

**STUDY ON CONCENTRATION AND DISTRIBUTION OF URANIUM
AND THORIUM IN SANDS OF POTENTIAL REGIONS OF
BANGLADESH**

MD. SAYDUR RAHMAN

MILITARY INSTITUTE OF SCIENCE AND TECHNOLOGY (MIST)

2021



STUDY ON CONCENTRATION AND DISTRIBUTION OF URANIUM
AND THORIUM IN SANDS OF POTENTIAL REGIONS OF
BANGLADESH

MD. SAYDUR RAHMAN

(B.Sc. Engg., PUST)


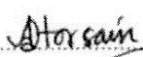


A THESIS SUBMITTED
FOR THE DEGREE OF MASTER OF ENGINEERING

DEPARTMENT OF NUCLEAR SCIENCE AND ENGINEERING (NSE)
MILITARY INSTITUTE OF SCIENCE AND TECHNOLOGY (MIST)

2021

The thesis titled “**Study on concentration and distribution of Uranium and Thorium in sands of potential regions of Bangladesh**” Submitted By: **Md. Saydur Rahman**, Roll No: **1016280012**, Session: April 2016 has been accepted as satisfactory in partial fulfillment of the requirement for the degree of M.Sc on Nuclear Science and Engineering.

BOARD OF EXAMINERS

1.  Chairman
Dr. Mohammad Amirul Islam (Supervisor)
Professor and Chief Scientific Officer
Reactor and Neutron Physics Division
INST, AERE, BAEC, Savar, Dhaka
2.  Member
Lt Col Md. Altab Hossain, PhD, EME (Co – Supervisor)
Instructor class A, Department of Nuclear Science and Engineering, MIST, Dhaka.
3. Member
Col Md. Abdur Razzak, SUP, psc (Ex-Officio)
Head, Department of Nuclear Science and Engineering, MIST, Dhaka.
4.  Member
Professor Dr. Md. Azizur Rahman
Department of Nuclear Science and Engineering, MIST, Dhaka.
5.  Member
Professor Dr. Mohammad Belal Hossen (External)
Head, Department of Physics, CUET, Chottogram.

“Declaration

I hereby declare that this thesis is my original work and it has been written by me in its entirety. I have duly acknowledged all the sources of information, which have been used in the thesis. The thesis (fully or partially) has not been submitted for any degree or diploma in any university or institute previously. ”



Name: Md. Saydur Rahman

Date: 25-02-2021

ABSTRACT

In this study, epithermal neutron activation analysis (ENAA) technique was implemented for the first time at 3 MW TRIGA research reactor of Bangladesh Atomic Energy Commission (BAEC) to determine elemental concentrations in soil/sand samples and characteristically compared it with instrumental neutron activation analysis (INAA) technique to find out advantageous features of ENAA over INAA. The assessment of three parameters (detection limit, analytical sensitivity and Q_0 values) using standard reference materials (IAEA-SL-1 and IAEA-Soil-7) indicate that ENAA is more preferable to INAA for the determination of low levels of Arsenic (As), Antimony (Sb), Samarium (Sm) and Uranium (U) in soil/sand samples.

In this study, total concentrations of 20 major, minor and trace elements including Th and U in surface and depth-wise inland and beach sands of potential regions of Bangladesh (Cox's Bazar, Potenga, Kuakata, Moulvibazar, Sylhet, Rajshahi and Sherpur) were determined by INAA and ENAA to deduce origin and extraction possibility of Th and U. The concentration ranges in the beach sands were from 4.63 to 382 mg/kg for Th and from 0.73 to 132 mg/kg for U whereas for inland sand, from 0.55 to 20.8 mg/kg and 0.97 to 4.57 mg/kg, respectively. From the concentration data of Th and U in inland and beach sands, the enrichment factor of Th and U, as well as their extraction possibilities, have been discussed. The depth-wise concentration variations for Th and U in beach and inland sands reveal that the overall U concentration variation decreases top to the deeper layer, but this variation in beach sands is higher than that of the inland sands of Bangladesh. The statistical analysis namely the Pearson correlation study reveals that high levels of Th and U in beach sands are due to the presence of Th and U rich heavy minerals in Cox's Bazar beach sands. The natural radioactivity concentrations (^{232}Th , ^{238}U and ^{40}K) and radiological hazard indices indicate that average radioactivity concentrations of ^{232}Th and ^{238}U in Cox's Bazar beach sands are relatively higher than those at other studied areas of Bangladesh as well as the world average values. However, external hazard indices at most of the sampling points are below the safe limit.

ACKNOWLEDGEMENTS

At first, I would like to express my gratitude to the Almighty, the Creator of the Universe, to whom all my praise is for helping me to complete my thesis. I am grateful to my honorable supervisor Dr. Mohammad Amirul Islam, Professor and Chief Scientific Officer, Reactor and Neutron Physics Division (RNPd), Institute of Nuclear Science and Technology (INST), Atomic Energy Research Establishment (AERE), Savar, Dhaka and co-supervisor: Lt. Col. Md. Altab Hossain, Ph.D, Instr Cl “A” & Post Graduate Program Coordinator, Dept. of Nuclear Science & Engineering (NSE), Military Institute of Science and Technology (MIST), Dhaka for their guidance, valuable suggestions and advice during this research work.

My sincere gratefulness goes to the Department of Nuclear Science and Engineering (NSE), the Military Institute of Science and Technology (MIST), and to the Institute of Nuclear Science and Technology (INST), AERE, for allowing their laboratory facilities and their 3 MW TRIGA Mark II research reactor to carry out this research. I would like to thank the Head, RNPd and other Scientists of the group for their help and encouragement during my research. Besides, my appreciation goes to the technical staff associated with this study, especially to the persons involved during sample preparation, irradiation and counting of the center for research reactor (CRR) and RNPd group, INST, BAEC.

I also convey my gratitude to, Col Md. Abdur Razzak, Col A Z M Salahuddin (Retd), Prof. Dr. Md. Azizur Rahman, Asst. Prof. Md. Ghulam Zakir, Capt. Md. Sifatul Muktadir and other teachers of the department of NSE, MIST, for their cordial affection, spontaneous assistance, untiring efforts, unprecedented motivation and deep assistance that played a significant role in the completion of my research work. This thesis work was difficult to complete without their invaluable assistance. I specially want to thank late Dr. M. A. Rashid Sarkar who helped me with a lot of guidance. Finally, I pray for the salvation of the souls of my parents from the core of my heart.

Table of Contents

ABSTRACT	v
ACKNOWLEDGEMENTS	vi
List of Symbols	x
List of Figures	xi
List of Tables	xii
CHAPTER 1	1
INTRODUCTION	1
1.1 Background	1
1.1.1 Neutron activation analysis	2
1.1.2 Basic principles of NAA	3
1.1.3 Classification of NAA method.....	3
1.1.4 Importance of NAA	5
1.2 Epithermal neutron activation analysis	6
1.3 Motivation of this study	7
1.4 Objectives of this study	8
1.5 Structure of this thesis	8
CHAPTER 2	9
LITERATURE REVIEW	9
CHAPTER 3	17
EXPERIMENTAL PROCEDURE	17
3.1 Study area.....	17
3.2 Instruments for collection of inland and beach sand samples.....	19
3.3 Samples collection	19
3.3.1 Surface samples collection.....	19
3.3.2 Depth wise samples collection.....	20
3.4 Instruments for preparing samples in the laboratory	21
3.5 Successive samples preparation for INAA	21
3.6 Samples analysis	23
3.6.1 Instrumental neutron activation analysis (INAA).....	23
3.6.2 Epithermal neutron activation analysis (ENAA)	24
3.7 Neutron sources.....	24
3.8 TRIGA MARK-II research reactor	25

3.9 Irradiation facilities of TRIGA MARK-II reactor	29
3.10 Experimental conditions for ENAA and INAA	29
3.11 Neutron irradiation	30
3.11.1 Irradiation for ENAA	30
3.11.2 Irradiation for INAA	30
3.11.2.1 Short irradiation	31
3.11.2.2 Long irradiation.....	31
3.12 High purity germanium (HPGe) detector system	32
3.13 Full specification of HPGe.....	33
3.14 Some important parameters of HPGe detector system	34
3.15 Gamma spectra analysis	36
3.16 Gamma-ray spectrometry system.....	37
3.17 Gamma-ray counting for ENAA and INAA	37
3.18 Determination of elemental concentration	38
3.18.1 Absolute NAA method.....	38
3.18.2 Comparative NAA method	41
3.19 Determination of activity concentration	41
3.20 Calculation for uncertainty.....	42
CHAPTER 4	44
EXPERIMENTAL RESULTS AND OBSERVATIONS.....	44
4.1 Detection limit.....	44
4.2 Analytical sensitivity.....	46
4.3 Comparison of Q_0 values of ENAA with INAA.....	47
4.4 Determination of elements by INAA technique	47
4.5 Quality control of the studied elements	48
4.6 Elemental abundances of inland surface sands of Bangladesh	51
4.7 Elemental abundances of beach surface sands of Bangladesh.....	56
4.8 The mean concentrations of Th and U in beach/inland sands.....	65
4.9 Elemental enrichment of the sands of Bangladesh	67
4.10 The mean value of enrichment factor (EF) for Th and U in beach/inland sands	74
4.11 Depth wise concentration Variations of Th and U in Beach/Inland sands	76
4.12 Conversion of elemental mean concentration into activity concentration.....	78
4.13 Calculation of radiological hazard indexes	78
4.13.1 Radium equivalent activity (Raeq)	79
4.13.2 External hazard index (H_{ex}).....	79

4.14 Activity concentration and radiological hazard indexes of inland and beach sands.....	80
4.14.1 Activity concentration of inland sands.....	80
4.14.2 Activity concentration of beach sands	83
4.15 Statistical analysis and inter-elemental correlation.....	89
CHAPTER 5	92
DISCUSSION ON RESULTS AND RELEVANCE.....	92
5.1 Comparison of the determined elemental concentration with literature data	92
5.2 Comparison of the determined activity concentration with literature data	94
CHAPTER 6	97
CONCLUSIONS AND RECOMMENDATIONS	97
6.1 Conclusions.....	97
6.2 Recommendations	98
References	99
Appendix A.....	106
Appendix B	107

List of Symbols

Abbreviation	Term
AERE	Atomic Energy Research Establishment
IAEA	International Atomic Energy Agency
NAA	Neutron Activation Analysis
INAA	Instrumental Neutron Activation Analysis
AAS	Atomic Absorption Spectrometry
AES	Atomic Emission Spectroscopy
XRF	X-Ray Fluorescence
ICP-MS	Inductively Coupled Plasma-Mass Spectrometry
RNAA	Radiochemical Neutron Activation Analysis
TNAA	Thermal Neutron Activation Analysis
ENAA	Epithermal Neutron Activation Analysis
PGNAA	Prompt Gamma-ray Neutron Activation Analysis
ICPS	Inductively Coupled Plasma Spectroscopy
SRM	Standard Reference Material
CRM	Certified Reference Material
NIST	National Institute of Science and Technology
SD	Standard Deviation
DGNAA	Delayed Gamma-ray Neutron Activation Analysis
FNAA	Fast Neutron Activation Analysis
TRIGA	Training, Research, Isotopes, General Atomics
GM	Geiger-Mueller
INST	Institute of Nuclear Science and Technology
NORM	Naturally occurring radioactive materials
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
ID	Identification Number
FWHM	Full width half maximum
UCC	Upper Continental Crust
BTRR	Bangladesh TRIGA Research Reactor
BAEC	Bangladesh Atomic Energy Commission

List of Figures

Figure 1.1: The processes of neutron capture by a target nucleus.	3
Figure 3.1: Samples collection from the studied area.	18
Figure 3.2: Required tools for collection of samples.	19
Figure 3.3: Instruments for preparing samples before irradiation at NAA Lab.	21
Figure 3.4: Samples preparation at NAA lab before irradiation.	23
Figure 3.5: TRIGA MARK-II research reactor at AERE, Saver, Dhaka.	26
Figure 3.6: Schematic diagram of the cross-sectional view of TRIGA MARK–II reactor core.	27
Figure 3.7: Core configuration and irradiation channels used for this study at BAEC research reactor.	30
Figure 3.8: Pneumatic irradiation facility of TRIGA Reactor at AERE, Savar, Dhaka.	31
Figure 3.9: HPGe detector used in this study at RNPD, AERE, Savar, Dhaka.	33
Figure 3.10: Energy resolution of the HPGe detector system (1173.9 keV).	35
Figure 3.11: Energy resolution of the HPGe detector system (1332 keV).	36
Figure 3.12: Gamm - ray Peak Analysis using a computer.	38
Figure 4.1: Comparison of ENAA and INAA spectra for IAEA-Soil-7 under the same experimental conditions.	45
Figure 4.2: Elemental concentrations of Th and U in sands of Moulvibazar.	53
Figure 4.3: Elemental concentrations of Th and U in Other’s regions (Sherpur, Rajshahi and Sylhet).	56
Figure 4.4: Elemental concentration of Th and U in sands of Patenga beach.	59
Figure 4.5: Elemental concentration of Th and U in sands of Cox’s Bazar beach.	62
Figure 4.6: Elemental concentration of Th and U in sands of Kuakata beach.	65
Figure 4.7: Th concentration of Beach sands/Inland sands.	66
Figure 4.8: U concentration of Beach sands/Inland Sands.	66
Figure 4.9: Enrichment factor (EF) of Th in beach/inland sands.	75
Figure 4.10: Enrichment factor (EF) of U in beach/inland sands.	75
Figure 4.11: Depth wise concentration Variations Th and U for Beach sands.	77
Figure 4.12: Depth wise concentration variations Th and U for Inland sands.	77
Figure 4.13: Activity concentration of ^{232}Th and ^{238}U in inland sands of Moulvibazar.	81
Figure 4.14: Activity concentration of ^{232}Th and ^{238}U in Other’s (Sherpur, Rajshahi and Sylhet) inland sands.	83
Figure 4.15: Activity concentration of ^{232}Th and ^{238}U in beach sands of Patenga.	85
Figure 4.16: Activity concentration of ^{232}Th and ^{238}U in beach sands of Cox’s Bazar.	87
Figure 4.17: Activity concentration of ^{232}Th and ^{238}U for beach sands of Kuakata.	89

List of Tables

Table 3.1: Detailed sample information with ancillary data.....	20
Table 3.2: Experimental conditions for ENAA and INAA.....	29
Table 3.3: Specification of the HPGe detector.	33
Table 3.4: Origin and typical magnitudes of uncertainties in NAA.	42
Table 4.1: Relevant nuclear data for the determination of five elements (As, Sb, Sm, Th and U).....	44
Table 4.2: Comparison of detection limit (mg/kg) for ENAA and INAA for IAEA-SL-1 and IAEA-Soil-7.....	45
Table 4.3: Comparison of analytical sensitivity (cps/g) for ENAA and INAA in the case of SL-1 and Soil-7.....	46
Table 4.4: Q_0 values for five elements (As, Sb, Sm, Th and U).....	47
Table 4.5: Studied radionuclides with their half-lives and gamma-ray energies.....	48
Table 4.6: Elemental abundances (mg/kg or in %) in repeated analyses of IAEA-SL-1(n=3) and NIST-1633b (n=8) of this study along with the certificate values.	50
Table 4.7: Elemental concentration (mg/kg or in %) in inland sands of Moulvibazar.	51
Table 4.8: Elemental concentration (mg/kg or in %) in inland sands of Sherpur, Rajshahi and Sylhet.	54
Table 4.9: Elemental concentration (mg/kg or in %) in beach sands of Patenga, Chittagong.	57
Table 4.10: Elemental concentrations (mg/kg or in %) in beach sands of Cox's Bazar, Chittagong.	59
Table 4.11: Elemental concentration (mg/kg or in %) in beach sands of Kuakata.	63
Table 4.12: Enrichment Factor for inland (Moulvibazar) sands in Bangladesh.	67
Table 4.13: Enrichment Factor for inland (Sherpur, Rajshahi and Sylhet) sands in Bangladesh. .	69
Table 4.14: Enrichment Factor for beach sands of Kuakata, Bangladesh.	70
Table 4.15: Enrichment Factor for beach sands of Patenga, Bangladesh.	71
Table 4.16: Enrichment Factor for beach sands of Cox's Bazar, Bangladesh.	72
Table 4.17: Elemental abundances (in $\mu\text{g/g}$, otherwise specified) in beach and inland sands with depth and spatial Variations.....	76
Table 4.18: Activity concentration and radiological hazard indexes for inland sands of Moulvibazar.....	80
Table 4.19: Activity concentration of Sherpur, Rajshahi and Sylhet inland sands.	82
Table 4.20: Activity concentration and radiological hazard indexes for beach sands of Patenga.	83
Table 4.21: Activity concentration and radiological hazard indices for Cox's Bazar beach sands.	85
Table 4.22: Activity concentration and radiological hazard indices for beach sands of Kuakata.	87
Table 4.23: Pearson correlation matrix for inter-element correlation in Beach sands of Bangladesh.....	91
Table 5.1: Comparison of elemental concentrations of K, Th and U in beach/inland sands in this study with some selective literature data in the world.....	93
Table 5.2: The mean activity concentration (Bq/kg) of radionuclide in beach sands and inland sands compared with literature data.	95

CHAPTER 1

INTRODUCTION

1.1 Background

In many different forms of rock, from sedimentary to volcanic, the element Uranium (U) is formed [1]. High concentrations of U are often caused by chemical conditions and occur significantly in some substances, such as deposits of phosphate rocks, minerals (such as lignite) and monazite sands in uranium-rich ores. The redistribution of U by wind, rain and geological processes in soil, rocks as well as water takes place in the environment. Moreover, it can dissolve in the water that any organism can drink. Naturally occurring radioactive elements (^{232}Th , ^{238}U and ^{40}K) are spread throughout geological and geochemical processes, and the vast majority of their radioactivity is found on Earth from primordial origins [2]. In nature, U is mainly contained as ^{238}U (99.2739-99.2752%), one of the heaviest elements that occur predominantly in the earth's crust with activity appearing in soil and sand [3].

In the earth's crust, the mean concentration of U ranges from 0.5 to 5 ppm [4], and in granite, it is 4 ppm [3]. Moreover, the radioactive Thorium (Th) substance occurs also naturally in association with other minerals, such as silica, in the earth's crust. In the form of ^{232}Th with a concentration range of 2-20 ppm, there are more than 99% of natural Th [4]. In underground mines, some rocks contain a more concentrated form of Th. These rocks are broken by natural weathering, such as wind and water action, and render the Th and all other rock components become part of the soil [5]. Also, soil containing high levels of clay materials has a higher Th content [6].

Instrumental neutron activation analysis (INAA) with a high-resolution gamma-ray spectrometry system is a powerful method for trace element analysis of sands and minerals.

Moreover, epithermal-NAA (ENAA) has the potential to determine the low level of Th, U and other elements in soil and sand samples. In this research, ENAA will be implemented for the first time at 3 MW TRIGA research reactor of Bangladesh Atomic Energy Commission (BAEC) to determine Th and U in sands of the potential regions of Bangladesh. The Th and U profile in Bangladeshi sands will be determined by INAA and ENAA to deduce their extraction possibility.

Th and U are naturally occurring elements used as fuel in the nuclear reactor. Traces of Th and U are found everywhere but commercial extraction is possible only in the locations where the proportions of Th and U are adequate. The largest deposits are found in Kazakhstan, Canada and Australia, etc. [7]. Among the different analytical techniques, instrumental neutron activation analysis (INAA) and epithermal neutron activation analysis (ENAA) can be used to determine concentrations of Th and U in soil and rocks due to the non-destructive and highly sensitive nature of these techniques [8]. Between ENAA and INAA, ENAA has the potential to improve detection limits to determine U. Therefore, it is important to implement ENAA in the BAEC TRIGA Research Reactor (BTRR) to determine the low concentration of U in soil and rocks of Bangladesh. Since ENAA was not applied in the BTRR earlier, it was needed to characterize this analytical technique at BTRR.

1.1.1 Neutron activation analysis

Neutron activation analysis (NAA) is a nuclear analytical technique used widely for the determination of elemental concentrations in a vast number of materials. In NAA, studied samples were excited by capturing neutrons and product isotopes emit gamma rays. It represents the identification and quantification of the elements in the sample.

1.1.2 Basic principles of NAA

When the incident neutrons from the reactor strike the target nucleus, it forms into a compound nucleus (shown in Figure 1.1). The excitation power of the compound nucleus is due to the neutron's binding energy to the nucleus. The compound nucleus then turns into a radioactive nucleus by emitting prompt gamma rays (emits within 10^{-12} to 10^{-15} s). Finally, the radioactive nucleus decays to the product nucleus by emitting beta particles and delayed gamma rays. These gamma rays are counted by an HPGe detector to determine the elemental contents. Figure 1.1 illustrates the process of neutron capture by a target nucleus followed by the emission of gamma rays [9, 10].

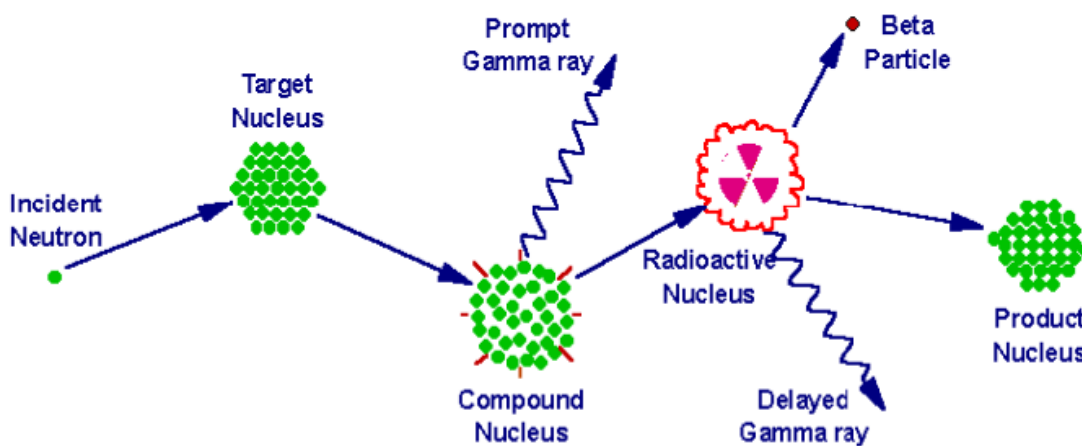


Figure 1.1: The processes of neutron capture by a target nucleus.

1.1.3 Classification of NAA method

There are many types of neutron activation analysis, which are as follows:

Instrumental neutron activation analysis (INAA)

Using automated sample handling, gamma-ray analysis with solid-state detectors, and computerized data processing, NAA can typically calculate more than thirty elements simultaneously without chemical processing in most sample forms. Instrumental neutron

activation analysis (INAA) is generally referred to as the implementation of strictly instrumental procedures [11]. INAA is often referred to as non-destructive NAA or NAA without post-irradiation radiochemical separation.

Radiochemical neutron activation analysis (RNAA)

NAA in which chemical separation is done to sample after irradiation to remove interference or to concentrate the radioisotope of interest, the technique is called radiochemical neutron activation analysis [11]. This technique is referred to as destructive NAA and is performed infrequently due to its high labor cost. The technique can also be categorized according to whether gamma-ray is measured neutron irradiation or at some time after the end of the irradiation.

Prompt gamma-ray neutron activation analysis (PGNAA)

The PGNAA method is usually performed by using a neutron beam collected through a port of the reactor beam. Fluxes are one million times lower on samples irradiated in the beam than on samples inside a reactor, but detectors can be positioned very close to the sample, compensating for much of the sensitivity loss due to flux. Most commonly, the PGNAA approach is used to evaluate lighter elements (H, C, N, O, F and S).

Delayed gamma-ray neutron activation analysis (DGNA)

For the vast majority of elements that create radioactive nuclides, DGNA is useful. With respect to time, the technique is versatile such that by waiting for the short-lived radionuclide to decay, the sensitivity of a long-lived radionuclide suffering from interference by a shorter-lived radionuclide can be increased. This is a primary advantage of DGNA over other methods of analysis.

Epithermal neutron activation analysis (ENAA)

Epithermal neutron activation analysis is useful when the nuclides of interest have a much higher nuclear cross-section by resonance capture of epithermal neutron than the thermal neutrons [12], while the interfering elements have not or have lower reaction cross-section, the actively by epithermal neutrons can considerably increase the sensitivity of the detection for an element.

Fast neutron activation analysis (FNAA)

When the measurement of elements (lighter elements such as C, N, O, F and S) by thermal neutron is not favorable because of the very low (n, γ) cross-section [12], the fast neutron (14 MeV) can be used to produce a reaction of type $(n, 2n)$, (n, α) , (n, p) and (n, n) . The advantage of 14 MeV NAA is fast and non-destructive, can be used on a routine basis, is ideal for short irradiation and is particularly sensitive for the detection of the lighter elements.

1.1.4 Importance of NAA

Various types of analytical tools used for the determination of elemental concentrations and studying the chemical composition of environmental geochemical samples, such as atomic absorption spectroscopy [13], inductively coupled plasma mass spectrometry [14, 15], inductively coupled plasma atomic emission spectrometry [16], inductively coupled plasma optical emission spectrometry [17], Ion chromatography (IC), Laser induced fluorimetry (LIF) and UV–Vis spectrophotometer [18], etc. Unlike the above-mentioned analytical methods, neutron activation analysis (NAA) is free from chemical digestion and is nondestructive as well as independent of the chemical form [19]. NAA is considered to be a primary method of measurement and possesses versatile applicability [19, 20]. So, in this study, we have used NAA for the determination of Th, U and other elements concentration in the inland and beach sand samples.

Neutron Activation Analysis (NAA) has become very important due to its various advantages such as:

a. Number of elements: When exposed to the neutron flux in a reactor, sixty-seven (67) common and rare earth elements become radioactive. About 50 of the 67 elements can be identified and calculated very easily. No other techniques are able to determine such many elements at a time.

b. Highly sensitive: The method allows all observable elements with high sensitivity to be calculated; many elemental concentrations can be measured in parts per million (ppm) or parts per billion (ppb).

c. Elemental analysis: This technique analyses many elements regardless of their chemical form.

d. Non-destructive: Unlike other techniques, the sample is not destroyed by the analysis, and can be re-analyzed if necessary.

e. Accuracy and precision: This technique is noted for its accuracy and precision.

1.2 Epithermal neutron activation analysis

An epithermal neutron activation analysis (ENAA) is a form of NAA with neutron energies ranging from 0.5 eV to 10 KeV. Nuclides with high epithermal neutron capture cross-sections are preferentially activated when irradiated under a cadmium filter. Therefore, elements with nuclides having high resonance integrals can potentially be determined by ENAA. Since the threshold energy of epithermal neutrons for cadmium (Cd) is 0.5 eV, a thin foil of Cd material (0.5-1 mm thick) strongly absorbs all neutron energies below this threshold energy [21]. In ENAA, a sample is irradiated in an epithermal neutron flux by covering it with cadmium foil or putting it in a borated capsule. This technique is used for the determination of trace amounts of elements in samples [22, 23]. In instrumental neutron activation analysis (INAA), thermal

neutrons can activate major matrix elements, such as Aluminum (Al), Manganese (Mn), Sodium (Na), Chromium (Cr), etc. in geological samples because of their high thermal neutron capture cross-sections, whereas ENAA can largely reduce their radioactivities and enhance the sensitivity for some trace elements determination such as arsenic (As), Antimony (Sb), Samarium (Sm) and Uranium (U), etc. [24, 25]. Therefore, ENAA can be a potential method for the determination of some elements, especially U in geological samples.

1.3 Motivation of this study

The 3 MW TRIGA Mark II research reactor of Bangladesh Atomic Energy Commission (BAEC) has been used for multiple purposes like training, education, radioisotope production and various R & D activities in the field of nuclear science and technology since 1986 [26,27]. Since its inception, BAEC TRIGA Reactor was used for the determination of major, minor and trace elements including Th and U in different sample matrices using INAA [8, 19, 28]. However, ENAA was not implemented at BAEC TRIGA Research Reactor (BTRR) due to the absence of a Cd-screened channel at the reactor. ENAA can also be implemented in the usual irradiation channel of a research reactor when samples along with standards are put into a Cd/B covered sample box.

The Rooppur nuclear power plant (2 x1200 MW) at Pabna, the first nuclear power plant in Bangladesh, is under construction. The U fuels for the plant will be supplied by Russia. However, it is very important to know the concentration level of U in soils and sands of Bangladesh to find out possible U-mining areas. Since ENAA can be used to determine a very low level of some elements like U in soil/sand samples, the characterization of ENAA at BAEC TRIGA reactor is needed to implement this technique.

1.4 Objectives of this study

The main objectives of this study are:

- a. To implement epithermal neutron activation analysis (ENAA) at TRIGA MARK-II research reactor for determination of Th and U.
- b. To assess Th and U profile in the sands of potential regions of Bangladesh to deduce their extraction possibility.
- c. To assess natural radioactivity concentration and different radiological hazard indices (radium equivalent activity, external hazard index) of the sand samples.

1.5 Structure of this thesis

a. Introduction: In this chapter, some basic information discussed instrumental neutron activation analysis (INAA) and epithermal neutron activation analysis (ENAA) and also listed down objectives of this study.

b. Literature review: In this chapter, some literatures related to my research are mentioned and written as a summary.

c. Experimental procedure: Two irradiation facilities one is neutron activation analysis and the other is epithermal neutron activation analysis were used for the determination of elemental concentration in Bangladeshi inland and beach sand samples. Samples and standards were irradiated in 3MW TRIGA Mark II research reactor, AERE, Savar, Dhaka

d. Experimental results and observations: Based on the experiment, determined elemental concentration, activity concentration and radiological hazard indexes were shown in some tables and analyzed studied elements also.

e. Discussion on results and relevance: In this section, the results were briefly explained and experimental results compare with national and international values.

CHAPTER 2

LITERATURE REVIEW

In this research, instrumental neutron activation analysis (INAA) technique and epithermal neutron activation analysis (ENAA) were used for the determination of elemental concentrations of 20 studied elements with special emphasis on Th and U in inland and beach sands of Bangladesh. There is some literature related to the activity concentrations of primordial radionuclides and elemental concentrations in different sample matrices using different analytical techniques worldwide. The literature studies most relevant to this study are mentioned below with proper information.

Acharya et al. 2013 [22] applied an epithermal neutron activation analysis (ENAA) technique to determine trace concentrations of Th in four samples of uranium oxide (U_3O_8) samples. At a reactor neutron flux of about $10^{12} \text{ cm}^{-2} \text{ s}^{-1}$, samples and Th standards, wrapped with cadmium foil, were irradiated. A Compton suppressed anticoincidence gamma-ray spectrometer consisting of HPGe-BGO detectors paired with MCA was used to perform radioactive testing. Th concentrations in these samples were in the 15-72 mg/kg range. Two types of chemical methods, ion chromatography (IC) and inductively coupled plasma atomic emission spectrometry (ICP-AES), compared the results of ENAA with those obtained. It was established that the results obtained by the three methods were in good agreement, indicating further validity of the proposed ENAA method.

Hou et al. 1996 [24] used the ENAA method to evaluate the cadmium and boron ratios of 44 elements and 66 nuclides at the internal and external irradiation sites of the Miniature Neutron Source Reactor (MNSR) at the China Institute of Atomic Energy. A permanent Cd-shielded epithermal neutron radiation site has been planned and built inside the outer part of the

beryllium reflector of this reactor. BN-shielded epithermal neutron activation analysis (ENAA) and Au, As, Sb, Th and U were analyzed by Cd-shielded ENAA in geological and environmental samples for elements such as I, Br, St, Si, Th and U in biological samples (foodstuffs, water and blood) (soil, rock, sediment and vegetable leaves). The results show that ENAA's detection limits of these elements are higher than those of traditional NAA by a factor of 1.5-7.

Parry et al. 1982 [29] analyzed soil samples and the cadmium ratios of 52 short-lived nuclides were measured by using the ENAA technique in the University of London Reactor Centre. Irradiation of the epithermal neutron decreases the behaviors of certain nuclei by factors of 20-30. Besides, the measured changes in the As, Sb, Th and U detection limits are in the ranges of 1-6. Theoretically, epithermal neutron activation under cadmium would enhance the identification of several short-lived nuclides in various types of rock. The method has been successfully applied to the determination of hafnium, silver and rhodium, and many elements such as Sc, Co and Se can be determined simultaneously, although applications are limited.

Tokay et al. 1985 [30] applied first epithermal neutron activation analysis (ENAA) technique in Sweden in 1985 and analyzed the existence of non-overlapping resonance peaks in the neutron absorption cross-section spectra for the nuclides present in the sample. By this method, it is possible to reduce some of the interfering activities with respect to the sought activity by using Cd filters.

Stuart et al. 1981 [31] applied epithermal neutron activation analysis (ENAA) in a SLOWPOKE nuclear reactor. A total of 69 elements have been calculated using cadmium and boron shielding. Additionally, the Th and U cadmium ratios were 2.8 and 1.2, respectively, while the Th and U advantage factors were 9.3 and 10.9, respectively.

Khan et al. 2017 [32] analyzed thirty sand samples of Cox's Bazar, Bangladesh and determined nineteen elemental concentrations (Na, K, Sc, Cr, Fe, Co, Zn, As, La, Ce, Sm, Eu, Gd, Tb, Yb, Hf, Ta, Th and U) for understanding the spatial and layer-wise distribution of heavy elements and their provenance. Sand samples were collected from six different stations and from each sampling station five samples were collected according to the sampling depth. These elements were determined by TRIGA Mark II research reactor-based instrumental neutron activation analysis (INAA). Heavy metals (Fe, La, Ce, Sm, Hf, Ta, Th and U) are enriched due to the presence of some heavy minerals (monazite, magnetite, ilmenite).

Yasmin et al. 2018 [33] determined elemental abundances of three elements (K, Th and U) in the different types of samples like soil, sand and sediment collected from Potenga sea beach area of Chittagong. The average mass concentrations of U, Th and K were found as 3.98 ± 0.51 ppm, 17.06 ± 0.91 ppm, $1.60 \pm 0.01\%$ for sand; 5.30 ± 0.46 ppm, 20.42 ± 1.19 ppm, $2.96 \pm 0.02\%$ for soil and 7.59 ± 0.65 , 29.93 ± 1.52 ppm and $1.56 \pm 0.02\%$ for sediment samples, respectively. Moreover, the deduced mass concentrations of the primordial radionuclides U, Th and K in the investigated samples are corresponding to the typical rocks, crustal minerals and granite rocks, respectively.

Ullah et al. 2018 [34] determined U concentrations in water, soil and stones collected from six different locations of Sherpur District, Bangladesh, by adsorptive cathodic stripping (ACS) voltammetric technique. The method is based on the uranium(VI)-chloranilic acid (CA) complex's adsorptive accumulation on a hanging mercury drop electrode, followed by a cathodic voltammetric scan using differential pulse modulation to minimize the complex. For the soil and stone samples, the elemental concentration of U (uranium) was 3.8 and 5.3 ppb, respectively. The 10.3 ppb value was obtained from a water sample.

The range of U concentration values measured in water, soil and stones was 8.9-16.4 ppb, 16.3-31.7 ppm and 19.2-161.6 ppm respectively.

Ullah et al. 2015 [35] analyzed and determined the concentration of U in the water of a stream branch, beach sands and U-rich rocks collected from different locations of Bangladesh, by adopting the trace U determination technique. The concentration of U in stream water, beach sands and rocks were estimated to be 21 ppb, 34.42-39.55 ppm and 530.08 - 108.89 ppm respectively. The present research appears to be the first investigation into the quantification of U in rocks containing high U by the 'Differential Pulse Cathodic Stripping Voltammetry (DPCSV)' trace level determination technique.

Rahman et al. 1993 [36] evaluated the geological materials such as radioactive rock samples collected from the Hararganj Anticline in Moulvibazar, Bangladesh and determined the elemental concentration of Th and U. In this case, in the qualitative and quantitative study of the rock samples for Th and U, the pure instrumental neutron activation analysis (INAA) technique was used. The samples were properly prepared together with their standards and irradiated simultaneously in the order of 10^{12} n cm⁻² neutron flux s⁻¹ using the TRIGA MARK II research reactor facility at the AERE, Savar, Dhaka. After activation, using a high purity germanium detection system, the samples were subjected to gamma-ray spectrometry.

Majumder et al. 2016 [37] analyzed a total of 19 stream sediments containing black minerals and sandy pebble type samples for Th and U mineralization in Kulaura and Baralekha Upazillas. Major, minor and trace elements of those studied samples were determined by the instrumental neutron activation analysis (INAA) technique. Besides, among these elements, the elemental concentration of Th and U were 6.0 ppm and 47.1 ppm, respectively. U is strongly associated with Sc, La, Ce, Sm, Tm and Ta.

Khalil et al. 2016 [38] determined the activity concentration of ^{238}U , ^{232}Th , and ^{40}K and distribution of these radionuclides, their particular activities in the sands and sediments of the Brahmaputra (Jamuna) River in Bangladesh, along with their mineral characteristics, were studied to assess the level of radiation and to establish a baseline database for potential comparison in the event of any shift in the region under research due to anthropogenic activity. Natural radioactivity radiological parameters were evaluated by measuring the radium equivalent activity, the hazard index, the absorbed dose rate and the annual effective dose. The mean concentrations of activity in sand and sediment of ^{226}Ra (^{238}U), ^{232}Th , and ^{40}K were found to be 59 ± 2 & 60 ± 2 Bq/kg, 113 ± 5 & 135 ± 5 Bq/kg, and 983 ± 42 & 1002 ± 43 Bq/kg, respectively. It was found that the measured mean absorbed dose rate and annual effective dose were 150 nGy/h and 0.18 mSv/year respectively. Such high values are correlated with sediment's mineral content. X-ray diffraction peaks of sand and sediment samples classify the minerals to be found in the samples: quartz, feldspar, rutile, zircon, monazite, uranium fluoride, hematite, kyanite, and uranium arsenide.

Zaman et al. 2016 [39] focused on the elevated levels of environmental radioactivity present in heavy mineral deposits located along a 120-km coastal section of Cox's Bazar on the eastern panhandle of Bangladesh. Gamma-ray spectroscopy has been used to examine bulk beach sands and individual mineral fractions. In the bulk beach sand samples, the activity concentrations of ^{238}U , ^{235}U , ^{232}Th and ^{40}K were found to be considerably high and associated positively with the concentration of heavy minerals in the sand. The highest concentrations of activity were contained in the mineral fractions in the zircon fraction, followed by garnet, rutile, ilmenite and magnetite.

Alam et al. 1999 [40] measured the radioactivity of ^{238}U , ^{232}Th and ^{40}K in beach sand minerals from the Plant of the Beach Sand Exploitation Center (BSEC) and soils from the tourist zone of Cox's Bazar, Bangladesh by using gamma-ray spectrometry to estimate the radiation hazard

as well as establish a database for radioactivity levels of the Plant and the tourist area nearby. The average activity concentrations of the corresponding nuclides were 6439, 1324 and 472 Bq/kg for zircon; 348, 388 and 59.7 Bq/kg for ilmenite; 22.0, 43.1 and 293 Bq/kg for magnetite; 3951, 7903 and 213 Bq/kg for garnet; 6643, 11670 and 182 Bq/kg for rutile; 2582, 4684 and 639 Bq/kg for heavy sand and 110, 213 and 501 Bq/kg for light sand, respectively.

Islam et al. 2012 [41] determined the activity concentrations of radionuclide in the sand and soil samples obtained from the Patuakhali district of Bangladesh's Kuakata sea beach using a 40 percent relative efficiency of high-resolution high-purity germanium (HPGe) detector. In the Kuakata seabeach sand samples, the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K ranged from 2.82 ± 4.89 to 87.96 ± 4.45 Bq/kg, 21.72 ± 16.27 to 290.93 ± 18.15 Bq/kg and 26.24 ± 0.35 to 852.05 ± 142.15 Bq/kg, respectively. The activity concentrations of corresponding radionuclides were observed for soil samples in the range of 20.98 ± 3.96 to 42.92 ± 4.76 Bq/kg, 59.25 ± 15.62 to 144.34 ± 18.52 Bq/kg and 570.43 ± 100.3 to 1165 ± 166.27 Bq/kg respectively.

Miah et al. 2013 [42] measured the activity concentrations of naturally occurring radionuclides ^{238}U , ^{232}Th and ^{40}K by using a Broad Energy Ge (BEGe) detector for soil samples collected from the Malnichara tea garden in Sylhet district of Bangladesh. In soil samples, the average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K were identified to be 55.284 ± 4.68 Bq/kg, 125.27 ± 5.81 Bq/kg and 497.91 ± 43.83 Bq/kg. For the corresponding nuclides, the results obtained are significantly higher than the global mean values of 30 Bq/kg, 35 Bq/kg, and 400 Bq/kg, respectively.

Zaman et al. 2012 [43] determined and quantified Th and U elemental concentration in zircon assemblages using NAA from beach placers of Cox's Bazar. In a mineral processing pilot plant, Zircon was extracted from beach sands. A polarizing petrographic microscope and X-ray diffraction techniques analyzed the separated zircon assemblages and found that more

than 75 percent pure zircon was present. Th and U concentrations in zircon were subsequently calculated by NAA and showed that zircon contained 94-141 ppm U and 127-506 ppm Th.

Tzortzis et al. 2004 [44] analyzed 115 soil samples and determined the elemental concentration of Th, U and K in the bedrock surface of the island based on the different lithological units of the study area. Elemental concentrations for Th (range 2.5×10^{-3} to 9.8 mg/kg), U (8.1×10^{-4} to 3.2 mg/kg) and potassium (1.3×10^{-4} to 1.9%) were determined from the measured gamma-ray spectra. From all samples, the measured arithmetic mean values (A.M \pm SD) are (1.2 ± 1.7) mg/kg, (0.6 ± 0.7) mg/kg and (0.4 ± 0.3) percent respectively, for Th, U and K which are three to six times lower than the world average values of 7.4 mg/kg (Th), 2.8 mg/kg (U) and 1.3% (K) from all world data available. The most suitable relationship between the concentrations of Th and K versus U and also of K versus Th is a linear form with a coefficient of correlation of 0.93, 0.84 and 0.90, respectively. The extracted Th/U, K/U and K/Th ratios (slopes) are equal to, respectively, 2.0, 2.8×10^3 and 1.4×10^3 .

Bajoga et al. 2019 [45] determined the elemental concentration of ^{238}U , ^{232}Th and ^{40}K in the ninety (90) surface soil samples of Kuwait. Obtained elemental concentration were in the ranges from 0.48 to 2.61 mg/kg, 0.87 to 5.23 mg/kg, and 0.24 to 2.23%, with a mean value of 1.39 mg/kg, 3.47 mg/kg, and 1.18%, for the ^{238}U , ^{232}Th and ^{40}K , respectively. In addition, the research was carried out among the five soil types established, i.e. Petrocalcids (S3), Petrogypsids (S4), Aquisalids (S1), Calcigypsid (S2), and Torripsammit (S5). The highest concentrations of Th and U radioactivity were reported in soil S2 (Calcigypsid), with values of 1.71 (Bq/kg) and 4.45 (Bq/kg), respectively. The minimum and maximum ^{40}K values are 1.1(percent) and 1.27(percent) and are prevalent in the soil forms of Aquisalids (S1) and Petrocalcids (S3). The elementary concentration ratios are 2.53, 0.09 and 0.03 for $^{232}\text{Th}/^{238}\text{U}$,

$^{40}\text{K}/^{238}\text{U}$, $^{40}\text{K}/^{232}\text{Th}$ across soil types, with a correlation coefficient of 0.92, 0.34, and 0.38, respectively.

Maxwell et al. 2013 [46] quantified the activity concentration of ^{232}Th , ^{238}U and ^{40}K of rock samples from site one (S1L1–S1L11, 70 m) and site two (S2L1–S2L9, 60 m) boreholes in Dei-Dei and Kubwa was presented and the first time in the region to be compared. Using a high-resolution co-axial HPGe gamma-ray spectrometer system, activity concentrations were measured. Site-one borehole activity concentrations ranged from 45 ± 1 to 98 ± 6 Bq/kg for ^{232}Th , 18 ± 2 to 37 ± 4 Bq/kg for ^{238}U and 254 ± 32 Bq/kg to 1195 ± 151 Bq/kg for ^{40}K . At site two, the activity concentration levels were 32 ± 3 to 84 ± 7 Bq/kg for ^{232}Th , 15 ± 2 to 52 ± 5 Bq/kg for ^{238}U and 119 ± 15 to 705 ± 94 for ^{40}K Bq/kg. Samples obtained from layers S1L7, S1L11 and S2L1 display a significantly higher concentration of ^{232}Th and ^{238}U .

In previous works, they determined the elemental concentration of thorium (Th) and uranium (U) as well as their radioactivity concentration in a limited number of soil/sand samples in the specific area of Bangladesh. In this study, Th, U and natural radioactivity concentrations in surface and depth-wise beach and inland soil/sands from potential regions (Cox's Bazar, Potenga, Kuakata, Sylhet, Moulvibazar, Sherpur and Rajshahi) of Bangladesh are determined to study Th and U distribution in the country and their extraction possibility.

CHAPTER 3

EXPERIMENTAL PROCEDURE

In this study, Instrumental Neutron Activation Analysis (INAA) and Epithermal Neutron Activation Analysis (ENAA) using Bangladesh TRIGA research reactor (BTRR) were used to analyze soil and sand samples of Bangladesh.

3.1 Study area

Bangladesh is located on the northern coast of the Bay of Bengal with many sea beaches containing heavy minerals. In this study, surface and depth-wise inland and beach sands were collected from potential regions (Cox's Bazar, Potenga, Kuakata and Moulivibazar, Sylhet, Rajshahi and Sherpur) of Bangladesh. Kuakata is a sandy beach of 11 km long and 3 km wide which is situated at Patuakhali district in southeastern Bangladesh [41]. Cox's Bazar and Potenga are important two sandy sea beaches, the most attractive places for foreign and local people [47]. Moulivibazar is another potential region containing rocks with Th and U bearing minerals in the Sylhet Division of Bangladesh. It is located close to the Meghalaya and Assam State of India where some U mines are located [42]. The sample collection areas are indicated as red star in the Map of Bangladesh (shown in Figure 3.1).



Figure 3.1: Samples collection from the studied area (Source: <http://www.maps-of-the-world.net/maps-of-asia/maps-of-bangladesh>).

3.2 Instruments for collection of inland and beach sand samples

In this study, many types of equipment were required to collect inland and beach sand samples (shown in Figure 3.2).



GPI Pipe



Zip lock pack



Hand gloves



Shovel



Note book



Marker

Figure 3. 2: Required tools for collection of samples.

3.3 Samples collection

In this study, surface and depth-wise inland and beach sand samples were collected to determine the studied 20 elemental concentrations.

3.3.1 Surface samples collection

A total of 49 surface inland and beach sand samples were collected from 49 sampling positions of Cox's Bazar, Potenga, Kuakata, Moulivibazar, Sylhet, Sherpur and Rajshahi area. Each sampling point was about 1 km away from the near one. From each sampling point, about 1 kg of soil/sand sample was collected and immediately stored in acid-rinsed polyethylene plastic bags to prevent cross-contamination and identified each sample by giving a unique

identification number (ID). All identified samples were then transported to the laboratory of the Instrumental Neutron Activation Analysis (INAA), Bangladesh Atomic Energy Commission (BAEC) for elemental analysis.

3.3.2 Depth wise samples collection

Depth-wise inland and beach sands were also collected from 2 locations, at every location four soil/sand samples were collected at a depth of 0–5 cm, 5–15 cm, 15–25 cm, 25–35 cm, respectively. During the sample collection, hand gloves were used to avoid contamination and giving identification numbers (ID) carefully in each sampling packet. The detailed information such as sample ID, their locations, GPS data as well as the depth from the surface is shown in Table 3.1.

Table 3.1: Detailed sample information with ancillary data.

Sample ID	Location	Latitude	Longitude	Depth from the surface (cm)
C-01				0-05
C-02	Cox's Bazar	21°25'90" N	91°58'06" E	05-15
C-03				15-25
C-04				25-35
M-01				0-05
M-02	Moulvibazar	24°30'01" N	92°8'16" E	05-15
M-03				15-25
M-04				25-35

3.4 Instruments for preparing samples in the laboratory

A lot of tools were also needed to prepare the samples before irradiation (Shown in Figure 3.3).



Mortar and pestle



Mesh



Petri dishes



Digital micro



Impulse sealer



De-ionized water



Teflon



Electric Oven

Figure 3.3: Instruments for preparing samples before irradiation at NAA Lab.

3.5 Successive samples preparation for INAA

To implement INAA and ENAA, the collected samples were prepared until irradiation using the following steps (shown in Figure 3.4).

Drying

Petri dishes were cleaned with de-ionized water and put into the oven for drying. The identification number (ID) was given on each Petri dish according to the sample ID. Collected samples were put into the Petri dishes corresponding to the given ID. The petri dish containing inland and beach sand samples was then allowed to dry in an electric oven at 50° C until gain constant weight [48].

Grinding

Using a mortar and pestle, the dried samples were then ground into a small grain size and homogeneous blend and sieved through a 0.25 mm aperture to extract organic materials, stones and lumps.

Weighing

At first, some small polyethylene bags were prepared for carrying the sample. The masses of the polyethylene bag with and without samples were measured separately by using a digital microbalance device. This step proceeded with intensive care. However, to get the sample mass, the bag mass was subtracted from the total mass (Bag plus sample mass). In this way, we get the actual mass. About 50 mg of each dried powder sample was weighed in a polyethylene bag and heat-sealed.

Irradiation vial

De-ionized water was also used to clean all necessary irradiation vials and the identification number (ID) was given on each vial according to the sample ID. Besides, for epithermal and instrumental neutron activation analysis, the prepared double heat seal samples and standards were placed into the vial.

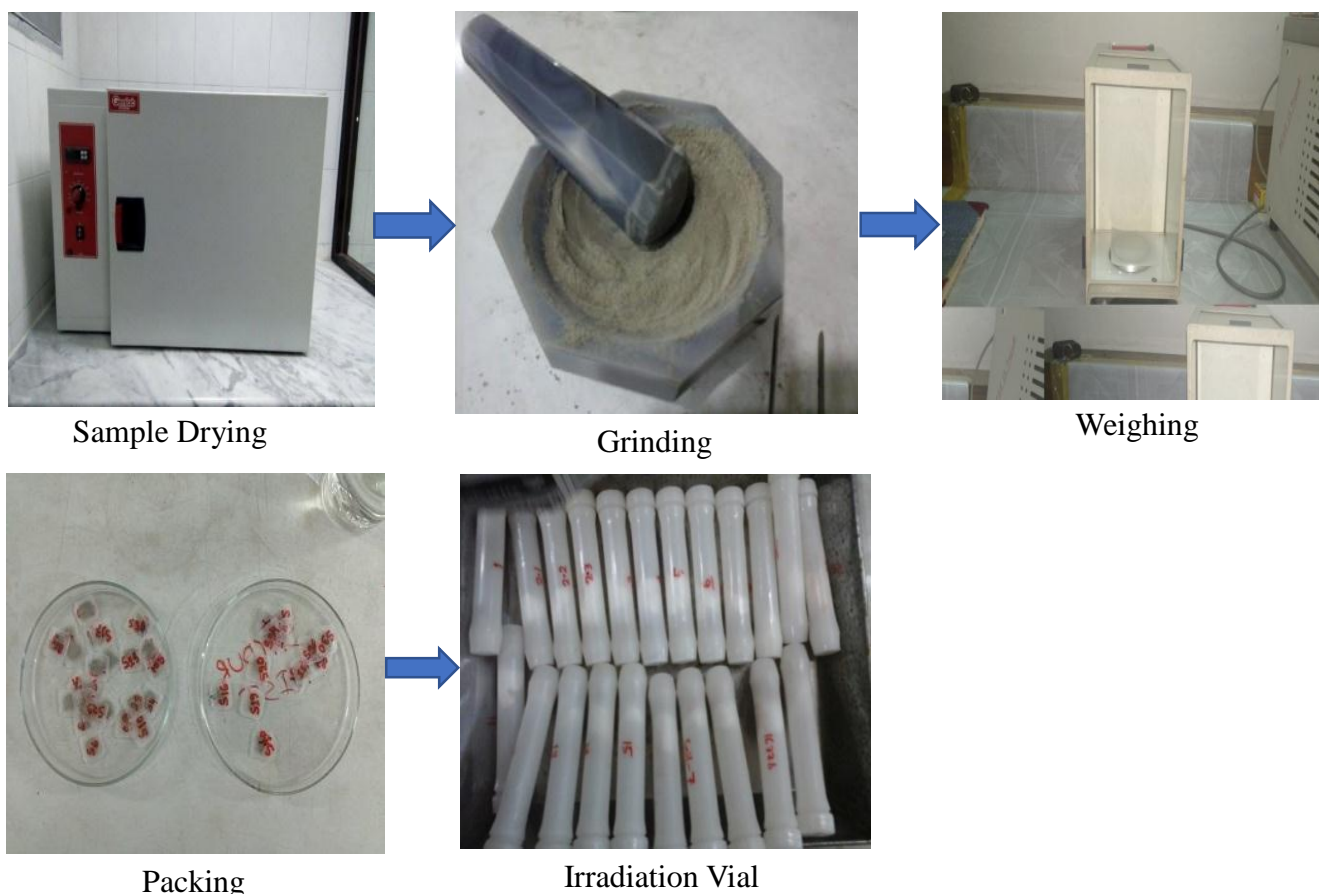


Figure 3. 4: Samples preparation at NAA lab before irradiation.

3.6 Samples analysis

3.6.1 Instrumental neutron activation analysis (INAA)

Two certified reference materials (CRMs): Soil-7 and IAEA-SL-1 (Lake Sediment), and one standard reference material: NIST-1633b (Coal Fly Ash), along with the inland and beach sand samples, were analyzed in this study. The relative standardization method of INAA was applied for the determination of elemental concentration. IAEA-Soil-7 was used as the standard, while IAEA-SL-1 and NIST-1633b were used as the control samples. The samples and standards were placed in a vial and irradiated using a pneumatic transfer (rabbit) system at the 3 MW TRIGA Mark-II research reactor of Bangladesh Atomic Energy Commission. Short-term and long-term types of irradiation were performed targeting radioactive nuclei with short and long-half-lives

3.6.2 Epithermal neutron activation analysis (ENAA)

To implement ENAA at TRIGA Mark II research reactor of BAEC, three reference standard materials: Certified reference material (CRM)-Soil-7 and SL-1 (lake sediment) from international atomic energy agency (IAEA) and standard reference material (SRM)-1633b from the national institute of standard and technology (NIST), USA were used. Two sets of reference materials-one set in Cd-covered box and another set in bare were prepared for neutron irradiation. About 50 mg of each reference material's powder sample was weighed in a polyethylene bag and double heat-sealed. The samples were then inserted into neutron irradiation vial.

3.7 Neutron sources

Neutron can be obtained from the reactor, accelerator and radio-isotopic neutron emitters.

Accelerator Source

The source of the accelerator is used to produce neutrons. The van-de Graff generator and cyclotron for the source of neutrons are seldom used. The electron accelerator, such as the Betatron, the microtone synchrotron, or a linear accelerator, produces high-energy brake radiation resulting in an (n, γ) reaction in the target. Such accelerator sources are used all over the world for regular nuclear research work.

Isotopic Source

In an activation analysis, there are essentially three kinds of isotopic sources used, photon neutron (n, γ) sources, alpha (n, n) sources and spontaneous source of fission. By using (n, γ) reaction on various targets, photon neutron sources are prepared. Deuteron and ^9Be isotopes (n, γ) are endoergic. Deuteron (^2H) has a 2.226 MeV threshold, while that of ^9Be is 1.66 MeV. Other isotopes used as a target for the preparation of sources of photon neutrons have a threshold greater than 6 MeV.

Reactor Source

The primary neutron source is the nuclear reactor that works in ^{235}U and ^{239}Pu on the theory of fission chain and is capable of generating the maximum neutron flux by far. The energy range of the fission neutron in the reactor is wide-ranging from 0.01 eV to 15 MeV, depending on the reaction form and the location within the reactor. Reactions are happened in the nuclear reactor by different neutron energies. Basically, three types of neutron energies are

1. Thermal neutron < **(0.5 eV)**
2. Epithermal neutron **(0.5 eV – 10 KeV)**
3. Fast neutron > **(0.5 MeV)**

3.8 TRIGA MARK-II research reactor

In this study, samples and standards were irradiated by 3 MW TRIGA MARK–II research reactor at the Atomic Energy Research Establishment (AERE), saver, Dhaka. A partial view of the 3 MW TRIGA MARK–II research reactor is shown in Figure 3.5. It is a multi-purpose reactor, capable of both steady-state and pulsing operation has been put into service in several disciplines since its commissioning. It is a light water-cooled graphite reflected reactor designed for continuous operation at a steady-state power of 3 MW. The detailed description of the components of TRIGA MARK –II research reactor is given below:



Figure 3. 5: TRIGA MARK-II research reactor at AERE, Saver, Dhaka (Source: www.usgs.gov).

Reactor Core

Each reactor consists of an active component in which the fission chain reaction is sustained and much of the core is released as fission energy. This active part is known as the core of the reactor, which required, contains the nuclear fuel and the moderator. There is a 0.63 cm thick wall with an inner diameter of 2 m and a depth of 8.2 m at the center of the TRIGA MARK II reactor. The cross-sectional view of the TRIGA MARK–II reactor core is shown in Figure 3.6. The core and reflector arrangement of the reactor is a cylinder with a diameter of about 1.1 m and a height of 0.89 m [49]. Figure 3.6 displays the inner view of the TRIGA reactor.

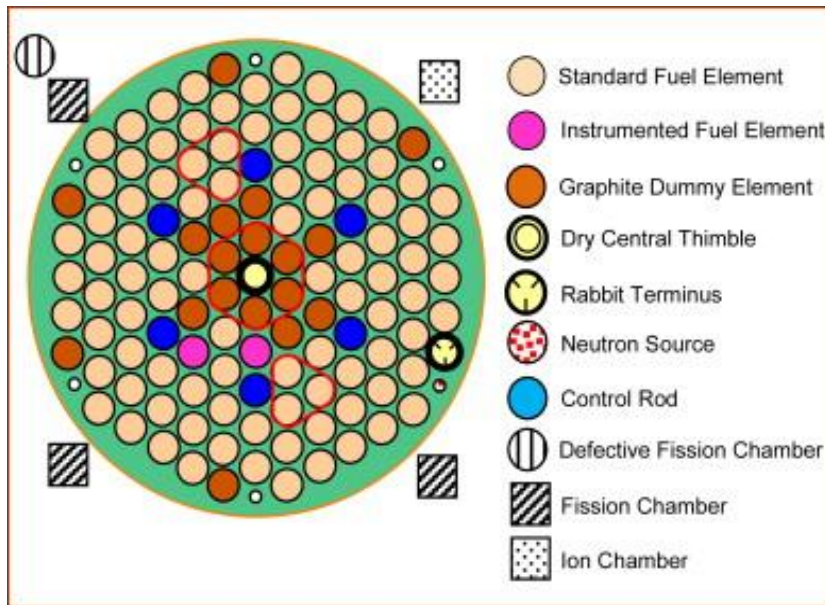


Figure 3.6: Schematic diagram of the cross-sectional view of TRIGA MARK-II reactor core (Source: www.sciencedirect.com).

Fuel

In the reactor core, a total of 100 fuel elements are present. The fuel is a solid homogeneous U-ZrH alloy mixture containing 20% U enriched to approximately 19.7% ^{235}U by weight and about 0.47% Erbium by weight. Each portion is clad with a 0.051 cm thick stainless steel can. The H/Zr ratio is around 1.6. The can, one above and one below the fuel, is of interest to two pieces of graphite to act as the top and bottom reflectors for the core.

Control Rods

The heat generated in the reactor is proportional to the fission rate, which depends on the neutron density of the core. Boron and cadmium, which have very high neutron absorption cross-sections, are mainly used as material for control rods. For ^{10}B , $\sigma_a = 3800$ barns (thermal) and ^{113}Cd , $\sigma_a = 20,000$ barns (thermal). The BAEC TRIGA MARK-II reactor is controlled by six boron carbide control rods. Each control rod (approximately 51 cm long) is a sealed aluminum tube containing powdered boron carbide as a neutron poison.

Reactor Tank

The reactor core is accommodated by the reactor tank (which is also called the pool liner). Near the bottom of the reactor tank, the reactor core is situated. The tank is made of a special aluminum alloy and is 8.23 m long and 1.98 m in diameter. It is filled with demineralized water of 24,865 liters.

Moderator

Most of the fission takes place from thermal neutrons in a reactor. The moderator's purpose is to slow the high energy neutrons through successive collisions to the level of thermal energy. Materials composed of elements with low mass numbers and low cross-sections for neutron absorption are the strongest moderators. Hydrogen, water, heavy water, beryllium or its oxide, carbon-like graphite, and hydrocarbons are examples. ZrH1.6 is primarily used as the moderator in the BAEC TRIGA MARK-II reactor. For that reason, light water is often used.

Reflector

A ring-shaped graphite block that radially surrounds the core is the reflector of the TRIGA reactors. It has a radial thickness of 30.5 cm, an inner diameter of 45.7 cm and a height of 55.6 cm. The graphite is covered by a leak-tight welded aluminum can from water penetration. The purpose of the reflector is to reduce the loss of core neutrons by reflecting some of the neutrons that appear to leak out or escape from the core.

Reactor shield

A neutron and gamma radiation source are a reactor. A reactor is often built within certain barriers capable of absorbing them to protect humans and the atmosphere subjected to these dangerous radiations, such barriers are primarily known as reactor shielding. A reinforced concrete frame, standing 7.9 m above the reactor hall level, is the reactor shield. Across the flats, the lower octagonal segment is 6.6 m. The beam ports are mounted through the concrete

shield and the reactor tank water in the shield system with tabular penetrations and end either at the reflector assembly or at the edge of the core of the reactor. At least 2.29 m of concrete with a minimum density of 2.75 g/cm³, 45.7 cm of water, 19 cm of graphite and 5 cm of lead are needed for the radial shielding of the heart.

3.9 Irradiation facilities of TRIGA MARK-II reactor

The TRIGA MARK-II reactor is designed to provide intense fluxes of ionizing radiation for research, training and isotope production. Pneumatic transfer system (Rabbit) and Dry central thimble (DCT) were used in this study. Experiments with the TRIGA reactor can be carried out using the following irradiation facilities:

1. Rotary specimen rack (Lazy Susan)
2. Pneumatic transfer system or Rabbit system
3. Dry central thimble (DCT)
4. Beam port facilities

3.10 Experimental conditions for ENAA and INAA

Irradiation facilities such as neutron flux, reactor power, irradiation time and counting time for ENAA and INAA technique are given in Table 3.2.

Table 3.2: Experimental conditions for ENAA and INAA.

Irradiation characteristics	ENAA	Short (INAA)	Long (INAA)
Irradiation facility	DCT	Rabbit	Rabbit
Neutron flux	$1.3 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$	$1.77 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$	$1.70 \times 10^{13} \text{ cm}^{-2} \text{ s}^{-1}$
Reactor power	500 KW	250 KW	2.4 MW
Irradiation time	60 minutes	1 min	7 min
Counting time	3600 – 10,000 s	300 – 600 s	1800 – 10,000 s

3.11 Neutron irradiation

3.11.1 Irradiation for ENAA

Two sets of standards were put into the irradiation vial and the vial was placed in the center of the reactor core for neutron irradiation. Three neutron flux monitors, Au-Al alloy (0.1% Au) at the bottom, middle and top of the stack of the samples were set to monitor neutron flux variation during irradiation. Samples were irradiated at dry central thimble (DCT) of the research reactor (shown in Figure 3.7) for 60 minutes at 500 KW power. The thermal and epithermal neutron fluxes at DCT were 3.01×10^{13} and $1.3 \times 10^{12} \text{ cm}^{-2}\text{s}^{-1}$, respectively.

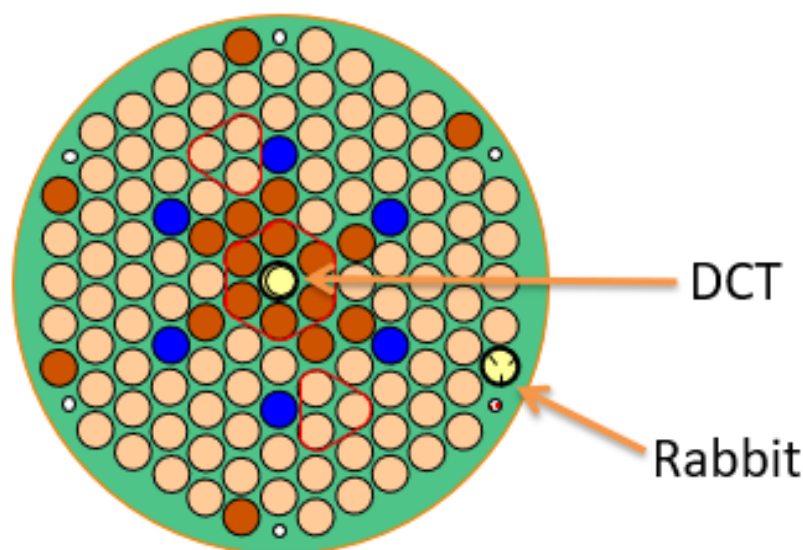


Figure 3.7: Core configuration and irradiation channels used for this study at BAEC research reactor (Source: www.sciencedirect.com).

Three days later the irradiation vial was carried out from the reactor core to reduce activity induced in the Cd filter as well as to decay short-lived radionuclides in the standard samples.

3.11.2 Irradiation for INAA

Two irradiation schemes were performed using a pneumatic transfer (rabbit) system at the 3 MW TRIGA MARK-II research reactor of Bangladesh Atomic Energy Commission, Savar. Two types of irradiation were used for the determination of short and long-lived radionuclides.

3.11.2.1 Short irradiation

Short irradiation was performed separately for each sample and standard with the thermal neutron flux of $1.77 \times 10^{12} \text{ cm}^{-2}\text{s}^{-1}$ for 1 min at 250 KW.

3.11.2.2 Long irradiation

Long irradiation was performed simultaneously with all the samples and standards with the thermal neutron flux of $1.70 \times 10^{13} \text{ cm}^{-2}\text{s}^{-1}$ for 7 min at 2.4 MW. In the case of long irradiation, to monitor neutron flux gradient within the sample stack, three IRMM530RA Al-0.1% Au (0.1 mm foil) monitor foils were also irradiated by placing them at the bottom, middle and top of the sample stack. After long irradiation, samples were turned into highly radioactive [50]. For this reason, they usually were not handled immediately. All samples and standards were put in a shielded place for 2 days. Figure 3.8 shows the irradiation of samples using the Pneumatic transfer system of TRIGA Research Reactor at AERE, Savar, Bangladesh.



Rabbit system at BTRR



Irradiated sample opening from the vial

Figure 3.8: Pneumatic irradiation facility of TRIGA Reactor at AERE, Savar, Dhaka.

3.12 High purity germanium (HPGe) detector system

Three types of detectors have been used to detect gamma-ray worldwide. These are- i) gas-filled detector, ii) scintillation detector and iii) semiconductor detector. The depletion depth of the semi-conductor type detector is less than 1 mm, which is adequate for charged particle spectroscopy or soft X-ray detection. For photon spectroscopy in the energy region of a hundred keV or several MeV, much thicker semiconductor detectors are needed. It needs a depletion depth of at least several cm. Ge has a much larger coefficient of linear attenuation due to its higher atomic number, which contributes to a shorter average free path. Ge is therefore chosen to achieve greater detection efficiency for hard X-ray or gamma-ray detection. HPGe crystal was first produced in the mid-1970s. Bulk germanium intended for the semiconductor industry is the starting material. The substance is further refined with the zone processing technique even though it is already of very high purity. In a crucible, the germanium is melted using radio frequency heating coils. The underlying theory is that in the process, impurities concentrate making the solid purer than the initial melt when a liquid freeze and appears solid.

HPGe gamma spectrometry system in the present experiment consists of the following parts.

- HPGe detector
- Digital gamma spectrometer
- Shielding arrangement

HPGe detector

Cryostat, Dewar and pre-amplifiers are the key components of an HPGe detector. While in theory, silicon detectors can work at room temperature, it is impossible to operate germanium detectors at room temperature due to the small band-gap energy that causes an unacceptable leakage current. Therefore, to lower the leakage current, germanium crystals must be cooled. When they are not in use, HPGe detectors are permitted to warm to room temperature, but they

are still held at the LN2 temperature (liquid nitrogen at 196 ° C or 77 K). Figure 3.9 displays the HPGe detector system used to detect gamma-ray in this study.



Figure 3.9: HPGe detector used in this study at RNPDP, AERE, Savar, Dhaka.

3.13 Full specification of HPGe

As a whole, the following are the specification of the HPGe detector, which is used in the present study shows in Table 3.3.

Table 3.3: Specification of the HPGe detector.

Detector	HPGe
Detector model	GC 4020
Serial number	07037658
Crystal geometry	Closed-end coaxial
Crystal diameter	6.2 cm
Crystal length	5.7 cm

Crystal active volume	172 cm ³
Crystal/window distance	0.5 cm
Dewar capacity	30 liters
Cooling temperature of the Ge crystal	77 K
The energy resolution of the detector at 1332 keV of ⁶⁰ Co γ – rays	2 keV (specified by the manufacturer)
Relative efficiency	25%
Peak to Compton ratio	54:1

3.14 Some important parameters of HPGe detector system

Some important parameters of the gamma-ray spectrometry system used for this study are given below.

Energy calibration

Energy calibration of the HPGe detector system for this study was performed using ¹⁵²Eu point source. The ¹⁵²Eu point source was counted for 3 minutes using the detector. The peak energies of 121.78, 778.90 and 1408.08 keV and their corresponding channel numbers for the ¹⁵²Eu point source were put in the software and energy calibration of the HPGe detector was performed.

Measurement of energy resolution

A spectrum was acquired to measure the energy resolution of the HPGe detector system using a ⁶⁰Co source positioned 10 cm away from the surface of the detector. To minimize the background contribution, the source has been shielded by the lead castle. Two cascades of 1173.9 keV and 1333 keV γ -ray energy were released from ⁶⁰Co, separated by 159.1 keV. Table 3.3 contains the data used for calculating the resolution of the detector. The energy resolution curves of the HPGe detector are shown in Figures 3.10-3.11. FWHM value was

found to be 4 channels at 1173.9 keV peak. The energy resolution was calculated following the equation.

$$\begin{aligned} \text{Energy resolution} &= \text{FWHM} \times \frac{E_2 - E_1}{C_2 - C_1} \dots\dots\dots(3.1) \\ &= 4.5 \times \frac{1333 - 1173.9}{3417 - 3008} \text{ keV} \\ &= 1.75 \text{ keV} \end{aligned}$$

Where, $C_1 = 3008$, $C_2 = 3417$ (C_1 and C_2 are two peak channels corresponding to characteristic gamma-ray energies $E_1 = 1173.9$ keV and $E_2 = 1333$ keV released from ^{60}Co).

The energy resolution of the detector obtained in this experiment is 1.75 keV at 1333 keV gamma-ray line of ^{60}Co source.

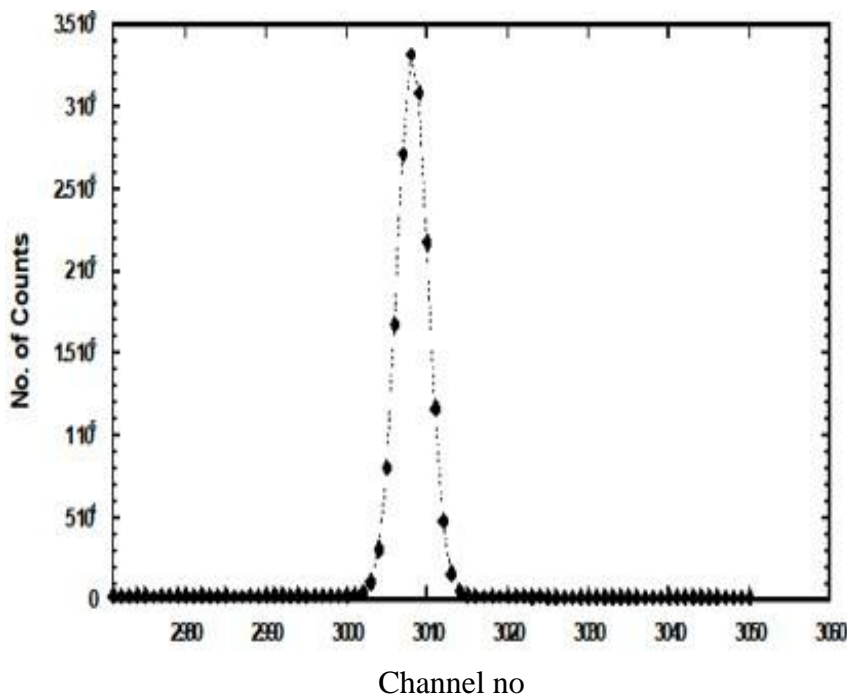


Figure 3.10: Energy resolution of the HPGe detector system (1173.9 keV).

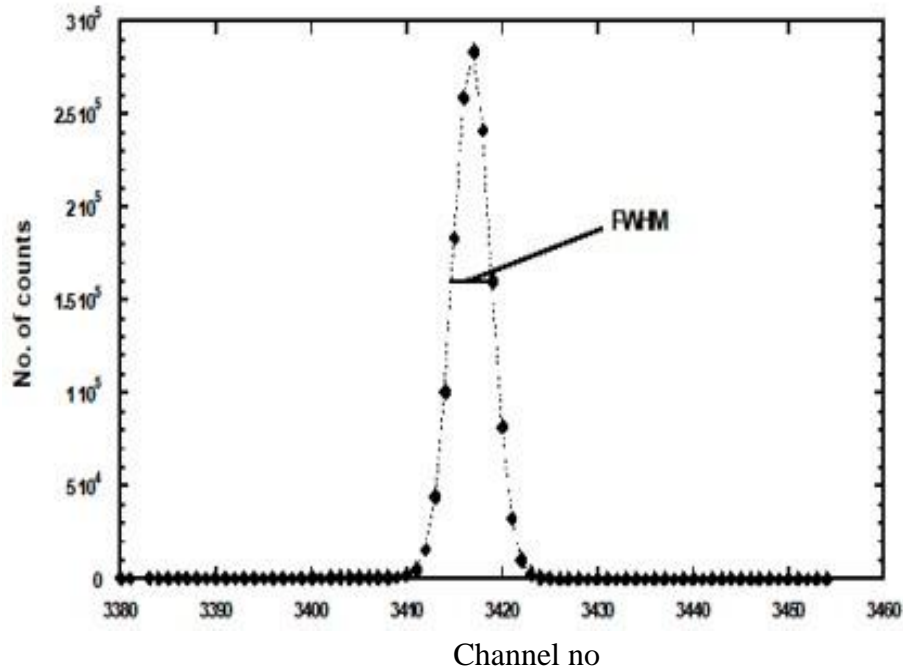


Figure 3.11: Energy resolution of the HPGe detector system (1332 keV).

Measurement of detector Efficiency

In this study, the comparative NAA method was used to determine elemental concentrations in the samples. Samples and standards have been calculated for this process under the same geometrical conditions. For that reason, the detector shows the same efficiency for a particular gamma peak for a sample and a standard that cancels out the effect of efficiency of the gamma-ray peaks at different energies. Therefore, in this study, the efficiency at different energies of the gamma-ray spectrometry system was not calculated.

3.15 Gamma spectra analysis

Using gamma spectrum analysis software (Hypermet PC version 5.12), the gamma peak analysis of samples and standards was carried out. The area under the peak of a gamma-ray from a radionuclide of interest in a gamma-ray spectrum represents the total count for that peak. That was placed into the excel sheet to measure the concentration of the corresponding element in the sample after obtaining the net count of a selected peak.

3.16 Gamma-ray spectrometry system

Gamma-ray spectrometry is a fast and non-destructive analytical technique that can be used in a sample to identify different radioactive isotopes. In gamma-ray spectrometry, a detector is used to determine the energy of incident gamma-rays. The identity of the emitter can be calculated by comparing the measured energy to the known energy of gamma-rays emitted by radioisotopes.

3.17 Gamma-ray counting for ENAA and INAA

After irradiation, the activities of irradiated standards and samples were measured using a high-resolution HPGe detector (resolution at FWHM is 1.88 keV at 1332.5 keV of ^{60}Co) coupled with a digital gamma-ray spectrometry system. For ENAA, the first and second gamma-ray counting was performed for 20 min and 1h after a decay time of 3 days and 7 days, while the third counting was performed for 2h after a decay time of 30 days.

In the case of INAA, for short irradiation, first counting was performed for 300 s after a decay time of about 300 s and second counting for 600 s after decay time of 2-3 h. For long irradiated samples, the first counting was performed for 60 min after a decay time of 2 days while the second counting was performed for 2h after a decay time of 7–10 days.

Short-lived and long-lived radionuclides were independently measured from the short and long irradiation. A PC-based HPGe detector coupled with a digital gamma spectrometry system was used to conduct the gamma spectrometry of all the irradiated samples and approved reference materials [51]. The data collection was carried out using Genie-2000 (Canberra) and MAESTRO-32 (ORTEC) software and the gamma peak analysis was carried out using the software Hypermet PC V.5.12 [51]. The gamma-ray spectrum analysis is shown in Figure 3.12.



Figure 3.12: Gamm - ray Peak Analysis using a computer.

3.18 Determination of elemental concentration

The elemental concentration of an element can be determined by the following two methods.

1. Absolute NAA method
2. Comparative NAA method

3.18.1 Absolute NAA method

In a neutron-induced reaction, the reaction rate varies based on the neutron flux. If the neutron flux increases, the neutron interaction will be improved. The activation rate is directly proportional to the neutron flux. It can be written as

$$\text{Activation rate} \propto \text{neutron flux } (\Phi)$$

The activation rate is also directly proportional to the number of target nuclei.

$$\text{Activation rate} \propto \text{number of nuclei present } (N)$$

The number of target nuclei present will depend on its isotopic abundance of the particular isotope of interest.

$$N = M N_A \frac{\theta}{W} \dots\dots\dots (3.2)$$

Here, M is mass in grams

N_A is Avogadro's number in atoms/mol

W is atomic weight in grams

θ is the isotopic abundance

The relationship between the activation rate, the number of target nuclei and the neutron flux (Φ in $n\text{ cm}^{-2}\text{s}^{-1}$) is expressed by the term "cross-section" (σ in barn):

$$\text{Activation rate} = \sigma \Phi N \dots\dots\dots(3.3)$$

Substituting $N = M N_A \frac{\theta}{W}$ into the expression for the activation rate (equation 3.3), it becomes:

$$\text{Activation rate} = \sigma \Phi M N_A \frac{\theta}{W} \dots\dots\dots(3.4)$$

If the nuclide of the product is radioactive, it will have a rate of decay which must be taken into account. The rate of decay of the nuclei is proportional to N^* .

$$\begin{aligned} \text{Decay rate } \frac{dN^*}{dt} &\propto -N^* \\ &= -\lambda N^* \dots\dots\dots(3.5) \end{aligned}$$

Where λ is the decay constant in s^{-1} .

If the equation is integrated between the limits N_0^* at time zero and N^* at time t . we can write as the following equation

$$N = N_0^* \exp (-\lambda t) \dots\dots\dots(3.6)$$

If the activation product is radioactive and decays with its characteristic half-life, the growth of the activity is governed by:

Production rate = activation rate – decay rate

$$\frac{dN^*}{dt} = \sigma \Phi N - \lambda N^* \dots\dots\dots(3.7)$$

$$N^* = \sigma \Phi N [1 - \exp (-\lambda t)] \dots\dots\dots(3.8)$$

The activity (Bq) or disintegration rate (A_0) at the end of the irradiation time t_i , then:

$$A_0 = \sigma \Phi N [1 - \exp (-\lambda t_i)] \dots\dots\dots(3.9)$$

Now, substituting $N = M N_A \frac{\theta}{W}$ into the above equation. It becomes

$$A_0 = \sigma \Phi M N_A \theta [1 - \exp (-\lambda t_i)] / W \dots\dots\dots(3.10)$$

In neutron activation analysis, the behavior of the radionuclide is typically calculated experimentally in a sample in order to deduce the unknown mass (M) of the substance.

Correction must be made for the decay period t_d and counting period t_c ,

Where,

Decay factor, $F_d = e^{-\lambda t_d}$ and

Counting factor, $F_c = \frac{1 - e^{-\lambda t_c}}{\lambda t_c}$

So, the basic equation for NAA calculation in absolute method can be written as

$$A_0 = \frac{MN_A\theta}{W} \sigma\varphi\{1 - e^{-\lambda t_i}\}\{e^{-\lambda t_d}\}\left\{\frac{1-e^{-\lambda t_c}}{\lambda t_c}\right\} \dots\dots\dots(3.11)$$

Hence,
$$M = \frac{A_0 W}{N_A \varphi \sigma \theta \{1 - e^{-\lambda t_i}\} \times F_d \times F_c} \dots\dots\dots(3.12)$$

All the factors on the right of the above equation are, in principle, known or can be measured. Thus, it can be possible to calculate the mass of the element [48].

3.18.2 Comparative NAA method

In the comparative NAA method, an element “x” in a sample and a known amount of the same element “x” as a standard are irradiated together and both sample and standard are counted under the same conditions by the same radiation detector. This procedure eliminates any uncertainty in the parameter σ , φ , λ and in the decay scheme and detection efficiency. The NAA equation by the comparative method is thus reduced to a simple form, as shown below

$$\frac{\text{Mass of element x in sample}}{\text{Mass of element x in standard}} = \frac{\text{Activity}(A_x) \text{ in sample} \times (e^{\lambda t_d})_{\text{sample}}}{\text{Activity}(A_x) \text{ in standard} \times (e^{\lambda t_d})_{\text{standard}}} \dots\dots\dots(3.13)$$

Knowing the activities of x in sample and standard, the sample and standard decay times and the mass of the element “x” in the standard, the mass of the element “x” in the sample is then calculated. A multi-elemental determination of 30 to 40 elements in the comparative method requires the use of certified reference material, whereas the absolute method requires only one standard [12].

3.19 Determination of activity concentration

The natural radioactivity in the surroundings is the main source of radiation exposure for human health. Natural radionuclide in soils and sands contributes a significant amount of background radiation exposure to the population through inhalation and ingestion.

The measured elemental concentrations are converted into activity concentration using the following equation [45].

$$F_E = \frac{M_E \cdot C \cdot A_E}{f_{A,E} \cdot N_A \cdot \lambda_E} \dots\dots\dots(3.14)$$

where F_E , M_E , λ_E , $f_{A,E}$ and A_E are the fraction of element E in the samples, the atomic mass in kg/mol, the decay constant in s^{-1} , the fractional atomic abundance in nature and the determined activity concentration in Bq/kg, respectively. N_A is Avogadro's number of 6.023×10^{23} atoms/mol, C is a constant with values of 10^6 for Th and U as well as 10^3 for potassium that converts the ratio of the elements to soils/sands mass into part per million or a percentage [44].

3.20 Calculation for uncertainty

In this investigation, measuring uncertainty is also an essential matter. It is a parameter associated to the measurement results that characterizes the dispersion of the values that may fairly be assigned to the calculated value. The sampling uncertainty was determined in accordance with the 1993 ISO Guide and applied to the measurement uncertainty [52]. The uncertainty results were added to the main result. Uncertainty component and its calculation have shown in Table 3.4.

Table 3.4: Origin and typical magnitudes of uncertainties in NAA.

Uncertainty Component	Origin	standard uncertainty	
U ₁	Sample and comparator preparation		
	U _{1a}	Mass determination of a sample	0.05 %
	U _{1b}	Mass determination of a comparator	0.2 %
	U _{1c}	Mass changes of samples due to moisture uptake during weighing	0.1 %
	U _{1d}	Blank variation and the necessary correction	0.5 %

		(due to analytical content in the irradiation vial)	
U ₂	Irradiation		
	U _{2a}	Irradiation geometry differences	0.10%
U ₃	Spectrometry measurement		
	U _{3a}	Counting statistics	Depend on spectrum
	U _{3b}	Counting geometry difference	3 %
	U _{3c}	Pulse-pileup losses (random coincidences)	0.5 %
	U _{3d}	Peak integration method	0.3 %

Where,

$$U_1 = \sqrt{\{(0.05)^2 + (0.2)^2 + (0.1)^2 + (0.5)^2\}} \% = 0.55\%$$

$$U_2 = 0.1\%$$

$$U_3 = \sqrt{\{(U_{3a})^2_{\text{sam}} + (U_{3a})^2_{\text{std}} + (U_{3b})^2 + (U_{3c})^2 + (U_{3d})^2_{\text{sam}} + (U_{3d})^2_{\text{std}}\}}$$

The total uncertainty calculation using the equation 3.15,

$$U_c = \sqrt{\{U_1^2 + U_2^2 + U_3^2\}} \% \dots\dots\dots(3.15)$$

This uncertainty is converted into absolute uncertainty as follows:

$$U_{\text{total}} = \frac{U_c * \text{Sample concentration}}{100} \dots\dots\dots(3.16)$$

The total uncertainty calculation of Uranium (U-238, 106 keV energy) for sample K3 (kuakata beach sands) has been shown in Appendix A.

CHAPTER 4

EXPERIMENTAL RESULTS AND OBSERVATIONS

In this study, research reactor-based Instrumental Neutron Activation Analysis (INAA) and Epithermal Neutron Activation Analysis (ENAA) techniques were applied to determine Th, U and other elemental concentrations in inland and beach sands of Bangladesh. ENAA method has never been applied at Bangladesh TRIGA Mark II Research Reactor (BTRR) at Atomic Energy Research Establishment (AERE), Savar, Bangladesh. In this research, ENAA was implemented for the first time at BTRR. Therefore, the advantages and disadvantages of ENAA over INAA are characterized first. To compare the advantages of ENAA over INAA, three parameters such as detection limit (DL), analytical sensitivity and Q_0 values can be used. In this study, the above three parameters were examined for the determination of As, Sb, Sm, Th and U using two standard reference materials IAEA-Soil-7 and IAEA-SL-1. The relevant nuclear data for the determination of these parameters are given in Table 4.1. ENAA is mainly used to determine a high Q_0 nuclides while a low Q_0 nuclides are acted as an interference [53].

Table 4.1: Relevant nuclear data for the determination of five elements (As, Sb, Sm, Th and U).

Elements	Product nuclide	Half-life	Energy(keV)
As [24, 54]	^{76}As	26.5 h	559
Sb [21, 54]	^{122}Sb	2.70 d	564
Sm[21, 54]	^{153}Sm	46.8 h	103
Th [21, 54]	^{233}Pa	27.0 d	312
U [21, 54]	^{239}Np	2.36 d	278

4.1 Detection limit

The detection limit represents the ability of a given analytical procedure to determine the minimum amounts of an element reliably. In this research, spectra were collected for ENAA and INAA under the same experimental conditions (mass =50 mg, Irradiation time = 60 min.

at DCT of BTRR, Decay time = 2 days; Counting time = 1200 s) are compared in Figure 4.1. It is observed that the ENAA spectrum has a lower background than the INAA spectrum. In NAA, the detection limit mainly depends on the irradiation, the decay time and the counting conditions. The DL was calculated using Currie's formula for the studied elements are tabulated in Table 4.2.

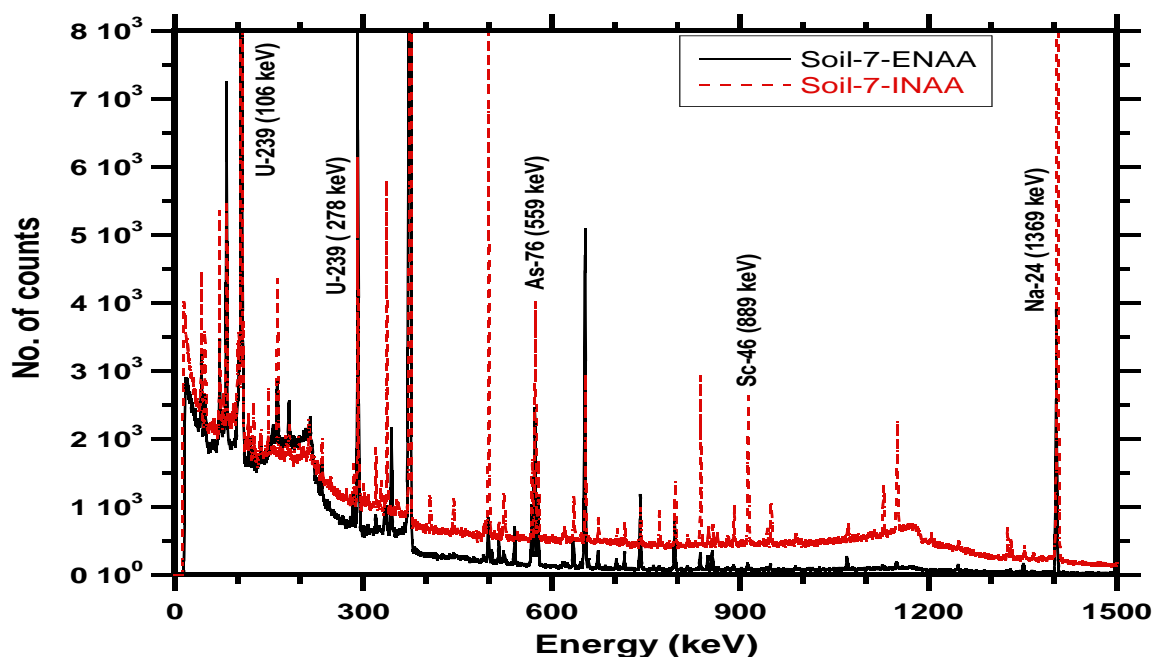


Figure 4.1: Comparison of ENAA and INAA spectra for IAEA-Soil-7 under the same experimental conditions.

Table 4.2: Comparison of detection limit (mg/kg) for ENAA and INAA for IAEA-SL-1 and IAEA-Soil-7.

Elements	IAEA-SL-1			IAEA-Soil-7		
	ENAA	INAA	Ratio*	ENAA	INAA	Ratio*
As	0.29	0.35	0.83	0.27	0.38	0.71
Sb	0.070	0.120	0.54	0.090	0.120	0.75
Sm	1.00	1.39	0.72	0.88	1.28	0.69
Th	0.48	0.31	1.55	0.51	0.25	2.04
U	0.40	0.53	0.75	0.34	0.52	0.65

Ratio* = DL of ENAA/DL of INAA.

From Table 4.2, it is observed that the DL of As, Sb, Sm and U for SL-1 obtained by ENAA are 17%, 46%, 28% and 25%, respectively lower than those obtained by INAA, whereas for

Th, the DL is 55% higher than the value of INAA. For Soil-7, the values for As, Sb, Sm and U obtained by ENAA are 29%, 25%, 31% and 35%, respectively lower than those obtained by INAA, whereas for Th, the DL is 104% higher than the value of INAA. Lower detection limits obtained by ENAA indicate better improvement of ENAA to determine studied elemental concentrations than those of INAA. Because a lower detection limit means the ability of a technique to determine fewer amounts of an element and hence improvement in detection by that technique.

4.2 Analytical sensitivity

The analytical sensitivity (count per sec./unit mass) data using two methods are tabulated in Table 4.3.

Table 4.3: Comparison of analytical sensitivity (cps/g) for ENAA and INAA in the case of SL-1 and Soil-7.

Elements	IAEA-SL-1			IAEA-Soil-7		
	ENAA	INAA	Ratio*	ENAA	INAA	Ratio
As	9.86E7	7.52E7	1.31	8.44E7	6.78E7	1.25
Sb	2.06E9	1.89E9	1.09	2.09E9	1.89E9	1.11
Sm	0.30E9	0.17E9	1.75	0.21E9	0.18E9	1.15
Th	0.50E9	1.44E9	0.35	0.47E9	1.37E9	0.34
U	0.50E9	0.39E9	1.28	0.52E9	0.35E9	1.48

*Ratio = Analytical sensitivity of ENAA/INAA

From Table 4.3, it is observed that for SL-1, the analytical sensitivities (cps/g) of As, Sb, Sm and U determined by ENAA are 31%, 9.0%, 75% and 28%, respectively higher than those obtained by INAA, whereas the value for Th is 65% lower than the value of INAA. For Soil-7, the values of the analytical sensitivities of As, Sb, Sm and U determined by ENAA are 25%, 11%, 15% and 48%, respectively higher than those obtained by INAA, whereas the value for Th is 66% lower than the value of INAA. In terms of detection limit and analytical sensitivities, it can be concluded that ENAA is better than INAA for the determination of As, Sb, Sm and U in soil/sand samples.

4.3 Comparison of Q_0 values of ENAA with INAA

The ENAA is effective if the nuclide of interest has higher Q_0 ($Q_0 > 10$) values, where Q_0 value for a nuclide is defined as the ratio of its resonance integral (I_0) to its thermal neutron capture cross-section (σ_0) [54, 55]. The Q_0 values for ^{76}As , ^{122}Sb , ^{153}Sm , ^{233}Pa (from ^{233}Th) and ^{239}Np (from ^{239}U) are calculated to 13.7, 20.3, 15.2, 11.5 and 102, respectively (shown in Table 4.4). These values indicate that activities of ^{76}As , ^{122}Sb , ^{153}Sm , ^{233}Pa and ^{239}Np will be reduced in ENAA. The use of epithermal neutron activation analysis (ENAA) is only helpful because all five isotopes have a high ratio (Q_0 values). So, activation products will make the gamma-ray spectrum simpler because of the reduction in activities in ENAA. Although Q_0 value for Th is theoretically 11.5 (more than 10), the practical values of DLs and analytical sensitivities do not show benefit for ENAA relative to INAA due to the presence of co-existing elements in the standards.

Table 4.4: Q_0 values for five elements (As, Sb, Sm, Th and U).

Elements	σ_0 (barn)	I_0 (barn)	Q_0 value
As[54]	4.6	63	13.7
Sb[21, 54]	6.25	127	20.3
Sm[21, 54]	206	3141	15.2
Th[21, 54]	7.26	83.7	11.5
U[21, 54, 55]	2.7	275	102

σ_0 = Thermal neutron capture cross section, I_0 = Epithermal neutron capture cross section,
 Q_0 value = Epithermal/thermal neutron capture cross section.

4.4 Determination of elements by INAA technique

In this study, total concentrations of 20 major, minor and trace elements (Na, Al, K, Sc, Ti, V, Cr, Mn, Fe, Co, As, Cs, La, Ce, Sm, Eu, Dy, Yb, Th and U) with special emphasis on Th and U in inland and beach sands of Bangladesh were determined by INAA technique.

Radionuclides with their half-lives and gamma-ray energies for the determination of studied elements by INAA are given in Table 4.5.

Table 4.5: Studied radionuclides with their half-lives and gamma-ray energies.

Elements	Product radionuclides	Half-life	Gamma-ray energy(keV)
Na ^a	²⁴ Na	14.7 h	1369
Al ^a	²⁸ Al	2.24 min	1779
K ^a	⁴² K	12.4 h	1525
Sc	⁴⁶ Sc	83.8 d	889, 1121
Ti ^a	⁵¹ Ti	5.76 min	320
V ^a	⁵² V	3.75 min	1434
Cr	⁵¹ Cr	27.7 d	320
Mn ^a	⁵⁶ Mn	2.58 h	847, 1811
Fe	⁵⁹ Fe	44.5 d	1099, 1292
Co	⁶⁰ Co	5.27 y	1173, 1332
As	⁷⁶ As	26.3 h	559
Cs	¹³⁴ Cs	2.06 y	796
La	¹⁴⁰ La	1.68 d	487, 1596
Ce	¹⁴¹ Ce	32.5 d	145
Sm	¹⁵³ Sm	46.7 h	103
Eu	¹⁵² Eu	13.3 y	122, 1408
Dy ^a	¹⁶⁵ Dy	140 min	94.7
Yb	¹⁷⁵ Yb	4.19 d	396
Th	²³³ Pa	27 d	312
U	²³⁹ Np	2.36 d	106, 278

^a Elements are determined by short irradiation

4.5 Quality control of the studied elements

In this research, three standards from International Atomic Energy Agency (IAEA)-Certified Reference Material (CRM)-Soil-7, IAEA-CRM-SL-1 (lake sediment) and National Institute of Standards and Technology (NIST)-Standard Reference Material (SRM)-1633b (Coal fly

ash) were identically prepared and analyzed along with the soil/sand samples. Analytical data quality of the studied 20 elements had been ensured by the repeated analysis of the reference materials IAEA-SL-1 and NIST-1633b. The average elemental concentration of those elements with standard deviation (SD, 1σ) and the certified/non-certified values for the studied elements are also tabulated in Table 4.6. Besides, to evaluate the laboratory performance we have determined three-parameters such as U-score, RB (relative bias) and the ratio of determined value to the certified value. U-score was calculated using the following equation [56].

$$U\text{-score} = \frac{|X_{lab} - X_{ref}|}{\sqrt{\mu_{lab}^2 + \sigma_{ref}^2}} \dots\dots\dots(4.1)$$

$$RB = \frac{X_{lab} - X_{ref}}{X_{ref}} \times 100 \dots\dots\dots(4.2)$$

where X_{lab} , μ_{lab} , X_{ref} , and σ_{ref} are the laboratory result, standard deviation, the reference value (certified/information) and uncertainty with the reference value, respectively. Based on the U-score value, the laboratory results will be satisfied if U-score is ≤ 1 [56].

It is noted that for the reference materials, the absolute U-score values are below 1.0 for most of the studied elements. Z-score values are not recorded for certain elements because of the unavailability of the uncertainty values with their non-certified values [8]. According to U-score parameters, the values are within 1.0 for most of the studied elements, which indicates satisfactory laboratory results. Moreover, deviations of the studied values from certified values for most of the elemental abundances for the repeated analysis of IAEA-SL-1 (n=3) and NIST-1633b (n=8) are less than 10%, which ensures the accuracy of the analysis.

Table 4.6: Elemental abundances (mg/kg or in %) in repeated analyses of IAEA-SL-1(n=3) and NIST-1633b (n=8) of this study along with the certificate values.

Elements	IAEA-SL-1 (n=3)					NIST-1633b (n=8)				
	Studied values	Certified values	U-score	RB %	Ratio	Studied values	Certified values	U-score	RB %	Ratio
Na%	0.173 ± 0.010	0.170 ± 0.005	0.268	1.76	1.02	0.202 ± 0.022	0.201 ± 0.003	0.045	0.498	1.00
Al%	9.66 ± 0.81	8.90 ± 0.18	0.917	8.54	1.09	14.4 ± 3.3	15.1 ± 0.3	0.201	-4.39	0.956
K%	1.46 ± 0.06	1.45 ± 0.04	0.162	0.759	1.01	1.80 ± 0.20	1.95 ± 0.03	0.755	-7.95	0.921
Sc	17.7 ± 1.5	17.3 ± 0.5	0.233	2.14	1.02	37.2 ± 3.4	41.0	-	-9.20	0.908
Ti%	0.451 ± 0.100	0.517 ± 0.028	0.635	-12.8	0.872	0.701 ± 0.165	0.791 ± 0.014	0.544	-11.4	0.886
V	186 ± 20	170 ± 4	0.808	9.41	1.09	309 ± 93	296 ± 4	0.137	4.33	1.04
Cr	108 ± 9	104 ± 6	0.386	4.04	1.04	190 ± 8	198 ± 5	0.884	-4.14	0.959
Mn	3419 ± 157	3460 ± 12	0.264	-1.20	0.988	124 ± 15	132 ± 2	0.495	-5.69	0.943
Fe%	7.26 ± 0.66	6.74 ± 0.03	0.786	7.67	1.08	7.37 ± 0.74	7.78 ± 0.23	0.533	-5.30	0.947
Co	20.2 ± 0.6	19.8 ± 0.4	0.491	1.92	1.02	47.0 ± 3.4	50.0	-	-5.96	0.940
As	30.1 ± 2.3	27.6 ± 0.4	1.05	8.99	1.09	130 ± 7	136 ± 3	0.830	-4.55	0.954
Cs	6.94 ± 0.41	7.00 ± 0.38	0.108	-0.857	0.991	9.52 ± 0.97	11.0	-	-13.4	0.866
La	49.1 ± 3.7	52.6 ± 0.5	0.949	-6.73	0.933	79.5 ± 7.8	94.0	-	-15.4	0.846
Ce	117 ± 19	117 ± 3	0.015	0.256	1.00	183 ± 20	190	-	-3.79	0.962
Sm	8.95 ± 0.47	9.25 ± 0.18	0.597	-3.24	0.968	16.5 ± 1.4	20.0	-	-17.6	0.825
Eu	1.70 ± 0.18	1.60 ± 0.10	0.472	6.00	1.06	3.69 ± 0.28	4.10	-	-9.98	0.900
Dy	6.89 ± 0.36	7.50 ± 0.53	0.963	-8.13	0.919	14.5 ± 1.9	17.0	-	-14.9	0.851
Yb	3.86 ± 0.61	3.42 ± 0.18	0.686	12.7	1.13	7.79 ± 0.97	7.60	-	2.43	1.02
Th	15.6 ± 1.7	14.0 ± 0.6	0.910	11.5	1.12	24.0 ± 2.8	25.7 ± 1.3	0.541	-6.46	0.935
U	4.18 ± 0.27	4.02 ± 0.28	0.425	4.05	1.04	8.27 ± 0.79	8.79 ± 0.36	0.598	-5.93	0.941

Ratio = Studied values/ Certified values.

4.6 Elemental abundances of inland surface sands of Bangladesh

The total elemental abundances of 20 major, minor and trace elements in surface inland sands of Bangladesh are tabulated in Tables 4.7 and 4.8. The studied elements are Na, Al, K, Sc, Ti, V, Cr, Mn, Fe, Co, As, Cs, La, Ce, Sm, Eu, Dy, Yb, Th and U. Mean concentration, minimum and maximum value, standard deviation (SD, 1σ), relative standard deviation (RSD, %) as well as the literature data [57] for the upper continental crust (UCC) are also given in Tables 4.7 and 4.8.

Table 4.7: Elemental concentrations (mg/kg or in %) in inland sands of Moulvibazar.

Elements	M1	M2	M3	M4	M5
Na%	0.250 ± 0.010	0.0681 ± 0.0029	0.638 ± 0.022	0.119 ± 0.005	0.146 ± 0.005
Al%	9.04 ± 0.29	5.10 ± 0.16	5.09 ± 0.16	4.33 ± 0.14	8.47 ± 0.27
K%	1.50 ± 0.09	0.688 ± 0.050	1.50 ± 0.09	1.64 ± 0.10	1.77 ± 0.10
Sc	9.30 ± 0.30	9.00 ± 0.30	9.36 ± 0.31	4.35 ± 0.15	16.3 ± 0.5
Ti%	0.364 ± 0.031	0.326 ± 0.027	0.299 ± 0.025	0.122 ± 0.014	0.492 ± 0.038
V	128 ± 8	78.3 ± 4.7	58.2 ± 3.6	38.6 ± 3.0	142 ± 8
Cr	61.8 ± 2.7	91.1 ± 3.84	79.9 ± 3.4	31.1 ± 1.5	121 ± 5
Mn	316 ± 4	104 ± 6	205 ± 3	142 ± 7	209 ± 5
Fe%	2.65 ± 0.10	3.00 ± 0.12	2.24 ± 0.09	1.62 ± 0.07	4.56 ± 0.17
Co	10.1 ± 0.5	6.30 ± 0.38	7.80 ± 0.45	3.07 ± 0.21	10.0 ± 0.5
As	6.10 ± 0.21	6.48 ± 0.23	6.35 ± 0.22	3.79 ± 0.14	3.91 ± 0.15
Cs	4.06 ± 0.22	5.38 ± 0.28	5.50 ± 0.29	2.63 ± 0.15	9.39 ± 0.45
La	29.4 ± 1.0	27.3 ± 0.9	35.4 ± 1.2	14.6 ± 0.5	46.4 ± 1.6
Ce	68.7 ± 2.5	64.4 ± 2.4	81.3 ± 3.0	42.0 ± 1.6	95.6 ± 3.4
Sm	4.26 ± 0.13	4.34 ± 0.14	5.90 ± 0.19	2.21 ± 0.07	7.62 ± 0.24
Eu	0.916 ± 0.100	0.854 ± 0.098	1.01 ± 0.11	0.598 ± 0.073	1.39 ± 0.14
Dy	3.62 ± 0.17	3.30 ± 0.16	4.39 ± 0.21	2.67 ± 0.14	6.12 ± 0.28
Yb	2.26 ± 0.13	2.79 ± 0.15	3.38 ± 0.18	1.17 ± 0.08	4.15 ± 0.22
Th	13.1 ± 0.5	13.4 ± 0.5	15.6 ± 0.6	9.73 ± 0.36	20.3 ± 0.7
U	2.29 ± 0.11	3.13 ± 0.15	2.92 ± 0.14	1.34 ± 0.07	3.70 ± 0.17

Table 4.7 continue

Elements	M6	M7	M8	M9
Na%	0.630 ± 0.022	0.0702 ± 0.0029	0.0785 ± 0.0032	0.120 ± 0.005
Al%	4.55 ± 0.15	3.19 ± 0.10	7.94 ± 0.26	7.23 ± 0.23
K%	1.50 ± 0.09	0.993 ± 0.065	1.55 ± 0.09	1.78 ± 0.11
Sc	6.91 ± 0.23	2.69 ± 0.09	15.0 ± 0.5	10.0 ± 0.3
Ti%	0.241 ± 0.022	0.183 ± 0.017	0.360 ± 0.030	0.261 ± 0.023
V	47.3 ± 3.2	30.1 ± 2.2	171 ± 10	74.2 ± 4.5

Cr	78.8 ± 3.4	17.9 ± 1.0	166 ± 7	49.4 ± 2.3
Mn	232 ± 8	115 ± 4	11.3 ± 14.0	248 ± 8
Fe%	1.85 ± 0.07	0.715 ± 0.034	9.61 ± 0.35	2.56 ± 0.10
Co	1.22 ± 0.12	0.338 ± 0.056	13.0 ± 0.7	6.53 ± 0.38
As	1.98 ± 0.09	2.73 ± 0.11	15.4 ± 0.5	10.6 ± 0.4
Cs	4.45 ± 0.24	1.53 ± 0.10	7.72 ± 0.38	5.41 ± 0.28
La	36.1 ± 1.2	9.34 ± 0.34	32.8 ± 1.1	29.0 ± 1.0
Ce	87.4 ± 3.2	30.0 ± 1.2	83.1 ± 3.0	79.3 ± 2.9
Sm	5.70 ± 0.18	1.57 ± 0.05	5.39 ± 0.17	4.83 ± 0.15
Eu	0.859 ± 0.096	0.232 ± 0.039	0.942 ± 0.103	0.968 ± 0.105
Dy	3.78 ± 0.18	1.38 ± 0.08	2.49 ± 0.13	3.86 ± 0.19
Yb	2.71 ± 0.15	0.85 ± 0.06	3.14 ± 0.17	2.99 ± 0.16
Th	16.0 ± 0.6	6.86 ± 0.26	19.5 ± 0.7	20.8 ± 0.8
U	2.31 ± 0.12	0.97 ± 0.06	4.57 ± 0.21	3.41 ± 0.16

Table 4.7 Continue

Elements	Min	Max	Mean	SD	RSD %	UCC
Na%	0.068	0.638	0.236	0.237	101	2.43
Al%	3.19	9.04	6.10	2.18	35.7	8.15
K%	0.688	1.78	1.44	0.395	27.5	2.32
Sc	2.69	16.3	9.22	4.76	51.6	14.0
Ti%	0.122	0.492	0.294	0.122	41.5	0.380
V	30.1	171	85.3	52.6	61.6	97.0
Cr	17.9	166	77.5	50.6	65.2	92.0
Mn	11.3	316	176	102	57.8	775
Fe%	0.72	9.61	3.20	3.03	94.7	3.92
Co	0.338	13.0	6.48	4.53	69.9	17.3
As	1.98	15.4	6.38	4.72	74.1	4.80
Cs	1.53	9.39	5.12	2.65	51.7	4.90
La	9.34	46.4	28.9	12.4	42.9	31.0
Ce	30.0	95.6	70.2	23.4	33.3	63.0
Sm	1.57	7.62	4.64	2.05	44.1	4.70
Eu	0.232	1.39	0.864	0.366	42.4	1.00
Dy	1.38	6.12	3.51	1.52	43.3	3.90
Yb	0.847	4.15	2.60	1.14	43.6	2.00
Th	6.86	20.8	15.0	5.06	33.7	10.5
U	0.974	4.57	2.74	1.24	45.2	2.70

For inland sands (Moulivabazar), among the studied elements most of the element's content do not vary over a long range (RSD: 27.5%–45.2%), whereas some element (Na, Sc, V, Cr, Mn, Fe, Co, As and Cs) contents vary relatively wide range (RSD: 51.6% - 101%) in the studied area. If we compare the mean concentrations of the studied elements with those of

upper continental crust (UCC), it is observed that the mean concentrations of As, Cs, Ce, Yb, Th and U show elevated values compared to UCC [57].

From Table 4.7, the mean concentrations of K, Th and U in the inland sands were found as 1.44%, 15.02 mg/kg and 2.74 mg/kg, respectively. However, the average abundance of Th is 43% greater than the UCC values. If the elemental concentrations of K, Th and U in sands are in the range of (0.3-4.5%), (3-30) mg/kg and (1-10) mg/kg, respectively, it represents the origin of typical rocks, crustal minerals and granite rock, respectively [58-60]. The results obtained from the studied area for U are the formation of granite rock (0.97-4.57 mg/kg of U), while the Th is the crustal minerals (6.86-20.8 mg/kg). Besides, the determined concentrations of K are in the range of typical rocks (0.69-1.78%).

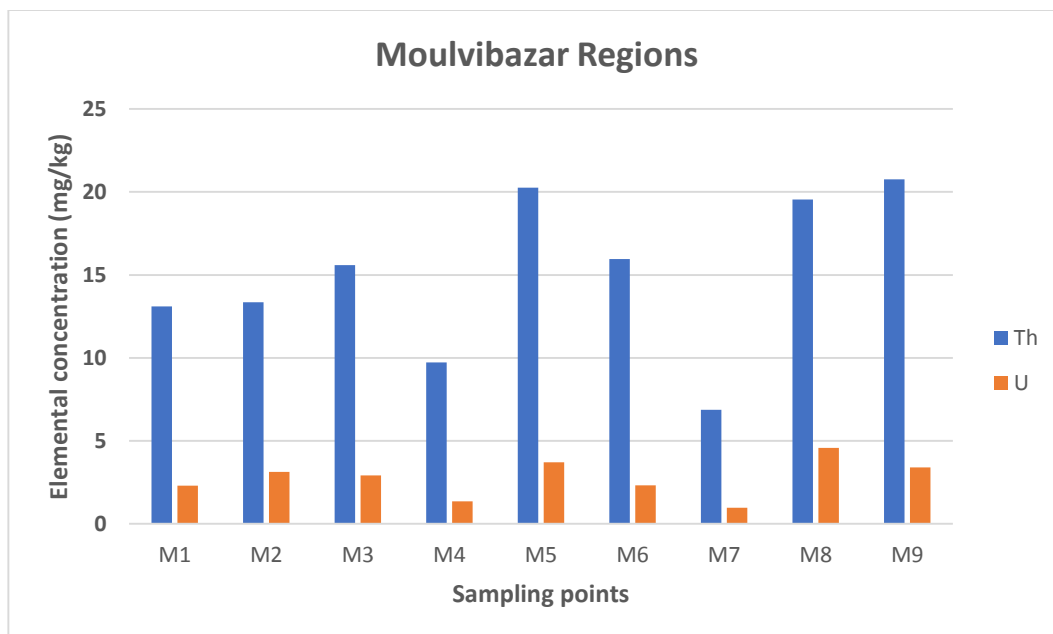


Figure 4.2: Elemental concentrations of Th and U in sands of Moulvibazar.

From Figure 4.2, it can be observed that the highest elemental concentrations for Th and U are found from the M9 and M8 sampling points with a value of 20.8 mg/kg and 4.57 mg/kg,

respectively, whereas the lowest values for both Th and U are obtained from the M7 sampling point with a value of 6.86 mg/kg and 0.97 mg/kg, respectively.

Table 4.8: Elemental concentrations (mg/kg or in %) in inlands sands of Sherpur, Rajshahi and Sylhet.

Elements	SH1	RAJ1	SH2	SYL1	SYL2
Na%	1.13 ± 0.04	0.871 ± 0.028	0.0167 ± 0.0001	1.10 ± 0.03	0.293 ± 0.009
Al%	4.39 ± 0.14	3.79 ± 0.12	0.943 ± 0.031	4.14 ± 0.13	2.23 ± 0.07
K%	1.48 ± 0.06	1.30 ± 0.05	0.283 ± 0.015	1.58 ± 0.06	1.53 ± 0.06
Sc	6.05 ± 0.20	5.56 ± 0.18	0.630 ± 0.027	1.14 ± 0.04	0.442 ± 0.020
Ti%	0.137 ± 0.012	0.0797 ± 0.0088	0.0361 ± 0.006	0.187 ± 0.015	0.104 ± 0.010
V	43.6 ± 2.2	18.2 ± 1.2	6.45 ± 0.63	34.0 ± 1.8	25.2 ± 1.5
Cr	24.9 ± 1.5	11.0 ± 0.9	211 ± 9	3.08 ± 0.40	3.99 ± 0.49
Mn	427 ± 14	515 ± 16	82.3 ± 2.8	301 ± 10	378 ± 12
Fe%	1.77 ± 0.07	1.42 ± 0.06	0.576 ± 0.030	0.295 ± 0.018	0.218 ± 0.016
Co	5.27 ± 0.32	3.08 ± 0.21	2.86 ± 0.20	0.92 ± 0.10	0.71 ± 0.09
As	2.45 ± 0.10	1.07 ± 0.05	1.82 ± 0.08	1.90 ± 0.08	4.12 ± 0.15
Cs	3.30 ± 0.18	3.32 ± 0.18	0.734 ± 0.063	1.04 ± 0.08	0.714 ± 0.062
La	36.3 ± 1.2	16.8 ± 0.6	2.28 ± 0.10	18.1 ± 0.6	6.34 ± 0.22
Ce	60.4 ± 2.4	23.9 ± 1.1	8.89 ± 0.55	6.11 ± 0.41	3.16 ± 0.30
Sm	6.05 ± 0.19	3.28 ± 0.10	1.86 ± 0.06	3.53 ± 0.11	1.51 ± 0.05
Eu	0.903 ± 0.084	0.441 ± 0.051	0.322 ± 0.042	0.315 ± 0.039	0.219 ± 0.033
Dy	4.60 ± 0.16	3.21 ± 0.12	10.5 ± 0.4	2.95 ± 0.11	1.78 ± 0.07
Yb	3.19 ± 0.17	3.49 ± 0.18	10.3 ± 0.5	2.34 ± 0.13	2.00 ± 0.11
Th	13.4 ± 0.5	5.06 ± 0.22	3.07 ± 0.15	1.05 ± 0.07	0.551 ± 0.047
U	2.85 ± 0.10	1.36 ± 0.05	2.96 ± 0.11	1.68 ± 0.06	1.54 ± 0.06

Table 4.8 continue

Elements	Min	Max	Mean	SD	RSD	UCC
Na	0.0167	1.13	0.682	0.502	73.5	2.43
Al	0.943	4.39	3.10	1.47	47.4	8.15
K	0.283	1.58	1.24	0.543	43.9	2.32
Sc	0.442	6.05	2.77	2.79	101	14.0
Ti	0.0361	0.187	0.109	0.0572	52.6	0.380
V	6.45	43.6	25.5	14.3	56.1	97.0
Cr	3.08	211	50.8	89.9	177	92.0
Mn	82.3	515	341	164	48.2	775
Fe	0.218	1.77	0.854	0.697	81.6	3.92
Co	0.715	5.27	2.57	1.86	72.2	17.3
As	1.07	4.12	2.27	1.14	50.4	4.80
Cs	0.714	3.32	1.82	1.37	74.9	4.90
La	2.28	36.3	16.0	13.2	82.8	31.0
Ce	3.16	60.4	20.5	23.7	116	63.0

Sm	1.51	6.05	3.24	1.79	55.3	4.70
Eu	0.219	0.903	0.440	0.271	61.5	1.00
Dy	1.78	10.5	4.61	3.45	74.9	3.90
Yb	2.00	10.3	4.27	3.43	80.4	2.00
Th	0.551	13.4	4.63	5.22	113	10.5
U	1.36	2.96	2.08	0.761	36.6	2.70

For inland sands (Sherpur, Rajshahi and Sylhet, shown in Table 4.8), among the studied elements following the elemental abundances (K, Al, Mn, As and U) do not vary over a long-range (RSD: 36.6% - 50.4%) in the studied area, whereas remaining the elements (Na, Sc, Ti, V, Cr, Fe, Co, Cs, La, Ce, Sm, Eu, Dy, Yb and Th) vary over a wide range (RSD: 52.6% – 177%).

If we compare the studied elements with the upper continental crust (UCC), it is observed that the mean concentrations of Dy and Yb show elevated values compared to UCC values [57].

From Table 4.8, the mean concentrations of K, Th and U in the inland sands were found as 1.24%, 4.63 mg/kg and 2.08 mg/kg, respectively. Besides, the mean abundances of Th and U are lower than those of UCC values. If the elemental concentrations of K, Th and U in sands are in the range of (0.3-4.5%), (3-30) mg/kg and (1-10) mg/kg, respectively, it represents the origin of typical rocks, crustal minerals and granite rock, respectively [58-60]. The results obtained from the studied area for U are the formation of granite rock, while the Th is the crustal minerals. Moreover, the determined concentrations of K are in the range of typical rocks.

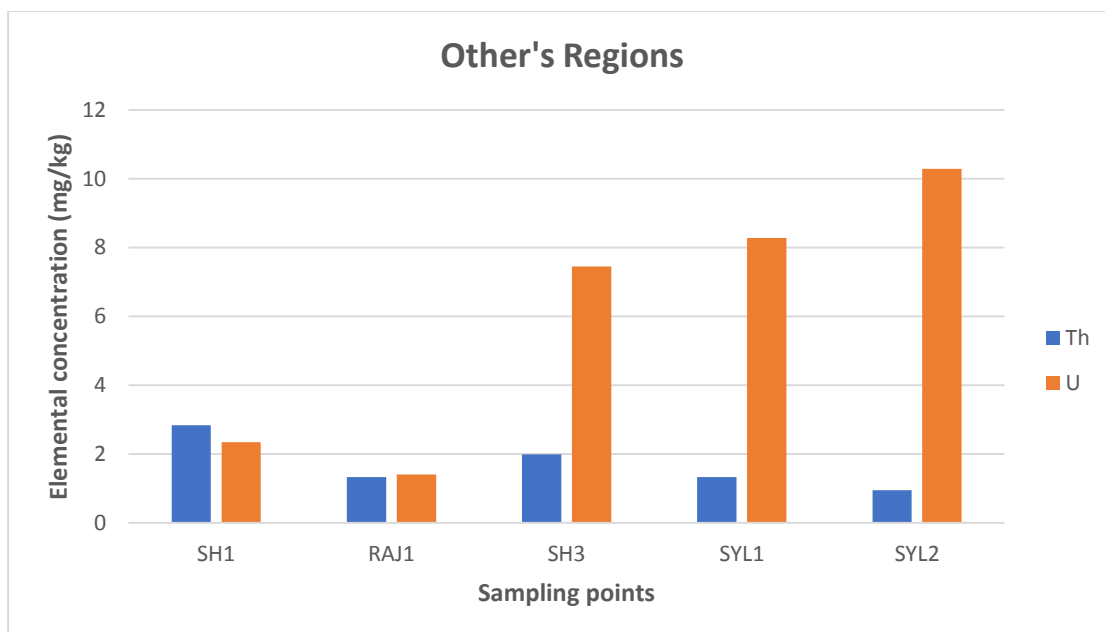


Figure 4.3: Elemental concentrations of Th and U in Other’s regions (Sherpur, Rajshahi and Sylhet).

From Figure 4.3, it can be observed that the maximum elemental concentrations of Th and U are obtained from the SH1 and SYL2 sampling points with a value of 2.83 mg/kg and 10.3 mg/kg, respectively, whereas the lowest values for Th and U are found from the SYL2 and RAJ1 sampling point with a value of 0.95 mg/kg and 1.40 mg/kg, respectively.

4.7 Elemental abundances of beach surface sands of Bangladesh

The total elemental abundances of 20 major, minor and trace elements in surface Beach sands of Bangladesh are tabulated in Tables 4.9 - 4.11. The studied elements are Na, Al, K, Sc, Ti, V, Cr, Mn, Fe, Co, As, Cs, La, Ce, Sm, Eu, Dy, Yb, Th and U. Mean concentration, minimum and maximum value, standard deviation (SD, 1σ), relative standard deviation (RSD, %) as well as the literature data [57] for the upper continental crust (UCC) are also given in Tables 4.9 - 4.11.

Table 4.9: Elemental concentrations (mg/kg or in %) in beach sands of Patenga, Chittagong.

Elements	P1	P2	P3	P4	P5
Na%	0.527 ± 0.019	0.687 ± 0.024	0.426 ± 0.015	0.744 ± 0.026	0.934 ± 0.032
Al%	3.07 ± 0.10	4.64 ± 0.15	2.98 ± 0.09	4.16 ± 0.13	4.38 ± 0.14
K%	1.39 ± 0.08	1.76 ± 0.11	1.08 ± 0.07	1.75 ± 0.10	2.04 ± 0.11
Sc	6.04 ± 0.21	8.54 ± 0.29	3.95 ± 0.14	4.86 ± 0.17	5.23 ± 0.18
Ti%	0.691 ± 0.043	0.368 ± 0.024	0.222 ± 0.016	0.245 ± 0.017	0.212 ± 0.015
V	52.9 ± 3.0	55.0 ± 2.8	32.5 ± 1.8	38.3 ± 2.1	38.8 ± 2.0
Cr	436 ± 21	66.2 ± 3.8	128 ± 6	43.7 ± 2.6	37.2 ± 2.3
Mn	469 ± 16	354 ± 12	193 ± 7	259 ± 9	309 ± 11
Fe%	1.89 ± 0.09	2.82 ± 0.13	1.46 ± 0.07	1.73 ± 0.08	2.00 ± 0.09
Co	5.64 ± 0.41	12.1 ± 0.7	4.76 ± 0.37	6.65 ± 0.45	8.51 ± 0.57
As	2.35 ± 0.12	4.18 ± 0.22	1.97 ± 0.10	2.07 ± 0.11	4.59 ± 0.22
Cs	1.19 ± 0.11	4.56 ± 0.28	1.80 ± 0.14	2.76 ± 0.18	3.51 ± 0.23
La	107 ± 4	27.8 ± 1.0	31.8 ± 1.2	31.7 ± 1.2	20.2 ± 0.7
Ce	237 ± 10	64.9 ± 3.0	79.6 ± 3.5	60.1 ± 2.7	45.9 ± 2.2
Sm	13.3 ± 0.4	4.50 ± 0.15	5.17 ± 0.17	4.74 ± 0.15	3.02 ± 0.10
Eu	1.74 ± 0.19	0.898 ± 0.110	1.03 ± 0.13	0.896 ± 0.106	0.709 ± 0.095
Dy	11.1 ± 0.5	3.29 ± 0.16	3.34 ± 0.16	3.66 ± 0.18	2.28 ± 0.12
Yb	7.92 ± 0.48	2.79 ± 0.20	2.67 ± 0.18	2.85 ± 0.19	1.48 ± 0.11
Th	85.3 ± 2.6	15.2 ± 0.7	14.2 ± 0.6	13.9 ± 0.5	14.8 ± 0.4
U	15.1 ± 0.6	2.31 ± 0.16	2.93 ± 0.19	3.65 ± 0.13	3.35 ± 0.09

Table 4.9 continue

Elements	P6	P7	P8	P9	P10
Na%	0.645 ± 0.023	0.579 ± 0.020	0.509 ± 0.018	0.498 ± 0.018	0.546 ± 0.019
Al%	3.23 ± 0.10	3.85 ± 0.12	2.82 ± 0.09	2.80 ± 0.09	2.78 ± 0.09
K%	1.41 ± 0.09	1.87 ± 0.10	1.08 ± 0.07	1.13 ± 0.07	1.53 ± 0.07
Sc	3.40 ± 0.12	4.77 ± 0.17	3.02 ± 0.11	3.40 ± 0.12	2.84 ± 0.10
Ti%	0.254 ± 0.018	0.233 ± 0.016	0.266 ± 0.019	0.286 ± 0.020	0.295 ± 0.022
V	28.8 ± 1.7	38.7 ± 2.0	28.5 ± 1.8	33.1 ± 1.9	27.9 ± 1.9
Cr	59.5 ± 3.3	63.1 ± 3.5	73.3 ± 4.0	216 ± 11	52.4 ± 3.1
Mn	241 ± 9	320 ± 11	180 ± 7	232 ± 8	179 ± 7
Fe%	1.14 ± 0.06	1.70 ± 0.08	1.02 ± 0.05	1.27 ± 0.06	0.942 ± 0.051
Co	4.27 ± 0.33	6.26 ± 0.45	4.42 ± 0.35	4.77 ± 0.37	4.33 ± 0.35
As	2.35 ± 0.12	3.68 ± 0.18	1.68 ± 0.09	2.10 ± 0.11	2.10 ± 0.12
Cs	1.46 ± 0.12	2.30 ± 0.17	1.22 ± 0.11	1.38 ± 0.12	1.71 ± 0.14
La	33.3 ± 1.2	25.9 ± 0.9	33.9 ± 1.2	29.0 ± 1.1	20.1 ± 0.8
Ce	65.4 ± 2.9	59.2 ± 2.7	72.0 ± 3.2	75.8 ± 3.4	47.9 ± 2.3
Sm	4.09 ± 0.13	4.24 ± 0.14	5.13 ± 0.17	7.02 ± 0.23	3.87 ± 0.13
Eu	0.780 ± 0.099	0.759 ± 0.098	0.826 ± 0.107	0.928 ± 0.115	0.493 ± 0.074
Dy	3.78 ± 0.18	2.66 ± 0.13	2.98 ± 0.15	3.16 ± 0.15	2.31 ± 0.12

Yb	2.33 ± 0.16	1.26 ± 0.10	2.37 ± 0.16	1.92 ± 0.14	1.44 ± 0.11
Th	15.2 ± 0.6	16.2 ± 0.5	40.0 ± 1.6	12.7 ± 0.5	12.1 ± 0.4
U	2.15 ± 0.14	4.56 ± 0.11	4.01 ± 0.25	2.76 ± 0.18	2.75 ± 0.13

Table 4.9 continue

Elements	Min	Max	Mean	STD	RSD%	UCC
Na%	0.426	0.934	0.609	0.149	24.4	2.43
Al%	2.78	4.64	3.47	0.714	20.6	8.15
K%	1.08	2.04	1.50	0.345	22.9	2.32
Sc	2.84	8.54	4.61	1.73	37.6	14.0
Ti%	0.212	0.691	0.307	0.142	46.3	0.380
V	27.9	55.0	37.4	9.66	25.8	97.0
Cr	37.2	436	118	124	106	92.0
Mn	179	469	274	91.7	33.5	775
Fe%	0.942	2.82	1.60	0.565	35.4	3.92
Co	4.27	12.1	6.18	2.49	40.3	17.3
As	1.68	4.59	2.71	1.04	38.2	4.80
Cs	1.19	4.56	2.19	1.12	51.0	4.90
La	20.1	107	36.0	25.3	70.3	31.0
Ce	45.9	237	80.8	56.1	69.4	63.0
Sm	3.02	13.28	5.51	2.93	53.1	4.70
Eu	0.493	1.74	0.906	0.328	36.2	1.00
Dy	2.28	11.1	3.86	2.59	67.3	3.90
Yb	1.26	7.92	2.70	1.92	71.1	2.00
Th	12.1	85.3	23.9	23.1	96.3	10.5
U	2.15	15.1	4.36	3.85	88.3	2.70

For Patenga beach sand, half of the elemental abundances (Na, Al, K, Sc, Ti, V, Mn, Fe, Co, As and Eu) among the studied elements do not vary over a long-range (RSD: 20.6% - 46.3%) in the studied surface area, whereas remaining the elements (Cr, Cs, La, Ce, Sm, Dy, Yb, Th and U) content vary over a wide range (RSD: 33.5% - 106%). Besides, it is observed that the mean concentrations of Cr, La, Ce, Sm, Th and U show elevated values compared to UCC [57].

From Table 4.9, the average concentrations of K, Th and U in the Patenga beach sands were found as 1.51%, 23.9 mg/kg and 4.36 mg/kg, respectively. From the ranges of Th and U, it can be concluded that those elements are also obtained from the crustal minerals and granite rocks [58, 59], while the K abundances are in the typical rocks [60].

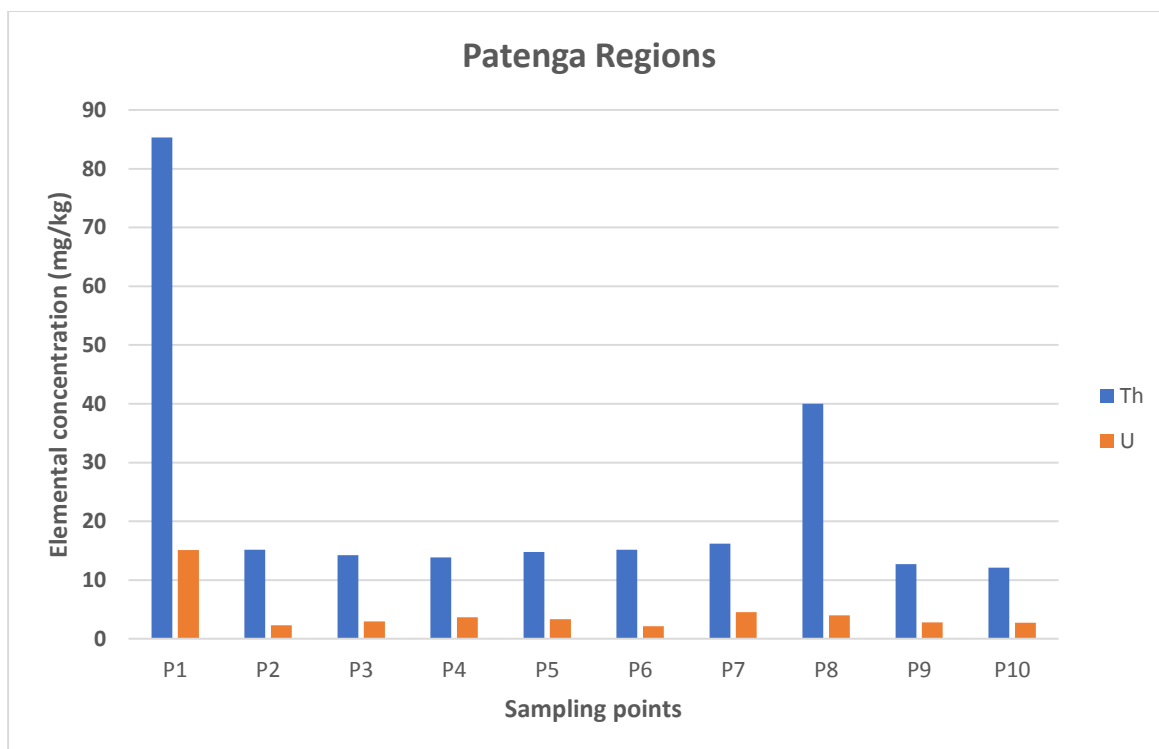


Figure 4.4: Elemental concentration of Th and U in sands of Patenga beach.

The maximum elemental concentration of both Th and U with a value of 85.3 mg/kg and 15.1 mg/kg, respectively, are observed at the P1 sampling point, whereas the lowest value of those elements is recorded at the P10 and P6 sampling points, respectively

Table 4.10: Elemental concentrations (mg/kg or in %) in beach sands of Cox's Bazar, Chittagong.

Elements	C1	C2	C3	C4	C5
Na%	0.560 ± 0.018	0.399 ± 0.014	0.570 ± 0.020	0.412 ± 0.015	0.628 ± 0.022
Al%	2.85 ± 0.09	3.20 ± 0.10	3.13 ± 0.10	4.55 ± 0.15	3.45 ± 0.11
K%	1.06 ± 0.07	0.939 ± 0.064	1.35 ± 0.09	0.651 ± 0.046	1.30 ± 0.08
Sc	4.31 ± 0.15	10.9 ± 0.4	4.94 ± 0.17	14.7 ± 0.5	3.27 ± 0.12
Ti%	0.148 ± 0.012	0.233 ± 0.017	0.231 ± 0.017	1.03 ± 0.06	0.0878 ± 0.0082
V	36.4 ± 1.9	52.4 ± 2.7	35.3 ± 2.0	114 ± 5	28.2 ± 1.6
Cr	21.5 ± 1.6	143 ± 7	27.5 ± 1.8	272 ± 12	21.6 ± 1.5
Mn	427 ± 15	699 ± 23	491 ± 16	1412 ± 47	512 ± 18
Fe%	1.51 ± 0.07	3.07 ± 0.14	2.20 ± 0.10	3.97 ± 0.16	1.20 ± 0.06
Co	5.66 ± 0.42	10.8 ± 0.7	7.59 ± 0.49	10.8 ± 0.58	5.74 ± 0.39
As	8.37 ± 0.40	5.40 ± 0.49	7.98 ± 0.57	6.63 ± 0.25	7.26 ± 0.10
Cs	1.85 ± 0.15	1.63 ± 0.13	2.06 ± 0.14	1.19 ± 0.09	1.04 ± 0.34
La	26.7 ± 1.0	91.5 ± 3.5	17.9 ± 0.8	122 ± 4	9.11 ± 0.91
Ce	54.5 ± 2.6	233 ± 10	47.4 ± 2.2	223 ± 8	15.5 ± 0.1

Sm	4.63 ± 0.15	11.6 ± 0.4	3.39 ± 0.12	18.0 ± 0.6	1.69 ± 0.02
Eu	1.02 ± 0.13	1.84 ± 0.19	0.907 ± 0.104	2.15 ± 0.09	0.460 ± 0.100
Dy	2.93 ± 0.14	5.71 ± 0.25	2.57 ± 0.13	13.6 ± 0.58	1.81 ± 0.01
Yb	2.52 ± 0.17	3.97 ± 0.32	1.12 ± 0.12	10.4 ± 0.6	0.804 ± 0.127
Th	8.06 ± 0.37	34.9 ± 1.4	64.0 ± 0.5	382 ± 3	6.63 ± 0.19
U	1.23 ± 0.09	2.90 ± 0.26	8.74 ± 0.17	132 ± 1	2.44 ± 0.06

Table 4.10 continue

Elements	C6	C7	C8	C9	C10
Na%	0.697 ± 0.024	0.451 ± 0.016	0.668 ± 0.023	0.613 ± 0.022	0.814 ± 0.028
Al%	3.35 ± 0.11	2.88 ± 0.09	3.27 ± 0.11	3.22 ± 0.10	3.32 ± 0.11
K%	1.17 ± 0.07	1.49 ± 0.05	1.35 ± 0.08	1.29 ± 0.08	1.03 ± 0.07
Sc	3.76 ± 0.13	4.26 ± 0.15	3.41 ± 0.18	1.51 ± 0.06	4.33 ± 0.15
Ti%	0.0869 ± 0.0080	0.214 ± 0.016	0.146 ± 0.013	0.0803 ± 0.0079	0.149 ± 0.012
V	31.3 ± 1.7	32.9 ± 1.8	25.8 ± 1.6	27.9 ± 1.6	36.5 ± 2.0
Cr	18.1 ± 1.3	18.0 ± 1.3	22.4 ± 3.8	10.0 ± 1.0	34.6 ± 2.2
Mn	473 ± 16	344 ± 12	463 ± 15.9	361 ± 12	468 ± 16
Fe%	1.50 ± 0.07	1.30 ± 0.06	1.70 ± 0.15	0.595 ± 0.035	1.57 ± 0.07
Co	6.56 ± 0.41	5.54 ± 0.37	8.33 ± 1.01	2.81 ± 0.24	6.38 ± 0.41
As	8.36 ± 0.31	6.14 ± 0.24	12.0 ± 0.8	7.16 ± 0.27	8.01 ± 0.30
Cs	1.62 ± 0.12	2.04 ± 0.14	4.85 ± 0.54	0.960 ± 0.091	1.78 ± 0.13
La	14.3 ± 0.5	22.0 ± 0.8	10.5 ± 0.4	8.61 ± 0.33	15.7 ± 0.6
Ce	28.2 ± 1.4	45.3 ± 2.0	23.7 ± 2.7	8.68 ± 0.64	30.9 ± 1.5
Sm	2.30 ± 0.07	3.39 ± 0.11	1.84 ± 0.07	1.54 ± 0.05	3.07 ± 0.10
Eu	0.740 ± 0.035	0.575 ± 0.029	0.381 ± 0.046	0.187 ± 0.013	0.680 ± 0.033
Dy	2.03 ± 0.11	2.37 ± 0.12	1.75 ± 0.10	1.30 ± 0.08	2.36 ± 0.12
Yb	1.22 ± 0.10	0.18 ± 0.03	1.62 ± 0.23	0.680 ± 0.073	1.84 ± 0.15
Th	4.63 ± 0.22	13.5 ± 0.4	4.88 ± 0.25	5.28 ± 0.15	4.94 ± 0.24
U	1.91 ± 0.12	5.65 ± 0.07	2.56 ± 0.22	3.56 ± 0.08	1.94 ± 0.13

Table 4.10 continue

Elements	C11	C12	C13	C14	C15
Na%	0.608 ± 0.021	0.414 ± 0.015	0.565 ± 0.020	0.413 ± 0.015	0.621 ± 0.022
Al%	3.15 ± 0.10	3.97 ± 0.13	3.39 ± 0.11	5.33 ± 0.17	3.76 ± 0.12
K%	1.00 ± 0.07	1.56 ± 0.04	1.30 ± 0.08	0.658 ± 0.048	1.18 ± 0.07
Sc	3.66 ± 0.13	2.66 ± 0.10	3.80 ± 0.13	26.0 ± 0.9	7.41 ± 0.25
Ti%	0.101 ± 0.010	1.11 ± 0.07	0.213 ± 0.016	2.74 ± 0.16	0.385 ± 0.026
V	33.6 ± 2.0	118 ± 6	30.2 ± 1.7	216 ± 10	54.6 ± 2.7
Cr	21.0 ± 1.5	18.5 ± 1.4	22.2 ± 1.6	710 ± 31	72.6 ± 3.8
Mn	350 ± 12	1533 ± 51	464 ± 16	2896 ± 95	534 ± 18
Fe%	1.40 ± 0.07	0.797 ± 0.044	1.34 ± 0.07	8.69 ± 0.34	2.40 ± 0.10
Co	6.48 ± 0.42	8.07 ± 0.50	5.38 ± 0.37	17.4 ± 0.9	9.69 ± 0.56
As	5.75 ± 0.23	9.78 ± 0.37	7.83 ± 0.32	10.6 ± 0.4	10.9 ± 0.4
Cs	1.59 ± 0.12	1.96 ± 0.15	2.22 ± 0.16	1.23 ± 0.11	2.49 ± 0.16
La	10.8 ± 0.4	235 ± 8	15.4 ± 0.6	423 ± 14	47.4 ± 1.6

Ce	19.6 ± 1.1	63.9 ± 2.8	24.9 ± 1.3	781 ± 29	85.9 ± 3.6
Sm	1.96 ± 0.06	33.9 ± 1.1	3.04 ± 0.10	57.3 ± 1.8	8.91 ± 0.28
Eu	0.482 ± 0.026	0.420 ± 0.023	0.496 ± 0.026	5.11 ± 0.20	0.982 ± 0.045
Dy	1.74 ± 0.10	17.7 ± 0.8	2.59 ± 0.13	41.8 ± 1.7	7.64 ± 0.33
Yb	1.25 ± 0.11	10.5 ± 0.7	1.71 ± 0.14	34.6 ± 2.0	3.24 ± 0.22
Th	7.65 ± 0.22	39.3 ± 1.0	12.3 ± 0.2	176 ± 5	21.0 ± 0.8
U	2.75 ± 0.08	24.7 ± 0.7	2.12 ± 0.07	32.0 ± 1.3	3.79 ± 0.22

Table 4.10 continue

Elements	C16	C17	Min	Max	Mean	STD	RSD%	UCC
Na%	0.594 ± 0.021	0.835 ± 0.029	0.399	0.835	0.580	0.132	22.8	2.43
Al%	3.21 ± 0.10	3.75 ± 0.12	2.85	5.33	3.52	0.623	17.7	8.15
K%	0.978 ± 0.062	1.26 ± 0.07	0.65	1.56	1.15	0.256	22.3	2.32
Sc	4.99 ± 0.17	5.92 ± 0.20	1.51	26.0	6.46	5.97	92.4	14.0
Ti%	0.197 ± 0.015	0.313 ± 0.023	0.080	2.74	0.439	0.665	152	0.380
V	34.8 ± 1.9	48.1 ± 2.6	25.8	216	56.3	49.7	88.3	97.0
Cr	34.1 ± 2.1	79.8 ± 4.2	10.0	710	90.9	172	190	92.0
Mn	580 ± 20	617 ± 21	344	2896	743	651	87.6	775
Fe%	1.81 ± 0.08	1.97 ± 0.09	0.595	8.69	2.18	1.86	85.6	3.92
Co	7.57 ± 0.46	8.19 ± 0.51	2.81	17.4	7.82	3.20	40.9	17.3
As	10.8 ± 0.4	10.5 ± 0.4	5.40	12.0	8.44	2.00	23.7	4.80
Cs	1.67 ± 0.12	2.25 ± 0.16	0.960	4.85	1.91	0.873	45.7	4.90
La	51.4 ± 1.7	31.3 ± 1.1	8.61	423	67.8	108	160	31.0
Ce	93.7 ± 3.9	54.6 ± 2.5	8.68	781	108	185	172	63.0
Sm	6.91 ± 0.22	4.20 ± 0.13	1.54	57.3	9.87	14.7	149	4.70
Eu	0.797 ± 0.037	0.678 ± 0.034	0.187	5.11	1.05	1.16	110	1.00
Dy	3.25 ± 0.15	3.51 ± 0.17	1.30	41.8	6.74	10.1	150	3.90
Yb	2.13 ± 0.14	1.94 ± 0.14	0.175	34.6	4.69	8.28	177	2.00
Th	22.0 ± 0.9	15.6 ± 0.5	4.63	382	48.4	95.4	197	10.5
U	2.24 ± 0.13	4.49 ± 0.09	1.23	132	13.8	31.6	229	2.70

For Cox's Bazar beach sands, among the studied elements, most of the elemental abundances (Sc, Ti, V, Cr, Mn, Fe, La, Ce, Sm, Eu, Dy, Yb, Th and U) vary significantly over a long-range (RSD: 85.6% - 229%) in the sampling points, whereas some elements (Na, Al, K, Co, Cs and As) content do not vary over a wide range (RSD: 17.7% - 45.7%). When compared with UCC, it is observed that the mean concentration of As, La, Ce, Sm, Eu, Dy, Yb, Th and U show elevated values.

From Table 4.10, the average concentrations of K, Th and U in Cox's Bazar beach sands were found as 1.15%, 48.4 mg/kg and 13.8 mg/kg, respectively. The high contents of Th and U are

found in the Cox's Bazar area due to the presence of Th and U enriched heavy minerals in that area [61, 62]. Besides, the concentrations of K are in the range of the typical rocks [60].

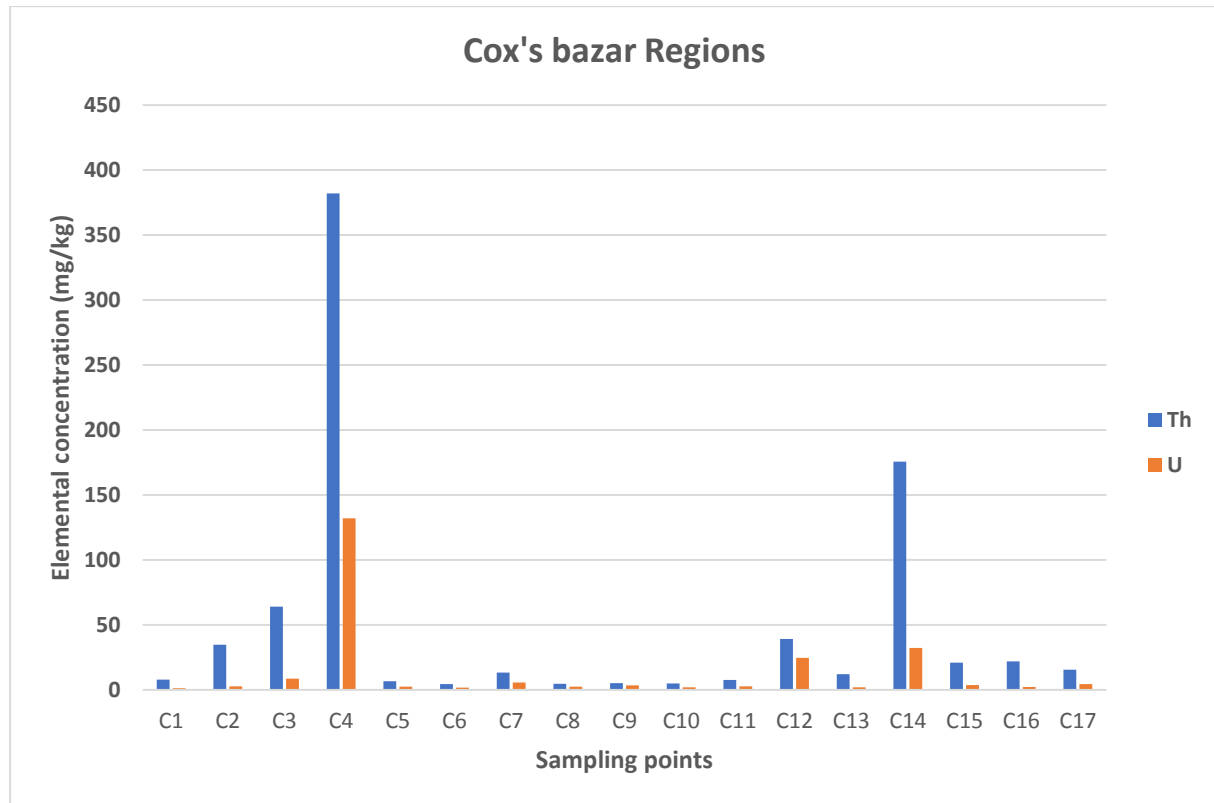


Figure 4.5: Elemental concentration of Th and U in sands of Cox's Bazar beach.

From the above Figure 4.5, it can be observed that the highest elemental concentration of both Th and U are found at the sampling point C4 with a value of 382 mg/kg and 132 mg/kg, respectively, whereas the lowest values are 4.63 mg/kg and 1.23 mg/kg, respectively, from the sampling points of C6 and C1.

Table 4.11: Elemental concentrations (mg/kg or in %) in beach sands of Kuakata.

Elements	K1	K2	K3	K4	K5
Na%	1.18 ± 0.04	1.11 ± 0.04	1.14 ± 0.04	1.10 ± 0.03	1.12 ± 0.04
Al%	3.85 ± 0.12	3.94 ± 0.13	4.30 ± 0.14	3.98 ± 0.13	3.91 ± 0.12
K%	1.20 ± 0.05	1.39 ± 0.06	1.45 ± 0.06	1.10 ± 0.04	1.07 ± 0.04
Sc	8.89 ± 0.29	5.65 ± 0.19	7.26 ± 0.24	6.53 ± 0.21	7.29 ± 0.24
Ti%	0.209 ± 0.017	0.117 ± 0.011	0.159 ± 0.014	0.236 ± 0.018	0.159 ± 0.013
V	50.8 ± 2.7	40.0 ± 2.2	52.5 ± 2.7	50.5 ± 2.5	43.6 ± 2.2
Cr	39.4 ± 2.0	26.1 ± 1.5	30.9 ± 1.7	27.2 ± 1.5	39.6 ± 2.1
Mn	358 ± 11	304 ± 10	401 ± 13	424 ± 14	306 ± 10
Fe%	1.96 ± 0.08	1.87 ± 0.08	2.80 ± 0.11	1.46 ± 0.06	1.70 ± 0.07
Co	6.63 ± 0.37	6.41 ± 0.36	10.5 ± 0.5	4.11 ± 0.26	5.10 ± 0.31
As	1.94 ± 0.08	4.56 ± 0.16	9.35 ± 0.32	1.59 ± 0.07	1.70 ± 0.07
Cs	2.01 ± 0.12	3.78 ± 0.20	4.54 ± 0.23	1.91 ± 0.12	1.92 ± 0.12
La	25.0 ± 0.8	17.6 ± 0.6	20.5 ± 0.7	36.5 ± 1.2	32.9 ± 1.1
Ce	56.8 ± 2.3	38.1 ± 1.60	42.6 ± 1.8	51.8 ± 2.09	63.9 ± 2.54
Sm	4.31 ± 0.14	2.80 ± 0.09	3.41 ± 0.11	5.76 ± 0.18	4.62 ± 0.15
Eu	1.09 ± 0.10	0.808 ± 0.077	0.879 ± 0.082	0.782 ± 0.075	1.11 ± 0.10
Dy	3.84 ± 0.14	2.45 ± 0.09	4.16 ± 0.15	6.42 ± 0.22	3.65 ± 0.13
Yb	2.27 ± 0.13	1.84 ± 0.11	3.43 ± 0.18	4.20 ± 0.22	1.77 ± 0.11
Th	11.9 ± 0.5	6.63 ± 0.27	8.41 ± 0.33	10.7 ± 0.4	12.9 ± 0.5
U	1.40 ± 0.05	0.729 ± 0.032	1.60 ± 0.06	1.65 ± 0.06	1.23 ± 0.05

Table 4.11 continue

Elements	K6	K7	K8	Min
Na%	1.09 ± 0.03	1.13 ± 0.04	1.08 ± 0.03	1.08
Al%	3.95 ± 0.13	4.02 ± 0.13	3.71 ± 0.12	3.71
K%	0.990 ± 0.041	1.09 ± 0.04	0.959 ± 0.040	0.959
Sc	8.94 ± 0.29	6.94 ± 0.23	6.74 ± 0.22	5.65
Ti%	0.227 ± 0.018	0.205 ± 0.017	0.138 ± 0.012	0.117
V	50.7 ± 2.6	47.6 ± 2.4	43.1 ± 2.2	40.0
Cr	36.9 ± 2.0	39.3 ± 2.1	25.6 ± 1.5	25.6
Mn	359 ± 11.5	293 ± 9	289 ± 9	289
Fe%	1.96 ± 0.08	1.71 ± 0.07	1.68 ± 0.07	1.46
Co	6.23 ± 0.35	5.28 ± 0.32	5.36 ± 0.32	4.11
As	1.98 ± 0.08	1.97 ± 0.08	2.38 ± 0.10	1.59
Cs	1.76 ± 0.11	1.65 ± 0.11	1.52 ± 0.10	1.52
La	36.3 ± 1.2	25.6 ± 0.8	30.0 ± 1.0	17.6
Ce	67.4 ± 2.7	45.7 ± 1.9	55.6 ± 2.3	38.1
Sm	5.24 ± 0.16	4.25 ± 0.13	5.18 ± 0.16	2.80
Eu	1.03 ± 0.09	0.863 ± 0.081	0.957 ± 0.088	0.782
Dy	4.34 ± 0.15	3.74 ± 0.13	3.35 ± 0.12	2.45
Yb	3.19 ± 0.17	2.89 ± 0.16	2.23 ± 0.13	1.77
Th	14.6 ± 0.6	8.10 ± 0.33	13.3 ± 0.5	6.63
U	1.57 ± 0.06	0.924 ± 0.039	1.65 ± 0.06	0.729

Table 4.11 continue

Elements	Max	Mean	STD	RSD%	UCC
Na%	1.18	1.12	0.0320	2.86	2.43
Al%	4.30	3.96	0.167	4.23	8.15
K%	1.45	1.16	0.179	15.5	2.32
Sc	8.94	7.28	1.13	15.6	14.0
Ti%	0.236	0.181	0.0439	24.3	0.380
V	52.5	47.3	4.59	9.70	97.0
Cr	39.6	33	6.32	19.1	92.0
Mn	424	342	51.8	15.2	775
Fe%	2.80	1.89	0.401	21.2	3.92
Co	10.5	6.21	1.93	31.1	17.3
As	9.35	3.18	2.67	83.7	4.80
Cs	4.54	2.39	1.12	47.1	4.90
La	36.5	28.1	7.05	25.1	31.0
Ce	67.4	52.7	10.2	19.4	63.0
Sm	5.76	4.45	0.982	22.1	4.70
Eu	1.11	0.940	0.127	13.5	1.00
Dy	6.42	4.00	1.14	28.4	3.90
Yb	4.20	2.73	0.850	31.2	2.00
Th	14.6	10.8	2.84	26.3	10.5
U	1.65	1.34	0.354	26.3	2.70

In the case of Kuakata beach sands, among the sampling points, most of the elemental abundances do not vary over a long-range (RSD: 2.86% - 47.1%) except As and the variation of As vary with a wide range (RSD: 83.7%). Moreover, the average concentration of Dy, Yb, and Th show enriched values compared to UCC [57]. This spatial variation of the trace elements for all studied sampling points can be attributed to variations in anthropogenic stresses, geo-chemical process, tidal settings and point/ non-point sources in that area [64].

From Table 4.11, the mean abundances of K, Th and U in Kuakata beach sands were found as 1.16%, 10.8 mg/kg and 1.34 mg/kg, respectively. The determined concentrations of K, Th and U are also corresponding to the formation of typical rocks, crustal minerals and granite rocks, respectively [58-60]. Moreover, the Variations of K, Th and U vary in the rocks due to the metamorphic process [63].

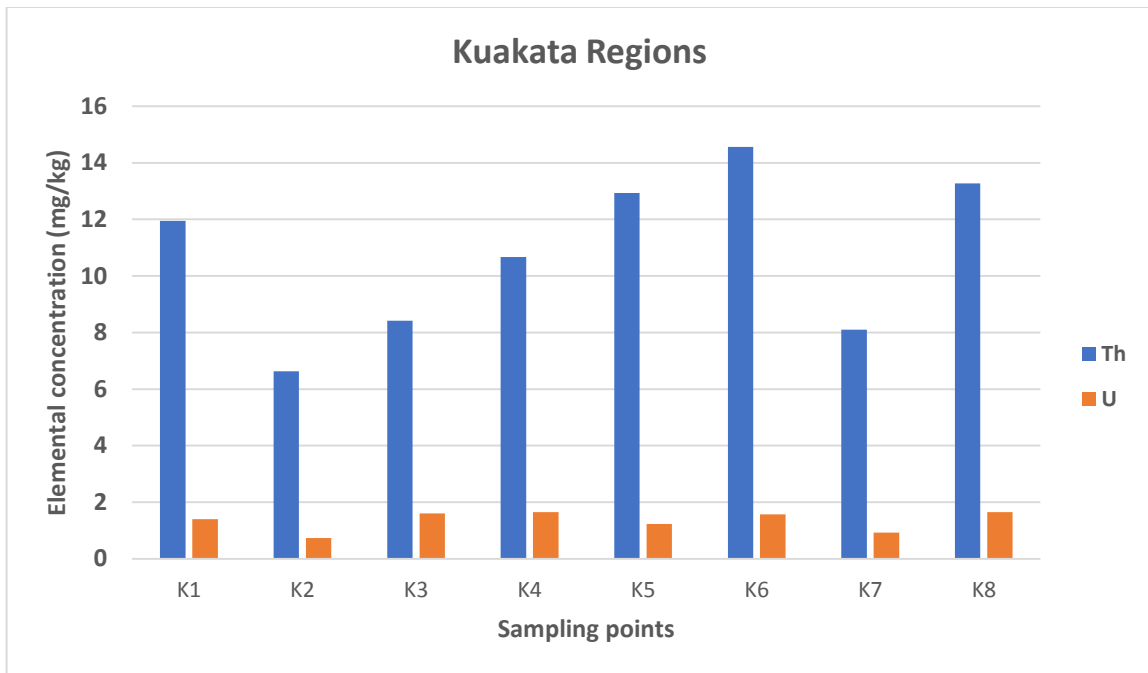


Figure 4.6: Elemental concentration of Th and U in sands of Kuakata beach.

From Figure 4.6, it can be observed that the maximum elemental concentrations of Th (14.6 mg/kg) and U (1.65 mg/kg) are observed at the sampling points of K6 and K8, whereas the lowest value of those elements is recorded at the same sampling point K2.

4.8 The mean concentrations of Th and U in beach/inland sands

The mean concentration of Th and U in different locations of inland sands, beach sands and UCC values are plotted in the graph (shown in Figure 4.7 and Figure 4.8). From the bar graphs 4.7 and 4.8, it can be observed that the highest mean values of both Th and U are found at Cox's Bazar beach sands than UCC and other locations.

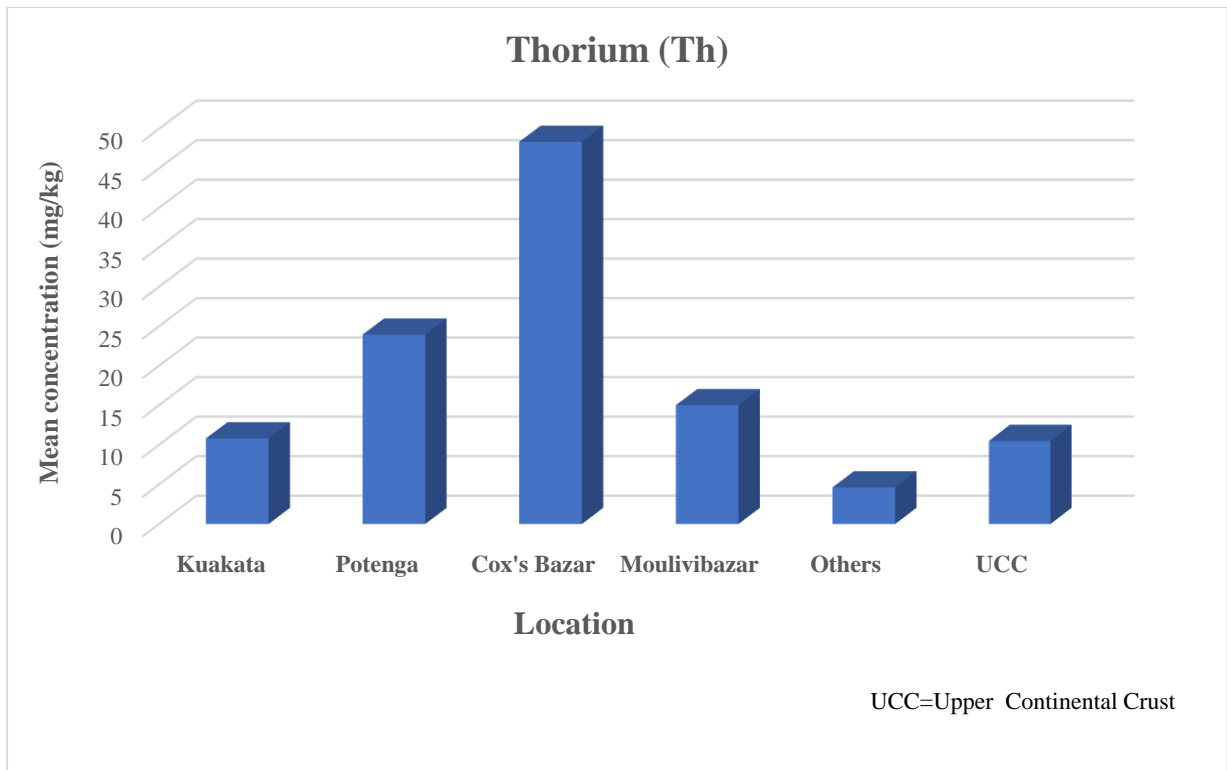


Figure 4.7: Th concentration of Beach sands/Inland sands.

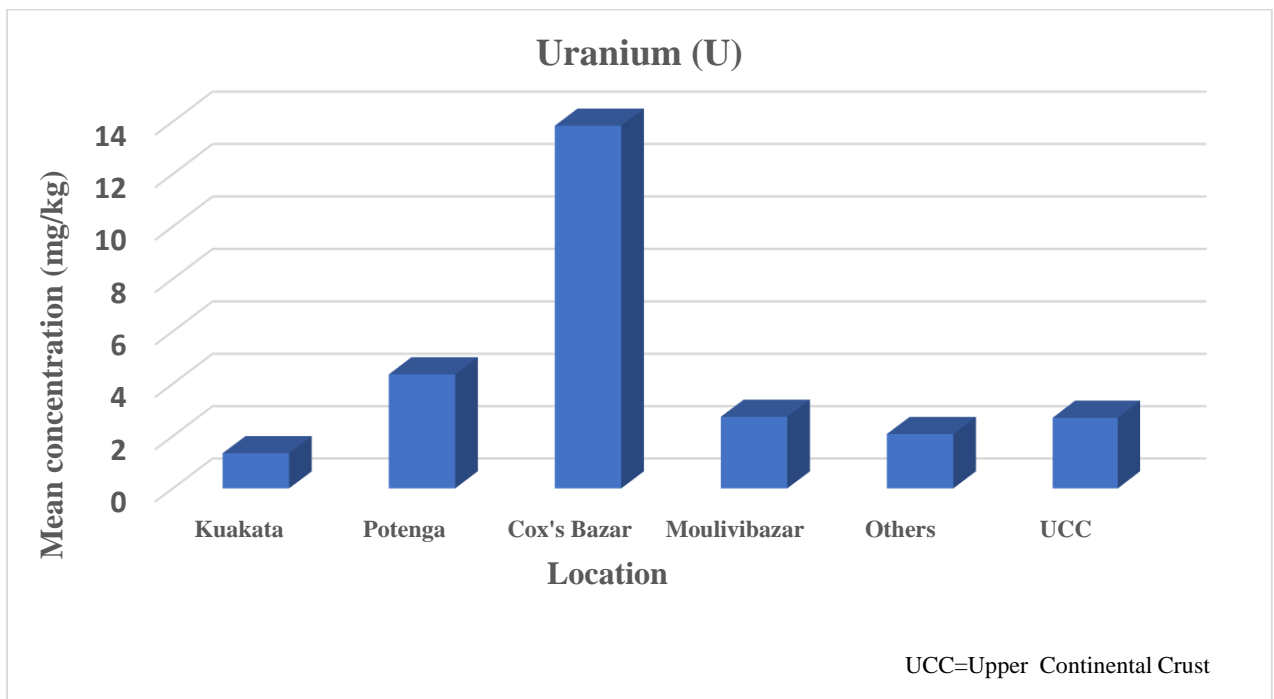


Figure 4.8: U concentration of Beach sands/Inland Sands.

4.9 Elemental enrichment of the sands of Bangladesh

The enrichment factor (EF) is an indicator used to assess the enrichment of elements on surface soil [78]. Thus, EF can be calculated using the following equation:

$$EF = \frac{(\frac{\text{Metal}}{\text{Fe}})_{\text{Sample}}}{(\frac{\text{Metal}}{\text{Fe}})_{\text{Background}}} \dots\dots\dots(4.3)$$

In this study, Fe was used as a reference element for geochemical normalization because of the following three reasons. The reasons are (i) Fe is associated with a fine solid surface. (ii) Its geochemistry is similar to that of many trace metals and (iii) Its natural concentration tends to be uniform. The unity EF indicates a crusted origin, those less than 1.0 suggest a possible mobilization or depletion of metals whereas $EF > 1.0$ indicates that the element is of anthropogenic origin. EF values of minor, moderate, severe and very severe enriched elements in sediments are 1.5-3.0, 3.0-5.0, 5.0-10 and >10 , respectively [65]. According to the EF equation, calculated EF values in inland and beach sands are tabulated in Tables 4.12 - 4.16.

Table 4.12: Enrichment Factor for inland (Moulvibazar) sands in Bangladesh.

Elements	M1	M2	M3	M4	M5	M6
Na	0.152	0.0366	0.459	0.119	0.0516	0.550
Al	1.64	0.817	1.092	1.29	0.893	1.19
K	0.959	0.388	1.13	1.71	0.656	1.37
Sc	0.984	0.841	1.17	0.751	1.00	1.05
Ti	1.42	1.12	1.38	0.775	1.11	1.34
V	1.95	1.06	1.05	0.962	1.26	1.04
Cr	0.994	1.29	1.52	0.818	1.13	1.82
Mn	0.604	0.175	0.463	0.444	0.231	0.635
Fe	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.861	0.476	0.789	0.429	0.498	0.150
As	1.88	1.77	2.31	1.91	0.701	0.874
Cs	1.23	1.43	1.96	1.30	1.65	1.93
La	1.40	1.15	2.00	1.14	1.29	2.47
Ce	1.61	1.34	2.26	1.61	1.30	2.95
Sm	1.34	1.21	2.20	1.13	1.39	2.57
Eu	1.36	1.12	1.77	1.45	1.20	1.82

Dy	1.38	1.11	1.97	1.65	1.35	2.06
Yb	1.68	1.83	2.95	1.42	1.78	2.88
Th	1.85	1.66	2.60	2.24	1.66	3.23
U	1.25	1.51	1.89	1.20	1.18	1.82

Table 4.12 continue

Elements	M7	M8	M9	Min	Max	Mean
Na	0.158	0.0132	0.0755	0.0132	0.550	0.180
Al	2.14	0.397	1.36	0.397	2.14	1.20
K	2.35	0.272	1.17	0.272	2.35	1.11
Sc	1.05	0.437	1.10	0.437	1.17	0.931
Ti	2.64	0.386	1.05	0.386	2.64	1.25
V	1.70	0.721	1.17	0.721	1.95	1.21
Cr	1.07	0.738	0.82	0.738	1.82	1.13
Mn	0.814	0.00594	0.489	0.00594	0.814	0.429
Fe	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.11	0.305	0.58	0.107	0.861	0.466
As	3.11	1.31	3.38	0.701	3.38	1.92
Cs	1.71	0.643	1.69	0.643	1.96	1.50
La	1.65	0.431	1.43	0.431	2.47	1.44
Ce	2.61	0.538	1.93	0.538	2.95	1.79
Sm	1.83	0.468	1.57	0.468	2.57	1.52
Eu	1.27	0.384	1.48	0.384	1.82	1.32
Dy	1.94	0.261	1.52	0.261	2.06	1.47
Yb	2.32	0.640	2.29	0.640	2.95	1.98
Th	3.58	0.759	3.03	0.759	3.58	2.29
U	1.98	0.690	1.93	0.690	1.98	1.50

For inland sands (Table 4.12), the ranges of EF values for the elements As, Cs, Ce, Sm, Yb, Th and U are within 1.5 – 3.0 which indicates that the studied areas are minorly enriched by those elements.

Moreover, the values of EF for Na, Sc, Mn and Co are less than 1.0 which suggests a possible mobilization or depletion of metals. Besides, EF values of Al, K, Ti, V, Cr, Fe, La, Eu and Dy are within 1.0 – 1.5 which indicates that the elements are of anthropogenic origin. The higher level of U represents the presence of ilmenite minerals in the inland sands [61].

Table 4.13: Enrichment Factor for inland (Sherpur, Rajshahi and Sylhet) sands in Bangladesh.

Elements	SH1	RAJ1	SH2	SYL1	SYL2	Min	Max	Mean
Na	1.04	0.992	0.0468	5.99	2.17	0.0468	5.99	2.05
Al	1.19	1.29	0.787	6.75	4.93	0.787	6.75	2.99
K	1.42	1.55	0.830	9.06	11.9	0.830	11.9	4.95
Sc	0.960	1.10	0.306	1.08	0.569	0.306	1.10	0.804
Ti	0.801	0.580	0.646	6.53	4.93	0.580	6.53	2.70
V	1.00	0.519	0.453	4.65	4.68	0.453	4.68	2.26
Cr	0.601	0.332	15.6	0.444	0.782	0.332	15.6	3.55
Mn	1.22	1.84	0.723	5.15	8.79	0.723	8.79	3.54
Fe	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.676	0.492	1.13	0.708	0.745	0.492	1.13	0.750
As	1.13	0.616	2.58	5.27	15.5	0.616	15.5	5.01
Cs	1.50	1.88	1.02	2.82	2.63	1.02	2.82	1.97
La	2.60	1.50	0.501	7.76	3.68	0.501	7.76	3.21
Ce	2.13	1.05	0.960	1.29	0.903	0.903	2.13	1.27
Sm	2.86	1.93	2.69	9.97	5.79	1.93	9.97	4.65
Eu	2.01	1.22	2.19	4.18	3.95	1.22	4.18	2.71
Dy	2.62	2.28	18.4	10.0	8.24	2.28	18.4	8.31
Yb	3.55	4.83	35.1	15.6	18.0	3.55	35.1	15.4
Th	2.83	1.33	1.99	1.33	0.946	0.946	2.83	1.69
U	2.34	1.40	7.45	8.28	10.3	1.40	10.3	5.95

In this research among the studied 20 elements for inland sands (shown in Table 4.13), the mean values of EF for the elements Na, Al, Ti, V, Cs, Eu, Th and U are within 1.5 – 3.0 which shows that the studied areas are minor enriched by those elements.

The values of EF for the elements of As, Dy and U are greater than 5.0 which indicates that those elements are severely enriched, whereas K, Cr, Mn, As, La and Sm are moderately enriched due to the range of 3.0 – 5.0. Besides, the Yb is very severely enriched by that element and the remaining elements are of crustal origin.

Table 4.14: Enrichment Factor for beach sands of Kuakata, Bangladesh.

Elements	K1	K2	K3	K4	K5	K6
Na	0.975	0.954	0.657	1.22	1.06	0.898
Al	0.945	1.01	0.739	1.31	1.11	0.968
K	1.04	1.26	0.873	1.27	1.07	0.851
Sc	1.27	0.845	0.727	1.25	1.20	1.27
Ti	1.10	0.644	0.586	1.67	0.962	1.19
V	1.05	0.862	0.759	1.40	1.04	1.04
Cr	0.858	0.594	0.471	0.795	0.993	0.799
Mn	0.924	0.820	0.726	1.47	0.910	0.926
Fe	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.767	0.775	0.853	0.639	0.679	0.718
As	0.810	1.99	2.73	0.892	0.818	0.822
Cs	0.819	1.61	1.30	1.05	0.901	0.717
La	1.62	1.19	0.927	3.17	2.44	2.34
Ce	1.80	1.26	0.948	2.21	2.34	2.14
Sm	1.84	1.25	1.02	3.29	2.27	2.23
Eu	2.18	1.69	1.23	2.10	2.56	2.05
Dy	1.97	1.32	1.50	4.43	2.16	2.22
Yb	2.27	1.92	2.41	5.64	2.04	3.18
Th	2.28	1.32	1.12	2.73	2.84	2.77
U	1.04	0.565	0.832	1.64	1.05	1.16

Table 4.14 continue

Elements	K7	K8	Min	Max	Mean
Na	1.06	1.04	0.657	1.22	0.983
Al	1.13	1.06	0.739	1.31	1.03
K	1.07	0.967	0.851	1.27	1.05
Sc	1.13	1.13	0.727	1.27	1.10
Ti	1.24	0.847	0.586	1.67	1.03
V	1.12	1.04	0.759	1.40	1.04
Cr	0.976	0.650	0.471	0.993	0.767
Mn	0.864	0.871	0.726	1.47	0.939
Fe	1.00	1.00	1.00	1.00	1.00
Co	0.699	0.724	0.639	0.853	0.732
As	0.938	1.16	0.810	2.73	1.27
Cs	0.770	0.727	0.717	1.61	0.987

La	1.89	2.26	0.927	3.17	1.98
Ce	1.66	2.07	0.948	2.34	1.80
Sm	2.07	2.58	1.02	3.29	2.07
Eu	1.97	2.24	1.23	2.56	2.00
Dy	2.19	2.01	1.32	4.43	2.23
Yb	3.30	2.60	1.92	5.64	2.92
Th	1.76	2.96	1.12	2.96	2.22
U	0.783	1.43	0.565	1.64	1.06

From Table 4.14, it is observed that the range of EF values for the elements La, Ce, Sm, Eu, Dy, Yb and Th are within 1.5 – 3.0 which indicates that the studied areas are minor enriched by those elements. The remaining elements are from crustal and anthropogenic origin.

Table 4.15: Enrichment Factor for beach sands of Patenga, Bangladesh.

Elements	P1	P2	P3	P4	P5	P6	P7
Na	0.449	0.393	0.471	0.693	0.754	0.916	0.549
Al	0.781	0.791	0.982	1.15	1.05	1.37	1.09
K	1.24	1.05	1.25	1.70	1.72	2.10	1.86
Sc	0.893	0.849	0.758	0.785	0.733	0.838	0.786
Ti	3.767	1.35	1.57	1.46	1.09	2.30	1.42
V	1.13	0.788	0.901	0.893	0.784	1.02	0.919
Cr	9.82	1.00	3.72	1.07	0.793	2.23	1.58
Mn	1.25	0.635	0.668	0.757	0.782	1.07	0.952
Fe	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.674	0.976	0.740	0.870	0.965	0.851	0.835
As	1.01	1.21	1.10	0.977	1.87	1.69	1.77
Cs	0.504	1.29	0.985	1.28	1.40	1.03	1.08
La	7.13	1.25	2.75	2.31	1.28	3.71	1.92
Ce	7.80	1.43	3.39	2.16	1.43	3.58	2.17
Sm	5.85	1.33	2.96	2.28	1.26	3.00	2.08
Eu	3.61	1.25	2.75	2.03	1.39	2.69	1.75
Dy	5.89	1.17	2.30	2.12	1.15	3.34	1.58
Yb	8.20	1.94	3.59	3.22	1.45	4.02	1.45
Th	16.8	2.01	3.64	2.98	2.76	4.98	3.56
U	11.6	1.19	2.92	3.06	2.43	2.75	3.89

Table 4.15 Continue

Elements	P8	P9	P10	Min	Max	Mean
Na	0.806	0.635	0.935	0.393	0.935	0.660
Al	1.33	1.06	1.42	0.781	1.42	1.10
K	1.79	1.51	2.74	1.05	2.74	1.70
Sc	0.830	0.751	0.844	0.733	0.893	0.807

Ti	2.70	2.33	3.23	1.09	3.77	2.12
V	1.13	1.06	1.19	0.784	1.19	0.982
Cr	3.07	7.28	2.37	0.793	9.82	3.29
Mn	0.892	0.925	0.959	0.635	1.25	0.890
Fe	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.984	0.854	1.04	0.674	1.04	0.879
As	1.35	1.35	1.82	0.977	1.87	1.42
Cs	0.958	0.872	1.45	0.504	1.45	1.09
La	4.21	2.90	2.70	1.25	7.13	3.02
Ce	4.40	3.72	3.17	1.43	7.80	3.33
Sm	4.20	4.63	3.42	1.26	5.85	3.10
Eu	3.18	2.87	2.05	1.25	3.61	2.36
Dy	2.94	2.51	2.46	1.15	5.89	2.55
Yb	4.56	2.98	2.99	1.45	8.20	3.44
Th	14.7	3.74	4.79	2.01	16.8	5.99
U	5.72	3.17	4.24	1.19	11.6	4.09

For Patenga beach sands (shown in Table 4.15), the mean value of EF for the elements K, Ti, Eu and Dy are within 1.5 – 3.0 which indicates that the studied areas are minor enriched by those elements. The value of EF for Th is greater than 5.0 which indicates that this element is severely enriched, whereas Mn, La, Ce, Sm, Yb and U are moderately enriched due to the range of 3.0 – 5.0.

Table 4.16: Enrichment Factor for beach sands of Cox’s Bazar, Bangladesh.

Elements	C1	C2	C3	C4	C5	C6	C7
Na	0.599	0.210	0.417	0.167	0.844	0.749	0.560
Al	0.910	0.502	0.684	0.551	1.38	1.07	1.07
K	1.18	0.517	1.04	0.277	1.84	1.32	1.93
Sc	0.801	0.992	0.627	1.04	0.764	0.701	0.918
Ti	1.01	0.783	1.08	2.67	0.755	0.598	1.70
V	0.976	0.691	0.646	1.16	0.951	0.844	1.02
Cr	0.607	1.98	0.532	2.91	0.767	0.514	0.59
Mn	1.43	1.15	1.13	1.80	2.16	1.60	1.34
Fe	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.852	0.800	0.780	0.614	1.09	0.991	0.966
As	4.54	1.44	2.95	1.36	4.95	4.55	3.86
Cs	0.984	0.425	0.748	0.239	0.696	0.864	1.26
La	2.24	3.77	1.03	3.87	0.961	1.21	2.14
Ce	2.25	4.73	1.34	3.49	0.806	1.17	2.17
Sm	2.56	3.15	1.28	3.79	1.17	1.28	2.17
Eu	2.65	2.35	1.61	2.12	1.50	1.93	1.73

Dy	1.96	1.87	1.17	3.45	1.52	1.36	1.83
Yb	3.27	2.53	0.992	5.12	1.31	1.60	0.264
Th	2.00	4.25	10.8	35.9	2.06	1.15	3.88
U	1.18	1.37	5.76	48.3	2.95	1.85	6.31

Table 4.16 continue

Elements	C8	C9	C10	C11	C12	C13	C14
Na	0.634	1.66	0.838	0.700	0.839	0.680	0.0767
Al	0.924	2.61	1.02	1.08	2.40	1.22	0.295
K	1.34	3.66	1.11	1.21	3.32	1.64	0.128
Sc	0.562	0.709	0.774	0.732	0.934	0.793	0.838
Ti	0.885	1.39	0.983	0.740	14.3	1.64	3.25
V	0.612	1.89	0.942	0.969	5.99	0.911	1.01
Cr	0.560	0.717	0.941	0.639	0.990	0.707	3.48
Mn	1.38	3.07	1.51	1.26	9.74	1.75	1.69
Fe	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Co	1.11	1.07	0.923	1.05	2.30	0.909	0.453
As	5.75	9.83	4.17	3.35	10	4.77	0.999
Cs	2.28	1.29	0.909	0.909	1.97	1.33	0.113
La	0.784	1.83	1.26	0.975	37.3	1.46	6.16
Ce	0.868	0.908	1.23	0.870	4.99	1.16	5.59
Sm	0.902	2.16	1.64	1.17	35.5	1.89	5.50
Eu	0.878	1.23	1.70	1.35	2.07	1.45	2.30
Dy	1.03	2.19	1.51	1.25	22.3	1.94	4.83
Yb	1.87	2.24	2.30	1.75	25.9	2.50	7.80
Th	1.07	3.31	1.18	2.04	18.4	3.41	7.54
U	2.19	8.69	1.79	2.85	45.0	2.30	5.39

Table 4.16 Continue

Elements	C15	C16	C17	Min	Max	Mean
Na	0.416	0.528	0.684	0.0767	1.66	0.624
Al	0.753	0.852	0.916	0.295	2.61	1.07
K	0.828	0.912	1.08	0.128	3.66	1.37
Sc	0.864	0.772	0.842	0.562	1.04	0.804
Ti	1.65	1.12	1.64	0.598	14.3	2.13
V	0.918	0.775	0.987	0.612	5.99	1.25
Cr	1.29	0.801	1.73	0.514	3.48	1.16
Mn	1.12	1.62	1.58	1.123	9.74	2.08
Fe	1.00	1.00	1.00	1.00	1.00	1.00
Co	0.913	0.946	0.942	0.453	2.30	0.982
As	3.69	4.89	4.37	0.999	10.0	4.44
Cs	0.829	0.739	0.915	0.113	2.28	0.970
La	2.49	3.58	2.01	0.784	37.3	4.30

Ce	2.22	3.21	1.72	0.806	5.59	2.28
Sm	3.09	3.18	1.78	0.902	35.5	4.25
Eu	1.60	1.72	1.35	0.878	2.65	1.74
Dy	3.20	1.80	1.79	1.03	22.3	3.24
Yb	2.64	2.30	1.93	0.264	25.9	3.90
Th	3.26	4.53	2.96	1.07	35.9	6.34
U	2.29	1.80	3.31	1.18	48.3	8.43

From the above Table 4.16, it can be concluded that the mean values of EF for the elements of As, La, Sm, Dy and Yb are within 3.0 – 5.0 which indicates that the studied elements are moderately enriched. The average values of EF for Th and U are 6.34 and 8.43, respectively, which reveals that elements are severely enriched by those elements. The high value of Th and U represents the presence of monazite and ilmenite minerals in the inland and beach sands [61].

4.10 The mean value of enrichment factor (EF) for Th and U in beach/inland sands

To compare the concentration of any elements at any point relative to UCC values EF is used. In this study, the determined values of EF for all locations are compared with each other and shown in Figure 4.9 and Figure 4.10. From Figure 4.9, it reveals that the concentrations of Th at Patenga and Cox's Bazar are 5.6 to 6 times higher than those values of UCC. Besides, Figure 4.10 shows the highest value at Cox's Bazar beach sands which is 8.43 times higher than UCC values.

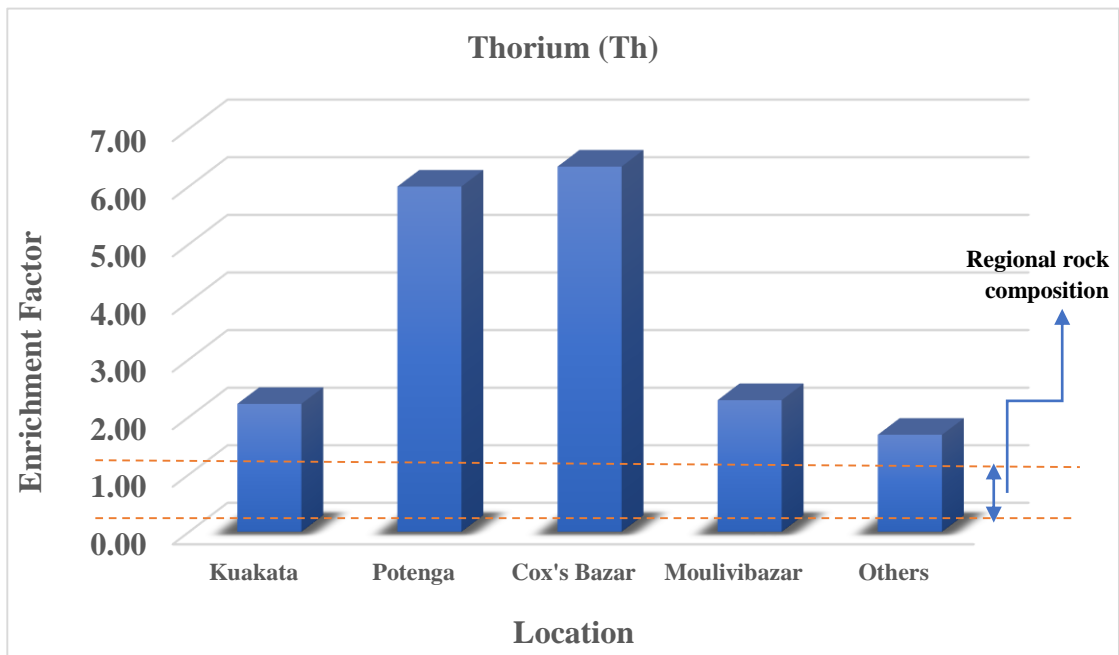


Figure 4.9: Enrichment factor (EF) of Th in beach/inland sands.

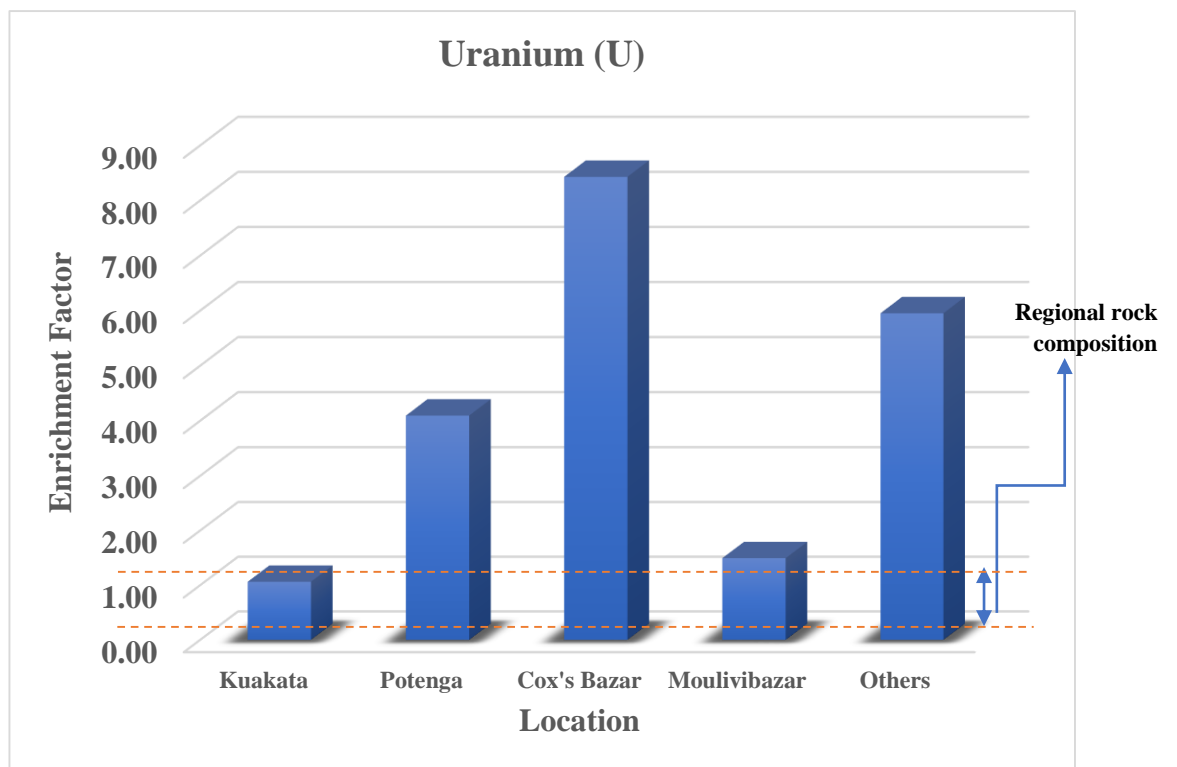


Figure 4.10: Enrichment factor (EF) of U in beach/inland sands.

4.11 Depth wise concentration Variations of Th and U in Beach/Inland sands

For depth-wise concentration variations, sand samples were collected by boring the beach and inland sands of each location for a specific depth of 0–05 cm, 05–15 cm, 15–25 cm, 25–35 cm with respect to the surface. The depth-wise elemental mean concentration of Th and U, their ranges, standard deviation, relative standard deviation as well as UCC (upper continental crust) are tabulated in Table 4.17.

Table 4.17: Elemental abundances (in mg/kg, otherwise specified) in beach and inland sands with depth and spatial Variations.

This work (sampling depth)	Beach Sands		Inland Sands	
	Th(mg/kg)	U(mg/kg)	Th(mg/kg)	U(mg/kg)
0-05 cm	25.5	14.2	20.3	3.71
05-15 cm	21.9	3.95	12.8	2.74
15-25 cm	24.1	3.36	15.7	3.03
25-35 cm	13.1	2.71	8.88	1.62
Mean	21.1	6.05	14.4	2.77
Rang	13.1-25.5	2.71-14.2	8.88-20.3	1.62-3.71
STD	5.59	5.44	4.81	0.87
RSD	26.5	90.1	33.3	31.3
UCC	10.5	2.7	10.5	2.7

UCC: Upper Continental crust

From Table 4.17, the range of the Th and U concentration for beach sands and inland sands are 13.1-25.58 mg/kg, 8.88-20.3 mg/kg and 2.71-14.2 mg/kg, 1.62-3.71 mg/kg, respectively whereas the mean concentration of Th and U are 21.2 mg/kg, 14.4 mg/kg and 6.05 mg/kg, 2.77 mg/kg, respectively. The depth-wise variation of Th and U are due to the high contents of Magnetite, Ilmenite in the beach and inland sands [62]. The depth-wise concentration (mg/kg) in the beach sands and inland sands are shown in Figure 4.11 and Figure 4.12.

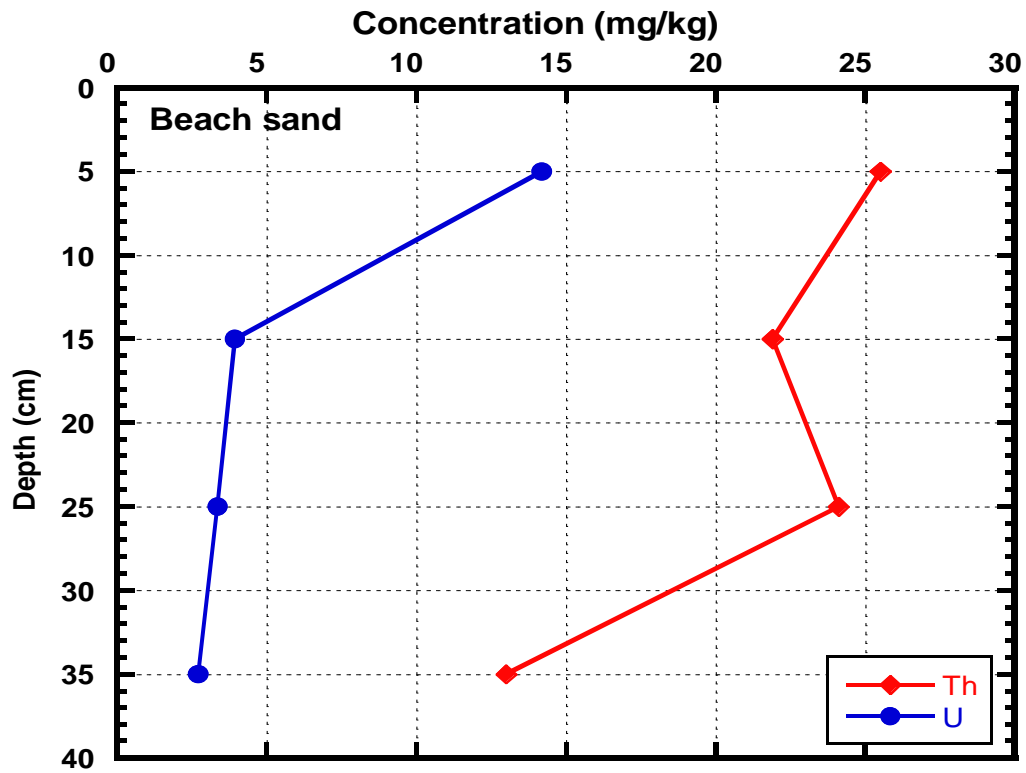


Figure 4.11: Depth wise concentration Variations Th and U for Beach sands.

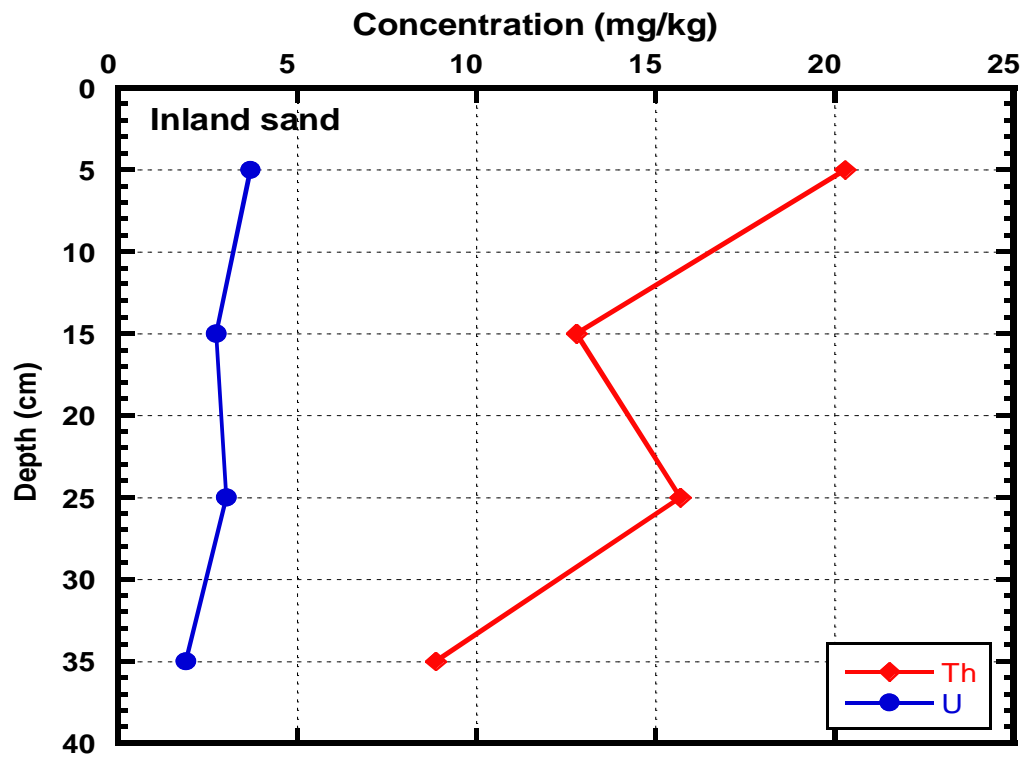


Figure 4.12: Depth wise concentration variations Th and U for Inland sands.

From Figures 4.11 and 4.12, it can be concluded that the overall Th concentration variation with depth (up to 35 cm) decreases from the top to the deeper layers for the beach and inland sands of Bangladesh. Moreover, the overall U concentration variations also follow the same pattern, however, the concentration variation for beach sands is greater than that of the inland sands of Bangladesh.

4.12 Conversion of elemental mean concentration into activity concentration

In this study, the activity concentration of ^{238}U , ^{232}Th and ^{40}K has been calculated from the elemental concentration of U, Th and K using equation (4.4). The activity concentrations of radionuclides such ^{238}U , ^{232}Th and ^{40}K in inland and beach sands of Bangladesh with their ranges (minimum and maximum), mean, standard, relative standard deviation as well the radiological hazard indexes (radium equivalent activity and external hazard index) in the studied samples are given in Tables 4.18 – 4.22.

$$F_E = \frac{M_E \cdot C \cdot A_E}{f_{A,E} \cdot N_A \cdot \lambda_E} \dots\dots\dots(4.4)$$

where F_E , M_E , λ_E , $f_{A,E}$ and A_E are the fraction of element E in the samples, the atomic mass in kg/mol, the decay constant in s^{-1} , the fractional atomic abundance in nature and the determined activity concentration in Bq/kg, respectively. N_A is Avogadro's number of 6.023×10^{23} atoms/mol, C is a constant with values of 10^6 for Th and U as well as 10^3 for potassium that converts the ratio of the elements to soils/sands mass into part per million or a percentage [44].

4.13 Calculation of radiological hazard indexes

The beach sands and inland sands in the study areas are the most important construction materials used by the local people. Besides, the selected elements from the sands are also

utilized for different industrial purposes and research uses. Because of the safe use of those beach sands and inland sands, the radiological hazard indices are the main concern issues. To assess the health effects, the radiation hazards such as radium equivalent activity (R_{eq}) and external hazard index (H_{ex}) have been calculated from the radionuclides (^{226}Ra , ^{232}Th and ^{40}K) using the equations (4.5) and (4.6), respectively, and their values have also given in Tables 4.18 – 4.22.

4.13.1 Radium equivalent activity (R_{eq})

The radium equivalent activity (R_{eq}) is an index that shows the total activities of ^{226}Ra , ^{232}Th and ^{40}K . Since ^{238}U is in secular equilibrium with its daughter ^{226}Ra , the activity concentrations of ^{238}U were used as activity concentrations of ^{226}Ra in the sands [66]. The distributions of those radionuclides activity in the soil and sands may vary. Variability concerning radiation exposure may also be defined in terms of radium-equivalent activity (R_{eq}) in Bq/kg. The radium-equivalent activity was calculated from the following equation (4.5) [67].

$$R_{eq} = (A_{Ra} + 1.43 A_{Th} + 0.077 A_k) \text{ Bq/kg} \quad \dots\dots\dots(4.5)$$

where A_{Ra} , A_{Th} and A_k are the specific activities of ^{226}Ra , ^{232}Th and ^{40}K , respectively, in Bq/kg.

4.13.2 External hazard index (H_{ex})

The external hazard index (H_{ex}) is the radiation dose rate due to the external exposure to gamma radiation in the construction materials of dwellings which was calculated by using formula (4.6) [68].

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_k/4810 \quad \dots\dots\dots(4.6)$$

Where, A_{Ra} , A_{Th} and A_k are the specific activities of ^{226}Ra , ^{232}Th and ^{40}K in Bq/kg, respectively.

4.14 Activity concentration and radiological hazard indexes of inland and beach sands

4.14.1 Activity concentration of inland sands

The radioactivity, radium equivalent activity as well as external hazard indexes for the studied samples of inland sand are tabulated in Table 4.18.

Table 4.18: Activity concentration and radiological hazard indexes for inland sands of Moulvibazar.

Sample	Activity Concentrations			Radiological Hazard Indexes	
	⁴⁰ K	²³² Th	²³⁸ U	Ra _{eq}	Hex
M1	466	53.0	26.9	139	0.374
M2	214	54.0	36.7	130	0.352
M3	464	63.1	34.3	160	0.433
M4	510	39.4	15.8	111	0.301
M5	549	82.0	43.5	203	0.548
M6	464	64.6	27.2	155	0.419
M7	308	27.8	11.4	74.9	0.202
M8	481	79.0	53.7	204	0.550
M9	552	84.0	40.0	203	0.547
Min	214	27.8	11.4	74.9	0.202
Max	552	84.0	53.7	204	0.550
Mean	445	60.8	32.2	153	0.414
UNSCEAR (2000)	474	36	33	370	1.00

From Table 4.18, it is observed that the minimum and maximum values of ²³²Th and ²³⁸U are 27.8 Bq/kg and 84.0 Bq/kg; 11.4 Bq/kg and 53.7 Bq/kg with the mean values of 60.8 Bq/kg and 32.2 Bq/kg, respectively. Also, the activity concentrations of ⁴⁰K are found in the range of 214 – 552 Bq/kg with a mean value of 445 Bq/kg. The overall mean activities of ²³²Th and ⁴⁰K in the studied samples were found higher than that of the world mean values of 36 and

474 Bq/kg, respectively, whereas the mean activity of ^{238}U was comparable to the world mean value of 33 Bq/kg [69].

It can be also observed from Table 4.18 that the total radium equivalent activity (R_{aeq}) due to the presence of ^{226}Ra , ^{232}Th and ^{40}K in Moulvibazar inland sands varied from 74.9 to 204 Bq/kg with an average value of 153 Bq/kg, which is lower than the world average value of 370 Bq/kg [70]. Moreover, the mean value of the external radiation hazard index (H_{ex}) is 0.41 which is less than 1.0 and ensures that it is safe to carry out the activities for the human in that area [42].

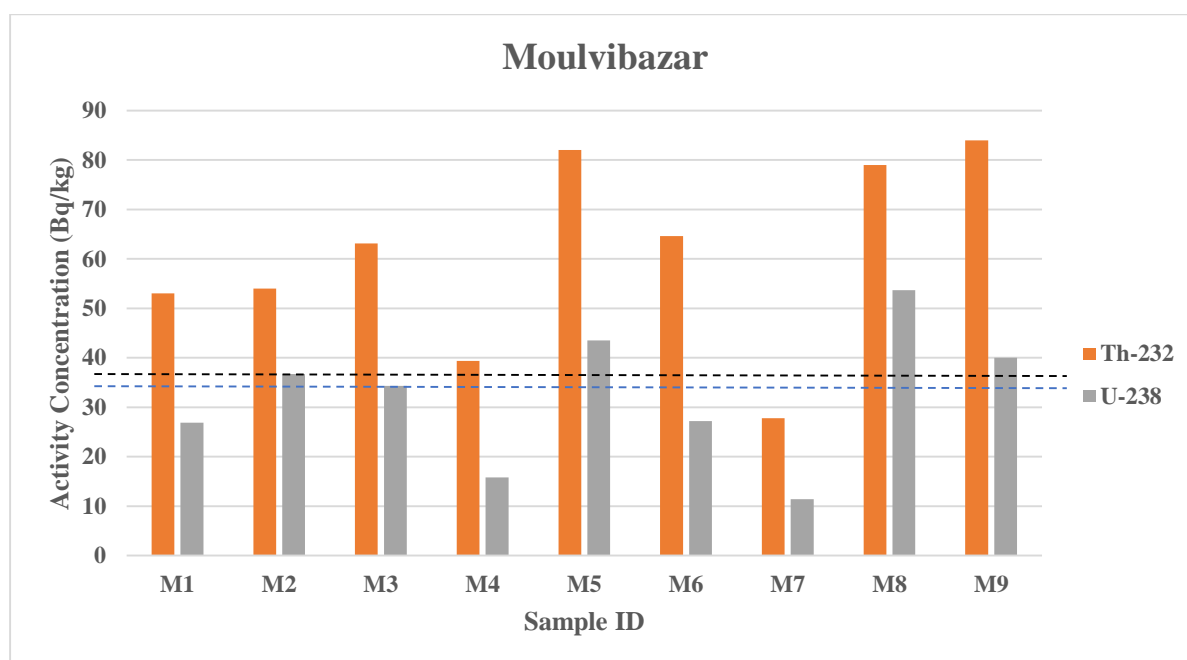


Figure 4.13: Activity concentration of ^{232}Th and ^{238}U in inland sands of Moulvibazar.

Figure 4.13 shows the graphical representation of the activity concentrations of all sampling points in Moulvibazar inland sands. In the above figure 4.13, it is observed that the maximum values of ^{232}Th and ^{238}U are at the M9 and M8 sampling points, respectively, whereas the minimum values of ^{232}Th and ^{238}U both are at the M7 sampling point.

Table 4.19: Activity concentration of Sherpur, Rajshahi and Sylhet inland sands.

Sample	Activity Concentrations			Radiological hazard indexes	
	⁴⁰ K	²³² Th	²³⁸ U	R _{eq}	H _{ex}
SH1	460	54.2	33.5	146	0.308
RAJ1	404	20.5	16.0	76.4	0.217
SH2	87.7	12.4	34.7	59.3	0.276
SYL1	491	4.25	19.8	63.7	0.290
SYL2	474	2.23	18.1	57.8	0.310
Min	87.7	2.23	16.0	57.8	0.217
Max	491	54.2	34.7	146	0.310
Mean	383	18.7	24.4	80.7	0.280

Table 4.19 shows the radioactivities and radiological hazard indexes of the studied nuclides in the samples collected from Sherpur, Rajshahi and Sylhet inland sands. The sample collected from the SYL1 and SYL2 have values close to each other (4.25 Bq/kg and 2.23 Bq/kg), and distinctly low. Besides, the higher value refers to the sample (SH1) is 54.2 Bq/kg with a mean value of 18.7 Bq/kg for ²³²Th. Moreover, the activity concentrations of ²³⁸U obtained in inland sands are in the range of 16.0 – 34.7 Bq/kg with a mean value of 24.4 Bq/kg. Also, the minimum and maximum values obtained are from 87.7 Bq/kg to 491 Bq/kg with an average value of 383 Bq/kg for ⁴⁰K. In this case, the overall mean activities of ²³⁸U, ²³²Th and ⁴⁰K in the studied samples were found lower than that of the world mean values of 33, 36 and 474 Bq/kg [69], respectively.

Table 4.19 also shows that the total radium equivalent activity (R_{eq}) calculated from the activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K in Other inland sands (Sherpur, Rajshahi and Sylhet) varied from 57.8 to 146 Bq/kg with an average value of 80.7 Bq/kg, which is lower than the world average value of 370 Bq/kg [70]. Moreover, the mean value of the external radiation hazard index (H_{ex}) is 0.28 which is less than 1 and indicates that the studied areas are safe for human health [42].

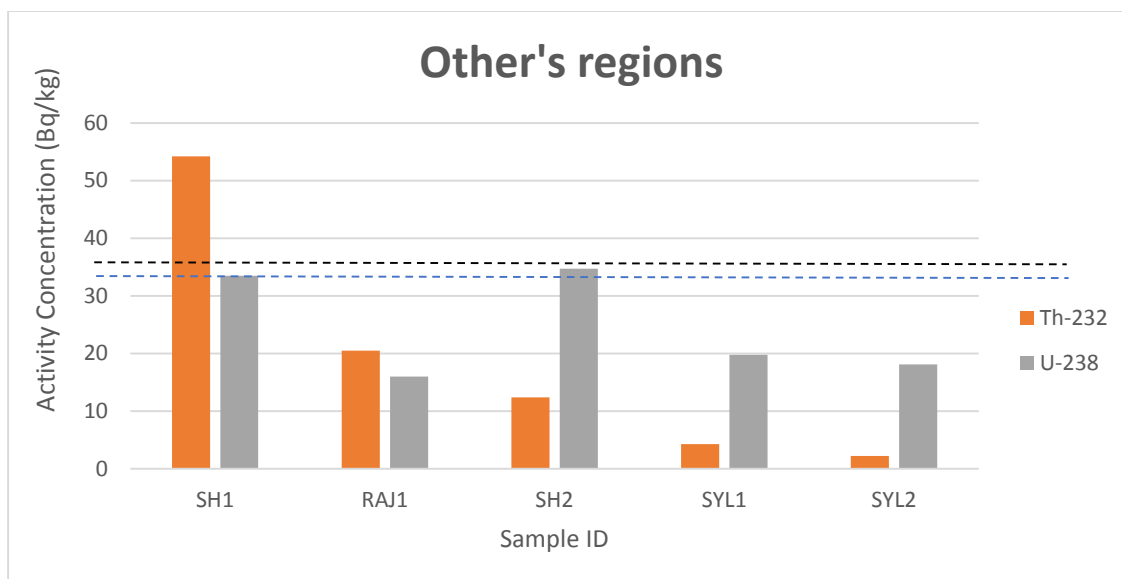


Figure 4.14: Activity concentration of ²³²Th and ²³⁸U in Other's (Sherpur, Rajshahi and Sylhet) inland sands.

Figure 4.14 represents the graphical representation of the activity concentrations of all sampling points in other inland sands (Sherpur, Rajshahi and Sylhet). From the above Figure 4.14, it is observed that the highest values of ²³²Th and ²³⁸U are found at the SH1 and M3 sampling points, respectively, whereas the lowest values of ²³²Th and ²³⁸U are at the SYL2 and RAJ1 sampling points, respectively.

4.14.2 Activity concentration of beach sands

The radioactivity, radium equivalent activity, as well as external hazard indexes for the studied samples of beach sands, are tabulated in Table 4.20.

Table 4.20: Activity concentration and radiological hazard indexes for beach sands of Patenga.

Sample	Activity Concentrations			Radiological hazard indexes	
	⁴⁰ K	²³² Th	²³⁸ U	R _{eq}	Hex
P1	432	345	177	704	1.90
P2	546	61.4	27.2	157	0.424
P3	334	57.5	34.5	142	0.385

P4	543	56.0	42.9	165	0.445
P5	633	59.8	39.4	174	0.469
P6	438	61.4	25.3	147	0.397
P7	581	65.6	53.6	192	0.519
P8	335	162	47.1	304	0.822
P9	352	51.3	32.5	133	0.359
P10	475	49.0	32.3	139	0.375
Min	334	49.0	25.3	133	0.359
Max	633	345	177	704	1.90
Mean	467	96.9	51.2	226	0.610

From Table 4.20, the minimum to maximum values ranges from (49.0 - 345) Bq/kg and (25.3 - 177) Bq/kg with the mean values of 96.9 Bq/kg and 51.2 Bq/kg for ^{232}Th and ^{238}U , respectively. Besides, a minimum of 334 Bq/kg to a maximum of 633 Bq/kg with a mean value of 467 Bq/kg is recorded for ^{40}K . In this case, the mean values of activity concentration of radionuclides such as ^{238}U , ^{232}Th and ^{40}K in the studied areas are found as higher values than that of the world mean values of 33, 36 and 474 Bq/kg, respectively [69].

Table 4.20 also shows that the calculated total Radium equivalent activity (Ra_{eq}) from the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in Patenga beach sands are in the range from 133 to 704 Bq/kg with an average of 226 Bq/kg, which is lower than the world average value of 370 Bq/kg [70]. Moreover, the mean value of the external radiation hazard index (Hex) is 0.61 which is less than 1 and indicates that the studied areas are safe for human health [42].

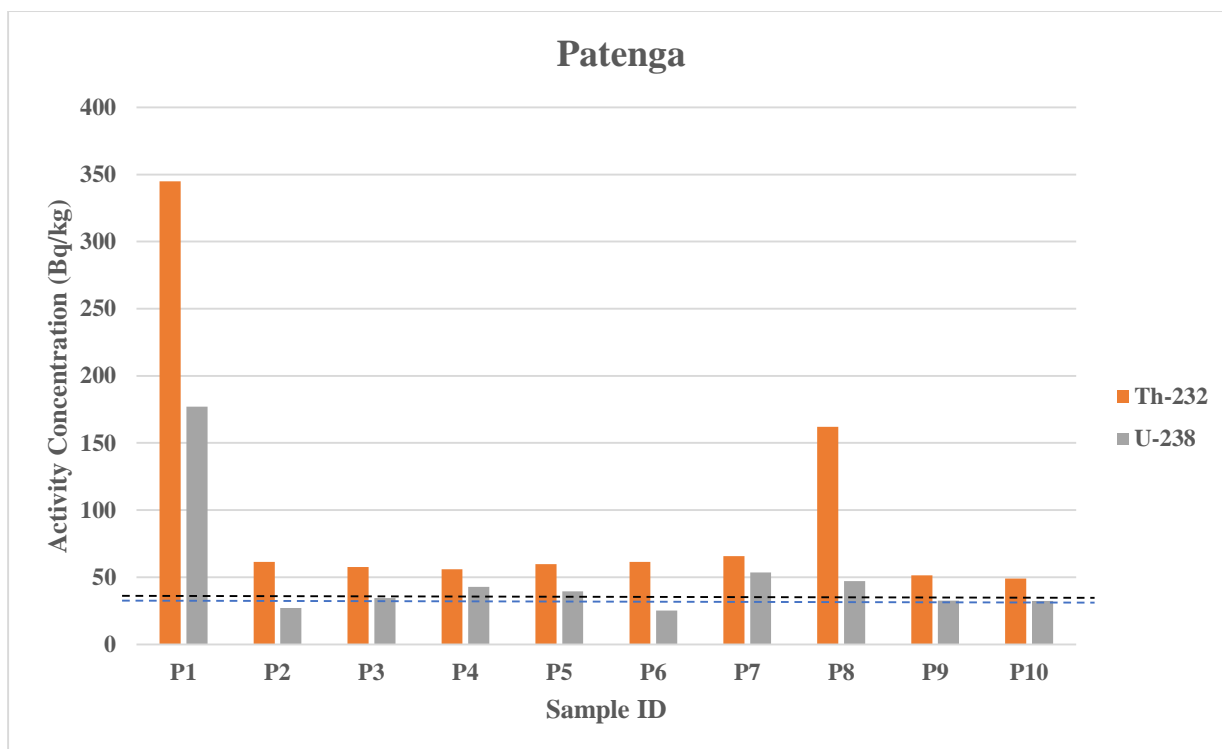


Figure 4.15: Activity concentration of ²³²Th and ²³⁸U in beach sands of Patenga.

Figure 4.15 shows the graphical representation of the activity concentrations of all sampling points in Patenga beach sands. From the above Figure 4.15, it can be observed that the highest values of both ²³²Th and ²³⁸U are found at the P1 sampling point, whereas the lowest values of ²³²Th and ²³⁸U are at the sampling points of P10 and P6, respectively.

Table 4.21: Activity concentration and radiological hazard indices for Cox's Bazar beach sands.

Sample	Activity Concentrations			Radiological hazard indexes	
	⁴⁰ K	²³² Th	²³⁸ U	R _{eq}	Hex
C1	328	32.6	14.4	86.3	0.233
C2	291	141	34.1	258	0.698
C3	419	259	103	505	1.36
C4	202	1546	1551	3777	10.2
C5	405	26.8	28.7	98.2	0.265
C6	362	18.8	22.4	77.2	0.208
C7	461	54.6	66.4	180	0.486

C8	417	19.7	30.1	90.5	0.244
C9	399	21.4	41.8	103	0.279
C10	318	20.0	22.7	75.8	0.205
C11	311	31.0	32.3	101	0.272
C12	485	159	290	555	1.50
C13	405	49.6	24.9	127	0.343
C14	204	710	380	1411	3.812
C15	365	84.9	44.5	194	0.524
C16	303	89.0	26.3	177	0.478
C17	390	63.1	52.8	173	0.467
Min	202	18.8	14.4	75.8	0.205
Max	485	1546	1551	3777	10.2
Mean	357	196	163	470	1.27

Table 4.21 shows that the minimum to maximum values ranges from (18.8 – 1546) Bq/kg and (14.4 – 1551) Bq/kg for ^{232}Th and ^{238}U , respectively. Also, a minimum of 202 Bq/kg to a maximum of 485 Bq/kg is recorded for ^{40}K . Besides, the average values of ^{40}K , ^{232}Th and ^{238}U are found as 357 Bq/kg, 196 Bq/kg and 163 Bq/kg, respectively.

Moreover, from Table 4.21 it can be concluded that the mean activity concentration of ^{232}Th and ^{238}U in Cox's Bazar beach sands are 5.43 times and 4.93 times higher than that of the world average value, whereas the mean activity concentration of ^{40}K is comparable with the world average value [69].

It can be also observed from Table 4.21 that the total Radium equivalent activity (Ra_{eq}) from the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in Cox's Bazar beach sands are in the range from 75.8 to 3777 Bq/kg with an average value of 470 Bq/kg, which is 1.27 times higher than the world average value of 370 Bq/kg [70]. Moreover, the mean value of the external radiation hazard index (Hex) is 1.27 which is greater than 1, and it indicates that the studied areas are not safe for human health [42].

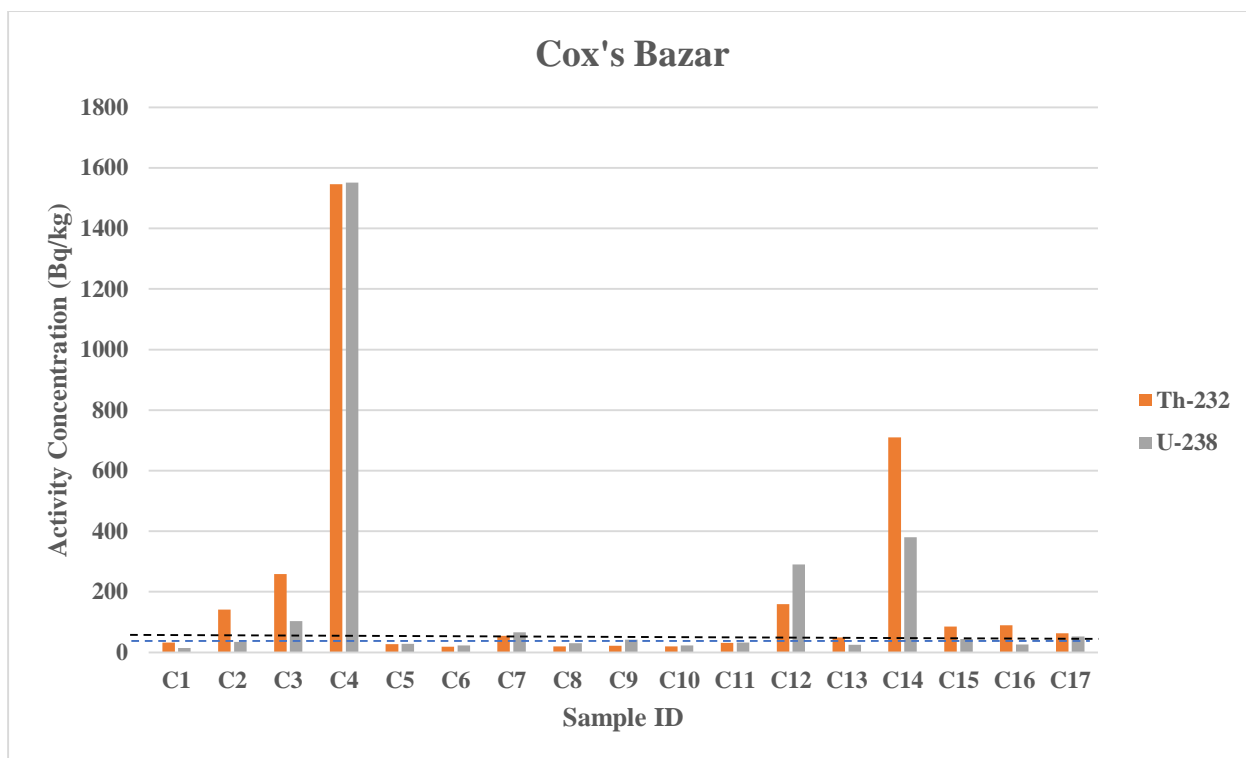


Figure 4.16: Activity concentration of ²³²Th and ²³⁸U in beach sands of Cox's Bazar.

Figure 4.16 shows the graphical representation of the activity concentrations of all sampling points in Cox's Bazar beach sands. From the above Figure 4.16, it can be observed that the highest values of both ²³²Th and ²³⁸U are at the C4 sampling point, whereas the lowest values of ²³²Th and ²³⁸U are at the C6 and C1 sampling points, respectively.

Table 4.22: Activity concentration and radiological hazard indices for beach sands of Kuakata.

Sample	Activity Concentrations			Radiological hazard indexes	
	⁴⁰ K	²³² Th	²³⁸ U	R _{aeq}	Hex
K1	372	48.3	16.4	114	0.31
K2	433	26.8	8.57	80.2	0.22
K3	448	34.0	18.8	102	0.28
K4	340	43.2	19.3	107	0.29
K5	333	52.3	14.4	115	0.31
K6	307	58.9	18.5	126	0.34

K7	337	32.8	10.9	83.7	0.23
K8	297	53.7	19.4	119	0.32
Min	297	26.8	8.57	80.2	0.22
Max	448	58.9	19.4	126	0.34
Mean	358	43.8	15.8	106	0.29

From Table 4.22, it can be observed that the activity concentration of ^{232}Th and ^{238}U in the investigated samples vary in the range from 26.8 Bq/kg to 58.9 Bq/kg and 8.57 Bq/kg to 19.4 Bq/kg, respectively with the mean values of 43.8 Bq/kg and 15.8 Bq/kg, respectively. However, the minimum and maximum values of ^{40}K are 297 Bq/kg and 448 Bq/kg, respectively with the mean value of 358 Bq/kg. In this case, the overall mean activities of ^{238}U and ^{40}K in the studied samples were found as lower than that of the world mean values of 33 and 474 Bq/kg [69], respectively.

However, the total radium equivalent activity (Ra_{eq}) calculated from the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in Cox's Bazar beach sands are in the range from 80.2 to 126 Bq/kg with an average value of 106 Bq/kg, which is lower than the world average value of 370 Bq/kg [70]. Moreover, the mean value of the external radiation hazard index (H_{ex}) is 0.29 which is less than 1, and it indicates that the studied areas are safe for human health [42]. Besides, it is noted that the activity concentration of ^{232}Th is comparable with the average activity concentrations in the continental crust [71].

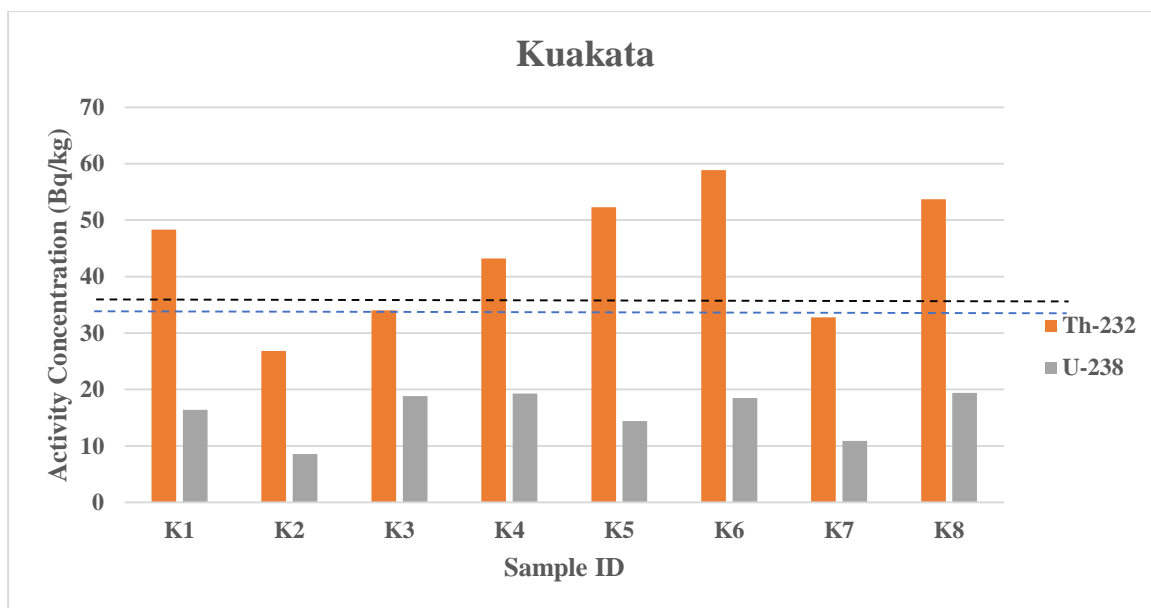


Figure 4.17: Activity concentration of ²³²Th and ²³⁸U for beach sands of Kuakata.

Figure 4.17 shows the graphical representation of the activity concentrations of all sampling points in Kuakata beach sands. From the above Figure 4.17, it can be observed that the maximum values of ²³²Th and ²³⁸U are at K6 and K8 sampling points, respectively, whereas the minimum values of both ²³²Th and ²³⁸U are at the K2 sampling point.

4.15 Statistical analysis and inter-elemental correlation

To study origin(s) and correlations of Th and U with other elements present in the beach sands, the Pearson correlation matrix was calculated using the software Statistica version 8. The correlation coefficients among the studied elements represent poor, strong, negative and positive correlations among the elements that are given in Table 4.23. According to the values of Pearson correlation coefficients, there is a strong positive correlation between Th and U (0.96) indicates the co-existence of these two elements in the beach sands.

Moreover, Th has strong positive correlations with Ti, V, Cr, Mn, Fe and REEs, which indicate that the Th rich minerals like Monazite [(REE, Th) PSYL_1] and Zircon [(Zr, Th, U, Y) SiSYL_1] predominant in the beach sands. Besides, poor or negative correlations of Na, As and Cs with Th and U indicate different origin or mechanism of Na, As and Cs than Th and U in the beach sands [72].

Table 4.23: Pearson correlation matrix for inter-element correlation in Beach sands of Bangladesh.

	Na	Al	K	Sc	Ti	V	Cr	Mn	Fe	Co	As	Cs	La	Ce	Sm	Eu	Dy	Yb	Th	U
Na	1.00																			
Al	0.14	1.00																		
K	0.43	0.05	1.00																	
Sc	-0.32	0.71	-0.49	1.00																
Ti	-0.42	0.67	-0.36	0.85	1.00															
V	-0.40	0.73	-0.39	0.87	0.98	1.00														
Cr	-0.41	0.45	-0.44	0.82	0.85	0.77	1.00													
Mn	-0.39	0.67	-0.47	0.84	0.94	0.97	0.71	1.00												
Fe	-0.25	0.71	-0.45	0.99	0.85	0.85	0.82	0.83	1.00											
Co	-0.11	0.80	-0.28	0.88	0.72	0.79	0.58	0.77	0.89	1.00										
As	0.10	0.25	-0.33	0.24	0.24	0.32	-0.02	0.48	0.28	0.43	1.00									
Cs	0.46	0.29	0.53	-0.09	-0.19	-0.16	-0.30	-0.20	-0.03	0.28	0.17	1.00								
La	-0.47	0.60	-0.36	0.79	0.97	0.96	0.79	0.95	0.79	0.70	0.27	-0.23	1.00							
Ce	-0.42	0.56	-0.48	0.93	0.91	0.87	0.93	0.84	0.93	0.74	0.15	-0.25	0.89	1.00						
Sm	-0.49	0.60	-0.37	0.78	0.97	0.97	0.78	0.95	0.78	0.70	0.28	-0.22	1.00	0.87	1.00					
Eu	-0.40	0.57	-0.52	0.95	0.88	0.85	0.92	0.81	0.95	0.75	0.12	-0.24	0.84	0.98	0.82	1.00				
Dy	-0.43	0.64	-0.39	0.85	0.99	0.97	0.84	0.95	0.85	0.72	0.28	-0.22	0.98	0.92	0.98	0.88	1.00			
Yb	-0.40	0.64	-0.43	0.87	0.99	0.96	0.87	0.94	0.88	0.73	0.25	-0.21	0.97	0.95	0.96	0.92	0.99	1.00		
Th	-0.42	0.50	-0.50	0.68	0.61	0.65	0.59	0.62	0.60	0.49	0.08	-0.27	0.53	0.56	0.53	0.62	0.57	0.58	1.00	
U	-0.37	0.45	-0.42	0.52	0.48	0.55	0.41	0.53	0.43	0.38	0.09	-0.22	0.41	0.37	0.42	0.43	0.44	0.43	0.96	1.00

Marked correlations are significant at $P < 0.01$.

CHAPTER 5

DISCUSSION ON RESULTS AND RELEVANCE

5.1 Comparison of the determined elemental concentration with literature data

The elemental concentration of the studied elements obtained in the beach/inland sands of Bangladesh were compared with available literature from different countries and the world mean value (shown in Table 5.1).

In this study, for Cox's Bazar beach sands, the minimum to maximum values obtained for U was from 1.23 mg/kg to 132 mg/kg with a mean value of 13.84 mg/kg which was significantly greater than those values of the literature (shown in Table 5.1), and the reason was high mineral deposition in that beach sands. The mean value of U was 5.13 times higher than the world average value (UNSCEAR, 2000). Besides, the mean value obtained for Th was 48.4 mg/kg which was also significantly greater than the literature data [73], almost 4.61 times higher than the world average value (UNSCEAR, 2000).

In Patenga beach sands, the obtained mean value (3.10 mg/kg) with the range of 1.25-10.2 mg/kg for U in this research was 1.15 times higher than the world average value (UNSCEAR, 2000) and comparable to Patenga, Kuakata, Iran, Thailand, China and Mexico, but much lower than Pakistan and Jordan. Besides, the mean elemental concentration (20.75 mg/kg) obtained for Th was almost two times higher than the world average value (10.5 mg/kg) and comparable to Patenga, China, Iran, Thailand and Mexico, but lower than Pakistan and Kuakata. This variation was because of the higher content of heavy minerals like Monazite, Kyanite, etc., in the beach sands. However, the distinction in the concentration of Th and U in the soil/sands was found due to the differences in geological properties of soil and their fractionation during weathering [33].

Table 5.1: Comparison of elemental concentrations of Th and U in beach/inland sands in this study with some selective literature data in the world.

In this research		Th(mg/kg)	U(mg/kg)
Cox's Bazar sand, Bangladesh	Range	4.63-382	1.23-132
	Mean	48.4	13.84
Patenga sand, Bangladesh	Range	9.07-67.4	1.25-10.2
	Mean	20.75	3.10
Kuakata sand, Bangladesh	Range	6.63-14.6	0.73-1.65
	Mean	10.82	1.34
Moulvibazar red soil, Bangladesh	Range	6.86-20.8	0.97-4.57
	Mean	15.02	2.74
Other's soil (Sherpur, Rajshahi, & Sylhet), Bangladesh	Range	0.55-13.4	1.36-2.96
	Mean	4.63	2.08
Literature data			
Cox's Bazar, Bangladesh [73]		52.83	-
Patenga, Bangladesh [74]		18.7	2.97
Patenga, Bangladesh [33]		17.1	3.98
Kuakata, Bangladesh [41]		23.01	2.37
Hurgada, Egypt [75]		5.50	1.66
Xianyang, China [76]		11.03	2.50
Persian Gulf, Iran [77]		6.38	2.81
Chalatat and Samila Beach, Thailand [78]		15.7	3.33
Peshwar, Pakistan [79]		20.6	5.23
Acapulco, Mexico [80]		7.67	2.60
Chachalacas, Mexico [81]		4.84	1.52
Rajasthan, India [82]		10.6-26.1	2.4-6.3
Russaifa, Jordan [83]		2.1-6.7	3.9-42.4
South Africa [84]		1.4-18.6	0.8-4.2
Cyprus [85]		< 1-9.8	< 1-3.2
Extractable range [86]		Not specified	>200mg/kg
World Average [57]		10.5	2.7

For Kuakata beach sands, the mean values due to U obtained in this work (1.34 mg/kg) were comparable to those for Egypt, Mexico and Cyprus, but lower than the remaining countries and the world average value. In the case of Th, the mean value (10.82 mg/kg) was similar to China, Mexico and the world average value, but lower than the remaining countries.

For inland sands (Moulvibazar and other sampling points), the mean values of U obtained in this research (2.74 and 2.08 mg/kg) were similar to those for Patenga, Kuakata, China, Iran and Mexico (Table 5.1). Besides, the derived values of Th in this research (15.02 mg/kg and 4.63 mg/kg) were comparable to China, Iran, Egypt, Mexico, Thailand. Moreover, the reported data for Cox's Bazar (52.83 mg/kg) was significantly higher because of the presence of high heavy mineral deposition in that beach sands. The higher content of heavy minerals like Monazite, Kyanite, etc., in the beach sands and inland sands of Bangladesh, cause this difference.

5.2 Comparison of the determined activity concentration with literature data

Naturally occurring radioactive materials (NORM) constituted about 85% of public exposure to terrestrial gamma radiation due to the presence of primordial ^{238}U and ^{232}Th and ^{40}K radionuclides in the atmosphere (UNSCEAR, 2000). They could be present in many ores and minerals, sands, clays, soils and stones, commodities, recycled residues, and other appliances used by humans. Although their distribution was not uniform, it depended on the lithology and degree of mineralization of the rock formation in a given area, as shale and phosphate rocks were relatively high levels of radiation [87, 88]. The activity concentration of radionuclides such as ^{238}U and ^{232}Th and ^{40}K for inland and beach sand samples of Bangladesh, their maximum and minimum values, mean, literature data, as well as the world average values, are given in Table 5.2.

Table 5.2: The mean activity concentration (Bq/kg) of radionuclide in beach sands and inland sands compared with literature data.

In this research	⁴⁰K	²³²Th (Bq/kg)	²³⁸U (Bq/kg)
Cox's Bazar	357 (202-485)	196 (18.8-1546)	163 (14.4-1551)
Patenga	467 (334-633)	96.9 (49.0-345)	51.2 (25.3-177)
Kuakata	358 (297-448)	43.8 (26.8-58.9)	15.8 (8.57-19.4)
Moulvibazar	445 (214-552)	60.8 (27.8-84.0)	32.2 (11.4-53.7)
Others	383 (87.7-491)	18.7(2.23-54.2)	24.4(16.0-34.7)
Literature data			
Cox's Bazar, Bangladesh (Zircon) [39]	-	10405	14849
Cox's Bazar, Bangladesh (Zircon sand) [89]	-	1324	6438
Patenga beach, Bangladesh [33]	512.2	69.5	49.5
Kuakata sea beach (sand), Bangladesh [41]	551.2	93.7	29.5
Kuakata sea beach (soil), Bangladesh [42]	874.9	90.7	29.2
Sylhet, Bangladesh [42]	497.91	125.3	55.3
Dhaka City, Bangladesh [90]	402 - 705	9 - 22	21 - 43
USA [3]	100 - 700	4 - 130	4 - 140
Louisiana (USA)(soil) [91]	43 - 729	50 - 190	43 - 95
Nile Delta (Egypt) (soil) [92]	316	19	17
Kalpakkam, Tamilnadu (soil) [93]	200 - 854	15 - 776	5 - 71
Kalpakkam, Tamilnadu (sand) [93]	324 - 405	352 - 3872	36 - 258
Peshwar, Pakistan [79]	646	84	65
Soil, Kuwait [45]	333.20	12.70	16.99
China [94]	9-1800	1-360	2-690
Malaysia [94]	170-430	63-110	49-86
Abuja, northcentral Nigeria [46]	119-750	45-98	18-37
Porto Seguro, Brazil [95]	25	1735	313
World Average [69]	474	36	33

From Table 5.2, it can be observed that the values of 163 Bq/kg and 51.2 Bq/kg, respectively were the mean activity concentration of ²³⁸U in the Cox's Bazar and Patenga beach sands of

Bangladesh and those were 4.94 times and 1.55 times higher than the world average value (33 Bq/kg) [69].

From literature data, Cox's Bazar beach sands were shown significantly higher values than the world mean value due to heavy mineral zircon and zircon sands in that area [22, 23]. Besides, the mean value obtained due to ^{238}U in this research (163 Bq/kg) was also significantly higher than those of Kuakata, Egypt, Tamilnadu, Pakistan, China, Kuwait, Malaysia, Nigeria, etc., but lower than Brazil (313 Bq/kg) [69]. Moreover, the results in the present work except Patenga and Cox's Bazar were also shown a good agreement with those reported in literature data. In general, all results except some countries exist (Table 5.2) within the range given in the United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR (2000). Finally, from the above discussion, it stated that the origin of Th and U concentrations in rocks and sands may vary because of alteration or metamorphic processes [63].

A total of 17 heavy mineral deposits were identified by the Beach Sand Minerals Exploitation Centre (BSMEC) of the Bangladesh Atomic Energy Commission (BAEC). Among them, ilmenite, magnetite, garnet, zircon and rutile were the principal potential mineral components [96]. In the 1960s, highly enriched U (greater than 1000 mg/kg) was mined, but today very low-grade ores are successfully extracted from the mines, down to around 0.02% U (200 mg/kg) [86]. While ore deposits contain a large amount of Th and U compared to the current values in placer sands, as seen in many Australian deposits, less concentration can be considered economically extractable [97]. Besides, Beach sands contain a large amount of Th and U in Cox's Bazar, Bangladesh, compared to other studied areas in this study. The presence of Th and U has found in the zircon and monazite. Moreover, if the heavy mineral zircon and monazite are enriched in the laboratory, Th and U can be extracted from the placer sands of Cox's Bazar [43].

CHAPTER 6

CONCLUSIONS AND RECOMMENDATIONS

6.1 Conclusions

In this study, an ENAA method with a cadmium filter was implemented at BAEC TRIGA research reactor for the first time to determine trace levels of As, Sb, Sm and U without any chemical dissolution and separation, and compared with INAA. It was confirmed that ENAA was more preferable to INAA for the determination of low levels of As, Sb, Sm and U in soil/sand samples. The ENAA and INAA techniques combinedly can be used for the determination of many elements, especially, trace levels of U in soil/sand samples to find out potential U-mining areas in Bangladesh.

A total of 20 elemental concentrations (Na, Al, K, Sc, Ti, V, Cr, Mn, Fe, Co, As, Cs, La, Ce, Sm, Eu, Dy, Yb, Th and U) with special emphasis on Th and U in inland and beach sands from 49 sampling points of Cox's bazar, Potenga, Kuakata, Moulvibazar, Sylhet, Sherpur and Rajshahi area were determined by INAA and ENAA techniques. The concentration data of Th and U in soil/sand samples from this research were compared with available literature data of the world which indicate that the mean concentration of Th (48.4 mg/kg) and U (13.8 mg/kg) in Cox's Bazar were significantly higher than the world average value and most of the literature data, whereas the concentrations of those elements at Potenga beach sands were comparable with UCC and literature data. However, the other study areas such as Kuakata, Moulvibazar, Sylhet, Rajshahi and Sherpur showed similar and sometimes lower concentration levels than the world average value and the literature data.

From this study, the depth-wise concentration variations for Th and U in beach and inland sands revealed that the overall U concentration variation decreased top to the deeper layer, but the concentration variation for beach sands was higher than that of the inland sands of Bangladesh.

To find out the origin and correlation of Th and U with other studied elements, statistical analysis like the Pearson correlation study revealed that high levels of Th and U in beach sands were due to the presence of Th and U rich heavy minerals in the Cox's Bazar beach sands. Finally, concentrations of U in Bangladeshi inland and beach sands obtained from this study indicate that the maximum concentration of U (132 mg/kg) was below the general extractable concentration range (>200 mg/kg), however, some efficient chemical method(s) can be used/developed to extract U from beach sands of Bangladesh.

The natural radioactivity concentrations (^{232}Th , ^{238}U and ^{40}K) and radiological hazard indices indicated that average radioactivity concentrations of ^{232}Th and ^{238}U in Cox's Bazar beach sands were relatively higher than those at other studied areas of Bangladesh as well as the world average values. However, external hazard indices were below the safe limit at most of the sampling points.

6.2 Recommendations

1. Some efficient chemical process(es) can be developed in the future to find out a cost-effective way to extract U from Bangladeshi soil/sands.
2. In the future, the concentrations of Th and U in soil/sand samples from all over Bangladesh can be collected and analyzed for mapping of Th and U distribution in the country.

References

- [1] New Mexico Bureau of Geology & Mineral Resources (NMBGMR), “*New Mexico Institute of Mining & Technology 1971*”. A Division of New Mexico Institute of Mining and Technology, Socorro, New Mexico, 1971.
- [2] Trimble, C.A, “Absolute counting of Alpha decay and the Radioactivity in water from hot spring national park”, *M.Sc. Thesis*, University of Arkansas, 1968.
- [3] UNSCEAR, “*Source and effect of ionizing radiation 1993*”. Report to the general assembly, Scientific Annexes, United Nations, 1993.
- [4] R. C. Ramola, V. Choubey, G. Prasad, G. S. Gusian Z. Tosheva, and A. Kies, “Radionuclide analysis in the soil of Kumaun Himalaya, India, using gamma ray spectrometry,” *Journal of Research Communication and Current Science*, vol. 100, no. 6, pp. 906-914, 2011.
- [5] Agency for Toxic Substances and Disease Registry (ATSDR), “*Toxicological profile for Thorium 1999*”. Department of Health and Human Services, Public Health Service, Atlanta, GA: U.S., 1999.
- [6] K. Harmsen, and F. A. M. De Haan, “Occurrence and behavior of uranium and thorium in soil and water,” *Netherlands Journal of Agricultural Science*, vol. 28, pp. 40-62, 1980.
- [7] World-nuclear.org2020/information-library/nuclear-fuel-cycle/mining-of-uranium.
- [8] M. Rahman, M. A. Islam, and R. A. Khan, “Characterization of chemical elements in common spices of Bangladesh for dietary intake and possible health risk assessment by INAA and AAS techniques,” *Journal of Radioanalytical and Nuclear Chemistry*, vol. 318, pp. 1347-1357, 2018.
- [9] G. V. Hevesy, and H. Levi, *The action of neutrons on the rare earth elements*. Germany: Ejnar Munksgaard, 1936.
- [10] H. J. M. Bowen, *Activation analysis*. Boston: MA, 1975.
- [11] M. D. Glascock, “An overview of neutron activation analysis for research reactor, University of Missouri, Colombia,” *Malayasian Magazine of Physics*, vol. 1, pp. 41-45, 1996.
- [12] J. C. Laul, “Neutron activation analysis of geological materials,” *Journal of Atomic Energy Review*, vol. 17, no. 3, pp. 603-695, 1979.
- [13] M. F. Soto-Jimenez, and F. Paez-Osuna, “Distribution and normalization of heavy metal concentrations in mangrove and lagoonal sediments from Mazatlan Harbor (SE Gulf of California),” *Journal of Estuarine, Coastal and Shelf Science*, vol. 53, no. 3, pp. 259-274, 2001.
- [14] R. Khan, N. Shirai, and M. Ebihara, “Chemical characteristic of R chondrites in the light of P, REEs, Th and U abundances,” *Earth and Planetary Science Letters*, vol. 422, pp. 18-27, 2015.
- [15] P. L. Barnard, A.C. Foxgrover, E. P. Elias, L. H. Erikson, J. R. Hein, M. McGann, and F. L. Wong, “Integration of bed characteristics, geochemical tracers, current measurements, and numerical modeling for assessing the provenance of beach sand in the San Francisco Bay Coastal System,” *Marine Geology*, vol. 345, pp. 181-206, 2013.

- [16] P. Schramel, and L. Q. Xu, "Determination of arsenic, antimony, bismuth, selenium and tin in biological and environmental samples by continuous flow hydride generation ICP-AES without gas-liquid separator," *Fresenius Journal of Analytical Chemistry*, vol. 340, no. 1, pp. 41-47, 1991.
- [17] H. Tuovinen, D. Vesterbacka, E. Pohjolainen, D. Read, D. Solatie, and J. Lehto, "A comparison of analytical methods for determining uranium and thorium in ores and mill tailings," *Journal of Geochemical Exploration*, vol. 148, pp. 174–180, 2015.
- [18] E. M. E. Gammal, and S. Ahmed, "Comparison of four analytical techniques for determining uranium in different rock samples," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 322, no. 3, pp. 1-8, 2019.
- [19] M. A. Islam, A. Al-Mamun, F. Hossain, S. B. Quraishi, K. Naher, R. Khan, and F. Nahid, "Contamination and ecological risk assessment of trace elements in sediments of the rivers of Sundarban mangrove forest, Bangladesh," *Marine Pollution Bulletin*, vol. 124, no. 1, pp. 356-366, 2017.
- [20] M. A. Rakib, M. E. Huda, S. M. Hossain, K. Naher, R. Khan, M. S. Sultana, and M. A. Patwary, "Arsenic content in inactive tissue: human hair and nail," *Journal of Scientific Research and Reports*, pp. 522-535, 2013.
- [21] P. J. Potts, *A handbook of silicate rock analysis*. New York, 1992.
- [22] R. Acharya, A. D. Shinde, S. Jeyakumar, M. K. Das, A. V. R. Reddy, "Determination of trace concentrations of thorium in uranium oxide matrix by epithermal instrumental neutron activation analysis," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 298, pp. 449-453, 2013.
- [23] N. Rodríguez, M. Yoho, S. Landsberger, "Determination of Ag, Au, Cu and Zn in ore samples from two Mexican mines by various thermal and epithermal NAA techniques," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 307, pp. 955–961, 2016.
- [24] X. Hou, K. E. wang, and C. Chai, "Epithermal neutron activation analysis and its application in the miniature neutron source reactor," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 210, pp. 137-148, 1996.
- [25] S. Y. Abdo, O. G. Dului, I. Zinicovscaia, "Epithermal neutron activation analysis of major and trace elements in Red Sea scleractinian corals," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 314, pp. 1445–1452, 2017.
- [26] M. S. Islam, M. M. Haque, M. A. Salam, M. M. Rahman, M. R. I. Khandokar, M. A. Sardar, P. K. Saha, A. Haque, M. A. Malek Sonar, M. M. Uddin, S. M. S. Hossain, and M. A. Zulquarnain, "*Operation Experience with the 3 MW TRIGA Mark-II Research Reactor of Bangladesh 2004*". Bangladesh Atomic Energy Commission, Dhaka, Bangladesh, 2004.
- [27] M. M. Zaved, M. A. Islam, and A. Hossain, "Experimental cross sections of the ^{174}Yb (n, γ) ^{175}Yb reaction at new energies of 0.0334 eV and 0.0536 eV," *Journal of Radiation Physics and Chemistry*, vol. 166, pp. 108471, 2020.
- [28] M. A. Islam, S. Mahmud, S. M. Hossain, A. Sk. Latif, and M. H. Ahsan, "Elemental analysis of raw materials of nuclear reactor shielding to develop low activation concrete," *Journal of Nuclear Science*, vol. 3, pp. 7-14, 2016.
- [29] S. J. Parry, "Epithermal Neutron Activation Analysis of Short-Lived Nuclides in Geological Material," *Journal of Radioanalytical Chemistry*, vol. 72, no. 1-2, pp. 195–207, 1982.

- [30] R. K. Tokay, M. Skalberg, and G. Skarnemark, "A Technique for Reducing Interferences in Epithermal Neutron Activation Analysis," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 3, pp. 265–271, 1985.
- [31] S. D. Craig, and D. E. Ryan, "Epithermal Neutron Activation Analysis with a SLOWPOKE Nuclear Reactor," *Canadian Journal of Chemistry*, vol. 59, no. 10, pp. 1470–1475, 1981.
- [32] R. Khan, A. Rouf, S. Das, and S. M. Hossain, "Spatial and Multi-Layered Assessment of Heavy Metals in the Sand of Cox's-Bazar Beach of Bangladesh," *Journal of Regional Studies in Marine Science*, vol. 16, pp. 171–180, 2017.
- [33] S. Yasmin, B. S. Barua, M. U. Khandaker, M. Kamal, M. A. Rashid, S. A. Sani, and D. A. Bradley, "The presence of radioactive materials in soil, sand and sediment samples of Potenga sea beach area, Chittagong, Bangladesh: Geological characteristics and environmental implication," *Journal of Results in Physics*, vol. 8, pp. 1268-1274, 2018.
- [34] A. K. M. A. Ullah, "Uranium Determination in Water, Soil and Stone Through Adsorptive Accumulation of U (VI) -Chloranilic Acid Complex," *Journal of Nuclear Science and Application*, vol. 27, no. 1, pp. 37–43, 2018.
- [35] A. K. M. Atique. Ullah, I. Khalil, and A. K. M. Fazle Kibria, "Determination of Uranium in Stones by Stripping Voltammetry Following Adsorptive Accumulation of the U (VI) - Chloranilic Acid Complex," *International Research Journal of Engineering & Technology (IRJET)*, vol. 2, pp. 705–712, 2015.
- [36] M. Rahman., "Determination of uranium and thorium in rock samples from Hararganj Anticline by instrumental neutron activation analysis," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 173, pp. 17-22, 1993.
- [37] M. R. Kumar; M. I. Khalil, S. Karmaker, and R. Khan, "Uranium Potentiality of Sandstones Collected from North - East Part of Bangladesh," *Journal of East China University of Technology*, vol. 39, pp. 25–31, 2016.
- [38] M. I. Khalil, R. K. Majumder, and M. Z. Kabir "Assessment of Natural Radioactivity Levels and Identification of Minerals in Brahmaputra (Jamuna) River Sand and Sediment, Bangladesh," *Radiation Protection and Environment*, vol. 39, no. 4, pp. 204-211, 2017.
- [39] M. Zaman, M. Schubert, and S. Antao, "Elevated Radionuclide Concentrations in Heavy Mineral-Rich Beach Sands in the Cox's Bazar Region, Bangladesh and Related Possible Radiological Effects," *Journal of Isotopes in Environmental and Health Studies*, vol. 48, no. 4, pp. 512–525, 2012.
- [40] M. N. Alam, M. I. Chowdhury, M. Kamal, and M. M. Ansary, "The ^{226}Ra , ^{232}Th and ^{40}K Activities in Beach Sand Minerals and Beach Soils of Cox's Bazar, Bangladesh," *Journal of Environmental Radioactivity*, vol. 46, pp. 243–250, 1993.
- [41] K. M. N. Islam, D. Paul, M. M. R. Bhuiyan, A. Akter, B. Neher, and S. M. Islam, "Study of environmental radiation on sand and soil samples from Kuakata sea beach of Patuakhali," *Journal of Environmental Protection*, vol. 3, pp. 1078-1084, 2012.
- [42] A. Miah, M. M. H. Miah, M. Kamal, M. I. Chowdhury, and M. Rahmatullah, "Natural radioactivity and associated dose rates in soil samples of Malnichera Tea Garden in Sylhet

district of Bangladesh," *Journal of Nuclear and Particle Physics*, vol. 2, no. 6, pp. 147-152, 2012.

[43] M. M. Zaman, M. Rajib, and M.Z. Kabir, "Presence of uranium and thorium in zircon assemblages separated from beach sands of Cox's Bazar, Bangladesh," *Journal of Science, Technology and Environment Informatics*, vol. 3, no. 1, pp. 161-169, 2016.

[44] M. Tzortzis, and H. Tsertos, "Determination of thorium, uranium and potassium elemental concentrations in surface soils in Cyprus," *Journal of Environmental Radioactivity*, vol. 77, pp. 325-338, 2004.

[45] A. D. Bajoga, A. N. Al-Dabbous, A. S. Abdullahi, N. A. Alazemi, Y. D. Bachama, and S. O. Alaswad, "Evaluation of elemental concentrations of uranium, thorium and potassium in top soils from Kuwait," *Journal of Nuclear Engineering and Technology*, vol. 51, pp. 1638-1649, 2019.

[46] O. Maxwell, H. Wagiran, N. Ibrahim, S. K. Lee, and S. Sabri, "Comparison of activity concentration of ^{238}U , ^{232}Th and ^{40}K in different Layers of subsurface Structures in Dei-Dei and Kubwa, Abuja, northcentral Nigeria," *Journal of Radiation Physics and Chemistry*, vol. 91, pp. 70-80, 2013.

[47] M. A. A. Mamun, M. K. Hasan, and S. A. K. M. Hossain, "Image of Cox's Bazar beach as a tourist destination," *Review of Business Research Papers*, vol. 9, no. 5, pp. 122-138, 2013.

[48] S. J. Parry, *Activation Spectrometry in Chemical Analysis*. John Wiley & Sons, 1991.

[49] General Atomic Company, "3 MW TRIGA MARK-II pulsing reactor and support facilities for Bangladesh Atomic Energy Commission 1980". Bangladesh Atomic Energy Commission, Dhaka, Bangladesh.

[50] M. Messaoudi, and S. Begaa, "Dietary intake and content of some micronutrients and toxic elements in two algerian spices (*Coriandrum sativum L.* and *Cuminum cyminum L.*)," *Biological Trace Element Research*, vol. 188, no. 2, pp. 508-513, 2019.

[51] A. Wytttenbach, "Coincidence losses in activation analysis," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 8, no. 2, pp. 335-343, 1971.

[52] M.A. Islam, M. Ebihara, Y. Toh, Y. Murakami, and H. Harada, "Characterization of multiple prompt gamma-ray analysis (MPGA) system at JAEA for elemental analysis of geological and cosmochemical samples," *Journal of Applied Radiation and Isotopes*, vol. 70, pp 1531-1535, 2012.

[53] An Advisory Group Meeting, "Use of research reactors for neutron activation analysis 1998". Report on IAEA-TECDOC-1215, Vienna, 1998.

[54] K. Kostadinov, and R. Djingova, "Trace element analysis of biological materials by thermal and epithermal neutron activation analysis," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 63, no. 1, pp. 5-12, 1981.

[55] A. Simonits, L. Moens, F. De Corte, A. De Wispelaere, A. Elek, and J. Hoste, "ko-measurements and related nuclear data compilation for (n, γ) reactor neutron activation analysis," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 60, no. 2, pp. 461-516, 1980.

[56] S. Begaa, M. Messaoudi, and A. Benarfa, "Statistical approach and neutron activation analysis for determining essential and toxic elements in two kinds of Algerian Artemisia plant," *Journal of Biological Trace Element Research*, 2020.

<https://doi.org/10.1007/s12011-020-02358-7>

[57] R. L. Rudnick, and S. Gao, *Composition of the continental crust*. University of Maryland, College Park: USA, 2014.

[58] International Atomic Energy Agency (IAEA), "*Construction and use of calibration facilities for radiometric field equipment 1989*". Technical reports no 309, Vienna, 1989.

[59] I. Alnour, N. Ibrahim, and I. Hossain, "Concentrations of ^{214}Pb , ^{214}Bi in ^{238}U series and ^{208}Tl , ^{228}Ac in ^{232}Th series in granite rock in (Kadugli) Sudan," *Indian Journal of Pure and Applied Physics*, vol. 50, pp. 285-288, 2012.

[60] NCRP: "*National Council on Radiation Protection and Measurements 1987*". Exposure of the population in the united states and Canada from natural background radiation, NCRP report no. 94, Maryland, 1987.

[61] R. Khan, S. Das, S. Kabir, M. A. Habib, K. Naher, M. A. Islam, and S. M. Hossain, "Evaluation of the elemental distribution in soil samples collected from ship-breaking areas and an adjacent island," *Journal of Environmental Chemical Engineering*, vol. 7, pp. 103189, 2019.

[62] S. Yasmin, B. S. Barua, M. U. Khandaker, M. Kamal, M. A. Rashid, S. A. Sani, and D. A. Bradley, "The presence of radioactive materials in soil, sands and sediment samples of Potenga sea beach area, Chittagong, Bangladesh: Geological characteristics and environmental implication," *Journal of Results in Physics*, vol. 8, pp. 1268-1274, 2018.

[63] M. Verdova, P. Chiozzi, and V. Pasquale, "Heat-producing radionuclides in metamorphic rocks of the Brianconnais-Piedmont Zone (Maritime Alps)," *Eclogae Geologicae Helvetiae*, vol. 94, pp. 213-219, 2001.

[64] D. R. Mayer, W. Kosmus, H. Poggitsch, and W. Beyer, "Essential trace elements in humans," *Journal of Biological Trace Element Research*, vol. 37, no. 1, pp. 27-37, 1993.

[65] G. M. S. Abraham, and R. J. Parker, "Assessment of heavy metal enrichment factors and the degree of contamination in marine sediments from Tamaki Estuary, Auckland, New Zealand," *Environmental monitoring and assessment*, vol. 136, pp. 227-238, 2008.

[66] H. Bagheri, M. Kamali, S. M. Mostajaboddavati, K. Raeissi, and M. R. Zare, "Radionuclide and trace elements geochemistry of the Urmia Lake Shore of Iran," *Journal of Environmental earth sciences*, vol. 78, pp. 373, 2019.

[67] J. Beretka, and P. J. Mathew, "Natural radioactivity of Australian building materials, industrial wastes and by-products," *Journal of Health physics*, vol. 48, pp. 87-95, 1985.

[68] X. Lu, and Z. Xiolan, "Measurement of natural radioactivity in sands samples collected from the Booje Weihe sands park, China," *Journal of Environmental Geology*, vol. 50, pp 977-988, 2006.

[69] UNSCEAR, "*Sources and effects of ionizing radiation 2000*", Report to General Assembly, with Scientific Annexes, United Nations, New York, 2000.

[70] K. A. Kabir, S. M. A. Islam, and M. M. Rahman, "Distribution of radionuclides in surface soil and bottom sediment in the district of Jessore, Bangladesh and evaluation of radiation hazard," *Journal of Bangladesh Academy of Sciences*, vol. 33, pp. 117-130, 2009.

[71] M. Eisenbud and T. F. Gesell, *Environmental radioactivity from natural, industrial and military sources*. USA, 1997.

- [72] U. Tamim, R. Khan, Y. N. Jolly, K. Fatema, S. Das, K. Naher, and S. M. Hossain, "Elemental distribution of metals in urban river sediments near an industrial effluent source," *Journal of Chemosphere*, vol. 155, pp. 509-518, 2016.
- [73] Ansary, M. M., "Study of radioactivity in beach sand minerals and soils for the estimation of radiation dose", *Doctoral dissertation, M. Sc. Thesis*, Chittagong University, Bangladesh, 1997.
- [74] S. Ghose, M. Kamal, M. I. Chowdhury, M. N. Alam, and M. N. Islam, "Gamma radiation dose from the naturally occurring radionuclides in soil of the Potenga Sea beach area of Bangladesh," *Journal of Nuclear Science and Applications*, vol. 12, pp. 31-36, 2003.
- [75] A. M. El-Arabi, "Natural radioactivity in sand used in thermal therapy at the Red Sea Coast," *Journal of Environmental Radioactivity*, vol. 81, pp. 11-19, 2005.
- [76] X. Lu, and X. Zhang, "Measurement of natural radioactivity in beach sands from Rizhao bathing beach, China," *Journal of Radiation protection dosimetry*, vol. 130, pp. 385-388, 2008.
- [77] M. R. Abdi, M. Kamali, and S. Vaezifar, "Distribution of radioactive pollution of ^{238}U , ^{232}Th , ^{40}K and ^{137}Cs in northwestern coasts of Persian Gulf, Iran," *Journal of Marine Pollution Bulletin*, vol. 56, pp. 751-757, 2008.
- [78] Benjakul, S., and Kessaratikoon, P., "Natural Radionuclide Distribution in Soil from Muang District in Songkhla Province", *Doctoral dissertation, M. Sc. Thesis*, Thaksin University, Songkhla, 2007.
- [79] S. Ali, M. Tufail, K. Jamil, A. Ahmad, and H. A. Khan, "Gamma-ray activity and dose rate of brick samples from some areas of North West Frontier Province (NWFP), Pakistan," *Science of the Total Environment*, vol. 187, pp. 247-252, 1996.
- [80] J. S. Armstrong-Altrin, "Provenance of sands from Cazonces, Acapulco, and Bahía Kino beaches, Mexico," *Revista Mexicana de Ciencias Geológicas*, vol. 26, no. 3, pp. 764-782, 2009.
- [81] J. S. Armstrong-Altrin, R. Nagarajan, V. Balaram, O. Natalhy-Pineda, "Petrography and geochemistry of sands from the Chachalacas and Veracruz beach areas, western Gulf of Mexico, Mexico: constraints on provenance and tectonic setting," *Journal of South American Earth Sciences*, vol. 64, pp.199-216, 2015.
- [82] N. Rao, S. S. Bhati, P. R. Seshu, and A. R. Reddy, "Natural radioactivity in soil and radiation levels of Rajasthan," *Journal of Radiation Protection Dosimetry*, vol. 63, no. 3, pp. 207-216, 1996.
- [83] J. Al-Jundi, "Population doses from terrestrial gamma exposure in areas near to old phosphate mine, Russaifa, Jordan," *Journal of Radiation Measurements*, vol. 35, no. 1, pp. 23-28, 2002.
- [84] F. B. Masok, P. L. Masiteng, R. D. Mavunda, P. P. Maleka, and H. Winkler, "Determination of uranium and thorium concentration and gross alpha and beta activity measurement in top soil samples from Richards Bay, South Africa," *Journal of Radiation Research and Applied Sciences*, vol. 11, no. 4, pp. 305-310, 2018.
- [85] M. Tzortzis, and H. Tsertos, "Determination of thorium, uranium and potassium elemental concentrations in surface soils in Cyprus," *Journal of Environmental Radioactivity*, vol. 77, pp. 325-338, 2004.

- [86] World-nuclear.org/information-library/nuclear-fuel-cycle/mining of Uranium/ Uranium-mining-overview.aspx.
- [87] G. Faure, 2nd Editions, *Principles of Isotope Geology*. Singapore: John Wiley & Sons, 1986.
- [88] M. T. Menager, M. J. Heath, M. Ivanovich, C. Montjotin, C. R. Barillon, J. Camp, and S. E. Hasler, "Migration of uranium from uranium-mineralised fractures into the rock matrix in granite: implications for radionuclide transport around a radioactive waste repository," *Radiochim. Acta*, vol. 66, pp. 47-83, 1993.
- [89] M. S. Alam, N. E. Huq, and M. S. Rashid, "Morphology and sediments of the Cox's Bazar coastal plain, south-east Bangladesh," *Journal of coastal Research*, pp. 902-908, 1999.
- [90] F. K. Miah, S. Roy, M. Touchiduzzaman, and B. Alam, "Distribution of radionuclides in soil samples in and around Dhaka city," *Journal of Applied Radiation and Isotopes*, vol. 49, pp. 133-137, 1998.
- [91] R. D. Delaune, G. L. Jones, and C. J. Smith, "Radionuclide concentrations in Louisiana soils and sediments," *Journal of Health Physics*, vol. 51, pp. 239-244, 1986.
- [92] N. M. Ibrahim, A. H. Abd El Ghani, S. M. Shawky, E. M. Ashraf, and M. A. Farouk, "Measurement of radioactivity levels in soil in the Nile Delta and Middle Egypt," *Journal of Health Physics*, vol. 64, pp. 620-627, 1993.
- [93] V. Kannan, M. P. Rajan, M. A. R. Ivengar, and R. Ramesh, "Distribution of natural and anthropogenic radionuclides in soil and beach sand samples of Kalpakkam (India) using hyper pure germanium (HPGe) gamma ray spectrometry," *Journal of Applied Radiation and Isotopes*, vol. 57, pp. 109-119, 2002.
- [94] United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR, "*Sources, Effects and Risks of Ionizing Radiations 1998*". Scientific Annexes, New York, 1998.
- [95] R. Veiga, N. Sanches, R. M. Anjos, K. Macario, J. Bastos, M. Iguatemy, and M. Baptista Filho, "Measurement of natural radioactivity in Brazilian beach sands," *Journal of Radiation Measurements*, vol. 41, pp. 189-196, 2006.
- [96] BSME, "*Beach Sand Minerals Exploitation Center Brochure Cox's Bazar 2020*". Scientific Information Division, Bangladesh Atomic Energy Commission, 2020.
- [97] Australia's thorium resources, "*Geoscience Australia Records 2008*". A review of the geochemical processes controlling the distribution of thorium in the earth's crust and Australia's thorium resources, Australia, 2008.

Appendix A

Uncertainty Calculation

The total uncertainty calculation of Uranium (U-238, 106 keV energy) for sample K3 (kuakata beach sands) is given below-

Various types of uncertainty parameter added in our experiment are as following

Sample and comparator preparation, $S= 0.55$

Irradiation, $T= 0.1$

Counting statistics for Sample, $C= \frac{100}{\sqrt{N_p}} = \frac{100}{\sqrt{2827}} = 1.88$

Where N_p is the peak area, $N_p= 2827$

Counting statistics for Comparator, $V= 1.34$ (this value varies for each sample)

Geometry difference, $W= 3.0$

Losses (random coincidences), $X= 0.5$

Correction, $Y= 0.3$

Calculation of Spectrometry, $Z = \sqrt{(C^2 + V^2 + W^2 + X^2 + Y^2)} = 3.83$

Uncertainty, $Un = \sqrt{(S^2 + T^2 + Z^2)} = 3.87$

The concentration of U in sample K3, $P= 1.60$ mg/kg

Total uncertainty for the sample K3 = $\frac{P*Un}{100} = 0.06$ mg/kg

Appendix B

Full Paper Publication

[1]. **M. S. Rahman**, M. A. Islam and A. Hossain, Comparison of Epithermal and Instrumental Neutron Activation Analysis for the Determination of U and Other Elements in Soil Samples using BAEC TRIGA Reactor, Accepted (Paper#226), Conference Proceedings of 'International Conference on Mechanical, Industrial and Energy Engineering 2020', 19-21 December, 2020, Khulna, Bangladesh.

[2]. **M. S. Rahman**, M. A. Islam and A. Hossain, Assessment of concentration and distribution of Thorium and Uranium in inland and beach sands of Bangladesh by NAA technique, Manuscript under preparation.

Conference Presentations

1. **M. S. Rahman**, M. A. Islam and A. Hossain, "Comparison of Epithermal and Instrumental Neutron Activation Analysis for the Determination of U and Other Elements in Soil/Sand Samples using BAEC TRIGA Reactor". Presented Oral at International Conference on Mechanical, Industrial and Energy Engineering 2020, Organized by KUET. Date 19-21 December, 2020.

2. **M. S. Rahman**, M. A. Islam, A. Hossain, K. Naher, R. Khan and U. Tamim, "Concentration and Distribution of Th, U and other Elements in Soil and Sands of the Potential Region of Bangladesh by NAA Technique". Presented Poster at 3rd International Conference on Physics for Sustainable Development and Technology 2019, Organized by CUET. Date: 18-19 December, 2019.

3. **M. S. Rahman**, M. A. Islam, M. Rahman, A. Hossain, "Enhancement of Analytical Sensitivity by Epithermal Neutron Activation Analysis to Determine Th and U in Sand of Sylhet Region". Presented Oral at International Conference on Physics-2018, organized by Bangladesh Physical Society, Dhaka University (DU). Date: 08-10 March, 2018.

