Measurement of Radionuclides in Agricultural Soil at Mandalay Region

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Abstract

This study aims at investigating the environmental impact of the use of phosphate fertilizers in environmental compartments, namely soils, by physical and nuclear techniques. The investigation focused on radionuclides present in fertilizers and cultivated soils, and their decay in time. The radionuclides, ^{228}Th, ^{228}Ra, ^{232}Th and ^{40}K were detected in soil samples treated with GTSP 46% and SSP 16% fertilizers. It is found that the decrease of radionuclides content in soil samples with time was due to the percolation of radionuclides along the soil profile.

Introduction

The lithosphere is an ultimate source of all phosphorus (P) in biosphere. Phosphorus is mainly contained in apatite, a naturally occurring phosphate mineral, which is one of the most easily weathered primary mineral. As a result, P occurs in sufficient supply in young, arid, and neutral soils, although there are some exceptions, depending on the nature of the parent material. Anthropogenic activities greatly enhanced phosphorus inputs from the lithosphere, mainly through phosphate mining and accelerated weathering, as deep soil is exposed as the result of surface erosion. As a consequence, P is accumulating in terrestrial ecosystems in both the industrial world and the developing world. In particular, P is building up in terrestrial soils due to the use of P fertilizers, and is leaking into rivers, lakes and coastal waters. P is also concentrated in sewage effluents and animal and industrial wastes, including P-containing detergents. This source makes a relatively small global contribution [1], although it may be important locally.

Since the 1950, the application of plants nutrients, including phosphate fertilizers, has increased substantially. The long-continued application of phosphorus fertilizers can redistribute and elevate uranium and toxic heavy metals, such as As, Cd and Pb in soil profiles and consequently cause their transfer to the food chain. In soils, one of the sources of radioactivity other than those of natural origin is mainly due to extensive use of fertilizers which is rich in phosphates used for agricultural purposes [2, 3]. Nowadays, chemical fertilizers, such as phosphate fertilizers, are an essential component of the agricultural activities that help to increase crop production and to improve the properties of the nutrient-deficient lands. However, a possible negative effect of phosphate fertilizers is the contamination of cultivated lands by trace metals and some naturally occurring radioactive materials (NORM) [2, 4]. Superphosphate fertilizers contain not only major elements necessary for plant sustainment and growth but also trace metal impurities such as Cd, Pb or Hg. Concentrations of other heavy metal and radionuclide contaminants in phosphate (P) fertilizers vary considerably, depending on the phosphate rock (PR) source. The main radionuclide contaminants in PR are uranium (U), radium (Ra), and thorium (Th) [4, 5].

Particularly, the application of phosphate fertilizers significantly increases the radioactivity level of cultivated soil when it is compared to soil from barren land [6, 7, 8]. The fertilizer industry is considered to be a potential source of heavy metals and natural radionuclides, because fertilizers contain a large amount of heavy metals as Hg, Cd, As, Pb, Cu, and Ni, and natural radionuclide such as ^{238}U, ^{232}Th, and ^{210}Po [9, 10]. These radionuclides elements emit alpha, beta, or gamma radiation when they decay [11], and this could be harmful

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to organisms and damage the soil [12]. The accumulation of heavy metals and radioactive materials in the cultivated soil due to excessive use of phosphate fertilizers in which constitute an increased indestructible poison to the environment.

Objectives

This study investigates the phosphorus dynamics in cultivated soils applied with phosphate fertilizers, through radioactivity analysis. The aims are to assess the radiological risk of use of phosphate fertilizers in cultivated soil from environmental hygiene point of view and to understand the impact on the environment. This is necessary for establishing rules and regulations relating to environmental safety.

Experimental Procedures

Study Area

The present study was carried out in the agricultural field of Marfels ground, Mangi Inn Village, Pyin Oo Lwin Township, Mandalay. Marfels agricultural field is located about 17 km southeast of Mandalay, 64 m (210 feet) above sea level (a.s.l), 21˚58'–21˚98' N, 96˚08'–96˚50' E and on the road to Yeywar hydroelectric power plant. This field lies along the Sagaing Fault [13].

Marfels field falls in a tropical wet and dry climate as Mandalay. Average temperature is around 21˚C in January, the coolest month, while the warmest month, April, average 31˚C. It is very hot in the months of April and May, with average high temperature easily exceeded 35˚C. It is also features wet and dry seasons of nearly equal length with the wet season (May–October) and the dry season (November–April) [14].

The soil profile before experiment is sandy loam soil type with pH is 8.04, organic content is 1.2676% (low), nitrogen (N) is 0.0036%, potassium oxide (K_2O) is 0.058% and phosphorus pentoxide (P_2O_5) is 75.22 lb/ac. Calcium and magnesium contents are 20.49 me/100g and 2.879 me/100g, respectively.

Sampling

In order to assess the concentration of primordials, two commonly used phosphate fertilizers namely triple superphosphate fertilizer (GTSP 46%) and single superphosphate fertilizer (SSP 16%) were collected from the local market. Soil sampling campaigns were carried out in the months of June–August, 2012 and 2013. Eighteen samples of soil were collected from the field of garlic chives in the Marfels agricultural field (Tab. 1). Barren soil was taken from the undisturbed areas. Control, soil sample, was taken from the field without fertilization. The soil samples were collected from the top surface layer to 20 cm depth in each plot using the soil sampler. The experimental beds were fertilized with GTSP 46% and SSP 16% at the rate of 12 g/kg of soil (800 kg per hectare). One soil sample was immediately collected after putting phosphate fertilizers and named “one day soil sample”. Also, the collection of soil samples was performed at one, two, three, four, five and six weeks after fertilization. Indeed, after six weeks from fertilization, garlic chives were harvested. Soil samples were dried in the room temperature for 5-6 days, grounded into fine powder using mortar and pestle and sieved through a 2 mm mesh. One kilogram of homogenized sample was weighted and packed in a polypropylene bag and sealed tightly for about one month to ensure the equilibrium has been reached between ^{226}Ra and its decay products of short half-life and ^{228}Ra and its decay products before being taken for analysis. A Gamma-ray spectrometer with

NaI(Tl) scintillation detector at the Physics department, University of Mandalay, was used for analysis of the specific activity concentration of radioactive substances in the samples. The physiochemical characters of soil and water (irrigation water used in the field) were also analyzed at the Department of Agricultural Research, Yezin, Nay Pyi Taw, Myanmar.

Table 1. List of the samples analyzed with NaI(Tl) scintillation detector (Gamma-ray spectrometer)

No.	Code Name	Sample Name
1	GTSP 46%	Triple superphosphate fertilizer
2	SSP 16%	Single superphosphate fertilizer
3	Barren soil	Soil sample where no crop was sown since long time
4	Control	Soil sample without use of fertilizers
5	1 day	Soil sample collected immediately after fertilization with GTSP 46% and SSP 16% at the rate of 12g/kg of soil
6	1 week	Soil sample collected after one week from fertilization
7	2 weeks	Soil sample collected after two weeks
8	3 weeks	Soil sample collected after three weeks
9	4 weeks	Soil sample collected after four weeks
10	5 weeks	Soil sample collected after five weeks
11	6 weeks	Soil sample collected after six weeks

Analysis with NaI(Tl) Scintillation Detector

For the measurement of all the prepared samples, a plastic container was used. It has a concentric cylindrical shape of inner in 8.6" (diameter), 9.3" (height) and outer in 9.6" (diameter) and 20.5" (height). Before measuring the sample, the environmental gamma background at the laboratory site has been determined with an empty beaker under identical measurement conditions for 3 hours (10800 s). Following, a soil powder sample, with net weight of one kilogram, was placed in the container and the spectrum was collected for 3 hours. The sample in the container covers the detector from all directions in equal thickness and density around the detector. The same procedure was used for all samples. After counting 3 hours, the spectra stored in PC with MCA and the measured spectra were elaborated with the Gamma Vision 32 software. The spectra of samples and background were compared. Peaks in the spectrum were marked “Region of Interest (ROI)” and checked with the library file to get peak information. From this, the gross area and the net area of full energy peak were obtained. The net counts of radioactive elements in the sample were obtained by subtracting the net area of the peak in the background spectrum from that of the peak in the sample spectrum.

Results and Discussion

Physicochemical Parameters of Cultivated Soil

The physicochemical parameters for the soil sample, control, without the use of fertilizers are the soil pH, EC, available N, P_2O_5 and K_2O, exchangeable-Ca and -Mg, organic

matter and soil texture. The soil type in the Marfels agricultural field is sandy loam. Available P_2O_5 of the soil is high. The soil pH is 8.04 and alkaline soil, and the Calcium content is very high (Tab. 2).

The analyzed physicochemical parameters for water (irrigation water used in the Marfels agricultural field) are pH, EC, SAR, Ca, Mg, Na, K, CO_3, HCO_3, Cl, SO_4. The Calcium content of water used in this study is high and pH is 7.78 (slightly alkaline) (Tab. 3).

Table 2. Physicochemical parameters for soil sample in the Marfels agricultural field

No.	Physicochemical parameters	Unit	Result	Classification
1	pH	-	8.04	Slightly alkaline
2	EC	d S/m	0.81	Salt free
3	Available N	%	0.0036	Low
4	Available P_2O_5	lb/ac	75.22	High
5	Available K_2O	%	0.058	Very high
6	Exchangeable-Ca	me/100g	20.49	Very high
7	Exchangeable-Mg	me/100g	2.873	High
8	Organic Matter	%	1.2676	Low
9	Textural class	-	Sand (63.72%), Silt (18.60%) and Clay (17.68%)	Sandy loam
dSm^{-1} = deciSiemens per metre				

Table 3. Physicochemical parameters for irrigation water sample in the Marfels agricultural field

No.	Physicochemical parameters	Unit	Result
1	pH	-	7.78
2	EC	d S/m	0.704
3	SAR	-	0.6573
4	Ca	Ppm	38.72
5	Mg	Ppm	11.50
6	Na	Ppm	3.294
7	K	Ppm	0.3516
8	CO_3	Ppm	Nil
8	HCO_3	Ppm	459.95
10	Cl	Ppm	57.15
11	SO_4	Ppm	23.1
dSm^{-1} = deciSiemens per metre, ppm = part per million			

Radioactivity in Cultivated Soil

The radioelements found in the phosphate fertilizers and in the soil, samples are ^{226}Ra (180 keV), ^{228}Th (239 keV), ^{226}Ra (276 keV), ^{228}Ra (343 keV), ^{232}Th (612 keV) and ^{40}K (891 keV) with net count rate (Tab. 4–6). Previous studies on the analysis of radioactivity in phosphate rocks and their products indicated that the phosphate fertilizers contain the radioelements ^{226}Ra, ^{228}Ra, ^{228}Th, ^{232}Th and ^{40}K, and their concentrations depend on the origin of fertilizer manufactured [15, 16, 17].

The radioelements, ^{226}Ra (180 keV), ^{228}Th (239 keV), ^{226}Ra (276 keV), ^{228}Ra (343 keV), ^{232}Th (612 keV) were found in GTSP 46% fertilizer (Fig. 1B) and also found in SSP 16% fertilizer (Fig. 2B).

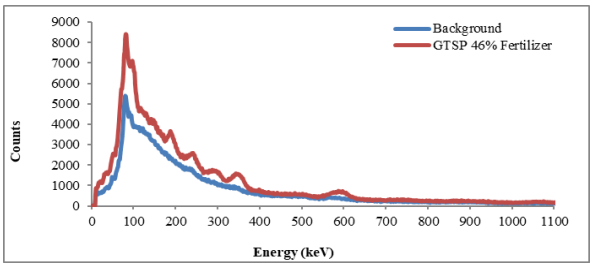

Figure 1(A) Comparison of gamma-rays' spectrum between background and GTSP 46% fertilizer

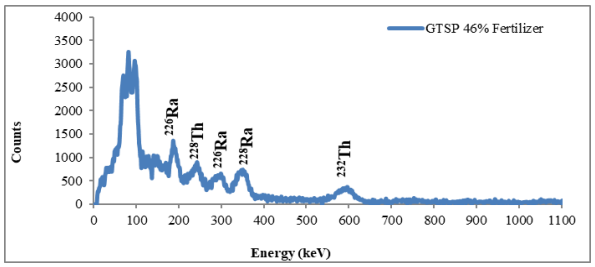

Figure 1(B) Background subtracted gamma spectrum of GTSP 46% fertilizer

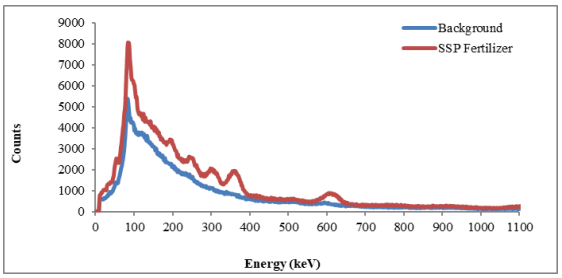

Figure 2(A) Comparison of gamma-rays' spectrum between background and SSP 16% fertilizer

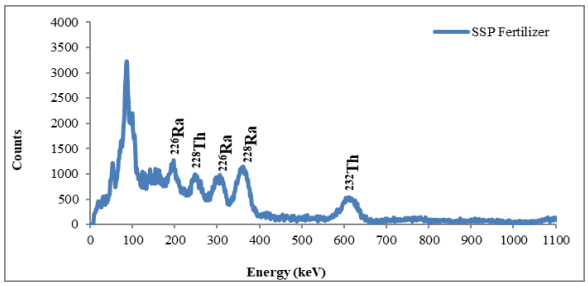

Figure 2(B) Background subtracted gamma spectrum of SSP 16% fertilizer

In GTSP 46%, the net count rates for radioelements are nearly the same for ^{228}Th (239 keV), ^{228}Ra (343 keV), ^{232}Th (612 keV) and followed by ^{226}Ra (180 keV) and ^{226}Ra (276 keV) (Tab. 5). However, in SSP 16%, the net count rate of ^{228}Ra (343 keV) is dominated and followed by ^{232}Th (612 keV), ^{226}Ra (276 keV), ^{226}Ra (180 keV) and ^{228}Th (239 keV) (Tab. 6).

The radioelements, ^{228}Th (239 keV), ^{228}Ra (343 keV), ^{232}Th (612 keV) and ^{40}K (891 keV) are also present in barren soil and control (Fig. 3B, 4B).

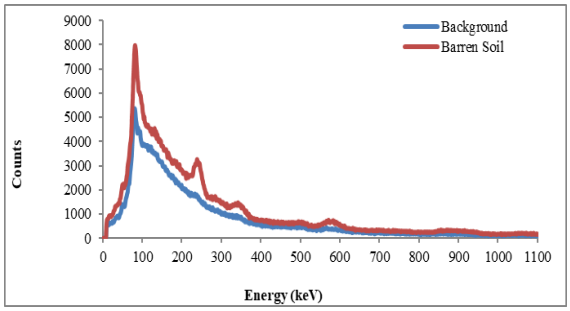

Figure 3(A) Comparison of gamma-rays' spectrum between background and barren soil

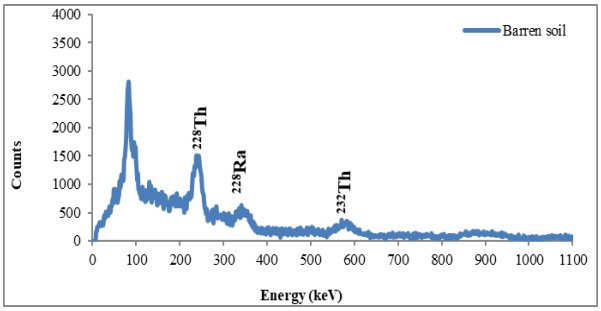

Figure 3(B) Background subtracted gamma spectrum of barren soil

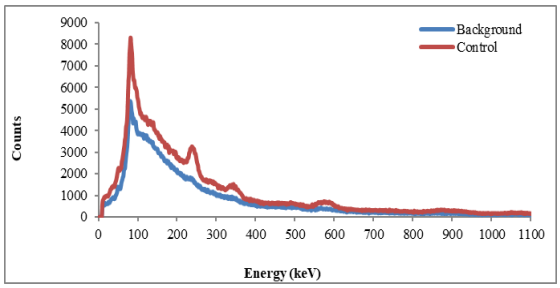

Figure 4(A) Comparison of gamma-rays' spectrum between background and control

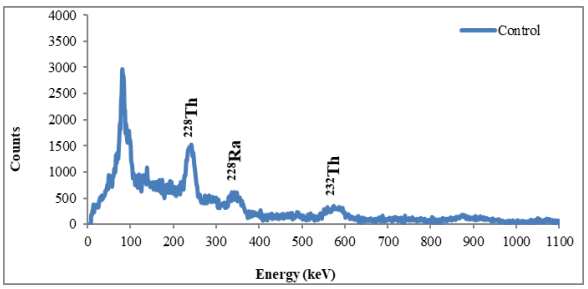

Figure 4 (B) Background subtracted gamma spectrum of control

The net count rate of ^{232}Th is higher than that of ^{228}Th and ^{228}Ra in barren soil. However, net count rate of ^{228}Th is higher than that of ^{232}Th and ^{228}Ra in control. Nevertheless, it is observed that the net count rate of each radionuclide in control is higher than those in barren soil sample (Tab. 5). The higher content of radionuclides in control is caused by the phosphate fertilizers application in the last years during the crop season.

The radionuclides ^{228}Th (239 keV), ^{228}Ra (343 keV), ^{232}Th (612 keV) and ^{40}K (891 keV) are also present in soil samples treated with GTSP 46% and SSP 16% fertilizers.

For ^{228}Th, the net count rate in soil sample treated with GTSP 46% fertilizer indicates that there is no significant change within 2 weeks and it gradually decreases after 2 weeks (R^2=0.786), reaching the same level as control. From the results, the net count rate of ^{228}Ra is high in 1 day soil sample, although it progressively decreases (R^2=0.879) to the same level of control, while the net count rate of ^{232}Th is higher than that in control until 3 weeks and then it also regularly decreases (R^2=0.660) to the same level of control (Fig. 5).

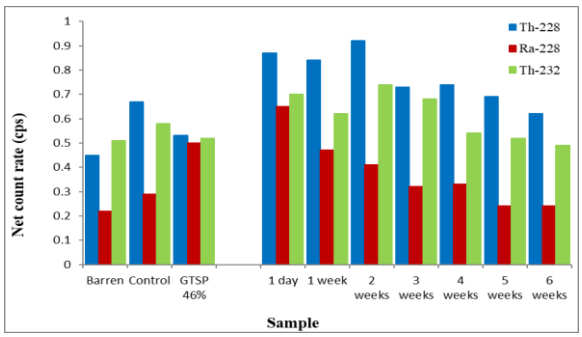

Figure 5. Comparison of the net count rate of radioelements in GTSP 46% fertilizer and soil samples

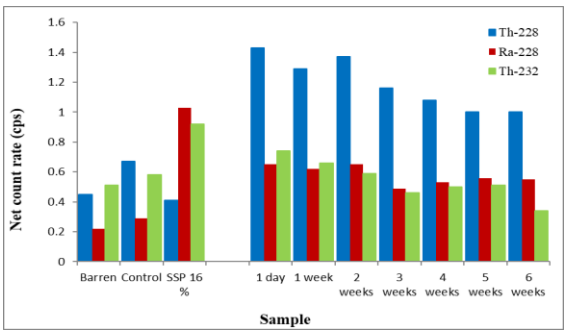

Figure 6. Comparison of the net count rate of radioelements in SSP 16% fertilizer and soil samples

The net count rate of ^{228}Th in soil sample with SSP 16% fertilizer is high in 1 day. It slightly decreases (R^2=0.899) after 3 weeks, however it does not reach the same level as control until 6 weeks. For ^{228}Ra, the net count rate in soil sample is significantly higher until the first 2 weeks, and then it decreases (R^2=0.447), although it does not reach the same level as control. The net count rate of ^{232}Th is elevated in 1-day soil sample and also regularly decreases (R^2=0.853) with time, reaching the level of control after 2 weeks (Fig. 6).

The decrease of radionuclides content in the soil sample with time is due to the percolation of radionuclides along the soil profile**.** In a previous study, Bolivar et al., (1995) pointed out the effect of radioactivity to the surroundings, since radionuclides migrate in various ways like transport, leaching and dissolution from soil to soil and from soils to the aquatic environment [18]. In the light of the above mentioned facts, the results of this study provide the radiological impact of the use of phosphate fertilizers in the soils. This is also confirmed by the IAEA report (2004) that the use of phosphate fertilizers containing radionuclides is the way in which the environment is exposed to enhanced doses of radiation [19].

Table 4. Radioelements found in phosphate fertilizers and soil samples with energy and half-live

No.	Radionuclides	Energy (keV)	Half-live
1	^{226}Ra	180	1.6×10^3 yrs
2	^{228}Th	239	1.91 yrs
3	^{226}Ra	276	1.6×10^3 yrs
3	^{228}Ra	343	5.76 yrs
4	^{232}Th	612	14.1×10^9 yrs
5	^{40}K	891	1.28×10^9 yrs

Table 5. Net count rate of the radioelements with count per second (cps) found in triple super phosphate fertilizer (GTSP 46%) and soil samples

No.	Sample	^{226}Ra 180 keV	^{228}Th 239 keV	^{226}Ra 276 keV	^{228}Ra 343 keV	^{232}Th 612 keV	^{40}K 891 keV
1	Barren		0.45		0.22	0.51	0.34
2	Control		0.67		0.29	0.58	0.44
3	GTSP 46%	0.39	0.53	0.39	0.50	0.52	-
4	1 day		0.87		0.65	0.70	0.47
5	1 week		0.84		0.47	0.62	0.31
6	2 weeks		0.92		0.41	0.74	0.49
7	3 weeks		0.73		0.32	0.68	0.44
8	4 weeks		0.74		0.33	0.54	0.44
9	5 weeks		0.69		0.24	0.52	0.40
10	6 weeks		0.62		0.24	0.49	0.25

Table 6. Net count rate of the radioelements with count per second (cps) found in single super phosphate fertilizer (SSP 16%) and soil samples

No.	Sample	^{226}Ra 180 keV	^{228}Th 239 keV	^{226}Ra 276 keV	^{228}Ra 343 keV	^{232}Th 612 keV	^{40}K 891 keV
1	Barren		0.45		0.22	0.51	0.34
2	Control		0.67		0.29	0.58	0.44
3	SSP 16%	0.42	0.41	0.66	1.03	0.92	-
4	1 day		1.43		0.65	0.74	0.34
5	1 week		1.29		0.62	0.66	0.36
6	2 weeks		1.37		0.65	0.59	0.34
7	3 weeks		1.16		0.49	0.46	0.36
8	4 weeks		1.08		0.53	0.50	0.40
9	5 weeks		1.00		0.56	0.51	0.32
10	6 weeks		1.00		0.55	0.34	0.33

Conclusion

This thesis reports a comprehensive study on the impact of phosphates in cultivated areas. Soil contamination was investigated in the agricultural field of Marfels ground, Mangi Inn Village, Pyin Oo Lwin Township, Mandalay. The radionuclides found in the phosphate fertilizers and in soil samples were ^{226}Ra, ^{228}Ra, ^{228}Th, ^{232}Th and ^{40}K. The radionuclides, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{232}Th were found in GTSP 46% fertilizer and in SSP 16% fertilizer, although there were differences in the net count rates. The radionuclides, ^{228}Ra, ^{228}Th, ^{232}Th and ^{40}K were also present in barren and control soils. The net count rate of each radionuclide in the control soil was higher than that in barren soil, likely caused by the application of phosphate fertilizers on the former during the crop season in the last years.

The radionuclides, ^{228}Ra, ^{228}Th, ^{232}Th and ^{40}K were also detected in soil samples treated with GTSP 46% and SSP 16% fertilizers. In the first case, during the first two weeks, radionuclides in soils were higher than those contained in the fertilizer and in the control soil, and generally decreased in the following weeks reaching the same level as the control soil. However, soils treated with SSP 16% fertilizer showed high net count rates, especially for ^{228}Th and ^{228}Ra, which gradually decreased over time but did not reach the levels of control soil during the observation period.

The decrease of radionuclides content in soil samples with time was due to the percolation of radionuclides along the soil profile. Barren soil was always lower than the control soil, and soils treated with phosphate fertilizers, in some instances, could not reach the net count rates of the control soil over the observation period, suggesting the danger of an accumulation of radionuclides in cultivated areas, which would result in an enhanced dose potentially affecting the environment.

Acknowledgements

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