ABSTRACT

Measuring background radiations (BR) and risk assessment due to the radiation exposure is very important, in terms of different perspectives, especially in view of health threat to public due to its carcinogenic nature. Human beings are always exposed to environmental radiations. Exposure of public to ionizing radiation may lead to stochastic effects. Many computational stochastic models, developed for simulated cells irradiation, utilize probabilities and probability distribution functions to describe biophysics of cells. Such models have established a sigmoidal connection between carcinogenic risk and radiation exposure.

This study has been accomplished in different parts. In the first part of study, an extensive review of literature has been carried out. In 2nd part of study ambient concentrations of indoor/outdoor radon and gamma dose rates (GDR) were measured, using RTM 1688-2 and Ludlum micrometer 19, in the Muzaffarabad city. For indoor radon measurements, radon concentrations varied in the range of 16 to 150 Bq m^{-3} whilst, for outdoor environment from 7 to 31 Bq m^{-3}. Average values of indoor and outdoor radon concentrations were found as 46.9 and 13.3 Bq m^{-3} respectively. For indoor measurements, GDR ranges from 419 to 1486 µGy y^{-1} with mean value of 846 µGy y^{-1}. Whilst outdoor GDR varied from 495 to 1029 µGy y^{-1} with mean value 777 µGy y^{-1}. For indoor measurements, annual effective dose (E_{Rn}) due to radon exposure ranges from 0.4 to 3.78 mSv y^{-1} with average value of 1.18 mSv y^{-1}. Radon doses delivered to lungs varied from 0.97 to 9.08 mSv y^{-1} with an average value of 2.84 mSv y^{-1}. Excessive lifetime cancer risk (ELCR) varied from 1.49 $\times 10^{-3}$ to 14.01$\times 10^{-3}$ with mean value 4.38$\times 10^{-3}$.

In the 3rd part of study, the specific activities of primordial radionuclides, gross alpha and gross beta activities in soil samples collected from 29 locations of Azad

Kashmir were estimated. Soil samples were analyzed, for possible radionuclide contents and relevant health implications, by high-resolution γ-ray spectroscopy and α/β counter ASC-950-DP Protean instrument. The alpha activity varied from 77.31 ± 9.95 to 440.08 ± 16.48 Bq kg^{-1} with an overall average value of 234.88 ± 1.69 Bq kg^{-1}. While beta activity varied from the minimum detection level, i.e., <MDL to 361.55±149.33 Bq kg^{-1}, with average value for all samples estimated as 235.65 ± 149.98 Bq kg^{-1}. Specific activities of ^{232}Th, ^{226}Ra and ^{40}K were calculated using γ-ray spectroscopy and were subsequently utilized for the estimation of radiation doses and radiological hazards. Activity concentrations due to ^{40}K, ^{232}Th and ^{226}Ra were found in the ranges 213.54 ± 17.22 to 1205.83 ± 12.82, 26.11 ± 3.72 to 84.70 ± 4.63 and 13.74 ± 1.46 to 62.23 ± 4.29 Bq kg^{-1}, with average values 616.22 ± 29.20, 55.83 ± 5.74 and 37.91 ± 2.35 Bq kg^{-1} respectively, whilst, activity concentration due to anthropogenic radionuclide ^{137}Cs was found in the range from minimum detection limit, i.e., ≤0.50 to 8.82 ± 0.83 Bq kg^{-1}.

Average value for ^{137}Cs sample was found as 3.43 ± 0.28 Bq kg^{-1}. Excess lifetime cancer risk (ELCR) for indoor occupation varied from 4.94×10^{-4} to 1.82×10^{-3} and for outdoor occupation 1.32×10^{-4} to 4.62×10^{-4}. Overall excess lifetime cancer risk (ELCR) for the current study was estimated as 1.55×10^{-3}.

In the 4th part of study gross alpha, grass beta activities in medicinal plants samples collected from different districts of Azad Kashmir, Pakistan have been measured. Measured activities have been used to assess age dependent annual effective doses for infants, one year, five year, ten years, fifteen years, and adult peoples. Effect of altitude on measured values of gross α/β activities has also been investigated. For medicinal plants consumption rate (MPCR) of 1.8 kg y^{-1}, the average gross alpha and beta annual committed effective dose (ACED), delivered to infants, one, five, ten, 15 years and adults ranged from 43±7 to 1732±18 μSv y^{-1}, 7±1 to 274±3 μSv y^{-1}, 5±1 to 192±2 μSv y^{-1}, 5±1 to 181±2 µSv y^{-1}, 6±1 to 248±3 µSv y^{-1} and 3±0 to 100±1 µSv y^{-1} with mean value 797±10, 274±2, 88±1, 83±1, 114±1 and 46±1 µSv y^{-1}. For higher values of MPCR, viz. 2, 4, 6, 8 and 10 kg y^{-1} respective gross alpha and gross beta ACED goes on increasing. Finding of study shows that, except ACED delivered to infants for MPCR of 1.8 kg y^{-1}, all other estimated values, at same MPCR, fall below the WHO recommended level (290 µSv y^{-1}) and that of as reported in UNSCEAR, 2000 (0.3 mSv y^{-1} or 300 µSv y^{-1}) report.

In the 5th part of study, high resolution gamma spectrometry (using HPGe detector) have been used to determine the radionuclides distribution within selected wild herbal species found in sloppy forests areas of Azad Kashmir. Results obtained showed that activity concentrations of ^{226}Ra, ^{40}K and ^{232}Th ranged from (≤MDA-8.67±1.08, ≤MDA-243.77±22.73, ≤MDA-7.45±0.76) Bq kg^{-1} with an average values 3.21±0.64, 112.75±14.11, 6.16±2.22 Bq kg^{-1} respectively. Measured radionuclidic activities have also been employed to evaluate age dependent annual committed effective doses received by general public for age ranges of <1year, 1 year, 5 year, 10 year, 15 year and >15 year and results were found consistent to UNSCEAR safe limit for ACED (300 µSv yr^{-1}). As a final point results have been summarized and recommendations for future work have been proposed in the last part of the study.

PUBLICATIONS

1. Chand Shahzadi, Muhammad Rafique, and Abdul Jabbar. Natural and Fall out Radionuclide Concentrations in Medicinal Plants: An Overview, *J. Rad. Nucl. Appl*. 5, No. 1, 29-41 (2020).

2. Shahzadi, C., Jabbar, A., Rafique, M., Khan, M., Dilband, M., & Hayat Satti, K. (2020). Study of gross alpha, gross beta and natural radioactivity in soil samples of district Muzaffarabad. *International Journal of Environmental Analytical Chemistry*, 1-18.

3. Rafique, M., Abbasi, S., **Shahzadi, C.** *et al.* Excessive Lifetime Cancer Risk Assessment due to Short-Term Indoor/Outdoor Ambient Radon and Gamma Dose Rate Exposures. *Iran J Sci Technol Trans Sci* **45,** 2181–2190 (2021). https://doi.org/10.1007/s40995-021-01192-3

SUBMITTED PAPERS

1. Shahzadi et al. Measurement of Age Dependent Radiation Ingestion Doses due to Gross Alpha and Gross Beta Exposure from Medicinal Plants submitted in the journal *Isotopes in Environmental and Health studies* with manuscript No. Ref.: Ms. No.GIEH-2021-0024.

2. Shahzadi et al. Radiological risk assessment of natural radioactivity in some selected medicinal plants {Prepared to submit in the *Journal of Radiation Protection and Dosimetry*}.

Chapter 1

INTRODUCTION

1.1. LAYOUT OF THESIS

This thesis is divided into three chapters. First chapter introduces background of the problem with title as **Introduction.** This chapter contains information about the natural and anthropogenic radioactivity and carries out a brief review of literature. Both radioactivities in the environment and herbal plants are discussed. Study objectives are also mentioned in the first chapter. Second chapter contains informations about the study area, sampling methodologies, and measurement techniques employed for radiometric assessments. Chapter three gives detailed description about results and discussion part of the study and also contains conclusions and future recommendation.

1.2. BACKGROUND

Clean air, in public buildings, is a fundamental requirement of peoples for healthy lives and their well-beings. Building materials and many human indoor activities may lead to broad range of health threats that prove fatal depending upon level of exposure. Globally, and especially in developing countries, indoor exposure from hazardous substances causes significant damage to health. Radon is one of the indoor and outdoor pollutants, sustained exposure of which may pose substantial health intimidations. From many residential epidemiological studies, the evidence of lung cancer risk from radon exposure have been established and it is classified as a human carcinogen, group I, by International Agency for Research on Cancer (IARC) (Mustafa & Vasisht, 1987; Nero, 1988; Rahman et al. 2009; WHO, 2010; Rafique et al., 2011).

Radon stems its origin in radium, with half-life of 1600± 7 y, which by itself is a decay product of ^{238}U present in uranium ore. ^{222}Rn, with half-life of 3.82 d, decays to a series of short-lived progenies viz. ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po. When radon is inhaled, being inert radioactive gas, it is often exhaled without producing any damage. However, radon progenies being atoms of heavy metals deposit their selves in human respiratory system and continuously irradiate lung tissues causing severe impairment (Sahu et al., 2014).

As described in WHO, (2010) guidelines report, that without the threshold, the relationship between radon exposure and response can be best described as linear. Based upon long term (30 years) average radon exposure, for every 100 Bq m^{-3} increase in radon concentration, the excess relative risk is about 16 %. Public exposure to radon usually comes from inhalation or ingestion. Radon is source of internal exposure, whilst background radiation levels (BRL), consisting of gamma particles, contribute towards external exposure. BRLs coming from naturally occurring radionuclides, from soil, sand, and rocks are the major source of external radiations. In the world some regions have high background radiations and are termed as high background radiation areas (HBRAs) (WHO, 2010).

BRLs depend mostly on geographical and geology of the area and vary worldwide from approximately 2 to 8×10^{3} μSvy^{-1} (Ojovan et al., 2019). Highest BRLs, due to high concentration of radioactive minerals, are found in countries like Brazil, India and China. Monazite sand deposits along beaches in Brazil are the source of external radiation levels up to ~50 μGyh^{-1}. Annual effective natural background radiation doses resulting from inhalation of radon ranges from 0.2 to 10 mSv with worldwide average of 1.26 mSv, whilst from terrestrial gamma rays annual effective background radiation doses ranges from 0.3 to 1 mSv with worldwide average of 0.48 mSv (Ojovan et al., 2019).

Human exposure to ionizing radiations arises due to natural and artificial sources. Natural sources contribute via radiations coming from outer space or from primordial radionuclides. Whilst, radiations with artificial origin are usually generated due to different human activities viz. in medical diagnostic and therapeutic processes; power generation through nuclear reactors; nuclear weapon testing; and nuclear reactor accidents (UNSCEAR, 2017).

Over the years radioactive particles have had been releasing into the environment originating from nuclear-based weapons testing and nuclear fuel cycle operations in nuclear power plants (IAEA, 2011). In past, few nuclear accidents viz., Three Mile Island in 1979, Chernobyl in 1986 and Japan earthquake along with tsunami lead Fukushima Daiichi Nuclear Power Plant (FDNPP) accidents in March 2011 have substantially increased human exposure from ionizing radiations. Radiation exposure after nuclear accidents is of great concern to the public and many organizations and scientists have proposed guidance in nuclear emergency conditions (Becker, 2004; N0, 2005; Bouville et al., 2014).

In case of Chernobyl nuclear reactor accidents, environmental radioactive contamination was primarily due to ^{137}Cs, ^{134}Cs, and ^{90}Sr. The exposure from the anthropogenic radionuclides viz. Caesium and Strontium is considered as potential threat to human health as they concentrate their selves in human muscle and bone, respectively (Moysich et al., 2002). In FDNPP accident, significant amount of ^{134}Cs, ^{137}Cs, ^{89}Sr, ^{90}Sr, and ^{131}I have been released and detected into the environment (Shimura et al., 2012; Kumamoto et al., 2017). While the radionuclides released have also reached and detected in other countries like Russia and Greece (Bolsunovsky & Dementyev, 2011). So, nuclear activities in any part of world may lead to increase in environmental background radiations throughout the globe.

On the other-hand background radiations, with origin in nature, are either originated from cosmogenic or primordial radionuclides sources. Cosmogenic radionuclides viz. ^{3}H, ^{7}Be, ^{14}C and ^{22}Na, are usually produced due to interaction of cosmic ray particles, mainly consisting of high energy proton, with earth atmospheric particles. Only those primordial radionuclides along with their progenies, formed due to nucleosynthesis process in stars, are still found on earth with half-lives comparable to the age of the earth. These primordial radionuclides are major contributors to radiation environment on earth. Source of terrestrial radiations are Uranium-238, Thorium-232 and radioactive isotope of Potassium-40. These radionuclides are widely distributed, in varying amount, throughout the earth crust and transferred to human beings through the number of known and unknown ways. Usually from soil, it is transferred to plants, food chain, and then to human beings (No, 1982).

Besides the external radiation exposure, human beings are also prone to internal radiations exposure that may come through ingestion or digestion processes. The alpha emitter radionuclides, such as ^{210}Po, ^{226}Ra, ^{232}Th, ^{234}U and ^{238} U and beta emitters like that ^{40}K, ^{210}Pb, and ^{228}Ra might be found as a main ingredient of soil, plants and water reservoirs (Barescut et al., 2011). As soon as radon abolished then net activity of alpha emitters defines gross alpha activity. Whereas, gross beta activity might be defined via net activity contents of beta emitters and ^{14}C, ^{3}H as well as other frail beta emitters exclusively. Radionuclide identification as well as radioactive contents within soil, plants and water reservoirs needed a much expensive and time-taking investigative techniques. But the simplest radioanalytical techniques are gross alpha and beta analysis that might be employed as prime screening step, being a quick, safe and secure as well as low-cost process (Ferdous et al., 2012).

Once these radionuclides are transferred within the human body, the cells, tissues and host organs become prone to energetic particles emitted by them during their decay process. Based upon energy of emitted particles the exposed cell and hence structure of exposed DNA may change, bringing many unsolicited biological effects to occur.

Radioactivity, either from natural or manmade sources, is a permanent feature of our environment. For human beings, besides having number of growing beneficial applications in industry, agriculture, nuclear medicine and power generation there are many harmful effects associated with radiation exposure. Sustained exposure of public from areas of high background radiation sources or occupational exposure of personals working in radiation field environment needs to be assessed and, if necessary, should be controlled (IAEA, 2014). These radiation exposures may come from; i). Contaminants of areas, having past nuclear activities without applying nuclear regularity control mechanism, by residual radioactive material and nuclear or radiological emergency. ii). from commodities i.e., food, drinking water, feed and construction material. iii). from natural radioactive sources like, ^{222}Rn, ^{220}Rn coming from ^{238}U and ^{232}Th decay chains. iv). exposure from cosmic radiations (IAEA, 2014).

Keeping in view the importance of subject, many researches have monitored radon and gamma radiations coming from background sources (Ulbak et al., 1988; Tufail et al., 1988, 1992; Yu et al., 1997; Virk & Sharma, 2000; Srivastava et al., 2001; Roth et al., 2002; Sannappa et al., 2003; Rafique et al., 2009, 2010, 2011a,b,c; Rahman et al., 2009, 2010a, 2010b, Nasir et al., 2014; Kearfott 2016). Scientists have developed new instruments and methodologies to understand the dynamics of radiations for reliable monitoring and assessment of risk associated with radiation exposure (Seekamp et al., 2020; Chung et al., 2020; Iqbal et al., 2020).

Several other researchers have conducted studies focusing on the measurement of radioactivity due to gross alpha, gross beta and naturally occurring radionuclides in environmental samples (Tahir et al., 2005; Yalcin & Gurler, 2007; Zorer et al., 2009; Agbalagba & Onoja , 2011; Rafique et al., 2011; Rahman et al., 2011; Rahman & Rafique, 2012; Sarap et al., 2012; Bal et al., 2012; Rahman et al., 2013; Lee et al., 2014;

Anekwe et al., 2013; Rafique et al., 2013; Ferdous et al., 2015; Ogundare & Adekoya , 2015; Adziz & Siong, 2018; Kurnaz et al., 2020; Turhan, 2020; Kadhim & Najam, 2020; Punniyakotti et al., 2020; Elsaman et al., 2020; Abbasi et al., 2020).

Considerable data is available in literature reporting activity concentrations of radionuclides in environmental and building material samples across the globe (Rahman et al., 2008; Rafique et al., 2013; Muhamad et al., 2019; Gomez et al., 1997; Wang et al., 1997; Shenber, 2011; Rafique et al., 2011; Rahman et al., 2012; Rafique and Rathore, 2013; Rafique, 2013; Rafique, 2014; Rafique et al., 2014; Reddy et al., 2017; Niranjan et al., 2018). Some of organizations and researchers have also conducted studies to find activity concentrations of radionuclides in food stuff, medicinal plants and estimated risk associated with the exposures (Alvim et al., 2006; WHO, 2007; El-Taher & Uosif, 2006; Hashem et al., 2013; Ahmad and Beg, 2001; Lee et al., 2008; Rokaya et al., 2010; Street, 212; Sharma et al., 2012; Shanmugam et al., 2012; Naidu et al., 1999; Balunas & Kinghorn, 2005; Gambari & Lampronti, 2006; Jager et al., 2010; El-Taher & Abdelhalim, 2014; Nyila et al., 2012; Durugbo et al., 2012; Robison et al., 1997; UNSCEAR, 2001; Narayana & Prakash, 2006; Turhan et al., 2007; Sussa et al., 2009; Ahmed et al., 2010; Desideri et al., 2010; Jevremovic et al., 2011; Oni et al., 2011; Sussa et al., 2011/2013; Oufni et al., 2013; Tettey-Larbi et al., 2013; Oprea et al., 2015; Kovacs et al., 2015; Pourimani et al., 2015; Najam et al., 2015; Njinga et al., 2015; Shatha et al., 2015; Harb, 2015; Shatha et al., 2015; Harb, 2015; Chandrashekara et al., 2015; Chandrashekara & Somashekarappa, 2016; Kareem et al., 2016; Abojassim et al., 2016).

Almost food of all kinds contains radionuclides in varying amount depending largely upon local geology, agricultural practices and climate of the area. Usually these radionuclides are transferred from soils to the crops and water to fish. Plants take up radioactive material contents with the nutrients needed for their growth. On consumption, these products may expose peoples to unwanted radiation.

For this purpose, it is appropriate to know the radionuclides concentrations in food and drinking water and to take necessary actions in controlling their distributions. In this part of dissertation an overview of the studies conducted across the globe for assessment of radionuclides concentration in medicinal plants have been made. Data of different radionuclides viz. ^{210}Po, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{238}U, ^{222}Rn, ^{220}Rn, ^{40}K, ^{90}Sr and ^{137}Cs, have been summarized for the sake of readers ease and interest. In literature, many studies have also reported transfer factor (TF) of radionuclides from soil to plant and estimated the values of average annual committed effective dose (AACED) due to the ingestion of Radionuclides present in medicinal plants. Knowledge of TF is very important in order to get reasonable predictive estimates for radionuclides concentrations and resulting radiation doses received by public from agricultural crops. These studies are source of baseline data that might be used in any radiological emergency or to formulate regulations related to radiological healthcare for medicinal plants of local origin

Usually plants are contaminated by radionuclide concentrations using two mechanisms; via root uptake or deposition of anthropogenic radionuclides on plants. Radionuclides occurring naturally in soil transfer via their roots and assimilated metabolically into plants. These Radionuclides transfer to human beings via major pathway of soil-plant-man (Kranrod et al., 2017; Falandysz et al., 2017).

In earliest history of humanity, ancient methods of utilizing medicinal plants and plant extracts have been employed to treat several diseases and ailments (Alvim et al., 2006). Numerous traditional systems of medicines have been employed therapeutically medicinal plants due to their significant aspect acknowledged world over. Several cultural and theoretical models formulate phytotherapy within doctrine scheme subjected to all traditional system like Amazonian to African medicinal system, Unani to Tibetan, and Ayurveda to Chinese traditional system of medicine. All such system practiced regularly and employed whole plant or parts like core ingredients of medicines. Radionuclides spontaneously exhibiting within medicinal plants are one sort of renowned residue and contaminants that might subject impairment to herbal medicinal consumers (WHO, 2007).

Soil comprises of radionuclides like ^{238}U, ^{232}Th, ^{226}Ra, ^{137}Cs, ^{40}K etc metabolically incorporated within plants and administered ultimately onto food chain. These varied radionuclides deposited in parts of plants might be reason of human exposure. Since, different parts of plants have had been used as chief medicinal ingredients, therefore, quantitative understanding might be necessary about human risk assessment linked to medicinal plants ingestion or other interrelated alleyways by which ultimately human is affected radiologically (El-Taher & Uosif, 2006).

Recently, across the globe, researchers have focused on medicinal plants exploration owing to potential as well as diversity of such medicinal plants like key ingredient of medicinal stuff (Hashem et al., 2013; Ahmad & Beg, 2001; Lee et al., 2008; Rokaya et al., 2010; Street, 212; Sharma et al., 2012). Therapeutic plants were investigated so as to depict active compound subsisting within medicinal plants to explore therapeutic features on scientific basis (Shanmugam et al., 2012; Naidu et al., 1999; Balunas and Kinghorn, 2005; Gambari & Lampronti, 2006; Jager et al., 2010; El-Taher & Abdelhalim, 2014; Nyila et al., 2012). National monitoring programmes, in many countries, has had been working to determine the levels of Radionuclides present in food. Such programmes mainly focus on finding the levels of man-made radionuclides such as ^{90}Sr, ^{137}Cs, ^{238}Pu and ^{239}Pu within food products (https://www.iaea.org/newscenter/news/naturalradioactivityinfoodexpertsdiscusharmonizng-international-standards).

Many studies found in literature have reported Radionuclides concentrations within animal as well plants metabolic system. Some of these studies have also focused on radionuclides impacts within medicinal plants (Durugbo et al., 2012).

Final section of the first part of dissertation reports radionuclide data obtained by several research groups in various locations of the world to aid as future reference database. Main focus of this section of thesis is to summarize radionuclide data obtained from medicinal plants and to asses resulting radiological doses.

1.3. STUDY AREA

Studies addressing the measurement of radionuclide concentrations, and resulting dose assessments, in medicinal herbal plants conducted in different parts of globe have been compiled and reported here. Results of studies conducted in Brazil, China, Egypt, Morocco, Slovakia, Serbia, Ghana, Iran, Iraq, Jordon, India, Italy, Turkey, Hungary, (Marshall Islands) SW Hawaii, Nigeria, Thailand, and Romania are presented here.

1.4. EXPERIMENTAL TECHNIQUES

Two major techniques, 1) gamma spectroscopy and 2) alpha spectroscopy, have been employed for the assessment of radionuclide concentrations in herbal plants. In some studies global alphas as well as beta counting technique have also been used. Gamma ray spectroscopy is usually carried out using High Purity Germanium (HPGe) detector and NaI(Tl) detector coupled to a computer interfaced multichannel analyzer (MCA). Radon (^{222}Rn) and Thoron (^{220}Rn) concentrations were measured using passive Solid State Nuclear Track techniques (SSNTD) CR-39 and LR115 type-II detectors were used for estimation of radon and Thoron. Radioactivity contents of ^{210}Po and gamma dose rate have been measured using electrochemical deposition and portable scintillator.

1.5. LITERATURE REVIEW

Results obtained from various studies conducted in different parts of the world are summarized in Table 1 and 2. Robison et al., (1997) have reported results of a survey conducted from September through November 1978 in Northern Marshall Islands to collect the radiological data and assessed resulting doses (Robison et al., 1997). They focused their study on anthropogenic radionuclides that may have contaminated the Northern Marshall Islands areas from atmospheric nuclear tests conducted at the Pacific Proving Grounds between 1946 and 1958. They have calculated external gamma exposure rate through aerial survey and radionuclide concentrations in soil, food crops, animals, well water, fish and native vegetation's (Robison et al., 1997). Samples were analyzed for ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{241}Am anthropogenic Radionuclides. Their reported results show that, via ingestion, 95 % of doses come from exposure to ^{137}Cs. Following ^{137}Cs, the second most significant contribution comes from ^{90}Sr. In case of external gamma exposure, dose via the inhalation pathway, ^{137}Cs accounts for 10 to 30 % dose and $^{239+240}$Pu and ^{241}Am are major contributors in this case. For the atolls of study, estimated maximum annual effective dose ranges from 2 to 2.1 mSv y^{-1}. Background dose was estimated as 2.4 mSv y^{-1}. Total dose, due to background and contribution from fallout radionuclides, ranges from slightly over 2.4 mSv y^{-1} to 4.5 mSv y^{-1}. Their estimated 50-y integral dose ranged from 0.5 to 65 mSv (Robison et al., 1997).

For the same Marshall Islands, UNSCEAR 2000 have reported that approximately 85–90 % of the nuclear test related dose delivered, via ingestion, to the resident population is derived from ^{137}Cs contained in locally grown food plants (UNSCEAR, 2001).

To improve the trustworthiness of predictive dose assessments from anthropogenic Radionuclides and to address resettlement and possible option of rehabilitation of Marshall Islands the Lawrence Livermore National Laboratory (LLNL) has developed an interactive internet application. This open access computer application has provided public a chance to assess radiological conditions in the Marshall Islands. User can calculate hypothetical ingestion doses from ^{137}Cs presence in food plants using the application of the ingestion dose calculator (Kehl et al., 2013).

Dufy et al. (1999) surveyed the medicinal plants in the Marshall Island, used in traditional medicine, for ^{137}Cs contents. ^{137}Cs activity concentration was measured using a high purity germanium detector (HPGe) with 40 % nominal efficiency. ^{137}Cs concentration in Polypodiumscolopendria was reported to be several folds higher as compared to other kind of plant species analyzed for the gamma spectroscopy. The highest reported ^{137}Cs contents was found in Polypodiumscolopendria ranging from (0.200 to 3) KBq kg^{-1} out of total investigated herbal plants and rest of observed medicinal plant have not significantly exhibited role for ^{137}Cs received dose (range, 0.001 to 1 KBq kg^{-1}) (Duffy et al., 1999).

Salamon and Haban, (2005) assessed some medicinal plants of Slovakia for radioactivity contents by employing gamma spectroscopy using HPGe detector. The medicinal edible plant parts like roots, herb, flowers and leaves were analyzed for ^{137}Cs and ^{134}Cs contents and activity concentrations were found in the range (0.40 to 3.20) Bq kg^{-1}. Analysis confirmed the radioactivity contents in medicinal plants verily count on radiation exposure at explicit place (Salamon & Haban, 2005).

Narayana et al., (2007) investigated ayurveda medicinal plants for radioactive contents by employing electrochemical deposition and portable scintillator and found contribution of ^{210}Po varies from 6.3 to 56.9 Bq kg^{-1} with mean value of 27.8 Bq kg^{-1}. For the investigated medicinal plants gamma dose rate was found to vary from 34.8 to 52.2 nGy/h with mean value of 43.5 nGy/h (Narayana et al., 2007).

Turhan et al., (2007) have reported radionuclide concentrations for edible mushrooms of Turkey using gamma ray spectroscopy carried out by HPGe detector. Concentrations of ^{232}Th and ^{228}Ra were reported below lower limit of detection. Activity concentrations of ^{40}K and ^{137}Cs varied from 715.5 ± 50.1 to 1779.0 ± 163.7 Bq kg^{-1} with mean value of 1150.8 ± 315.2 Bq kg^{-1} (dry matter) and 2.4 ± 0.3 -109.0 ± 7.3 Bq kg^{-1} with mean value of 28.4 ± 27.2 Bq kg^{-1} (dry matter). The mean annual effective doses due to ^{40}K and ^{137}Cs from mushrooms were found to be (0.13 ± 0.03) μSv and (7.0 ± 6.0) × 10^{-3} μSv respectively. The plants intake of ^{137}Cs was found quite low and no significant contamination was recorded for the mushroom species of local origin. Morchellaesculenta and Strophariacoronilla plant species were found with having comparatively higher contents of ^{137}Cs and ^{40}K among the all analyzed mushroom samples (Turhan et al., 2007).

Sussa et al., (2009) reported stable elements as well as radioactive concentrations in Brazillian medicinal plants through employing techniques of alpha, beta counting and neutron activation analysis. The activity concentration of ^{228}Ra, ^{210}Pb and ^{226}Ra were found as (29 ± 3-65 ± 4), (32 ± 3-76 ± 8) and (< 2.2-18.4 ± 0.2) Bq kg^{-1} respectively (Sussa et al., 2009).

Ahmed et al., (2010) using gamma spectroscopic analysis by HPGe detector, have estimated external as well internal radiation exposure due to radionuclides present in herbal plants of Egypt. Radium contents were found as 7.71 ± 0.25 Bq kg^{-1} within green tea, while, 115.08 ± 0.49 Bq kg^{-1} for gawafa. For the fall out radionuclide the concentrations of ^{137}Cs varied from minimum detection limit (MDL) to 12.62 ± 0.42 Bqkg^{-1} (Ahmed et al., 2010).

Desideri et al., (2010) have estimated activity concentrations due to anthropogenic as well as natural radioactive contents by employing alpha and HPGe spectrometer in the medicinal plants. Using alpha spectrometry, ^{238}U estimated values fall within range <0.1 to 7.32 Bq kg^{-1} and <0.12 to 30.3 Bq kg^{-1} for ^{210}Po.While, for ^{137}Cs, ^{214}Pb–^{214}Bi, ^{40}K and ^{210} Pb activity concentrations varied from <0.3 to10.7, <0.3 to 16.6, 66.2 to 3582.0 and <3 to 58.3 Bq kg^{-1} respectively (Desideri et al., 2010).

Jevremovic et al., (2011) investigated radioactivity contents within medicinal herb samples and calculated effective doses through ^{137}Cs intake and Radionuclides contents within herbal tea stuff available at Serbian market. They have employed gamma ray spectroscopic technique using HPGe spectrometer. The radioactivity contents due to ^{137}Cs, ^{238}U, ^{40}K, ^{232}Th varied from 0.3 to 8.8, 0.6 to 8.2, 126 to 1243.7 and 1.7 to15.1 Bq kg^{-1} respectively. Whilst, annual body doses via intake of ^{137}Cs as well as natural Radionuclides within herbal tea through medicinal herb consumption were reported as (2.5469.9) nSv in case of ^{137}Cs, (1026.0-132.0) nSv for ^{40}K, (0.7-9.7) nSv for ^{238}U and (0.3-2.8) nSv for ^{232}Th. Estimated doses for their study showed insignificant hazardous effects due to Radionuclides present in herbal plants (Jevremovic et al., 2011).

Oni et al., (2011) have found natural radionuclides concentrations in medicinal plants in Ughelli. These medicinal plants namely; lemon grass (Cymbopogan citrates), Spear grass (Imperata cylindrical) and Carpet grass (Eleusinindicageartin) were collected around oil and gas factories. Concentrations of primordial radionuclides were found by gamma spectroscopy using NaI(TI) detector. Average values of ^{238}U, ^{232}Th and ^{40}K estimated for lemon grass are (15.3 ± 1.7 Bq kg^{-1}), (1.1 ± 2.7 Bq kg^{-1}) and (67.9 ± 7.4 Bq kg^{-1}) respectively. For spear grass, ^{238}U, ^{232}Th and ^{40}Kwere reported as (15.8 ± 2.4 Bq kg^{-1}), (1.7 ± 4.3 Bq kg^{-1}) and (69.3 + 9.4 Bq kg^{-1}) respectively. For carpet grass ^{238}U, ^{232}Th and ^{40}Kwere reported as (16.0 ± 1.9 Bq kg^{-1}), (1.6 ± 4.2 Bq kg^{-1}) and (70.2 ± 11.6 Bq kg^{-1}) respectively. For the measured concentrations of primordial radionuclides, the effective dose equivalent (ADE) was calculated for three species of medicinal plants. It was reported that for each species of medicinal plant ADE were found to be lower than the recommended limit of 1 mSv in a year (Oni et al., 2011).

Sussa et al., (2011/2013) studied common medicinal herb Peperomia pellucida and its surrounding soils for radionuclide concentrations of ^{238}U, ^{232}Th, ^{230}Th, ^{226}Ra, ^{228}Ra and ^{210}Pb by alpha spectrometry and gross alpha and beta counting. Their reported radionuclide activity levels ranged from 4.3-38 Bq kg^{-1}, 1.7-124 Bq kg^{-1}, 2.1-38 Bq kg^{-1}, 8.5-37 Bq kg^{-1}, 3.2-46 Bq kg^{-1}, 39-93 Bq kg^{-1}, respectively (Sussa et al., 2011/2013).

Oufni et al., (2013) observed thoron and radon activity in several medicinal plant used in Moroccan cooking and traditional medicine. Radon (^{222}Rn) and Thoron (^{220}Rn) concentrations were measured using passive Solid-State Nuclear Track techniques (SSNTD) CR-39 and LR-115 type-II detectors were used for estimation of radon and Thoron. ^{222}Rn and ^{220}Rn levels were measured in soil, from where medicinal plants were collected. ^{222}Rn and ^{220}Rn levels are reported to be varying from 0.87 ± 0.06 Bqkg^{-1} to 6.20 ± 0.47 Bqkg^{-1} and from 30 ± 2.30 mBqkg^{-1} to 195±16 mBqkg^{-1} respectively. Higher values were reported for roots of studied plants as compared to stem and leaves (Oufni et al., 2013).

Tettey-Larbi et al., (2013) reported radioactivity level for several medicinal plants in Ghana using gamma ray spectroscopy by HPGe spectrometer. Their reported results depicted that mean activity concentration of ^{238}U, ^{232}Th and ^{40}K in the medicinal plants were found as 31.8±2.8 Bq kg^{-1}, 56.2±2.3 Bq kg^{-1} and 839.8±11.9 Bq kg^{-1} respectively. Highest activity concentration of ^{238}U and ^{232}Th were reported for Khayaivorensis plant and for ^{40}K highest value was observed for Lippiamultiflora plant**.** The total annual committed effective doses calculated for medicinal plants ranged from 0.026±0.001 to 0.042±0.002 mSv a^{-1} with an average value of 0.035±0.001 mSv a^{-1}. The average annual committed effective dose, 0.3 mSv a^{-1} for ingestion of natural Radionuclides, estimated for medicinal plants for current study was below the world average annual committed effective dose as reported in UNSCEAR 2000 report (Tettey-Larbi et al., 2013; UNSCEAR, 2000).

Oprea et al., (2014) investigated medicinal plants viz.; Tiliacordata, Matricariachamomilla, Calendula officinalis, Ocimumbasilicaum, Achilleamillefolium and Hypericumperforatum in Romania for radionuclidic contents. They have adopted global alpha as well as beta counting techniques (Oprea et al., 2014).

For radionuclides ^{210}Po and ^{238}U the maximum levels were recorded in Ocimumbasilicum (8 mBq kg^{-1}) and Achilleamillefolium (40 m Bqkg^{-1}). Highest values of ^{210}Pb were found in Matricariachamomilla, Achilleamillefolium and Hypericumperforatum (30 mBq kg^{-1}) and highest value of radionuclide ^{232}Th was found in Achilleamillefolium and Hypericumperforatum (60 mBq kg^{-1}). The radionuclides ^{210}Pb, ^{210}Po, ^{232}Th and ^{238}U have shown strongest tendency for accumulation in the Achilleamillefolium.

Kavocas et al., (2015) studied ^{226}Ra, ^{210}Po, ^{137}Cs, ^{210}Pb and ^{40}K contents within medicinal plant by employing alpha and gamma spectrometry. Activity concentrations of all Radionuclides were estimated via gamma spectrometry except ^{210}Po which was determined through alpha spectrometry. For the radionuclide ^{210}Po highest activity levels (10-19 Bq kg^{-1}) were recorded for herbs consisting of only leaves, whilst lowest ($\leq$ 2 Bq kg^{-1}) were reported for medicinal herbs consisting of only flowers. Same pattern was observed for ^{210}Pb. No definite relation was observed for primordial Radionuclides in different kind of herbs. For anthropogenic radionuclide, ^{137}Cs, highest values (0.4-20) Bq kg^{-1} were reported for wild grown samples as compared to cultivated medicinal herbs (0.4-1.6) Bq kg^{-1} (Kavocas et al., 2015).

Pourimani et al., (2015) have carried out study for the estimation of natural and anthropogenic Radionuclides in 8 medicinal and edible plant species including: Saliva nemorsa L., Triticumaestivum L., Peganumharmala L., Vitisvinifera cv. Shirazi, Medicagosativa L., Gondeliatournefortii L., Descorainiasophia (L.) Webb et Berth and Achilleavermicularis Trin. Activity concentrations of natural ^{226}Ra, ^{232}Th, ^{40}K and anthropogenic ^{137}Cs Radionuclides were determined using gamma ray spectrometry by HPGe detector. Activity concentrations reported for ^{226}Ra, ^{40}K, ^{232}Th and ^{137}Cs ranged from 2.27±0.45 to 7.43±0.60, MDA to (2.75±0.01) $\times 10^3$, MDA to 7.79±1.40 and MDA to 1.02±0.35 Bq kg^{-1} respectively. Internal and external hazard indices calculated for all herb samples were reported to be less then unity, which shows no significant health threats are posed by Radionuclides presence in medicinal plants (Pourimani et al., 2015).

Najam et al., (2015) have assessed nine medicinal plant samples used in Iraq for the determination of radionuclide activity concentrations. They have used Gross alpha, beta and gamma spectrometry (Proportional counter + NaI(Tl) detector) and HPGe detector). For ^{40}K, their reported activity concentrations varied from 124.1 Bq kg^{-1} in Crust sample to 88.3 Bq kg^{-1} in Chamomile sample, gross alpha varied from not detectable limit in Flax sample to 0.4 cpm in Anise sample, while beta activity varied from 5.7 cpm in Flax sample to 25.6 cpm in Latency sample and gamma activity varied from 0.6 cpm in Thyme sample to 5.10 cpm in Coriander and Flax samples (Najam et al., 2015).

Njinga et al., (2015) conducted preliminary research on medicinal plants of Nigeria for investigating Radionuclides concentration using NaI(TI) detector. He reported ^{40}K activity concentration in medicinal plants varying from (74.59 ± 2.19 to 324.18 ± 8.69) Bq kg^{-1} with average value of (324.18 ± 8.69) Bq kg^{-1}. Highest ^{40}K activity concentration was reported for A. indica whilst lowest for A. occidentale.

Activity concentrations of ^{226}Ra varied from (10.79 ± 4.24−42.47 ± 2.76) Bq kg^{-1} with average value of (25.02 ± 3.18) Bq kg^{-1}. Lowest and highest activity was recorded for P. guajava and V. paradoxa herbal samples respectively. The activity concentration of ^{232}Th varied from (27.76 ± 1.02 − 41.05 ± 1.05) Bq kg^{-1} with average value of (35.09 ± 0.71) Bq kg^{-1}. Lowest and highest ^{232}Th activity was reported for V. paradoxa and T. catappa herbal plants respectively. Due to ingestions of naturally occurring radionuclides in herbal plants the average annual committed effective doses (AACED) received by public range from (4.26±0.50 to 6.86±0.44) ×10^{-3} mSv y^{-1} with an average of (5.38±0.35) ×10^{-3} mSv y^{-1} (Njinga et al., 2015).

Highest values of AACED were found for A.occidentale whilst lowest for P.guajava herbal plants. AACED reported for this study are far below the worldwide average of 0.3 mSv y^{-1} (UNSCEAR, 2000 report) showing insignificant contribution to radiological health risk by Radionuclides found in herbal plants (Durugbo et al., 2012).

Shatha et al., (2015) determined natural radionuclide concentrations in 46 medicinal plant samples collected from Jordanian shop. He has used gamma spectroscopy using HPGe detector. Highest values estimated for ^{228}Ra, ^{40}K and ^{226}Ra were found as 15.33 ± 0.1, 2034 ± 57 and 15.6 ± 0.46 Bq kg^{-1} respectively. Whereas, lowest values of ^{228}Ra, ^{40}K and ^{226}Ra were respectively found as 1.47 ± 0.5, 24 ±1.6 and 0.26 ± 0.05 Bq kg^{-1} in herbal plants (Shatha et al., 2015).

Harb, (2015) reported natural Radionuclides concentrations in some medicinal plants available in Egypt. ^{226}Ra, ^{228}Ra and ^{40}K activity concentrations were determined using gamma spectrometry by HPGe spectrometer. The activity concentrations for ^{40}K, ^{228}Ra, ^{226}Ra varied from 140 ± 6−1538 ±54, <0.3-42.3±5.9, 0.4 ± 0.2 − 21.0 ± 1.2 Bq kg^{1}. Annual effective dose due to natural radionuclide presence in herbal plants varied from 0.003 to 0.073 mSv y^{-1} with mean value of 0.02 mSv y^{-1} (Harb, 2015).

Chandrashekara et al., (2015) reported ^{226}Ra, ^{210}Pb, ^{232}Th, ^{40}K and ^{137}Cs activity concentration for the medicinal plants Justicaadhatoda L., Careyaarborea Roxb., Mimosa pudica L., Azadirachtaindica A Jus. and Plectranthusamboinicus (Lour) Spreng. They have employed gamma spectroscopic method using HPGe detector for the determination of activity concentrations of different radionuclides. Their results showed that activity concentration due to anthropogenic radionuclide ^{137}Cs for all medicinal plant samples was below detection limit (BDL). Contributions from other radionuclides viz. ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K fall in the range from (BDL to 9.59, BDL to 6.40, 9.07 to 320.34 and 443.50 to 3401.29) Bq Kg^{-1} respectively. Authors have also reported activity concentration of same Radionuclides for soil samples and thereby calculated soil to plant transfer factor. Their transfer factor reported values for Radionuclides ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K vary in the range from (BDL to 0.17, BDL to 0.068, 0.12 to 3.73, and 2.94 to 28.66) Bq kg^{-1} respectively (Chandrashekara et al., 2015).

Chandrashekara and Somashekarappa, (2016) estimated Radionuclides contents and average effective doses by ingestion of various medicinal plants collected from Malnad area of Karnataka in south India. They have used High Purity Germanium Detector for gamma spectrometry. The observed variation in ranges of activity concentration were (BDL−87.03, 93.79−6831.40, 2.66−11.27 and2.42−8.72) Bq kg^{-1} for ^{210}Pb, ^{40}K, ^{226}Ra and ^{232}Th respectively. The average effective doses via ingestion of such Radionuclides were assessed as (0.0075−0.1067) mSv y^{-1}. These doses were much less than the world accepted dose standards (Chandrashekara & Somashekarappa, 2016).

Kareem et al., (2016) estimated primordial radionuclide concentrations in selected medicinal plants sample of Iraq. They have used NaI (TI) spectrometer for the purpose of gamma spectroscopy of samples. The activity concentrations of ^{40}K, ^{238}U and ^{232}Th fall in range from (219.134±2.24, 4.953± 0.37, 2.916± 0.12) Bq kg^{-1} respectively (Kareem et al., 2016).

Abojassim et al., (2016) reported radon concentrations in forty medicinal herbs collected from different stores of Iraq. They have used Solid State Nuclear Track detectors (SSNTD) technique for the determination of radon. Their reported values for radon concentrations ranged from 10.66 to 53.30 Bqm^{-3} within medicinal plants respective risk due to radon (Abojassim et al., 2016).

Kranrod et al., (2016) surveyed Thai herbal plants for the presence of natural radioactivity contents. They have used gamma spectroscopy using HPGe spectrometer. The activity concentration due to ^{226}Ra, ^{40}K and ^{228}Ra ranged from 0.20 to 6.67, 159.42 to 1216.25 and 0.10 to 9.69 Bq kg^{-1} respectively. Concentrations of ^{228}Ra and ^{226}Ra were recorded highest in Gotu kola. Whereas, highest ^{40}K was recorded in ginger. The annual effective doses via consumption of several herbal plants ranged from 0.0028 to 0.0097 mSv y^{-1} with average value of 0.0060±0.0001 mSv y^{-1}. Consequently, Thai medicinal plants found to be safe in health perspective (Kranrod et al., 2016).

Falandysz et al., (2017) estimated anthropogenic and ^{40}K activity concentrations in Boletus species of edible mushrooms from state of China by employing HPGe technique. The ^{137}Cs activity concentration ranged from 4.4 to 83±3 Bq kg^{-1} dry biomass in caps and from <3.8 to 37±3 Bq kg^{-1} dry biomass in stipes. Whilst, activity concentration for ^{40}K ranged from (420± 41 to 1300± 110 and 520± 61 to1300± 140) Bq kg^{-1} dry biomass. The estimated internal dose rate per 1 kg intake of mushrooms per annum ranged from <0.003 to 0.047±0.003 µSv and 0.22± 0.04 to1.2± 0.1 µSv for ^{137}Cs and ^{40}K respectively (Falandysz et al., 2017).

This study, for the first time, presents the results of activity concentration determinations for ^{137}Cs and ^{40}K in a high number (21 species, 87 composite samples, and 807 fruiting bodies) of mushrooms of the genus Boletus from across Yunnan in 2011-2014 and Sichuan (Boletus tomentipes) using high-resolution high-purity germanium detector. Activity concentrations of ^{137}Cs demonstrated some variability and range from <4.4 to 83 ± 3 Bq kg^{-1} dry biomass in caps and from <3.8 to 37 ± 3 Bq kg^{-1} dry biomass in stipes, and of ^{40}K, respectively, from 420 ± 41 to 1300 ± 110 and from 520 ± 61 to 1300 ± 140 Bq kg^{-1} dry biomass. No significant variations were observed regarding ^{137}Cs and ^{40}K activity concentrations among the same Boletus species from different sampling sites. No activity concentrations from ^{134}Cs were detected in any mushrooms.

Internal dose rates estimated were from intake of 1 kg of mushrooms per annum for ^{137}Cs range for species and regions from around <0.0031 to 0.047 ± 0.003 μSv, while those for ^{40}K were from around 0.22 ± 0.04 to 1.2 ± 0.1 μSv. The overall intake of ^{137}Cs was low, since low contamination was found in Boletus species. Worldwide studies relevant to radioactivity measurements within medicinal plants at various time periods via employing several techniques are hereby tabulated in Table 2.1 and Table 2.2.

World wide data reported so far via several research groups just about health perilous linked to radioactivity existing within medicinal plants has compiled. Reported studies depicted that a limited data available on radioactivity assessment within medicinal plants around the globe. NaI(TI) and HPGe spectrometer have been employed for radioactivity assessment. Radioactivities have also been evaluated via Gross α, β, CR-39 and LR-115 type II devices.

The compiled natural and anthropogenic data for medicinal plants shows area based variations. These variations may be attributed due to features involving geology, ecology, topography, soil and plant type. For current study compiled data, maximum radium contents, 115.08 ± 0.49 Bqkg^{-1}, was reported for Brazilian medicinal herbs and lowest value, below detection limit, has been reported for Turkey herbs.

Table 1.1. Radioactivity measurements in medicinal plants through employing various techniques

Findings	Technique	Studied Material	Investigated Area	References
^{137}Cs content	NaI spectrometer	Medicinal plants	(Marshall Islands) SW Hawaii	Robison et al., 1997
Radioactive content for ^{134}Cs and ^{137}Cs	HPGe detector	Medicinal plants	BanskaBystrica, Slovakia	UNSCEAR, 2001
Radioactivity content of ^{210}Po and gamma dosage rate	Electrochemical deposition portable scintillator and	Ayurvedic medicinal plant	Moodabidri nearby Mangalore	Narayana & Prakash, 2006
^{232}Th,^{228}Ra, ^{137}Cs and ^{40}K content	HPGe spectrometer	Medicinal Plants	Turkey	Turhan et al., 2007
Activity of ^{226}Ra,^{210}Pb and ^{228}Ra	Alpha as well as beta counting	Medicinal Plants	Brazil	Sussa et al., 2009
^{226}Ra, ^{137}Cs, ^{232}Th and ^{40}K content	HPGe spectrometer	Herbal plants	Egypt	Ahmed et al., 2010
Radioactivity content of ^{210}Po, ^{238}U and ^{214}Pb-Bi,^{210}Pb, ^{137}Cs, ^{40}K	AlPha and HPGe spectrometer	Medicinal Plants	Urbino, Italy	Desideri et al., 2010
Radioactivity content of ^{238}U, ^{40}K, ^{232}Th, ^{137}Cs and annual whole-body dosage	High Purity Germanium detector	Medicinal Herbs	Serbia	Jevremovic et al., 2011
Radioactive contents for ^{238}U, ^{40}K and ^{232}Th	NaI(Tl) detector	Medicinal Plants	Nigeria	Oni et al., 2011
Activity content of ^{238}U, ^{232}Th, ^{230}Th, ^{228}Ra, ^{210}Pb and ^{226}Ra	Gross α, β counting and alpha spectrometry	Medicinal Herb	Brazil	Sussa et al., 2011/2013
Radon as well as Thoron level	CR-39 and LR-115 type-II	Medicinal plants	Morocco	Oufni et al., 2013
Annual committed effective dosages and ^{238}U, ^{40}K ,^{232}Th	HPGe detector	Medicinal Plants	Ghana	Tettey-Larbi et al., 2013
Content of ^{210}Po, ^{232}Th, ^{210}Pb, ^{238}U and ^{90}Sr, ^{137}Cs	Global alpha as well as beta counting	Medicinal plants	Romania	Oprea et al., 2015
Radioactivity content of ^{226}Ra, ^{210}Po, ^{137}Cs, ^{210}Pb and ^{40}K	HPGe spectrometer and alpha spectrometry	Medicinal herbs	Hungary	Kovacs et al., 2015
Radioactivity content of ^{226}Ra, ^{40}K, ^{232}Th,^{137}Cs and internal as well as external hazards indices	High Purity Germanium detector	Medicinal Plants	Shazand, Iran	Pourimani et al., 2015

Table 1.1. continued...

Table 1.1. continued…

Findings	Technique	Studied Material	Investigated Area	References
α, β, ϒ activity and activity content of ^{40}K	Gross alpha, Beta, Gamma spectroscopic proptional counter, NaI detector and HPGe detector	Herbal Plants	Iraq	Najam et al., 2015
Radioactivity content of ^{226}Ra, ^{40}K, ^{232}Th and annual effective dosages	NaI detector	Medicinal Plants	Nigeria	Njinga et al., 2015
Radioactivity content of ^{40}K, ^{228}Ra, ^{226}Ra and heavy metallic content	HPGe spectrometer	Medicinal plants	Jordon	Shatha et al., 2015
Annual committed effective dosages and radioactivity content of ^{226}Ra, ^{40}K ,^{228}Ra	HPGe detector	Medicinal Plants	Qena, Upper Egypt	Harb, 2015
^{226}Ra,^{40}K, ^{232}Th, ^{137}Cs,^{210}Pb contents	HPGe spectrometer	Medicinal Plants	India	Shatha et al., 2015
Annual effective dosages and radioactivity content of ^{40}K, ^{210}Pb,^{226}Ra, ^{232}Th	HPGe detector	Medicinal Plants	South India	Harb, 2015
Activity content of ^{40}K, ^{238}U, ^{232}Th, ^{226}Ra and internal hazardous index	NaI detector	Medical Plants	Iraq	Chandrashekara et al., 2015
^{222}Rn Content	CR-39	Medicinal Plants	(Al-Najaf) Iraq	Chandrashekara & Somashekarappa, 2016
Radioactivity content of ^{226}Ra, ^{40}K, ^{228}Ra and Annual effective dosages	HPGe detector	Medicinal herbs	Thailand	Kareem et al., 2016

Table 1.2. Radioactivity contents (Bq kg^{-1}) subsisting within medicinal plants via applying various techniques

Year of Study	Country	^{40}K	^{137}Cs	^{230}Th	^{232}Th	^{234}U	^{238}U	^{226}Ra	^{228}Ra	^{210}Po	^{210}Pb	^{222}Rn	References
1999	(Marshall Islands) SW Hawaii	-	$(0.001-1)\times10^3$, Polypodiumscolopendria $(0.200-3)\times10^3$	-	-	-	-	-	-	-	-	-	Robison et al., 1997
2007	Slovakia	-	0.400-3.200	-	-	-	-	-	-	-	-	-	UNSCEAR, 2001
2007	Moodabidri nearby Manglore	-	-	-	-	-	-	-	-	6.3-56.9	-	-	Narayana & Prakash, 2006
2007	Turkey	715.5±50.1-1779.0±163.7	2.4±0.3-109.0±7.3	-	<BDL	-	-	<BDL	-	-	-	-	Turhan et al., 2007
2009	Brazil	-	-	-	-	-	-	29±3-65±4	<2.2-18.4±0.2	-	32±3-76±8	-	Sussa et al., 2009
2010	Egypt	-	MDL-12.62±0.42	-	-	-	-	-	7.71±0.25 - 15.08±0.49	-	-	-	Ahmed et al., 2010
2010	Italy	66.2-3582.0	-	-	<0.3-10.7	-	<0.1-7.32	-	-	<0.12-30.3	<3-58.3	-	Desideri et al., 2010
2011	Serbia	126-12437	0.3-8.8	-	1.7-15.1	-	0.6-8.2	-	-	-	-	-	Jevremovic et al., 2011
2011	Nigeria	67.9±7.4,70.2±11.6, 15.8±2.46	-	-	1.1±2.7, 1.6±4.2, 1.7±4.3	-	15.3±1.7(lemon grass), 16±1.9(carpet grass), 69.3±9.4(spear grass)	-	-	-	-	-	Oni et al., 2011
2011/ 2013	Brazil	-	-	2.1-38	1.7-124	42-129	4.3-38	8.5-37	3.2-46	-	39-93	-	Sussa et al., 2011/2013
2013	Morocoo	-		-	-	-	-	-	-	-	-	0.87±0.06-6.20±0.47	Oufni et al., 2013
2013	Ghana	839.8±11.9	-	-	56.2±2.3	-	3.18±2.8	-	-	-	-	-	Tettey-Larbi et al., 2013
2014	Romania	-	-	-	<DL	-	40×10^{-3}	-	60×10^{-3}	8×10^{-3}	30×10^{-3}	-	Oprea et al., 2015
2015	Hungary	-	-	-	-	-	-	-	-	0.4-20	-	-	Kovacs et al., 2015

Table 1.2. Continued…

Table 1.2. Continued…

Year of study	Country	^{40}K	^{137}Cs	^{230}Th	^{232}Th	^{234}U	^{238}U	^{226}Ra	^{228}Ra	^{210}Po	^{210}Pb	^{222}Rn	References
2015	(Shazand) Iran	MDA-(2.75±0.01) $\times10^{-3}$	10-15(leafy Parts), ≤2(Flowering Parts)	-	MDA-7.79±1.40	-	-	-	MDA-7.43±0.60	-	-	-	Pourimani et al., 2015
2015	Iraq	88.3(Chammile),124.1(Crust)	MDA-1.02±0.35	-	-	-	-	-	-	-	-	-	Najam et al., 2015
2015	Nigeria	74.59±21.9-324.18±8.69	-	-	27.76±1.02-41.05±1.05	-	-	-	10.79±4.24-42.47±2.76	-	-	-	Njinga et al., 2015
2015	Jordon	24±1.6-2034±57	-	-	1.47±0.5-15.33±0.1	-	-	0.26±0.46-15.6±0.46	-	-	-	-	Shatha et al., 2015
2015	Egypt	140±6-1538±54	-	-	-	-	-	0.4±0.2-21.0±1.2	<0.3-42.3±5.9	-	-	-	Harb, 2015
2015	India	443.50-3401.29	BDL	-	BDL-6.40	-	-	BDL-9.59	-	-	9.07-320.34	-	Shatha et al., 2015
2016	South India	93.79-6831.40	-	-	2.42-8.72	-	-	-	2.66-11.27	-	BDL-87.03	-	Harb, 2015
2016	Iraq	-	-	-	-	-	-	-	-	-	-	(10.6602.07−53.3034.64) $\times10^{3}$	Chandrashekara et al., 2015
2016	(Al-Najaf) Iraq	219.134 ±2.24	-	-	2.916 ±0.12	-	4.953±0.37	-	-	-	-	-	Chandrashekara & Somashekarappa, 2016
2017	Thailand	159.42-1216.25	-	-	-	-	-	0.20-6.67	0.10-9.69	-	-	-	Kareem et al., 2016
2017	SW China	420± 41-1300± 140	<3.8- 83±3	-	-	-	-	-	-	-	-	-	Abojassim et al., 2016

DL: Detection limit, BDL: Below Detection Limit, MDA: Minimum Detectable Activity

For anthropogenic radionuclides, higher activity concentrations of ^{137}Cs (2.4 ± 0.3 - 109.0 ± 7.3) Bq kg^{-1} are found in literature for Turkey herbal species and smallest were found (0.400-3.200) Bq kg^{-1} within Slovakian and Indian medicinal plant. ^{40}K maximum contents (93.796831.40) Bq kg^{-1} were reported within medicinal plants of South India and lowest (24± 1.6) Bq kg^{-1} was reported for Jordon medicinal plants.

Highest values for ^{210}Pb was found within Indian medicinal herbs (9.07- 320.34) Bqkg^{-1} and lowest, (BDL87.03) Bq kg^{-1}, were reported for medicinal plant of South India. Highest values of Polonium contents (<0.12-30.3) Bq kg^{-1} were found for the medicinal plants of Italy while lowest, ≤ 2 Bq kg^{-1}, for Hungary medicinal flowers. Nigerian medicinal plants found to have highest uranium contents (69.3 ± 9.4) Bq kg^{-1} whilst, smallest (<0.1-7.32) Bq kg^{-1} are found within medicinal plants of Italy. Brazilian medicinal herbs are reported for highest thorium contents (1.7-124) Bq kg^{-1} and smallest, <BDL, are reported within medicinal plant of Turkey and Romania.

To conclude, this chapter has reviewed and compiled natural and anthropogenic data reported in literature, especially for last 2 decades, for medicinal plants. Researchers across the globe have employed different spectroscopic techniques for the measurements of radionuclide concentrations. Considerable variations in reported data can be observed. Higher activity concentrations were reported for the South Indian medicinal plants whilst, lowest for the Romania medicinal plants (Achilleamillefolium, Matricariachamomilla and Hypericumperforatum). Maximum annual doses were reported for Egyptian herbs (Tilia). Much of the data available in literature is relevant with measurement of gamma emitting Radionuclides, and hence assessment of external dose exposure, in herbal plants. On the other hand, ^{226}Ra, ^{222}Rn, ^{220}Rn and ^{210}Po radionuclides are alpha emitting. Their elevated concentrations might results in excess of internal dose exposure in humans.Very limited numbers of studies addressing ^{222}Rn,

^{220}Rn measurements for herbal plants are reported in literature. These studies have provided a baseline data for future assessment, in case of any undesirable radiological emergency, and may lead in formation of standards of environmental safety regulations related to radiological healthcare due to use of medicinal plants.

1.6. SCOPE OF THE STUDY

With a heavy nuclear infrastructure for power production programs, use of radioactive isotopes in nuclear medicines, agriculture**,** in industrial radiography, mineral analysis, gauging applications etc., have increased the chances of radiation exposure of general public. Through many studies the carcinogenic nature of artificial and natural radionuclides has already been established. Keeping in view the health threat due to sustained radiation exposure the developed countries/organizations has had set regulations to establish standards for the the purpose of protection against ionizing radiations that results from activities carried out under licences issued by the Nuclear Regulatory Commission. In title 10, part 20, of the the report “Standards for Protection Against Radiation” from Code of Federal Regulations (10 CFR Part 20), the dose limits arising from the radiation exposure have been establish for occupational and non-occupational cases. For whole body exposure, they have suggested 5,000 mrem year^{-1} or 50 mSv year^{-1} occupational dose limit and similarly 100 mrem year^{-1} or 1 mSv year^{-1} dose limits in non-occupational cases.

Pakistan Nuclear Regulatory Authority, in a report “The Gazzet of Pakistan” part II published on, Tuesday, October 5, 2004 have proposed whole body effective occupational dose of 20 mSv for workers above age of 18 years and non-occupational doe limit for general public as 1 mSv year^{-1}.

Under normal controlled conditions background radiation exposure is supposed to be less threatening but situation has drastically changed especially, following the Tōhoku earthquake that happened on 11 March 2011. Earthquake lead tsunami off the coast of Japan, initiated the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident that resulted in meltdowns of three cores of reactors due to failure of the emergency cooling system for lack of electricity supply. FDNPP was a most serious nuclear accident since the Chernobyl nuclear reactor disaster. FDNPP accident resulted release of radioactivity to air and also water leakage from Fukushima Daiichi. Many kind of fission products and radionuclides emitted from the fuel contained volatile iodine-131, with half-life of 8 days and caesium-137, which has a 30-year half-life and is a strong gamma emitter. They are carried in a plume, and when it lands it may contaminate land for some time. The presence of this radionuclide was not only detected in Japan but also in far off countries.

Beside Nuclear accidents, many other activities, including use of radioactive isotopes in nuclear medicines, agriculture, in industrial radiography, mineral analysis, and gauging applications etc., increases radiation exposure.

This study aims at measurement of background radiations in soil and herbal plants especially in post FDNPP accident scenario. Results will be helpful for future studies in terms of availability of a baseline data for the purpose of comparison in any nuclear emergency condition. Another part of study is related with economical trade of herbal plants from Azad Kashmir to other part of country and abroad. Azad Kashmir is rich of wild plants and in recent past many ethnobotanical studies have been conducted to asses' economic, nutritional and medicinal values of these medicinal plants for human beings. Azad Government of the state of Jammu & Kashmir is trying to set legislation about commercial use of these plants in near future. Radiometric analysis of herbal plants can assure its suitability to be used as medicine against deceases. Current research intends not merely to assess radioactivity levels and provide background statistics but also to measures the external dosages caused by soil, medicinal herbs and air, while internal by inhaled air and by ingested medicinal herbs, which are practiced by a local community and throughout the country as well.

1.7. AIMS AND OBJECTIVES OF STUDY

This study aims at achieving underlying following objectives;

(1) To asses natural and anthropogenic radionuclides in soil samples.

(2) To measure gross alpha, gross beta activities in soil samples.

(3) To evaluate natural and anthropogenic radionuclides in medicinal plants of local origin.

(4) To measure gross alpha and gross beta activities in medicinal plants of local origin.

(5) To measure radiation doses and asses life time cancer risk due to exposure from natural and anthropogenic, gross alpha and gross beta activities in herbal plants and soil.

(6) To generate baseline data of background radiations within soil, air and medicinal herbs and that may serve a guidline in case of any nuclear emergency.

(7) Medicinal herbs radiological research will truly suggest possibly predictive, discriminative as well as informative analysis to resolve problematic issues and deliver significant data to pharmaceutical industries.

(8) To check suitability of medicinal herbs employed as traditional remedies for respective ailments.

Chapter 2

MATERIALS AND METHODS

2.1. STUDYAREA

The Azad Jammu and Kashmir state lies in western Himalayas and has a latitude 33.9259° N and longitude 73.7810° E. This region constitute of about 5134 miles of area, while mean annual growth rate is 2.69 with a population of 4.045 million (Rehman et al., 2017). The state of Azad Jammu and Kashmir is comprised of ten districts including Neelum, Hattian Bala, Bhimber, Haveli, Muzaffarabad, Bagh, Kotli, Mirpur, Sudhanuti and Poonch. For the current study sampling was carried out from five districts namely Poonch, Muzaffarabad, Hattian Bala, Bagh and Neelum valley. The precise description of these districts is as under.

2.1.1. Muzaffarabad District

Muzaffarabad district is one amongst the largest Azad Kashmir (AK) districts. The Muzaffarabad city is a capital of the state of Azad Jammu and Kashmir. It was named in 1952 as Muzaffarabad after Bombay Dynasty Chief; Sultan Muzaffar Khan. The capital city reflects the ancient history by Black as well as Red forts which are located at its central region.

Muzaffarabad district covers the area of about 1642 Km^2 and it is situated at the conjunction of the two rivers namely; Neelum and Jhelum between longitude 73.4769° E and latitude 34.3551° N, whereas its borders are linked with Khyber Pakhtoonkhwa and Punjab Provinces as well as to Bagh and Neelum districts of Azad Kashmir. Its population, according to 2017 census, is 0.650370 million along with growth rate of 2.80

%. It is situated at 76 km distance from Abbotabad and 145 km distance from Islamabad Pakistan.

The state administrative headquarter is situated in Muzaffarabad city and it is a facinatingtourists place. Muzaffarabad is a nucleus for the cultural as well as political activities. Jhelum and Neelum rivers confluxes at Domail, site of Muzaffarabad city which is surrounded via lofty mountains and have a vital role for the micro-climate of the district.

2.1.2. Poonch District

District Poonch has 33°83′ N and 73°88′ E coordinates along with mean elevation of 1,006 m above the sea level and has 0.476835 million inhabitants, acoording to 2011 estimation, which covers the area of about 1,674 km^2. Poonch district is widely extended with the mountain’s assembly like Pir Panjal and Toli Pir being the highest climax of the district. Poonch district shared the western boundaries that running across the River Jhelum from North towards South. The occupied Jammu and Kashmir adjoined to Pir Panjal derives the River Poonch and thus formed a south eastern periphery of the district. District’s climate constantly fluctuates with an altitude. As, typically cold climate exist in North eastern areas and hot summer as well as cold winter subsist in lower valleys of the district. Temperature typically fluctuates between 16 °C and 26 °C for the month of June, while In January, temperature fluctuations drops to 1 °C and 7 °C as well. While average yearly rainfall roughly 18000 millimeters were observed for the Poonch district.

2.1.3. Bagh District

The Bagh district has 33°97′ N and 73°79′ E coordinates along with typically average elevation of 5499 ft. The Bagh district has 0.395 million inhabitants which folds a region of a 770 km^2.

2.1.4. Hattian Bala District

Hattian Bala district is relatively situated across the bank of Jhelum river at 34° 16’ N and 73° 74’ E. It encloses the area of 854 km^2 along 0.265 million inhabitants according to the 2015 census. The elevation level of central valley usually finds within 500 m and larger than 2000 m, whereas sub Himalayan are located inside the district area. The steep climax covered with glaciers might have elevation larger than 3000 m. Whereas, Murree Formation predominantly incorporate the area with lithological deposits like broad colluvium deposit, fewer fluvial terraces along parallel elevated surfaces and proglacial plains in terms of lofty valley loads (Beg, 2015).

2.1.5. Neelum Valley District

District Neelum has 34°58′ N and 73°91′ E coordinates with a mean elevation of 5299 ft and considered to be a largest district with respect to area of AK state. Neelum was estimated to have 0.201 million populations and area of about 3621 km^2 that is merely a 27.2 % of the entire AJK area, while it is extended approximately 144 km parallel to river Neelum.

The tectonic units like Lesser Himalayan Crystalline Unit (LHC) as well as Higher Himalayan Crystalline Unit (HHC) separated out through a Main Central Trust (MCT), the outline of Neelum valley. These tectonic units demonstrated an equivalent stratigraphy and are usually located with the lesser rating metamorphic throng (Malik et al., 1996).

2.2. SAMPLING METHODOLGY

For the estimation of activity concentrations of natural and manmade radionuclides associated with environmental samples including soil, medicinal herbs invistigated samples have been collected from five districts of the state of Azad Jammu and Kashmir.

2.2.1. Sampling Approach for Spectroscopy

For measurement of gross alpha/beta activities and gamma spectrometric analysis soil and medicinal herbs samples were collected from different parts of five districts of the state of Azad Jammu and Kashmir. A detailed sampling approach of each sample for gross alpha/beta and gamma measurements are illustrated in the section 2.2.1.1, 2.2.1.2, and 2.2.1.3.

2.2.1.1. Soil sampling and pre- treatment

In current study, IAEA standards described in TECDOC-14515 was strictly pursued for the collection of soil samples at various sites of several Azad Kashmir districts (IAEA, 2004). The underlying step by step approach involved in respective soil and herbal sampling has been carried out.

a) Pre-sampling approach

Several processes were conducted prior to leaving for field study to collect the sample. Respective site selection, road map of study plan, tour team selection as well as travelling kit packed with essential necessities were arranged. The underlying tools were kept within the kit.

(1) Coring tool of precise depth for soil sample collection.

(2) Transparent polythene bags with 5 kg volume to maintain and carry the required sample.

(3) Black permanent marker employed for labelling of each individual bag.

(4) Global positioning system (GPS) device for the measurement of the sampling spot parameters.

(5) Note-book as well as pencil for documenting parameters of every visited spots.

(6) Pair of gloves employed for skin protection.

(7) Dust mask to prevent from dust inhalation.

(8) Steel ruler for depth measurement.

(9) Transparent tape to secure the written sampling sites information from rain or moisture of the environment.

Several sites along with tool kits were visited and corresponding soil samples were collected.

b) Sample collection

For every individual sample, a sampling area of (1×1 m^2) was selected. The extraneous material like stones, wood pieces, leaves and glass pieces were eradicated to get the natural soil for the respective sites. Coring tool was employed to obtain the 2 kg soil at the depth of 5 cm. The obtained soil was sealed in the polythene bags. The site name, date and sample code were indexed properly upon the bag via permanent marker. The accessories were washed and dried to prevent any possible contaminants prior to leaving for next sampling site. All soil samples were collected under the same sampling procedure.

c) Site parameters

The global positioning system known as GPS device was employed for the determination of the geological parameters like altitude, longitude as well as latitude at concerned sampling site. A twenty-nine soil samples were collected, sealed, indexed and then brought to Solid State Nuclear Track Detector laboratory, of Physics Department, University of AJ&K Muzaffarabad for the further process.

Table 2.1. The geographical features of collected soil sample at investigated zone of five districts

Sr. No.	Site Name	District	Latitude	Longitude	Altitude (ft)
1	Nagdar	Neelum valley	34°40.288′′	073°56.214′′	5997
2	Sudhan Gali	Bagh	34°04.647′′	073°44.587′′	7310
3	Peer Chinasi	Muzaffarabad	34°23.386′′	073°33.028′′	9165
4	Rashian	Hattian Bala	34°15.5164′′	073°49.187′′	7287
5	Tolipeer	Poonch	33°54.003′′	073°51.037′′	7181

2.2.1.2. Medicinal herb sampling and pre-treatment

Prior to visit the sampling sites, tool kit with same equipment's were packed and a total of five district of Azad Kashmir with several locations were selected for medicinal herb sampling. All relevant informative data were indexed properly upon the filled with samples as well as sealed polythene bags. These packed sample bags were placed in the Nuclear Physics Labartory to conduct pretreatment. Snapshots of respective investigated medicinal herbs are shown in figure 2.10.

Table 2.2. The geographical features of collected medicinal herbs from Muzaffarabad

Sr. No.	Site Name	District	Latitude	Longitude	Altitude (ft)
1	Pir chinasi	Bistorta amplexicule(Roots, Leaves)	34°23.386′′	073°33.028′′	9165
2	Pir chinasi	Bergenia ciliate (Roots, Leaves)	34°22.676′′	073°31.484′′	6947
3	Centre plate	Mentha Longifollia (Leaves)	34° 22.789′′	073° 28.047′′	2259
4	Lower Plate	Nastrition officinal (Leaves)	34° 22.015′′	073° 28.038′′	2221
5	Pir chinasi	Polygonum aviculare (whole herb without root)	34°23.362′′	073°33.043′′	9193

Table 2.3. The geographical features of collected medicinal herbs from Poonch district

Sr. No.	Site Name	District	Latitude	Longitude	Altitude (ft)
1	Toli Peer	Bistorta amplexicule (Roots, Leaves)	34°40.019′′	073°56.074′′	5856
2	Toli Peer	Bergenia ciliate (Roots, Leaves)	33°53.689′′	073°52.879′′	7536
3	Ali Sojal	Mentha Longifollia (Leaves)	33° 53.035′′	073°352.023′′	5574
4	Ali Sojal	Nastrition officinal (Leaves)	33° 53.035′′	073° 52.023′′	5574
5	Toli peer	Polygonum aviculare (whole herb without root)	33°54.003′′	073°51.037′′	7181

Table 2.4. The geographical features of collected medicinal herbs from Bagh district

Sr. No.	Site Name	District	Latitude	Longitude	Altitude (ft)
1	Sudhan Gali	Bistorta amplexicule (Roots, Leaves)	34°04.731′′	073°44.470′′	7353
2	Sudhan Gali	Bergenia ciliate (Roots, Leaves)	34°04.767′′	073°44.313′′	7322
3	Bagh	Mentha Longifollia (Leaves)	33° 58.775′′	073° 46.349′′	3413
4	Bagh	Nastrition officinal (Leaves)	33° 58.755′′	073° 46.349′′	3410
5	Sudhan Gali	Polygonum aviculare (whole herb without root)	34°04.825′′	073°44.969′′	7826

Table 2.5. The geographical features of collected medicinal herbs from Hattian Bala district

Sr. No.	Site Name	District	Latitude	Longitude	Altitude (ft)
1	Reshian	Bistorta amplexicule(Roots,Leaves)	34°15.561′′	073°49.281′′	7274
2	Reshian	Bergenia ciliate (Roots, Leaves)	34°15.718′′	073°48.927′′	7081
3	Reshian	Mentha Longifollia (Leaves)	34° 15.566′′	073° 49.300′′	7251
4	Reshian	Nastrition officinal (Leaves)	34° 15.580′′	073° 49.305′′	7257
5	Rashian	Polygonum aviculare (whole herb without root)	34°15.500′′	073°49.169′′	7278

Table 2.6. The geographical features of collected medicinal herbs from Neelum valley district

Sr. No.	Site Name	District	Latitude	Longitude	Altitude (ft)
1	Nagdar	Bistorta amplexicule (Roots, Leaves)	34°40.288′′	073°56.214′′	5997
2	Nagdar	Bergenia ciliate (Roots, Leaves)	34°40.019′′	073°56.074′′	5856
3	Nagdar	Mentha Longifollia (Leaves)	34° 39.956′′	073° 55.782′′	5734
4	Nagdar	Nastrition officinal (Leaves)	34° 39.980′′	073° 55.810′′	5602
5	Nagdar	Polygonum aviculare (whole herb without root)	34°40.101′′	073°56.416′′	5858

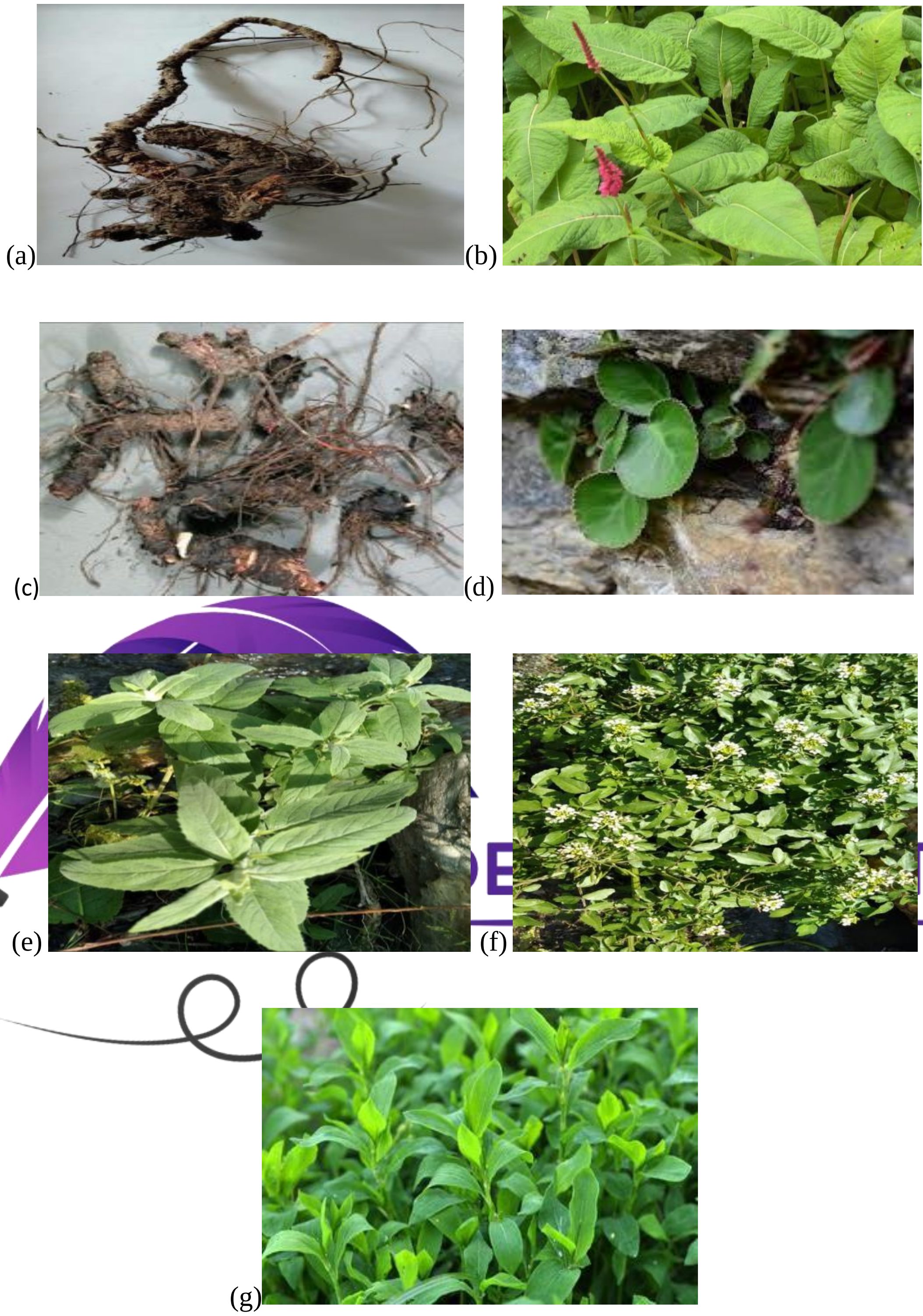

Figure 2.1. Snapshots of respective investigated medicinal herbs (a) Bistorta amplexicule roots, (b) Bistorta amplexicule leaves (c) Bergenia ciliate roots (d)Bergenia ciliate leaves (e) Mentha Longifollia (f) Nastrurtium Officinal (g) Polygonum Aviculare

Table 2.7. Name, examined edible parts and medicinal properties of observed medicinal herbs (Medicinal properties available at https://en.wikipedia.org/wiki/Mentha longifollia/Polygoum aviculare/Bistorta amplexicule/Bergenia ciliate/ Nastrition officinal)

Sr. No.	Scientific Name	Local Name	Edible parts	Medicinal uses
1	Bistorta amplexicule	Masloonr	Root, Leaves	Rheumatic Pains, Backache, Fever, Flu, Wound Treatment
2	Bergenia ciliate	Butt Peva, Butt Keva	Root, Leaves	Antibacterial, Antidiabetic, Anticancer, Antiinflammatory, Kidney disorder
3	Mentha Longifollia (Leaves)	Chitta Poodna,	Leaves	Antiasthmatic, Flatulence, Antiseptic, Antipasmodic, Carminative, Indigestion stimulant, Headache, Cold, Cough
4	Nastrition officinal officinal (Leaves)	Tara Meera, Chulura	Leaves	Antiscorbutic, Depurative, Stomachic, Diuretic, odontaglic, Expectorant, Hypoglycaemic, Purgative, Stimulant
5	Polygonum aviculare (whole herb without root)	Tarrobra	Leaves	Diarrhea, Hemerrhoids, Dysentry, Kidney Stones, Jaundice, Gonorrhea, Lung Cancer, Throat, Skin disorder, Sore mouth

2.2.1.3. Sample preparation

IAEA guide lines were followed for the preparation of the sample (IAEA, 1989). The brief description is as under.

Collected sample might not be in appropriate physical form to conduct the assessment at a laboratory. The estimation of activity concentrations for a sample requires its dryness, size contraction and homogenized form. Collected samples were dryed that process have decreased the volume and weight of the sample and ensured preservance of sample for longer period. Adequate period at constant temperature was used to dry the sample in order to get a persistent dehydrated weight. Measurements for both wet or fresh and dry weight of the sample were required. Contamination was prevented throughout the drying process.

Figure 2.2 shows an oven employed to dry the investigated samples at Nuclear Physics Laboratory of Department of Physics, University of AJ&K Muzaffarabad.

Figure 2.2. Drying oven

While, electronic digital balance employed for weighing the soil and medicinal herb samples is shown in Figure 2.3.

Figure 2.3. Electronic Digital Balance

The dried, grinded as well as homogenized powdered soil samples of weigh 250 g, and medicinal herb samples of weight 100 g were packed and then sealed for two months to get secular equilibrium state prior to subject for gamma spectroscopic analysis via High Purity Germanium (HPGe) detector.

While for the purpose of gross alpha/beta counting, about 1g and 0.5-0.7g of each soil and herbs powdered samples were taken for further pretreatment. These samples were placed in the planchet (stainless steel) and evenly distributed via ethyl alcohol which further evaporated by IR lamp (250 W). Next samples were oven dried for two hours at 105°C and dried weight were noticed and kept into the desiccator to avoid any moisture until these samples were subjected to gross alpha/beta counter measurements.

2.3. MONITORING METHODS

Gross alpha/beta spectrometric counter (ASC-950-DP) and gamma ray spectroscopic system (HPGe) were employed to obtain the afore-mentioned goals.

2.3.1. Gross Alpha/Beta Spectrometry

2.3.1.1. Theory

ASC-950-DP counter also termed as dual phosphor counter is a high speed portable counter supporting gasless operation. The detector is able to stop contribution from cosmic backgrounds using anti –coincidence Guard Detector. Three sensing components of latest DP design includes Zinc Sulphide layer (alpha sensitive), plastic scintillator (beta/gamma sensitive) and Photo-multiplier tube (light sensitive). As soon as the alpha particles enter into the outer window, these particles after striking on ZnS produces scintillations. These light pulses are projected from plastic scintillator on to the Photo Multiplier Tube (PMT). While, beta particles are passed through the layer of ZnS and are detected via plastic layer, next light pulses from this plastic scintillator are projected onto the PMT. Since plastic material is technically gamma sensitive, that’s why extremely thin layer is employed to diminish the gamma impacts on the DP counter. Practically, DP counter has about zero gamma efficiency. Both scintillator materials produce the characteristic pulses which might be differentiated on the basis of their pulse height. As, an alpha particles produces pulses of relative higher amplitude upon striking to the layer of the ZnS and low amplitude pulses are generated after striking of beta particles on the plastic layer.

2.3.1.2. Operational setup for the current study

The protean α/β counter (ASC-950-DP) was employed for the gross α/β counting of soil and medicinal herbs samples. Operating voltage of 705 V and 1260 V were correspondingly selected for alpha and beta modes. Calibration of the system was achieved by standard alpha and beta sources viz. Am-241 and Sr-90 on 15-16 October 2019 with activities 0.6938 and 0.4092 KBq. Sample's counts as well as geometry corrections were achieved by drawing self-absorption curve separately for alpha and beta. Soil and medicinal herbs samples were counted for time span of 3000s consecutively for beta and alpha modes. Instrumental calibration parameters including attenuation term, background, efficiency as well as sample's parameter including sample volume, mass residue and counting time span provided the minimum detection limit (Zorer et al., 2009). The employed counting setup is shown in Figure 2.4

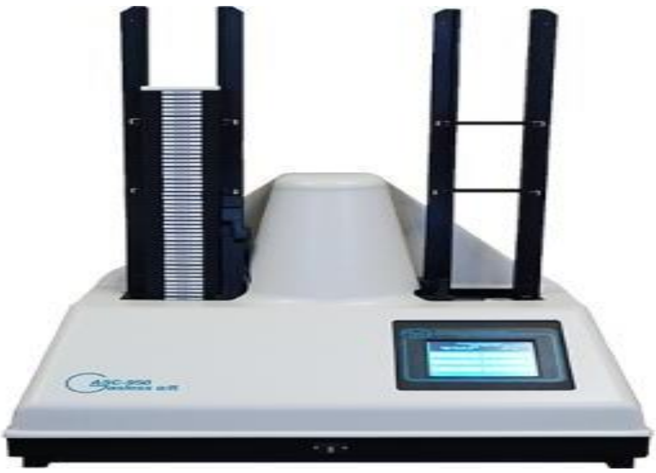

Figure 2.4. The α/β counter employed for the assessments of several environmental sample

2.3.1.3. Mechanism of gross α/β measurement

2.3.1.3.1. Background measurement

For background assessments, empty planchet was counted for the same preset conditions including time and operating voltage that were employed in the case of investigated sample's counting. Counts obtained for each sample was corrected by subtracting background readings to get corrected value.

$$C_N = C_n - C_b \tag{2.1}$$

Where, C_N is corrected measured count rate, C_n is measured count rate and C_b is the background count rate.

2.3.1.3.2. Calculations for gross alpha/beta activity

Gross alpha/beta activity levels within soil and medicinal herb samples have been calculated and details for the measurements are mentioned in the chapter 3 under the section 3.2.5.3.1.

2.3.1.3.3. Minimum detectable activity

Minimum detectable activity (MDA) of several environmental samples was evaluated and details are mentioned under section 3.2.5.3.1 of chapter three.

2.3.2. Gamma Spectroscopy

A gamma spectrometric setup provides a non-destructive, a rapid and multi elemental technique that was employed for the radioactivity assessment within environmental samples. Gamma spectrometric setup could accomplish both quantitative as well as qualitative assessment of the samples. Spectrum of investigated samples could present the identification of radionuclides corresponding to their intensity points and energy for the case of qualitative assessments, while radio-nuclidic activities are governed by a quantitative estimation. For the current study, gamma spectrometric system was employed for radionuclidic assessments by estimating the precise efficiency of the detector. The arguably important technique for the assessment of radioactivity in environmental samples is surely a gamma spectrometry (IAEA, 2003).

When gamma ray undergoes an interaction with the material and being uncharged, doesn't make a direct excitation or ionization of the targeted material. Whenever gamma ray imparts its energy either partially or wholly to an electron of the material or through interaction with the detector which could critically determines the gamma ray, two different functions are carried out by the detector to serve as a gamma spectrometer. Firstly, production of the secondary (one or more) electrons upon the most probable impacts of incident gamma ray. Furthermore, for such secondary electrons it essentially acts like a conventional detector (Mlwilo et al., 2007).

2.3.2.1. HPGe detector

2.3.2.1.1. General features

Semiconductor germanium setup was firstly introduced in 1962. Later on, an additional improvement was achieved, when semiconductor material of high purity germanium was fabricated. The 10^{10} atoms cm^{-3} as a substitute of 10^{13} atoms cm^{-3} was employed as high purity content of germanium. A high resistivity of material is proportionally linked with square of the width of its depletion region. To detect gamma radiation, currently a high purity germanium setup has developed as a setup of choice and highly preferred device. Low impurity content, enlarge conductivity, briskly response, enlarge depletion region, germanium of relatively high proton number, electron hole pair developed by less energy, superb peak symmetry, excellent operational setup available for gamma energy ranges, little noise operation and compact dimension makes HPGe setup as an exceptional detector (Khandaker, 2011).

2.3.2.1.2. Theory

In a nuclear decay, whenever an excited nucleus decays to lower levels then gamma rays with energy ranges 0.1-20 MeV are ejected. The radiation detection procedure is initiated soon after gamma ray strikes the targeted crystal. Energy is exclusively transmitted in ideal states. Whenever, gamma ray falls on to the crystal that leads to three type of processes including; 1) photo-electric affect, 2) Compton scattering and 3) Pair production.

An interaction of less energy photon to the inner shell electron of the orbit called as photoelectric effect. The energy of the collided photon is wholly imparted to orbital electron and thus it is ejected from the inner shell. This ejected electron is called as photoelectron. This mechanism occurs only, whenever energy of incident photon exceeded to the energy of orbital electron. Outer shell electrons come to cover the vacancy created by the inner shell emitted electron, then auger electron or characteristics x-rays are ejected (Sorenson & Phelps, 1987: Knoll, 2000).

The mechanism of Compton scattering proceeds only whenever a gamma ray photon imparts its energy to electron of the detector (Adams & Dams, 1970). The deflection with an angle (θ) along the original direction of incoming photon and projectile gamma ray then transmitted a partial energy and momentum to the free electron and therefore formed a recoil electron. The transmitted energy to electron can have energy from zero to large fraction of incoming gamma ray, with scattering, possibly at all angles (Knoll, 2000).

The process of pair production proceeds only in the powerful electric field of the protons of nuclei of the absorbing material that resulted into the formation of electron and positron at the fully annihilated junction of projectile gamma ray. Pair production proceeded by energetic gamma ray having energy higher to double the rest mass energy of the single electron. A back to back ejection of annihilated photons with energy m_oc^2 could replace the pair of electron and positron and thus both would disappear (Knoll, 2000).

2.3.2.1.3. Operational principal

A reverse biased approach is employed in semiconductor HPGe setup. The interaction between ionizing radiation and depletion region leads to the formation of electron hole pair that further is swept to the opposite electrodes via electric field. The integrated electrical pulses are transmitted to voltage signal via preamplifier as soon as electrodes gets the charge carrier and additionally this signal is further intensified from milli to few volts through amplifier. Thus, scalar would register this unit pulse as a single count. The amplitudes of very individual electric pulse entirely depend on the energy associated with incoming rays. The detection efficiency of device is directly linked to the receptive degree of instrument that further relies on diode dimensions as well as active width of intrinsic region (Roth et al., 1984). Thickness of depletion region varies inversely with the impurity content of that material. While, purity content of detector might be enhanced to enlarge the detection efficiency of the instrument.

2.3.2.1.4. Configuration

Even though, coaxial and planar blueprint of HPGe setup have been employed historically, however, coaxial designs are mostly employed for germanium detector. Detector called as planar detector if and only if germanium crystal cuts into disc like shape. The other one of hollow core like cylindrical crystal shape is named as coaxial detector. A negative electrode lies at outer region of P-type coaxial geometry and positive located at inner region of axial void core thus makes the junction as reversed biased and this configuration is known as close ended coaxial geometry (Knoll, 2000).

2.3.2.1.5. Biasing

Drift velocity of respective charge carriers as well as thickness of corresponding depletion region might be enhanced via enlarging reverse biased voltage of operated HPGe diode. As drift velocity of respective charge carriers is directly linked to biased voltage and at specific biased voltage, it gets saturated as 10^7 cm/sec through minimizing the collection time. The width of depleted region plays a crucial role for the detection of the specific energy of the incoming radiations. It is theoretically stated that square root of biasing voltage is linked directly to the depth of depletion region. Biasing voltage of several thousand volts could be applied to have the absolute depletion region within the two electrodes of respective diode thereby extending the sensitivity of depletion region all over to its active dimensions. Entire depletion region might get extended electric field within the respective electrodes and efficiency of setup could be enhanced via enlarging charge collection (Krane, 1988: Canberra, 2008).

2.3.2.1.6. Detection efficiency

Efficiency of detecting setup might be determined so as to know that at what extent the observed counts may found closer to actual source activity. Determination of actual source activity requires a sufficiently high efficiency of detection setup. While, contiguous environment as well as physical factor of counting setup have relatively high influences upon efficiency parameter. HPGe setup could define several efficiencies like intrinsic efficiency and filled energy photo-peak, relative as well as absolute efficiency. In case of gamma detection setup, full energy photo peak efficiency considered to be a crucial one that varies with soild angle on detector, distance of source from detector and energy of incoming photon but independent of detector's dimension and it can be evaluated by the following term;

$$E = \frac{C}{I \times A} \times 100\ \% \qquad (2.2)$$

where, net counts s^{-1} are described by ‘C’, gamma ray intensity is showed by ‘I’ and actual source activity is represented by ‘A’ (Khandaker, 2011).

2.3.2.1.7. Shielding

In laboratory, setups are always surrounded by background radiations that are obtained from spontaneous radioactive traces exhibiting on ground, building materials and in air as well as from cosmic origin. Therefore, setup must be shielded to avoid any background radiation impinging onto the detector sensitive surface, while dealing with low level radiation of observed samples as shielding minimizes the external radiation impacts. Lead is a common shielding material with roughly thickness of 1cm along with internal lining of copper or cadmium which lessens the background X-rays impacts raised via lead itself (Gordon, 1995).

2.3.2.1.8. Cooling

Normally, low temperature less than 120 K is a pre requisite for operational setup of HPGe. The thermal electron pair creation is avoided by means of such less temperature known as cooling of setup. As germanium crystal has less band gap that makes probably the thermal diffusion of electron from valence towards conduction band possible just at room temperature. This electrical noise rises thermally, which could be avoided via cooling procedure and this less temperature requirement could merely fulfilled through liquid Nitrogen. N_2 may offered 77K (-196 °C) as liquefaction temperature for normal operational setup. While, Room temperature leads to non-operational condition for HPGe setup (Krane, 1988).

2.3.2.1.9. Operational setup for current study

For gamma ray detection, HPGe setup is considered to be the best choice and this setup gives directly counts or voltage signal for every incoming gamma radiation. Soil, and medicinal herb samples were investigated in the current study via P-type coaxial HPGe setup (Canberra Industries inc., USA) with active volume of 180 cm^3 having Model No. GC2520-7500SL. Table 2.8 portrays clearly the operating parameters as well as physical features of HPGe setup used for the current study.

Table 2.8. Operating parameters as well as physical features of HPGe setup

Parameters	Specifications
Physical Dimensions	
Geometry	P-Type coaxial close- ended
Length	4.75cm
Diameter	5.56cm
Distance from Window	0.5cm
Crystal category	upright dipstick
Operating Parameters	
Leakage Current at B.V	-0.01nA
Baising Voltage	+ 3000Volts
Peak toward Compton Ratio	54: 1

The DC preamplifier of 100mV/MeV charge sensitivity manufactured by CANBERA was additionally employed for signal amplification.

The setup was shielded by lead layer of about 4.6 cm at top, its sides are shielded by three layers with inner one of copper (3 mm) thick, tin (4 mm) and outer one is again of lead having 15cm that was employed to diminish the background radiation impacts. While detecting active samples, shielding must be required for safety of laboratory workers.

For current study, a dipstick cryostat system was employed to house the detector vacuum compartments with dipstick just like cold finger that was introduced within the neck of dewar consisting of liquid nitrogen. Thermal noise could be reduced during its operational mechanisms as generated heat could be absorbed within the detector assemblage through dipstick and transmitted a liquid N_2 further to Dewar. Spectroscopic software Genie 2000 of version 2.1 (Canberra, USA) was installed as well as employed to display, and also to analyze a gamma ray spectrum via multichannel analyzer (MCA). Fundamental spectrum analysis plus reporting, MCA control and central display as well as manipulation be the parameters that could be performed via Genie 2000.

Figure 2.5. Depicts clearly the systematic diagram of detection setup coupled with accessories

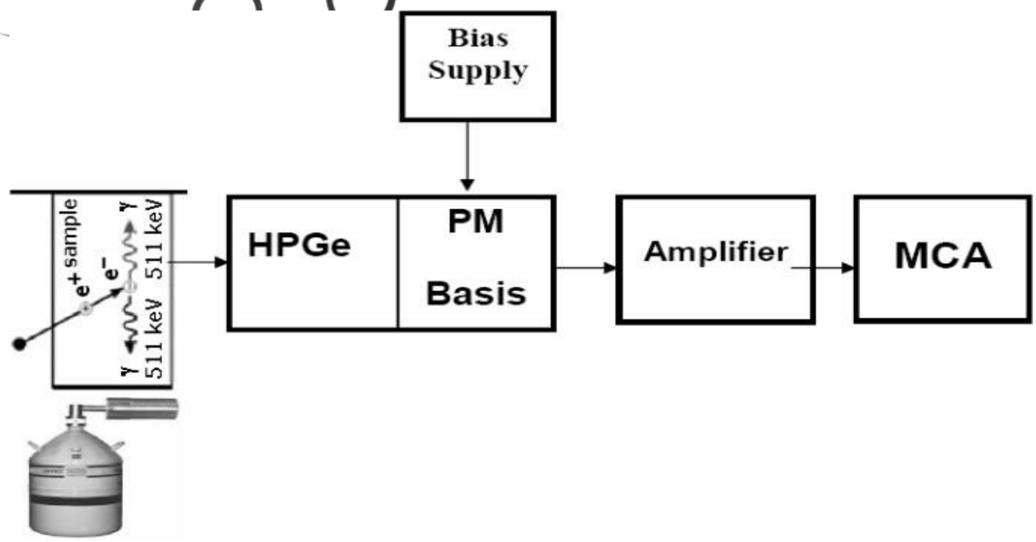

Figure 2.6. Block diagram for HPGe setup

2.3.2.1.9.1. System calibration

Any analyst acquired the one of the crucial jobs, which is the setting of analytical system. All the obtained results will be wrong if the incorrect calibration was performed. The crucial calibration was needed for efficiency, resolution and energy relationship (Sutton et al., 1993). A germanium detector was employed that provides energies of gamma rays corresponding to location of the photo-peaks.

2.3.2.1.9.2. Energy calibration

Qualitative analysis leads to energy calibration of the prepared samples of radioactive nuclides. Detector provides the spectrum that presents the location of photo-peak which is significantly crucial for gamma emitter. The multichannel analyzer (MCA) represents the launching of channel number whereas; pulse height analyzer corresponding to energy of gamma rays that leads to energy calibration (IAEA, 1989).

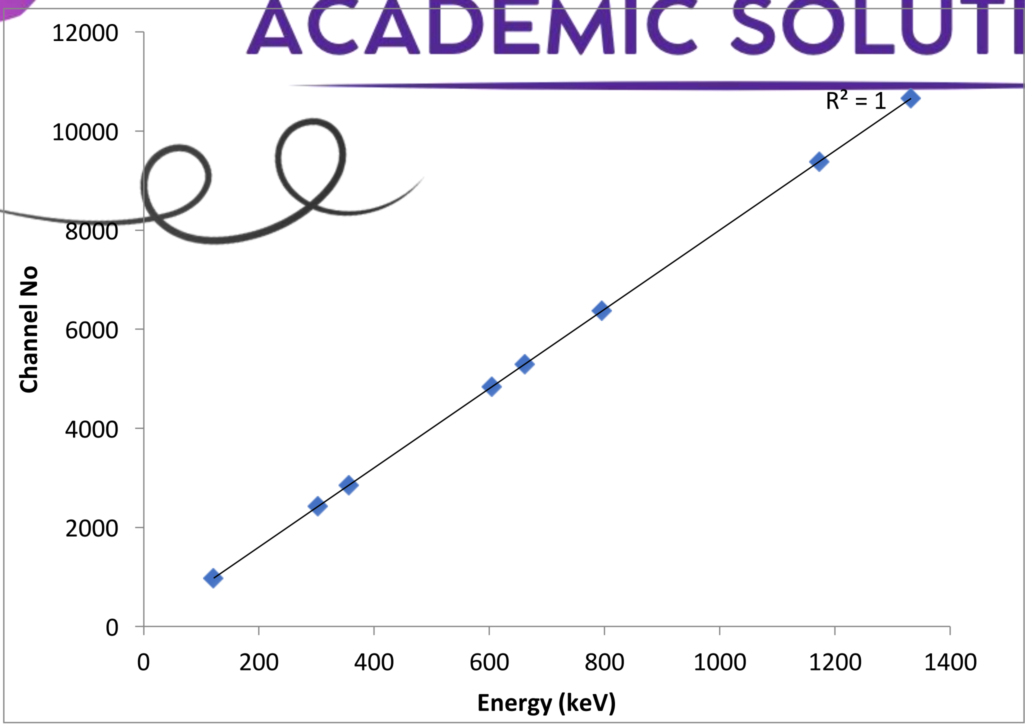

Figure 2.7. Energy calibration for HPGe setup

2.3.2.1.9.3. Efficiency calibration

Quantitative analysis leads to an efficiency calculation of the tested sample which comprises of radioactive nuclides. An estimation of efficiency graph is important for reliable, accurate and correct activity assessment. Relative efficiency generally describes performance of detection system that portrays detection efficiency of gamma ray of ^{60}Co with energy 1332 KeV of HPGe relative to NaI(TI) standard detector. For appropriate spectrometric calibration, a standard for matrices with comparable density, alike content for the related radionuclides and similar dimension and counting configuration just similar to real samples should be established (IAEA, 1989 and Vukanac et al., 2008). In current work, detector was calibrated for absolute efficiency by using full energy peaks of standard mixed source ^{152}Eu. The calculated efficiency of HPGe system is 30 % relative to NaI(TI) setector, whereas energy resolution of HPGe detector for gamma ray with energy 1332 KeV from gamma source ^{60}Co was found to be 2.0 KeV.

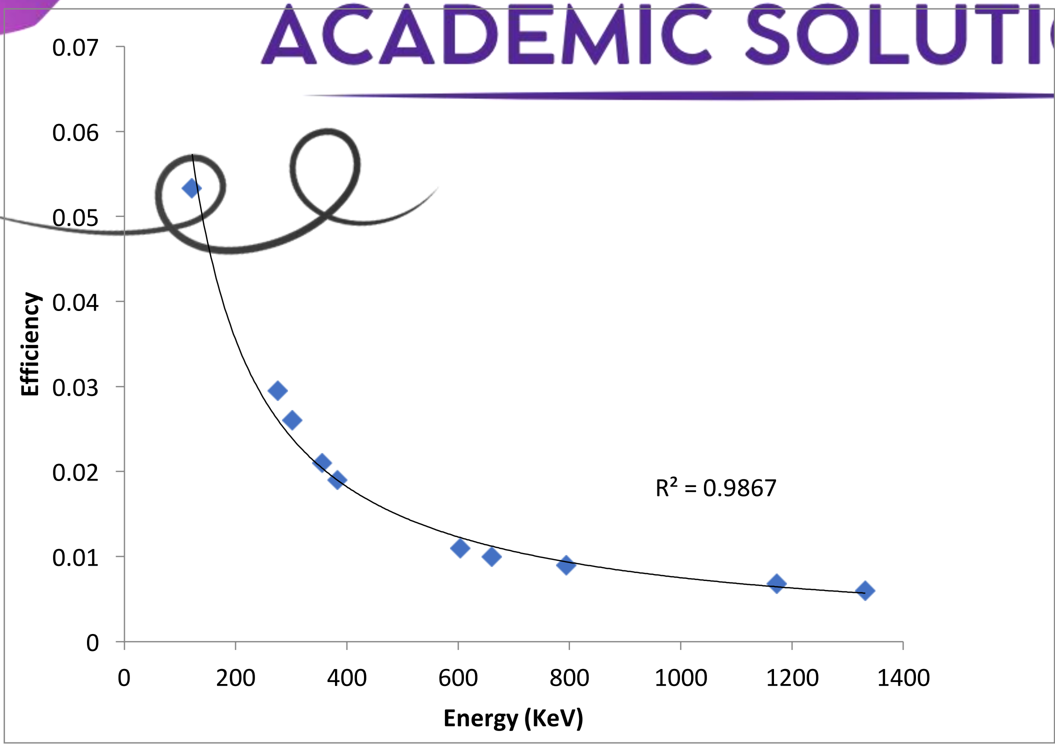

Figure 2.8. Efficiency calibration of HPGe setup

2.3.2.1.10. Quantitative radioactivity measurement

2.3.2.1.10.1. Background measurement

A hollow Marinelli beaker was employed for spontaneous background assessment with somewhat alike dimension that was employed for samples case and after that every estimated value of the sample was subtracted from the spontaneous background value.

$$C_n = C_s - C_b \qquad (2.3)$$

Whereas, net counts of sample are represented by C_n, sample counts by C_s and background counts by C_b.

2.3.2.1.10.2. Activity concentration

The following mathematical term was employed for the estimation of activity concentration for every individual observed sample (Beretka & Mathrew, 1985).

$$\text{Activity concentration} = \frac{Cn}{t \times E_\gamma \times m \times P_\gamma} \qquad (2.4)$$

where, net counts are shown by C_n, detection efficiency by E and yield % age of certain radionuclide or ejection probability of specific gamma ray by P_γ, sample mass in grams by m and counting time span of sample in seconds by t.

Equation 2.5 was employed for the evaluation of uncertainty for activity concentration measurements.

$$\frac{\Delta A}{A} = \sqrt{\left(\frac{\Delta s}{s}\right) + \left(\frac{\Delta P_\gamma}{P_\gamma}\right) + \left(\frac{\Delta CR}{CR}\right) + \left(\frac{\Delta M}{M}\right)} \qquad (2.5)$$

Where, A and ΔA are the activity concentration and its uncertainty, ε, Δs are the efficiency, P_γ and ΔP_γ are the emission probability of the gamma ray of interest and its uncertainty, CR and ΔCR are the net counts of gamma ray photopeak of interest and uncertainity, M and ΔM are the mass of the sample and its uncertainity.

2.3.2.1.10.3. Minimum detectable activity (MDA)

MDA is employed to detect the ability of computing system. MDA is the minimum expected activity that might be detected with 95 % of confidence level (Gilmore & Hemingway, 2008). ^{232}Th, ^{137}Cs, ^{226}Ra and ^{40}K radionuclides could be checked for MDA by this mathematical term:

$$MDA = \frac{4.66 \times \sqrt{Continum\ Counts + Background\ Peak\ Counts}}{t \times E, \times m \times P,} \quad (2.6)$$

Where, statistical coverage term is 4.66, unit of Bq kg^{-1} employed for MDA, while all other terms are likely common.

2.3.2.1.10.4. Radium equivalent activity (Ra_{eq})

Material comprises of radionuclides (^{232}Th, ^{226}Ra and ^{40}K) that are assessed for hazardous impact by widely employed radiological term called as Ra_{eq} (Xinwei, 2005). As collected sample don't have any uniformity in radionuclidic content and its distribution. Therefore, for radiation threat assessments, it is oftenly employed. It is the mere term that truly link with activity concentartion of Ra-226, K-40 and Th-232 and rely on the evidence which suggested that 259 Bq kg^{-1} for ^{232}Th, 4810 Bq kg^{-1} for ^{40}K and 370 Bq kg^{-1} for ^{226}Ra gives off the somewhat similar gamma ray dose rate and are represented as;

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_k \quad (2.7)$$

Where, activity concentration for ^{232}Th, ^{226}Ra and ^{40}K were expressed via respective terms A_{Th}, A_{Ra} and A_K. External dosage must not exceed to 1.5 $mGyY^{-1}$ iff Ra_{eq} value is less than to the maximum limit of 370 Bq kg^{-1}(Huy & Luyen, 2005).

2.3.2.1.10.5. Internal hazard index (H_{in})

Alpha particles are ejected from radon as well as its progenitors and might have serious threats to the respiratory trail of the humans. Thus, internal hazard index which computes the internal hazards originated from radon and its progenitors, and are described by the relation as (Krieger, 1981; Singh, 2011).

$$H_{in} = \frac{ARa}{185} + \frac{Ath}{259} + \frac{Ak}{4810} \quad (2.8)$$

Radiation impacts are trivial if and only if H_{in} must not exceed to unity.

2.3.2.1.10.6. External hazard index (H_{ex})

The assessment of gamma radiation hazards caused by spontaneous sources might be referred by the term H_{ex} (Ibrahim, 1999). The gamma dosage equivalent was permissibly limited to 1 $mSvy^{-1}$ and also presented via H_{ex} term (Singh, 2011). Beretka and Mathew model (Beretka and Mathew, 1985) was employed for the assessment of H_{ex};

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (2.9)$$

Radiation threats for corresponding radionuclides were insignificant, If and only if H_{ex} term was found to be less than unity. The H_{ex} (370 Bq kg^{-1}) is well determined radium equivalent activity which accounts for the determined unity value (Xinwei et al., 2005).

2.3.2.1.10.7. Exposure indices

Internal exposure index, external exposure index and alpha index, are collectively known as exposure indices. In current study, these indices were also computed. These terms are given as (Xinwei, 2005).

$$I_{ex} = \frac{A_{Ra}}{370} + \frac{A_K}{42000} + \frac{A_{Th}}{259} \quad (2.10)$$

$$I_{\alpha} = \frac{A_{Ra}}{200} \quad (2.11)$$

Where, A_k, A_{Ra} and A_{Th} be activities of respective radionuclide.

The other radiological terms like gamma index as well as representative index might be computed and are as under (NEA, O., 1979: Righi & Bruzzi, 2006).

$$I_\gamma = \frac{A_{Th}}{200} + \frac{A_{Ra}}{300} + \frac{A_K}{3000} \qquad (2.12)$$

$$I_r = \frac{A_K}{1500} + \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} \qquad (2.13)$$

I_r must not exceed the unity in order to preserve the insignificant radiation threats.

2.3.2.1.10.8. Absorbed dose rate (D)

Ionizing radiations which transmit energy for every mass of targeted substance per exposure time are described by radiation absorbed dose rate. Gray is its SI unit (1Gray = 1J kg^{-1}, 100 rad = 1 Gray) and rad is the old one unit (1rad= 100 $ergsgm^{-1}$).

Current study provides the absorbed dose rate caused by gamma rays that are computed through activity concentrations of respective radionuclides ^{232}Th, ^{40}K and ^{226}Ra as well as their precursors are assumed to be likely at radioactive equilibrium. The relation was employed as under;

$$D = (0.462 \times A_{Ra}) + (0.0417 \times A_K) + (0.604 \times A_{Th}) + (0.030 \times A_{Cs}) \qquad (2.14)$$

A unit $Bqkg^{-1}$ was employed for computing the activity concentration of respective radionuclide K, Ra, Th and Cs are depicted through A_k, A_{Ra}, A_{Th} and A_{Cs}. Though, these radionuclides has conversion factors like 0.0147 (nGy h^{-1}/ Bq kg^{-1}), 0.462(nGyh^{-1}/ Bq kg^{-1}), 0.604 (n Gy h^{-1}/ Bq kg^{-1}) and 0.030 (n Gy h^{-1}/ Bq kg^{-1}) respectively (Godoy et al., 1998; UNSCEAR, 2000).

2.3.2.1.10.9. Annual effective dose equivalent (AEDE)

The following relation was employed to compute the AEDE,

$$AEDE = Q \times D_{air} \times t \times O \times 10^{-6} \ (mSvy^{-1}) \qquad (2.15)$$

Where, Q known as conversion factor which is 0.7, In air the absorbed dose rate be D_{air} measured in (nGy Y^{-1}), ‘‘t’’ be time span in hours of a year (8760 hrs) as well as occupancy factor ‘‘O’’ give off AEDE in mSv y^{-1}(Ahad, 2004). It has values 0.8 and 0.2 for indoor and outdoor exposure respectively (UNSCEAR, 1988; Xinwei, 2005).

2.3.2.1.10.10. Excess lifetime cancer risk (ELCR)

At certain exposure, ELCR deals with the possibility of developing cancerous cells in a whole life-time. Following equation was employed to compute ELCR stimulated via gamma radiation.

Excess lifetime cancer risk (ELCR) = Average life duration (DL) × Risk Factor (RF) × AEDE (2.16)

Whereas, annual effective dose equivalent represented by AEDE, (DL) shows the duration of life that is 66 years (Rafique et al., 2010). While, fatal cancer risk as well as risk factor in Sv^{-1}. Since stochastic impacts could be produced by less dose background radiations, the public exposure might have value of 0.05 which is suggested in ICRP 60 technical report (ICRP, 1991; Taskin et al., 2009).

2.4. SURVEY METERS

Ludlum Micrometer-19, RTM 1688-2 and Garmin manufactured Global Positioning system (GPS) were employed to record the data in the field survey.

2.4.1. Ludlum Micrometer-19

The external or open-air gamma exposure rates as well as radon exposures could be evaluated with the help of portable surveyed meter named as “Model 19, Micro R Meter’’ fabricated by Ludlum Measurements INC (SWEETWATER, TEXAS). This employed setup within micrometer has a NaI (2.5 ×2.5 cm) with measurement ranges fall in 0 – 5000 µR/hr. Being highest efficiency of this device makes its wide employability among all of the gamma or radon survey meters. But different physical separation resulted into different grades of every product which shows its variation within values of survey meter. Survey meter has a 1 µRh^{-1} being the lowest scale that is approximately equivalent to 8.7n Gyh^{-1} (IAEA, 1979). Dose rates assessment was established corresponding to height level of 1000 mm from the earth surface. Next, these measurements were helpful in derivation of absorbed dose rates and outdoor as well as indoor annual effective doses within air over terrestrial basis. The exposure rates are expressed in µRh^{-1}, next it is converted into average absorbed dose rates via employing following equation (2.17).

$$1\ \mu Rh^{-1} = 8.7n\ Gyh^{-1} \qquad (2.17)$$

Figure 2.9 shows a Ludlum micromteter that was employed for current field study.

Figure 2.9. Ludlum micromteter-19 used in current field study

2.4.2. GPS Meter

GPS meter (explorist) was employed to record the sampling site coordinates for traceability purpose of the sampling sites in future. Figure 2.10 shows a GPS device that was employed to record coordinates for current field study.

Figure 2.10. GPS device used in current field study

2.4.3. RTM 1688-2

RTM 1688-2(SARAD GmbH D-01159 Dresden Germany) was employed to measure the indoor as well as outdoor radon concentration at various locations of Muzaffarabad, Azad Kashmir (see Figure 2.11).

Figure 2.11. RTM 1688-2(SARAD GmbH D-01159 Dresden Germany) used for field study

Further working principle details are given in chapter 3 under the section 3.1.5.1.

Chapter 3

RESULTS AND DISCUSSION

3.1. EXCESSIVE LIFETIME CANCER RISK ASSESMENT DUE TO SHORT-TERM INDOOR/OUTDOOR AMBIENT RADON AND GAMMA DOSE RATE EXPOSURES

This study aims at the ambient measurements of radon and gamma radiations dose rates in indoor and outdoor environment of Muzaffarabad. Radiation concentrations have been used for the risk assessment for the inhabitants of the area.

3.1.1. Study Area

The present study has been carried out in the city of Muzaffarabad, the state capital of Pakistani administered part of Jammu and Kashmir. It is located near the confluence of river Jhelum and Neelum at the location with coordinates 34.3551^0 N, 73.4769^0 E with an average height of 737 m (2418 ft) from sea level (https://en.wikipedia.org/wiki/Muzaffarabad). According to 2017 census the population of Muzaffarabad city is 149913 peoples (Statistical Yearbook, 2019). Muzaffarabad is the wettest region and the average rainfall exceeds 1400 mm (https://www.ajk.gov.pk/). Both, summer and winter seasons are extreme in terms of rise and fall of temperature.

3.1.2. Geology Of The Area

Geological map of the area, displaying different formations, is shown in the Figure 3.1. Area of study is exposed by Hazara, Muzaffarabad and Murree formations. The Hazara Formation consists of slates, phyllite and shales with smaller occurrences of graphite, gypsum and limestone layers. Slates are black to dark brown, weathered, highly jointed and fine grained. These slates are the metamorphic product of shales. This rock unit also hosts the quartz veins. According to Calkins et al. (1969), thickness of limestone beds is up to 150 m and calcareous phyllite and gypsum are 30-120 m thick in area. This formation is corelated with the Precambrian Salt Range Formation based on presence of evaporites in both the formations (Latif, 1973).

Muzaffarabad Formation is composed of stromatolitic and cherty dolomite, grey and cherty white bands limestone and black color shales (Baig & Snee, 1995). The Miocene age Murree Formation is well exposed in the study region and comprised of interbedded siltstones, sandstones, claystones, and shales. Sandstone is the dominant rock unit of the Murree Formation along with clays and shales. The Holocene alluvial deposits are also present along Neelum and Jhelum River in Plate, Chellah Bandi, Dhanni, Maira Tanolian and Dherian areas.

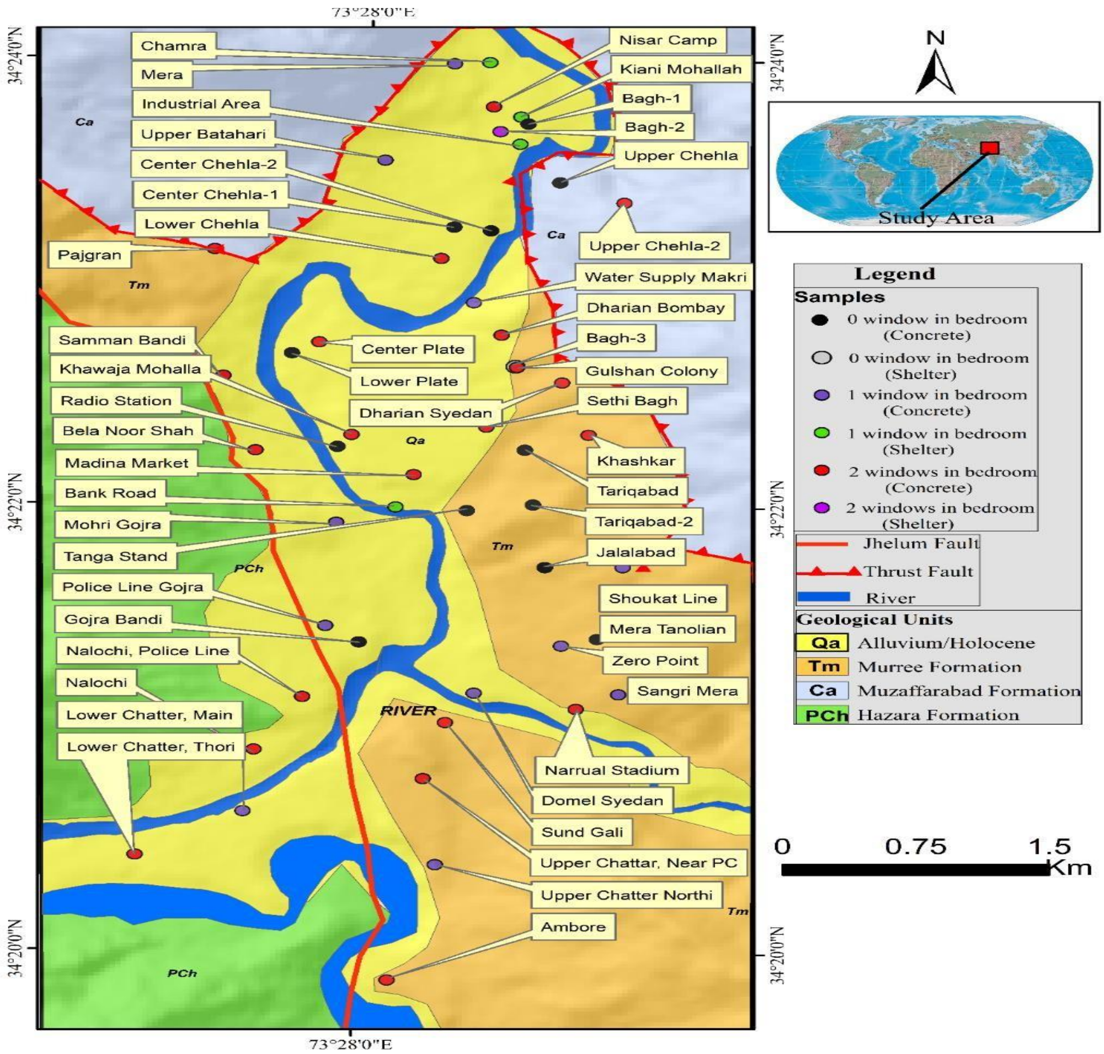

Figure 3.1. Geological map showing different formations in investigated study area

3.1.3. Materials and Methods

3.1.3.1. Measurement methodology for ambient indoor/outdoor radon

Ambient values of radon were measured using RTM 1688-2 (SARAD GmbH D-01159 Dresden Germany). The detector consists of optimized high voltage chamber. Concentration of ^{222}Rn is measured indirectly from its short living progenies. After the decay of radon, some of atomic orbital electrons get scattered by emitted alpha particles, the remaining ^{218}Po nuclei become positively charged for a short period of time. Electric field forces collect these ions on the surface of semiconductor. Within the chamber, the radon concentration is proportional to the number of collected ^{218}Po ions. Polonium 218, being radioactive having half-life 3.05 minutes, decays and about 50 % of all decays are registered by the detector. The equilibrium between radon decay rate and Polonium-218 activity is achieved almost after 5 half-lives i.e. ~15 minutes.

This time is considered as minimum time span needed to record radon concentration. The RTM 1688-2 works in two calculation modes for the Radon concentration measurement, one (Slow mode) includes measurement of both, Po-218 and Po-214 decays and the other one includes Po-218 only (Fast). The detector is insensitive to humidity. Measurement range of the instrument is 0 to 10 MBqm^{-3}. It is provided with the sensors to record relative humidity (0 ... 100 %), Temperature (-20 ... 40 °C) and barometric pressure (800 ... 1200 mbar). For the current study data for each site was recorded for the consecutive three days at specific times and measurement interval of detector was set for 40 minutes. Average of radon concentration was calculated that is taken as representative value of particular site.

3.1.3.2.Measurement methodology for ambient indoor/outdoor gamma dose rates (GDR)

A portable radiometric instrument, Ludlum micrometer 19 (manufactured by Ludlum measurements Inc. USA) have been used for the ambient indoor/outdoor gamma dose rate measurements in air one meter above ground at fifty different locations of Muzaffarabad. The instrument is a highly sensitive gamma microR meter employing an internally housed, 2.5 x 2.5 cm (1″ x 1″) NaI detector. The measuring range of the instrument is 0 to 5000 μRh^{-1}. Sensitivity of the device against gammas emitted from ^{137}Cs is approximately 175 cpm per μRh^{-1}.

Three measurements were taken for each location, spanning over time period of 2 minutes. Gamma dose rates presented in the current study are mean values of three measurements taken at each location. The exposure rate measured in μRh^{-1} was converted into absorbed dose rate μGyy^{-1} using the conversion factor (Rafique et al., 2014):

$$1\frac{\mu R}{hr} = 76.212\ \mu Gy/y \qquad (3.1)$$

3.1.4. Results and Discussion

3.1.4.1. Analysis of indoor/outdoor radon concentrations

As discussed earlier, short-term radon and gamma dose rate measurements were taken in indoor and outdoor environments at fifty locations of Muzaffarabad. Statistical parameters obtained from the study are presented in Table 3.1. For both indoor and outdoor radon concentrations a step horizontal connection graph that creates a right-angle connection between data points have been drawn, with an initial horizontal line. The data points are not displayed on the graph (see Figure 3.2). For indoor radon measurements, radon concentrations varied in the range of 16 to 150 Bq m^{-3}. Minimum concentration was found on location number 34 i.e., Gojra Bandi, whilst maximum concentration was recorded at location number 24 i.e., Tariqabad. For outdoor radon measurements, minimum concentration i.e., 7 Bq m^{-3} was recorded at location 34 whilst maximum concentration 31 Bq m^{-3} was found on location number 10 (center Chehla-1) and 34. Average values of indoor and outdoor radon concentrations were found as 46.9 and 13.3 Bq m^{-3} respectively. Normal distribution fit has been applied on indoor and outdoor radon data as shown in Figure 3.3. Normal distribution fitting on indoor radon data shows clearly that indoor radon is affected by several parameters like door, windows, embrasure etc. Range of data can observe from Figure 3.4, which also shows outlier values in the data.

Table 3.1. Statistical parameters associated with indoor and outdoor radon concentrations

Attributes	Indoor Radon (Bq m^{-3})	Outdoor Radon (Bq m^{-3})	Indoor Gamma Dose rates (µGy y^{-1})	Outdoor Gamma DoseRates (µGy y^{-1})
Total Measurements	50	50	50	50
Min	16	7	419	495
Max	150	31	1486	1029
Mean	46.9	13.3	846	777
Std. error	3.58	0.77	28.87	16.36
Variance	640.7	29.85	41682	13394
Stand. Dev	25.31	5.46	204	116
Median	32	12	800	762
Skewness	1.92	1.52	0.81	0.031
Kurtosis	4.56	2.79	1.68	0.119

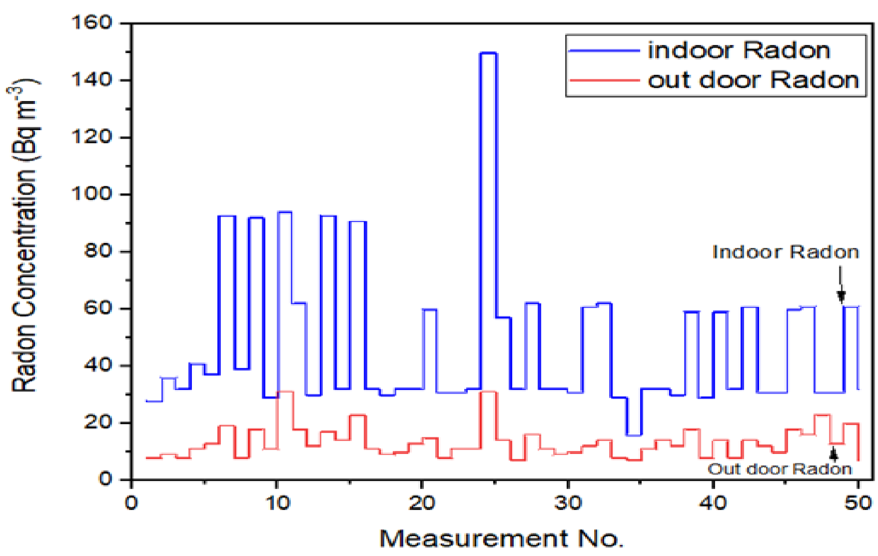

Figure 3.2. Indoor and outdoor radon concentrations a step horizontal connection graph

For normality check, goodness of fit test has been employed on indoor/outdoor radon data. For indoor radon, using Kolmogorov-Smirnov goodness-of-fit test (Chakravarti & Roy, 1967), the p-value was obtained as 5.21631×10^{-4}. Since p-value is less than the level of significance (0.05), this shows indoor radon does not follow the normal distribution. For outdoor radon, estimated p-value was 0.10201, confirming that the data follows normal distribution trend.

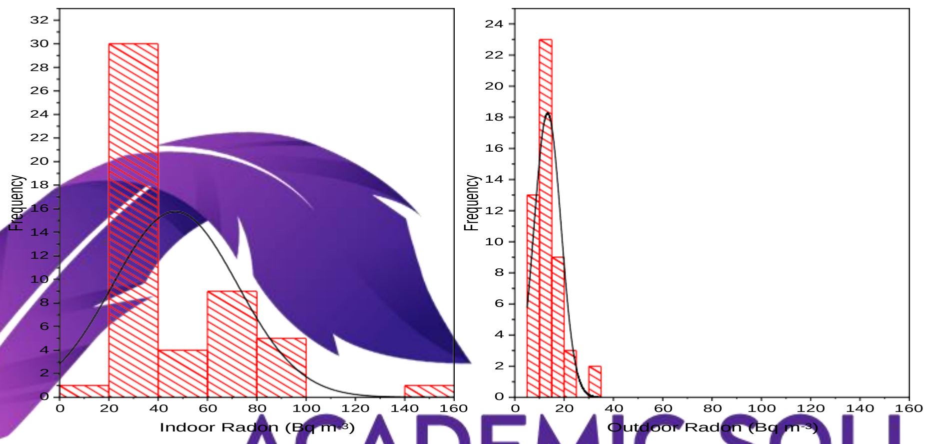

Figure 3.3. Normal distribution fitting on indoor and outdoor radon data

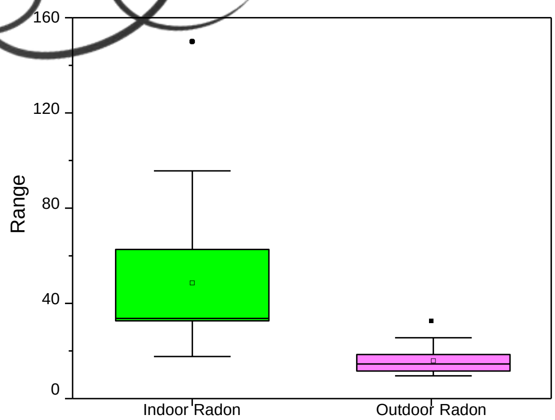

Figure 3.4. Box plot of indoor and outdoor radon data

Maximum indoor and outdoor radon concentration 150 and 31 Bq m^{-3}, outliers, were obtained inside one of Tariqabad house and in open air nearby it. Tariqabad is lying in Murree formation, which is well exposed of interbedded siltstones, sandstones, claystones, and shales. Sandstone along with claystone's and shales are the dominant rock units of the Murree Formation. Conventionally on the average, black shale contains 10 ppm of uranium concentration, whilst other shale types contain 3 ppm of uranium. On the other hand, sandstones contain 2.2 ppm of uranium (Nagda, 1994).

Most likely, rock types with uranium concentration greater than 5 ppm raises indoor radon problems. Rocks which are expected to raise indoor radon problems includes, carbonaceous black shales with uranium-bearing organic compounds; autunite; tyuyamunite, glauconite bearing sand stones with radium and uranium-bearing iron-oxides and some fluvial sandstones with uraninite, coffinite, pitchblende and secondary uranium minerals etc. (Nagda, 1994). The other factor that might have contributed in high values of indoor radon is size of room. Investigated room was without any window, embrasure and with only one door.

3.1.4.2. Calculation of annual effective dose, ELCR and LCC associated with radon exposure

Subsequent ambient values of indoor and outdoor radon have been used to calculate Annual Effective dose (DT), Annual Effective dose to lungs (ET), ELCR, and lung cancer cases per year per million person (LCC).

The annual effective dose (E_{Rn}), due to radon exposure have been estimated using equation 3. 2 (UNSCEAR, 2000).

$$E_{Rn} = C_{Rn} \times 0.4 \times T \times O_{indoor} \times DCF_{Rn} \quad (3.2)$$

Where, C_{Rn} is concentration of radon, 0.4 is the equilibrium factor for radon and its decay products, T are hours per annum (24 h × 365 days =8760 hy^{-1}), O_{indoor} is indoor occupancy factor and DCF_{Rn} is dose conversion factor (9 ×10^{6} mSvh^{-1} per Bqm^{-3}).

The Annual effective dose delivered to lungs (E_{lungs}) due to radon exposure have been estimated by equation 3.3:

$$E_{lungs} = D_{annual\ absorbed} \times W_R \times W_T \qquad (3.3)$$

Where,

$D_{annual\ absorbed}$ = annual absorbed dose (mSvy^{-1}); W_R is radiation weighting factor (20 for alpha particles as recommended by the ICRP); and W_T is tissue weighting factor (0.12 for lung) (IARC, 1988).

The Excess lifetime cancer risk (ELCR), due to radon exposure, have been evaluated using the Equation 3.4 (Vaeth & Pierce 1990; Rafique et. al 2014):

$$ELCR = E_{Rn} \times D_L \times CR_F \qquad (3.4)$$

Where E_{Rn} is the annual effective dose, D_L is the average duration of life estimated to a 66.7 years and CR_F is the fatal cancer risk per Sievert (5.5 ×10^{-2} Sv^{-1}) recommended by ICRP, (103).

The lung cancer cases per year per million person (LCC) is estimated using the risk factor for lung cancer induction 18 × 10^{-6} mSv^{-1} using the Equation 3.5 (Fakhri et al., 2015; Özen et al., 2018):

$$Lung_{cancer\ cases} = E_{Rn} \times 18 \times 10^{-6} \qquad (3.5)$$

For indoor measurements, annual effective dose (E_{Rn}) due to radon exposure ranges from 0.4 to 3.78 mSvy^{-1} with average value of 1.18 mSvy^{-1}. Radon doses delivered to lungs varied from 0.97 to 9.08 mSv/yr with an average value of 2.84 mSvy^{-1}. Excess lifetime cancer risk (ELCR) varied from 1.49 ×10^{-3} to 14.01×10^{-3} with mean value 4.38×10^{-3}.

Data of current study shows that lung cancer cases per year per million person ($Lung_{cancer\ cases}$) ranges between 7.26 to 68.1 per million persons per year with mean value of 21.2 per million persons per year. These results are lower than as reported by Sherafat et al., 2019, where $Lung_{cancer\ \ cases}$ values ranged between 5.04 to 199.2 with mean value of 46.8 per million persons per year. Present study results reported for $Lung_{cancer\ \ cases}$ are also lower than ICRP recommended values viz. 170–230 per million persons (ICRP, 1993).

Table 3.2. Annual Effective dose (E_{Rn}), Annual Effective dose to lungs (E_{lungs}), ELCR, and lung cancer cases per year per million person ($Lung_{cancer\ cases}$)

Attribute	E_{Rn} (mSvy^{-1})	E_{lungs} (mSvy^{-1})	ELCR $\times 10^{-3}$	$Lung_{cancer\ cases}$
For Indoor Measurements				
Average	1.18	2.84	4.38	2.12×10^{-5}
Minimum	0.40	0.97	1.49	7.26×10^{-6}
Maximum	3.78	9.08	14.01	6.81×10^{-5}
For outdoor Measurements				
Average	0.126	0.30	0.46	2.26×10^{-6}
Minimum	0.06	0.159	0.24	1.19×10^{-6}
Maximum	0.29	0.70	1.08	5.27×10^{-6}

3.1.4.3. Relation between indoor and outdoor radon concentrations

Data obtained for indoor radon have been plotted against outdoor radon levels to explore, if there exists, relationship between two types of data set as shown in Figure 3.5.

The value of coefficient of determination (R square) is obtained as 0.67. Which shows a positive relationship between indoor and outdoor radon concentrations. Especially it was noticed that indoor values of radon concentration were following increasing or decreasing pattern of outdoor radon concentration. Relationship between indoor and outdoor radon is shown by equation 3.6.

$$Rn_{outdoor} = a + b \times Rn_{indoor}$$

$$Rn_{indoor} = \frac{Rn_{outdoor} - a}{b} \qquad (3.6)$$

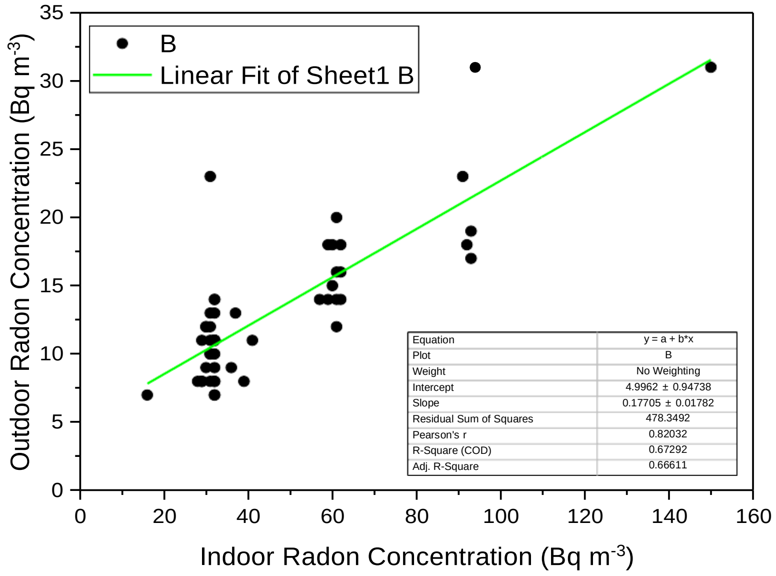

Figure 3.5. Plot of graph between indoor and outdoor radon concentrations

Where, Rn_{indoor} is radon concentration in indoor environment, whilst $Rn_{outdoor}$ is radon concentration in outdoor environment also a is intercept and b is slope of linear fit on the data. As may be seen from Figure 3.5 that the intercept 'a' has the value 4.9962±0.9473 and slope 'b' has value 0.17705±0.01782.

3.1.4.4. Analysis of indoor/outdoor gamma dose rates (GDR)

Statistical parameter obtained for GDR are displayed in Table 3.1. For indoor measurements, GDR ranges from 419 to 1486 µGy y^{-1} with mean value of 846 µGy y^{-1}. Whilst outdoor GDR varied from 495 to 1029 µGy y^{-1} with mean value 777 µGy y^{-1} as can be seen in figure 3.6 and 3.7. The ratio of indoor to outdoor gamma dose rate was found as 1.088, which is nearly equal to unity. The reason for indoor to outdoor ratio close to unity may be that majority of the dwellings in the city have been built by construction material of local origin. The dwellings with higher gamma dose rates have particularly tiled, cemented floors, decorative stones like granite, marble toping and flooring, concrete walls and ceilings. Granite and marble are usually procured from a different cities and these construction materials, may have greater radioactivity content than the construction material of local origin. In order to check, if the data follows normal distribution, goodness of fit test has been applied on indoor/outdoor GDR data. For indoor GDR using Kolmogorov-Smirnov goodness-of-fit test (Chakravarti & Roy, 1967), the obtained p-value was 0.57. This shows indoor GDR follows normal distribution. Similarly, for outdoor GDR, p-value was 0.31 confirming again that data follows normal distribution as can be seen in figure 3.8.

The indoor and outdoor gamma dose rates, 1486 and 953 μGy y^{-1} respectively, were found highest inside and outside of house situated at Narrual. Location of measurement was at the boundary of Murree formation and Holocene alluvial deposits. Gamma dose rates are usually provided by either neutron activated radionuclides or naturally occurring radionuclides like, ^{40}K, ^{238}U, ^{232}Th, ^{226}Ra etc. Again, geology of the area plays important role in establishing and understanding of levels of background radiations and higher GDR may be due to presence of naturally occurring and anthropogenic radionuclides in the area.

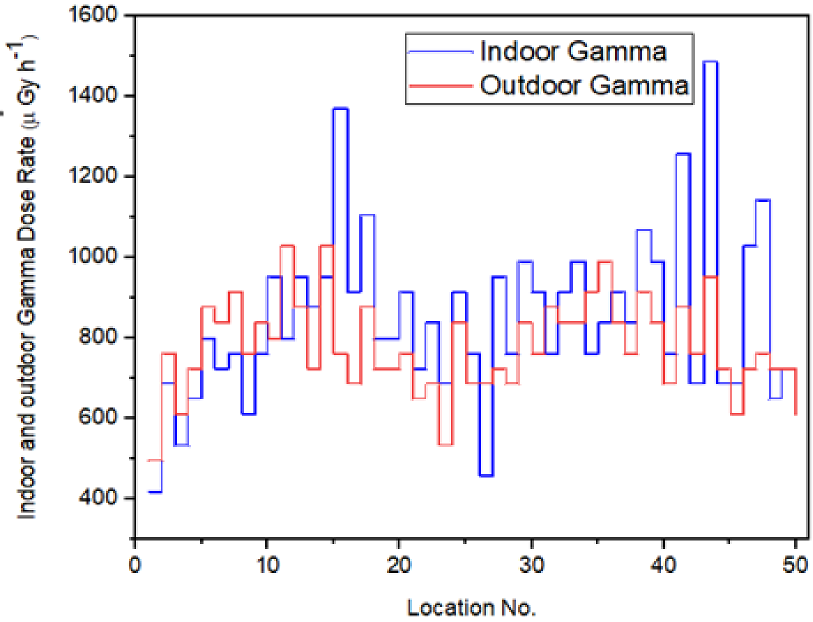

Figure 3.6. Indoor and outdoor GDR as a step horizontal connection graph

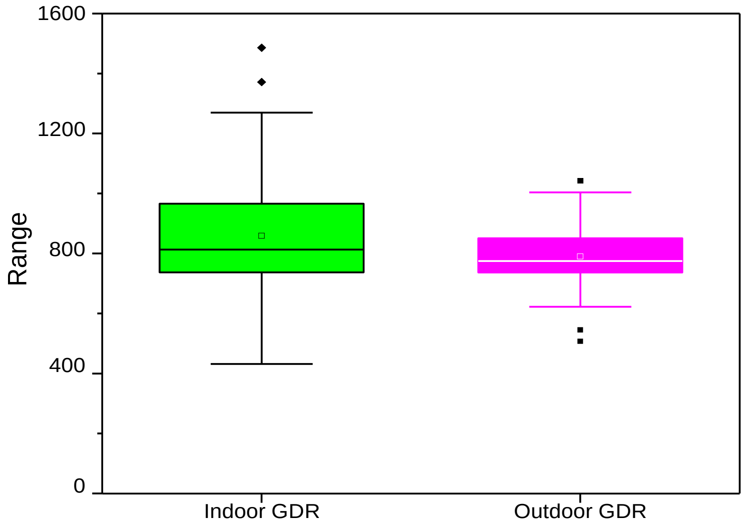

Figure 3.7. Box plot of indoor and outdoor GDR data

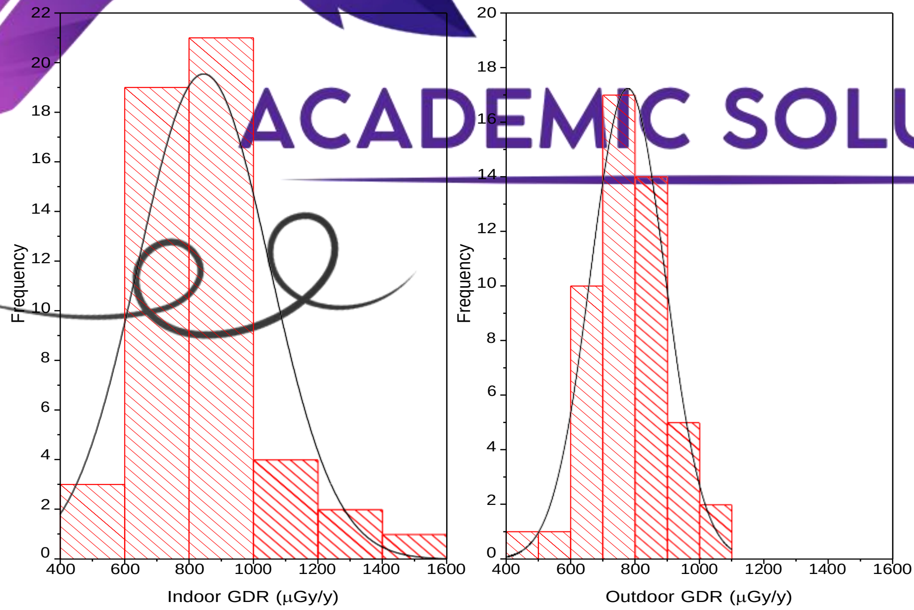

Figure 3.8. Normal distribution fitting on indoor and outdoor GDR

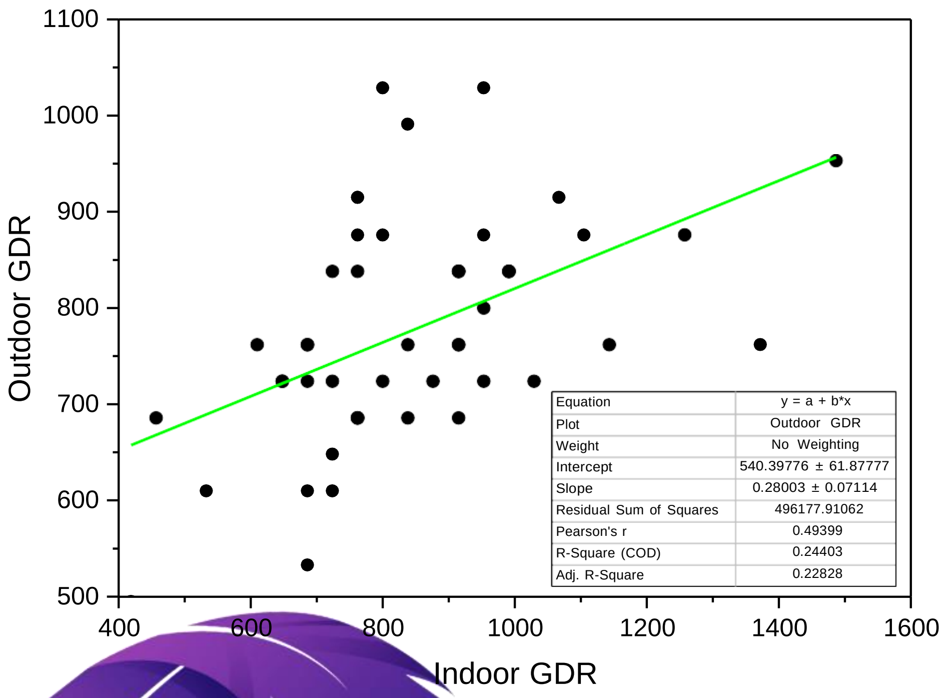

Figure 3.9. Plot of graph between indoor and outdoor GDR

Figure 3.9 shows plot of graph between indoor and outdoor gamma dose rates. For predictive analysis linear fitting on data has been done. The obtained R-square value i.e., 0.244 shows a weak positive relationship between indoor and outdoor GDR.

3.1.4.5. Estimation of annual effective dose equivalent (AEDE)

Quantified absorbed GDR have been used to calculate the annual effective dose equivalent (AEDE) received by inhabitants of study area. Dose conversion factor of 0.7 Sv/Gy and the occupancy factor (O.F) for indoor and outdoor was 0.80 (UNSCEAR, 1988), and 0.20 (UNSCEAR, 1988) have been used for the purpose of estimation of AEDE. In the UNSCEAR, (1993) report the Committee used 0.7 Sv/Gy for the conversion coefficient from absorbed dose in air to effective dose received by adults. The annual effective dose equivalent (AEDE) for indoor and outdoor environment was calculated using equation 3.7 and 3.8 (Rafique et al., 2014);

$$AEDE_{Indoor}\left(\frac{mSv}{y}\right) = GDR\left(\frac{\mu Gy}{y}\right) \times 10^{-3} \times \frac{8760h}{y} \times \frac{0.7Sv}{Gy} \times 0.8 \quad (3.7)$$

$$AEDE_{Outdoor}\left(\frac{mSv}{y}\right) = GDR\left(\frac{\mu Gy}{y}\right) \times 10^{-3} \times \frac{8760h}{y} \times \frac{0.7Sv}{Gy} \times 0.2 \quad (3.8)$$

For the indoor measurements, min, max and mean values of AEDE were found as 0.21, 0.42 and 0.73 mSv y^{-1}. Whilst for outdoor measurements, min, max and mean values of AEDE were found as 0.06, 0.13 and 0.1 mSv y^{-1}.

3.1.4.6. Excess lifetime cancer risk (ELCR)

AEDE values have been used for the estimation of Excess Lifetime Cancer Risk (ELCR) using equation (3.9) (Rafique et al., 2014).

$$ELCR = AEDE\ (mSv/y) \times D_L \times CR_F \quad (3.9)$$

D_L is the average duration of life estimated to 66.7 years and CR_F is the fatal cancer risk per Sievert (5.5×10^{-2} Sv^{-1}) recommended by ICRP, (103).

For low dose background radiations which are considered to produce stochastic effects, ICRP, (60) uses values of 0.05 for the public exposure (Taskin et al., 2009; Rafique et al., 2014).

For indoor GDR measurements, min, max and mean values of ELCR were found as 0.69×10^{-3}, 2.45×10^{-3} and 1.38×10^{-3}. For outdoor GDR measurements, min, max and mean values of ELCR were found as 0.2×10^{-3}, 0.42×10^{-3} and 0.32×10^{-3}

3.2. STUDY OF GROSS ALPHA, GROSS BETA AND NATURAL RADIOACTIVITY IN SOIL SAMPLES OF DISTRICT MUZAFFARABAD

3.2.1. Abstract

Results for the activitiy concentrtion of primordial radionuclides, gross alpha and gross beta activities in soil samples collected from 29 locations of Azad Kashmir are presented. Soil samples were analysed, for possible radionuclide contents and relevant health implications, by high-resolution γ-ray spectroscopy and α/β counter ASC-950-DP

Protean instrument. The alpha activity varied from 77.31 ± 9.95 to 440.08 ± 16.48 Bq kg^{-1} with an overall average value of 234.88 ± 1.69 Bq kg^{-1}. While beta activity varied from the minimum detection level, i.e., <MDL to 361.55±149.33 Bq kg^{-1}, with average value for all samples estimated as 235.65 ± 149.98 Bq kg^{-1}. Activitiy concentrtion of ^{232}Th, ^{226}Ra and ^{40}K were calculated using γ-ray spectroscopy and were subsequently utilized for the estimation of radiation doses and radiological hazards. Activity concentrations due to ^{40}K, ^{232}Th and ^{226}Ra were found in the ranges 213.54 ± 17.22 to 1205.83 ± 12.82, 26.11 ± 3.72 to 84.70 ± 4.63 and 13.74 ± 1.46 to 62.23 ± 4.29 Bq kg^{-1}, with average values 616.22 ± 29.20, 55.83 ± 5.74 and 37.91 ± 2.35 Bq kg^{-1} respectively, whilst, activity concentration due to anthropogenic radionuclide ^{137}Cs was found in the range from minimum detection limit, i.e., ≤0.50 to 8.82 ± 0.83 Bq kg^{-1}. Average value for ^{137}Cs sample was found as 3.43 ± 0.28 Bq kg^{-1}. Excess lifetime cancer risk (ELCR) for indoor occupation varied from 4.94 × 10^{-4} to 1.82 × 10^{-3} and for outdoor occupation 1.32 × 10^{-4} to 4.62 × 10^{-4}. Overall excess lifetime cancer risk (ELCR) for the current study was estimated as 1.55 × 10^{-3}. The average values of radionuclide, in presently investigated soil samples, were within the worldwide range (50 Bq Kg^{-1}) for radium radionuclide contents, whilst higher for thorium and potassium radionuclides contents. Current study results can serve as baseline data for any nuclear emergency in future and may be helpful in setting radiological map of country and devising nuclear regulatory standards for background radiations in country.

3.2.2. Introduction

Current study is aimed to measure the gamma radionuclide, gross alpha and gross beta activity contents within soil samples collected from Muzaffarabad district of Azad Kashmir. Radiological health hazard, excessive lifetime cancer risk assessment has been carried out and results for present study have been compared with the data available in literature.

3.2.3. Area Study

The study has been carried out in Muzaffarabad, capital of Pakistani administered part of Jammu and Kashmir. Muzaffarabad district is surrounded from west by Pakistani province Khyber-Pakhtunkhawa, from east by Kupwara and Baramulla districts of Indian administered Jammu and Kashmir, and from north Neelum district of Azad Kashmir (https://en.wikipedia.org/wiki/Muzaffarabad).

3.2.4. Sampling

A total of 25 representative area surface soil samples were collected about 2 cm depth from surface during the month of July 2019 from Muzaffarabad metropolitan ranges such as Makri, Thallah Makri, Bella Makri, Upper Plate, Centre Plate, Residential Block Chellah University, Vermi Tech Unit Chellah Campus, Gulshan Colony, Madina Market, Janjua Market, Nisar Camp, Domail, Ambore, Naloochi, Hassan Gallian, Lamiyan Pattian, Gori Phear, Garri Dupatta, Pir Chinasi, Sathra Upper Jalalabad, Tariqabad, Baldiya Office and Sethi Bagh. Four (04) more samples from different districts Rawalakot, Jhelum valley, Bagh and Neelum valley at respective sites Tolipeer, Reshian, Sudhan Gali and Nagdar were collected for the purpose of comparison of radionuclide contents and hazards. Sample collection sites, in Muzaffarabad, have been shown in figure 3.10.

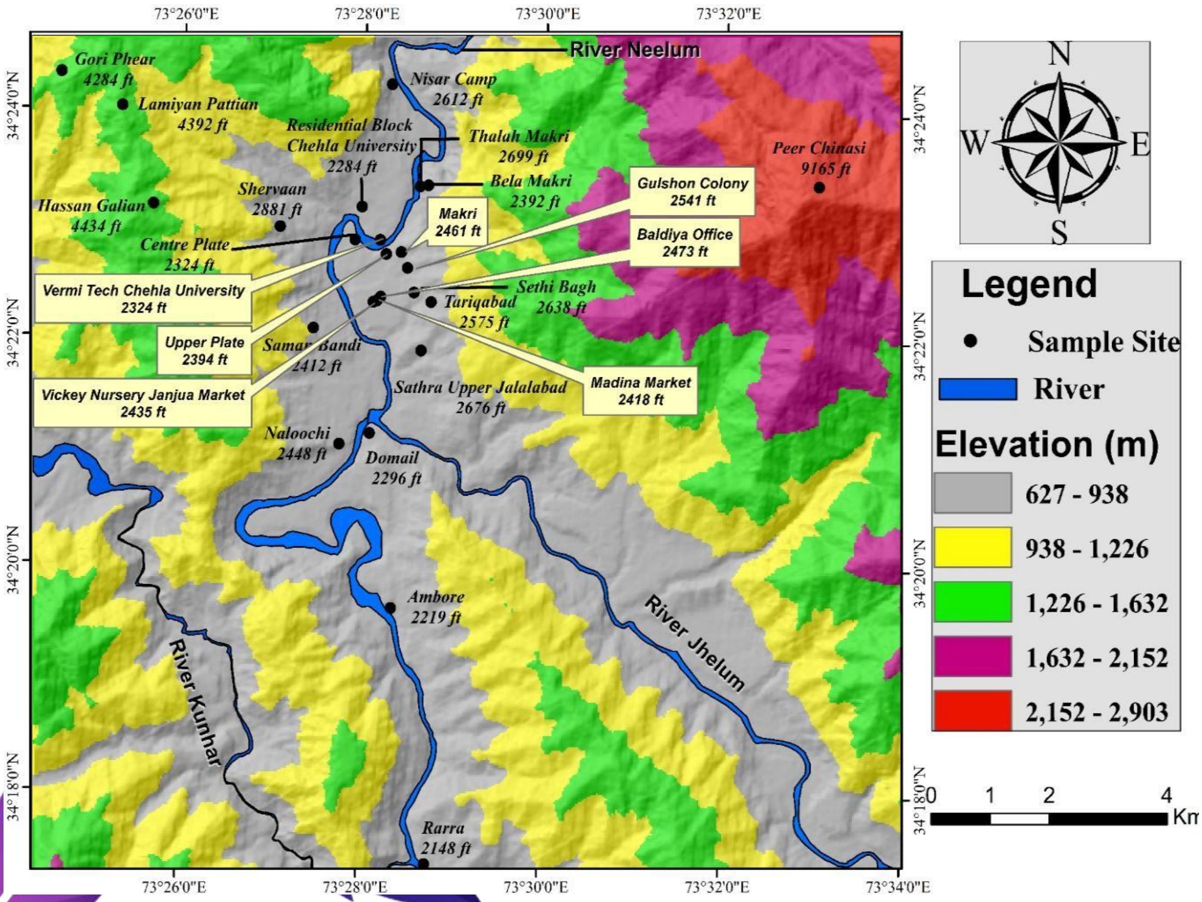

Figure 3.10. Samples collection sites in Muzaffarabad, Azad Kashmir

Soil samples weighing approximately 2 kg were packed inside polyethylene bag with proper cataloguing and GPS coordinates were measured using Magellan Explorist 210 GPS Receiver 2005, before bringing samples to the Radiation Physics Laboratory of Physics Department of the University of Azad Jammu and Kashmir. The purification of soil samples was ensured by exterminating stone pieces, grasses, lefty roots and residual fragments. Then samples were subsequently crushed and homogenised via mortar, pestle and sieved, respectively. The procedure adopted in pre-sampling and sampling processes was in line with the guidelines mentioned in IAEA-TECDOC-295 (IAEA, 1989).

3.2.5. Experimental

3.2.5.1. Pretreatment for gamma measurements

Samples were housed within oven for the period of six (6) hours at temperature of 110° C to make soil samples dried and moisture free. The soil samples each weighing 200 g were then packed in Merinelli beakers, labelled and hermetically sealed. In order to achieve secular equilibrium among the different progenies of ^{238}U and ^{232}Th decay series the sealed samples were preserved for the period of 28 days (Rafique et al., 2011; Rahman et al., 2011; Rahman et al., 2012).

3.2.5.2. Pretreatment for gross alpha and gross beta measurements

Powdered soil sample of 1 g each was placed within Planchet of stainless steel. Ethyl alcohol was employed to distribute the soil sample evenly within the planchet. Afterwards, alcohol was evaporated via IR lamp kept just above the planchet and then immediately placed within the detector. Counting time of 3000 s was selected for each gross alpha and beta activities.

3.2.5.3. Gross α/β detection

α/β counter (ASC-950-DP) Protean instrument corporation was used to get an estimate of gross alpha and gross beta activity contents in soil samples. System was calibrated for beta and alpha activity via respective standard sources like ^{90}Sr having activity 409.2 Bq and ^{241}Am having activity 693.8 Bq. The background counts were detected via hollow planchet counted for 3000 s. Then, background counts were subtracted through net counts to obtain samples counts. Quality of program was assured to make radiological monitoring rationally effective and reasonably valid. Population dose was predicted by vital factors like quality of sampling and calculated uncertainty and measurement and validation monitoring objective to approve whether the results could be accepted or unaccepted. The reproducibility as well as accuracy of counter was verified periodically on weekly basis. Detector and shielding ensured for non-contaminated radioactive contents via monitoring it without source to assess net background counts. ^{90}Sr and ^{241}Am sources were employed respectively to check beta and alpha efficiencies of low-level α/β counter. Detection performance of instrument for alpha background is 0.2 cpm and for beta background 40 cpm.

3.2.5.3.1. Calculation for gross α/β activity

Equation (3.10) was used to get an estimate of gross α/β activity (Kurnaz et al., 2020).

$$A_{\alpha\beta} = \frac{I}{m \times 60} \quad (3.10)$$

Where, sample's activity of gross α or gross β (Bq kg^{-1}) is represented by '$A_{\alpha\beta}$' and 'm' stands for sample weight within planchet in (kg) and I is obtained by the relation given as:

$$I = \frac{(N - B)}{s}$$

Where, N stands for count rate of sample in (1/s), and B represents count rate from background in (1/s) and 'ε' shows efficiency of detector for both alpha as well beta calculations.

While, Equations (3.11 and 3.12) was employed to calculate minimum detectable activity and measured uncertainty respectively:

$$MDA = \frac{2.71 + 4.65\sqrt{R_b \times t}}{m \times t \times s} \quad (3.11)$$

$$O = \frac{\sqrt{sam\ \ le\ count\ rate + backgroundcount\ rate}}{t} \times 2 \times \frac{1}{s \times 60 \times m} \quad (3.12)$$

Where, background count rate is represented by 'R_b'(cps) and counting time for background and sample denoted by 't' in seconds, m is mass of the sample, while, counting efficiency is shown by 'ε' (Görür et al., 2011).

3.2.5.4. Gamma spectroscopy using HPGe

PC-based high-resolution gamma spectrometry (HPGe) system was used to analyze soil samples for gamma activity (Debertin & Helmer, 1988). P-type closed-end coaxial high purity germanium detector (Canberra Industries Inc., USA) coupled with PC-based Multi-Channel Analyser (MCA) was used as spectroscopic system. The detector had an active volume of 180 cm^3. As compared with NaI(Tl) detector the relative efficiency of HPGe detector was 30 % with active volume of 180 cm^3. For gamma ray photon of energy 1332 keV from standard calibration ^{60}Co source the energy resolution was found to be 2.0 keV (FWHM).

The detector was shielded, to reduce background radiations, with 15 cm thick lead having inner lining of 3 mm thick copper and 4 mm thick tin layers. Shield cavity was kept with 25 × 25 cm^2 inner size area (Heusser et al., 1989; Westmeier, 1992). IAEA soil-326 was used to calibrate system and soil-375 was used as reference material to ensure reliability of counting efficiency. ^{152}Eu was used as a standard source for the purpose of energy calibration of the spectrum as it covers both low and high energy ranges. Each soil sample was counted continuously for the period of 18 hrs. Software Genie 2000 version 2.1 (Canberra, USA) was used for acquiring, displaying and analyzing the gamma spectrum from XMCA spectroscopy. The radionuclides ^{226}Ra, ^{137}Cs, ^{232}Th and ^{40}K were identified from the spectrum peaks appearing at energies 351.99 keV(^{214}Pb), 661.62 keV(^{133} Ba), 911.07 keV(^{228}Ac) and 1460.75 keV respectively (Lucia et al., 2006).

The minimum detectable activity (MDA) for the radionuclides ^{232}Th, ^{226}Ra, ^{40} K and ^{137}Cs was evaluated using following equation (Ellison & Williams, 2003).

$$MDA = \frac{K(N_C+N_B)^{0.5}}{y(E).P_\gamma.T_C.M} \qquad (3.13)$$

Where, MDA is expressed in Bq kg^{-1} and the multiplying factor K=4.66 is a statistical coverage factor, N_C continuum counts, N_B is background peak counts, $5(E)$ is the photopeak efficiency, P_γ is gamma ray emission probability i.e., yield, T_C is counting time and M is dry weight of sample in kg. MDA for the radionuclide's ^{232}Th, ^{226}Ra, ^{137}Cs and ^{40}K were found as 2.25, 3.60, 1.35 and 6.70 Bq Kg^{-1} respectively.

The activity concentration levels of radionuclides ^{226}Ra, ^{232}Th, ^{137}Cs, and ^{40}K were calculated using the relation suggested by (Lucia et al., 2006).

$$A = \frac{N_i}{y_i \times P_{\gamma i} \times C_i \times t} \quad (3.14)$$

Where, N_i is the difference of net peak counts for sample and background counts, 5_i is the efficiency of the detector for the corresponding peak, $P_{\gamma i}$ is the emission probability, C_i are net counts, and t is counting time.

3.2.5.5. Estimation of hazard indices and dose rates

Different hazard indices, see in Table 3.3, were estimated which include radium equivalent activity, Ra_{eq}, internal, external hazard indices (H_{in}, H_{ex}) (Beretka & Mathew, 1985), Gamma index (I_γ) (EC, 1999; Righi & Bruzzi, 2006), the representative level index I_r (NEA, 1979), and Alpha index (I_α) (Righi & Bruzzi, 2006; Xinwei et al., 2006). Also, mathematical expressions for dose rate, annual effective dose, outdoor and indoor annual effective doses are mentioned in Table 3.4.

Table 3.3. Formulae for estimation of hazard indices from activity mass concentrations in soil samples of different origins

Hazard Index / Formula	Remarks / Characteristics	Reference
Radium equivalent activity, $Ra_{eq}\left(\frac{Bq}{Kg}\right)$		
$Ra_{eq} = \left(A_{Ra} + \frac{370}{259}A_{Th} + \frac{370}{4810}A_k\right)$ (3.15)	The criterion $Ra_{eq} \leq 370\,\frac{Bq}{Kg}$ should be met, to limit the external dose rate $D \leq 1.5\ mG/y$	Ellison & Williams, 2003
External Hazard Index (H_{ex}) $H_{ex} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right)$ (3.16)	*Criterion* $H_{ex} \leq 1$, $Ra_{eq} \leq 370$ Bq kg^{-1} For D ≤ 1.5 mGy y^{-1}	Ellison & Williams, 2003
Internal Hazard Index (H_{in}) $H_{in} = \left(\frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right)$ (3.17)	H_{in} must be less than unity for the radiation hazard to be negligible and $Ra_{eq} \leq 370$ Bq kg^{-1} D ≤ 1.5 mGy y^{-1}	Ellison & Williams, 2003
Gamma Index (I_γ) and representative level index (I_r) $I_\gamma = \left(\frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000}\right)$ (3.18) and	For bulk material if $I_\gamma \leq 0.5$ ⇒ D ≤ 0.3 mSv y^{-1} and if $I_\gamma \leq 1.0$ ⇒ D ≤ 1.0 mSv y^{-1} For restricted material (Tiles etc.) If $I_\gamma \leq 2$, ⇒ D ≤ 0.3 mSv y^{-1} and if $I_\gamma \leq 6$, ⇒ D ≤ 1.0 mSv y^{-1} $I_r \leq 1$	Ben Fredj et al., 2005; Beretka & Mathew, 1985
$I_{(r)} = \left(\frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}\right)$ (3.19)		EC, 1999
Alpha Index (I_α) $I_a = \left(\frac{A_{Ra}}{200}\right)$ (3.20)	xemption level = 100 Bq kg^{-1} ecommended upper level =200Bqkg^{-1}	Beretka & Mathew, 1985; Righi & Bruzzi, 2006

A_K, A_{Ra} and A_{Th} represent the activity concentrations respectively of ^{40}K, ^{226}Ra and ^{232}Th in Bq kg^{-1}.

Table 3.4. Mathematical expressions for dose rate, annual effective dose, outdoor and indoor annual effective doses

Hazard Index / Formula	Remarks/ Characteristics	Reference
Absorbed gamma dose rate in air (nGy/h) $D = (0.462 \times A_{Ra}) + (0.604 \times A_{Th}) + (0.0417 \times A_K)$ (3.21)	While, estimating D it is assumed that all decay products of ^{226}Ra and ^{232}Th are in radioactive equilibrium with their precursors.	NEA, 1979
Annual effective dose eq. E ($mSvy^{-1}$) $E = 0 \times t \times O \times D \times 10^{-6}$ For indoor public occupation, the indoor annual effective dose equivalent is: $E_{in} = 0.7 \times 8760 \times 0.8 \times D \times 10^{-6}$ (3.22) For outdoor public occupation, the outdoor annual effective dose equivalent is: $E_{out} = 0.7 \times 8760 \times 0.2 \times D \times 10^{-6}$ (3.23)	"t" is the time in hours for one year, i.e. 8760 hours, "D" is the dose rate in $nGyh^{-1}$ and "O" is the occupancy factor for outdoor (0.2) and indoor (0.8) circumstances.	Xinwei et al., 2006
Excess Life -time Cancer Risk ($ELCR$) $ELCR = LD\|_{average} \times RF \times E$ (3.24)	Whereas, E denotes annual effective dose equivalent, $LD\|_{average}$ = 66 years, shows the average life time and RF denotes lethal cancer risk per Sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public	UNSCEAR, 2006

3.2.6. Results and Discussion

3.2.6.1. Analysis of soil samples for gross alpha and beta activities

Results for the activity contents of gross alpha and gross beta estimated for twenty-five soil samples can be seen from figure 3.11. The alpha activity varied from 77.31 ±9.95 to 440.08 ±16.48 Bq kg^{-1} with overall average value of 234.88 ±1.69 Bq kg^{-1} (see Figure 3.11 a, b). While beta activity varied from minimum detection limit (MDL) to 361.55 ±149.33 Bq kg^{-1}, with an average value, for all samples, 235.65 ±149.98 Bq kg^{-1} (see Figure 3.11a, 3.11b and Table 3.5).

The maximum alpha activity, i.e., 440.08 ±2.13 Bq kg^{-1} was found for soil sample collected from Hassan Gallian and lowest activity, 77.31 ±1.28 Bq kg^{-1} was observed for soil sample collected from Chehla campus of the University. Whilst for beta activity, highest concentration, i.e., 361.55 ±149.33 Bq kg^{-1} was found for soil samples collected from Shervaan site, and lowest value, below MDL, was found for soil samples taken from Centre plate site. Lowest, highest, average and other statistical parameters associated with gross alpha and gross beta measurements are listed in Table 3.5.

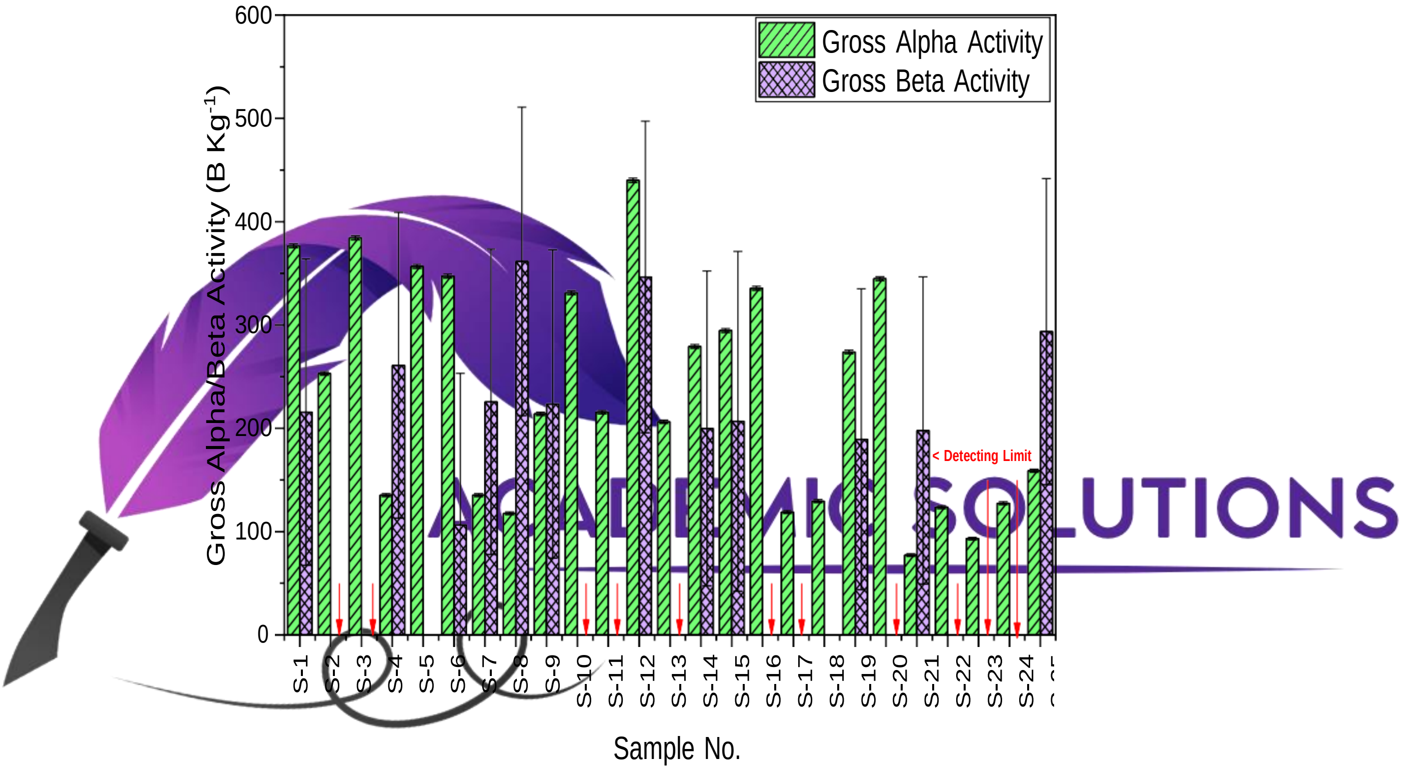

Figure 3.11. Gross alpha and gross beta activities of soil samples from Muzaffarabad

Table 3.5. Average, minimum, maximum and statistical properties of gross alpha and beta activities

Characteristics	Activity (Bq kg^{-1}) ± 2ó	
	Gross alpha	Gross beta
Mean	234.88 ±1.69	235.65±149.98
Minimum	77.31±1.28	<MDL
Maximum	440.08±2.13	361.55±108.50
Range	77.31-440.08	<MDL -361.55
Standard deviation	109.03	70.82
Skewness	0.19	0.36
Kurtosis	-1.35	0.36

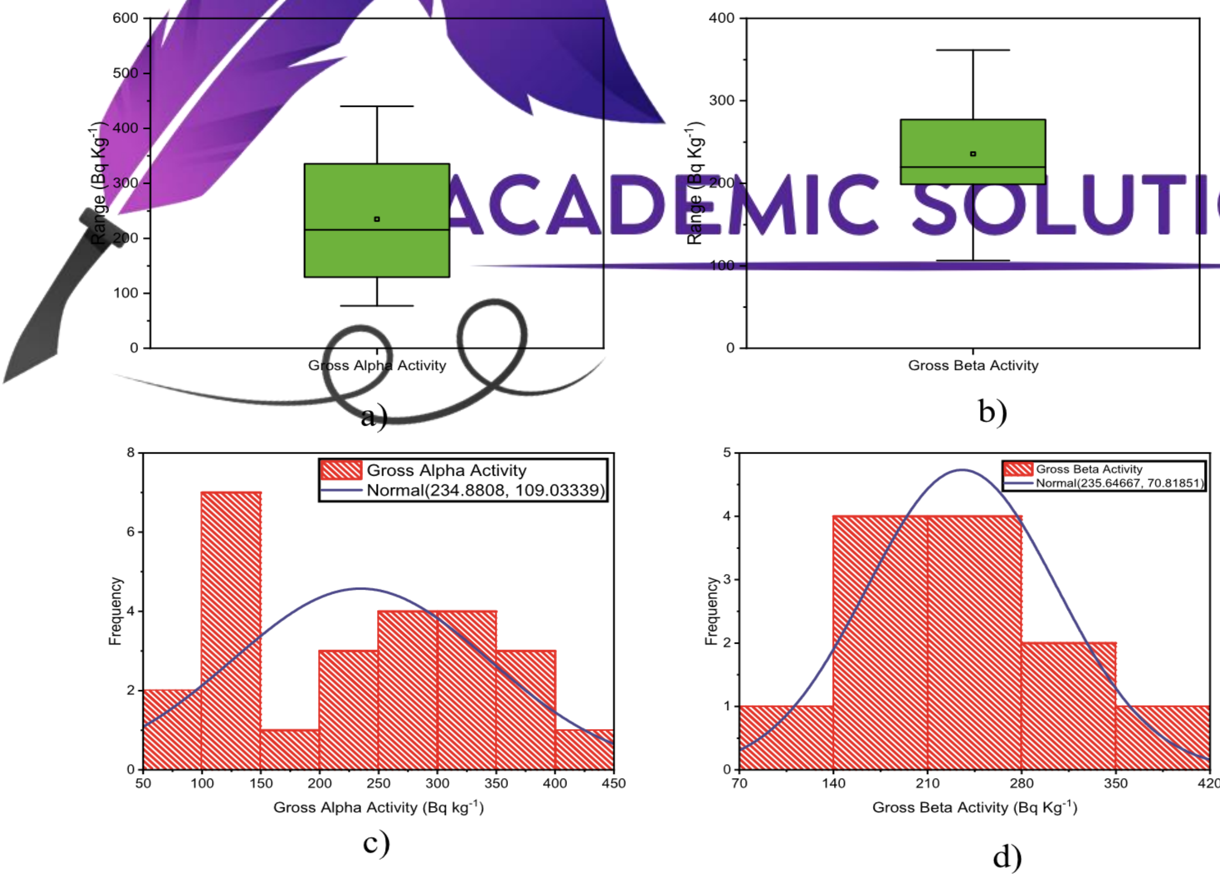

Figure 3.12. a) Boxplot of gross alpha activity, b) Boxplot of gross beta activity, c) Normal distribution fit on gross alpha activity, d) Normal distribution fit on gross beta activity

Figure 3.12. (a, b) shows Boxplot of gross alpha and Beta activities, while figure 3.12. (c, d) shows Normal distribution fit on gross alpha and gross Beta activity

The box plots have been drawn to compare the medians and variation of gross alpha and gross beta activities with respect to location of measurements.

Gaussian distribution fitting, both on gross alpha and gross beta activities, or slightly positively skewed towards right. For gross alpha activities, as can be seen from Table 3.5, the value of Kurtosis is negative (−1.35) showing that the distribution is platykurtic distribution with light tails. While for gross beta activities, the value of Kurtosis is positive (0.36) showing that the distribution is a leptokurtic with heavier tails. As from figure 3.12(d), it can be seen that distribution is sharply peaked with heavy tails and gross beta activities are closer to the mean value when compared with distribution drawn for the gross alpha activities. For gross alpha distribution, very few activity values are close to mean showing that the curve has flat peak and more dispersed activity values (McLeod, 2019).

It was observed that the variations in radioactive alpha and beta contents are also subject to variations in local geology and lithology. The occurrence of gross beta activity, in soil sample, may be attributed due to the presence of ^{40}K. At the same time other radionuclides, in soil samples, like ^{3}H and ^{90}Sr may also be responsible for beta activity besides ^{40}K presence.

3.2.6.2. Gamma activity using HPGe

As discussed earlier twenty-nine soil samples were investigated for gamma activity and gross alpha, beta activities. Results obtained for primordial and anthropogenic radionuclides in district Muzaffarabad and other districts of Azad Kashmir (Jhelum valley, Neelum valley, Rawalakot and Bagh) are presented in Tables 3.6 and 3.7 respectively. Gamma activity due to ^{40}K, ^{232}Th and ^{226}Ra was found in the range 213.54

±17.22 to 1205.83 ±12.82, 26.11 ±3.72 to 84.70 ±4.63 and 13.74 ±1.46 to 62.23 ±4.29 Bq kg^{-1} (see Figure 3.13). Whilst, activity concentration due to anthropogenic radionuclide ^{137}Cs was found in the range from minimum detection limit, i.e., ≤0.50 to 8.82 ±0.83 Bq kg^{-1}. Average values of all samples collected from district Muzaffarabad for ^{40}K, ^{232}Th and ^{226}Ra were found as 616.22 ±29.20, 55.83 ±5.74 and 37.91 ±2.35 Bq kg^{-1} respectively. Where, for ^{137}Cs the average value was found as 3.43 ±0.28 Bq kg^{-1}. As can be seen from Table 4.4 that for ^{40}K, current study reported value for range of measurements (213.54 ±17.22 to 1205.83 ±12.82 Bq kg^{-1}) and average value (616.22 ±29.20 Bq kg^{-1}) exceeds worldwide range (140–850 Bq kg^{-1}) and world average value (400 Bq kg^{-1}). The same trend can be seen for ^{232}Th. Minimum to maximum values, for current study, 26.11 ±3.72 to 84.70 ±4.63 Bq kg^{-1} and average value 55.83 ±5.74 Bq kg^{-1} exceeds worldwide range of 11–64 Bq kg^{-1} and world average value of 30 Bq kg^{-1}. For ^{226}Ra, the current study findings (13.74 ±1.46 to 62.23 ±4.29 Bq kg^{-1} for range and 37.91 ±2.35 Bq kg^{-1} average value) are comparable with the values reported for worldwide range 17–60 Bq kg^{-1} and world average value 35 Bq kg^{-1}.

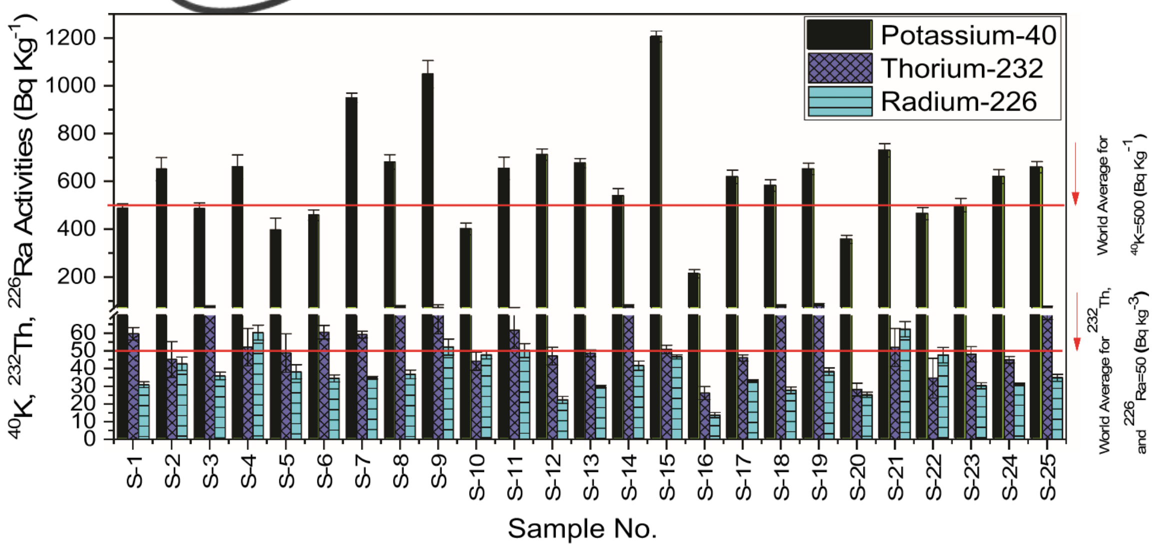

Figure 3.13. Gamma activities due to ^{40}K, ^{226}Ra, ^{232}Th in soil samples (District-MZD)

Table 3.6. Gamma (K-40, Ra-226, Th-232, and Cs-137) activity and statistical parameters for soil samples (District-Muzaffarabad)

Characteristic	Radioactivity (Bq kg^{-1}) ± 1ó			
	K-40	Th-232	Ra-226	Cs-137
Minimum	213.54±17.22	26.11±3.72	13.74±1.46	≤0.50
Maximum	1205.83±12.82	84.70±4.63	62.23±4.29	8.82±0.83
Average	616.22±29.20	55.83±5.74	37.91±2.35	3.43±0.28
Current-Study Range	213.54-1205.83	26.11-84.70	13.74-62.23	≤0.50-8.82±0.83
World-wide Range	140-850	11-64	17-60	----
World average	400	30	35	----
Standard deviation	214.90	16.02	11.40	2.44
Skewness	0.95	0.10	0.30	1.22
Kurtosis	1.74	-0.69	0.19	1.49

Current study results have also been compared with gamma activity concentrations of soil samples, collected from four other districts. For this purpose, single representative samples were collected from districts Bagh, Poonch, Neelum and Jhelum valley. The current study average value of ^{40}K (616.22 ±29.20 Bq kg^{-1}) was found greater than the values reported for Bagh (340.41 ±21.70 Bq kg^{-1}), Poonch (443.83 ±21.05 Bq kg^{-1}) and Jhelum Valley (449.52 ±20.29 Bq kg^{-1}) whilst smallest than the value reported for Neelum district (634.20 ±25.03 Bq kg^{-1}). For district Muzaffarabad the gamma activities due to ^{232}Th (55.83 ±5.74 Bq kg^{-1}) were greater than as reported for districts Bagh (54.93 ±4.81 Bq kg^{-1}) and Neelum (48.99 ±5.37 Bq kg^{-1}), whilst smaller than as reported for districts Poonch (61.66 ±4.68 Bq kg^{-1}) and Jhelum valley (95.27 ±4.72 Bq kg^{-1}).

In case of ^{226}Ra the average value reported for Muzaffarabad (37.91 ±2.35 Bq kg^{-1}) was greater than the value reported for Neelum district (26.80 ±2.12 Bq kg^{-1}) whilst less than the values reported for district Bagh (47.67 ±1.99 Bq kg^{-1}), Rawalakot (38.23 ±1.88 Bqkg^{-1}) and Jhelum valley district (39.85 ±1.82 Bq kg^{-1}). For anthropogenic radionuclide Cs-137, the average value reported for district Muzaffarabad (3.43 ±0.28 Bq kg^{-1}) was found greater than the values reported for district Poonch (1.46 ±0.11 Bq kg^{-1}), Neelum (2.49 ±0.16 Bq kg^{-1}) and Jhelum valley (≤0.50 Bq kg^{-1}) districts and less than the value reported for district Bagh (3.92 ±0.19 Bq kg^{-1}).

Table 3.7. Gamma (K-40, Ra-226, Th-232, Cs-137) activity in soil samples (District I-IV)

Sample code with Location	Activity (Bq kg^{-1}) ±1ó			
	K-40	Th-232	Ra-226	Cs-137
S-9(Sudhan Gali, Bagh)	340.41±21.70	54.93±4.81	47.67±1.99	3.92±0.19
S-33(Toli peer, RWT)	443.83±21.05	61.66±4.68	38.23±1.88	1.46±0.11
S-1(Nagdar, Neelum V)	634.20±25.03	48.99±5.37	26.80±2.12	2.49±0.16
S-25(Rashian, Jhelum V)	449.52±20.29	95.27±4.72	39.85±1.82	≤0.50

Minimum and maximum ^{226}Ra activity was estimated for Makri and Chehla University campus respectively (Figure 3.14a), whilst for ^{232}Th radionuclide, minimum and maximum activity were recorded for the soil samples collected from the locations of Makri and Sathra Upper Jalalabad. Minimum and maximum values of ^{40} K were recorded for the locations of Makri and Thallah Makri respectively (Figure 3.14(a, b, c)). Radioactivity content of anthropogenic radionuclide ^{137}Cs was found maximum at the location of Madina Market.

The radionuclides concentration in soil samples were found to follow the following pattern: ^{40}K > ^{232}Th > ^{226}Ra. The contribution from ^{40}K was found to be greater as compared to ^{232}Th and ^{226}Ra radionuclides. The reason for the higher values of ^{40}K activity concentration may be due to high potassium feldspar (K-feldspar) mineral contents in the soil samples (Ferdous et al., 2015). Since Potassium is an important nutrient applied as fertilizer for increasing crop amount and agricultural practices and live stock's system might be responsible for higher values of potash as compared to world average value. Current values of ^{40}K are influenced by water bodies' losses by runoff and leaching processes. So, it is possible that the same trend, of ^{40}K, may not exist for the longer period of time and get slightly changed subject to the impact of influencing parameters. There are some values of outliers in measured values of ^{40}K as may be seen from figure 3.14(c). Gaussian distribution fitting on radium, thorium and potassium has been shown in figure 3.14(d, e, f). The contents of anthropogenic radionuclide ^{137}Cs varied from below lower of detection (≤1.35) to 8.82 ±0.83 Bq kg^{-1}. It is formed in high fission yield and considered as highly mobile in some environments. Largest contents of caesium are distributed in soils due to nuclear reactor accidents like Fukushima Daiichi Nuclear Power Plant (NPP), Chernobyl nuclear reactor etc., or other origin may be nuclear testing and use of nuclear radionuclides for medical purposes. Radioactive due to Fukushima Daiichi Nuclear Power Plant (NPP) accident has been detected not only in soil of Japan (Yasunaria et al., 2011) but also, in many other countries (Kumamoto et al., 2017). ^{137}Cs activity concentration in soil of local origin might be due to one of reason mentioned above.

Table 3.8 shows the radium equivalent activity, absorbed dosage rate, annual effective dosages, indoor as well as outdoor radiation hazardous index and net (gamma) representative level index for soil samples collected from Muzaffarabad district. Minimum radium equivalent activity value (67.52 Bq kg^{-1}) was found at Makri and maximum (235.46 Bq kg^{-1}) at Lamiyan Pattiyan. The mean value of outdoor hazard index was found as 0.45 Bq kg^{-1} having minimum value (0.18 Bq kg^{-1}) at Makri and maximum 0.64 Bq kg^{-1} at Lamiyan Pattiyan while mean value of indoor hazard index was 0.55 Bq kg^{-1} having minimum value 0.22 Bq kg^{-1} at Makri and maximum 0.78 Bq kg^{-1} at Lamiyan Pattiyan. The smallest (30.98 nGyh^{-1}) and highest (111.17 nGyh^{-1}) values of absorbed dose rates have also been reported at Makri and Lamiyan Pattiyan respectively. The average values of indoor and outdoor annual effective dose rates were found as 0.38, 0.09 mSv y^{-1} respectively. Minimum values were recorded (0.15 and 0.04) mSv y^{-1}, at Makri and maximum (0.55 and 0.14) mSv y^{-1}, at Lamiyan Pattiyan. The maximum value of (gamma) representative level index (1.76 Bq kg^{-1}) was recorded for Lamiyan Pattiyan whilst minimum value (0.50 Bq kg^{-1}) is reported for the Makri site.

Average value of radium equivalent activity, Ra_{eq} (165.20 Bq kg^{-1}), estimated for the current study is well below the world average value 370 Bq kg^{-1}. Average values of Annual effective dose for indoor environment in current study (0.38 mSv y^{-1}) are smaller than world average (0.42 mSv y^{-1}), whilst annual effective dose for outdoor environment in current study (0.09 mSv y^{-1}) is comparable with the world average (0.08 mSv y^{-1}). Values for indoor, H_{in} (0.55) and out hazard index, H_{out} (0.45) are within the range of unity. Mean value of (gamma) representative level index for current study soil samples (I_r = 1.22 Bq kg^{-1}) is greater than permitted safe value of unity. Samples collected from Bela Makri, Sethi Bagh and Makri have permissible levels of (gamma) representative level index as compared to samples collected other site locations. Likewise, mean value of gamma index level (I_Υ= 0.61) for current study soil samples is found equal to safe value of unity. The same trend can be seen in other districts representative samples (see Table 3.9).

Table 3.8. Radium equivalent activity, absorbed dose rates, Annual effective dose as well as hazardous indices associated to observed soil samples of Azad Kashmir district Muzaffarabad

Characteristics	Ra_{eq} (Bq kg^{-1})	D (nGyh^{-1})	E_{in} (mSvy^{-1})	E_{out} (mSvy^{-1})	H_{in} (Bqkg^{-1})	H_{Ex} (Bqkg^{-1})	I_r (Bqkg^{-1})	I_Υ (Bqkg^{-1})
Minimum	67.52	30.98	0.15	0.04	0.22	0.18	0.50	0.24
Maximum	235.46	111.17	0.55	0.14	0.78	0.64	1.76	1.03
Mean	165.20	76.89	0.38	0.09	0.55	0.45	1.22	0.61
World average	370	55	0.42	0.08	≤ 1	≤ 1	≤ 1	≤ 1

Table 3.9. Doses and hazards associated with soil (District I-IV)

Sample code	Ra_{eq} (Bq kg^{-1})	D (nGyh^{-1})	E_{in} (mSvy^{-1})	E_{out} (mSvy^{-1})	H_{in} (Bqkg^{-1})	H_{Ex} (Bqkg^{-1})	I_r (Bqkg^{-1})	I_Υ (Bqkg^{-1})
S-1(Nagdar)	145.69	68.41	0.34	0.08	0.47	0.39	1.09	0.54
S-9(Bagh)	152.43	69.28	0.34	0.08	0.54	0.41	1.09	0.54
S-25(Reashian)	210.71	94.46	0.46	0.12	0.68	0.57	1.52	0.75
S-33(Toli peer)	160.58	73.30	0.36	0.09	0.54	0.43	1.17	0.58

Excess lifetime cancer risk (ELCR) for indoor occupation varied from 4.94×10^{-4} to 1.82×10^{-3} and for outdoor occupation 1.32×10^{-4} to 4.62×10^{-4}. Overall excess lifetime cancer risk (ELCR) for the current study was estimated using equation (3.15) and its value was found as 1.55×10^{-3}.

Activity concentration of ^{40}K and ^{137}Cs (as may be seen from Table 3.7) have positive skewness and the curve is well skewed towards right and right tail is longer with most of distribution is at left, whilst ^{232}Th is slightly and ^{226}Ra is moderately skewed towards right.

For, ^{232}Th activities, as can be seen from Table 3.7, the value of Kurtosis is negative (−0.69) showing that the distribution is platykurtic distribution with light tails. While for ^{40}K, ^{226}Ra and ^{137}Cs activities, the value of Kurtosis is 1.74, 0.19 and 1.49 respectively showing that the distribution, for these radionuclides, is a leptokurtic with heavier tails. As from Figure 3.14.(e, f,) it can be seen that distribution is sharply peaked with heavy tails and ^{40}K, ^{226}Ra activities are closer to the mean value when compared with distribution drawn for the ^{232}Th activities. For ^{232}Th activities distribution, very few activity values are close to mean showing that the curve has flat peak and more dispersed activity values (McLeod, 2019).

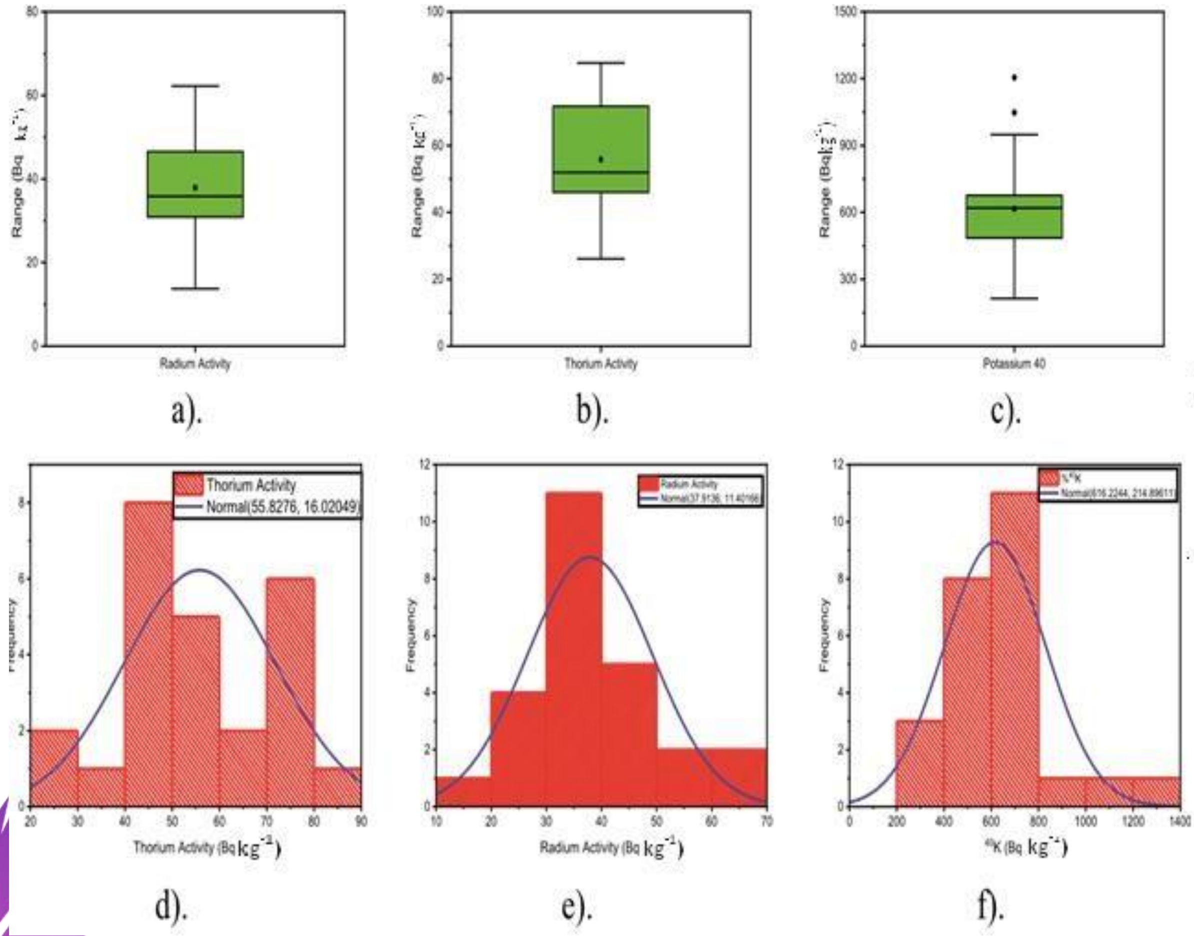

Figure 3.14. Box plot and normal distribution of Radium (a,d), Box plot and normal distribution of Thorium (b,e), box plot and normal distribution of Potassium (c,f)

3.2.7. Comparison of Results with Data Available In Literature

Results obtained in present study for primordial radionuclides have been compared with data available in literature (see Figure 3.15). Mean value of ^{222}Ra activity (37.91 ±2.35 Bq kg^{-1}) obtained for present study is found relatively higher than the values reported at national and international level viz. Punjab Pakistan (35 Bq kg^{-1}) (Tahir et al., 2005), Bangladesh (33 Bq kg^{-1}) (Miah et al., 1998), Southern Punjab Pakistan (21.7 Bq kg^{-1}) (Fatima et al., 2008), Lahore Pakistan (25.8 Bq kg^{-1}) (Akhtar et al., 2005), China (32 Bq kg^{-1}) (Xinwei et al., 2006), Venezuela (27 Bq kg^{-1}) (LaBrecque, 1994), Syria (20 Bq kg^{-1}) (Xinwei et al., 2006), Taiwan (30 Bq kg^{-1}) (Lin et al., 1987), USA (35 Bq kg^{-1}) (Xinwei et al., 2006), Indian Punjab (33 Bq kg^{-1}) (Singh et al., 2005). Whilst, smaller than as reported for the world global average (50 Bq kg^{-1}) (Righi & Bruzzi, 2006), Turkey (79 Bq kg^{-1}) (Baykara & Doğru, 2009), S. Jordon (42.5 Bq kg^{-1}) (Al-Hamarneh & Awadallah, 2009), Malaysia (67 Bq kg^{-1}) (Xinwei et al., 2006), Zanjan-Iran (88.5 Bq kg^{-1}) (Saghatchi et al., 2010), Hong Kong (95 Bq kg^{-1}) (Xinwei et al., 2006) and Rechna Doab Pakistan (48.2 Bq kg^{-1}) (Jabbar et al., 2010).

Likewise, Th-232 mean value (55.83 ±5.75 Bq kg^{-1}) measured for the Azad Kashmir district Muzaffarabad is relatively higher than as reported for S. Jordon (26.7 Bq kg^{-1}) (Al-Hamarneh & Awadallah, 2009), USA (40 Bq kg^{-1}) (Xinwei et al., 2006), Taiwan (44 Bq kg^{-1}) (Lin et al., 1987), Syria (20 Bq kg^{-1}) (Xinwei et al., 2006), Venezuela (31 Bq kg^{-1}) (LaBrecque, 1994), China (41 Bq kg^{-1}) (Xinwei et al., 2006), Lahore Pakistan (49.2 Bq kg^{-1}) (Akhtar et al., 2005), S. Punjab Pakistan (31.1 Bq kg^{-1}) (Fatima et al., 2008), Bangladesh (16 Bq kg^{-1}) (Miah et al., 1998), Punjab Pakistan (41 Bq kg^{-1}) (Tahir et al., 2005) and global mean average (50 Bq kg^{-1}) (Xinwei et al., 2006). Whilst, lower than to the reported values for Rechna Doab Pakistan (61.4 Bq kg^{-1}) (Jabbar et al., 2010), Hong Kong (59 Bq kg^{-1}) (Xinwei et al., 2006), Turkey (62 Bq kg^{-1}) (Baykara & Doğru, 2009), Malaysia (82 Bq kg^{-1}) (Xinwei et al., 2006) and Indian Punjab (87.4 Bq kg^{-1}) (Singh et al., 2005).

Mean value of ^{40}K radioactivity (616.22 ±29.20 Bq kg^{-1}) for current study was found relatively higher than as reported for the Southern Punjab Pakistan (393 Bq kg^{-1}) (Fatima et al., 2008), China (440 Bq kg^{-1}) (Xinwei et al., 2006), Venezuela (357 Bq kg^{-1}) (LaBrecque, 1994), Indian Punjab (143 Bq kg^{-1}) (Singh et al., 2005), Syria (270 Bq kg^{-1}) (Xinwei et al., 2006), Taiwan (431 Bq kg^{-1}) (Lin et al., 1987), Zanjan Iran (497 Bq kg^{-1} (Saghatchi et al., 2010), Punjab Pakistan (615 Bq kg^{-1}) (Tahir et al., 2005), USA (370 Bq kg^{-1}) (Xinwei et al., 2006), Malaysia (310 Bq kg^{-1}) (Xinwei et al., 2006), Lahore Pakistan (562 Bq kg^{-1}) (Akhtar et al., 2005), Bangladesh (574 Bq kg^{-1}) (Miah etal., 1998), Hong

Kong (530 Bq kg^{-1}) (Xinwei et al., 2006), Turkey (574 Bq kg^{-1}) (Baykara & Doğru, 2009), S. Jordon (291 Bq kg^{-1}) (Al-Hamarneh & Awadallah et al., 2009) and global average radioactivity (500 Bq kg^{-1}) (Xinwei et al., 2006). Whilst, lower than as reported for Rechna Doab Pakistan (648.9 Bq kg^{-1}) (Jabbar et al., 2010).

^{137}Cs mean activity content (3.69 ±0.28 Bq kg^{-1}) measured for the current study are comparable to data reported for Punjab Pakistan (1.11–6.4 Bq kg^{-1}) (Tahir et al., 2006), Rechna Doab Pakistan (0–8.1 Bq kg^{-1}) (Jabbar et al., 2010), Eastern India (1.8–7.5 Bq kg^{-1}) (Chakrabarty et al., 2009), and lesser than reported for Cairo Egypt (1.6–19.1 Bq kg^{-1}) (Higgy & Pimpl, 1998), Northern Taiwan (1.5–27 Bq kg^{-1}) (Wang et al., 1997), Majorca Spain (10–60 Bq kg^{-1}) (Gomez et al., 1997), Mont Coast Yugoslavia (1.5–28.4 Bq kg^{-1}) (Vuković et al., 1996), and Dhaka Bangladesh (3–10 Bq kg^{-1}) (Miah et al., 1998).

Range of ^{137}Cs activity (≤0.50 to 8.82 ±0.83 Bq kg^{-1}) and average value for all soil samples (3.69 ±0.28 Bq kg^{-1}) measured for district Muzaffarabad in present study are found greater than the range and average value estimated for another district Mirpur of Azad Kashmir (0.076 ±0.071 to 2.94 ±0.17 with mean value 1.39 ±0.17 Bq kg^{-1}). Mirpur study was conducted back in 2010 and published in 2011. One of reason of this change may be attributed due to radioactive release in environment due to possible reactor accident during the period.

Results obtained for activity contents of gross alpha (range 77.31–440.08 Bq kg^{-1}) and gross beta (range ≤30.00–361.55 Bq kg^{-1}), in the present study, have been compared with data available for different countries in literature. The range of gross alpha activity measured for current study is relatively higher than as reported for Power plant Gacko, Republic of Srpska (66.7–102.4 Bq kg^{-1}) (Sarap et al., 2012), Serbia (66.7–102.4 Bq kg^{-1}) (Vuković et al., 1996), Nigeria (8–40 Bq kg^{-1}) (Anekwe et al., 2013), Roper

Nuclear Power Plant area, Bangladesh (1.13–5.66 Bq kg^{-1}) (Ogundare and Adekoya, 2015), whilst smaller than Malaysia (15–9634 Bq kg^{-1}) (Lee et al., 2014), Turkey (206–837 Bq kg^{-1}) (Bal et al., 2012), Segamat District Malaysia (<MDA–4360 Bq kg^{-1}) (Rafique et al., 2013), Ipoh Malaysia (1555–5345 Bq kg^{-1}) (Kurnaz et al., 2020), Republic of Macedonia (221–1360 Bq kg^{-1}) (Dimovska, 2012), Bayelsa state, Nigeria (49–960 Bq kg^{-1}) (Meindinyo & Agbalagba, 2012), Dhaka Bangladesh (468–1710 Bq kg^{-1}) (Bose et al., 1993), and Van, Turkey (686–4713 Bq kg^{-1}) (Zorer et al., 2009), Rajasthan, India (175–2260 Bq kg^{-1}) (Shanthi et al., 2009).

For gross beta range (<MDL-361.55 Bq kg^{-1}) measurements, present study results were found greater than Roper Nuclear Power Plant area, Bangladesh (30.74–132 Bq kg^{-1}) (Ogundare & Adekoya, 2015), and Obrenovac, Serbia (93–262 Bq kg^{-1}) (Vuković et al., 1996), whilst smaller than as reported for Rajastan India (38,420–53,940 Bq kg^{-1}) (Shanthi et al., 2009), Power plant Gacko, Republic of Srpska (285.7–607.4 Bq kg^{-1}) (Sarap et al., 2012), Marmara Turkey (500–830 Bq kg^{-1}) (Yalcin & Gurler, 2007), Kinta District-Malaysia (142–6173 Bq kg^{-1}) (Lee et al., 2014), Eastern Anatolia Region-Turkey (111–423 Bq kg^{-1}) (Bal et al., 2012), Segamat District, Malaysia (70–4690 Bq kg^{-1}) (Rafique etal., 2013), Repository facility, Bukit Kledang, Ipoh, Malaysia (1645–5780 Bq kg^{-1}) (Kurnaz et al., 2020), Republic of Macedonia (438–1052 Bq kg^{-1}) (Dimovska, 2012), Bayelsa state, Nigeria (1520–119 910 Bq kg^{-1}) (Meindinyo & Agbalagba, 2012), Dhaka, Bangladesh (303–1125 Bq kg^{-1}) (Bose et al., 1993), and Van, Turkey (73–11,773 Bq kg^{-1}) (Zorer et al., 2009).

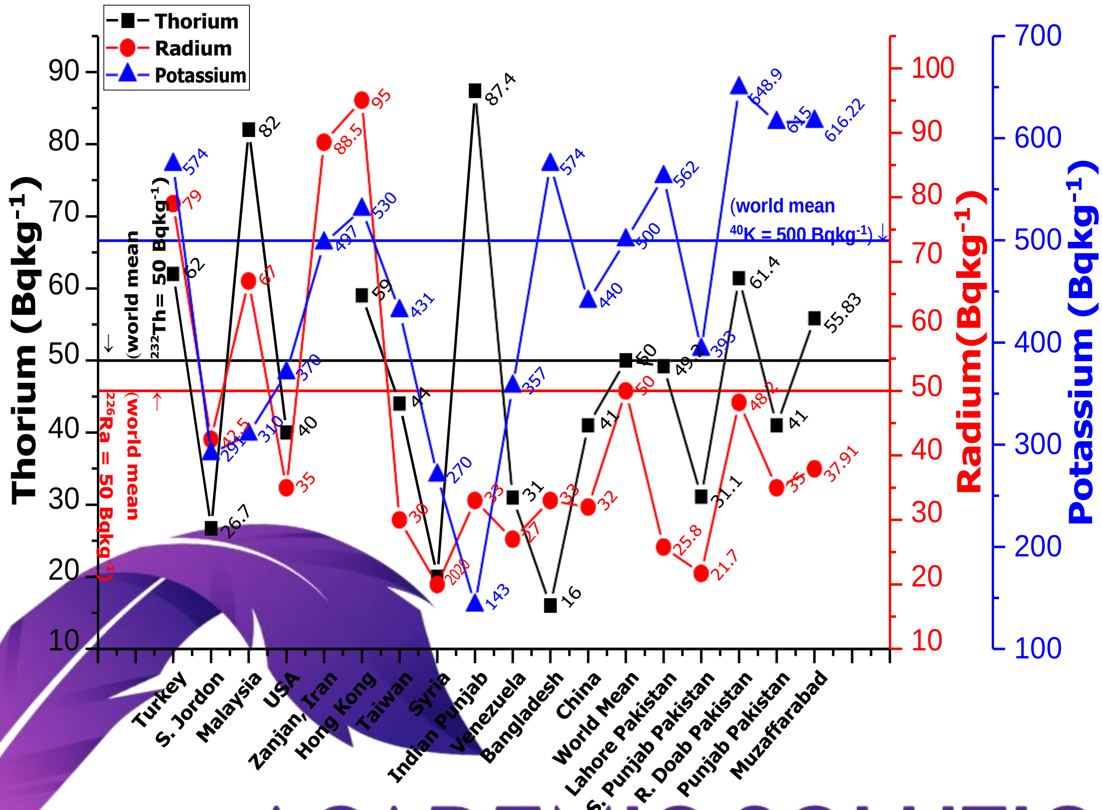

Figure 3.15. Comparison of current study result with data available in literature

Table 3.10. Comparison of current study data with several countries available data of gross alpha as well beta contents within soil samples

Country name	Gross alpha activity (Bq kg^{-1})	Gross beta activity (Bq kg^{-1})	References
Marmara, Turkey	-----	500 –830	Yalcin & Gurler, 2007
Power plant Gacko, Republic of Srpska	66.7 – 102.4	285.7 – 607.4	Sarap et al., 2012
Kinta District, Malaysia	Mean=1143, Range=15– 9634	Mean=1112, Range=142 – 6173	Lee et al., 2014
Nigeria	Range=152.11-322	Range= 311.15-615.5	Anekwe et al., 2013
Eastern Anatolia Region, Turkey	Range=206 – 837	Range=111 – 423	Bal et al., 2012
Roper Nuclear Power Plant area, Bangladesh	Mean=2.78, Range=1.13 – 5.66	Mean=71.85, Range=30.74 – 132	Ogundare & Adekoya, 2015
Segamat District, Malaysia	Mean=1020,Range=<MDL–4360	Mean=1071, Range= 70-4690	Rafique et al., 2013
Repository facility, Bukit Kledang, Ipoh, Malaysia	Mean=3747, Range= 1555 – 5345	Mean= 3493, Range=1645 – 5780	Kurnaz et al., 2020
Obrenovac, Serbia	-----	93–262	Vuković et al., 1996
Republic of Macedonia	Mean=522, Range=221–1360	Mean=681, Range=438 –1052	Dimovska et al., 2012
Bayelsa state, Nigeria	Mean=526, Range=49– 960	Mean=29.29, Range=1.520–119.91	Meindinyo & Agbalagba, 2012
Dhaka, Bangladesh	Mean=1020, Range=468–1710	Mean=635, Range=303 –1125	Bose et al., 1993
Van (Turkey)	Range=686-4713	Range=73-11773	Zorer et al., 2009
Rajastan India	Range=175-2260	Range=38420-53940	Shanthi et al., 2009
Azad Kashmir (Muzaffarabad)	Mean=234.88 ±1.69 Range=77.31-440.08	Mean=235.65±149.98 Range=<MDL-361.55	Present study

Currently, no regulatory limit for background radiations, including primordial radionuclides along with gross alpha and gross beta, has been proposed by Pakistan Nuclear Regulatory Authority (PNRA). However, the data presented here are first time attempt to characterize the study area for background radiations and it may serve as baseline data, in future, for any nuclear radiation emergency.

3.3. MEASUREMENT OF AGE DEPENDENT RADIATION INGESTION DOSES DUE TO GROSS ALPHA AND GROSS BETA EXPOSURE FROM MEDICINAL PLANTS

3.3.1 Abstract

This article presents the results of study conducted to measure the gross alpha, grass beta activities in medicinal plants samples collected from different districts of Azad Kashmir, Pakistan. The ASC-950-DP gasless high-speed counter was used for the measurement of gross α/β activities. Measured activities have been used to assess age dependent annual effective doses for infants, one year, five year, ten years, fifteen years, and adult peoples. Effect of altitude on measured values of gross α/β activities have also been investigated. For medicinal plants consumption rate (MPCR) of 1.8 kg y^{-1}, the average gross alpha and beta annual committed effective dose (ACED), delivered to infants, one, five, ten, 15 years and adults ranged from 43±7 to 1732±18 µSv y^{-1}, 7±1 to 274±3 µSv y^{-1}, 5±1 to 192±2 µSv y^{-1}, 5±1 to 181±2 µSv y^{-1}, 6±1 to 248±3 µSv y^{-1} and 3±0 to 100±1 µSv y^{-1} with mean value 797±10, 274±2, 88±1, 83±1, 114±1 and 46±1 µSv y^{-1}. For higher values of MPCR, viz. 2, 4, 6, 8 and 10 kg y^{-1} respective gross alpha and gross beta ACED goes on increasing. Finding of study shows that, except ACED delivered to infants for MPCR of 1.8 kg y^{-1} , all other estimated values, at same MPCR, fall below the WHO recommended level (290 µSv y^{-1}) and that of as reported in UNSCEAR, 2000 (0. 3 mSv y^{-1} or 300 µSv y^{-1}) report. In is concluded that, the radiological hazard related with consumption of the natural radionuclides in the medicinal plants is inconsequential. These results may serve as baseline data, for relevant agencies of the country, while establishing regulations, and guidelines concerning with the radiation protection due to the use of medicinal plants.

3.3.2. Introduction

Public exposure, from radiations originating from naturally occurring sources, is unavoidable. Cosmic radiations are believed to be the oldest source came in to being, about 13-14 billion years ago, with the creation of universe. Primordial radioactive elements are 2nd source of radiations and originated, about 4.5 billion years ago, with the birth of earth. Cosmogenic radioactivity is considered as third source of radiations whose production is ongoing process. Cosmogenic radionuclides are produced when cosmic radiations interact with atmospheric particles (Cember & Johnson, 2009). In addition to above said source peoples, from their birth, have radioactive potassium-40, carbon-14, lead-210, and other isotopes inside their bodies. The internal radioactive material contributes average annual dose to a person of about 40 millirems/year (US NRC Technical Training Center). Manmade sources including, nuclear medicine, nuclear reactors, atomic bomb testing are not only source of radiation exposure to public but are also responsible for occupational dose to individual workers during their duty timings. Number of radiation sources, discussed earlier, contribute towards radioactive contamination of soil, water, and food. Ubiquitous presence of trace amounts of uranium, thorium, and their progenies becomes source of radiation exposure in varying amount. Some of radioactive materials are ingested with water and food and some are inhaled e.g., radon. Food, grown from soil, contains radioactive material. Radionuclides are transferred from rocks and minerals, present in the soil, into the plants and crops. Radionuclides of thorium, uranium series and their progenies are present in the biotic system of plants, soil, animals, air, and water (Shanthi et al., 2009). Reactor accidents or from any other source of artificial radioactivity production, may contaminate the food by anthropogenic radionuclides, and thus contributing towards internal and external radiation doses. Radioactive materials are part of essential elements that serve as food and plants take it up from soil. In this way radioactive substance get into the plants and becomes part of the food chain. Plants are contaminated from radionuclides of uranium series through direct and indirect ways viz. directly from surface deposition, soil deposition, roots uptake and transference to bark, leaves, flower, seed, fruits, and berries (Desideri et al., 2010). Contamination level of these plants further increases with the use of fertilizers (Biswas et al., 2015). The geological and geographical location effects the amount of background radiation levels. Soils are usually having relatively higher values of activities due to ^{137}Cs, ^{134}Cs, ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{90}Sr. As reported by Hoshi et al., 1994 and Sahin et al., 2012, almost 100 % of ^{137}Cs activity is delivered to the interacting material. Many alpha emitters viz. ^{238}U, ^{226}Ra and ^{210}Po and beta emitter radionuclides viz. ^{40}K, ^{228}Ra and ^{210}Pb are dissolved in water from soil and up taken by plants from roots. Alpha emitter radionuclides are considered as potential source of internal radiation exposure. As reported in UNSCEAR, (1988) report that ^{210}Po contributes about 7 % of effective dose equivalent to man from all-natural ingested internal radiation sources (UNSCEAR, 1988; Bal, 2016). Keeping in view the importance of subject many researchers has measured natural radioactivity (Naryana & Prakash, 2007; Desideri et al., 2010; Sussa et al., 2013; Larbi et al., 2013; Shatha et al., 2015; Pourimani et al., 2015; Harb, 2015; Njinga et al., 2015;Najam et al., 2015; Espinosa et al., 2016; Kranrod et al., 2017; Gülçin & İlyas, 2019; Erkan Kiris 2020; Tomşe et al., 2020) and gross alpha and gross beta radioactivities in medicinal plant samples (Tettey-Larbi et al., 2013; Bal et al., 2016; Shahzadi et al., 2020a,b). Peoples use plants for multiple purposes, including its major use as herbal medicine, across the world. Medicinal plants, in Azad Kashmir, also serve as one of the sources for generating income for high altitude inhabitants. Azad Kashmir is rich in herbal plants which are being used as traditional medicinal ingredients. There are certain active chemical constituents in medicinal plants which produce a curing physiological therapeutic response against various ailments in human (Khan et al., 2012; Ahmed et al., 2017). Medicinal plants are the basic source of health care in the Pearl Valley District

Poonch and also for other districts of Azad Kashmir (Shaheen et al., 2017). For current study, five herb species (Bistorta amplexicule (roots, leaves), Bergenia ciliate (roots, leaves), Mentha longifollia, Nastrtion officinale, and Polugonum aviculare) from five Azad Kashmir districts were analyzed for gross alpha and gross beta activities. This study will help to assess the presence of radioactivity in medicinal plants to get estimate the radiation exposure to peoples resulting from the ingestions.

3.3.3. Sample Collection

To measure gross alpha and gross beta activities in medicinal plants (MPs), thirty-five samples were collected from five districts of Azad Jammu and Kashmir. Five different medicinal herb species viz. Bistorta Amplexicule, Bergenia Ciliate, Mentha Longifollia, Nastrition Officinal and Polygonum Aviculare were tested for activity concentrations. The pre-sampling as well as sampling strategies were made strictly in accordance with IAEA guidelines and protocols (IAEA, 1989: IAEA, 2004).

3.3.4. Pretreatment

All medicinal samples were brought at the Department of Physics and treated in Radiation Physics laboratory University of Azad Jammu & Kashmir. The MPs samples were collected and placed in plastic bags to prevent them from atmospheric humidity. Samples were washed by distilled water and left to dry at room temperature. MPs samples were then oven dried at 800 ºC for the period of 4 hours thereafter, grinded with mill. All the samples were sieved using stainless steel sieve and converted into the powder form. After drying the MPs samples, weighing approximately 0.5-1 g, were placed within planchet of stainless steel and minute amount of ethyl alcohol was employed to uniformly distribute the samples on planchet. Prior to house the samples within detector they were dried again by placing within the oven at 105 ° C for two hours period.

3.3.4.1. Gross alpha/beta counting system

The ASC-950-DP gasless high-speed counter was used for the measurement of gross α/β activities in MPs samples. It has 2 in. (5.1 cm) standard planchet diameter with 50 samples capacity. It is a lightweight gasless dual phosphor scintillator detector. Ability to export data directly to a computer using the PIC Link option makes it more versatile. With prior setting of guard and lead combination the detector can be optimized for lower and higher energy background betas measurements. Operating voltage of 705 V and 1260 V was correspondingly selected for alpha and beta mode. Calibration of the system was achieved by standard alpha and beta sources respective to Am-241 and Sr-90 on 15-16 October 2019 with activities 0.6938 and 0.4092 KBq (Shahzadi et al., 2020b). Sample's counts and geometry corrections were achieved by drawing selfabsorption curve separately for gross alpha and beta measurements. Samples were counted for time span of 3000 s for beta and alpha mode. Instrumental calibration parameters including attenuation term, background, efficiency as well as sample's parameter like sample mass, mass residue and counting time span provided the minimum detection limit (Zorer et al., 2009).

3.3.4.2. Gross alpha/beta activity

Gross alpha/beta activity concentrations were estimated in thirty-five MPs samples using Equation (3.25) (Kurnaz et al., 2019; L'Annunziata, 2003).

$$\text{Activity } (\alpha\beta) = N/(m \times \text{s} \times 60) \tag{3.25}$$

Where, gross alpha/beta activity is in Bq/kg, net count rate is represented by 'N', efficiency of counter by 'ε', 60 used as cpm to cps conversion term and 'm' represents mass of the sample in Kg or sample's volume in L.

3.3.4.3. Dose assessment

The average annual gross alpha or beta committed effective dose for a particular sample was determined by averaging the individual annual committed effective doses contributed by the major alpha or beta emitters in the ^{238}U and ^{232}Th series of the naturally occurring radionuclides as shown in equation (3.26) (Tettey-Larbi et al., 2013; Hao et al., 2017):

$$D_{avg}(\alpha/\beta) = \frac{IMP}{N_{R(\alpha/\beta)}} \sum_{i}^{R(\alpha/\beta)} A_{\alpha(\beta)} \times DCF_{ing(\alpha/\beta)} \qquad (3.26)$$

Where $D_{avg}(\alpha/\beta)$ is the average gross annual alpha or beta committed effective dose in the medicinal plant sample, $A_{\alpha/\beta}$ is the gross alpha or beta activity concentration in the medicinal plant sample, IMP is the consumption rate for the intake of the medicinal plants, $N_R(\alpha/\beta)$ is the number of radionuclides considered as major alpha or major beta emitters in the ^{238}U and ^{232}Th series of the naturally occurring radionuclides and $DCF_{ing}(\alpha/\beta)$ is the dose convection factor for ingestion of the natural radionuclides for an adult taken from UNSCEAR, (2000) report. Since there was no prior information regarding exact value of MPs consumption rate so a reference intake consumption rate of 1.8 kg y^{-1} was considered as was previously taken by Tettey-Larbi et al. (2013).

3.3.5. Results and Discussion

Results of gross alpha and gross beta activities in medicinal plants are shown in Figure 3.16. Mean gross alpha activity (GAA) and gross beta activity (GBA), in all samples, were found as 172 and 337 Bq Kg^{-1} respectively (see Table 3.11). GAA ranged from minimum detection limit (MDL) of instrument i.e., ≤4.5 to 374.65±2.49 Bq Kg^{-1}. MDL for GAA was observed in Nastrition officinal (Leaves, shoots) sample collected from Nagdar situated in Neelum Valley district, whilst maximum value of GAA was observed in Bistorta amplexicule (Roots) samples collected from Pir Chinasi Muzaffarabad.

Similarly, GBA ranged from minimum detectable activity to 481.07±166.32 Bq Kg^{-1}. MDL for GBA was observed in samples taken from different locations including, Pir Chinasi Muzaffarabad (in Bistorta amplexicule in Roots), Toli Peer Rawalakot

(Bistorta amplexicule samples both in Roots and leaves, Bergenia ciliate sample only in Roots and Polygonum aviculare sample only in leaves), Sudhan Gali Bagh (Bistorta amplexicule in Roots, Bergenia ciliate in Roots), Reshian in district Jhelum valley (Bergenia ciliate in both leaves and Roots and Polygonum aviculare sample in leaves), and in Nagdar district Neelum (only in Mentha Longifollia sample of Leaves). Maximum value of GBA was found in Nastrition officinal (Leaves, shoots) samples collected from Reshian district Jhelum Valley.

Table 3.11 shows the descriptive statistics of gross alpha and gross beta activity concentrations in herbal plants. For GAA the range of measurement, for all samples, was found as 365 Bq Kg^{-1} and for GBA the range of measurement was found as 323 Bq Kg^{-1} (see Figure 3.17). Minimum GAA, following MDL value, was found as 9.4 Bq Kg^{-1} and maximum was found as 375 Bq Kg^{-1}. Similarly, minimum GBA, following immediately after MDL value, was found as 158 Bq Kg^{-1} and maximum as 481 Bq Kg^{-1} (see Figure 3.17).

Table 3.11 shows that the values of variance for GBA and GAA was found as 11396 and 10284 respectively. To measure symmetry, or the lack of symmetry, skewness of the data was calculated. Skewness for GBA and GAA were found as 0.153 and -0.374 respectively. For GAA, mean of the data (172) is greater than median (165) and mode, showing that the distribution is positively skewed. This shows that most of the outlier lies on the right side of the distribution. For GBA, mean of the data (337) is less than median (356) and mode showing that the distribution is negatively skewed. This shows that for the GBA data the distribution is negatively skewed, and outliers lies on left side of distribution.

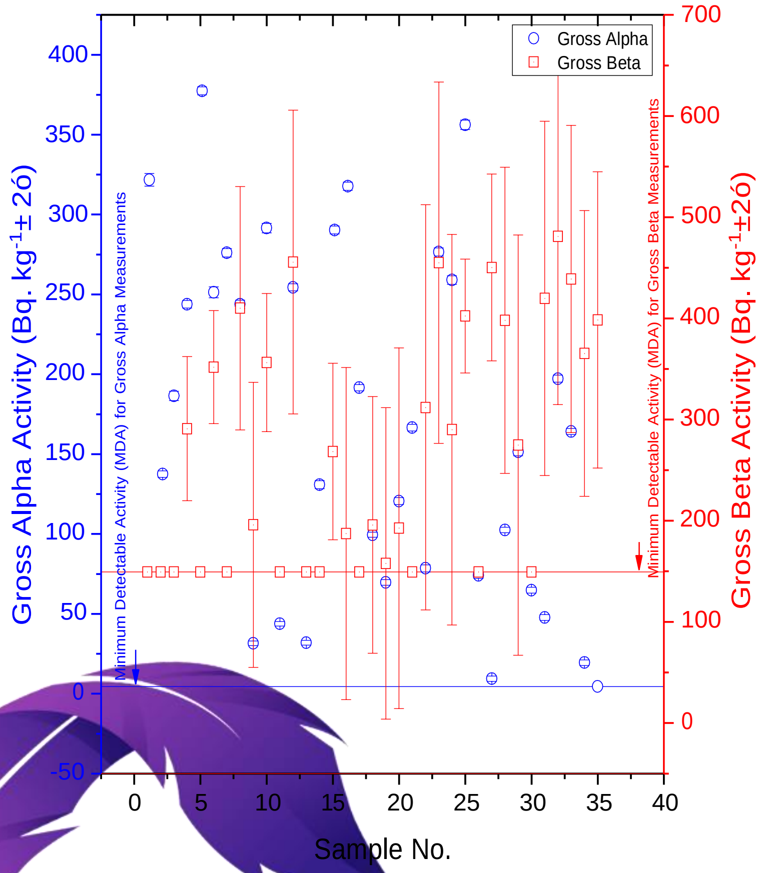

Figure 3.16. Gross alpha and gross beta activities in 35 samples of medicinal plants

Table 3.11. Descriptive statistics of gross alpha and gross beta activity concentrations in Herbal Plants

Variable	Gross Alpha Activity (Bq Kg^{-1})	Gross Beta Activity (Bq Kg^{-1})
No. of Observations	35	35
Mean	172	337
Median	165	356
Range (maximum-minimum)	365	323
SE. Mean	18	21
St. Dev	107	101
Variance	11396	10284
Skewness	0.153	-0.374
Kurtosis	-1.186	-1.185

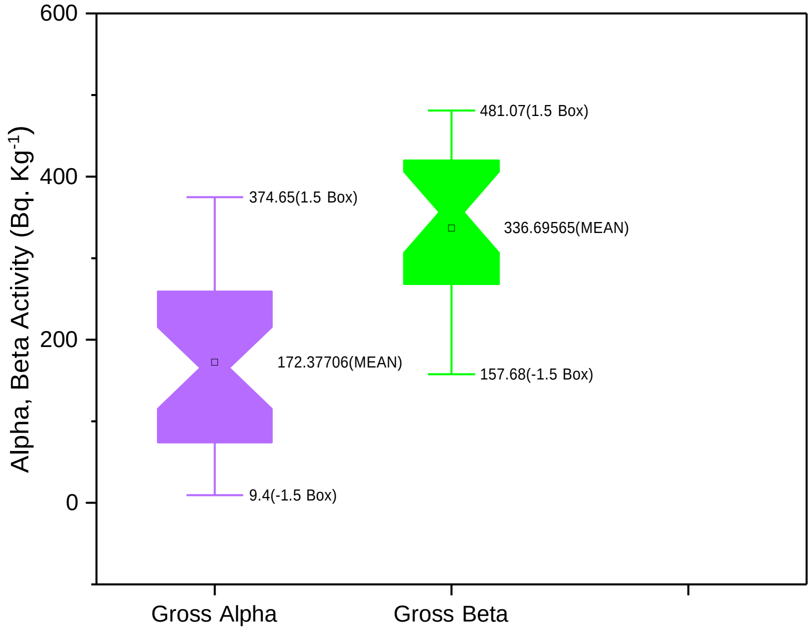

Figure 3.17. Mean, minimum and maximum values of gross alpha and gross beta activities

To find whether GAA and GBA data are heavily tailed or lightly tailed relative to normal distribution, kurtosis have been calculated. Kurtosis for GBA and GAA was found as -1.186 and -1.185. Both GAA and GBA have low values of kurtosis and resulting light tails with lack of outliers. Mean values of GAA and GBA in root samples of herbal plants follow the pattern as Bistorta amplexicule (238) > Bergenia ciliate (125) (see Figure 3.18). In case of root samples of Bistorta amplexicule, the mean value of GAA was found as 238, whilst for GBA as 196.

For leaves samples (see Figure 3.20), GAA follows following pattern in herbal plants; Bistorta amplexicule (232) > Polygonum aviculare (227) > Bergenia ciliate (184) > Nastrition officinal (107) > Mentha Longifollia (80). Similarly, GBA for leaves samples, follows following pattern in herbal plants; Nastrition officinal (421) > Mentha Longifollia (374) > Polygonum aviculare (365) > Bistorta amplexicule (351) > Bergenia ciliate (258).

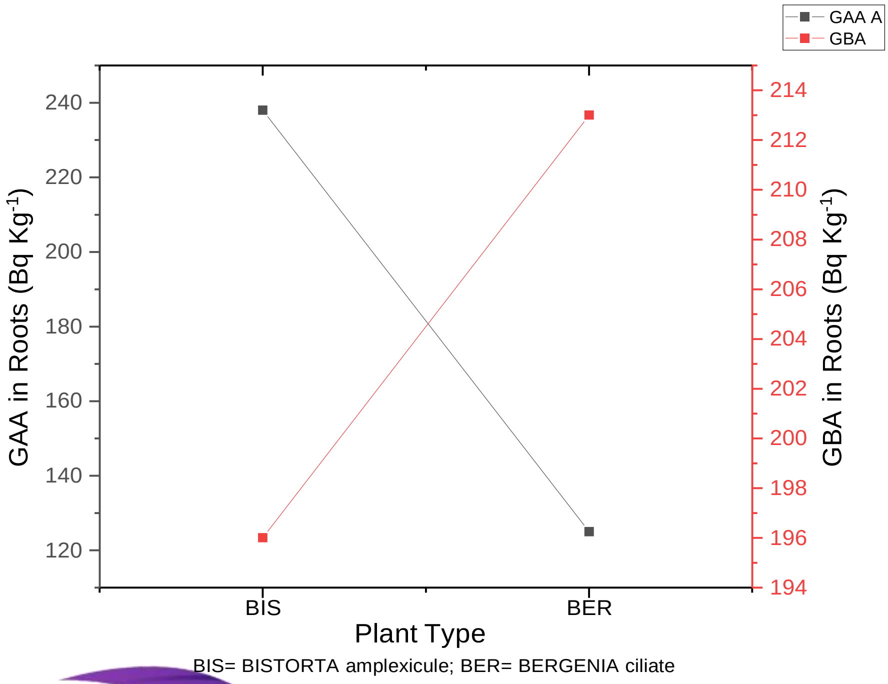

Figure 3.18.Average values of GAA and GBA in roots samples of Bistorta amplexicule and Bergenia ciliate MPs.

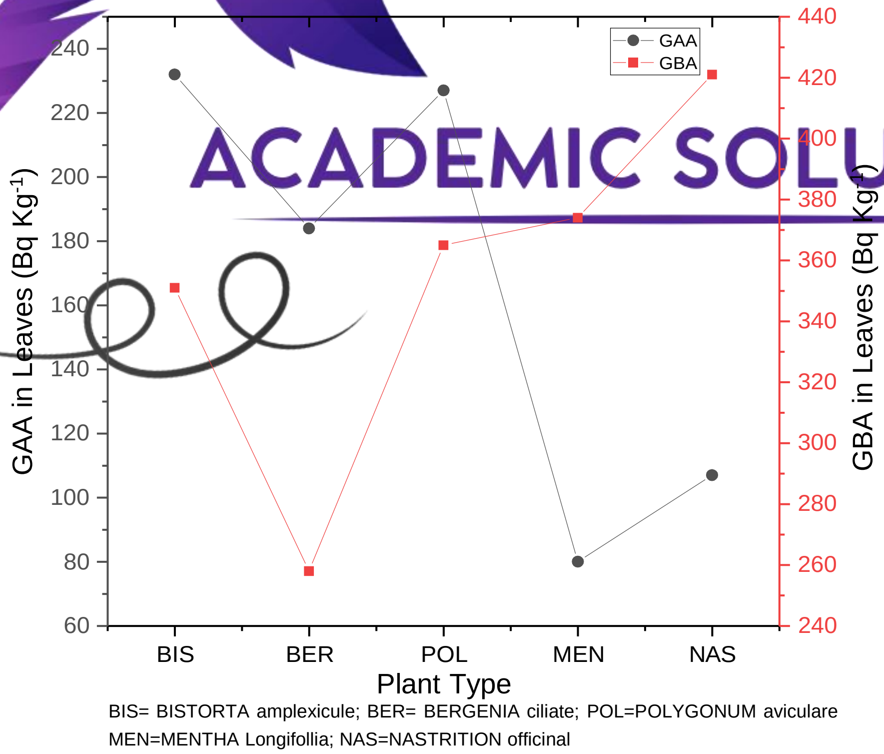

Figure 3.19. Average values of GAA, GBA in leaves samples of Bistorta amplexicule, Polygonum aviculare, Bergenia ciliate, Nastrition officinal and Mentha Longifollia MPs

3.3.5.1. Effect of altitude on gross alpha and gross beta activities

To show height and location of sample collection sites, surfer software has been used. Surfer software package is a full function 2D and 3D mapping, modeling, and analysis software. Figure 3.20 shows a 2D contour map representing height of medicinal plants sampling sites. Most of MPs samples were collected from the places situated above 5500 feet. About 85. 6 % of samples were collected from heights ranging from 5500 to 9193 feet. As may be seen from Figure 3.21 in region 1, only one sample have gross alpha activity value of 374.65±2.49 Bq Kg^{-1} and other values of gross alpha and beta activities are either low or below than BDL. Gross Beta activities in samples collected from sites with height greater than 9000 feet are found either lower than gross alpha activities or values less than BDL. Within height range of 5500 to 8000 feet usually gross beta activities were found higher than gross alpha activities.

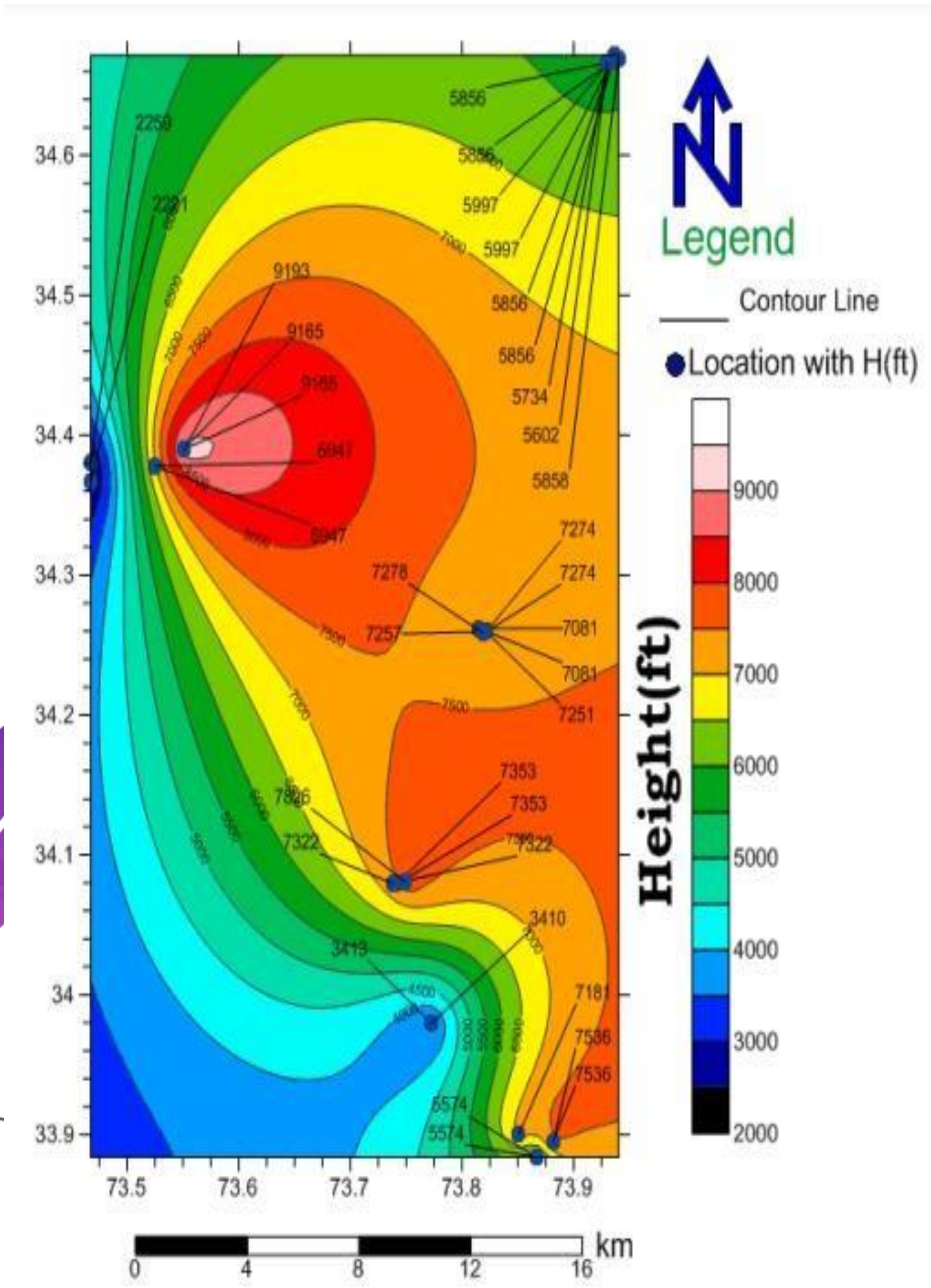

Figure 3.20. 2-D contour map representing height of the sampling sites

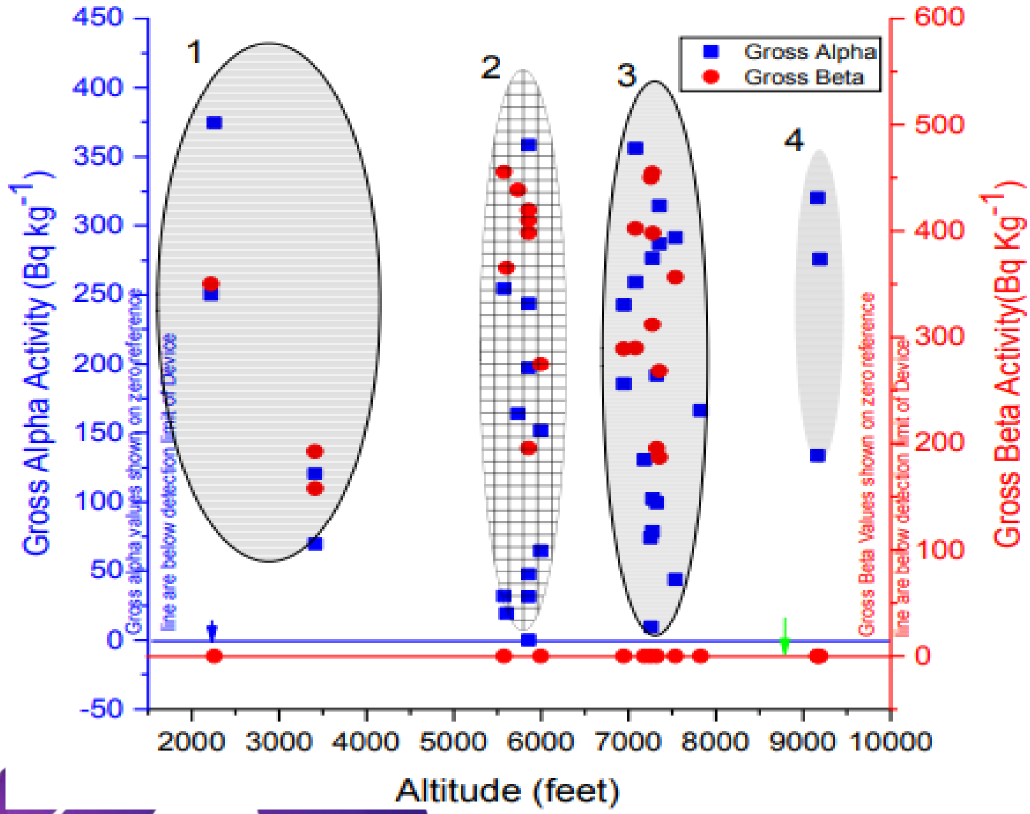

Figure 3.21. Variation of GAA and GBA with respect to altitude

3.3.5.2. Age dependent annual alpha and beta effective dose

The average gross alpha or beta annual committed effective dose (ACED) for a particular sample was determined by equation (3.26). ACED was calculated for different age groups including infants, one (01), five (05), ten (10). Fifteen (15) years and for adults. For medicinal plants consumption rate (MPCR) of 1.8 kg y^{-1}, the ACED, due to gross alpha activities, delivered to infants, one, five, ten, 15 years and adults ranges from 43±7 to 1732±18 µSv y^{-1} , 7±1 to 274±3 µSv y^{-1} , 5±1 to 192±2 µSv y^{-1} , 5±1 to 181±2 µSv y^{-1} , 6±1 to 248±3 µSv y^{-1} and 3±0 to 100±1 µSv y^{-1} with mean value 797±10, 274±2, 88±1, 83±1, 114±1 and 46±1 µSv y^{-1} . For other MPCR, viz. 2,4, 6, 8 and 10 kg y^{-1} respective gross alpha ACED goes on increasing as can been seen from Figures 3.22 and 3.23. Mean, minimum and maximum doses received due to GAA and GBA for different age groups are shown in Figure 3.24.

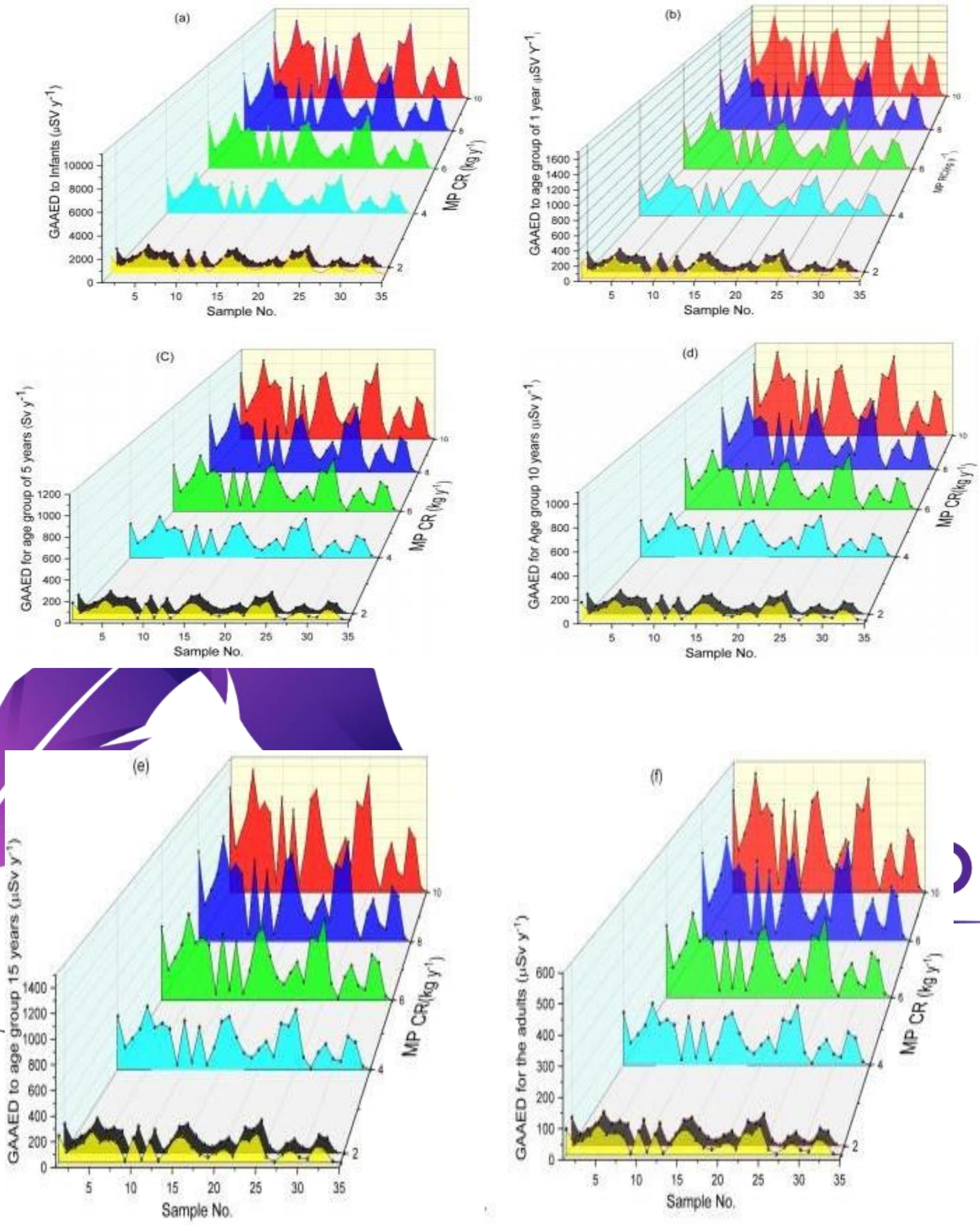

Figure 3.22. Gross Alpha Annual Effective Doses (GAAED) to a) infants; b) 1 year; c) 5 years; d) 10 years; e) 15 years and f) Adult population due to Medicinal Plants ingestion

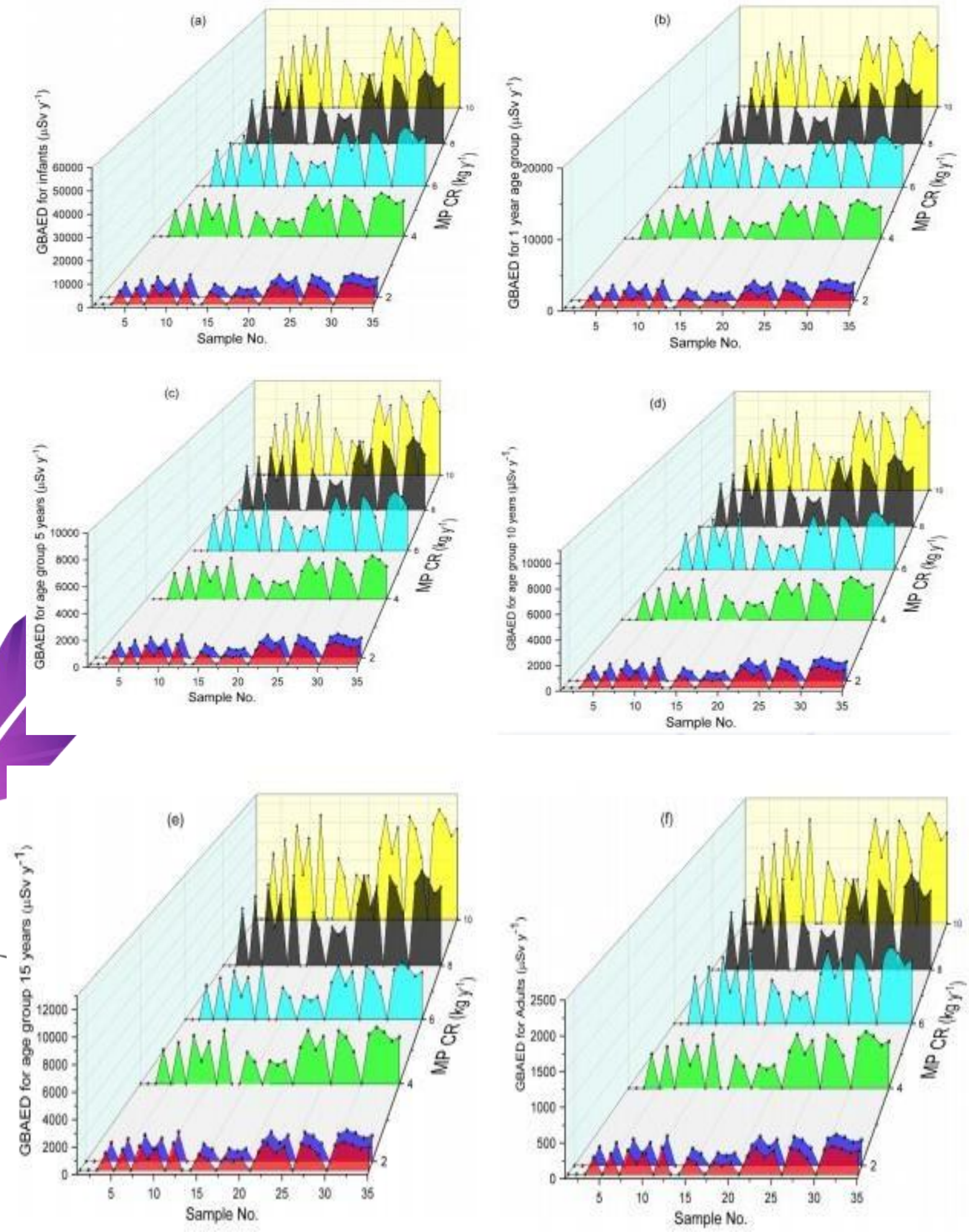

Figure 3.23. Gross Beta Annual Effective Doses (GBAED) to a) infants; b) 1 year; c) 5 years; d) 10 years; e) 15 years and f) Adult population due to Medicinal Plants ingestion

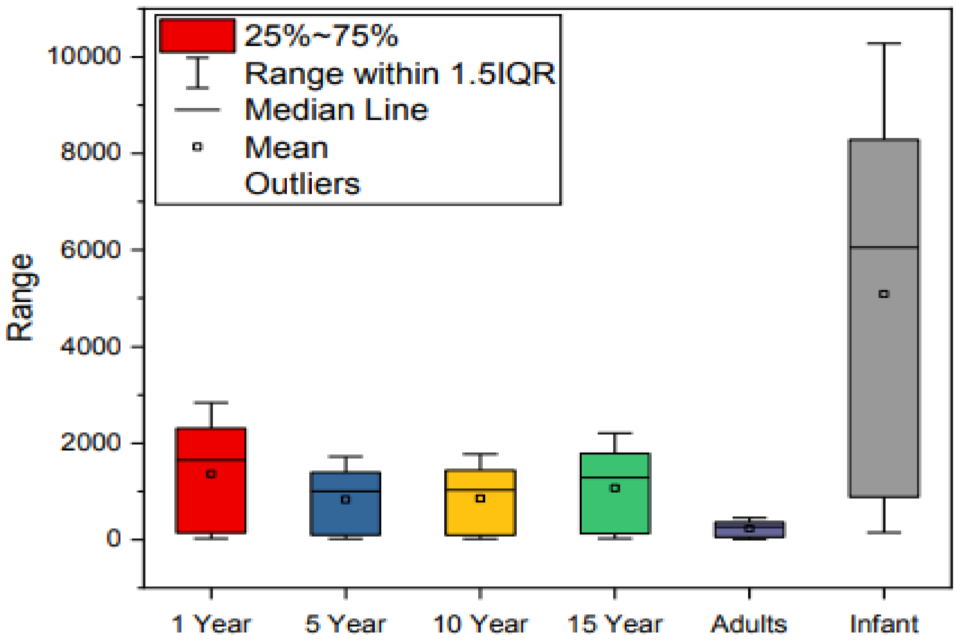

Figure 3.24. Mean, minimum and maximum doses (in µSv y^{-1}) for different age groups of peoples

3.3.6. Comparison of Results with Other Studies

Current study data obtained for gross alpha and beta activities have been compared with results found in literature for different countries as enlisted in Table 3.12.

Table 3.12. Comparison of current values of gross alpha and beta radioactivity with literature

Country name	Investigated Sample	Gross alpha activity range (Bq kg^{-1})	Gross beta activity range (Bq kg^{-1})	References
Bitlis, Turkey	Medicinal, aromatic plants	173±89- 989±224	347±241-3850±441	Bal et al., 2016
Ghana	Medicinal plants	11.73±0.61-132.67±7.22	124.34±11.28-790.58±13.19	Tettey-Larbi et al., 2013
Iraq	Nine medicinal plants	ND-0.4(cpm)	5.7-25.6(cpm)	Najam et al., 2015
Romania	Medicinal plants	3.20-10.75	214.5-429.40	Oprea et al., 2014
Azad Kashmir	Five medicinal herbs	≤MDA-374.65±2.49	≤MDA-481.07±166.32	Current study

For gross beta activities current study reported values ranged from (≤MDA481.07±166.32) Bq Kg^{-1}, which is greater than range of gross beta activities 214.5-429.40 Bq Kg^{-1} reported for Romania (Oprea et al., 2014), and less than the values ranging from (124.34±11.28- 790.58±13.19) Bq Kg^{-1} reported for Ghana (Tettey-Larbi et al., 2013) and the range of gross beta activities (347±241-3850±441) Bq Kg^{-1} reported for Bitlis Turkey (Bal et al., 2016)

For gross alpha activities current study reported values ranged from ≤MDA to 374.65±2.49 Bq Kg^{-1}, which is greater than range of gross alpha activities (11.73±0.61-132.67±7.22) Bq Kg^{-1} reported for Ghana (Tettey-Larbi et al., 2013), Romania (3.20-10.75) Bq Kg^{-1} (Oprea et al., 2014) and less than range of gross alpha activities (173±89-989±224) Bq Kg^{-1} reported for Bitlis Turkey (Bal et al., 2016).

3.4. RADIOLOGICAL RISK ASSESMENT OF NATURAL RADIOACTIVITY IN SOME SELECTED MEDICINAL PLANTS

3.4.1. Abstract

In this part of chapter, high resolution gamma spectrometery (using HPGe detector) have been used to determine the radionuclidic distribution within selected wild herbal species found in sloppy forests areas of Azad Kashmir. Contour maps have been drawn utilizing kriging interpolation method for activity distribution within medicinal herbs of sampled as well as unsampled areas. Results obtained showed that activity concentrations of ^{226}Ra, ^{40}K and ^{232}Th ranged from (≤MDA-8.67±1.08, ≤MDA-243.77±22.73, ≤MDA-7.45±0.76) Bq kg^{-1} with an average values 3.21±0.64, 112.75±14.11, 6.16±2.22 Bq kg^{-1} respectively. Effect of altitude on measured radionuclidic activities has been discussed. ^{226}Ra, ^{40}K and ^{232}Th transfer factors from soil constituent's to medicinal herbs (roots and leaves) were determined and found as 0.05, 0.21 and 0.03 respectively. Measured radionuclidic activities have also been employed to evaluate age dependent annual committed effective doses received by general public for

age ranges of <1year, 1 year, 5 year, 10 year, 15 year and >15 year and results were found consistent to UNSCEAR safe limit for ACED (300 µSv yr^{-1}). Study shows that investigated herbs are safe in terms of effective doses, cancer risks and radioactivity impacts. Suitability of medicinal herb against various ailments was also checked and Bistorta amplexicule (leaves) was found to be the most appropriate choice of potassium absorption. Comparisons were also made with the data available in literature for radioactivity distribution in medicinal plants/herbs and transfer factors from constituent's soil. Also, a trial was conducted to develop the national limits (intervention level) of selected medicinal herbs, which may help herbal medicine practitioner and pharmaceutical industries in diverse formulation as well as in an international trade.

3.4.2. Introduction

Ayureda is one amongst the vital therapeutic methods that has been practiced widely throughout the world. In order to combat with several diseases, these methods were developed by the numerous dynasties. It is an old-aged practice to employ medicinal herb for therapeutic purposes. We found diverse nature and treasure housed biodiversity of plants that are employed to treat several ailments of animals and humans. Several literatures about various medicinal properties of the herbs have been reported (Parchure, 1983). Preliminary plant's content was also evaluated by the various studies of different areas (Karunakara et al., 2003; Somashekarappa et al., 1996). Azad Kashmir is renowned land with herbs embedded forest that has been employed as folklore and traditional medicinal ingredients. Any comprehensive radiometric studies on such herbs are sparse for this geographical zone and elsewhere. Five herb species (Bistorta amplexicule (roots, leaves), Bergenia ciliate (roots, leaves), Mentha longifollia, Nastrition officinale, and Polygonum aviculare) from five Azad Kashmir districts were collected and analyzed for the purpose of their radiological consequences.

Bistorta amplexicule and Bergenia ciliate are among profound populous wild herbal species that is confined on Azad Kashmir mountainy forest that employed against various ailments. Durable relation seems to be developed for native herbs by the peoples of adjoining locality and clear rich culture observed in utilizing herbs for personnel requirements in one way or the other. Bergenia ciliate is used to cope with cough, fever, pulmonary affections, vomiting, wounds healing and diarrhea (Bhakuni et al., 1947). Genus Bergenia comprises of two specie namely B. strachey and B. ciliate and is a perennial herb with creeping rhizomes grown closely to rocks with enlarged leafy portions. Researches indicated that decoction of Bergenia ciliate roots with table salt employed to cope asthma in North west as well as trans Himalyan in Pakistan, Jummu and Kashmir. Urinary or oral calcii treated with extracted hot aqueous of whole plant B. ciliate, while roots employed as anti-scorbutic and tonic (Islam et al., 2002). Herbal B. ciliata rhizomes reported to have antioxidant, antibacterial, anti -inflammatory and anti urolithiatic properties (Anonymous, 1988; Kirtikar & Basu, 2006; Warrier et al., 1994; Sharma et al., 2006; Garimella, 2001; Sinha, 2001).

Mentha longifollia locally called as safaid podina which is well known herbal specie found to exhibit in Egypt, Arab countries, Sudan and Africa. Its leaves are popularly employed for tea flavoring, but herbalists practiced the whole herb as carminative (Banthorpe et al., 1981).

Bistorta amplexicule is a family member of polygonaceae, Masloon and Rain are its local common names in Azad Kashmir and Pakistan (Saqib & Sultan, 2005). While in India it is known to be Sarbguni (Sharma et al., 2004). It is characterized via dull pinkish or reddish inflorescence (Akeroyd, 2013), having erect, simple stem with fewer leaves, perennial, 50 to 70 cm tall, glabrous rhizomatous herb. June to September is its flowering time span in Pakistan and Azad Kashmir (e flora of Pakistan). In Pakistan, its leaves and rhizomes respectively serve as summer vegetable and medicinal product, while its roots were employed in cooking tea, which effectively cure the fever, flu and joints panic (Qureshi et al., 2007). Wound and sores were healed by applying its rhizomes and Bistorta amplexicule roots employed for tonic, cough and dysentery and also its rootstalk comprises of Anjubar drug with tannins that effectively used by the Ayurvedic and Unani medicinal practitioner. Its leaves and roots have high medicinal properties and employed typically in Swat Pakistan for the medication of ulcer, backache, rehematic pain, eyesight and gout (Hamayun et al., 2006) and has also a blood purifier status within Pakistan (North west) (Adnan & Holscher, 2010). Leafy paste of Bistorta amplexicule relieves dysentery and heals wound (Bhat, 2013).

Polygonum aviculare(knotweed) entirely appear as self-fertilizing herbal plant having flower size ranges 2 mm to 4 mm while anther dehisce in local proximity for stigma earlier to anthesis (Geber, 1990). Polygonum aviculare complex member found to colonize extensively at open areas and man-made homes or habitats with several disturbing types and regimes as well (Grime et al., 1988; Meerts, 1995). Trampled regime like cart tracks and walking or foot paths explored to have such genotype of little leafy types and seeds (Lousley & Kent, 1981; Grime et al., 1988; Meerts, 1995). Relatively larger leafy herbs tend to occur at two different untrampled habitations, short lived genotype having greater biomass distribution to seed at a regime with recurrent weeding actions and long-lived genotype with greater biomass distribution to vegetative portions tend to exhibit at stable region enclosed via denser plants (Meerts, 1992). Polygonum aviculare (knotweed) is one amongst the polygonaceae complex members which has astringent, antihypertensive, insecticide and diuretic features and used traditionally since over (Yin et al., 2005; Robu et al., 2008). Other aliments like dysentery, hemorrhage, arthiritis, hemoptysis, gout, diarrhea and hemorrhoids has been treated by this herbal plant (Robu et al., 2008). Polygonum aviculare exhibits high chemical contents flavonoids and phenolics that lead manifold biological impacts as DNA defensive activities, antitumour and antioxidant activities having capability to forage free radicals (Habibi et al., 2011). Flowering organs, leaves and woody portions including bark and stem of this herb that likely comprises of phenolic acid, lignans, tannins, stilbenes and flavonoids (Larson, 1988). Reports suggested that this herb has a preventive role against aging interrelated ailments, heart diseases and cancer growth (Hsu, 2006).

Several researchers have been focusing on herbal drugs to substitute with conventional medications which applied to cope with cancer types and numerous other ailments (Vickers, 2002). In Irani province, some therapists traditionally employed stewed knotweed extracts to cope cancer at Azarbaijan and good outcomes has reported (Habibi et al., 2011). However, radiological assessment within knotweed (Polygonum aviculare) has not ever been reported.

As plants extracts or raw parts are considered to be imperative source of safe and secure medication around the globe. Around 25 % modern medicine derived from medicinal herbs, mainly from flowering portions and more than 250,000 flowering herb species was estimated which being served as primary resources of new drug products and traditional orthodox (Gurib-Fakim & Kasilo, 2010).

World health organization explain traditional medication used for therapeutic purpose which has been practiced, since the emergence of human's immemorial time, and are still in practices (WHO, 1991). In fact, existed radionuclides migrated to human thru several alleys like drinking or food chain. Plants were contaminated via numerous contributed pathways such as directly surface deposition, soil deposition, also roots uptake and transference to bark, leaves, flower, seed, fruits and berries (Desideri et al., 2010).

Radioactivity level founds within the environment in dynamic pattern. Radionuclides revolves within troposphere and earth planet as crucial ingredient of soil, air, vegetation, water and rocks that might be subjected to ecosphere and eventually deposited upon the food shackle, which is one of the vital parameters that enhance the radiation doses to the common public, and it is needed to assess the radionuclidic burden on environmental materials and accompanied radiation hazards as well.

Radiation hazards to the environment and individuals largely relied on the contamination level, contaminants specific nature as well as spread range. As there is not any such radiation sensory way that can estimate the existence and extent of radiation. In current study medicinal herbs of Azad Kashmir districts have been scrutinized for radiation dose and dose consumption rate. Radioactivity transfer factor (T.F) from soils to medicinal herbs was also evaluated. Internal radiation dose arises through radioactivity intakes as consumption of medicinal herbs and food by the local community of the studied areas was also estimated. In this perceptive, peoples could be aware of most prospective radiation hazards, through suitable and proper knowledge of it.

3.4.3. Sample Collection

To explore radionuclidic distribution within medicinal herbs, five herbal species including Bergenia Ciliate, Polygonum Aviculare, Mentha Longifollia, Bistorta Amplexicule and Nastrurtium Officinale; collected from five districts of Azad Kashmir namely Neelum Valley (Nagdar), Muzaffarabad (Plate and Pir chinasi), Hattian Bala (Reshian), Bagh (Sudhan Gali) and Poonch (Ali sojal and Toli peer). Total of thirty-five samples were investigated for gamma spectroscopy. Pre sampling and sampling strategies were carried in accordance to IAEA guide lines (IAEA, 1989).

3.4.3. Pretreatment

All herb samples were tested from Botanist for identification and declaration of the herbal species. In next step, herbal samples were thoroughly cleaned with distilled water and then dried and ashed to get homogenized form of samples in line with IAEA guide lines (IAEA, 1989; IAEA, 2004).

3.4.4.1. Gamma spectroscopy using High Purity Germanium detector (HPGe)

P-type coaxial tubular HPGe high resolution setup was used for evaluating radionuclidic contents prevailing within medicinal herbs samples obtained from various locations of five Azad Kashmir districts.

3.4.4.2. Radioactivity

The activity contents were determined for ^{238}U at relative photopeak energies (609.31KeV) of ^{214}Bi-214 and (351.92KeV) of ^{214}Pb, for ^{232}Th at mean energy (911.21 KeV) of ^{228}Ac and (238.63 KeV) of ^{212}Pb, and for ^{40}K-40 directly at 1460.83KeV energy. The following equation (3.27) was employed for assessing the activity content (Bq kg^{-1}) (Ebaid, 2010).

$$A = \frac{N}{P \times T \times m \times s} \quad (3.27)$$

Where, net detected counts of radionuclides within sample are represented via "N", detector's absolute efficiency by "ε", live time span via "T(s)", mass of the sample by "m (kg)" and "*P*" depicts the yield of gamma ray.

3.4.4.3. Average committed effective dose (ACED)

Tetty-larbi et al. (2013) used a formula for ACED calculation for ingested NORMs via medicinal herbs described as under;

$$E_{av} = A_i \times DCF_i \times I \quad (3.28)$$

Where, annual committed effective dose represented by E_{av}, 'I' denote annual intake and DCF_i is dose conversion factor for ith radionuclide. DCF_{Th} (mSv/Bq), DCF_K

(mSv/Bq) and DCF_{Ra} (mSv/Bq) be respective dose conversion factors for ^{232}Th, ^{40}K and ^{226}Ra for adult, children and infants group (UNSCEAR, 2000). 'I' denote the intake rate that assumed to be 1.8 Kg y^{-1}, as there is nowhere intake rate for medicinal herbs. We have used the assumption, earlier used by Tetty-larbi et al. (2013) (CSRPM, 2011), that any patient intakes 100 ml/day of herbal ingredients for treatment phase. Only 5 % herb/plant ingredients employed for medicinal products which is (2 tbs) average dose of full (30 ml) that will be taken thrice time daily similar to 90 ml/d (Appiah, 2012). This may lead 5 g of herbal ingredient employed to yield 100 ml.

There is nowhere any standard dosage for the practice of such medicinal herbs in Pakistan, but with increase of consumption rate by patients that continuously employed these herbs as remedies for numerous ailments would equivalently enhances via her/his annual committed effective dose. Threshold consumption rate for each medicinal herb samples were calculated by following formula:

$$C_r = \frac{3E_{av}}{\sum_{i=1}^{3} [Ai \times DCFi]} \qquad (3.29)$$

Where, E_{av} depicts threshold average effective dosage raised by ingesting NORMS of medicinal herbs as mentioned in UNCEAR publications (UNSEAR, 2000).

3.4.4.4. Transfer factor (T. F)

Radionuclides exhibited in soil transfer in to plant through roots; are indicated by the transfer factor (T. F) known as soil to plant transfer factor, which is computed via determining the radionuclidic activity level within plant as well as of soil constituent (IUR, 1994).

$$T.\,F = \frac{radionuclidic\ activity\ level\ in\ herb(dried\ weight)\ (\frac{Bg}{kg})}{radionuclidic\ activity\ level\ in\ dried\ soil\ (dried\ weight)\ (\frac{Bg}{kg})} \qquad (3.30)$$

3.4.5. Geo-spatial Analysis

Since present investigated samples, for natural primordial radionuclides are representative of a single sampling site and cannot be considered for the rest of unsampled sites of entire zone, so to cover the entire region in order to get information of unsampled places geo-statistical procedure have been used (Sarma, 2010). Kriging interpolation technique, developed by D. Krige (Krige, 1966), was employed to demonstrate the spatial dispersals of primordial radionuclides activities within herbs samples of investigated and non-investigated zones. The Kriging formula is as under (Dindaroğlu, 2014).

$$Z\,(S_o) = \sum_{i=1}^{N} \lambda_i Z\,(S_i) \qquad (3.31)$$

Where, unknown weight for observed data corresponding to ith spot depicted via 'λ_i', estimated data respective to ith region via $Z\,(S_i)$ and observed location was depicted by S_o.

Based upon the regional scale, 2D contour maps having chiefly spatial resolutions have been drawn for primordial radionuclide activities within herb samples.

3.4.6. Results and Discussion

As discussed earlier, thirty five smples belonging to five herb species were investigated for natural radionuclides presence. Results of radionuclidic concentrations of ^{40}K, ^{232}Th and ^{226}Ra activities in medicinal herbs samples are shown in figure 3.25. Mean activity concentrations of ^{40}K, ^{232}Th and ^{226}Ra in all samples were found as 112.75±14.11, 6.16±2.22 and 3.39±0.66 Bq kg^{-1} respectively (see Table 3.13). Radionuclide concentrations of ^{40}K ranged from minimum detectable activity (MDA) of measurements i.e, ≤1.74 to 243.77±22.73 Bq kg^{-1}. MDA for the radionuclide ^{40}K was found in Bergenia Ciliate (roots) sample collected from Pir Chinasi situated in

Muzaffarabad district, whereas, greatest value of radionuclide ^{40}K was found in Mentha Longifollia (leaves) samples collected from Reshian of Hattia Bala district.

Likewise, activity concentration of ^{232}Th radionuclide ranged from minimum detectable activity (MDA), i,e. ≤0.62 to 7.45±0.76 Bq kg^{-1}. ^{232}Th radionuclide concentration was found below detection limit for almost all samples except samples collected from Pir Chinasi district Muzaffarabad (in only leaves samples of Bistorta Amplexicule and Bergenia Ciliate), Toli Peer district Poonch in only in roots sample (Bistorta Amplexicule), Bagh (in Polygonum Aviculare (leaves) and Bistorta Amplexicule (leaves) of Sudhan Gali) and in Reshian district Hattian Bala (in leaves samples for both Bistorta Amplexicule, Mentha Longifollia and in roots (Bergenia Ciliate)). Highest value of radionuclide (^{232}Th) was noticed in Bistorta Amplexicule (leaves) samples taken from Pir Chinasi district Muzaffarabad.

Similarly, concentration of radium radionuclide (^{226}Ra) ranged from minimum detectable activity (MDA), i.e. ≤0.30 to 8.67±1.08 Bq kg^{-1}. MDA for radionuclide (^{226}Ra) was observed in samples taken from different locations viz. Pir Chinasi district Muzaffarabad (Bistorta Amplexicule in roots only and in Bergenia Ciliate both in roots and leaves), in leaves both for (Nastrition Officinal and Polygonum Aviculare), Toli peer district Poonch (in Bergenia Ciliate both in roots and leaves), Sudhan Gali district Bagh (only in Bergenia Ciliate samples of leaves) and in Nagdar district Neelum Valley (in Bistorta Amplexicule (roots, leaves), Bergenia Ciliate (roots) and in leaves samples (Polygonum Aviculare, Nastrition Officinal, Mentha Longifollia). Maximum value of radionuclide (^{226}Ra) was observed in Bistorta Amplexicule (root) samples of Reshian district Hattian Bala.

Anthropogenic radionuclide, ^{137}Cs, was detected (0.20±0.02) Bq kg^{-1} in Bistorta Amplexicule (leaves) samples for the sampling site Toli Peer of district Poonch.

Higher radioactivity level of ^{40}K was observed for almost all tested herbs except Bergenia ciliate (roots) sample collected from Pir Chinasi district Muzaffarabad for which activity concentration was below minimum detection limit. It can be seen from table 3.13 that medicinal herbs have higher values of ^{40}K contents but lowest for the radionuclide ^{226}Ra.. It was noticed that radionuclidic contents within medicinal herbs is greatly influenced by the type of radioisotope and by herbal specie as well. Some investigators reported about K-Cs ratio which implied that higher the potassium content then lower be the cesium observed (Marchner, 1995). Also, the enormous difference between ^{137}Cs and ^{40}K contents within the plants has been reported (Berden et al., 2003). However, it is commonly found high ^{137}Cs content in cultivated plants than the uncultivated.

However, ^{40}K activity within all investigated district's samples found of higher magnitude compared to other radionuclides (^{226}Ra and ^{232}Th) and further in roots its concentration found to be far smaller than leafy portions comparatively. Evidently higher ^{40}K content in herbal and non-herbal plants with somewhat similarity has been presented by various other investigators (Karunakara, 1997; Desideri, 2010; Chethan, 2012).

The observed variability in activity contents for medicinal herbs might be outcomes of different geological formations as well as radiochemical conformation of soil where herbal plant relatively grown wildly. Also, activity content of radionuclides describes variation with half-life, longer radionuclide then larger will be the documented activity. Radioactivity is not normalized world over and it may be associated with capability of some herbs to more absorb certain elements comparative to other (Miah et al., 1998). Several other wild vegetation types of the same forest area might be the answer to such different radioactive levels and site characteristics. Soil-water ratio, soil organic contents, rainfall rate, biochemical progression and soil drainage also contributed to radiation level (Abu-Khadra & Eissa, 2008). All tested herbs have higher potassium content comparative to other radionuclides which is not much astonishing as potassium non-radioactive contents within plants comparatively high therefore about half of internal radiation lead by organism through fertilizer and nutrients mostly derived by ^{40}K (UNSCEAR, 2000). Our work demonstrated that ^{226}Ra contents found smaller than ^{232}Th content which is much astonishing fact due to immobility of thorium comparative to radium. But generally, most of plants samples have low thorium content as presented (Vandenhove et al., 2009; Tomazini et al., 2009). However, our observed data found to be consistent with the data reported for plants activity by Kovacs et al., 2015; Matiullah et al., 2008, where, respectively ^{232}Th ranges (MDA-3.65 Bq kg^{-1}) and (2.37-7.20 Bq kg^{-1}) found greater to ^{226}Ra ranges (MDA-1.9 Bq kg^{-1}) and (1.35-1.71 Bq kg^{-1}). Current study results are in accordance with results available in literature that shows ^{226}Ra concentration within herbs, might be greatly influenced through various parameters including phosphate content, PH and Ca-Mg ion content exhibited in soil constituent(Soudek et al., 2007; Tykva & Podracka, 2005).

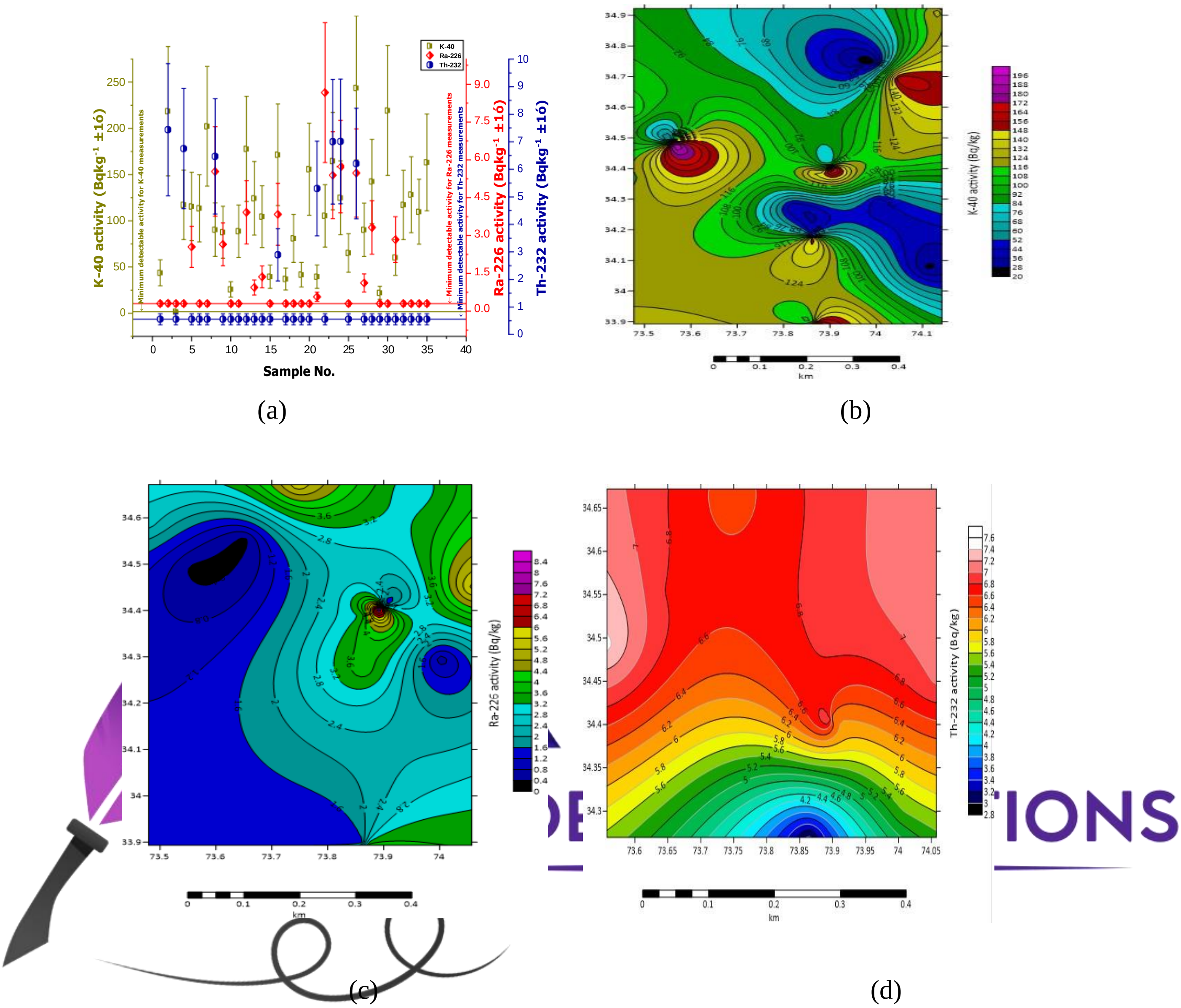

Figure 3.25. (a) Radionuclidic (^{40}K, ^{232}Th and ^{226}Ra) activities in 35 samples of medicinal herbs; (b, c, d) 2D contour maps respectively drawn for (^{40}K, ^{232}Th and ^{226}Ra) activities

Radionuclidic activity distribution within five herbal species (total 35 samples) is also depicted by 2D contour maps shown in Figure 3.25(b, c, d). The higher radionuclidic activity within investigated herbal species signified by denser lines as well as color scale thereby presenting the higher dissolved radionuclide areas respective to radionuclidic activity.

3.4.6.1. Remedial action

Bistorta amplexicule (leaves) sample mostly has higher potassium contents in comparison with other herb species. Therefore, it might be an appropriate choice for potassium absorption. The recorded higher ^{40}K contents within Bistorta amplexicule (leaves) samples might aid it's therapeutic purpose for the medications of high blood pressure (B.P) as high blood pressure patients had blood streams of relative low potassium content (HBPI, 2012). Almost every diabetic found to be hypertensive and this herb could also be employed for diabetic patients as remedial source (Apiah, 2012). It is also a better choice of potassium absorption by supporting their medicinal applications against various other respiratory ailments including cough, asthma and persistent disruptive pulmonary complications, since enriched potassium channels are highly needed against such ailments as health supplements (Pourimani et al., 2016).

3.4.6.2. Health hazards

Bergenia ciliate leaves have rough thick morphology structure which might help in trapping atmospheric deposition as well as absorption that is one of the reasons of higher ^{226}Ra (^{210}Pb) contents in leafy fragments than its roots (Somashekarappa, 1993). Due to higher water solubility of radium compared to thorium, subsequently then radium transmitted to aerial portions of herb by its root. Being alkaline earth metal, Radium isotope exhibit chemically alike features as Mg and Ca have, which essentially needed for plant's nutrition as well as growth. Due to radium availability, herbs might take up radium instead of Mg and Ca. Being similarity of toxic radium with calcium, despite of its uses, uptaken radium might be then ingested by the consumer and are rapidly deposited onto the bones and soft tissues and hence causes a head sinus as well as bone cancer (El-Gamal et al., 2019). It is concluded that Bergenia ciliate (leaves) with associated toxic radium then might be resulted into developing of cancer risks; therefore, it should be investigated further, in order to ensure safety of consumer or patients.

Highest value of ^{232}Th (^{228}Ac) was noticed in Bistorta amplexicule (Leaves) sample of Pir Chinasi site of Muzaffarabad district. For adult patients or consumer, presence of high thorium concentration with age is highly probable. Since, Wrenn et al., 1981 and Ibrahim et al. 1983 reported about the sluggish removal of thorium from bones (Wrenn et al. 1981; Ibrahim et al. 1983). So, Bistorta amplexicule (leaves) of Pir chinasi sites has a radiological risk and it should be used for medication only after investigation.

For all tested herbs type, no artificial radionuclidic activity was observed. However, ^{137}Cs (0.2 ±0.02) Bq kg^{-1} was detected in herb type Bistorta amplexicule (leaves) samples only from Toli peer site of Poonch district. These findings thus signified that ingestion of tested herb Bistorta amplexicule (leaves) of Poonch sites are of a little radiological risk and are strongly prohibited for the indigenous patients as well as consumers.

Table 3.13. Descriptive statistics of radionuclidic (^{40}K, ^{232}Th and ^{226}Ra) activity concentrations in medicinal herbs

	Activity (Bq kg^{-1} ±1ó)		
Radionuclides	^{40}K	^{232}Th	^{226}Ra
No. of observations	35	35	35
Maximum	243.77±22.73	7.45±0.76	8.67±1.08
Minimum	≤MDA	≤MDA	≤MDA
Range	≤MDA-243.77±22.73	≤MDA-7.45±0.76	≤MDA -8.67±1.08
Standard deviation	58.86	1.45	2.37
Skewness	0.41	-1.89	0.63
Kurtosis	-0.45	3.78	0.03
Mean	112.75±14.11	6.16±2.22	3.39±0.64
Median	111.67	6.63	3.08
Variance	3725.32	7.31	5.30

Table 3.13 portrays the descriptive statistics of radionuclidic activity concentrations in medicinal herbs. For radionuclide (^{40}K), range of observations for all investigated samples, was reported (≤MDA-243.77±22.73) Bq kg^{-1} and for radionuclide (^{232}Th), the range of measurements, was documented as (≤MDA-7.45±0.76) Bq kg^{-1}, while for radionuclide (^{226}Ra), range of measurements, was observed (≤MDA-8.67±1.08) Bq kg^{-1} (see Figure 3.26). Smallest radionuclidic activity (^{40}K), following MDA$_{k-40}$ value was noticed as (22.07) Bq kg^{-1} and (243.77) Bq kg^{-1} being greatest. Likewise, lowest radionuclidic activity (^{232}Th), following immediately after MDA$_{Th-232}$, was observed as (2.94) Bq kg^{-1}, but greatest as (7.45) Bq kg^{-1}. Similarly, smallest radionuclidic activity (^{226}Ra), following MDA$_{Ra-226}$ value, was noticed as (0.31) Bq kg^{-1} while with (8.67) Bq kg^{-1} being highest.

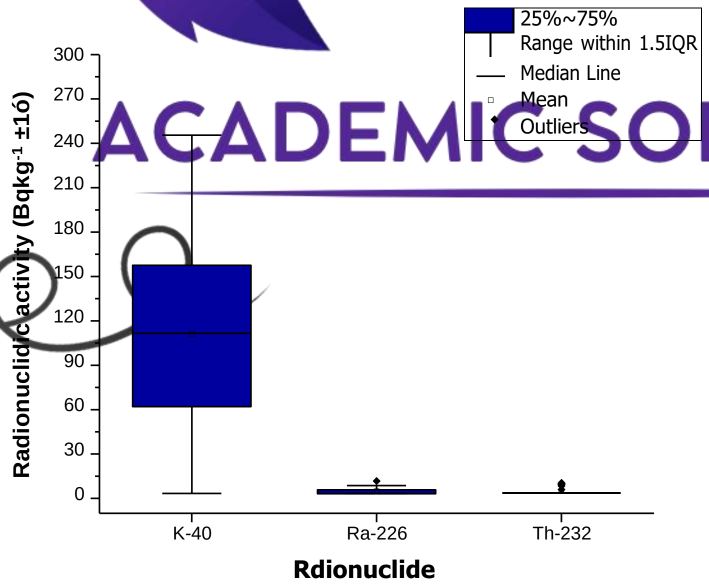

Figure 3.26. Minimum, maximum and mean values of radionuclidic activities

Table 3.13 portrays the values of variance for radionclidic ^{40}K, ^{232}Th and ^{226}Ra respectively. The estimated values are 3725.32, 7.31 and 5.32 for potassium, thorium and radium radionuclides. In order to check symmetry skewness of the investigated data was also calculated. Positive and small skewness (0.41) was observed for radionuclide ^{40}K that indicates the growing trends for radionuclidic (^{40}K) activity which might be associated to the potassium growth at investigated locations. It was observed that radionuclidic (^{232}Th) activity have negative skewness (-1.89) presenting about falling radioactivity trend. Positive skewness coefficient (0.61) was noticed for ^{226}Ra case, which illustrates growing radioactivity level that might be associated to high radon gas exhibited at constituents' soil of investigated herbs.

To investigate radionuclidic activities data, whether it is lightly or heavily tailed comparative to normal distribution, kurtosis coefficients were also determined. Kurtosis value for radionuclidic (^{40}K, ^{232}Th and ^{226}Ra) activities was respectively found as -0.45, 3.78 and 0.03. For ^{40}K case, negative kurtosis showed that distribution is platykurtic with tail heavier to normal as listed in Table 3.13. Also, positive but small kurtosis presents leptokurtic distribution for ^{226}Ra, while positive and large kurtosis for radionuclide (^{232}Th) revealed about leptokurtic distribution with lighter tail comparative to normal ones.

To check the existed ratios among three radionuclides of herbs, correlation was drawn for them along with data points (see Figure 3.27). Correlation coefficients were determined respectively for ^{226}Ra and ^{232}Th, ^{226}Ra and ^{40}K, ^{232}Th and ^{40}K, via regression method. In case of ^{226}Ra and ^{232}Th, positive and negligible regression was calculated, but for, ^{226}Ra and ^{40}K, ^{232}Th and ^{40}K, both show positive as well as small regression. This signified that ^{226}Ra and ^{232}Th have a somewhat similar origin. While, ^{40}K was noticed as independent source compared to both ^{232}Th and ^{226}Ra. Nevertheless, positive correlation might be associated to properties of soil's constituent, where these radionuclides exhibited under dissimilar weathering conditions.

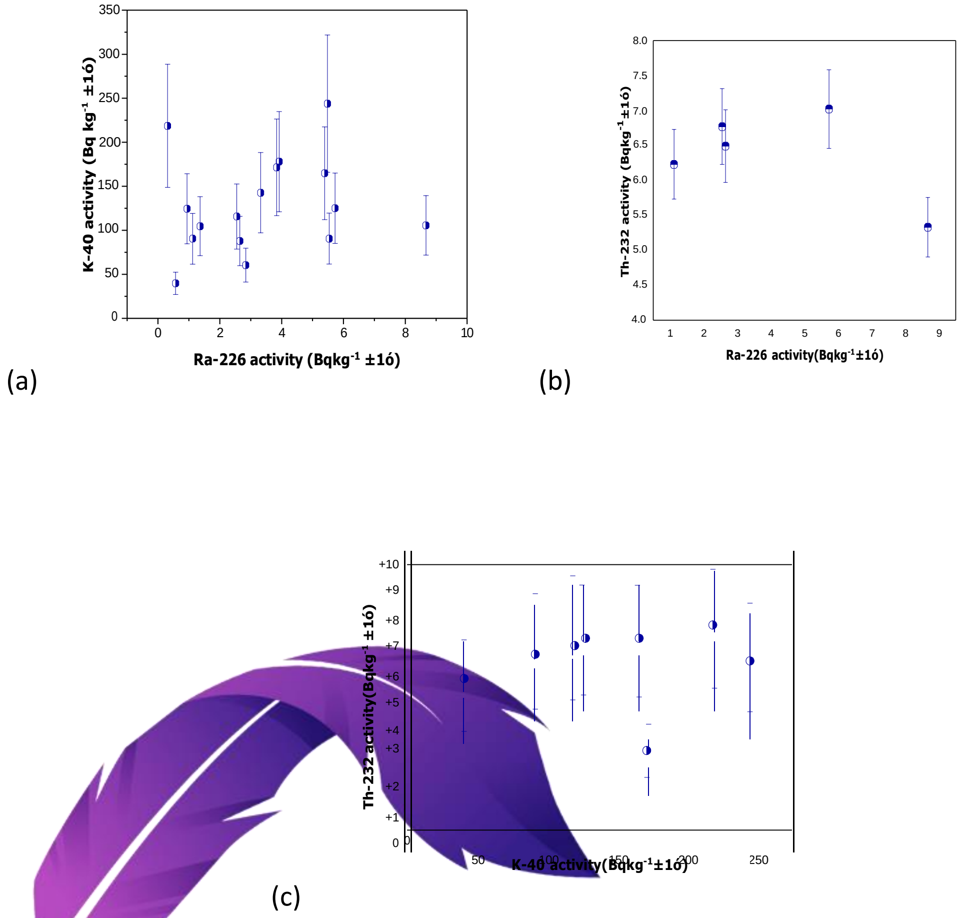

Figure 3.27. Correlation drawn for five medicinal herb species (a) between ^{226}Ra and ^{232}Th activity level (b) between ^{226}Ra and ^{40}K activity level (c) between ^{232}Th and ^{40}K activity level

Mean values of radionuclidic activity (^{40}K) Bq kg^{-1} in only root samples of medicinal herb species follow the order as; Bergenia Ciliate (62.05) > Bistorta Amplexicule (60.26), while for radionuclidic activity (^{232}Th) Bq kg^{-1} in only root samples of herbs follow the pattern as; Bergenia Ciliate (7.03) > Bistorta Amplexicule (6.49) but radionuclidic activity (^{226}Ra) Bq kg^{-1}, follow the order as; Bistorta Amplexicule (7.11) > Bergenia Ciliate (4.29) for root samples case only (see Figure 3.28).

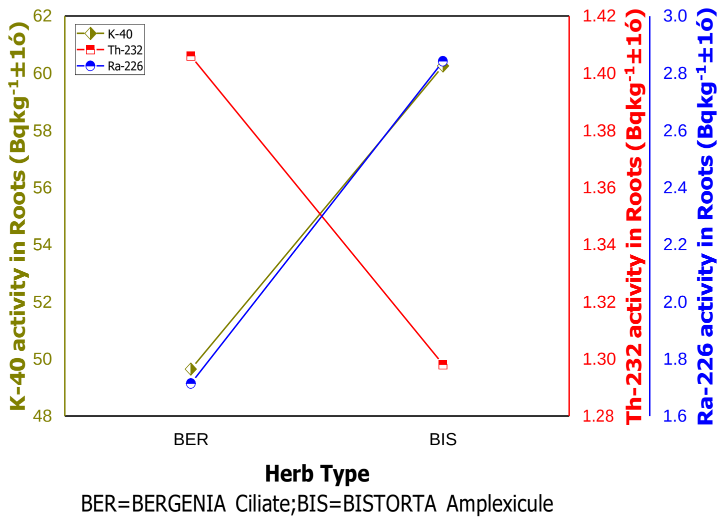

Figure 3.28. Mean values of radionuclidic activities (^{40}K, ^{232}Th and ^{226}Ra) in roots samples of Bergenia Ciliate and Bistorta Amplexicule medicinal herbs

For herbal species (leaves) samples (see Figure 3.29), radionuclidic activity (^{40}K) Bq kg^{-1} follows the following order as; Bistorta Amplexicule (172.32) > Mentha Longifollia (141.42) > Polygonum Aviculare (130.44) > Nastrition Officinal (118.76) >Bergenia Ciliate (93.88). Likewise, mean values of radionuclidic activity (^{232}Th) Bq kg^{-1}, follows the following order for herbal species (leaves) samples; Bergenia Ciliate (6.77) > Mentha Longifollia (6.23) > Bistorta Amplexicule (5.80) > Polygonum Aviculare (5.33) > Nastrition Officinal (≤MDA). Similarly, radionuclidic activity (^{226}Ra) Bq kg^{-1}, the pattern for herbal species (leaves) samples as follow; Mentha Longifollia (3.98) > Bistorta Amplexicule (3.05) > Polygonum Aviculare (1.75) > Nastrition Officinal (1.03) > Bergenia Ciliate (≤MDA).

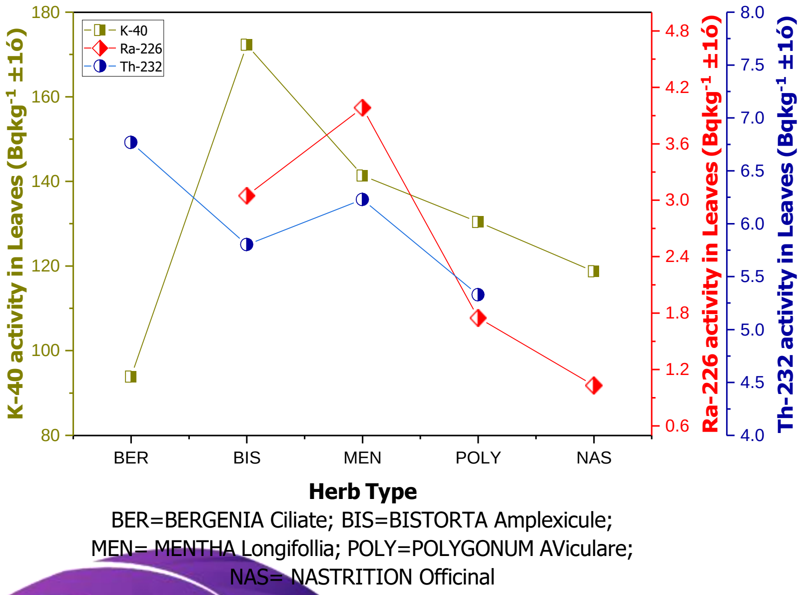

Figure 3.29. Mean values of radionuclidic activities (^{40}K, ^{232}Th and ^{226}Ra) in leaves samples of Bistorta Amplexicule, Mentha Longifollia, Bergenia Ciliate, Polygonum Aviculare and Nastrition Officinal medicinal herbs

3.4.6.3. Effect of altitude on radionuclidic activities

Most of the herbs were taken from places situated at higher altitude of about greater than 5500 feet. About 80.6 % of herbs samples were taken from locations with height ranged from 5500-9193 feet. It is clear from region 1(see Figure 3.30), only two samples were observed with ^{40}K activity and one sample with ^{226}Ra activity, also two samples were found to have either below or less than MDA. But, a few samples were lies within this height range (2200-3500 feet). In region 2, at height ranged 5500-6500 feet, nine samples with ^{40}K activity and five samples with ^{226}Ra, but only one sample with ^{232}Th was reported, while four samples were found to have less than MDA for ^{232}Th and ^{226}Ra case. Most of the samples with radionuclidic activities were noticed to lie within region 3, with height ranges 6500-8000 feet, while, radionuclide (^{40}K) activity usually found higher than the others but rest of the samples for radionuclides (^{226}Ra and ^{232}Th) were observed either below or less than MDA. In region 4, with height ranged 9000-9500 feet; a very few number of samples with radionuclidic activities was noticed but mostly with higher activity contents.

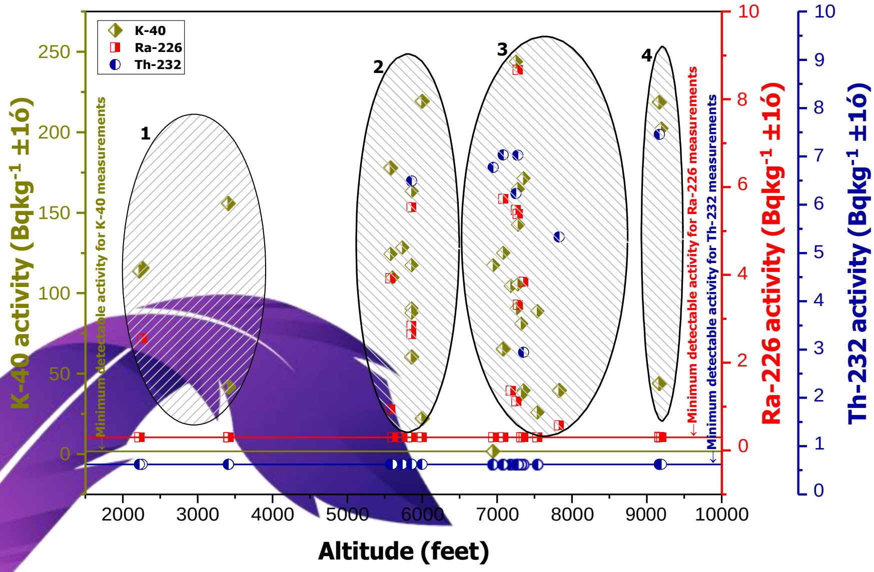

Figure 3.30. Variation of radionuclides (^{40}K, ^{232}Th and ^{226}Ra) with respect to altitude

3.4.6.4. Age dependent Annual (^{232}Th and ^{226}Ra) effective dose

The average radionuclidic annual committed doses ($ACED_{Ra\text{-}226}$ and $ACED_{Th\text{-}232}$) for a particular sample were calculated by equation (3.28). Here, ^{40}K is not investigated for doses because ^{40}K isotope being homeostatic, body itself stabilizes it (Jabir et al., 2007). $ACED_{Ra\text{-}226}$ and $ACED_{Th\text{-}232}$ were determined for different age groups including adults (>15 yr), fifteen year (15 yr), ten year (10 yr), five year (05 yr), one year (01yr) and less than one year (<1yr). For medicinal herbs consumption rate (MHCR) of 1.8 kgyr^{-1}, $ACED_{Ra\text{-}226}$(μSvy^{-1}) directed to adults (>15 yr), fifteen year (15 yr), ten year (10 yr), five year (05 yr), one year (01yr) and less than one year (<1yr) ranged from BDL to 4.37, BDL to 23.41 , BDL to 12.48, BDL to 9.68, BDL to 14.98, and BDL to 88.95 with an average values (0.78, 4.18, 2.23, 1.73, 2.68 and 15.90) µSv y^{-1} respectively. Likewise, with same CR of 1.8 kg yr^{-1}, $ACED_{Th\text{-}232}$ directed to adults (>15 yr), fifteen year (15 yr), ten year (10 yr), five year (05 yr), one year (01yr) and less than one year (<1 yr) ranged from BDL to 3.08, BDL to 3.35, BDL to 3.89, BDL to 4.69, BDL to 6.03 and BDL to 21.46 with an average values (0.58, 0.63, 0.73, 0.89, 1.14 and 4.05) $µSvy^{-1}$ respectively. For others MHCR, viz 2, 4, 6, 8 and 10 $kgyr^{-1}$, $ACED_{Ra\text{-}226}$ and $ACED_{Th\text{-}232}$ goes on increasing respectively with increase of CR, as it is vividly shown in figure (3.31) and figure (3.32), but fall within safe ACED limit (300 µSv y^{-1}). Thus, all investigated herbs of selected areas are safe in terms of effective doses, and radioactivity impacts except Bistorta amplexicule (leaves) sample from sampling site Toli pir of Poonch district, that is explored with anthropogenic contents (^{137}Cs). Also, these findings help out to explore the safe usage of medicinal herbs along their radioactivity distribution which might assist to pharmaceutical industries and herbal practitioner in diversity of formulation.

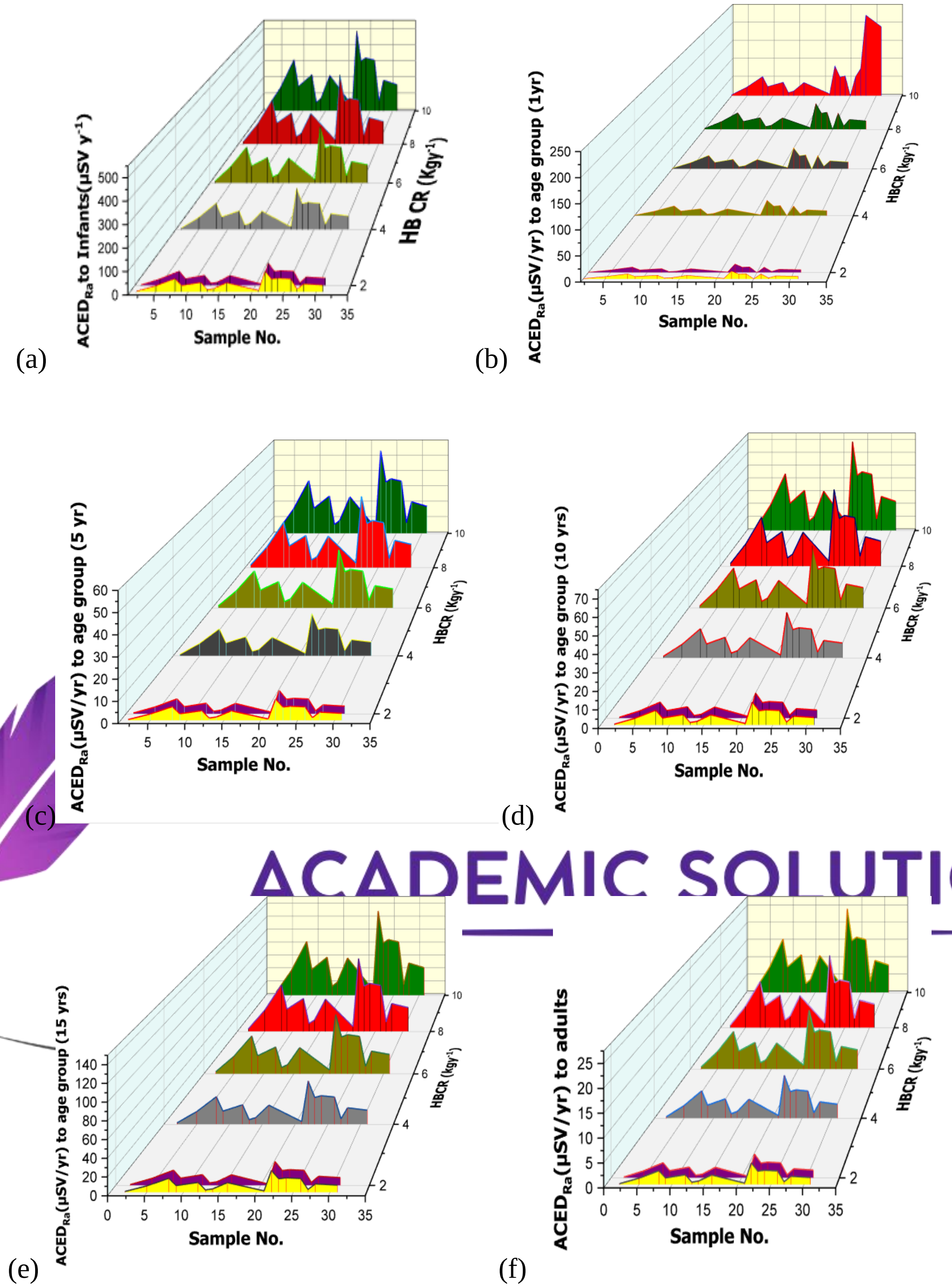

Figure 3.31. Radionuclidic doses (ACED$_{Ra-226}$) to a) < 1 year; b) 01 year; c) 05 years; d) 10 years; e) 15 years and f) > 15 year populace due to Medicinal herbs ingestion

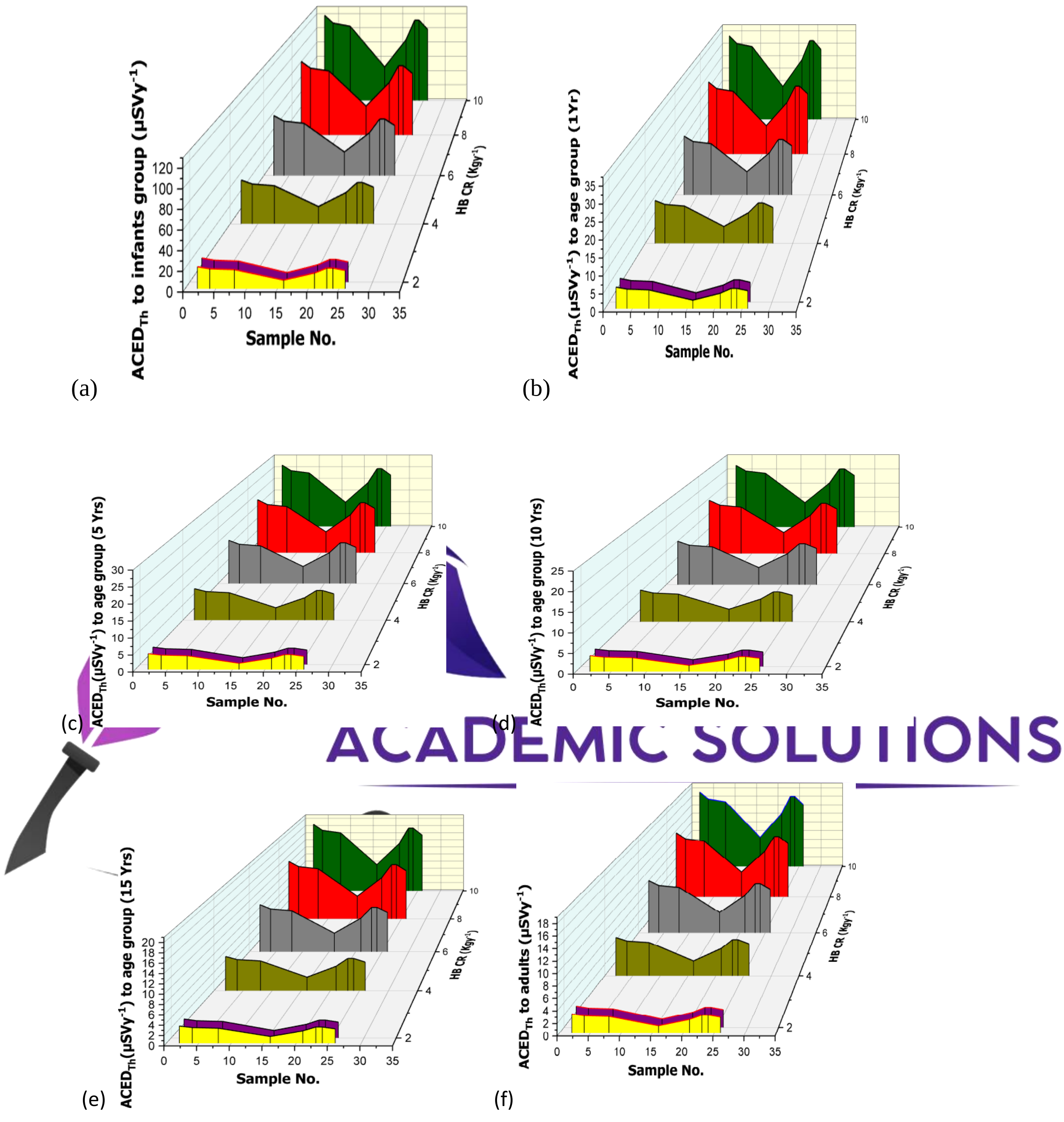

Figure 3.32. Radionuclidic doses (ACED$_{Th-232}$) to) < 1 year; b) 01 year; c) 05 years; d) 10 years; e) 15 years and f) > 15 year populace due to Medicinal herbs ingestion

3.4.6.5. Transfer factor soil-medicinal herb

Owed to spatial inconsistency as well as complexity of soil-plant system, which is rather difficult to count radiation uptakes of medicinal herb through soil, radionuclide transfer factor is usually calculated. Transfer coefficient referred as T. F and defined as ratio between radionuclidic activity within plant sample to soil (Dowdall et al., 2005; Greger, 2004).

Transfer factor relies upon the soil features, type of plant and radionuclides as well (Velasco, 2008; IAEA, 2009). Activity content of medicinal herb portrayed in table 3.13 and of soil illustrated (Shahzadi et al., 2020) through which calculated transfer factors were obtained and are enlisted in table 3.14.

Owing to assumption for consumption rate about (1.8) kg y^{-1}, ACED by ingested ^{40}K, ^{232}Th and ^{226}Ra within medicinal herb of all five Azad Kashmir districts samples were evaluated by employing corresponding values of each radionuclides and are tabulated (see Table 3.14). The highest $ACED_t$ (8.06) µSv y^{-1} was carried by the Reshian sample (Mentha Longifollia) of Jhelum valley district, while Pir chinasi sampling site of district Muzaffarabad explored with lowest as (0.00) µSv y^{-1} for sample Bergenia ciliate(roots). The calculated $ACED_t$ for medicinal herbs from five districts of Azad Kashmir ranges (0.00-8.06) µSv y^{-1} with an average value (2.55) µSv y^{-1}.

$ACED_t$ values by ingesting NORMs within medicinal herbs Bistorta amplexicule (roots, leaves), Bergenia ciliate(roots, leaves), Nastrium officinale, Mentha longifollia and Polygonum aviculare of all investigated five Azad Kashmir districts were observed less than global average ACED limit (300) µSv y^{-1} as published in UNSCEAR report (UNSCEAR, 2000)(see Figure 3.33). Figure 3.34 presented the $ACED_t$ function of numerous threshold consumption rate ranges (0.00-6577.31) kg y^{-1}.

Table 3.14. Transfer coefficients of the enlisted radionuclides from soil to medicinal herbs samples, $ACED_t$ (adults group) and threshold consumption rate (kg y^{-1})

Sampling district	Sampling sites	Sample code	Investigated herb	Transfer Coefficients			$ACED_t$ (μSvy^{-1})	Threshold Consumption rate(kgy^{-1})
				^{40}K	^{226}Ra	^{232}Th		
Muzaffarabad	Pir chinasi	1	Bistorta amplexicule (Roots)	0.07	0.00	0.00	0.16	3311.92
	Pir chinasi	2	Bistorta amplexicule (Leaves)	0.33	0.01	0.10	1.89	285.24
	Pir chinasi	3	Bergenia ciliate (Roots)	0.00	0.00	0.00	0	0.00
	Pir chinasi	4	Bergenia ciliate (Leaves)	0.18	0.01	0.09	1.37	394.03
	Centre plate	5	Mentha Longifollia (Leaves)	---	---	---	0.86	629.27
	Plate	6	Nastrurtium officinal (Leaves)	---	---	---	0.42	1278.39
	Pir chinasi	7	Polygonum aviculare	0.31	BDL	BDL	0.75	717.66
Poonch	Toli Peer	8	Bistorta amplexicule (Roots)	0.20	0.14	0.11	2.16	249.69
	Toli Peer	9	Bistorta amplexicule (Leaves)	0.20	0.07	0.00	0.77	700.09
	Toli Peer	10	Bergenia ciliate (Roots)	0.06	0.00	0.00	0.1	5602.52
	Toli Peer	11	Bergenia ciliate (Leaves)	0.20	0.00	0.00	0.33	1635.62
	Ali Sojal	12	Mentha Longifollia (Leaves)	---	---	---	1.32	409.12
	Ali Sojal	13	Nastrurtium officinal (Leaves, shoots)	---	---	---	0.62	870.37
	Toli peer	14	Polygonum aviculare	0.24	0.04	BDL	0.62	874.79
Bagh	Sudhan Gali	15	Bistorta amplexicule (Roots)	0.12	0.00	0.00	0.15	3669.40
	Sudhan Gali	16	Bistorta amplexicule (Leaves)	0.50	0.08	0.05	1.69	319.82
	Sudhan Gali	17	Bergenia ciliate (Roots)	0.11	0.00	0.00	0.14	3915.87
	Sudhan Gali	18	Bergenia ciliate (Leaves)	0.24	0.00	0.00	0.3	1793.00
	Bagh	19	Mentha Longifollia (Leaves)	---	---	---	0.16	3478.58
	Bagh	20	Nastrurtium officinal (Leaves, shoots)	---	---	---	0.58	931.54

Table 3.14. continued….

Table 3.14. continued…

Sampling district	Samplin g sites	Sample code	Investigated herb	Transfer Coefficients			$ACED_t$ (μSvy^{-1})	Threshold Consumption rate(kg y^{-1})
				^{40}K	^{226}Ra	^{232}Th		
	Sudhan Gali	21	Polygonum aviculare	0.12	0.01	0.10	0.98	551.76
Hattian Bala	Reshian	22	Bistorta amplexicule (Roots)	0.23	0.22	0.00	1.85	292.09
	Reshian	23	Bistorta amplexicule (Leaves)	0.37	0.14	0.07	2.49	217.15
	Reshian	24	Bergenia ciliate (Roots)	0.28	0.14	0.07	2.4	225.23
	Reshian	25	Bergenia ciliate (Leaves)	0.15	0.00	0.00	0.24	2226.06
	Reshian	26	Mentha Longifollia (Leaves)	---	---	---	2.69	200.95
	Reshian	27	Nastrurtium officinal (Leaves, shoots)	---	---	---	0.52	1030.24
	Reshian	28	Polygonum aviculare	0.32	0.08	BDL	1.09	496.25
NeelumValley	Nagdar	29	Bistorta amplexicule (Roots)	0.03	0.00	0.00	0.08	6577.31
	Nagdar	30	Bistorta amplexicule (Leaves)	0.35	0.00	0.00	0.82	661.78
	Nagdar	31	Bergenia ciliate (Roots)	0.10	0.11	0.00	0.7	769.93
	Nagdar	32	Bergenia ciliate (Leaves)	0.18	0.00	0.00	0.44	1238.26
	Nagdar	33	Mentha Longifollia (Leaves)	---	---	---	0.48	1131.24
	Nagdar	34	Nastrurtium officinal (Leaves, shoots)	---	---	---	0.41	1322.17
	Nagdar	35	Polygonum aviculare	0.26	BDL	BDL	0.61	889.47

Threshold consumption rate as indicated by table 3.14 that might be considered as limiting data below which $ACED_t$ value remains consistent to (300) µSv/y for any specie of medicinal herb. It is also clear from Table 3.14 and Figure (3.34) that higher the threshold value for medicinal herb, smaller be the $ACED_t$ which signified that patients having consumption rate less than threshold value would resulted in trivial radiological hazards but consumption rate fairly higher than threshold value will definitely pose a substantial health hazards.

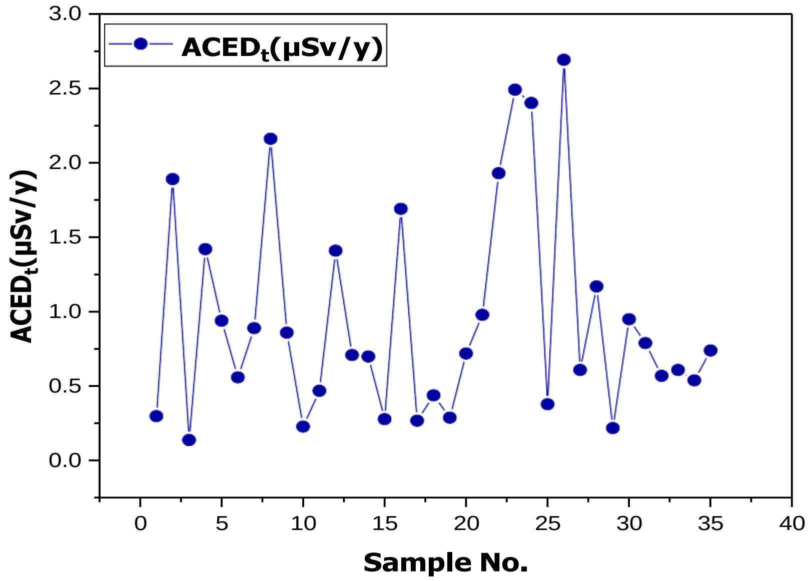

Figure 3.33. ACED$_t$ (µSvy^{-1}) to adults for medicinal herb samples from five Azad Kashmir districts

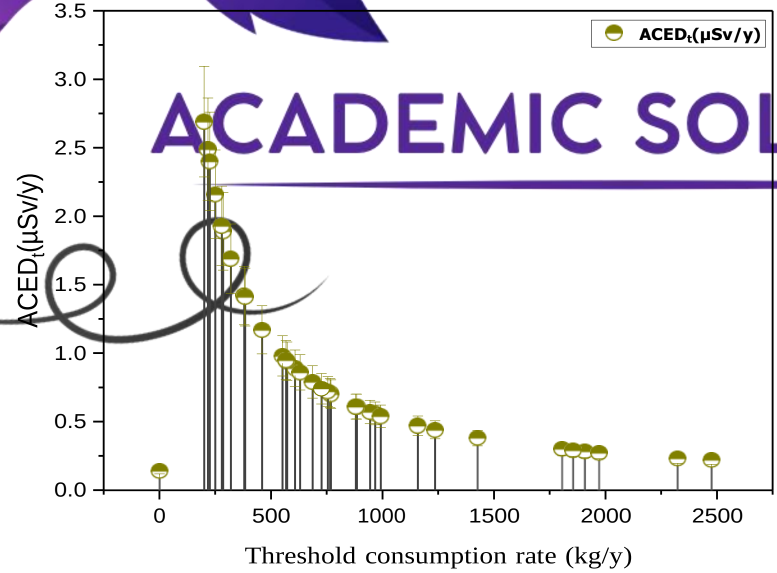

Figure 3.34. Consumption rate (kg y^{-1}) vs ACED$_t$ (µSv y^{-1}) for medicinal herbs

In case of ^{226}Ra, the minimum T.F (BDL) was observed in samples Polygonum aviculare at Nagdar and Pir chinasi sampling sites of respective districts Neelum valley and Muzaffarabad while, District Poonch sample Bistorta amplexicule(root) at Toli peer site and Hattian Bala district samples (Bistorta amplexicule (leaves), Bergenia ciliate(roots)) at site Reshian depicted the largest 0.14.

For case of ^{232}Th, a significant transfer factor (0.11) was reported at Toli peer of district Poonch sample Bistrorta amplexicule (root), but Muzaffarabad, Poonch, Neelum as well as Hattian Bala districts revealed with least (BDL) in Polygonum aviculare samples at respective sites Pir chinasi, Toli peer, Nagdar as well as Reshian. For ^{40}K, the higher transfer coefficient (0.37) was explored at Reshian sample Bistorta amplexicule (leaves) of Hattian Bala district and Muzaffarabad district delivered the lowest (0.00) in Bergenia ciliate (roots) at sampling site Pir chinasi.

The vast gap for ^{40}K activity was observed which is the consequence of massive difference in soil's activity contents as potassium uptake relies on several aspects including clay content, cation exchange bulk, texture, PH and also upon the plant portions and growth phase (Velasco et al., 2008). Hence wide range for TF data was relatively observed. The average value of T.F for medicinal herb samples from five different Azad Kashmir districts were (0.05), (0.21) and (0.03) that correspondingly noticed for ^{226}Ra, ^{40}K and ^{232}Th radionuclides. The TF of ^{40}K, ^{226}Ra and ^{232}Th and ranges (0.00-0.37), (BDL-0.14) and (BDL-0.11) respectively (see Figure 3.35).

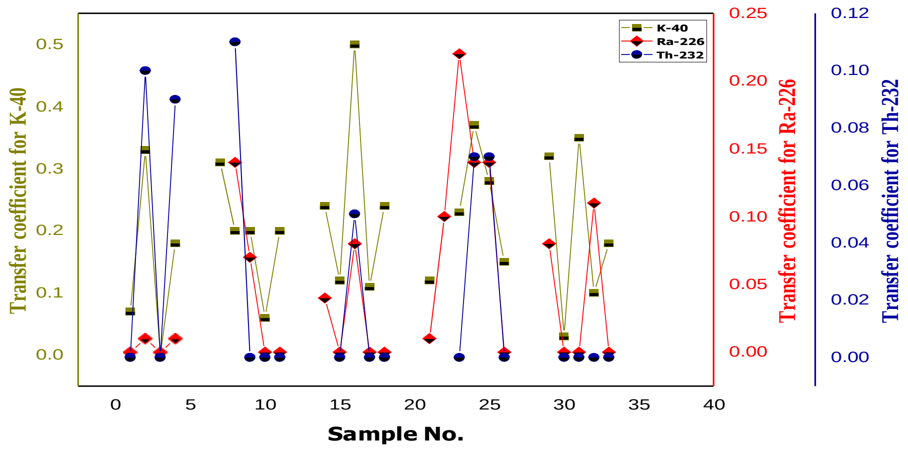

Figure 3.35. Comparison of transfer factors with radionuclides in medicinal herbs samples from five Azad Kashmir districts

Comparison was made for transfer coefficients of present research with available literature TF of herbs/plants shown by the Figure 3.36 and comparative report enlisted in table 3.15.

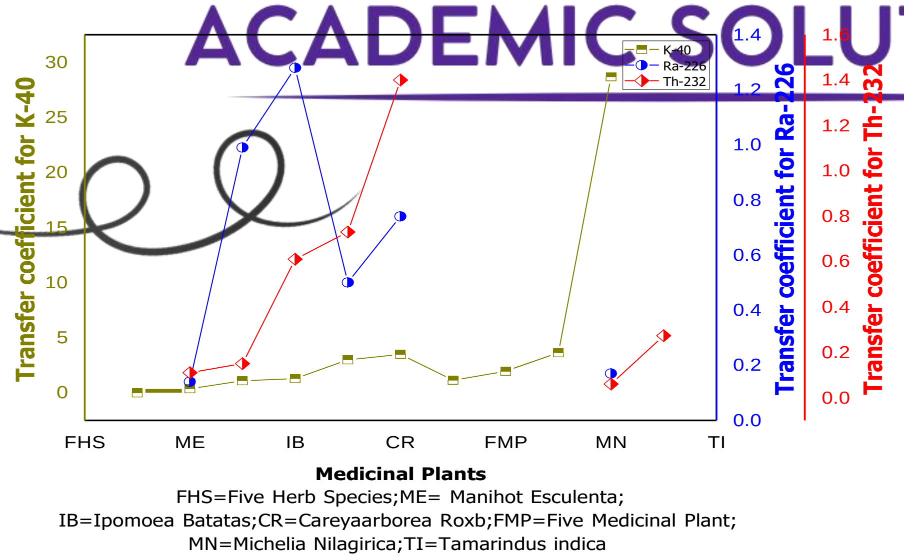

Figure 3.36. Comparison of transfer factor of different countries' species of medicinal plants/herbs

Table 3.15. Currently investigated medicinal herbs transfer factor's comparison with available literature

Radionuclide	Plant species	T. F	References
K-40	Five medicinal herbs (Polygonum Aviculare, Nastrituim offisinal, Mentha Longifollia, Bistorta amplexicule, Bergenia ciliate)	0.00-0.37	Current study
	Manihot esculenta	1.10-1.29	Asaduzzaman et al., 2014
	Ipomoea batatas	3.0-3.5	Asaduzzaman et al., 2014
	Careyaarborea Roxb	1.14-1.96	Karunakara, 1997
	Five medicinal plants (Careya arborea Roxb, Plectranthus amboinicus spreng, Mimosa pudica L, Justica adhatoda, Azadirachta indica A jus.)	3.63-28.66	ChandrashekaraK et al., 2015
Ra-226	Five medicinal herbs	BDL-0.14	Current study
	Manihot esculenta	0.99-1.28	Asaduzzaman et al., 2014
	Ipomoea batatas	0.50-0.74	Asaduzzaman et al., 2014
	Tamarindus indica	BDL	Karunakara, 1997
	Five medicinal plants (Careya arborea Roxb, Plectranthus amboinicus spreng, Mimosa pudica L, Justica adhatoda, Azadirachta indica A jus.)	BDL-0.17	ChandrashekaraK et al., 2015
Th-232	Five medicinal herbs	BDL-0.11	Current study
	Manihot esculenta	0.15-0.61	Asaduzzaman et al., 2014
	Ipomoea batatas	0.73-1.4	Asaduzzaman et al., 2014
	Michelia nilagirica	0.273	Manigandan, 2009
	Five medicinal plants (Careya arborea Roxb, Plectranthus amboinicus spreng, Mimosa pudica L, Justica adhatoda, Azadirachta indica A jus.)	BDL-0.06	ChandrashekaraK et al., 2015

3.4.7. Comparison of Data Results with Available Literature

Figure 3.37 presented a previously published work for NORM's activity contents within medicinal herbs/plants of various countries and their mean data as well as range was compared to current calculated values. It is clear from Figure (3.37) that current measured average ^{40}K activity content (112.75±14.11) Bq kg^{-1} was found less than Ghana (839.8 Bq kg^{-1}) (Tetty-larbi, et al., 2013), Serbia (589.6 Bq kg^{-1}) (Jevremovic et al., 2011), Brazil (976.3 Bq kg^{-1}) (Scheibel & Appoloni, 2007), Nigeria (171.7 Bq kg^{-1}) (Njinga et al., 2015) and Italy (654.7 Bq kg^{-1}) (Desideri et al., 2010) except for Nigeria (Ugelli) reported (67.9 Bq kg^{-1}) (Oni et al., 2011).

Average ^{232}Th (Ac-228) activity in this work (6.16±2.22 Bq kg^{-1}) found less compared to those reported by Ghana (56.2 Bq kg^{-1}) (Tetty-larbi et al., 2013), Nigeria (Ugeli) (8.5 Bq kg^{-1}) (Oni et al., 2011), Serbia (7.4 Bq kg^{-1}) (Jevremovic et al., 2011), Brazil (21.7 Bq kg^{-1}) (Scheibel & Appoloni, 2007) and Nigeria (35.1 Bq kg^{-1}) (Njinga et al., 2015).

Whereas, ^{226}Ra activity (3.39±0.66 Bq kg^{-1}) was observed greater to Serbia (2.6 Bq kg^{-1}) (Jevremovic et al., 2011) and Italy (0.4 Bq kg^{-1}) (Desideri, et al., 2010), but smaller to Nigeria (Ugelli) (15.6 Bq kg^{-1}) (Oni et al., 2011), Ghana (31.8 Bq kg^{-1}) (Tetty-larbi et al., 2013), and Nigeria (25 Bq kg^{-1}) (Njinga et al., 2015) as shown in Figure 3.37.

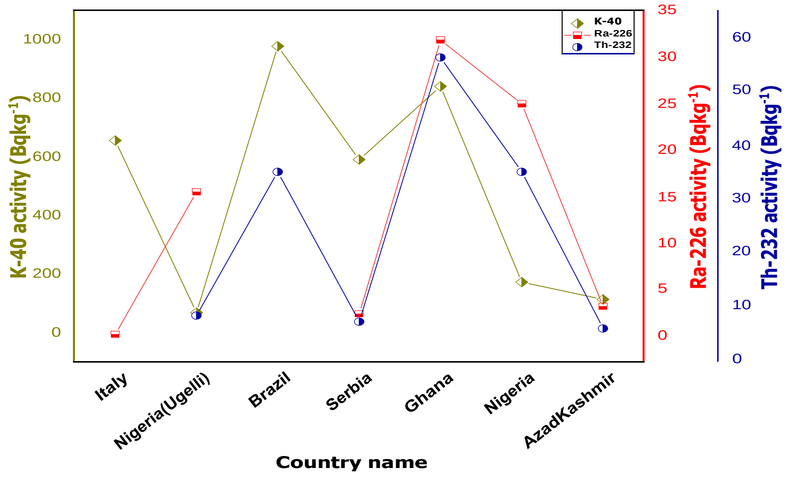

Figure 3.37. World activity comparison with current activity (Bq kg^{-1}) of medicinal herbs/plants samples

3.4. CONCLUSIONS

To conclude, it is impossible for public to avoid exposure from background radiations. Lifetime exposure of background radiations produces risk of developing cancer of about 1 % of the population. Followings are the outcomes of study;

- Average value of indoor radon in current study was found as 46.9 Bq m^{-3}, which is slightly higher than world average of 40 Bq m^{-3}. Average value of outdoor radon concentrations was found as 13.3 Bq m^{-3}, which is comparable with other countries, where outdoor radon varies from 5 to 15 Bq m^{-3}. For indoor measurements, mean value of GDR was found as 846 µGy y^{-1}. Mean value of outdoor GDR was found as 777 µGy y^{-1}. For indoor measurements, average value of annual effective dose (E_{Rn}) due to radon exposure was estimated as 1.18 mSv y^{-1} which is slightly above the recommended worldwide average of 1.15 mSv y^{-1}. Average value of radon doses delivered to lungs was found as 2.84 mSv y^{-1}. Excess lifetime cancer risk (ELCR) varied from 1.49 ×10^{-3} to 14.01×10^{-3} with mean value 4.38×10^{-3}. Results reported for $Lung_{cancer\ cases}$ are lower than ICRP recommended values viz. 170–230 per million persons.

- Gross alpha, gross beta and specific activities due to primordial radionuclides have been determined in soil samples taken from cultivated as well as non-cultivated lands. Average value of activity concentration from ^{226}Ra (37.91 ±2.35 Bq kg^{-1}) was within the range of worldwide average (50 Bq kg$^{−1}$), though ^{232}Th (55.83 ±5.74 Bq kg$^{−1}$) and ^{40}K (616.22 ±29.20 Bq kg$^{−1}$) activities for current study exceeded worldwide averages (50 Bq kg$^{−1}$, and 500 Bq kg$^{−1}$) for respective radionuclides. Average value of radium equivalent activity, Ra_{eq}, annual effective dose for indoor is within world averages, whilst annual effective dose for outdoor environment is comparable with the world average. Indoor, H_{in} (0.55) and out hazard index, H_{out} (0.45) is within the range of unity and mean value of representative level index (I_r = 1.22 Bq kg^{-1}) is greater than permitted safe value of unity. Average value of Excessive Lifetime Cancer Risk (ELCR) for current study (1.55 ×10$^{−3}$) is higher than the world average value (0.29 ×10$^{−3}$).
- Gross alpha/beta activities have been estimated in herbal wild plants. Gross alpha activity (GAA) ranged from minimum detection limit (MDL) of instrument i.e., ≤4.5 to 374.65±2.49 Bq kg^{-1}. Gross beta activity (GBA) ranged from MDL to 481.07±166.32 Bq Kg^{-1}. Minimum GAA, following MDL value, was found as 9.4 Bqkg^{-1} and maximum was found as 375 Bqkg^{-1}. Similarly, minimum GBA, following immediately after MDL value, was found as 158 Bq kg^{-1} and maximum as 481 Bq kg^{-1}. For medicinal plants consumption rate (MPCR) of 1.8 kg y^{-1}, the average gross alpha or beta annual committed effective dose (ACED), delivered to infants, one, five, ten, 15 years and adults ranges from 43±7 to 1732±18 µSv y^{-1}, 7±1 to 274±3 µSv y^{-1}, 5±1 to 192±2 µSv y^{-1}, 5±1 to 181±2 µSv y^{-1}, 6±1 to 248±3 µSv y^{-1} and 3±0 to 100±1 µSv y^{-1} with mean value 797±10, 274±2, 88±1, 83±1,

114±1 and 46±1 µSv y^{-1}. For higher values of MPCR, viz. 2, 4, 6, 8 and 10 kg y^{-1} respective gross alpha and gross beta ACED goes on increasing. Finding of study shows that, except ACED delivered to infants for MPCR of 1.8 kg y^{-1}, all other estimated values, at same MPCR, fall below the WHO recommended level (290 µSv y^{-1}) and that of as reported in UNSCEAR, 2000 (0. 3 mSv y^{-1} or 300 µSv y^{-1}) report.

- Natural radioactivities have been measured in medicinal plants. Results obtained showed that activity concentrations of ^{226}Ra, ^{40}K and ^{232}Th ranged from (≤MDA-8.67±1.08, ≤MDA-243.77±22.73, ≤MDA-7.45±0.76) Bq kg^{-1} with an average values 3.21±0.64, 112.75±14.11, 6.16±2.22 Bq kg^{-1} respectively. ^{226}Ra, ^{40}K and ^{232}Th transfer factors from soil constituent's to medicinal herbs (roots and leaves) were determined and found as 0.05, 0.21 and 0.03 respectively. Measured radionuclidic activities have also been employed to evaluate age dependent annual committed effective doses received by general public for age ranges of <1year, 1 year, 5 year, 10 year, 15 year and >15 year and results were found consistent to UNSCEAR safe limit for ACED (300 µSv y^{-1}). Study shows that investigated herbs are safe in terms of effective doses, cancer risks and radioactivity impacts.

Limitations of the Current Study

Though excessive lifetime cancer risks have been calculated based upon current study data, but due to unavailability of actual mortality and morbidity statistics, we couldn't be able to assess real time health hazards faced by the inhabitants.

3.5. FUTURE RECOMMENDATION

Current study is a preliminary invistigation for estimation of radionuclides contents and assessment of health hazrads and risk associated posed by radionuclide concentrations present in soil, atmosphere and herbal plants of local origin. Since current study have been conducted in specific part so it cannot meet the subject thoroughly, therefore, to get further deep insight about the radiological risks and toxic radiological contents present in the environmental samples of the area, various recommendations are hereby enlisted for relevant potential future research area:

- Current study is a preliminary work for some selected medicinal herbs of five districts of Azad Kashmir, it can be extended to the other districts of Azad Kashmir where some other kinds of herbal plants are available.
- The systematic elemental profile of medicinal herbs should be built by developing a systematized grid for medicinal herbs samples of studied area.
- Medicinal herbs samples should be tested for various other potentially hazardous heavy elemental contents.
- Same medicinal herb species and constituent soil should be assessed by other analysis technique like, Laser induced Breakdown Spectroscopy, Neutron Activation Analysis (NAA), X-ray fluorescence spectroscopy (XRF) etc.
- Radiometric analysis should be carried out for other environmental samples like water, vergin soil, and soil used for cultivation purpose, food, meat, chicken, and fruits.
- Mapping the area for possible environmental radiations.
- Development of mathematical models for risk assessment of radiations.
- Models of excessive risk of cancer development need to be developed.

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