Estimation of Radioactivity Levels in Biochar Amended Soilless Growth Media Collected from Agricultural Research Site in Quincy, Florida

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Abstract

Many organisms on earth are exposed to radiation, both natural and anthropogenic. Research to date indicates varying levels of radionuclides in agroecosystems, particularly in soil, plants and water bodies. Soil amended with biochar has been proposed to enhance long-term atmospheric carbon sequestration and plant productivity. However, little is known about possible radionuclides (natural and anthropogenic) in soilless media with biochar amendments. Soilless media amended with different levels of biochar were sampled from Florida Agricultural and Mechanical University Research and Extension Center (FAMU-REC) in Quincy, Florida, to assess possible radionuclides activities using gamma spectrometry. The mean activity concentration values for ^{235}U, ^{226}Ra, ^{232}Th, ^{40}K, *and* ^{137}Cs *found in the samples were 0.92±0.02, 3.32±0.19, 1.35±0.46, 22.1±053 and 0.36±0.04 Bq* kg^{-1}, *respectively. The estimated external* (H_{ex}) *and internal* (H_{in}) *radiation hazard mean values were 0.02 and 0.03, respectively. The results are within estimated safe radiological limits for plant growing media.*

Keywords: radioactivity, radionuclides, soilless growth media, biochar, pyrolysis, containerization, anthropogenic

Introduction

Many organisms are exposed to both natural and anthropogenic radiation. The presence of radionuclides in the environment poses risk to human health and the environment(Sunovska et al., 2012). Evaluation of radionuclides in environmental studies is important to better understand the distribution of radionuclides in different media in order to establish environmental radioactivity baseline levels for various media in the environment (Jordan et al. 1997).

Extensive research has been conducted on radioactivity levels in agroecosystems relating to soils, plants, and water (Yamaguchiet al., 2016; Guidottiet al., 2015; Uosif et al., 2014; Gulinet al., 2013; Ohno et al., 2012; Yasunari et al., 2011; Zarie& Al Mugren, 2010; Al-Hamarneh&Awadallah, 2009; Al-Kharouf et al., 2008; Bolcaet al., 2007; Papastefanouet al., 2006; Akhtar et al., 2005).

The perceived limits to producing food for the growing global population which according to the United Nations is currently at 7.6 billion, projected to be 8.6 billion in 2030, 9.8 billion in 2050 and 11.2 billion in 2100, has been a source of debate and preoccupations for ages (UN DESA, 2017; Alexandratos&Bruinsma, 2012). Global population growth impacts of climate change and food insecurity have led to significant shift in agricultural practices over the past decade (FAO, IFAD, UNICEF, WFP and WHO, 2018; Thrall et al., 2010). A global agricultural technique, currently gaining significant attention that could increase production yields, is greenhouse cultivation (Mendez et al., 2015).

Soilless systems, which involve containerization of plant roots within a porous rooting medium, substrate or growing medium, can be defined as any method of growing plants without the use of traditional soil as the rooting medium. It entails the use of soilless system as plant growth medium without using the traditional soil as the rooting medium (Barret et al., 2016; Savvas et al., 2013) is now attracting attention globally. Soilless media have been reported to produce higher yields and bumper harvests from smaller area, higher water and nutrients use efficiency, and more cost effective as compared to traditional soil system (RezaiNejad and Ismaili, 2014).

The charred organic matter that remains after pyrolysis of biomass or manure, known as biochar, has been reported as one of the very few technologies that can actively sequester carbon (C) from the atmosphere when amended with soil in agricultural cropping practices (Prasad et al., 2018; Lehmann and Joseph, 2015; Altland and Locke, 2012). Amending soilless media with different levels of biochar have been shown to increase plant growth. However, soilless-biochar amendment system has only recently been gaining increasing attention worldwide (Huang and Gu, 2019; Mendez et al., 2017).

The available research suggests that work on possible radionuclides (natural and anthropogenic) in soilless media with biochar amendments is very limited. This study was conducted on soilless media comprising a mixture of coconut fiber (coir) and fine pine bark amended with different levels of biochar. Samples were collected from Florida Agricultural and Mechanical University Agricultural Extension farm in Quincy, Florida, located about 18 miles west from Tallahassee, Florida, to estimate the presence of possible radionuclides. Measurements of radioactivity levels of the samples were performed via gamma spectroscopic techniques using high purified germanium detector (HPGe). The findings will provide information on the presence and concentration of radionuclides in soilless media amended with biochar.

Materials and methods

2.1 Geographical Location of the Research Site

The study site was Florida Agricultural and Mechanical University (FAMU) Research and Extension Center (REC) located in Quincy, in North Florida District of Gadsden County near the Florida-Georgia States line (30^{o}67'N and 84^{o}61'W). The research site is about 30 miles from the main campus of FAMU, Tallahassee, the State's capital. FAMU Research and Extension Center (FAMU-REC) is located on the more than 200 acres of farms, pines, lake, animal research laboratories and other facilities. It has an annual high and low temperatures of 26.11^{o}C (79.0^{o}F) and 12.94^{o}C (55.3^{o}F), respectively, with average temperature of about 19.53^{o}C (67.15^{o}F).

The annual precipitation is around 59.67 inches and humidity level also around 94%. The research area is also in the Panhandle of Florida with sandy loam soil (FL039) (Thomas et al., 1961). FAMU-REC farming program was developed to assist and equip underserved agricultural populations, including small farmers, farm laborers and their families to attain sustainable lifestyles. It also provides access to agricultural sustainability production and management systems to better equip small growers through educational and hand-on training.

2.2Sampling

A total of 27 samples were collected from 9 treatments, three composited samples from each treatment. The collected samples from FAMU-REC were stored in labelled plastic ziplock bags and transported on ice in cooler to FAMU main campus School of the Environment laboratory. Collected samples were of composited triplicates from 9 treatments of growth media comprising control (0%), control + 1%, + 2%, + 3%, +4%, +6%, +8%, +10% and +12% biochar amendments. For each treatment 3 composited containerized pots content were collected and placed into a cement mixer for homogeneity. While the mixer was running, roots and other debris in the samples were removed. 2 cup size good cook plastic cup was used to scooped about 450g of samples into the labelled ziplock bags.

The process was repeated for the remaining treatments rolls before the samples were transported to the lab. In the laboratory, subsamples were stored in refrigerator at 4 ^{o}C prior to analysis.

In the laboratory, subsamples were oven-dried at 105^{o} C to constant mass for moisture content analysis (Jackson, 1967) and sample pH was determined with 1 g fresh sample in 20 ml of deionized water (DI) shaken for 1.5 hours and left for 5 mins equilibration time before pH measurement with Fisher Scientific Accument Basic AB15 pH meter (Ngatia et al., 2017; Rajkovich et al., 2012; Yao et al., 2012) prior to samples preparation for radionuclides analyses. Uptake, retention,and distribution of radionuclides in plants are impacted by media characterization and properties such as organic matter content, pH, soil amendments and nutrient composition (Bolca et al., 2007; Pulhani et al., 2005).
Samples were prepared for radionuclides analysis by transferring into 500 ml Marinelli beakers, covered, and sealed with parafilm to limit any possible escape of radon. The prepared samples were left for at least 30 days to reach secular equilibrium with radon and its daughters.Samples were handled carefully, and proper measures were taken by changing gloves during sample preparation to minimize cross contamination.

2.3Analysis

Gamma spectrometry analyses was performed on the samples using a high purity germanium detector (HPGe) manufactured by Canberra industries to determine the activity concentrations of the gamma emitters' radionuclides. The detector is shielded with a thick lead shield with Cu inner layer. The active shield reduces the integral background. A pre-amplifier and amplifier spectra channeled to multichannel analyzer (MCA) with two digital converters were connected directly with a PC equipped with Canberra Genie 2000 software in which measured gamma spectra were stored and analyzed. The software internally calculates activity concentrations of radionuclides from all prominent gamma lines with background subtraction (Bikit et al., 2011). The instrument has an energy resolution of 0.5keV full width at half of maximum (FWHM) for a 1332 keV channel (using of Co-60) and a relative photo peak efficiency of 35%. The instrument was calibrated for energy and efficiency over the photon energy range of 2 to 2000 keV using a National Institute of Standards and Technology (NIST) traceable mixed gamma standard. Each sample was counted for a period of 86400s.

2.3^{235}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs

The activity concentrations of the natural radionuclides^{235}U and ^{40}K, and the anthropogenic radionuclide ^{137}Cs were determined directly from their photopeak energies lines of 185.7 (54.0%) 1460.8 (10.7%), and 661.7 (85.1%) keV, respectively. The weighted mean photopeakenergy lines of^{214}Pb (295.2 and 351.9) and^{214}Bi (609.3 and 1120.3) were used to estimate the activity concentration value of ^{226}Ra.The weighted mean photo peaks energies lines of^{212}Pb (238.6),^{212}Bi (727.2) and ^{228}Ac (338.3, 911.6 and 969.1) were used to determine the activity concentration value of ^{232}Th (Khol'nov et al., 1982;Alnour et al., 2012).Using the weighted meanphotopeaks procedure for multiple energy lines gives more accurate results with lower errors comparedto using only one of the photopeak line (Papp et al., 1997). The measured and estimated activity concentrations values of the radionuclides reported in Bqkg^{-1} are presented in Table 2.

Table 1: Physical and Chemical Properties of Media from FAMU-CE Media

ID	pH	MC (%)	OM (%)
0%	6.15±0.03	69.74	73.29
1%	6.02±0.01	70.27	71.93
2%	6.03±0.06	70.83	70.54
3%	6.23±0.09	72.38	72.70
4%	6.47±0.05	71.28	71.95
6%	6.46±0.02	70.93	74.17
8%	6.68±0.02	70.71	71.74
10%	6.76±0.01	69.57	70.89
12%	6.84±0.02	67.05	70.56

Table 2: Activity Concentration, A, of Radionuclides (Bq kg^{-1}) from FAMU-CE Media

ID	^{235}U	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
0%	0.92±0.05	2.93±0.12	1.01±0.04	0.00	0.21±0.02
1%	0.92±0.05	2.54±0.11	0.68±0.08	0.00	0.19±0.02
2%	0.99±0.05	2.94±0.12	1.02±0.11	22.54±0.54	0.22±0.02
3%	0.94±0.05	3.10±0.13	4.98±0.18	21.10±0.52	0.39±0.03
4%	0.92±0.05	3.78±0.14	0.57±0.09	21.37±0.53	0.37±0.03
6%	0.81±0.04	2.89±0.13	0.90±0.04	19.57±0.51	0.39±0.03
8%	0.90±0.05	3.63±0.14	0.88±0.10	22.18±0.54	0.48±0.03
10%	0.96±0.05	3.93±0.14	1.05±0.10	24.23±0.57	0.45±0.02
12%	0.94±0.10	4.14±0.29	1.02±0.22	23.70±1.13	0.55±0.06

2.4 Radiological Hazard Indices

2.4.1 Radium Equivalent activity

To compare the specific activity and the radiation hazard associated with the samples with respect to the natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K), the widely used radiation hazard index, the radium-equivalent activity, (Ra_{eq}), as expressed by Fonseca and Pecequilo (2015); Beretka and Matthew (1985); UNSCEAR (1982) was used; Equation 1.

$$Ra_{eq} = 370\left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{C_K}{4810}\right) \quad \text{Eqn} \quad (1)$$

Eqn (1) can be rewritten as Eqn (2)

$$Ra_{eq}\,(Bqkg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \text{Eqn} \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Ra_{eq} is the weighted sum of the afore mentioned natural radionuclides. The relation implies that 370 or 1 Bq kg^{-1} of ^{226}Ra, 259 or 1.43 Bq kg^{-1} of ^{232}Th and 4810 or 0.077 Bq kg^{-1} of ^{40}K produce the same γ-ray dose rate (Alzubaidi et al., 2016).

2.4.2Absorbed Dose Rate

Depending on the radionuclides in the sample, a parameter used to assess radiation exposure and radiological hazard from radionuclides in media, the absorbed dose rate (D_R) was also estimated (Alzubaidi et al., 2016). Thisγ-ray absorbed dose rates (D_R) wasestimated using UNSCEAR (2000) conversion factors as stated in equation (3) (Hamidalddin, 2014; Veiga et al., 2006; UNSCEAR, 2000).

$$D_R(nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad \text{Eqn} \quad (3),$$

where D_R is dose rate at 1 m above ground, A_{Ra}, A_{Th} and A_K are the calculated activity concentrations of ^{226}Ra,^{232}Th and ^{40}K in Bq kg^{-1}from the measured samples, respectively.

2.4.3Annual effective dose

The annual effective dose which considers the absorbed dose rate (D_R), the conversion coefficient of 0.7(Sv Gy^{-1}) and the average outdoor spent time fraction of 0.2 (UNSCER, 2000) was calculated using equation (4):

$$AED(\mu Svy^{-1}) = D_R(nGyh^{-1}) \times 8766\,hy^{-1} \times 0.2 \times 0.7(SvGy^{-1}) \times 10^{-3} \quad \text{Eqn} \quad (4)$$

2.4.4External and Internal hazard index

Other criteria used to limit natural radiation exposure to the population under the external hazard index (H_{ex}) and the internal hazard index (H_{in}) which describes the radon and its progeny risk to internal organs (Saleh and Shayeb, 2014) were also estimated in the study. For safety requirement, H_{in} is used for purposes of reducing ^{226}Ra acceptable activity concentration to half the normal limit of less than or equal to unity (Beretka and Mathew, 1985). The relations of H_{ex} and H_{in} are expressed in equations (5) and (6), respectively.

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} = \leq 1 \quad \text{Eqn} \quad (5)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad \text{Eqn} \quad (6)$$

Summary of the estimated values of $Ra_{eq}(Bqkg^{-1})$,$D_R(nGyh^{-1})$,$AED(\mu Svy^{-1})$, H_{ex} and H_{in} are reported in Table 3.

Table 3: Estimated Values of Ra_{eq} (Bq kg^{-1}), D_R *(nGy h^{-1}), AED (μSv y^{-1})*, H_{ex} (≤1), and H_{in} from the measured samples from FAMU-REC

ID	Ra_{eq}	D_R	AED	H_{ex}	H_{in}
R0	4.37	1.96	2.41	0.01	0.02
R1	3.52	1.59	1.95	0.01	0.02
R2	6.12	2.91	3.57	0.02	0.02
R3	11.85	5.32	6.53	0.03	0.04
R4	6.24	2.98	3.66	0.02	0.03
R6	5.69	2.70	3.31	0.02	0.02
R8	6.60	3.14	3.85	0.02	0.03
R10	7.30	3.46	4.25	0.02	0.03
R12	7.42	3.51	4.31	0.02	0.03

3.0 Results and Discussion

3.1 Natural radionuclides

The main aim of this study was to evaluate the possible presence of radioactivity levels in growth media at FAMU-REC and establish baseline data for the local communities, and Florida as a whole. The media samples activity concentration values were expressed as mean values (Bq kg^{-1}). The main gamma emitting natural and anthropogenic radionuclides activity concentrations (Bqkg^{-1}) measured from the media samples are shown in Table 2. The activity concentrations of the measured radionuclides ranged from 0.81±0.04 to 0.99±0.05, 2.89±0.13 to 4.14±0.29, 0.68±0.08 to 4.98±0.18and 19.57±0.51 to 24.23±0.57 Bq kg^{-1} for ^{235}U, ^{226}Ra, ^{232}Th and^{40}K [13], respectively. The mean activity concentrations for both natural and anthropogenic radionuclides, ^{235}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs, for the media samples analyzed were 0.92, 2.99, 1.25, 22.10 and 0.36Bq kg^{-1}, respectively. ^{40}K was not identified in 1% and 2% media biochar amendments (Table 2). The distributions of the radionuclides measured in the samples are represented in Figure 1.

The estimated ^{226}Ra, ^{232}Th and ^{40}K activity concentrations values of the analyzed samples were compared to similar studies of soil worldwide. The world ^{226}Ra, ^{232}Th and ^{40}K activity concentrations values (Bq kg^{-1}) ranged between 17 – 60, 11 – 64, and 140 – 850, with mean activity concentration values (Bq kg^{-1}) of 35, 30 and 400, respectively, according to UNSCEAR (2000). It was determined from this study that all the estimated and mean activity concentration values were below the world and United States values as shown in Table 4.

Table 4: Comparison of ^{226}Ra, ^{232}Th and ^{40}K activity concentrations levels around the world in Bq kg^{-1}.

Country	226Ra		232Th		40K		Reference	
	Ranged	Mean	Ranged	Mean	Ranged	Mean		
U.S.A	8 160	40	4 – 130	35	100 700	170	Myrick et al., 1983	
Ghana	10.72-40.74	23.8	20.22-76.66	43.64	80.55-245.44	199.69	Adjirackor et al., 2014	
Malaysia	7–222	57	10–158	68	104–1225	427	Almayahi et al., 2012	
Jordan	47.3-77.8	57.7	14.0-20.8	18.1	31.3-251.5	138.1	Saleh & Abu Shayeb, 2014	
Worldwide	17 - 60	35	11 – 64	30	140 - 850	400	UNSCEAR, 2000	
Present Study	2.54 -4.14	3.32	0.57 – 4.98	1.35	19.57-24.23	17.19	Osei et al., 2019	

The highest activity concentration values of 4.14±0.29 Bq kg^{-1} and 0.55±0.06 Bq kg^{-1} for ^{226}Ra and ^{137}Cs, respectively, were found in sample 12% media biochar amendment. The highest value of 24.23±0.57 Bq kg^{-1} activity concentration ^{40}K was detected in sample 10%, with no detection in samples 0% and 1% media biochar amendments. The mean Ra_{eq} of 6.57 Bq kg^{-1} in the present study was lower than recommended maximum radium equivalent level of 370 Bq kg^{-1} in soil (Almayahi et al., 2012). These levels are below what is considered suitable for the growth of agricultural plants.

The studied absorbed dose rate ranged from 1.59 to 5.32 $nGy\ h^{-1}$ with a mean value of 3.06 $nGy\ h^{-1}$, as compared with the world ranged and mean of 18 to 93 and 57 $nGy\ h^{-1}$, respectively (UNSCEAR, 2000). The annual effective dose of samples was determined to range between 1.93 and 6.53 $\mu Sv\ y^{-1}$ with a mean of 3.76 $\mu Sv\ y^{-1}$. The external (H_{ex})and internal (H_{in}) radiation hazards in the present study ranged from 0.01 to 0.02 and 0.02 to 0.04 with means of 0.02 and 0.03, respectively. All the (H_{ex})and (H_{in}) values are less than unity, suggesting that the media are radiologically hazard free, as shown in Table 3. The distributions of the radionuclides measured in the samples are represented in Figure 1.

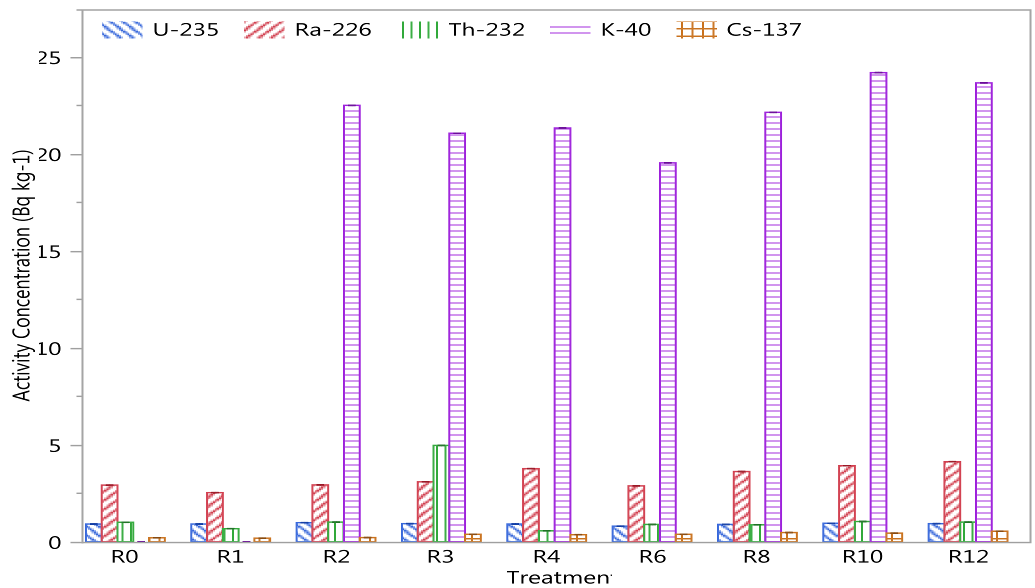

Figure 1: Distribution of radionuclides within different treatments.

3.2 Anthropogenic radionuclide (^{137}Cs)

^{137}Cs in the environment has historically been the result of the 1945 to 1980, 1986 nuclear weapon testing in the atmosphere, the Chernobyl accident and the catastrophic earthquake and tsunami which occurred in northeastern

Japan causing a severe destruction at the Fukushima Daichi Nuclear Power Plant in March 2011, respectively, (Yasunari et al., 2011). It was reported that by the end of 1970s, approximately 5.2 kBq m^{-2} of ^{137}Cs deposit density was estimated values for 40^o-50^o of the North Latitudes (UNSCEAR, 2000 Annex C). The deposition density corrected for decay by September 2011 was 2.5 kBq m^{-2}(UNSCEAR, 2000 Annex C), and was approximately 2.48 kBq m^{-2} by September 2019. In the present study, ^{137}Cs was detected in all the treatments. The activity concentrations of ^{137}Cs in the present study ranged from 0.19±0.02to 0.55±0.06Bq kg^{-1}, with mean activity concentration of 0.36±0.04 Bq kg^{-1}, values less than levels in agricultural soil (Asiani et al., 2003).

4.0 Conclusion

The study established baseline data for the application of soilless media with various levels biochar amendments as plant growth media in radiological health perspective. The reported values (0.92±0.02, 3.32±0.19, 1.35±0.46, 22.1±053 and 0.36±0.04 Bq kg^{-1} for ^{235}U, ^{226}Ra, $^{23\ 2}$Th, ^{40}K, and ^{137}Cs, respectively) were significantly lower compared to world average and other reported literature (Table 4). Based on the results, it can be concluded that soilless media use for planting is a safe technique and does not increase radioactivity levels in agricultural practices. Similar research using different organic media and different levels of biochar amendments need to be conducted in the future to better to evaluate and validate the data in this study., It will also enhance our understanding of the activity concentration of the individual medium for applications and combinations of suitable media for plants growth. This research provides data that is useful for assessing possible radiation exposure to humans and contribute to the selection of the media. Even though the levels of the detected radioactivity were low compared to worldwide levels, accumulation in humans via media to plant transfer for long period may pose radiological risk.

Based on the findings of this study, it is recommended that additional work is needed in the following areas:

1) Understanding transfer factor of the detected radionuclides into food produced in this area; and
2) Possible radionuclides concentrations of fertilizers used in such management practices.

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