ACTIVITY LEVEL OF ^{214}BI AND ^{228}AC IN LOCALLY AVAILABLE FERTILIZER USING HPGE DETECTING SYSTEM

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Abstract: Fertilizers are mainly derived from phosphate rock, natural gas as raw material and others chemicals. These materials contain few amounts of radionuclides. The aim of this study is to investigate the activity concentration of ^{214}Bi & ^{228}Ac in fertilizer. High purity Germanium detecting system was employed to determine the activity level of ^{214}Bi & ^{228}Ac in the fertilizer sample. The mean activity concentration of ^{214}Bi is found to be 142.4±6.3 Bq.kg^{-1} ranges from 11.2±3.8 Bq.kg^{-1} to 921.4±34.3 Bq.kg^{-1} and that of ^{228}Ac is found to be 61.3±7.7 Bq.kg^{-1} ranges from 31.3±10.1Bq.kg^{-1} to 397.7±17.5 Bq.kg^{-1}. This result indicates that the activity concentrations of the radionuclides in the fertilizers of Gazipur in Bangladesh are within the range of values reported in other countries.

Keywords: HPGe Detector, Radionuclides, Fertilizer, Activity Concentration, Efficiency curve

1. Introduction

Natural radioactivity originates from extra terrestrial sources as well as from primordial radioactive elements in the earth crust. Most widely spread natural radionuclides are from the family of Uranium (^{238}U), Thorium (^{232}Th), Actinium (^{235}Ac) and Kalium [1]. ^{214}Bi and ^{228}Ac are the member of Uranium (^{238}U), Thorium (^{232}Th) respectively.

There are many human activities that lead to the production of radioactivity. Mining processing and massive use of fertilizer materials caused redistribution of natural radionuclides throughout the environment, particularly uranium, radium, and radium decay products. While typical rock contains an average of 2.8ppm of uranium, a phosphate rock contains up to 300 ppm uranium [2]. Fertilizer generally produced from natural gas as raw materials, phosphate rock and other chemicals. It is reported that approximately 80% of Radium-226 follow the gypsum, whereas 86% of the uranium and 70% of the thorium are found in phosphoric acid [3].

Earth crust contains radioactive uranium, radium and their products. So while collecting the natural gas from the earth crust, it may contain those radioactive elements. Phosphate materials used for production of phosphate fertilizers contain a minor quantity of radioactive material, mainly various members of the uranium and thorium series, and radio potassium [4].

Phosphoric acid is the starting material for TSP and ammonium phosphate fertilizers. Natural uranium can substitute for calcium in the phosphate rock structure and over a period of time, accumulates in phosphate reserves. Thus uranium is present in fertilizers manufactured from phosphate rock. In NPK fertilizers, potassium component augments the natural radioactivity because of the presence of radioactive ^{40}K, whose natural abundance in potassium is 0.00118% [3].

The presence of radionuclide in fertilizer can result in elevated radionuclide level in soil and environment [5]. The radionuclides present in

the fertilizers at elevated concentrations are persistent in the environment, and the ongoing addition of fertilizers containing these radionuclides to soil can result in their accumulation in the soil over time.

The natural radionuclides of concern are mainly Uranium, Thorium and Potassium, and the radionuclides that are created as their radioactive decay chain. Emanation of Radon gas (e.g. ^{222}Rn and ^{220}Rn of life time 3.8 d and 55.6 s respectively) into air occurs as a product of uranium ^{238}U and thorium ^{232}Th decay chain respectively. The short lived decay products of radon are responsible for most of the hazard by inhalation. The hazard of Radon comes from its radioactive progeny, which use their physical properties to spread or attach like aerosols do, trapped in the lung and depositing their alpha-particle energies in the tissue, producing higher ionization density than beta particles or gamma-ray. Lung cancer, skin cancer and kidney diseases are the health effects attributed to inhalation of Radon decay products [6].

Assessment of any release of radioactivity to the environment is important for the public health; especially if the released radioactivity is a matter of population exposure and can enter into the food chain [7]. The aim of this study is to measure the activity concentration ^{214}Bi and ^{228}Ac (which are the daughter of ^{238}U and ^{232}Th respectively) in fertilizers using HPGe Digital Gamma-Ray Spectrometry System.

2. Experiments
2.1. Sample Collection and Preparation
Thirteen fertilizer samples were collected from different shops and one was produced locally of Gazipur district in Bangladesh, serially named as Urea, MOP, SSP, Feovit Mixture (NPKS), Raina Mixture(NPKS), TSP, Green Mag, Bio fertilizer, Cow dung, Bone dust, Agri Chelated, DAP, Gypsum. All the samples were dried in an oven at a temperature of about 85˚C until they attained constant weight. Each dried fertilizer samples were grounded in an agate mortar separately to make the samples powder and ensure homogeneity. Thus, homogenized sample was transferred to

sealable cylindrical plastic container of 7 cm height and 5 cm in diameter, marked individually with identification parameters. After packing, the weight of each samples were taken with microbalance. All the sample containers was sealed tightly with cap and wrapped with Teflon and thick vinyl tapes around their screw necks. Then the prepared samples were stored for minimum four weeks prior to counting, allowing establishment of secular equilibrium between the long lived ^{238}U, ^{232}Th and their decay products.

2.2. Parameters of Gamma Spectrometry System
The performance of the detector depends on the high resolving power and the higher value of efficiency. The most important parameters of a detector are energy calibration, energy resolution and efficiency calibration.

2.2.1. Energy Calibration of the detecting System
Energy calibration is the relation between the channel number in the spectrum and the corresponding gamma ray energy. In the present experiment ^{133}Ba, ^{57}Co, ^{60}Co, ^{137}Cs, ^{22}Na, ^{54}Na, ^{152}Eu sources have been used for energy calibration. A graph has been plotted with energy along X-axis and number of channel along Y-axis, which is shown in Fig-1. Normally the gamma energy follows a linear relationship with the channel number.
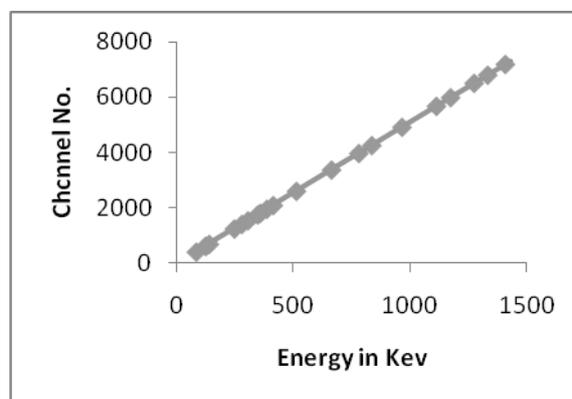

Fig 1: Energy calibration curve of HPGe detector

The empirical relations between the channel and the energies are therefore, as follows,

$$Y = 5.096\,X - 0.167 \qquad\qquad 1$$

Where Y and X are the energy in keV and channel No. respectively.

2.2.2. Energy Resolution

The resolution is a measure of a detector's capacity of distinguishing two closely spaced peaks in gamma energy spectrum. A calibrated ^{60}Co sources was placed on the surface of the detector. The output of the detector was collected in a multichannel analyzer through a preamplifier and an amplifier. In ^{60}Co spectrum there are two gamma ray peaks at 159.23 keV apart, one at 1173.23 keV and the other at 1332.51 keV. The energy resolution curve of the HPGe detector is shown in Fig. 2. The detector resolution obtained in this measurement is 1.69 keV at 1332.51 keV line of the ^{60}Co source.
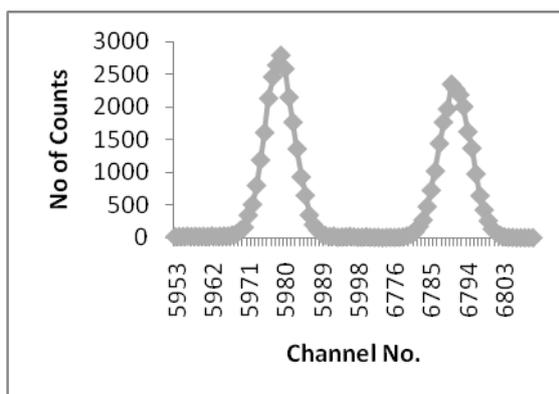

Fig. 2: The ^{60}Co spectrum for Energy Resolution of HPGe Detector

2.2.3. Efficiency Calibration of HPGe Detector

The efficiency of a detector is a measure of the number of gamma rays detected out of the total number of gamma rays that are actually emitted by the source.

The full energy peak detection efficiency curve was constructed using the Al_2O_3 based ^{226}Ra standard containing 800 Bq activities. The standard was prepared by homogenously mixing 800 Bq liquid ^{226}Ra with powdered Al_2O_3. Gamma ray counts of the standard (Al_2O_3 based ^{226}Ra) were performed for 5000 seconds at the surface of the detector. Since standard contain ^{226}Ra, which was radioactive substance, it decays successively by emitting gamma rays. The daughter nuclides of ^{226}Ra are ^{214}Pb, ^{214}Bi and others which characterized by their gamma energy. After taking counts, calculation was done for different energies of different nuclides. A graph has been plotted with energy along X-axis and surface efficiency along Y-axis, which is shown in Fig. 3. The efficiency curve was used to calculate the efficiencies of gamma energies emitting from different radionuclides determined in the present experiment.
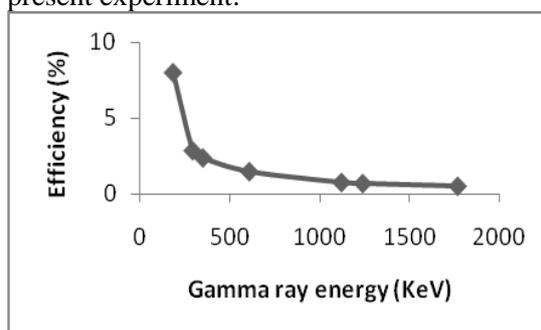

Fig. 3: Efficiency curve of the HPGe detector

The empirical equation for the relations between the efficiency (%) and the gamma ray energies are as follows,

$$Y = 2240 \cdot X^{-1.13} \qquad\qquad (2)$$

Where, Y is efficiency expressed in percentage and X is gamma-ray energy in keV

2.3. Counting of Sample and calculation

A high purity germanium detector was employed to measure the natural radioactivity produced by radioactive elements. Samples were placed at the surface of the HPGe detector and counted with digital gamma spectrometry system coupled with PC based Genei-2000, multi-channel analyzer acquisition software. The counting periods were 20,000 seconds for the samples. A background count was taken for 20000 seconds to obtain the background contribution in the samples. The software provides the

corresponding gamma spectra collected for both samples and standards. The spectra for samples were analyzed in two ways: manually and using gamma software package Hypermet PC Version 5.17. The spectra obtained from data acquisition (Genei-2000) software were binary mode. Then it was converted into ASKII data file, because Hypermet PC can only read the ASKII data file. The area under the peak in a gamma ray spectrum represents the number of counts collected for only that gamma ray energy. These peak areas were used for determination of radioactivity concentration of the radionuclides present in the sample. The software peak area was also checked manually. The net count of the sample is obtained by subtracting a linear background distribution of the pulse height spectra from the corresponding peak energy area. From the sample net counts activity of the sample were calculated using the formula

$$A = \frac{CPS \times 1000}{\varepsilon(abs) \times I_r(abs) \times W} \quad (3)$$

where, A is the activity in Bq.kg^{-1}, CPS is the net peak counts per second of the samples, W is the weight of the sample in gm, ε(abs) is the absolute gamma peak detection efficiency, $I_\gamma(abs)$ is the absolute gamma intensity of the corresponding gamma ray energy. Gamma rays

intensities were taken from the literature [8]. The peak detection efficiencies were calculated from the full energy peak detection efficiency curve constructed using Al$_2$O$_3$ based ^{226}Ra standard as shown **Fig 3**. The error was expressed in terms of the standard deviation $(\pm 1\sigma)$.

3. Result and Discussions

Using HPGe digital gamma ray spectroscopy system the activity concentration of ^{214}Bi and ^{228}Ac were measured. The gamma lines 609.31 keV, 768.36 keV, 1120.29 keV, 1238.11 keV, 1764.49 keV (due to ^{214}Bi) and 338.4 keV, 911.07 keV, 969.11 keV (due to ^{228}Ac) were considered to measure the activity concentration of these radionuclides. The measured mean activity concentration of ^{214}Bi was found to be 142.4±6.3 Bq.kg^{-1} range from 11.2±3.8 Bq.kg-1 to 921.4±34.3 Bq.kg^{-1} and the mean activity concentration of ^{228}Ac was found to be 61.3±7.7 Bq.kg^{-1} ranged from 31.3±10.1 Bq.kg^{-1} to 397.7±17.5 Bq.kg^{-1}. The detailed result is shown in Table 1. Table 2 shows the range and mean values of activity concentration of ^{214}Bi and ^{228}Ac. Table 3 shows the comparison between this work with other works performed all over the world.

Table 1: Activity concentration of ^{214}Bi & ^{228}Ac in Bq.kg^{-1} in different fertilizer samples

Sample ID	Fertilizer Name	^{214}Bi	^{228}Ac
S-1	Urea	ND	397.7±17.5
S-2	Muriate of Potass(MOP)	ND	ND
S-3	Simple Super Phosphate(SSP)	61.1±4.3	ND
S-4	Feovit Mixture(NPKS)	55.9±4.4	ND
S-5	Raina Mixture(NPKS)	11.2±3.8	ND
S-6	Triple Super Phosphate(TSP)	738.1±20.9	ND
S-7	Green Mag(Sulpher & Magnesium)	ND	163.7±21.4
S-8	Bio Fertilizer	28.2±6.7	85.6±16.9
S-9	Cow dung	ND	85.4±23.2
S-10	Bone Dust	13.3±4.8	33.93±12.0
S-11	Agri Chelated(Chelated Zinc)	22.7±3.9	31.3±10.1
S-12	Di-Ammonium Phosphate(DAP)	ND	ND
S-13	Gypsum	921.4±34.3	ND

Table 2. Range and mean value of activity concentration of ^{214}Bi & ^{228}Ac in Bq.kg^{-1} in different fertilizer samples

Radion-uclides	Minimum	Maximum	Mean
^{214}Bi	11.2±3.8	921.4±34.3	142.4±6.3
^{228}Ac	31.3±10.1	397.7±17.5	61.3±7.7
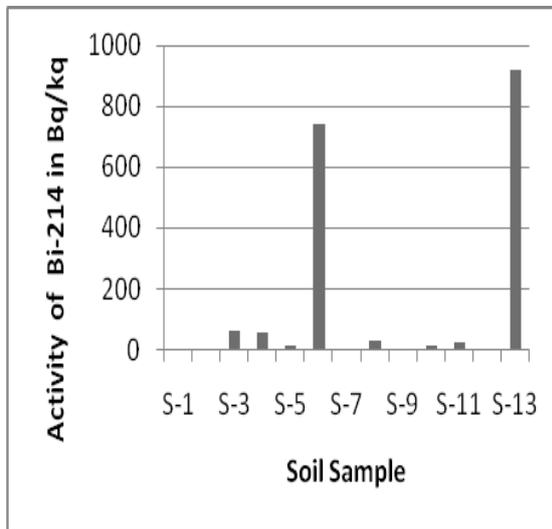
Fig. 4: Activity concentration of ^{214}Bi (^{238}U) in Bq.kq^{-1} in fertilizer samples
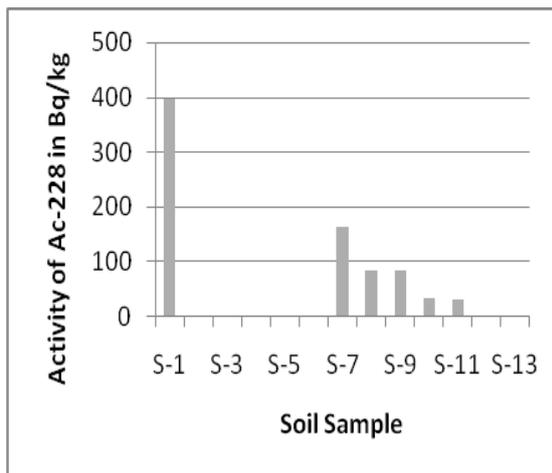
Fig 5: Activity concentration of ^{228}Ac (^{232}Th) in Bq.kq^{-1} in fertilizer samples

Since ^{214}Bi and ^{228}Ac are the member of ^{238}U and ^{232}Th series respectively, so the present study is compared with other work in which the activities of ^{238}U and ^{232}Th, and their daughters were measured.

Table 3: Comparison of the Present Study with Other Study in Home and Abroad

Country	Actinity concentration in Bq.kq^{-1}		References
	^{214}Bi(^{238}U)	^{228}Ac(^{232}Th)	
Gazipur	142.4±6.3	61.3±7.7	Present Study
Lebanon	401±8	-	5
Nigeria	1038.8	257	9
Egypt	301	125-239	10
Pakistan	386	38	11

4. Conclusion

In the present study the activity concentration of ^{214}Bi & ^{228}Ac were found in fertilizer samples. The result indicates that the activity concentrations of these radionuclides in the fertilizer of Gazipur in Bangladesh are within the range of values reported in other countries. but the activity levels are higher than the permissible activity levels which are in general 41.0, 52.2 and 230 Bq.kg^{-1} for ^{238}U and ^{232}Th respectively[6]. The result also reflects that the activity of radionuclides in imported fertilizer (in TSP and Gypsum imported from Morocoo) is higher than locally made fertilizer. This is a great indication that locally made fertilizers are safer than imported fertilizer. In Bangladesh, generally Urea, MOP and SSP fertilizers are used in agricultural land. The activity of radionuclides in Urea, MOP and SSP fertilizer are lower than other fertilizers. The use of fertilizer which contains radionuclides above the safe limit on agricultural land can result in impacts to the environment and human health. Ongoing additions of these fertilizers to agricultural land will result in accumulative impacts to the land. These impacts can potentially result in elevated levels of many of these contaminants in crops grown on that land [5]. Since the activity concentration of radionuclides in most of the fertilizers are

slightly above levels in soil, the need for specific measures to control radiological hazards to individuals and the environment is very limited. This study could be useful as baseline data for radiation exposure to fertilizers, and their impact on agricultural land and human health.

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