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STUDY OF PUBLIC EXPOSURE AND ASSESSMENT OF RADIOLOGICAL RISK FROM NATURALLY OCCURRING RADIOACTIVE MATERIALS IN FIVE DISTRICTS OF AZAD KASHMIR

By

Chand Shahzadi

Reg. No. 2007-Gmdg-4561

Session: 2015-2018

Department of Physics

Faculty of Science

UNIVERSITY OF AZAD JAMMU AND KASHMIR,

MUZAFFARABAD

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A Thesis

Submitted in partial fulfillment of the requirements for the degree of

Doctor of Philosophy

In

Physics

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Faculty of Science

UNIVERSITY OF AZAD JAMMU AND KASHMIR,

MUZAFFARABAD

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Certification

Certified that contents and form of thesis entitled **"Study of public exposure and assessment of radiological risk from naturally occurring radioactive materials in five districts of Azad Kashmir''** submitted by **Chand Shahzadi h**ave been satisfactory for the requirement of **Doctor of Philosophy** in **Physics**.

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ABBREVIATIONS

ACED	:	Annual committed Effective Dose
AEDE	:	Annual Effective Dose Equivalent
AGDE	:	Annual Gonadal Dose Equivalent
BDL	:	Below Detection Limit
CL	:	Confidence Level
CPS	:	Count Per seconds
ELCR	:	Excessive Lifetime Cancer risk
GAA	:	Gross Alpha Activity
GBA	:	Gross Beta Activity
GDR	:	Gamma Dose Rate
H_{ex}	:	External hazard Index
H_{in}	:	Internal hazard Index
HPGe	:	High Purity Germanium Detector
IAEA	:	International Atomic Energy Agency
ICRP	:	International Commission for Radiation Protection
LLC	:	Lung Cancer Cases
ppm	:	parts per million
Sv	:	Sievert
Gy	:	Gray
MCA	:	Multi Channel Analyzer
MDA	:	Minimum Detectable Activity
NORMs	:	Naturally Occurring Radioactive Materials
PINSTECH	:	Pakistan Institute of Nuclear Science and Technology
UNSCEAR	:	United Nations Scientific Committee on the Effects of Atomic Radiation
USA	:	United State of America
WHO	:	World Health Organization

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‘‘Glory is to that **Allah**, Lord of Lords and world, the genuine spirit, the owner of the day of the judgment and the most Benevolent, the most Compassionate, and the most merciful’’. And my humble gratitude is due to **HOLY PROPHET (Peace Be Upon Him)** who enlighten our souls with the true belief in **ALMIGHTY ALLAH**, who emphasized to seek knowledge and to work for the benefit of humanity.

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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 ×10^{-3} to 14.01×10^{-3} with mean value 4.38×10^{-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

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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

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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

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(https://www.iaea.org/newscenter/news/naturalradioactivityinfoodexpertsdiscusharmonizing-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

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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).

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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 Stropharia coronilla 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

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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 (≤ 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) ×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(Tl) 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

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

Year of Study	Country	^{40}K	^{137}Cs	^{226}Th	^{232}Th	^{235}U	^{238}U	^{226}Ra	^{228}Ra	^{210}Po	^{210}Pb	^{222}Rn	References
1999	(Marshall Islands) SW Hawaii	-	(0.001-1)×10^{4}, Polypodiumscolopendria (0.200–3)×10^{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…

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Table 1.2. Continued…

Year of study	Country	^{40}K	^{137}Cs	^{226}Th	^{232}Th	^{235}U	^{238}U	^{226}Ra	^{228}Ra	^{210}Po	^{210}Pb	^{222}Rn	References
2015	(Shazand) Iran	MDA-(2.75±0.01) ×10^{-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	-	-	-	-	-	-	-	-	-	-	(10.6602.07–53.303 4.64) ×10^{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

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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}.

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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

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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.

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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 km². 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

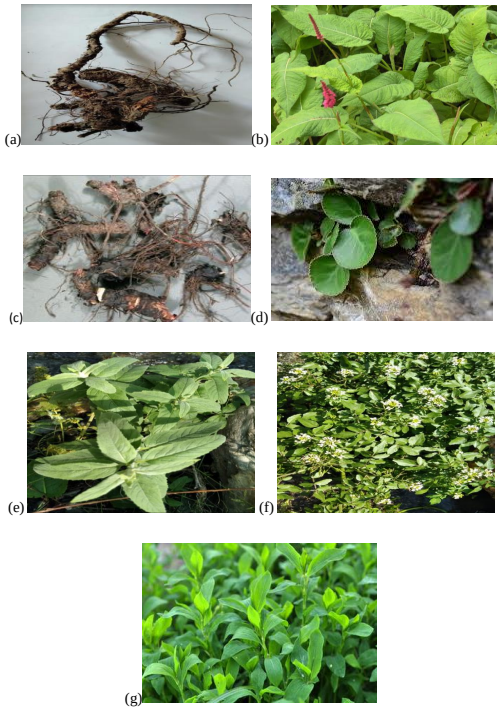

Figure 2.1. Snapshots of respective investigated medicinal herbs (a) Bistorta amplexicule roots, (b) Bistorta amplexicule leaves (c) Bergenia ciliate roots (d)Bergenia ciliate leaves (e) Mentha Longifollia (f) Nastrurtium Officinal (g) Polygonum Aviculare

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.

$$C_N = C_n - C_b \qquad (2.1)$$

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

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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).

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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

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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

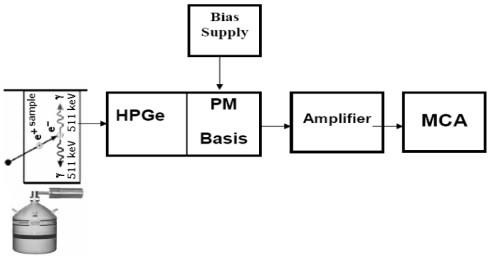

Figure 2.6. Block diagram for HPGe setup

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).

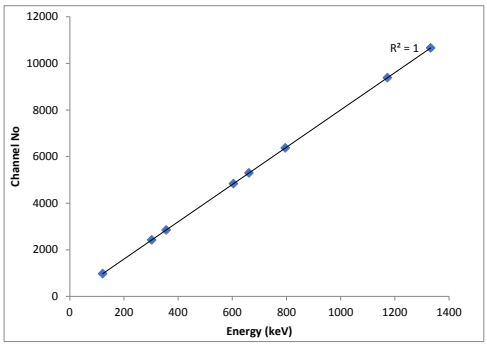

Figure 2.7. Energy calibration for HPGe setup

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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.

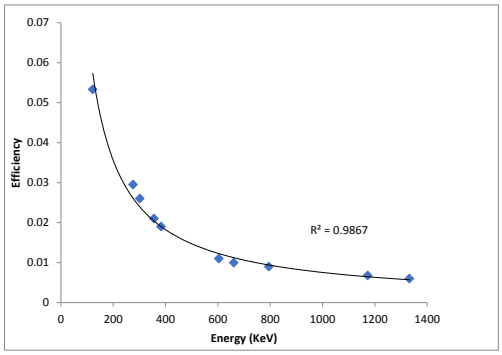

Figure 2.8. Efficiency calibration of HPGe setup

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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).

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

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.

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

Where, A and ΔA are the activity concentration and its uncertainty, ε, $\Delta\varepsilon$ are the efficiency, P_γ and ΔP_γ are the emission probability of the gamma ray of interest and its

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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:

$$\text{MDA} = \frac{4.66 \times \sqrt{Continum\ Counts + Background\ Peak\ Counts}}{t \times E_\gamma \times m \times P_\gamma} \qquad (2.6)$$

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;

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

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.

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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).

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

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

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;

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

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,

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

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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

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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).

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

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.

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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

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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.

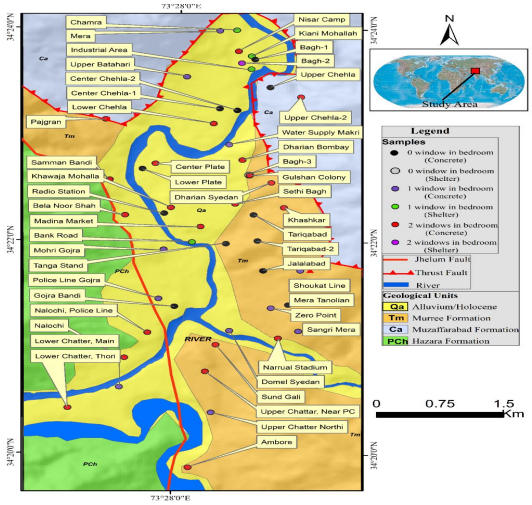

Figure 3.1. Geological map showing different formations in investigated study area

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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,

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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 ($\mu Gy\ y^{-1}$)	Outdoor Gamma DoseRates ($\mu Gy\ y^{-1}$)
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

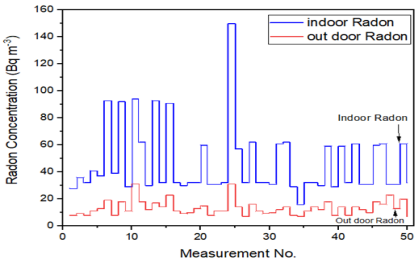

Figure 3.2. Indoor and outdoor radon concentrations a step horizontal connection graph

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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.

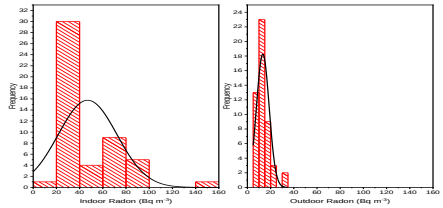

Figure 3.3. Normal distribution fitting on indoor and outdoor radon data

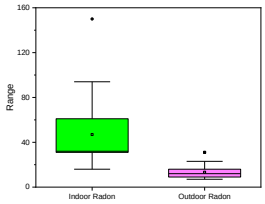

Figure 3.4. Box plot of indoor and outdoor radon data

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

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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).

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

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:

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

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):

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

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):

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

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 ×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.

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

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

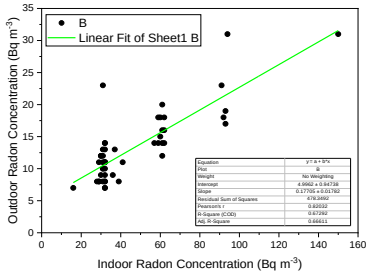

Figure 3.5. Plot of graph between indoor and outdoor radon concentrations

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.

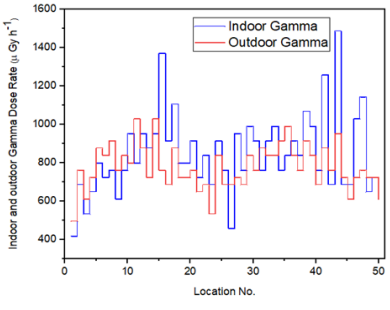

Figure 3.6. Indoor and outdoor GDR as a step horizontal connection graph

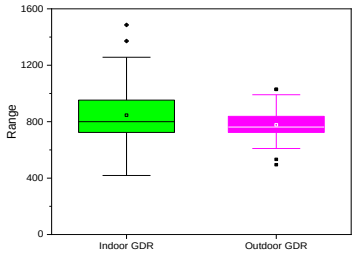

Figure 3.7. Box plot of indoor and outdoor GDR data

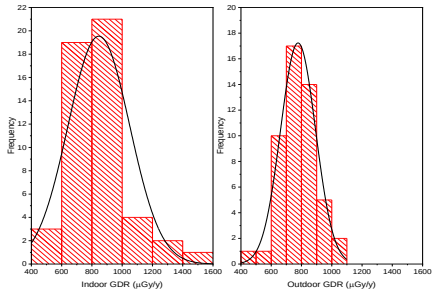

Figure 3.8. Normal distribution fitting on indoor and outdoor GDR

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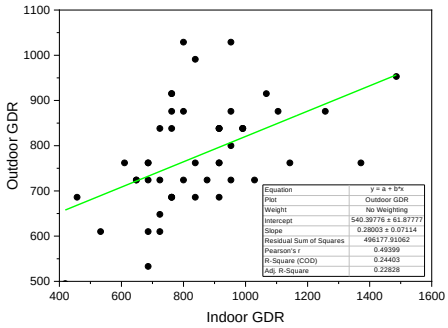

Figure 3.9. Plot of graph between indoor and outdoor GDR

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);

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

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

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).

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

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 $\times 10^{-2}$ 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

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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.

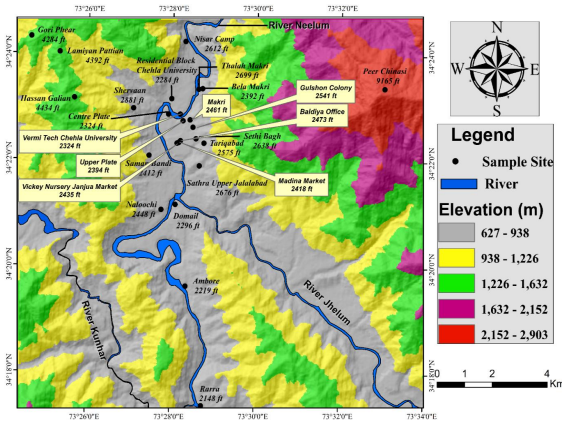

Figure 3.10. Samples collection sites in Muzaffarabad, Azad Kashmir

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).

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

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:

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

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:

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

$$\text{Ơ} = \frac{\sqrt{sample\ count\ rate + background count\ rate}}{t} \times 2 \times \frac{1}{\varepsilon \times 60 \times m} \qquad (3.12)$$

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).