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RESEARCH ARTICLE

INVESTIGATIONS OF NATURAL RADIOACTIVITY LEVELS AND THE POSSIBLE RADIATION HAZARDS IN FLORICULTURE SOIL, HOLETA, SHOA, ETHIOPIA, USING GAMMA RAY SPECTROMETRY

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ABSTRACT

The aim of this study is to investigate the natural radioactivity levels and possible hazards in floriculture soils. The observed results were compared with the same study done for Virgin land and Agricultural farm land, from same study area, Holeta, Shoa, Ethiopia. The radioactivity concentration of ^{238}U , $^{232}\text{T h}$ and ^{40}K were determined using gamma-ray spectrometry with NaI (TI) detector. Moreover, the elemental concentrations, R_{eq} , D_{R} , AD_{R} , H_{ex} and H_{in} were calculated. The results were compared to international standards, soil-375 and soil-6 given by IAEA. The average values of ^{238}U , $^{232}\text{T h}$ and ^{40}K in Floriculture soil were found to be 142.29 ± 27.67 , 7.82 ± 0.54 and 259.62 ± 44.98 Bq/Kg respectively, in agricultural farm land these were found to be, 133.85 ± 20.49 , BDL and 287.82 ± 35.68 Bq/Kg respectively, and in virgin land. 30.51 ± 5.32 , 14.52 ± 1.695 and 100.29 ± 19.81 Bq/Kg respectively. The activity concentration of ^{238}U is increasing faster than the other NORM for soils exposed to artificial fertilizers and increasing rate is more for the case of floricultural soils. The hazardous measuring parameters were found to be maximum as compared with the virgin soils. This may be as a result of more phosphate fertilizers added to the floriculture soil.

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INTRODUCTION

Measurements of environmental natural radioactivity level will create an awareness for human beings to protect themselves from hazardous radiations. In most cases the levels are raised by the activities of human beings in different industrial areas. We found that floriculture industry is one of the like places in which much labor forces are invested and factors increasing the levels of natural radiations are frequently used. The industry had been using excess, several types of fertilizers fabricated from sedimentary rock by adding of sulphuric acid and nitric acid (Cioroianu Bunus *et al.*, 2000). By the processes, the two known types of fertilizers, triple super phosphate (TSP) and Di Ammonium Phosphate (DAP) can be produced. The natural phosphates from sedimentary represent about 85% of fertilizer, in which it contains natural radioactive materials (Mesay Adugna Kassa, 2017). Fertilizers usually employed in the agriculture contain traces of heavy metals and naturally occurring radionuclides, such as potassium, thorium and uranium with their decay products. Out of these radioactives, Uranium represent more concentration than the remaining natural radiation sources (Ione Makiko Yamazakia and Luiz Paulo Geraldo, 2003). In our study areas, the floriculture farm can be used more than two times per year.

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In each cycle they add more fertilizers, TSP and DAP, and chemicals to increase the productivity of flowers as we made interview with some of the employee. Since those fertilizers are coming with naturally existing radioactive materials, workers of the fields has the chance to be affected by radiations from the like fertilizers and chemicals. Some studies reports that floriculture industries have social and environmental impacts due to the used fertilizers and chemicals. In Sebeta Floriculture, 74.9% of the workers were females, with 93% of study subjects showing at least one health symptom. 67.8% had at least one skin problem and 81.1% had at least one respiratory health symptom in 12 months. The highly prevalent disease symptoms were also observed. These symptoms had been observed on those who did not have full personal protective equipment (Atkure Defar, Ahmed Ali, 2013). Many diseases such as Methemoglobinemia, Japanese encephalitis (JE), cancer etc, have been noted due to use of chemical fertilizers. Methemoglobinemia (blue-baby syndrome) occurs when the excess nitrates remain in the soil move into the ground water and used for drinking. This will also cause cancer, especially stomach cancer (Mulugeta Getu Sisay, 2009). Chemicals and fertilizers used for floriculture industries have a different character and react differently when they applied to the soil and affect its texture, acidic value, and fertility by destroying nitrogen-fixing bacteria and many other micro- and macro-organism of the soil (Mesay Adugna Kassa, 2017).

The floriculture industry were taken as a solution for economic development and the generation of employment, these advantages of the industry are at the expense of social and environmental disadvantage. The majority of workers in these farms are young women and easily affected by pesticide and fertilizers than men (Gudeta Degytnu Tilahun, 2012). According to the reports of studies above, the reported social and environmental symptoms of risks should be from the radiations and chemicals used in the fields. Specifically no report showing for specific disease from radiation. As the reports of many researchers, Naturally Occurring Radioactive Materials (NORM), in virgin lands are so weak that may not cause such serious problems. Therefore fertilizers are frontal sources in creating both social and environmental problems in farming lands. In this work, we are going to present the activity concentration and the possible health hazards of floriculture by using gamma Spectrometry. The samples were collected near Holeta town, West shoa, Ethiopia, from the oldest floriculture of the area. We also measured activities in soils from normal agricultural farming land and virgin land. Virgin land we considered is not exposed to fertilizers as far as we obtained information from residents. Finally we are going to compare the results of the three areas, and see the social and environmental effects of the NORM from floriculture field.

MATERIALS AND METHODS

Area of the Study: The study area is near Holeta town, located at 30Km to the western part of the capital city of the country, Addis Ababa. Globally it is covering 1 KM radius to the Lat. 9° , 04.2828° N and longtd. 38° , 31.135° E in a maximum inaccuracy of $\pm 8m$. The study area consists of fertile soil, basically local farmers have been using for the production of cereal crops. Farmers started using fertilizer since 1980s, as residents of the area speaks. In nature, the soil types in the study areas are the same and variations of concentrations of radioactive materials may not be expected to the exaggerated difference. But in order to get more products, the peoples are using some radioactive containing fertilizers and chemicals.

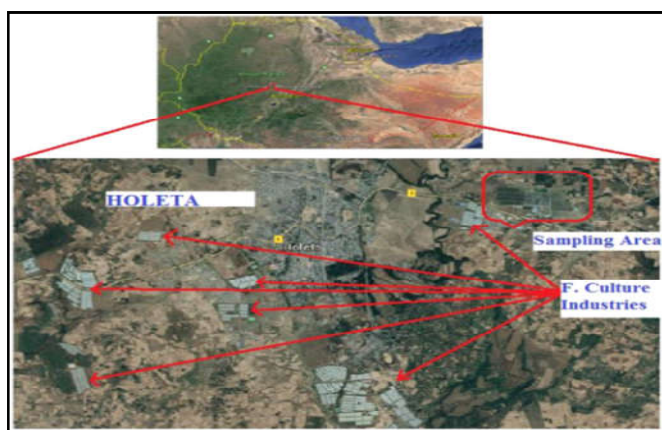


Figure 1. Holeta town and its surroundings referring the sampling areas and floriculture industries (Satellite image taken from Google map)

Sampling and Sample Preparation: A total of 30 soil samples have been collected which represent the predominant soil formations of study area. We collected 10 from floriculture, 10 from agriculture land and 10 from virgin land. In each sampling areas, we used surface sampling techniques to the maximum depth of 20cm. The 10 samples from virgin land were prepared in separate two beakers, where for

agriculture land also we did same things. Samples from floriculture were reduced from 10 to 5 sampling beakers and we had prepared a total of nine (9) sampling beakers. The nine sampling beakers were prepared after Oven drying of each sample by a temperature of $80 - 110^{\circ}C$. The dried samples were grinded to a fine powder, sieved through 0.075mm mesh sieve (ISO-9001 and ISO-2000 quality approved) to the optimum size enriched in heavy mineral for the reduction self-absorption. The final nine samples were packed in pvc plastic beakers of the shape of marinelli beakers, whose volume is 150ml. Then we weighed and stored for a minimum period of 28 days to allow the in-growth of uranium and thorium decay products and achievement of equilibrium with their respective progeny (Abd El-Hadi El-Kamel *et al.*, 2012, Mohammed Mahmud *et al.*, 2013). Finally the nine data were counted for 20-24 hours. For the analysis of spectrum from unknown samples, we used Soil-375 and Soil-6 given by IAEA as references. The two references were prepared and measured by same methodology as the collected soil samples.

Experimental Setup: Each sample was measured by gamma spectrometry with an FJ374 scintillation crystal cascaded with electronic circuits and Genie 2000 multichannel analyzer of 4096 channels. It is shielded in a chamber of two layers starting with the inner part stainless steel of 3 mm thick and lead of 45 mm thick to reduce background radiations (Gulcin Bilgici Cengiz, Asl Caglar, 2017). We fixed the detector in the center of the chamber to minimize the effect of the scattered radiation from the shielding materials. Then we kept the sample over the detector for 20-24 hours. The spectra were evaluated manually by comparing with the spectra from references given by IAEA listed above for the energies of 238 keV of Pb-212 for Th-232 identification, 351 keV of Pb-214 and 609 keV of Bi-214 for U-238 identification and 1460.9 keV gamma line for K-40 activity concentration (Faanu and Adukpo *et al.*, 2016).

Calculation of elemental concentrations: Following the spectrum analysis, for photo peaks representing Uranium, Thorium and Potassium, we calculated activity per mass unit and elemental (radioactive source) concentration in the sampled soils. The specific activity,

A_{Ei} (in Bq/kg) of a nuclide i for a peak at energy E , is given by;

$$A_{Ei} = \frac{\text{Net Peak Area}}{(\text{Live time})(\text{Efficiency})(\text{yield})(M_{\text{sample}})} \quad (1)$$

Where yield is the branching ratio fraction and live time is the actual counting time, Aide-de-camp (ADC) live time in seconds (Mirions Technologies, 2017). We also used this formula for the calculation of activities of reference soils. The geometry of sampling materials (beakers) are almost the same and we filled the prepared fine powder of samples in the same way. As it is already known, the activity of radioactive elements in reference soil was already known, and we deduct the current activity of sources from the date of issued by IAEA. After we found the current activities we used the comparative way to measure the activity of our samples.

The final equation should be;

$$A_{sEi} = A_{REi} * \frac{\text{Net peak Area}_{(\text{samples})}}{\text{Net peak Area}_{(\text{reference soils})}} * \frac{M_{\text{standard}}}{M_{\text{samples}}} \quad (2)$$

Where A_{sEi} is activity of photo peak from our sample (Michalis Tzortzis *et al.*, 2012; IAEA 1998/1999, Mirions Technologies, 2017). Based on the radiological concentrations we calculated the elemental concentrations of Uranium,

Table1. The activity concentration levels of natural radionuclide per Kg in samples from Virgin, Agricultural and Floriculture soil, near Holeta town

Sample Cite	Sample Code.	²³⁸ UBq/Kg	²³² ThBq/Kg	⁴⁰ KBq/Kg
Virgin Land	1	34.98 ± 4.60	12.21 ± 0.67	73.27 ± 14.65
	2	26.04 ± 2.68	16.83 ± 1.55	127.31 ± 13.34
Agricultural Land	3	137.50 ± 12.42	—	272.97 ± 23.53
	4	130.20 ± 16.30	—	302.67 ± 26.82
F.Culture Land	5	122.18 ± 3.44	—	282.44 ± 20.84
	6	83.80 ± 8.62	7.64 ± 0.45	274.24 ± 20.78
	7	169.43 ± 14.38	8.00 ± 0.29	360.40 ± 21.20
	8	138.84 ± 13.72	—	214.12 ± 17.13
	9	197.18 ± 16.88	—	166.89 ± 20.35

Table2. The elemental concentration of natural radioactive in samples from Virgin, Agricultural and Floriculture soil, near Holeta town

Sample Cite	S. Code	²³⁸ U Concent.(ppm)	²³² Th Concent.(ppm)	⁴⁰ K Concent.(%)
Virgin Land	1	2.83 ± 0.37	3.00 ± 0.16	0.23 ± 0.046
	2	2.11 ± 0.22	4.14 ± 0.38	0.40 ± 0.042
Agricultural Land	3	11.14 ± 1.01	—	0.86 ± 0.074
	4	10.55 ± 1.32	—	0.95 ± 0.084
F.Culture Land	5	9.90 ± 1.09	—	0.89 ± 0.065
	6	6.79 ± 0.70	1.88 ± 0.11	0.86 ± 0.065
	7	13.72 ± 1.16	1.97 ± 0.07	1.13 ± 0.066
	8	11.25 ± 1.11	—	0.67 ± 0.054
	9	15.97 ± 1.37	—	0.52 ± 0.064

Table 3. The radium equivalent (Ra_{eq}), absorbed doses (DR), Annual effective dose (ADR) the external (Hex) and the internal (Hin) hazard index of soil samples collected from Virgin, Agricultural and Floriculture near Holeta town

S. Code	Req in Bq Kg	DR in nGy/y	ADR in mSv/y	Hex	Hin
1	58.08 ± 2.86	25.69 ± 1.64	0.032	0.157	0.252
2	59.91 ± 2.36	27.07 ± 1.36	0.033	0.161	0.232
3	158.52 ± 12.55	70.45 ± 5.40	0.086	0.429	0.800
4	153.51 ± 16.43	68.61 ± 7.05	0.084	0.415	0.767
5	143.93 ± 13.53	64.31 ± 5.81	0.079	0.389	0.719
6	115.84 ± 4.26	52.33 ± 2.44	0.064	0.312	0.539
7	208.62 ± 5.94	92.83 ± 3.38	0.110	0.564	1.022
8	155.33 ± 13.78	68.49 ± 5.91	0.084	0.419	0.794
9	210.03 ± 16.95	91.38 ± 7.26	0.113	0.568	1.101

Thorium, and Potassium according to the following equation.

$$F_E = \frac{M_E * C}{\lambda_E * N_E * f_{AE}} * \frac{1}{n} \sum_{i=1}^n A_i \tag{3}$$

where FE is the fraction of element E in the sample, M_E is the atomic mass in kg/mol, λ_E is the decay constant in 1/s, N_A is Avogadro’s number in atoms/mol, f_{AE} is the fractional atomic abundance of ²³²T h, ²³⁸U or ⁴⁰K in nature, C is a constant (with a value of 100 or 1,000,000) that converts the ratio of the element’s mass to soil mass, into a percentage or parts per million (ppm), and A_i is the radiological concentration of selected daughter radionuclides in the decay series of ²³²T h and ²³⁸U, and ⁴⁰K. The value of n is two for ²³²T h and ²³⁸U, which is equivalent to the photon number represent the radioisotopes. The value is reported in ppm, whereas for potassium this value is one and reported in percent (%) (Michalis Tzortzis *et al.*, 2012).

Radiological Hazard Assessment: Radium Equivalent Calculation (Raeq): Gamma-ray radiation hazards caused by specific radionuclides found in our samples were evaluated using different indices. (Raeq), is the weighted sum of activities of the three radionuclides based on the supposition that 370Bq/kg of ²²⁶Ra, 259Bq/kg of ²³²T h, and 4810Bq/kg of ⁴⁰K will produce the same gamma-ray dose rate.

Therefore Raeq can be calculated from this concept as;

$$R_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \tag{4}$$

Where A_{Ra}, A_{T h} and A_K are activity concentrations in Bq/Kg of ²³⁸U, ²³²T h and ⁴⁰K respectively. (Ghazwa Alzubaidi *et al.*, 2016).

Absorbed Dose Rate in Air (DR): This parameter, (DR) is measured at a distance of 1m above the surface that ensures a uniform distributions of the three radionuclides for almost the same activities. At this distance the absorbed Dose rate (DR) can be calculated as,

$$DR = 0.427A_{Ra} + 0.623A_{Th} + 0.043A_K \tag{5}$$

Where the dose rate, DR is in nGy/h and A stands for activity in Bq/Kg for U-238, Th-232 and K-40. This dose rate indicates the received dose at outdoors from radiation emitted by radionuclides in environmental materials. The limit for this dose is 59nGy/h (UNSCEAR (2000, Vol. I; Ghazwa Alzubaidi *et al.*, 2016).

The Annual Effective Dose Rate (ADR): Annual effective dose rate can be calculated to assess the health effects of the absorbed dose in a year. Mathematically it can be represented as;

$$ADR = DR(mGy/h) * 8760h/y * 0.2 * 0.7Sv/Gy * 10^{-6} \tag{6}$$

where AD_R is in mSv/y and 0.7SvG/y is to transform absorbed dose in air to the effective dose received by humans at 1m high, 0.2 is an outdoor occupancy of 20% and 80% for the indoors (Mohammed Mahmud *et al.*, 2014; Ghazwa Alzubaidi *et al.*, 2016). This factor may be changed according to the pattern of human life in the study area. The worldwide average annual effective dose is approximately 2.4mSv/y (Ghazwa Alzubaidi *et al.*, 2016; UNSCEAR, 2000, vol. I).

External (H_{ex}) and Internal (H_{in}) Hazard Index

The external radiation hazard index, H_{ex} , corresponding to the investigated radionuclides is calculated using the following equation

$$H_{ex} = \frac{A_{Ra}}{370Bq/Kg} + \frac{A_{Th}}{259Bq/Kg} + \frac{A_K}{4810Bq/Kg} \tag{7}$$

The maximum value of H_{ex} should be 1 corresponding to the maximum value of Req, which is 370Bq/Kg.

The hazard levels from the inhalation of alpha particles emitted from the radon short-lived radionuclides such as 222-Rn, the daughter product of 226-Ra, and 220-Rn, the daughter product of 224-Ra, can be quantified by the internal hazard index, H_{in} as;

$$H_{ex} = \frac{A_{Ra}}{370Bq/Kg} + \frac{A_{Th}}{259Bq/Kg} + \frac{A_K}{4810Bq/Kg} \tag{8}$$

(Ayham Assie *et al.*, 2016).

RESULTS AND DISCUSSION

From the experimental set up seen in Fig. 2, we found the spectrum for each samples as shown in Fig. 3. The statistical parameters of the observed photo peaks, for 238-U, 232-Th and 40-K radionuclides of Virgin land, Agricultural land and Floriculture land collected from Holeta town are converted into activity concentrations using eq:1 as presented in Table 1. The elemental (radioactive) concentrations of ²³⁸U, ²³²T h and 40K in the three sites, virgin, agricultural and floriculture soils shows variations as compared with each other as seen in table 2. The elemental concentrations were calculated based on eq: 2, and values are showing maximum for uranium concentration and non-exaggerated for thorium and potassium concentrations. The average concentration of ²³⁸U in virgin soil is less than that of agricultural soil for the same soil type and geological structures. When we see again the average concentration of ²³⁸U in the agricultural soil, it is less than that of floriculture soil. Here the age of soils in getting fertilizers is less for floriculture land. The average concentration of 40K is below the recommended value given by much UNSCEAR-2000 in the whole samples of study. But it is more for the area that has been exposed to fertilizers. The remaining radio isotopes (²³²T h is Below Detection Limit (BDL) in agricultural land and very far from the recommended value. Based on the measured data, floriculture soil gets more fertilizers than agricultural soil, and soil from virgin land does not get any fertilizer. Adding radioactive materials containing fertilizers to the soil, specifically increases more the concentrations of ²³⁸U, where ⁴⁰K was not showing variation to the exaggerated value. The recommended reference levels of ²³⁸U, ²³²T h and ⁴⁰K in the soil are 35, 30, and 400 Bq/kg,

respectively, as listed in the world average concentrations published by UNSCEAR (2000 Vol. I, Annex B).



Figure 2. Gamma spectrometry with FJ374 scintillation crystal and Electronic circuits

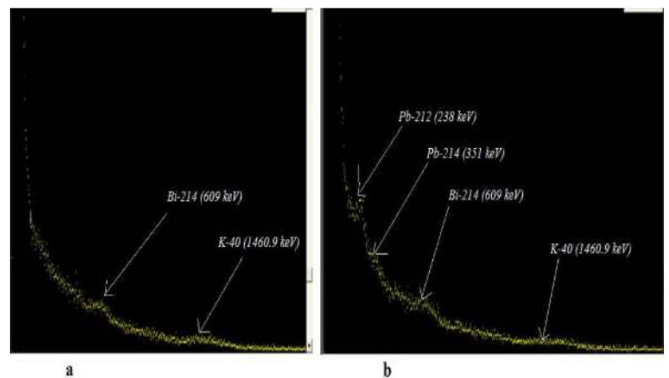


Figure 3. Spectrums showing; a, Background measurement and b, Sample from study area

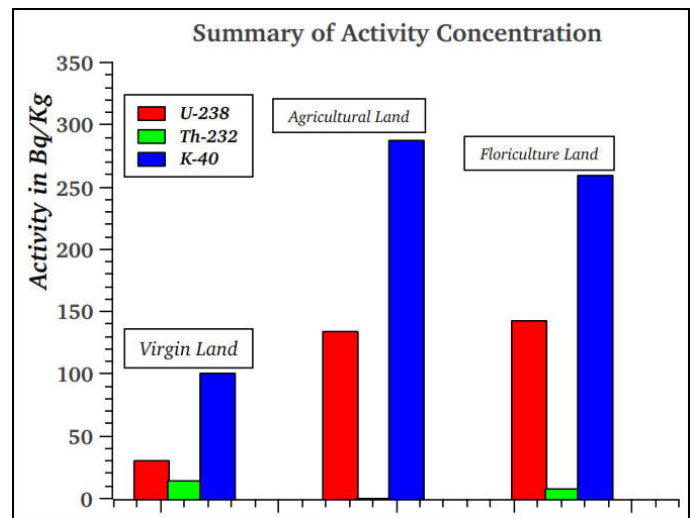


Figure 4: Histogram showing Activity concentrations of ²³⁸U, ²³²T h and 40K in the three sites

The average concentrations of ²³⁸U obtained in the present study for agriculture and floriculture crosses this recommended values. This may cause serious health problems. According to the report of (Atkure Defar, Ahmed Ali, 2013; Mulugeta Getu Sisay, 2009). majorities of the workers in floriculture sites were with breathing system problem, skin problems, cancer etc, within the period of 12 months. In the area where the concentration of short and long lived gamma are maximum, all the above reported problems are expected. Not only gamma radiation, the problems can also happen from ionizing radiations if the dose crosses the limit.

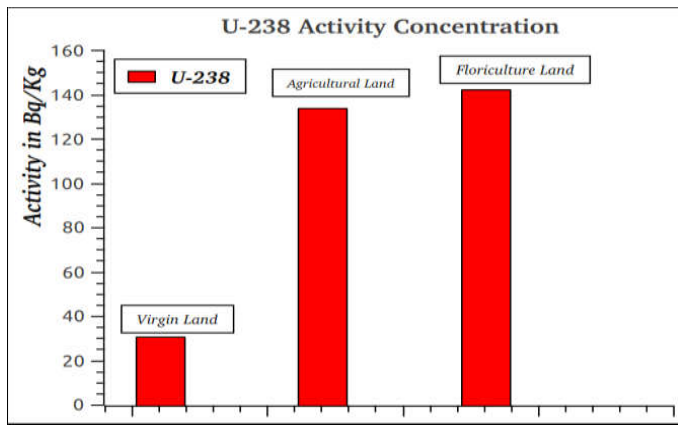


Figure 5. Histogram showing Activity concentration of ^{238}U in Virgin land, Agricultural land and Floriculture land



Figure 6. Unsafe grazing of cattle in floriculture industry

Those radiations are from ^{238}U , $^{232}\text{T h}$, ^{40}K and their decay progenies. The elemental concentrations in our samples also increases with the activity concentrations. For the purpose of the study we could not obtain the recommended values of elemental concentration in the soils. If the concentration of ^{238}U is maximum, like we found in floriculture soil; breathing system problems, skin problems and cancer deceases are expected. Rn-222 and any gas radioactive can cause such problems if they crosses the dose limit for the organs. The area is not safe even after the investor leave the places as seen in fig. 6. This can cause environmental pollution at the farming and at a distance by the mixing of floriculture soil with running waters and winds. Specifically ^{238}U is soluble in water and plants can take easily as a nutrients for their growth (Mamdouh F. Abdel-Sabour, 2014). In the below table, hazard assessments including radioactivity level index are presented. As it is observed from table 3 R_{eq} , D_{R} , AD_{R} , H_{ex} and H_{in} were calculated for the collected samples. We found less values of these parameters in samples collected from virgin lands. The concentrations of $^{238}\text{U}/^{226}\text{Ra}$ in virgin land, (sample 1 and 2) are much less than the samples collected from sites exposed to fertilizers (agriculture and floriculture). For the case of agricultural land, samples 3 and 4, fertilizers were given to the land once per year or once in two years for the production of cereal crops. The R_{eq} , D_{R} , AD_{R} , H_{ex} and H_{in} in the samples taken from this sites are more than that of virgin land. The

geographical and soil type of the two sites are almost the same. The variation is as a result of radioactive fertilizer used by farmers, TSP and DAP. In the long processes, this will produce a radioactive environment in which it becomes difficult for human being to live on it. Up to the time of this study, the average values of the above parameters are showing fast increasing as compared with virgin land. In the case of floriculture land, investors had been using fertilizers more than two times per year for the same floriculture farm. This condition increases the concentrations of radioactive materials in the soil artificially. As we tried to list in Table 3, the average values of R_{eq} , D_{R} , AD_{R} , H_{ex} and H_{in} in floriculture farm is higher than the values in the other sites of study, virgin and agricultural land. This will cause more internal and external health problems than the others. As the workers stay with soils rich in fertilizers, they will accumulate more doses. In the site, workers stays for a minimum of 10-hours per day. In the calculation we used 20% for environmental exposure. But those workers will get more than 45% per day. Therefore the values of ADR reported in table 3 will increase. This may cause the problems reported by (Atkure Defar, Ahmed Ali, 2013; Mulugeta Getu Sisay, 2009). As high radiation exposure rate affect human beings, it also affects microorganisms available in the soil. Microorganisms are essential organisms for the natural fertility of soil. According to (Mesay Adugna Kassa, 2017), more fertilizers can reduces nitrogen fixing bacteria in a soil, which means acidic level of soil will increases. In general, as more fertilizers used in floriculture farm, more radioactive materials are added to the soil. This situation will be at the expense of social and environmental disadvantage (Gudeta Degytnu Tilahun, 2012).

Conclusion

The investigations of this work from the three sampling areas shows a clear differences of activity concentrations of naturally occurring radioactive materials. This is from the phosphate fertilizers added to the soil for the increasing of production. In phosphate fertilizers, sedimentary rock cover more than 80% in which it is with natural radioactive materials. We found that adding such fertilizers to the soil frequently will increases the concentrations of radioactive materials in the soil. From our study, we found more radioactive materials and health hazardousness in the samples collected from fertilizer exposed soils than Virgin soils. This is harmful for humans and environments beyond the safety limit and can creates social and environmental disadvantages. Therefore we recommend the sector to give attention for human life, well inspection of working areas and take a care for the natural environment. Finally, floriculture management strategy should be implemented to avoid the negative impacts of the industry.

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