15 minor and trace elements from chromite rock samples collected from El-Robshi area in the Eastern Desert, Egypt [37]. Na, Ga, As, La, Sc, Cr, Fe, Co, Zn, Zr, Ce, Ce, Yb, Lu, Hf and Ta were determined in varying concentrations.

5. Conclusion

This article provided an overview on the level of natural radioactivity contamination of drinking water sources, its detection methods as well as the human health hazard indices associated with it. Studies on radioactivity in water were in most cases centered around the levels of naturally occurring radioactive materials of terrestrial origin. The studies indicated a wide variation of activity concentration levels in different environmental media from different parts of the world. The reported Human health radiological indices in many studies predicted high radiation exposure to human populations in some areas having high activity concentration values hence high dose rates. The effects due to natural radioactivity, many studies reveals, is dependent on the geologic formation of an area, which in some cases greatly enhanced by human practices.

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