



Distribution of natural radionuclides in the production and use of phosphate fertilizers in Brazil

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Abstract

The Brazilian phosphate fertilizer is obtained by wet reaction of igneous phosphate rock with concentrated sulphuric acid, giving as final product, phosphoric acid and dehydrated calcium sulphate (phosphogypsum) as by-products. Phosphoric acid is the starting material for triple superphosphate (TSP), single superphosphate (SSP), monoammonium phosphate (MAP) and diammonium phosphate (DAP). The phosphate rock used as raw material presents in its composition radionuclides of the U and Th natural series. Taking this into account, the main aim of this paper is to evaluate the fluxes of natural radionuclides and radioactive disequilibria involved in the Brazilian industrial process of phosphoric acid production; to determine the content of radioactivity in several commercial fertilizers produced by this industry; to estimate their radiological impact in crop soils and the long term exposure due to their application. Radiological characterization of phosphate rock, phosphogypsum and phosphate fertilizers was performed by alpha and gamma spectrometry. The fertilizer samples, which are derived directly from phosphoric acid, MAP and DAP, presented in their composition low activity concentrations for ^{226}Ra , ^{228}Ra and ^{210}Pb . As for U and Th, the concentrations found in MAP and DAP are more significant, up to 822 and 850 Bq kg^{-1} , respectively. SSP and TSP, which are obtained by mixing phosphoric acid with different amounts of phosphate rock, presented higher concentrations of radionuclides, up to 1158 Bq kg^{-1} for ^{238}U , 1167 Bq kg^{-1} for ^{234}U , 1169 Bq kg^{-1} for ^{230}Th , 879 Bq kg^{-1} for ^{226}Ra , 1255 Bq kg^{-1} for ^{210}Pb , 521 Bq kg^{-1} for ^{232}Th , 246 Bq kg^{-1} for ^{228}Ra and 302 Bq kg^{-1} for ^{228}Th . Long term exposure due to successive fertilizer applications was evaluated. Internal

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doses due to the application of phosphate fertilizer for 10, 50 and 100 years were below 1 mSv y^{-1} , showing that the radiological impact of such practice is negligible.

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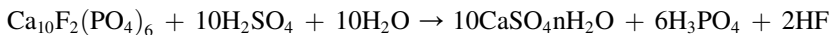
Keywords: Natural radionuclides; Phosphate fertilizers industry; TENORM

1. Introduction

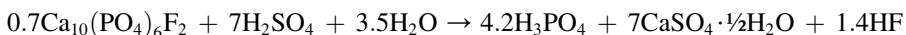
Fertilizers, which contain mainly N, P and K, form an important segment of the Brazilian industry. The common phosphate fertilizers generally used on agricultural lands in Brazil are triple superphosphate (TSP), single superphosphate (SSP), monoammonium phosphate (MAP) and diammonium phosphate (DAP). Igneous phosphate rock is the starting material for the production of all phosphate fertilizer products. Their production involves milling of the phosphate rock, chemical attack with sulphuric acid and finally preparation of commercial blends.

The basic raw material used in the fertilizer industry is the apatite, which is found in phosphate rocks enriched in P_2O_5 . In the apatite lattice, $\text{Ca}_{10}\text{X}_2(\text{PO}_4)_6$, where X stands for chloride, fluoride, carbonate or hydroxyl, both phosphate and calcium ions can be replaced by ionic exchange. In the first case, phosphate is substituted by small amounts of $(\text{VO}_4)^{3-}$, $(\text{AsO}_4)^{4-}$, $(\text{SiO}_4)^{4-}$, $(\text{CO}_3)^{3-}$ or $(\text{SO}_4)^{2-}$; in the second calcium is substituted by magnesium, manganese, strontium, lead, sodium, cadmium, yttrium, zinc, uranium, thorium or lanthanides. Usually, uranium replaces calcium in the apatite structure as U^{4+} and, more seldom, by the adsorption of U^{6+} as uranyl ion (UO_2^{2+}) in the surface of apatite crystals (Rutherford et al., 1994). This substitution is possible due to the similarity in the ionic radius. The same explanation is also valid for Th^{4+} , which presents an ionic radius (0.102 nm) close to calcium (0.099 nm).

In the Brazilian factories, the method used for the production of phosphoric acid is based on drastic treatment of the primary rock with sulphuric acid. This process forms a precipitate of calcium sulphate, called phosphogypsum, as a by-product, which is stockpiled and is considered waste due to its elevated levels of impurities. Phosphogypsum produced during the process is filtered off and pumped as slurry to nearby ponds, where it stays for a period sufficient to allow complete deposition. This waste is then moved to nearby storage areas, the so-called “gyp-stacks”. The production of phosphoric acid can be described by the following reaction:



During the chemical attack of the phosphate rock, different compounds can be formed, depending upon the experimental conditions and the stoichiometry of the reaction. Single superphosphate (SSP) is formed by reacting sulphuric acid with phosphate rock, according to the reaction:



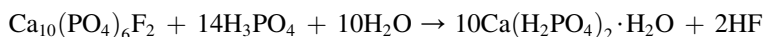
The phosphoric acid produced reacts with phosphate rock in excess giving a final product, monocalcium phosphate:



The total reaction can be described as follows:



In the production of triple superphosphate (TSP), phosphoric acid reacts with apatite according to the reaction:



The chemical fertilizers with ammonium, monoammonium phosphate (MAP) and diammonium phosphate (DAP) are obtained by reacting phosphoric acid with different amounts of NH_3 :



In the phosphate rock, the natural U and Th decay-series are in equilibrium. During the industrial process this equilibrium is disrupted and the radionuclides migrate to intermediate, final products and by-products according to the solubility and chemical properties of each element. Therefore, it is important to measure natural radioactivity, not only in the phosphate rock, but also in different types of fertilizers and by-products, because the high radioactive contents may lead to a significant exposure of miners, manufacturers and end users. The main aim of this paper is to evaluate the fluxes of natural radionuclides and radioactive disequilibria involved in the industrial process of phosphoric acid production; to determine the content of radioactivity in several commercial fertilizers produced by this industry; to estimate their radiological impact in crop soils and the long term exposure due to their application.

2. Experimental

Analysis was carried out on samples of phosphate rock (12), phosphogypsum (12) and fertilizers (18), obtained from the three main factories named A, B and C. After collection, the samples were dried at about 60 °C to remove moisture and crushed to fine powder (30 to 60 mesh). The following experimental procedure was adopted.

Activity concentrations of ^{226}Ra , ^{228}Ra and ^{210}Pb were measured in samples of phosphate rock, phosphogypsum and fertilizers by gamma spectrometry with a hyper-pure germanium detector, EGNC 150–190 R, from Eurisys Measures, with a resolution of 1.8 keV for the 1332 keV ^{60}Co photopeak and 15% efficiency. The detector was calibrated using natural soil, rock and water spiked with radionuclides certified by Amersham. Samples were packed in 100 cm³ cans and sealed for about 4 weeks prior to the measurement in order to ensure that equilibrium has been reached between ^{226}Ra and its decay products of short half-life. The ^{226}Ra activities were determined by taking the mean activity of three separate photopeaks of its daughter nuclides: ^{214}Pb at 295 keV and 352 keV, and ^{214}Bi at 609 keV. The ^{228}Ra content of the samples was determined by measuring the intensities of the 911 keV and 968 keV gamma ray peaks from ^{228}Ac . The concentration of ^{210}Pb was determined by measuring the intensity of the 46.5 keV peak. Self-absorption correction was applied to ^{210}Pb since the attenuation for low energy gamma rays is highly dependent upon sample composition. This approach used was suggested by Cutshall et al. (1983).

Typical lower limits of detection for gamma spectrometry were 5.0 Bq kg^{-1} for ^{226}Ra , 3.0 Bq kg^{-1} for ^{228}Ra and 19.0 Bq kg^{-1} for ^{210}Pb , for a counting time of 50 000 s.

Activity concentrations of the alpha emitters ^{226}Ra , U and Th isotopes were measured in samples of phosphate rock, phosphogypsum and fertilizers by alpha spectrometry. The radiochemical procedure was based on the total dissolution of the samples (200 mg) followed by purification with anionic exchange resins. In the first column the solution is eluted in a Dowex 1×8 resin in 9 M HCl: U is retained whereas Th isotopes and Ra flow through the resin. U is eluted with 0.1 M HCl and the final solution electrodeposited in a disk for alpha counting. The eluate containing Th and Ra is passed through two columns: the first one (Dowex 1×8 resin in 8 M HNO_3) for the retention of Th and the second one (Dowex 1×8 resin in 0.75 M HBr) for the purification of Ra. Th is eluted with 9 M HCl and is electrodeposited for alpha counting. A seeding suspension of BaSO_4 is added to the solution containing Ra; the microprecipitated BaRaSO_4 formed is retained in a Polypropylene membrane filter and counted on the alpha spectrometer. All the measurements were performed on a surface barrier detector, EG&G Ortec. Typical lower limits of detection for the alpha measurement of the radionuclides were 1.9 Bq kg^{-1} for ^{238}U , 1.7 Bq kg^{-1} for ^{234}U , 1.3 Bq kg^{-1} for ^{230}Th , 1.3 Bq kg^{-1} for ^{226}Ra , 2.0 Bq kg^{-1} for ^{232}Th and 1.8 Bq kg^{-1} for ^{228}Th , respectively.

3. Results and discussion

Results obtained for the radionuclides ^{238}U , ^{234}U , ^{230}Th , ^{226}Ra , ^{210}Pb , ^{232}Th , ^{228}Ra and ^{228}Th on samples of phosphate rock, phosphogypsum and fertilizers (TSP, SSP, MAP and DAP) obtained from the three main factories named A, B and C, are presented in Table 1.

Box plot graphic was used to evaluate the distribution of the experimental results for the activity concentration of phosphate rock, fertilizers and phosphogypsum, along the process, in industries A, B and C (Fig. 1). This methodology allows determining the mean, the median and the dispersion of the results. The plot obtained for industry A shows clearly the distribution of radionuclides along the process. The radioactive series of U (^{238}U , ^{234}U , ^{230}Th , ^{226}Ra , and ^{210}Pb) and of Th (^{232}Th , ^{228}Ra and ^{228}Th) are in equilibrium in the rock samples. After the chemical attack this equilibrium is disrupted and the radionuclides migrate according to their chemical properties. U isotopes (^{238}U and ^{234}U) are found in the fertilizers TSP and SSP, whereas Th isotopes (^{232}Th , ^{228}Th and ^{230}Th), radium isotopes (^{226}Ra and ^{228}Ra) and ^{210}Pb are distributed uniformly in the fertilizers TSP and SSP and in phosphogypsum. The results obtained are in agreement with Mazzilli et al. (2000), which found enrichment factors from phosphate rock to phosphogypsum of 90% for ^{226}Ra , 80% for ^{232}Th and ^{230}Th and 100% for ^{210}Pb .

Box plot obtained for the activity concentration along the process of industry C shows the distribution of radionuclides in the production of fertilizers MAP and DAP. U isotopes (^{238}U and ^{234}U) concentrate in the fertilizers, whereas, Th isotopes (^{232}Th , ^{228}Th and ^{230}Th) are distributed, following the same pattern, among fertilizers and phosphogypsum. Ra isotopes and Pb are found predominantly in phosphogypsum. Both industries, A and C, use phosphate rock from Catalão-Goiás, a phosphorite, which is an igneous rock constituted by apatite, magnetite and olivine cut by abundant veins of carbonatites. Although the mines explored by both industries are close, a difference between the concentrations of the U series can be observed; whereas, Th decay-series presents almost the same concentration in both industries. Such differences can be explained by the fact that U is associated to the mineral apatite and Th to aluminum phosphate, which present different behavior in the milling of the primary rock sample (Santos et al., 2006).

Table 1

Activity concentration (mean \pm standard deviation, range) in Bq kg⁻¹ for radionuclides in phosphate rock, fertilizers and phosphogypsum samples from industry A, B and C

	PR		SSP		TSP		PG	
	M \pm DP	Range	M \pm DP	Range	M \pm DP	Range	M \pm DP	Range
<i>Industry A</i>								
U-238	1361 \pm 404	841–1868	920 \pm 186	706–1158	848 \pm 19	834–861	48 \pm 5	42–53
U-234	1323 \pm 396	962–1841	936 \pm 181	735–1167	869 \pm 53	831–906	59 \pm 4	54–63
Th-230	1354 \pm 369	837–1694	836 \pm 232	665–1169	743 \pm 158	631–855	782 \pm 144	631–978
Ra-226	1195 \pm 224	948–1518	720 \pm 111	626–879	546 \pm 3	544–548	744 \pm 160	550–940
Pb-210	1310 \pm 223	1085–1698	1084 \pm 106	973–1255	904 \pm 60	844–964	1061 \pm 132	834–1163
Th-232	413 \pm 126	334–577	217 \pm 47	188–287	206 \pm 23	189–222	226 \pm 31	189–257
Ra-228	346 \pm 95	236–492	196 \pm 33	155–246	157 \pm 24	133–181	242 \pm 43	210–273
Th-228	387 \pm 127	244–559	190 \pm 35	170–243	189 \pm 33	166–212	217 \pm 40	166–253
<i>Industry B</i>								
U-238	376 \pm 202	158–705	433 \pm 142	264–620	273 \pm 69	182–377	37 \pm 19	50–23
U-234	527 \pm 208	320–736	410 \pm 190	237–613	329 \pm 51	276–378	65 \pm 35	40–89
Th-230	291 \pm 25	270–319	349 \pm 116	248–476	286 \pm 36	244–308	50 \pm 25	32–68
Ra-226	212 \pm 77	139–319	10 \pm 4	7–14	105 \pm 28	88–138	186 \pm 50	122–236
Pb-210	409 \pm 96	373–517	<19		141 \pm 51	101–198	182 \pm 65	136–228
Th-232	332 \pm 29	279–356	446 \pm 82	339–546	401 \pm 74	331–521	86 \pm 21	60–103
Ra-228	316 \pm 30	290–349	197 \pm 105	<3–368	189 \pm 17	171–204	152 \pm 39	124–179
Th-228	338 \pm 18	318–353	285 \pm 66	215–346	295 \pm 10	284–302	125 \pm 40	97–153
<i>Industry C</i>								
U-238	484 \pm 108	344–672	650 \pm 121	520–799	688 \pm 148	520–797	49 \pm 13	40–58
U-234	585 \pm 88	522–714	713 \pm 116	592–822	739 \pm 140	581–850	50 \pm 3	48–52
Th-230	578 \pm 64	492–636	164 \pm 32	131–195	244 \pm 77	155–290	293 \pm 59	251–334
Ra-226	361 \pm 143	222–581	9 \pm 6	<1.3–17	5 \pm 4	<1.3–11	344 \pm 65	280–434
Pb-210	543 \pm 94	463–666	33 \pm 12	19–42	<19		347 \pm 44	316–378
Th-232	395 \pm 75	246–457	105 \pm 20	81–127	145 \pm 26	109–174	204 \pm 33	172–243
Ra-228	320 \pm 51	255–367	99 \pm 47	46–133	72 \pm 43	44–122	219 \pm 40	191–247
Th-228	392 \pm 55	359–473	68 \pm 30	36–95	79 \pm 29	46–102	189 \pm 15	178–199

The raw material used in industry B comes from Tapira-Minas Gerais, which is made up of carbonatite and piroxenite. The Box plot for the results obtained along the process of industry B shows the distribution of radionuclides in the fertilizers MAP and TSP. U and Th isotopes are distributed homogeneously in the two fertilizers, their concentrations, however, are low in phosphogypsum. Th presents a different behavior when compared with industries A and C, since its concentration is low in the phosphogypsum and higher in the phosphoric acid. Ambiguous behavior was also found for ²²⁸Ra, which presents higher concentrations in the fertilizer MAP. The radionuclides ²²⁶Ra and ²¹⁰Pb are found preferentially in the fertilizer TSP and in phosphogypsum.

Although several publications were found related with the characterization of Brazilian phosphate rock and phosphogypsum (Mazzilli et al., 2000; Silva, 2001; Silva et al., 2001; Santos, 2002; Santos et al., 2002), no specific reference was found concerning the radiological characterization of the Brazilian phosphate fertilizers and the long term exposure due to its application along the years.

The amount of phosphogypsum produced in Brazil is about 4.5×10^6 tons per year. From this total, 1.7×10^6 tons per year is used as soil amendment and about 0.7×10^6 tons per

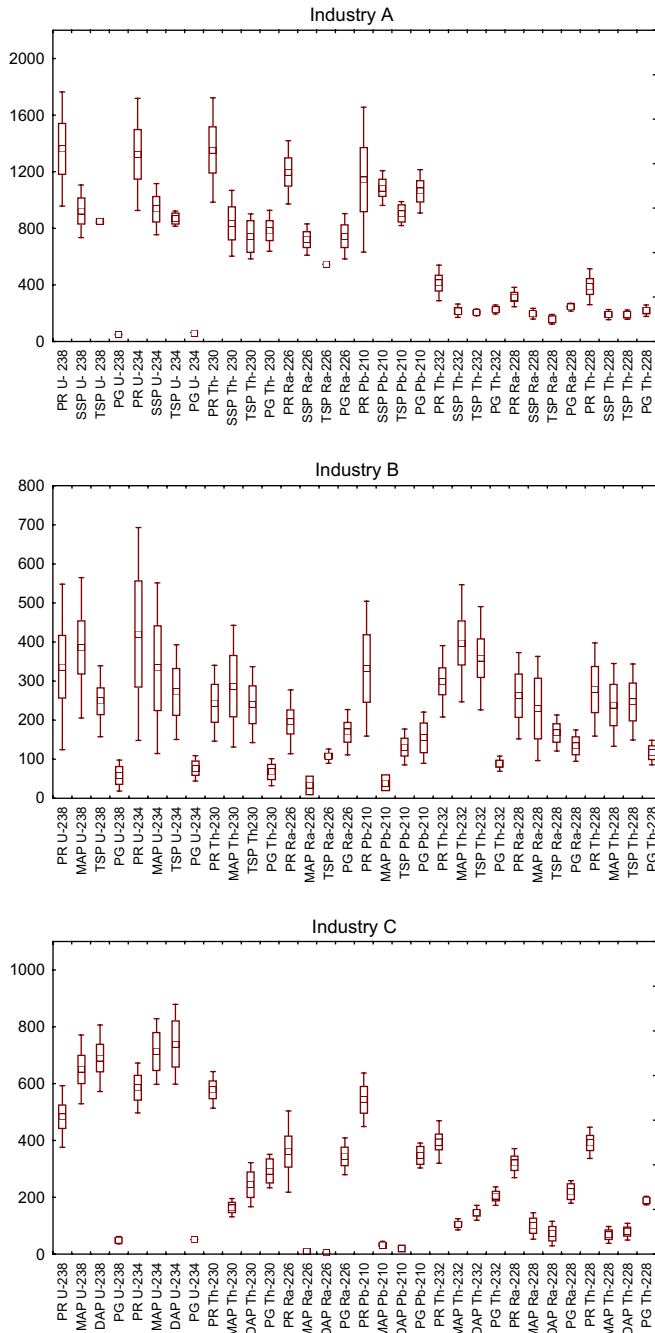


Fig. 1. Distribution of radionuclides in phosphate rock (PR), fertilizers (SSP, TSP, MAP and DAP) and phosphogypsum (PG) samples from industries A, B and C.

year are used in the cement industry, and the remaining 2.1×10^6 tons stored in piles in the open air. Knowing the amount of phosphogypsum produced per year by these Brazilian industries and the corresponding activity concentration it can be concluded that a total amount of 12.4 TBq (0.27 TBq of ^{238}U , 0.35 TBq of ^{234}U , 2.3 TBq of ^{230}Th , 2.7 TBq of ^{226}Ra , 3.3 TBq of ^{210}Pb , 1.0 TBq of ^{232}Th , 1.3 TBq of ^{228}Ra and 1.1 TBq of ^{228}Th) is released annually to the phosphogypsum piles of the factories. The radiological environmental impact of these piles is being published elsewhere (Santos et al., in press).

From the results obtained for the radionuclides' content in the Brazilian fertilizers, its environmental impact can be predicted. Considering that a maximum of 80 kg of P_2O_5 per ha (for grain crop) and a maximum of 700 kg of P_2O_5 per ha (in green crop) is added to the soil annually, the total activity released to the environment can be evaluated. Since these radionuclides are homogeneously distributed in the upper 20 cm of the soil with an apparent density of 1.3 g cm^{-3} , an increase of concentration up to 0.87 (grain crop) and 7.6 Bq kg^{-1} (green crop) are found in the soil, depending upon the fertilizer used (Tables 2–3). This added amount does not reach 2% of the normal content of these radionuclides in an undisturbed soil. This scenario, however, considered the environmental impact of one single fertilizer application. A more conservative approach should consider a long term exposure due to successive fertilizer applications along the years. In order to estimate such impact it is necessary to evaluate the transfer of radionuclides and their concentration distribution in the environmental matrices such as soil, vegetation, milk, etc. and the individual doses by using mathematical models. The estimation of the activity concentrations in the environmental compartments were estimated according to the model recommended by International Atomic Energy Agency (IAEA, 1989) and the results considered as intermediate in the food chain and used directly in the evaluation of the dose. The scenario takes into account application of 700 kg of P_2O_5 per ha for green crop and of 80 kg of P_2O_5 per ha for grain crop; application of SSP and TSP fertilizers from industry A and application of MAP and DAP fertilizers from industry C; concentration in the soil after successive annual fertilizer application for 10, 50 and 100 years; uptake of radionuclides from soil to vegetation through roots, intake of radionuclides by animals and transfer to milk and meat.

To estimate the increment of activity concentration in the soil it was assumed that the fertilizer is added to the soil before the harvesting and is incorporated to the rooting zone depth forming a uniform mixture. The equation used for this prediction is

$$C_{s,i} = \frac{C_{f,i} a_f (1 - e^{-\lambda_{E,i} t_d})}{P \lambda_{E,i}}$$

Table 2

Fertilizer recommendation (kg ha^{-1}) and radionuclides' concentration in land (Bq kg^{-1}) as a consequence of fertilizer application to grain crop

Fertilizer	Fertilizer recommendation grain crop	^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra	^{228}Th	Total activity
SSP	444	0.16	0.16	0.14	0.12	0.2	0.04	0.03	0.03	0.87
TSP	195	0.06	0.07	0.06	0.04	0.07	0.02	0.01	0.01	0.33
MAP	166	0.04	0.05	0.01	0.001	0.002	0.01	0.006	0.004	0.12
DAP	177	0.05	0.05	0.02	0.001	0.001	0.01	0.005	0.005	0.14

Table 3

Fertilizer recommendation (kg ha⁻¹) and radionuclides' concentration in land (Bq kg⁻¹) as a consequence of fertilizer application to green crop

Fertilizer	Fertilizer recommendation green crop	²³⁸ U	²³⁴ U	²³⁰ Th	²²⁶ Ra	²¹⁰ Pb	²³² Th	²²⁸ Ra	²²⁸ Th	Total activity
SSP	3888	1.4	1.4	1.2	1.1	1.6	0.3	0.3	0.3	7.6
TSP	1707	0.5	0.6	0.5	0.4	0.6	0.1	0.1	0.1	2.9
MAP	1458	0.4	0.4	0.1	0.01	0.02	0.06	0.06	0.04	1.1
DAP	1555	0.4	0.4	0.1	0.01	0.01	0.1	0.04	0.05	1.1

$C_{s,i}$ = radionuclide activity concentration i in soil (Bq kg⁻¹)

$C_{f,i}$ = radionuclide activity concentration i in fertilizer (Bq kg⁻¹)

a_f = surface application rate of fertilizer (kg m⁻² y⁻¹)

$\lambda_{E,i}$ = effective rate constant for reduction of the activity concentration of radionuclide i in the root zone of soil (y⁻¹) $\lambda_{E,i} = \lambda_i + \lambda_s$

λ_i = rate constant for radioactive decay of radionuclide i (y⁻¹)

λ_s = rate constant for reduction of the concentration of material deposited in the root zone of soils owing to processes other than radioactive decay (y⁻¹)

t_d = time of deposition in soil (y)

P = surface density for the effective root zone in soil (kg m⁻²).

The deposition rate for each radionuclide i in the soil is represented by the product of the activity concentration of the radionuclide in the fertilizer, $C_{f,i}$, by the surface application rate, a_f (given by the ratio between the recommended dosage and the frequency of application). In this scenario, it was assumed just one fertilizer application per year in the soil and the dosage recommended by *Boletim Técnico-100 (1997)* for each fertilizer and farming; the results are presented in *Tables 2 and 3*.

The removal factor for each radionuclide present in the soil is obtained by the application of a factor exponentially proportional to the deposition rate. Two mechanisms contribute to the reduction of activity in the root zone: the radioactive decay and the radionuclides' infiltration to deeper layers of soil. Both are implicit in the effective removal constant of the activity of radionuclide i from the root zone, $\lambda_{E,i}$, given by the summation of the radionuclide decay constant, λ_i , and the physical removal constant, λ_s , which accounts for the reduction by natural processes such as water percolation and erosion. In this case λ_s was considered as zero. The surface density for the effective root zone in soil represents the product of the soil density by the depth of the root zone. Both parameters depend on the type of soil, its classification and crop composition; the values assumed were 1.3 kg m⁻³ and 20 cm, respectively. The parameters used in the evaluation of the radionuclide concentration in soil were taken from *IAEA (2001)*.

To estimate the uptake from soil by edible portions of vegetation, it was assumed that the foliar absorption is negligible and that the uptake from soil to plant is mainly by the root. The equation used for this evaluation is

$$C_{v,i} = C_{s,i} F T_{v,i} e^{-\lambda_i t_h}$$

$C_{v,i}$ = radionuclide activity concentration i in the edible part of plant v (Bq kg⁻¹ fresh weight plant)

$C_{s,i}$ = radionuclide activity concentration i in soil (Bq kg⁻¹)

$FT_{v,i}$ = transfer factors for terrestrial foods (kg dry kg^{-1} fresh)
 λ_i = rate constant for radioactive decay of radionuclide i (d^{-1})
 t_h = average time interval between harvest and consumption of the food (d).

The transfer factor, $FT_{v,i}$, is defined as the ratio between the radionuclide activity concentration in the edible part of the plant in the harvest period and in the soil. The term $\exp(-\lambda_i t_h)$ takes into account the radioactive decay of radionuclide i , between the harvest and the consumption of the food.

For the evaluation of the activity concentration in milk and meat, the following equation was used:

$$C_{p,i} = C_{s,i} FT_{p,i} e^{-\lambda_i t_h}$$

$C_{p,i}$ = radionuclide activity concentration i in animal flesh and milk (Bq kg^{-1} fresh or Bq L^{-1})

$C_{s,i}$ = radionuclide activity concentration i in soil (Bq kg^{-1})

$FT_{p,i}$ = transfer factors from soil to meat and milk (kg dry kg^{-1} fresh)

λ_i = rate constant for radioactive decay of radionuclide i (d^{-1})

t_h = average time between slaughter or collection and human consumption of meat or milk (d).

For the evaluation of the transfer factors from soil to the animal products, $FT_{p,i}$, a more conservative approach was applied, which takes into account the direct transfer of the radionuclides from the soil to the meat and the milk. For the parameter t_h , the values of 14 and 60 days were assumed for green crop and grain crop, respectively. All the parameters used were taken from IAEA (2001).

The effective dose due to ingestion of terrestrial food is given by:

$$E_v = \sum_i C_{v,i} U_v \text{FCD}_{\text{ing},i}$$

E_v = committed effective dose due to terrestrial food (Sv y^{-1})

$C_{v,i}$ = activity concentration of radionuclide i in the edible part of plants (Bq kg^{-1})

U_v = ingestion rate (kg y^{-1})

$\text{FCD}_{\text{ing},i}$ = dose conversion factor due to ingestion of radionuclide i (Sv Bq^{-1}).

The effective dose due to ingestion of the meat and the milk is given by:

$$E_p = \sum_i C_{p,i} U_p \text{FCD}_{\text{ing},i}$$

E_p = effective dose due to ingestion of the meat and the milk (Sv y^{-1})

$C_{p,i}$ = activity concentration of radionuclide i in meat and milk (Bq kg^{-1} or Bq L^{-1})

U_p = ingestion rate (kg y^{-1})

$\text{FCD}_{\text{ing},i}$ = dose conversion factor due to ingestion of radionuclide i (Sv Bq^{-1}).

For the evaluation of the doses a deterministic assumption was made for 10, 50 and 100 years of fertilizers' (SSP, TSP, MAP and DAP) application. The values of ingestion rate were taken from IBGE, and refer to the annual per capita intake. The dose conversion factors due to radionuclides' ingestion were taken from ICRP 67. The effective doses due to ingestion

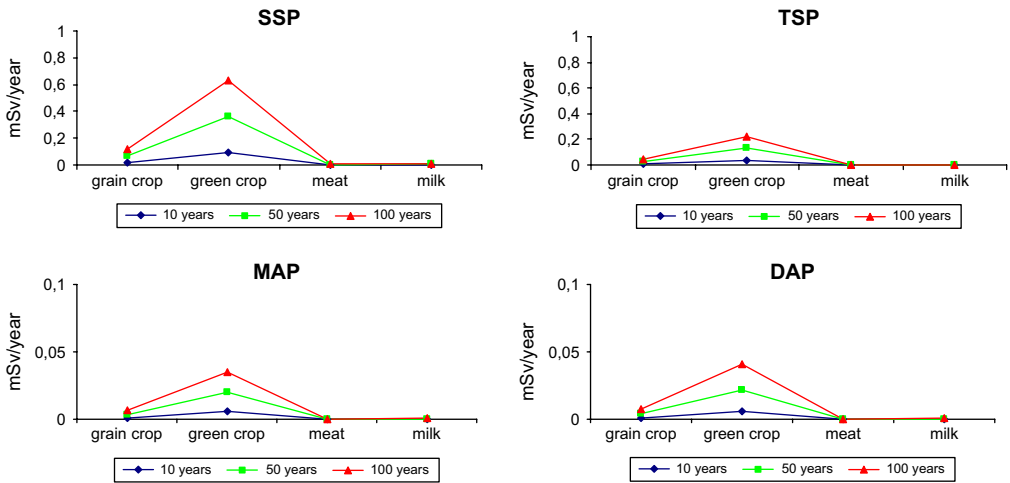


Fig. 2. Effective doses due to ingestion of radionuclides after successive annual fertilizer application in soil (mSv y^{-1}).

of radionuclides after successive annual fertilizer application in soil for 10, 50 and 100 years are presented in Fig. 2. It can be seen that the doses are quite below 1 mSv y^{-1} in all cases except for the SSP application in green crop during 100 years. It can be concluded that the radioactive impact of the use of phosphate fertilizers is negligible.

4. Conclusion

Brazilian phosphate fertilizers, which are produced directly from phosphoric acid, MAP and DAP, present in their composition low concentrations of radionuclides ^{226}Ra , ^{228}Ra and ^{210}Pb . As for the radionuclides U and Th, the concentrations observed are higher, reaching values up to 822 Bq kg^{-1} and 850 Bq kg^{-1} , respectively. SSP and TSP, which are obtained by reacting phosphoric acid with phosphate rock in different proportions, presented higher concentrations of all studied radionuclides, reaching values up to 1158 Bq kg^{-1} for ^{238}U , 1167 Bq kg^{-1} for ^{234}U , 1169 Bq kg^{-1} for ^{230}Th , 879 Bq kg^{-1} for ^{226}Ra , 1255 Bq kg^{-1} for ^{210}Pb , 521 Bq kg^{-1} for ^{232}Th , 246 Bq kg^{-1} for ^{228}Ra and 302 Bq kg^{-1} for ^{228}Th .

The results obtained show that the levels of radioactivity present in the fertilizers are of the same order of magnitude of those found in the phosphogypsum. The doses due to the application of phosphate fertilizer for 10, 50 and 100 years were below 1 mSv y^{-1} , showing that the radiological impact of such practice is negligible.

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