Radiological impact of the application of phosphogypsum in agriculture

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ABSTRACT

Phosphogypsum, a waste by-product derived from the wet process production of phosphoric acid, represents a serious problem facing the phosphate industry in Brazil. This by-product (mainly calcium sulphate dihydrate) precipitates during the reaction of sulphuric acid with phosphate rock and is stored at a rate of about $5.5x10^6$ tons per year on several piles in, Brazil. Phosphogypsum can be used in agriculture as a soil amendment. It can be used for highly weathered soils, with relatively low exchange capacities and/or low levels of extractable nutrients; for soils with high sodicity; for acid soils with high levels of Al and for calcareous soils. In order to study the radiological impact of using phosphogypsum in agriculture, a conservative scenario was defined considering a long term exposure due to successive annual applications of phosphogypsum in agriculture for 10, 50 and 100 years. The doses evaluated due to the ingestion of food produced by using soils tilled with phosphogypsum were always below 0.5 mSv/y, and of the same order of magnitude as those obtained by using Brazilian phosphate fertilizers.

INTRODUCTION

In Brazil, several NORM industries are in operation, which require an adequate environmental and occupational radiological control. Depending upon the level of radioactivity the NORM industries are subjected to the recommendations given by CNEN, which include compliance with the radiological protection regulations (CNEN-NN-4.01 and CNEN-NN-3.01).

Among these installations are the phosphate fertilizer industries, which generate a by-product known as phosphogypsum. This residue is produced by precipitation during wet sulphuric acid processing of phosphate rocks, thus posing serious problems with its utilization and safe disposal. In Brazil, three main phosphate industries are responsible for the production of approximately $5.1x10^6$ tons per year of phosphogypsum. The material 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 residue is then moved to nearby storage areas. The phosphogypsum stacks can pose serious environmental impact, not only from a chemical, but also from a radiological point of view. All the countries that produce phosphate fertilizer by acid wet processing of phosphate rock are facing the same problem of finding solutions for the safe application of this residue, in order to minimize the impact caused by the disposal of large amounts of phosphogypsum.

In Brazil, one possibility is the use of phosphogypsum in agriculture as a soil amendment. Phosphogypsum is useful for highly weathered soils, with relatively low exchange capacities and/or low levels of extractable nutrients; for soils with high sodicity; for acid soils with high levels of Al and for calcareous soils. Its application, however, should take into account the presence of radionuclides in phosphogypsum and their bioavailability for the environment, specially the contamination of draining water soil and the absorption by plants. In order to study the radiological impact of using phosphogypsum in agriculture, a conservative scenario was defined considering a long term exposure due to successive annual applications of phosphogypsum in agriculture for 10, 50 and 100 years. The doses evaluated due to the ingestion of food produced by using soils tilled with phosphogypsum were compared with the doses obtained by using Brazilian phosphate fertilizers (TSP, SSP, MAP and DAP).

MATERIAL AND METHODS

Activity concentrations of ^{238}U, ^{234}U, ^{230}Th, ^{226}Ra, ^{210}Po and ^{232}Th were measured in phosphogypsum samples by alpha spectrometry. The experimental procedure consisted of a chemical attack with strong acids, followed by spontaneous deposition in silver plate and counting on a surface barrier detector. Activity concentrations of ^{210}Pb and ^{228}Ra were measured by gamma spectrometry with a hyper-pure germanium detector. Details of the experimental procedure can be found in Saueia and Mazzilli (2005, 2006).

In order to estimate the radiological impact of the use of phosphogypsum in agriculture, the scenario chosen considered a long term exposure due to successive phosphogypsum applications along the years. In this scenario, the phosphogypsum is spread on land thus leading to a surface contamination which depends on the radionuclide concentration in the phosphogypsum and on the thickness of the layer on the land. The use of this land for agricultural production (vegetables, animal products) will then lead to an exposure of the public via the ingestion path. In this scenario, spreading 0.2 kg of phosphogypsum per m² is assumed according to the recommendations of the manufacturers (2000 kg/ha). 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.
To estimate the increment of activity concentration in the soil it was assumed that the phosphogypsum 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:

$$C_{s,i} = \frac{C_{f,i} \cdot a_f \cdot \left(1 - e^{-\lambda Ei \cdot td}\right)}{P \cdot \lambda_{E,i}}$$

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

$C_{f,i}$ = radionuclide activity concentration *i* in phosphogypsum (Bq kg^{-1})

a_f = surface application rate of phosphogypsum (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 $\left(y^{-1}\right)$. $\lambda_{E,i} = \lambda_i + \lambda_s$

λ_i = rate constant for radioactive decay of radionuclide *i* $\left(y^{-1}\right)$

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 $\left(y^{-1}\right)$

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 phosphogypsum, *Cfi*, by the surface application rate, *Af (*given by the ratio between the recommended dosage and the frequency of application). In this scenario, it was assumed just one phosphogypsum application per year in the soil and the dosage recommended by Boletim Técnico 100 (1997) for phosphogypsum and farming.

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, λe,i, given by the summation of the radionuclide decay constant, λi, and the physical removal constant, λs, which accounts for the reduction by natural processes such as water percolation and erosion. In this case λs 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:

$$C_{v,i} = C_{s,i} \cdot FT_{v,i} \cdot e^{-\lambda i \cdot th}$$

$C_{v,i}$ = 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, *FTv,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 (-λi.th) 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:

$$C_{p,i} = C_{s,i} \cdot FT_{p,i} \cdot e^{-\lambda i \cdot th}$$

$C_{p,i}$ = 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 meet and 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:

$$E_v = \sum_i C_{v,i} \cdot U_v \cdot FCD_{ing.i}$$

E_v = committed effective dose due to terrestrial food in 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})

$FCD_{ing.i}$ = dose convertion factor due to ingestion of radionuclide i (Sv Bq^{-1})

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

$$E_p = \sum_i C_{p,i} \cdot U_p \cdot FCD_{ing.i}$$

E_p = effective dose due to ingestion of meat and 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})

$FCD_{ing.i}$ = dose convertion factor due to ingestion of radionuclide i (Sv Bq^{-1})

For the evaluation of the doses it was made a deterministic assumption for 10, 50 and 100 years of phosphogypsum application. The values of ingestion rate were taken from IBGE, and reefer to the annual per capita intake. The dose conversion factors due to radionuclides ingestion were taken from ICRP 67.

RESULTS AND CONCLUSIONS

The results obtained for the doses are presented in table 1. The doses due to the application of phosphogypsum for 10, 50 and 100 years were always below 0.5 mSv/y, showing that the radiological impact of this practice is negligible. The higher dose was obtained from the use of phosphogypsum in grain crop for 100 years. In Figure 1 the results are compared with data obtained from the use of Brazilian phosphate fertilizers, SSP, TSP, MAP and DAP (Saueia and Mazzilli, 2006). It is interesting to note that the doses due to the use of phosphogypsum in the agriculture are of the same order of magnitude as those derived from the use of the fertilizers SSP and TSP for green crop, grain crop, meat and milk. Doses derived from the ingestion of meat and milk were always two orders of magnitude lower than those derived from the ingestion of green crop and grain crop, for both fertilizers and phosphogypsum. Doses reported by the European Commission, Report EUR 19264 (2000), on the radiological impact resulting from the use of the fertilizer products, using a similar scenario, ranged from 0.1 to 1 mSv/y. It can be concluded that the doses derived from the use of phosphogypsum in agriculture are in the range observed for the use of fertilizers in Europe and in Brazil, and therefore, pose no health risks to the final consumers.

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Table 1 – Effective doses due to ingestion after successive annual application of phosphogypsum in soil.

Effective doses mSv y^{-1}			
	Phosphogypsum from industry A		
	10 y	50 y	100 y
Grain crop	$7.6\ 10^{-2}$	$2.8\ 10^{-1}$	$4.9\ 10^{-1}$
Green crop	$4.6\ 10^{-2}$	$1.7\ 10^{-1}$	$2.9\ 10^{-1}$
Meat	$3.8\ 10^{-4}$	$1.4\ 10^{-3}$	$2.5\ 10^{-3}$
milk	$1.7\ 10^{-4}$	$2.8\ 10^{-3}$	$4.9\ 10^{-3}$
	Phosphogypsum from industry B		
	10 y	50 y	100 y
Grain crop	$2.2\ 10^{-2}$	$7.0\ 10^{-2}$	$1.2\ 10^{-1}$
Green crop	$1.3\ 10^{-2}$	$4.2\ 10^{-2}$	$7.0\ 10^{-2}$
Meat	$1.2\ 10^{-4}$	$3.9\ 10^{-4}$	$6.6\ 10^{-4}$
milk	$2.5\ 10^{-4}$	$7.7\ 10^{-4}$	$1.3\ 10^{-3}$
	Phosphogypsum from industry C		
	10 y	50 y	100 y
Grain crop	$3.9\ 10^{-2}$	$1.3\ 10^{-1}$	$2.3\ 10^{-1}$
Green crop	$2.3\ 10^{-2}$	$8.0\ 10^{-2}$	$1.4\ 10^{-1}$
Meat	$2.1\ 10^{-4}$	$6.8\ 10^{-4}$	$1.2\ 10^{-3}$
milk	$4.1\ 10^{-4}$	$1.3\ 10^{-3}$	$2.3\ 10^{-3}$

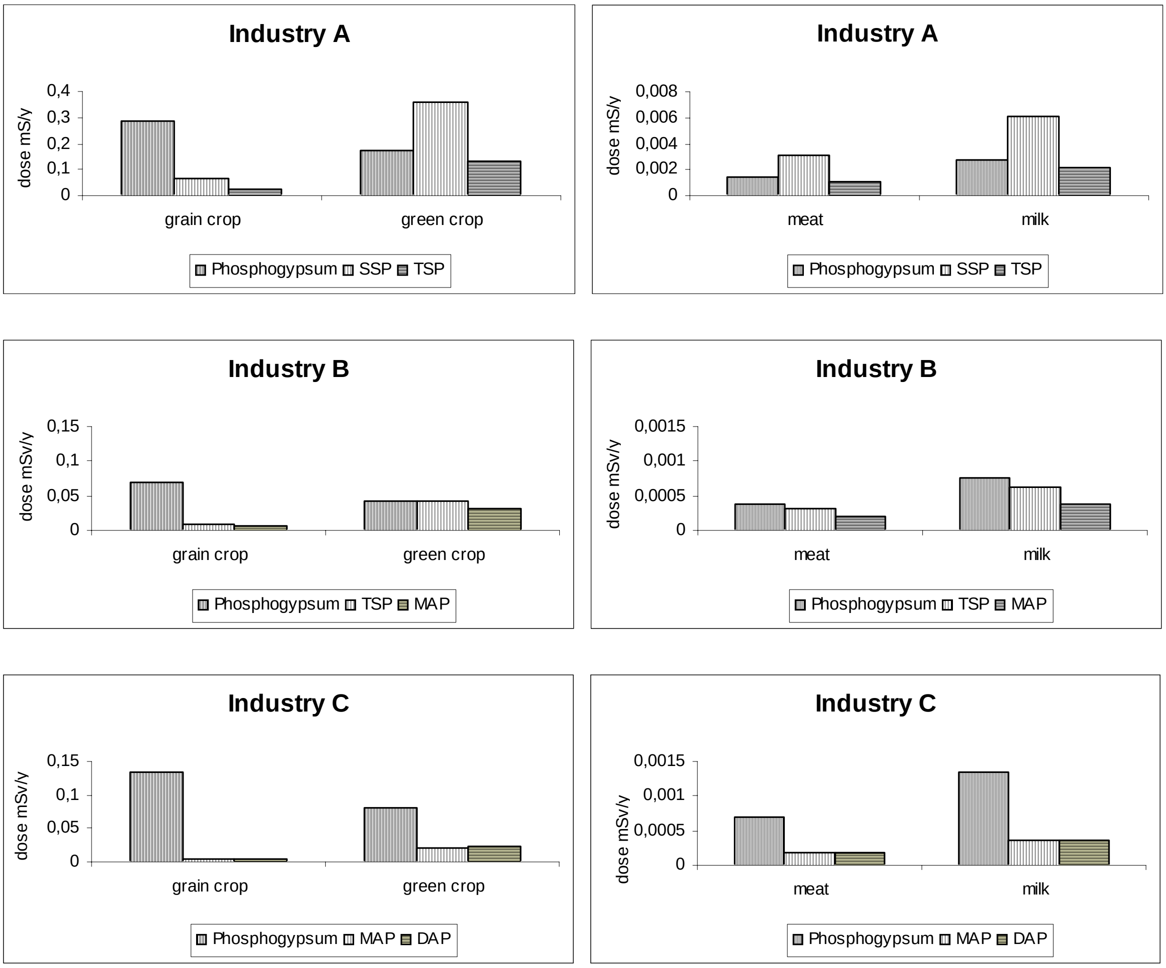

Figure 1. Effective 50 years dose per annual application of phosphogypsum and fertilizers (from Saueia and Mazzilli, 2006).