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Natural and Fall out Radionuclide Concentrations in Medicinal Plants: An Overview

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Abstract: Almost food of all kinds contains radionuclides in varying amount depending largely upon local geology, agricultural practices and climate of the area. Usually these radionuclides are transferred from soils to the crops and water to fish. Plants take up radioactive material contents with the nutrients needed for their growth. On consumption, these products may expose peoples to unwanted radiation. For this purpose, it is appropriate to know the Radionuclides concentrations in food and drinking water and to take necessary actions in controlling their distributions. This review article presents overview of the medicinal plants Radionuclides concentration data obtained across the globe in different studies. 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.
Keywords: Average Annual Committed Effective Dose; local geology; medicinal plants; Radionuclides; Transfer Factor

1 Introduction

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 back ground radiation sources or occupational exposure of personals working in radiation field environment needs to be assessed and, if necessary, should be controlled [1]. These radiation exposures may come from; I). contamination 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. IIIa). from natural radioactive sources like, ^{222}Rn, ^{220}Rn coming from ^{238}U and ^{232}Th decay chains. IIIb). Radionuclides having natural origin. IIIc). Materials, in which the radionuclide activity concentration coming from either the uranium or the thorium decay chain does not exceed 1 Bq/g and the activity concentration from radioactive ^{40}K does not exceed 10 Bq/g. IV). Exposure from cosmic radiations [1].

Wealth of data is available in literature reporting activity concentrations of Radionuclides in environmental and building material samples across the globe [2-12]. 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 [13-57].

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 [58, 59].

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In earliest history of humanity, ancient methods of utilizing medicinal plants and plant extracts have been employed to treat several diseases and ailments [13]. Numerous traditional systems of medicines have been employed therapeutically medicinal plants due to their significant aspect acknowledged world over. Several cultural and theoretical models formulate phytotherapy within doctrine scheme subjected to all traditional system like Amazonian to African medicinal system, Unani to Tibetan, and Ayurveda to Chinese traditional system of medicine. All such system practiced regularly and employed whole plant or parts like core ingredients of medicines. Radionuclides spontaneously exhibiting within medicinal plants are one sort of renowned residue and contaminants that might subject impairment to herbal medicinal consumers [14]. 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 [15]. 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 [16-21]. Therapeutic plants were investigated so as to depict active compound subsisting within medicinal plants to explore therapeutic features on scientific basis [22-28]. 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 [29].

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 [30]. This article reports radionuclide data obtained by several research groups in numerous parts of the world to aid as future reference database. Main focus of current study is to summarize radionuclide data obtained from medicinal plants and to asses resulting radiological doses.

2 Area of Study

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 in this article. 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.

3 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 LR-115 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.

4 Results and Discussion

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 [31]. 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 [31]. 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 [31].

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 [32].

To improve the trustworthiness of predictive dose assessments from anthropogenic Radionuclides and to address resettlement and possible option of rehabilitation of Marshall Islands the Lawrence Livermore National Laboratory (LLNL) has developed an interactive internet application. This open access computer application has provided public a chance to assess radiological conditions in the Marshall Islands. User can calculate hypothetical ingestion doses from ^{137}Cs presence in food plants using the application of the ingestion dose calculator [33].

Dufy *et al.*1999 [34] 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}).

Salamon and Haban 2005 [35] assessed some medicinal plants of Slovakiafor 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.

In 2007, Narayana *et al.* [36] investigated ayurvedic 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.

Turhan *et al.* 2007 [37] have reported radionuclide concentrations for edible mushrooms of Turkey using gamma ray spectroscopy carried out by HPGe detector. Concentrations of ^{232}Th and ^{228}Ra were reported below lower limit of detection. Activity concentrations of ^{40}K and ^{137}Cs varied from 715.5 ± 50.1 to 1779.0 ± 163.7 Bq kg^{-1} with mean value of 1150.8 ± 315.2 Bq kg^{-1} (dry matter) and 2.4 ± 0.3 -109.0 ± 7.3 Bq kg^{-1} with mean value of 28.4 ± 27.2 Bq kg^{-1} (dry matter). The mean annual effective doses due to ^{40}K and ^{137}Cs from mushrooms were found to be (0.13 ± 0.03) μSv and (7.0 ± 6.0) × 10^{-3}μSv respectively. The plants intake of ^{137}Cs was found quite low and no significant contamination was recorded for the mushroom species of local origin. Morchellaesculenta and Strophariacoronilla plant species were found with having comparatively higher contents of ^{137}Cs and ^{40}K among the all analyzed mushroom samples.

Sussa *et al.* 2009 [38] 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.

Ahmed *et al.* 2010 [39] 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 ± 0.42Bqkg^{-1}.

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

Jevremovic *et al.* 2011 [41] 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 (2.5-469.9) 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.

Oni *et al.* 2011 [42] 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.

Sussa *et al.*2011/2013 [43, 44] 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.

Oufni *et al.* 2013 [45] 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 Bq.kg^{-1} to 6.20 ± 0.47 Bq.kg^{-1} and from 30 ± 2.30 mBq.kg^{-1} to 195±16 mBq.kg^{-1}, respectively. Higher values were reported for roots of studied plants as compared to stem and leaves.

Tettey-Larbi *et al.* 2013 [46] 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 [30].

In 2014, Oprea *et al.* [47] 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.

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

In 2015, Kavocas *et al.* [48] 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) 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 were reported for wild grown samples as compared to cultivated medicinal herbs (0.4-1.6)Bq/kg.

In 2015, Pourimani *et al.* [49] 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 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.

Najam *et al.*2015 [50] 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 Bqkg^{-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.

Njinga *et al.* 2015 [51] conducted preliminary research on medicinal plants of Nigeria for investigating Radionuclides concentration using NaI(TI) detector. He reported ^{40}K activity concentration in medicinal plants varying from (74.59 ± 2.19 to 324.18 ± 8.69)Bq kg^{-1} with average value of (324.18 ± 8.69) Bq kg^{-1}. Highest ^{40}K activity concentration was reported for A. indica whilst lowest for A. occidentale. Activity concentrations of ^{226}Ra varied from (10.79 ± 4.24−42.47 ± 2.76)Bq kg^{-1} with average value of (25.02 ± 3.18) Bq kg^{-1}. Lowest and highest activity was recorded for P. guajava and V. paradoxa herbal samples respectively. The activity concentration of^{232}Th varied from (27.76 ± 1.02 − 41.05 ± 1.05) Bq kg^{-1}with average value of (35.09 ± 0.71) Bq kg^{-1}. Lowest and highest ^{232}Th activity was reported for V. paradoxa and T. catappa herbal plants respectively. Due to ingestions of naturally occurring radionuclides in herbal plants the average annual committed effective doses (AACED) received by public range from (4.26±0.50 to 6.86±0.44) ×10^{-3}mSv/yr with an average of (5.38±0.35) ×10^{-3} mSv/yr.

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/yr (UNSCEAR 2000 report) showing insignificant contribution to radiological health risk by Radionuclides found in herbal plants [30].

Shatha *et al.* 2015, [52] 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.

Harb 2015 [53] 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 $mSvy^{-1}$ with mean value of 0.02 $mSvy^{-1}$.

Chandrashekara *et al.* 2015 [54] reported ^{226}Ra, ^{210}Pb, ^{232}Th, ^{40}K and ^{137}Cs activity concentration for the medicinal plants Justicaadhatoda L., CareyaarboreaRoxb., 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) $BqKg^{-1}$ 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) $Bqkg^{-1}$ respectively.

Chandrashekara and Somashekarappa in 2016 [55] 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) $Bqkg^{-1}$ 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) $mSvy^{-1}$. These doses were much less than the world accepted dose standards.

Kareem *et al.* 2016, [56] 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.

Abojassim *et al.* 2016 [57] 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 53.30Bq/m^3 within medicinal plants respective to average 26.53Bq/m^3 value. Their findings showed consumption of medicinal herbs pose insignificant health risk due to radon.

Kranrod *et al.* 2016, [58] 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 $Bqkg^{-1}$ 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 $mSvy^{-1}$ with average value of 0.0060±0.0001 $mSvy^{-1}$. Consequently, Thai medicinal plants found to be safe in health perspective.

Falandysz *et al.* 2017 [59] 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 $Bqkg^{-1}$ 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) $Bqkg^{-1}$ 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.

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

Table 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	[31]
Radioactive content for ^{134}Cs and ^{137}Cs	HPGe detector	Medicinal plants	BanskaBystrica, Slovakia	[32]
Radioactivity content of ^{210}Po and gamma dosage rate	Electrochemical deposition and portable scintillator	Ayurvedic medicinal plant	Moodabidri nearby Mangalore	[36]
^{232}Th, ^{228}Ra, ^{137}Cs and ^{40}K content	HPGe spectrometer	Medicinal Plants	Turkey	[37]
Activity of ^{226}Ra, ^{210}Pb and ^{228}Ra	Alpha as well as beta counting	Medicinal Plants	Brazil	[38]
^{226}Ra, ^{137}Cs, ^{232}Th and ^{40}K content	HPGe spectrometer	Herbal plants	Egypt	[39]
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	[40]
Radioactivity content of ^{238}U, ^{40}K, ^{232}Th, ^{137}Cs and annual whole-body dosage	HighPurity Germanium detector	Medicinal Herbs	Serbia	[41]
Radioactive contents for ^{238}U, ^{40}K and ^{232}Th	NaI(Tl) detector	Medicinal Plants	Nigeria	[42]
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	[43,44]
Radon as well as Thoron level	CR-39 and LR-115 type-II	Medicinal plants	Morocco	[45]
Annual committed effective dosages and ^{238}U, ^{40}K, ^{232}Th	HPGe detector	Medicinal Plants	Ghana	[46]
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	[47]
Radioactivity content of ^{226}Ra, ^{210}Po, ^{137}Cs, ^{210}Pb and ^{40}K	HPGe spectrometer and alpha spectrometry	Medicinal herbs	Hungary	[48]
Radioactivity content of ^{226}Ra, ^{40}K, ^{232}Th, ^{137}Cs and internal as well as external hazards indices	HighPurity Germanium detector	Medicinal Plants	Shazand, Iran	[49]
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	[50]
Radioactivity content of ^{226}Ra, ^{40}K, ^{232}Th and annual effective dosages	NaI detector	Medicinal Plants	Nigeria	[51]
Radioactivity content of ^{40}K, ^{228}Ra, ^{226}Ra and heavy metallic content	HPGe spectrometer	Medicinal plants	Jordon	[52]
Annual committed effective dosages and radioactivity content of ^{226}Ra, ^{40}K, ^{228}Ra	HPGe detector	Medicinal Plants	Qena,Upper Egypt	[53]

^{226}Ra,^{40}K, ^{232}Th, ^{137}Cs,^{210}Pb contents	HPGe spectrometer	Medicinal Plants	India	[54]
Annual effective dosages and radioactivity content of ^{40}K, ^{210}Pb,^{226}Ra, ^{232}Th	HPGe detector	Medicinal Plants	South India	[55]
Activity content of ^{40}K, ^{238}U, ^{232}Th, ^{226}Ra and internal hazardous index	NaI detector	Medical Plants	Iraq	[56]
^{222}Rn Content	CR-39	Medicinal Plants	(Al-Najaf) Iraq	[57]
Radioactivity content of ^{226}Ra, ^{40}K, ^{228}Ra and Annual effective dosages	HPGe detector	Medicinal herbs	Thailand	[58]
Radioactivity content of ^{137}Cs, ^{40}K	HPGe spectrometer	Medicinal plant	SW china	[59]

Table 2: Radioactivity contents (Bq/kg) subsisting within medicinal plants via applying various techniques.

Year of Study	Country	^{40}K	^{137}Cs	^{230}Th	^{232}Th	^{234}U	^{238}U	^{226}Ra	^{228}Ra	^{210}Po	^{210}Pb	^{222}Rn	References
1999	(Marshall Islands) SW Hawaii	-	$(0.001\text{-}1)\times10^{3}$, Polypodiumscolopendria $(0.200\text{-}3)\times10^{3}$	-	-	-	-	-	-	-	-	-	[31]
2007	Slovakia	-	0.400-3.200	-	-	-	-	-	-	-	-	-	[32]
2007	Moodabidri nearby Manglore	-	-	-	-	-	-	-	-	6.3-56.9	-	-	[36]
2007	Turkey	715.5±50.1-1779.0±163.7	2.4±0.3-109.0±7.3	-	<BDL	-	-	<BDL	-	-	-	-	[37]
2009	Brazil	-	-	-	-	-	-	29±3-65±4	<2.2-18.4±0.2	-	32±3-76±8	-	[38]
2010	Egypt	-	MDL-12.62±0.42	-	-	-	-	-	7.71±0.25-15.08±0.49	-	-	-	[39]
2010	Italy	66.2-3582.0	-	-	<0.3-10.7	-	<0.1-7.32	-	-	<0.12-30.3	<3-58.3	-	[40]
2011	Serbia	126-12437	0.3-8.8	-	1.7-15.1	-	0.6-8.2	-	-	-	-	-	[41]
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)	-	-	-	-	-	[42]
2011/ 2013	Brazil	-	-	2.1-38	1.7-124	42-129	4.3-38	8.5-37	3.2-46	-	39-93	-	[43,44]
2013	Morocoo	-	-	-	-	-	-	-	-	-	-	0.87±0.06-6.20 ±0.47	[45]
2013	Ghana	839.8±11.9	-	-	56.2±2.3	-	3.18±2.8	-	-	-	-	-	[46]
2014	Romania	-	-	-	<DL	-	40×10^{-3}	-	60×10^{-3}	8×10^{-3}	30×10^{-3}	-	[47]
2015	Hungary	-	10-15(leafy Parts), ≤2(Flowering Parts)	-	-	-	-	-	-	0.4-20	-	-	[48]
2015	(Shazand) Iran	MDA-$(2.75\pm0.01)\times10^{-3}$	MDA-1.02±0.35	-	MDA-7.79±1.40	-	-	-	MDA-7.43±0.60	-	-	-	[49]

2015	Iraq	88.3(Chammile),124.1(Crust)	-	-	-	-	-	-	-	-	-	-	[50]
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	-	-	-	[51]
2015	Jordon	24±1.6-2034±57	-	-	1.47±0.5-15.33±0.1	-	-	0.26±0.46-15.6±0.46	-	-	-	-	[52]
2015	Egypt	140±6-1538±54	-	-	-	-	-	0.4±0.2-21.0±1.2	<0.3-42.3±5.9	-	-	-	[53]
2015	India	443.50-3401.29	BDL	-	BDL-6.40	-	-	BDL-9.59	-	-	9.07-320.34	-	[54]
2016	South India	93.79-6831.40	-	-	2.42-8.72	-	-	-	2.66-11.27	-	BDL-87.03	-	[55]
2016	Iraq	-	-	-	-	-	-	-	-	-	-	(10.6602.07−53.3034.64) $\times 10^3$	[56]
2016	(Al-Najaf) Iraq	219.134 ±2.24	-	-	2.916 ±0.12	-	4.953±0.37	-	-	-	-	-	[57]
2017	Thailand	159.42-1216.25	-	-	-	-	-	0.20-6.67	0.10-9.69	-	-	-	[58]
2017	SW China	420± 41-1300± 140	<3.8- 83±3	-	-	-	-	-	-	-	-	-	[59]

DL: Detection limit, BDL: Below Detection Limit, MDA: Minimum Detectable Activity 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 1 and Table 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 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 Bq kg^{-1}, was reported for Brazilian medicinal herbs and lowest value, below detection limit, has been reported for Turkey herbs.

For anthropogenic Radionuclides, higher activity concentrations of ^{137}Cs (2.4 ± 0.3 - 109.0 ± 7.3) $Bqkg^{-1}$ are found in literature for Turkey herbal species and smallest were found (0.400-3.200) $Bqkg^{-1}$ within Slovakian and Indian medicinal plant. ^{40}K maximum contents (93.79-6831.40) $Bqkg^{-1}$ were reported within medicinal plants of South India and lowest (24± 1.6) $Bqkg^{-1}$ 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, (BDL-87.03) $Bqkg^{-1}$, were reported for medicinal plant of South India. Highest values of Polonium contents (<0.12-30.3) $Bqkg^{-1}$ were found for the medicinal plants of Italy while lowest, ≤ 2 $Bqkg^{-1}$, for Hungary medicinal flowers. Nigerian medicinal plants found to have highest uranium contents (69.3 ± 9.4) $Bqkg^{-1}$ whilst, smallest (<0.1-7.32) $Bqkg^{-1}$ are found within medicinal plants of Italy. Brazilian medicinal herbs are reported for highest thorium contents (1.7-124) $Bqkg^{-1}$ and smallest, < BDL, are reported within medicinal plant of Turkey and Romania.

5 Conclusions

To conclude, this article 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.

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