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Radiological Analysis of Selected Organic Fertilizers in Zaria Local Government Area Council, Kaduna State, Nigeria: Possible Health Implications

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Abstract: *The activity concentrations of naturally occurring radionuclides (^{40}K, ^{226}Ra ^{232}Th) in nine organic fertilizers were measured using NaI (Tl) gamma ray spectrometry. The results show that ^{40}K mean activity concentration was 305.33±0.196Bqkg^{-1} ranging from 120Bq/kg to 907 Bqkg^{-1}, ^{232}Th mean concentration was 45.04Bqkg^{-1} ranging from 14.66 Bq/kg to 60.20Bqkg^{-1}. While ^{226}Ra mean concentration was 24.87±0.02Bqkg^{-1} with a range of 12.83 to 65.69Bqkg^{-1}. Radiological hazard indices were calculated and the mean air absorbed dose rate was 51.59nGyh^{-1} it ranged from 30.33 to 101.23nGyh^{-1}. The annual effective dose rate averaged 61.33µSvy^{-1}(0.061) which is lower than the world average (0.07µSvy^{-1}) it ranged from 33.39 to 124.51µSvy^{-1}. The Raeq was 112.90Bqkg^{-1} which is far below the 370Bqkg^{-1}maximum.The Hex and Hin averaged 0.31 and 0.37respectively. From Radiological point, organic fertilizers do not impart significant radiological risk, because the radiological hazard indices are lower than Bench marks.*
Key words: *Activity concentrations, Radionuclides, Health implications, Gamma ray, organic fertilizers*

I. Introduction

Fertilizers are very important in agriculture for sustaining soil fertility and productivity. They however, may constitute one source of radioactive materials obtained from Phosphate ore used in the production of inorganic fertilizers. Among these fertilizers, NPK (Nitrogen- Phosphorous-Potassium) are the major components in association with natural radioactive materials (Uranium-234, Thorium-232, and Potassium-40) transferred from phosphate ore during production (Ogunleye, Mayaki, & Amapu, 2001). The radioactive materials could emit alpha, beta or gamma radiation. The addition of NPK fertilizers may affect the environment by contaminating it with radioactive materials (Santos *et al.,* 1975), and this could be harmful to soil microorganisms and damaging to the soil (GRPC, 1977). The exposure to radioactive materials has been associated with some health problems like cancer. The objective of this study was to screen organic fertilizers as alternative to phosphate fertilizers which contain naturally occurring radioactive materials (^{226}Ra, ^{232}Th, and ^{40}K) obtained from the phosphate ore. The presence of naturally occurring radioactive materials in the soil above threshold levels may contaminate the environment and cause health problems to humans and plants. Plants, like all living organisms are made up of cells. Within these cells occur numerous metabolic reactions that are responsible for growth and reproduction. Plants depend upon chemical nutrients in the soil to provide the basic metabolic reactions (Mills & Jones, 1996). The supply of plant nutrients is limited and depleted with every harvest, thus causing further reduction in quality and yield in crop plants. The depletion of these plant nutrients is exacerbated by leaching and runoff during raining season. If the yield of plants is to improve or maintained, macro and micronutrients that have been depleted from the soil must be replenished by adding fertilizers.
Radiological hazard indices were measured for organic fertilizers since these fertilizers are spread on our farms and gardens and are used by plants as source of energy and growth which could affect humans through the food chain.

Background radiation exposures vary widely across the Earth, but the average is about 2.4mSv/year, the highest local levels being up to 10 times higher without any detected increased health risks from population studies; 0.1mSv therefore represents a small addition.
The decay of naturally occurring radionuclides in the soils produces a radiation field that transcends the soil-air interface to produce significant human exposure. Raeq was used in calculating radiological hazard indices.

II. Materials and Methods.

Two Cow dung (white Fulani- open-field herd and white Fulani-milk-shade herd), one Sheep dropping hay flock and one Layers litter Shika brown were collected from NAPRI (National Animal Production Research Institute), while one Layers litter coded Layers litter BZ farm was obtained from Abulude's poultry farm and one Sheep dropping grazing flock was obtained from Mamuda's pen in Samaru; these nine samples were transferred into polythene bags, labelled and taken to Centre for Energy Research and Training, Ahmadu Bello University for analysis

2.1 Sample Preparation for Gamma Spectrometry

The fertilizer samples were crushed into fine powder by use of an agate mortar and pestle. Three hundred grams (300g) were weighed and packaged into Radon-impermeable cylindrical plastic containers, selected to fit the space allocated to the detector vessel. A triple-stage sealing system was made for each packaging to avoid Radon-222 from escaping. The inner rims of each container lid were smeared with Vaseline wax; the lid was filled with candle wax to block the gap between lid and container, and finally tight –sealing lid container with a masking adhesive tape. The prepared samples were left alone for thirty days to enable Radon and its progenies to reach radioactive equilibrium prior to the laboratory analysis (Ibeanu, 1999). For the analysis of specific activity concentrations of natural radioactive substances in the fertilizers, gamma –ray spectrometer with NaI (Tl) detector, Canberra type at the Centre for Energy Research and Training, Ahmadu Bello University was used with spectral windows as shown in Table 1

Table 1 Spectral Energy Windows used in Analysis using NaI (Tl) Gamma Spectrometric System

Element	Isotope used	γ-ray energy(KeV)	Energy window (KeV)	Calibration factor CPS/BqKg^{-1})	Detection limits BqKg-^{1}
^{226}Ra	Bi	1764.00	1620-1820	8.632	3.84
^{232}Th	Tl	2614.50	2480-2820	8.768	9.08
^{40}K	K	1460.00	1380-1550	6.451	14.54

Radium equivalent activity was employed in comparing the activity concentrations of samples. Radium equivalent is used as a common index which takes into account the radiation hazard associated with ^{226}Ra, ^{232}Th, and ^{40}K. It provides a useful guideline in regulating safety standards on radiation protection for the public. It is defined on the assumption that 10BqKg-1 of ^{226}Ra, 7 BqKg-1 of ^{232}Th and 130Bqkg-1 ^{40}K produce the same gamma dose rate. It is calculated by use of the following equation:

$Raeq = A_{Ra} + 1.43A_{Th} + 0.077A_K$ (Beretka & Mathew, 1985).

The letters A_{Ra}, A_{Th} and A_K are activity concentrations of Radium, Thorium and Potassium respectively. The recommended value of radium equivalent is 370Bq kg-^{1} as stated by Beretka & Mathew, (1985).The decay of naturally occurring radionuclides in the soils produces a radiation field that transcends the soil-air interface to produce significant human exposure. The quantification of this exposure factor is done by external hazard index, Hex which is given by the expression;

$Hex = \frac{ARa}{370} + \frac{ATh}{259} + \frac{AK}{4810} \leq 1$(UNSCEAR, 2000).

Taking into consideration the hazardous effects of Radon, a progeny of Radium to the respiratory organs, its internal exposure is quantified by the internal hazard index which is given by the expression;

$Hin = \frac{ARa}{185} + \frac{ATh}{259} + \frac{AK}{4810} \leq 1$

The absorbed dose rate (D) due to gamma radiation in air at 1m above the ground surface for uniform distribution of naturally occurring radionuclide was calculated by use of the expression:

$D\ (nGyh^{-1}) = 0.0461A_{Ra} + 0.623A_{Th} + 0.0417A_K$ (UNSCEAR, 2000

III. Results and Discussions

3.1 Specific Radioactivity

The specific radioactivity of ^{226} Ra, ^{232} Th and ^{40} K in organic fertilizers and the Radium equivalent activity (Raeq) are shown in Table 2. The ^{40}K specific activity concentration of organic fertilizers ranged from 120.16 to 907.98Bqkg^{-1} having a mean of 305.33Bqkg^{-1}.

The ^{226}Ra activity concentration in the organic fertilizer had a mean of 24.8749±0.02 Bqkg^{-1} with a range of 121.83 to 65.68Bqkg^{-1}.

^{232}Th has mean activity concentrations of 45.04±0.05 Bqkg^{-1} with a range of 14.66 to 60.21Bqkg^{-1}. Royal fertilizer plus recorded the highest concentration activity of ^{40}K at 907.98±0.58Bqkg^{-1}. The higher ^{40}K activity concentration in Royal fertilizer plus, this so call organic fertilizer could not be of organic source. The organic fertilizers show no significant difference in the Raeq values. From Table 3 means with the same letters are not significantly different at P< 0.05.

Mega green and Sheep dropping hay flock have Potassium activity at 142.09Bq/kg and 137.64Bq/kg which are not significantly different. Layers litter Shika brown and Layers litter BZ farm have ^{226}Ra activity that is not significantly different at P< 0.05.

Cow dung milking herd and Sheep dropping hay flock have ^{232}Th activity that is not significantly different. Table 4.14 shows the Radium equivalent (Raeq) activity of organic fertilizers. The mean is 112.34

Bq/kg. The Raeq values for organic fertilizers are well below the maximum limit of 370Bq kg^{-1} which corresponds to the dose rate limit of 1 mSv of radiation for the public (Jibiri *et al.,* 2009).
Table 4.17 shows the values of Hex (external radiation hazard) and Hin (internal radiation hazard) expressing radiation hazard in organic fertilizers. The mean Hex and Hin values are 0.30 and 0.37 respectively. The highest value of Hex and Hin are 0.57and 0.75 respectively. Sheep dropping hay flock had the lowest value of Hex at 0.22.

The lowest Hin value 0.21 is recorded for Cow dung grazing herd. These indices indicate that organic fertilizers pose no radiological hazards to the environment, Humans, plants and microorganisms

Tables 2: The Results of the Specific Activities/Raeq of Radionuclide in Organic Fertilizers

0rganic fertilizer	K-40	error±	Ra-226	error±2	Th-232	error±3	Raeq. Bq/kg
CRC14	907.98	0.59	65.68	0.06	53.01	0.04	212.34±0.68
CRC15	142.84	0.09	12.83	0.01	48.19	0.04	92.74±0.15
CRNL16	227.01	0.15	25.18	0.02	42.19	0.04	102.99±0.20
CRNL17	361.56	0.23	24.18	0.02	43.19	0.04	113.79±0.29
CRND18	207.88	0.13	20.22	0.02	14.67	0.01	57.19±0.164
CRND19	244.70	0.16	18.77	0.02	52.11	0.05	112.13±0.22
CRNS20	120.16	0.08	18.66	0.02	52.59	0.05	103.12±0.14
CRNS21	139.34	0.09	13.67	0.01	39.22	0.03	80.49±0.14
CRCD22	396.52	0.26	24.68	0.02	60.21	0.05	141.32±0.33
MEAN	305.33	0.19	24.87	0.02	45.04	0 .04	112.90±0.26

KEY
CRC14 =Royal fertilizer plus
CRC15b= Mega Green
CRNL16= Layers litter (Shika brown)
CRNL17= Layers litter (BZ farm),
CRND18= Cow dung (open grazing herd)
CRND19= Cow dung (milking herd non-grazing)
CRNS20=Sheep dropping (open grazing flock)
CRNS21= Sheep dropping (open hay flock),
CRCD22= food concentrate for Cows

Types of Fertilizers	K-40	$Bqkg^{-1}$	Ra-226	$Bqkg^{-1}$	Th-232	$Bqkg^{-1}$
	Mean±S.E	Mean±S.E	Mean±S.E	Mean±S.E	Mean±S.E	Mean±S.E
CRC14	0.580±0.004a	902.38±5.61a	0.06±0.0007a	64.890.789a	0.046±0.001b	52.12±0.89b
CRC15B	0.091±0.001g	142.09±0.75g	0.01±0.0001d	12.69±0.140d	0.042±0.0003c	47.73±0.35c
CRNL16	0.144±0.003e	223.14±3.87e	0.02±0.0016b	24.47±0.713b	0.036±0.0007d	41.45±0.74d
CRNL17	0.231±0.002c	358.85±2.71c	0.02±0.0002b	22.82±1.797b	0.037±0.0011d	41.97±1.22d
CRND18	0.132±0.002f	205.36±2.50f	0.02±0.0015c	18.46±1.758c	0.013±0.0002f	14.47±0.20f
CRND19	0.156±0.002d	241.85±2.85d	0.02±0.0009c	17.73±1.043d	0.045±0.0008b	51.25±0.86b
CRNS20	0.076±0.001h	118.61±1.56h	0.02±0.0003c	18.36±0.300c	0.046±0.0003b	52.25±0.35b
CRNS21	0.089±0.001g	137.64±1.71g	0.01±0.001d	12.98±0.695d	0.034±0.0004e	38.77±0.46e
CRCD22	0.253±0.002b	392.86±3.65b	0.02±0.001b	23.87±0.811b	0.052±0.001a	59.12±1.08a
Mean	0.195±0.036	302.53±55.86	0.02±0.003	24.039±3.647	0.039±0.003	44.348±2.96

Table 3: The Results of Statistical Comparison of Activity Concentrations of Organic Fertilizer Samples

Note: Means with the same letter are not significantly different at P<0. 05
Key
CRC14 =Royal fertilizer plus
CRC15b= Mega Green,
CRNL16= Layers litter (Shika brown),
CRNL17= Layers litter (BZ farm),
CRND18= Cow dung (open grazing herd),
CRND19= Cow dung (milking herd non-grazing)
CRNS20=Sheep dropping (open grazing flock)
CRNS21= Sheep dropping (open hay flock),
CRCD22= food concentrate for Cows

Table 4: Organic Fertilizer Dose Rates and Radiation Hazard Indices

S/N	Sample Code	Air absorbed Dose rate nGy/h	Annual effective dose µSv/y	Raeq	Hex	Hin
1	CRC14	101.23	124.513	212.343	0.571	0.749
2	CRC15	41.9	51.54	92.737	0.251	0.285
3	CRNL16	47.38	58.28	102.989	0.278	0.346
4	CRNL17	53.16	65.39	113.786	0.307	0.372
5	CRND18	27.15	33.39	57.198	0.154	0.209
6	CRND19	51.34	63.15	112.13	0.303	0.359
7	CRNS20	46.4	57.07	103.124	0.282	0.329
8	CRNS21	30.33	37.30	80.493	0.217	0.254
9	CRCD22	65.45	80.50	141.317	0.383	0.448
	Mean	51.59	61.33	112.90	0.305	0.372

Table 5: Summary of Radiological indices

S/N	Radiological Indices	Organic fertilizers	Bench Marks	References
1	Activity Conc. (Bq/kg)			
	^{40}K	305.33±0.02	140.0-850.0	UNSCEAR 2000&USCR, Washington2000
	^{226}Ra	24.81±0.02	17.0- 60.0	UNSCEAR 200 & USCR Washington 2000
	^{232}Th	45.04±0.04	11.0-64.0	UNSCEAR 200 & USCR Washington 2000
2	Dose Rate (n Gy/h)	51.9	56	UNSCEAR, 1993
3	Effective Dose Rates(µSv/y)	61.33	70	UNSCEAR, 1993
4	Raeq (Bq/kg)	112Bq/kg	370(max)	Beretka & Mathew,1995
5	H_{ex}(Bq/kg)	0.305	< 1	UNSCEAR,2000
6	H_{in} (Bq/kg)	0.372	< 1	UNSCEAR,2000

IV. Conclusion

From radiological point of view, the organic fertilizers have lower Raeq values as compared to the values obtained from Phosphate fertilizers from other countries. The radiological hazard indices indicate that organic fertilizers pose no hazards to the environment, humans, plants and microorganisms.

References

[1]. Abdelaziz, M. E., Hanafy, A. H., Ahmed, M. M., & Shaaban, R. P. (2007). Fresh weight and yield of lettuce as affected by organic manure and bio-fertilizers. Conference of Organic farming, Czech Univ. Agric., Czech Republic, 212-214.

[2]. Khater, A. E. M., & AL-Sewaidan, H.A. (2008). Radiation exposure due to agricultural uses of phosphate fertilizers. Journal of Agricultural Food Chemistry, 16, 231-284.

[3]. Belivermis, M., Onder, K., Yavuz, C. V., & Sayhan, T. (2009). The effects of physiochemical properties on gamma emitting natural radionuclides levels in the soil profile of Istanbul. Environmental monitoring assessment, 4(2), 176-185.

[4]. Boukhenfouf, W., & Boucenna, A. (2011). The Radioactivity Measurements in Soils and Fertilizers using Gamma Spectrometry Technique, Journal of Environmental Radioactivity, 102 (4), 336-339.

[5]. Beretka, J., & Mathew, P.J. (19985). Natural radioactivity of Australian building materials Industrial waste and by products. Health Physics, 48, 87-95.

[6]. Cardoso, L.X., Souza, O.S., Ferreira, L.C.F., Ferreira, C.O., Barboza, E., & Ashanti, E.C. (2011). Determination of the Specific Activity of Soils and fertilizers in Serpige-Brazil. Journal of Environmental Quality, 22(4), 22-28.

[7]. Cowart, J. B., & Burnett, W.C. (1994). The Distribution of Uranium and Thorium Decay- Series Radionuclides in the Environment: A Review, Journal of Environmental Quality, 23(4), 23-28.

[8]. Elkhangi, F.A., Abdalla, I. A., & Ahmed, M. M. O. (1994). Evaluation of natural radioactivity and heavy metal content in Sudanese phosphate Rocks used as low cost Fertilizers. Fertilizer Research, 44,129-131.

[9]. Erdem, E., Tinkilic N., Yilmaz V.T., & Uyaink- Olmez H. (1996). Distribution of uranium in the production of triple superphosphate (TSP) fertilizer and phosphoric acid. Fertilizer Research, 44, 129-132.

[10]. German Radiation Protection Commission (1977). Comment of the radiological Protection Commission September. Bundsersanzeiger, 29, 1-3.

[11]. Glover-Kervlient, J. (1995). Environmental Assault on Immunity: Environmental Health Perspectives, 103, 236-240.

[12]. Guimond, R.J., & Hardin, J.M. (1989). Radioactivity released from phosphate containing fertilizers and from gypsum. Radiation Physical Chemistry, 34: 309–315.

[13]. http//www.madehow.com/vol-3/fertilizer.html: How fertilizers are made- materials, production process, making history, use and components.

[14]. International Atomic Energy Agency (2005). Naturally Occurring Radioactive Materials (NORM IV) Proceedings of an international conference held in Szczyrk, Poland, 17–21 May 2004.

[15]. IAEA (1990). Measurement of radionuclides in food and the environment-: Technical Reports Ser, No 295, IAEA, Vienna, 5-27. Journal of Environmental Radioactivity, 12(4), 5-7.

[16]. Ibeanu, I.G.E. (1999). Assessment of Radiological Tin mining activities in Jos and its Environs. A thesis (unpublished) submitted to the Post Graduate School of Ahmadu Bello University, Zaria, Nigeria.
[17]. http//www.tutorvista.com, 2012: Indiscriminate use of fertilizers.
[18]. International Atomic Radiation Committee (1994). Special report: Policy- a review of human carcinogens Part C: Metals, Arsenic, dust http: www.thelancet.com/oncology.
[19]. Jibiri, N.N., & Fasea, K. P. (2009). Radiological hazard indices due to activity concentrations of natural radionuclides in farm soils from two high background radiation areas in Nigeria, International Journal of Low Radiation, 6 (2), 79-95.
[20]. Liyu, W. (2004). WINSPAN, 2004, Amulti-purpose Gamma Ray Spectrum analysis software, CIAE Beijing, China.
[21]. Mazzilli, B.P., Palmiro, V., Saueia, C. & Nisti, M.B. (2000). Radiochemical characterization of Brazilian phosphogypsum, Journal of Environmental Radioactivity, 49, 113-122.
[22]. Mills, K., & Jones, E. (1996). United Nations Scientific Committee on the effects of Atomic Radiation, Report NY, Journal of Radio-Analytical and Nuclear Chemistry, 6, 21-24.
[23]. Munoz, L., Gras. N. Guzman. C., Thieck, M., & Vanska, L. (1993). Radionuclides in the environment. Journal of Radio Analytic & Nuclear Chemistry, 167(1), 97-101.
[24]. National Institute of Safety & Health (1988b). NIOSH testimony to the USA, Department of Labour: Statement of the National Institute of Safety and Health presented at the public hearing on OSHA PELS/Crystallization of silica July 1988. NIOSH policy statement. Cincinnati, Ohio: US Department of Health and Human Services, Public Health service, Centre for Disease Control, National Institute for Occupational Safety and Health.
[25]. Organization for Economic Cooperation and Development (1979). Exposure to radiation from natural radionuclides in building materials .Report by a group of experts of the OECD Nuclear Energy Agency, OECD. Paris, France.
[26]. Okeyode, I.C., & Oluseye, A.M. (2010). Studies of terrestrial outdoor Gamma Ray Dose rate Levels in Ogun-Osun, River Basins Development Authority Headquarters in Abeokuta, Nigeria. Physics International, 6(1), 1-8.
[27]. Ogunleye, P.O., Mayaki, M.C., & Amapu, I.Y. (2001). Radioactivity and heavy metal composition of Nigerian phosphate rocks: possible environmental implications, Journal of Environmental Radioactivity, 62, 39–48.
[28]. Popescu, H.L., Hilt, M., & Lancranjan, I. (2009). Evolution of Biological changes noticed in three cases of accidental mild gamma irradiation. Journal of Occupational Medicine, 14, 317-320.
[29]. Righi, S., Luciallib, P., & Bruzzia, L. (2005). Health and environmental impacts of fertilizer plant- part 1: assessment of radioactive pollution. Journal of Environmental Radioactivity, 82 (2), 167-182.
[30]. Rossler, C.E., Smith, Z. A., Bolch, W.E., & Prince, R.J. (1979). Uranium and Radium-226 in Florida phosphate materials, Journals of Health Physics, 37, 269-277.
[31]. Santos, A.J.G., Mazzilli, B.P., Fa'varo, D.I.T., & Silva, P.C. (2005). Partitioning of radionuclides and trace elements in phosphogypsum and its source materials based on sequential extraction methods. Journal of Environmental Radioactivity Series Radionuclides in the Environment: A Review Journal of Environmental Quality, the production of triple superphosphate (TSP) fertilizer and phosphoric acid. Fertilizer to the general Assembly, with annexes, United Nations, New York.
[32]. Santos, P.L., Gouvea, R.C., & Dutra, I.R. (1995). Human occupational radiation contamination from use of phosphate fertilizers, Science Total Environment, 162 (1), 19-22.
[33]. Shaffer, M. (2001). Waste Lands: The Threat of Toxic Fertilizers, CALPIRG Charitable Trust, the State PIRGs.
[34]. United Nation Scientific Committee on the Effects of Atomic Radiation (1993). Ionizing Radiation Sources and Biological Effects, 162 (1), 19–22.
[35]. UNSCEAR (2000). Sources, effects and risks of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, Report to general Assembly, with annexes, United Nations, New York.