A DOSIMETRIC MODEL FOR DETERMINING THE EFFECTIVENESS OF SOIL COVERS FOR PHOSPHOGYPSUM WASTE PILES

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***Abstract*—Phosphogypsum (PG) is a by-product of the phosphoric acid production process that contains high concentrations of U-series radionuclides. PG piles formed during the last 30 years cover about 1,200 hectares and are located close to the town of Huelva (Spain) on a salt-marsh. The regional government of Andalusia restored the area beginning in 1990 by covering it with a 25-cm-thick layer of natural soil. With this restoration, the external gamma-dose rate in the zone has decreased drastically, approaching near environmental background values. This conclusion is based on results obtained through *in-situ* monitoring measurements and through a dosimetric model developed for that particular radiation source. As the model uses average parameters of the studied site, its output does not show a correlation point by point with the *in-situ* monitoring measurements. However, a good agreement is observed in average values over the covered piles. The model gives an average dose rate of 0.41 mGy y^{-1} and the *in situ* monitoring 0.40 mGy y^{-1}. Based on this model, it is possible to calculate the necessary thickness of soil to reduce the dosimetric contribution from a similar extension of PG until the desired level is reached. In our conditions, in a 25-cm-thick soil, about 0.19 mGy y^{-1} is the increase produced by the PG layer in relation to an infinitum soil layer. Consequently, no radiological concern exists in the restored zones with respect to the external gamma radiation.**

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Key words: soil; dose assessment; waste management; uranium

INTRODUCTION

A LARGE industrial complex, which includes several factories devoted to phosphoric acid production, is since 1965 located near the estuary formed by the confluence of the Tinto and Odiel river mouths, nearby Huelva town (Southwestern Spain) (Fig. 1). Another factory devoted to the Cu extraction, whose wastes were also released with the PG, is located in this industrial complex.

In the chemical process, phosphate rocks are treated with sulfuric acid, generating in addition to phosphoric acid the PG, which is mostly calcium sulphate:

$$Ca_{10}(PO_4)_6F_2 + 10\ H_2SO_4 + 20\ H_2O \rightarrow 6\ H_3PO_4 + 10\ CaSO_4H_2O + 2\ HF. \quad (1)$$

In the phosphoric acid factories, owned by Fertiberia and FMC Foret companies, imported sedimentary phosphate rocks are treated that contain elevated amounts of natural radionuclides from the U-series [concentrations 30–50 times higher than uncontaminated soils (Bolívar et al. 1996a)]. ^{40}K and Th-series radionuclides concentrations in these rocks are much lower than in normal soils. The secular equilibrium between ^{238}U and its progeny found in the mineral is broken in the process of P_2O_5 production. PG contains the most of the ^{226}Ra and ^{210}Po originally present in the phosphate rock (between 80–100%). In addition to that, there are smaller percentages of uranium (20%) and thorium isotopes (30–40%) (Baestlé 1991; Bolívar 1995). A large fraction of this by-product is pumped as a suspension in sea water through a system of pipes into a salt-marsh zone adjacent to the factories (Fig. 1). In this area, PG is accumulated by deposition on big piles which are about 5 m high. About 2.5×10^6 tons of PG per year are stored in each pile, covering an area of 1,200 hectares, and containing a total of $60–70 \times 10^6$ tons of PG.

The PG piles form a potential radiological hazard source, containing high amounts of natural radionuclides, a large fraction of which are gamma emitters. Additionally, one of the radionuclides in high concentrations in these piles is ^{226}Ra, which decays to ^{222}Rn, an alpha emitter noble gas that can emanate from the PG into the atmosphere and can generate an increase in the total dose by inhalation.

In 1990, the regional government of Andalusia proposed the restoration of this zone. The objective was to correct the environmental impact (not only radiological) produced by the deposited wastes and to reduce the increasing contamination of the waters and sediments of

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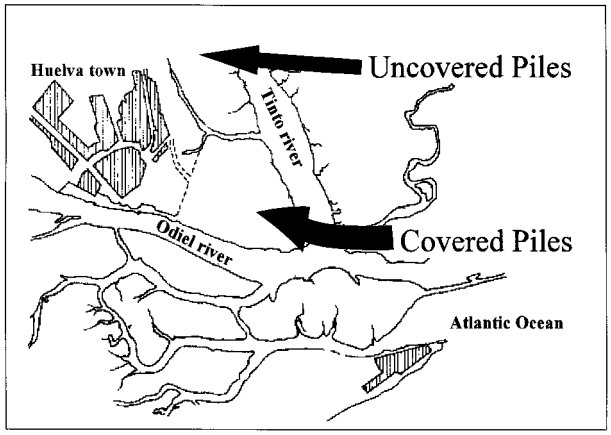

Fig. 1. Map of the estuary formed by the Odiel and Tinto River mouths. Arrows point to covered and uncovered phosphogypsum piles locations.

Tinto river affected by the wastes. The technological solution adopted for this restoration was to cover PG piles with a layer of soil with an average thickness of 25 cm, ranging from 19 to 36 cm in our measurements (Fig. 2). Radiologically, this solution will decrease the gamma radiation emitted in air coming from the phosphogypsum.

Until now, about 450 hectares have been restored, which correspond to the oldest deposits (20–30 y). Revegetation of the added soil layer has been accomplished in this area in order to use it as a recreative zone. This project requires a detailed radiological study, which addresses

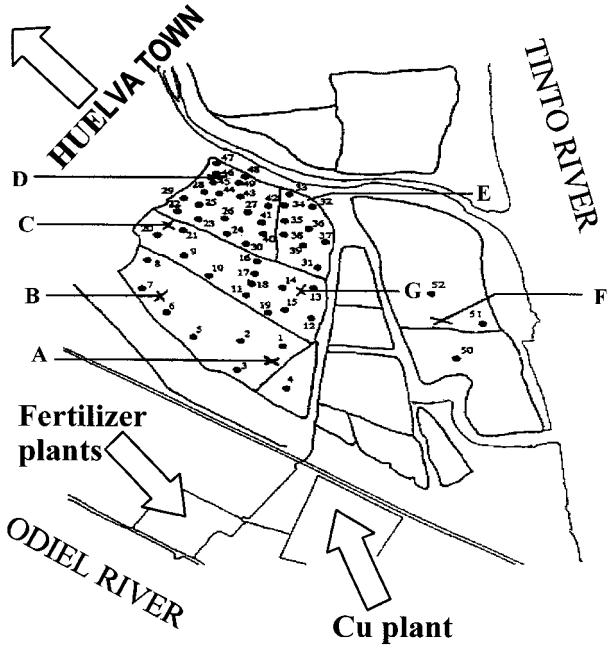

Fig. 2. Map of the restored phosphogypsum zone, the 52 monitored regions (1,2, . . . , 52) and the locations of the 7 collected cores (A, B, . . . , G).

1. The estimation of outdoor terrestrial gamma dose rates over the restored area; and
2. The measurement and analysis of dose for ^{222}Rn concentrations in air over the restored area and nearby surroundings, during a minimum of 1 y.

The phosphogypsum deposited in this zone was accumulated during 15 y and, for that reason, its radionuclide concentrations are not uniform depending on the origin of the phosphate rock treated and the effectiveness of the process of phosphoric acid extraction. In addition, the piles (formed mainly by phosphogypsum) can also contain wastes from the Cu-extraction factory in not considerable but variable amounts. These Cu-wastes contain significantly lower amounts of radioactive nuclides than the phosphogypsum, inducing (depending on its proportion in the piles) variable dilution degrees in the radionuclide concentrations.

This paper focuses on determination of the present absorbed dose rate in air over the restored zone and theoretical evaluation about the effect of soil layer thickness covering the PG on external dose rate. A dosimetric model for the covered piles was developed that allows calculation of outdoor external dose rate for any soil thickness. A study addressing measurement of ^{222}Rn concentrations is in progress, and thus it not dealt with here.

MATERIALS AND METHODS

In-situ measurements

Measurements of outdoor terrestrial gamma dose rates have been made using an environmental radiation monitor[‡] that contains an ionization chamber working as a Geiger-Müller detector. The detector is sensitive to gamma radiation and x rays, and the readings were taken at 1 m above the ground. The 450 hectares of the study zone were divided in 52 portions, whose size depended on its frequency of utilization in the future (Fig. 2). Ten measurements were made in each portion. From the average readings at each portion, the correct value of the outdoor terrestrial gamma dose rate was calculated by subtracting the background value due to the cosmic rays and its electronic noise. Contribution of the electronic noise (0.105 mGy y^{-1} for this device) was measured putting the monitor inside a lead shield in a 4π geometry and 10 cm thick; the dose rate at this latitude from cosmic rays is 0.28 mGy y^{-1} (UNSCEAR 1988). The total background was also directly measured by placing the monitor on the sea surface to avoid the contribution of the gamma ray fraction from the soil. The value found through this last method was 0.46 mGy y^{-1} (a similar result was obtained by the first method).

For comparison purposes, absorbed dose rates were also measured *in situ* over two uncovered PG piles, sited in the North-East of the recovered zone. The measurements were done in these uncovered piles at 15 points separated by at least 200 m.

[‡] FAC, model FH40F1, Berthold Systems, Inc., Aliquippa, PA, USA.

Radionuclide concentrations

Outdoor terrestrial gamma dose rates also can be estimated indirectly from measured concentrations of the gamma-emitters (U-series, Th-series, ^{40}K) at different depths in the terrestrial zone using a dosimetric model based on dose rate factors (Kocher and Sjoreen 1985).

To check the results obtained from the *in-situ* survey, seven cores were collected, which included the upper added soil layer and the 60 cm of the underlying phosphogypsum. PG cores were sliced in six layers of 10-cm thickness. Every sample was dried at 65°C (to avoid the loss of its hydration water) until constant weight and then ground. To study the distribution of several radionuclides in the PG block, radioactive measurements were done in 14 samples (two samples per core of the seven cores collected: the upper and bottom layers of each PG core).

In aliquots of the PG layers studied, as well as of soil layer, the specific activities of U-isotopes and ^{210}Po (in secular equilibrium with ^{210}Pb due to the age of these PG piles) were determined by alpha-particle spectrometry, while the ^{226}Ra, ^{228}Ra, ^{228}Th, and ^{40}K concentrations were measured by gamma-ray spectrometry.

For the determination of alpha emitters, an aliquot sample (1–2 g of soil or 0.3–0.5 g of PG) traced with known amounts of ^{232}U and ^{208}Po (radionuclides with the same chemical behavior as U-isotopes and ^{210}Po, respectively) was dissolved by wet digestion, and the polonium and uranium fractions were sequentially isolated by using a solvent extraction method with TBP (Holm and Fukai 1977). Finally, the uranium fraction was electrodeposited onto stainless-steel planchets while the polonium fraction was self-deposited onto silver planchets.

Counting of the obtained radioactive sources was done in an alpha spectrometry system formed by four independent chambers with 450 mm^2 ion-implanted silicon detectors that have a nominal resolution of 20 keV and a counting efficiency of ~25% for a sample-detector distance of 5 mm.

The ^{226}Ra, ^{228}Ra, ^{228}Th, ^{40}K and other gamma emitters were determined using a hyperpure coaxial germanium detector, which has been previously calibrated for soil and phosphogypsum samples at a fixed geometry of the sample-detector (Bolívar et al. 1994; Bolívar et al. 1996b). The detector has an active volume of 68 cm^3, 14% relative efficiency, and is surrounded by a 10-cm lead passive shield.

Dosimetric model

From the specific activities of ^{40}K and the gamma emitter radionuclides of the uranium and thorium series in the PG and the soil layers, the external gamma dose rate at 1 m over ground can be calculated applying a dosimetric model based on the dose rate conversion factors (DRF) published by Kocher and Sjoreen (1985) for monoenergetic sources from 0.01 to 3 MeV, uniform activity concentrations, and covering an infinite extension. These dose rate factors have been tabulated for various thicknesses of the source and for infinitely thick sources.

For a monoenergetic emission from a radionuclide, X_k, homogeneously distributed in a block (covering an infinite extension) between its surface $z = 0$ up to a depth z, with a gamma-emission probability of unity, and activity concentration $C(0, z; X_k)$ (Bq cm^{-3}), the absorbed dose rate in air $D(0, z; X_k)$ (Gy y^{-1}), produced at 1 m above the block is given by

$$D(0, z; X_k) = DRF(0, z; X_k)C(0, z; X_k), \quad (1)$$

where the $DRF(0, z)$ is the dose rate conversion factor. In our case, the restored zone can be considered as an infinite block (the spatial dimensions of piles are much bigger than 1 m) formed by three layers, where in each one of them the concentrations of the different gamma and x-emitters can be assumed to be uniform:

- Layer 1: soil layer from 0–25 cm;
- Layer 2: PG layer 5 m thick, under layer 1; and
- Layer 3: geological substratus in Huelva under layer 2, which extends infinitely in depth.

In these layers, it is assumed that the DRFs are independent of the composition because the linear attenuation coefficients of the soil and the PG are similar for gamma energies exceeding 120 keV (Bolívar 1995). On the other hand, gamma emissions coming from PG block (layer 2) with energies below 120 keV will contribute negligibly to absorbed dose rates in air. Kocher and Sjoreen (1985) demonstrated that 1 m of soil reduces the absorbed gamma dose rate in air to 0.01% of the total absorbed dose rate produced in the absence of the soil cover.

Thus, in our case, it is possible to eliminate layer 3 in the calculation of the external gamma dose rate because its contribution will be negligible and to assume (for an easier calculation) that the PG layer (layer 2) is of infinite depth beneath the 25-cm soil cover. Consequently, if a monoenergetic gamma-emitter, X_k, is distributed homogeneously in each of the two blocks, the first layer extended from 0 to a cm and the second from a to z cm, respectively, with different activity concentrations $C(0, a; X_k)$ (Bq m^{-3}) and $C(a, z; X_k)$ (Bq m^{-3}), the contributions of both sources to the total dose rate (once the DRF for this emitter is known in each layer) can be calculated from

$$\begin{aligned} D(0, z; X_k) &= D(0, a; X_k) + D(a, z; X_k) \\ &= DRF(0, a; X_k)C(0, a; X_k) \\ &\quad + DRF(a, z; X_k)C(a, z; X_k). \end{aligned} \quad (2)$$

But these DRF are only tabulated for layers from the interface (0 cm) to specific depths, z. This problem can be solved applying the superposition principle, considering that the dose rate from the a–z PG layer with $C(a, z; X_k)$ concentration, $D(a, z; X_k)$, is the difference between the contributions of a PG block extended from 0 to z cm, $D'(0, z; X_k)$, and another PG block with 0–a in thickness, $D'(0, a; X_k)$, both containing a $C(a, z; X_k)$ concentration. So, it is determined that

$$\begin{aligned} D(a, z; X_k) &= D'(0, z; X_k) - D'(0, a; X_k) \\ &= [DRF(0, z; X_k) \\ &\quad - DRF(0, a; X_k)]C(a, z; X_k) \\ &= DRF(a, z; X_k)C(a, z; X_k), \end{aligned} \tag{3}$$

and, consequently, we can obtain the $DRF(a, z; X_k)$ in function of the tabulated $DRF(0, z; X_k)$, which allows us to determine the contribution to the total absorbed dose of a layer (a, z) with an activity concentration $C(a, z; X_k)$, in Bq cm^{-3}, for the considered radionuclide. In our case, the densities of both soil and PG layers were 1.40 and 1.25 g cm^{-3}, respectively.

Then, by combining eqns (2) and (3), the total terrestrial dose rate in air at 1 m above the restored zone produced by a monoenergetic gamma-emitter radionuclide, X_k, can be calculated as follows:

$$\begin{aligned} D(0, z; X_k) &= DRF(0, a; X_k)C(0, a; X_k) \\ &\quad + [DRF(0, z; X_k) - DRF(0, a; X_k)] \\ &\quad \times C(a, z; X_k). \end{aligned} \tag{4}$$

However, DRFs are only tabulated for some specific energies and depths. For that reason it is necessary to obtain them as a function of the energy for each depth 0–z. For each layer (0–z cm), we have fitted the tabulated DRF values on the energy using the following polynomial functions:

$$DRF(0, z; E) = \sum_{i=0}^{n} c_i(z)\, E^i. \tag{5}$$

In this last equation, $n = 1$ for $E > 100$ keV (linear function), and $n = 2$ (quadratic) for $E < 100$ keV, while $c_i(z)$ are parameters independent on the energy. Table 1 shows the parameters of the linear fit ($E > 100$ keV) used to calculate the DRF. The goodness of fits is reflected in the high regression coefficients, which are all higher than 0.99. Similar results were obtained for gamma-energies lower than 100 keV.

Table 1. c_0, c_1 and r regression coefficients obtained in the linear fittings of eqn (5) used to calculate the DRF for different blocks $(0, z)$ and $E > 100$ keV.

Layer	c_0 (Gy y^{-1})/ (Bq cm^{-3})	c_1 (Gy y^{-1})/ (MeV Bq cm^{-3})	r
(0,25 cm)	3.70×10^{-5}	1.43×10^{-3}	0.997
(0,40 cm)	-1.20×10^{-5}	1.61×10^{-3}	0.998
(0,50 cm)	-3.01×10^{-5}	1.66×10^{-3}	0.9990
(0,60 cm)	-4.30×10^{-5}	1.69×10^{-3}	0.9991
(0,80 cm)	-5.42×10^{-5}	1.72×10^{-3}	0.9994
(0,100 cm)	-5.83×10^{-5}	1.73×10^{-3}	0.9994
(0,160 cm)	-5.97×10^{-5}	1.73×10^{-3}	0.9995
(0,∞)	-5.97×10^{-5}	1.73×10^{-3}	0.9995

Consequently, if a radionuclide X_k is considered homogeneously distributed in the layer 0–z, with m_k gamma-emissions at energies $E_{j,k}$ ($j = 1,2,\ldots, m_k$) and probability emissions $P_{j,k}$, the DRF for this isotope can be calculated from

$$DRF(0, z; X_k) = \sum_{j=1}^{m_k} P_{j,k} \sum_{i=0}^{n} c_i(z)\, E^i_{j,k}. \tag{6}$$

Finally, if we consider all the gamma emissions from all the radionuclides (k) of the different natural series that can be present in the source, the total external dose rate at 1 m over ground produced by a block (0–z) can be calculated as

$$\begin{aligned} D(0, z) = &\sum_{k=1}^{N_{U-238}} \left[\sum_{j=1}^{m} P_{j,k} \left(\sum_{i=0}^{n} c_i(z) E^i_{j,k} \right) \right] C(0, z; X_k) \\ &+ \sum_{k=1}^{N_{U-235}} \left[\sum_{j=1}^{m} P_{j,k} \left(\sum_{i=0}^{n} c_i(z) E^i_{j,k} \right) \right] C(0, z; X_k) \\ &+ \sum_{k=1}^{N_{Th-232}} \left[\sum_{j=1}^{m} P_{j,k} \left(\sum_{i=0}^{n} c_i(z) E^i_{j,k} \right) \right] C(0, z; X_k) \\ &+ P_{K-40} \left(\sum_{i=0}^{n} c_i(z) E^i_{K-40} \right) C(0, z; X_{K-40}), \end{aligned} \tag{7}$$

where $P_{j,k}$ is the j gamma emission probability of the k radionuclide (with m_k gamma emissions) belonging to ^{238}U, ^{235}U, or ^{232}Th series, which contain a total of $N_{U\text{-}238}$, $N_{U\text{-}235}$, and $N_{Th\text{-}232}$ radionuclides, respectively.

Then, by combining eqns (4) and (7), applying them in our specific study by adding the contributions of the different radionuclides in every layer and grouping the contributions of the radionuclides that are present with the same concentration because of they are in secular equilibrium (half-life smaller than four times the age of the piles), we obtain the following expressions that give the total external dose rate (in Gy y^{-1}) for both soil and PG layers:

Soil layer: 0–25 cm.

$$\begin{aligned} D_{soil}(0, 25\text{ cm}) = &\ 9.21 \times 10^{-8} C(0, 25; {}^{232}\text{Th}) \\ &+ 3.52 \times 10^{-3} C(0, 25; {}^{228}\text{Ra}) \\ &+ 2.06 \times 10^{-5} C(0, 25; {}^{238}\text{U}) \\ &+ 2.45 \times 10^{-7} C(0, 25; {}^{234}\text{U}) \\ &+ 2.57 \times 10^{-3} C(0, 25; {}^{226}\text{Ra}) \\ &+ 1.50 \times 10^{-6} C(0, 25; {}^{210}\text{Pb}) \\ &+ 8.16 \times 10^{-4} C(0, 25; {}^{235}\text{U}) \\ &+ 2.37 \times 10^{-4} C(0, 25; {}^{40}\text{K}). \end{aligned} \tag{8}$$

PG layer :25–∞ cm.

$$\begin{aligned} D_{PG}(25, \infty) = {} & 3.14 \times 10^{-16} C(25, \infty; {}^{232}\mathrm{Th}) \\ & + 2.02 \times 20^{-5} C(25, \infty; {}^{228}\mathrm{Ra}) \\ & + 1.81 \times 10^{-6} C(25, \infty; {}^{238}\mathrm{U}) \\ & + 1.70 \times 10^{-8} C(25, \infty; {}^{234}\mathrm{U}) \\ & + 3.08 \times 10^{-4} C(25, \infty; {}^{226}\mathrm{Ra}) \\ & + 1.12 \times 10^{-7} C(25, \infty; {}^{210}\mathrm{Pb}) \\ & + 7.31 \times 10^{-5} C(25, \infty; {}^{235}\mathrm{U}) \\ & + 3.63 \times 10^{-5} C(25, \infty; {}^{40}\mathrm{K}), \end{aligned} \quad (9)$$

where $C(a, b; X)$ is the activity concentration (in Bq cm^{-3}) of the radionuclide X on the supposition of an homogeneous concentration in the considered layer.

RESULTS AND DISCUSSION

In-situ measurements

The outdoor terrestrial gamma dose rate values obtained in the restored zones by *in-situ* measurements are compiled in Table 2. The value assigned in Table 2 to each one of the 52 regions is the average of the 10 measurements done in each region. An average value of 0.40 ± 0.03 mGy y^{-1} (46 nGy h^{-1}; the uncertainty is the standard deviation of its average value) can be assigned to the restored zone. This value is similar to those obtained in gardens from the nearby Huelva town, showing the effectiveness of the restoration in decreasing the outdoor terrestrial gamma dose rate.

On the contrary, the average absorbed dose rate at 1 m over the unrestored piles was 2.48 ± 0.15 mGy y^{-1}, which is about six times higher than the rate measured in the restored area.

It is interesting to note the large range in the values obtained for the gamma dose rate in the restored zone, from 0.12 to 0.74 mGy y^{-1}. These results are not surprising considering the following facts:

1. The different thickness of the soil layers measured over the PG piles in the seven sampling points: from 19 to 36 cm; and
2. The phosphogypsum deposited in this zone was accumulated during several years. In addition, and as it was stated in the introduction, the piles can also contain wastes from the Cu-extraction factory. These Cu-wastes contain significantly lower amounts of radioactive nuclides than the phosphogypsum so that (depending on their proportion in the piles) variable dilution degrees of the radionuclide in PG are generated. For example, in 14 samples the measured ^{226}Ra activity concentrations ranged from 360 to 1,320 Bq kg^{-1}. This radionuclide and its progeny are the major contributors to the external gamma dose rate as it will be demonstrated.

The fluctuations of the gamma dose rate values in the restored zone are essentially governed by the dispersion of radionuclide concentrations in the piles and the thickness of the soil layer.

Application of the dosimetric model

Since the total external dose rate in the restored zone comes from the two considered contributions (the soil cover and the underlying phosphogypsum block), we can apply the model that was previously explained and developed for this system.

Table 3 shows the average concentrations obtained for the soil layer as well as the average values obtained in the phosphogypsum layers (the uppermost and deepest ones). These values have been used for the determination of the outdoor external gamma dose rates through the previously explained dosimetric model and explicitly through the application of the eqns (8) and (9). Obviously, due to the high dispersion of the radionuclide concentrations in the PG piles and the hypothesis of the dosimetric model, it makes no sense to do a point-by-point comparison. So, the average results for the external gamma dose rates calculated with the model and the *in-situ* measurements will be compared.

Introducing the average activity concentrations, we have obtained a contribution to the external gamma dose rate from the cover soil of 0.17 mGy y^{-1} and for the underlying phosphogypsum layer 0.24 mGy y^{-1}, which gives a total external gamma dose rate of 0.41 mGy y^{-1}—in a good agreement with the *in-situ* average determination in the PG piles. This value is similar to the obtained one in the gardens of Huelva town (average of 0.40 mGy y^{-1}) where the natural radioactivity content in these gardens is about double that in the soil used for the covering of the PG piles, and it is also in agreement with the external dose rates measured in many parts of the world (UNSCEAR 1988; Baeza et al. 1993; Leung et al. 1990).

Table 2. External gamma dose rates measured by *in-situ* measurements (mGy y^{-1}) at 1 m above the covered piles and its location code.

M1	**M2**	**M3**	**M4**	**M5**	**M6**	**M7**	**M8**	**M9**	**M10**	**M11**	**M12**	**M13**
0.67	0.12	0.46	0.30	0.34	0.37	0.39	0.18	0.37	0.41	0.42	0.55	0.27
M14	**M15**	**M16**	**M17**	**M18**	**M19**	**M20**	**M21**	**M22**	**M23**	**M24**	**M25**	**M26**
0.66	0.18	0.60	0.50	0.60	0.69	0.41	0.72	0.53	0.21	0.51	0.46	0.48
M27	**M28**	**M29**	**M30**	**M31**	**M32**	**M33**	**M34**	**M35**	**M36**	**M37**	**M38**	**M39**
0.34	0.37	0.25	0.42	0.14	0.27	0.39	0.46	0.23	0.25	0.74	0.30	0.20
M40	**M41**	**M42**	**M43**	**M44**	**M45**	**M46**	**M47**	**M48**	**M49**	**M50**	**M51**	**M52**
0.55	0.25	0.25	0.37	0.20	0.32	0.49	0.63	0.67	0.35	0.34	0.28	0.63

Table 3. Average radionuclide concentrations (Bq kg^{-1}) determined in the soil and the phosphogypsum layers in the seven collected cores.

Layer	^{238}U	^{234}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra	^{228}Th	^{40}K
Soil	25	22	14	21	13	13	13	166
Gypsum	200	200	600	600	4	11	4	<30

Table 4. External gamma dose rates at 1 m on the covered piles (mGy y^{-1}) obtained through the dosimetric model and the specific activities in the different cores.

Core	A	B	C	D	E	F	G
Soil layer	0.194	0.227	0.167	0.156	0.127	0.161	0.107
Gypsum layer	0.177	0.284	0.270	0.314	0.192	0.190	0.228
Total	0.370	0.511	0.437	0.471	0.319	0.352	0.335

The model has been also applied core by core giving the results shown in Table 4. A high dispersion of results is observed, so that the calculated external gamma dose rates in the seven cores range from 0.32 to 0.51 mGy y^{-1}. This fact agrees with the observed dispersion by the *in-situ* measurements as shown in Table 2.

If we replace the PG layer by the soil used to cover the former, the total dose rate calculated through the model will be 0.21 mGy y^{-1}. Therefore, we can conclude that the increase produced by the underlying PG layer is about 0.20 mGy y^{-1}, a dose rate smaller than the maximum values recommended for the public (ICRP 1990).

If the cover soil is eliminated, the remaining PG source becomes an infinite-thickness block source (from the point of view of the application of the dosimetric model). Making the respective calculations (considering the average activity concentrations shown in Table 3), a total external gamma dose rate of 2.23 mGy y^{-1} is calculated, which is in agreement with the average dose measured *in situ* at the uncovered piles of 2.48 ± 0.15 mGy y^{-1}.

Table 5 shows the average contributions of the different subseries in the total external gamma dose rate produced by soil and PG layers. For the soil layer (0–25 cm) similar contributions of the U-, Th-series and ^{40}K are obtained, being about 0.06 mGy y^{-1} for each one of them. On the contrary, for the PG layer about 99% of the total external gamma dose (0.23 mGy y^{-1}) is due to the ^{226}Ra and its short half-life progeny, since the activity concentrations in the PG of Th-series radionuclides and ^{40}K are very low, as shown in Table 3.

Once the model can be considered validated, and as an example of its possible predictive capacity, it can be questioned what thickness of a soil layer covering the phosphogypsum piles is necessary to decrease the PG contribution to the external gamma dose rate below a desirable level. The contribution to the external dose rate of the PG block for different soil thicknesses was determined in a similar way in that the soil layer was assumed to be 25 cm thick. In Table 6 the respective contributions from PG to the external gamma dose rates are presented for different thicknesses of soil coverage. From these data, it is demonstrated that our model can evaluate the necessary thickness of soil to reduce the PG contribution in the gamma dose rate below any fixed maximum level.

CONCLUSION

A dosimetric model has been developed to evaluate the external gamma dose rates in air at 1 m above the ground. This model considers a flat radioactive source conformed by two blocks, the uppermost layer of soil with a thickness of *a* cm and another one conformed by the underlying PG block. Based on this model, it is possible to calculate the external dose rate as a function of the thickness of soil layer; therefore, we could estimate the cover soil thickness necessary to obtain a desired dose level.

This model has been applied to the 450 ha of PG piles with a 25 cm soil cover. The average of the total outdoor gamma dose rate calculated from the radionuclide concentrations by the developed model (0.41 mGy y^{-1}) is in agreement with the average obtained from *in-situ* measurements done with an environmental monitor (0.40 mGy y^{-1}). This agreement gives confidence and validates the methodology used in this work.

It was also demonstrated that the contribution of the soil layer is similar to the contribution of the underlying

Table 5. Terrestrial external gamma dose rates (mGy y^{-1}) for the different subseries calculated from the specific activities determined in the collected cores.

Layer	^{238}U	^{234}U	^{226}Ra	^{210}Pb	^{232}Th	^{228}Ra	^{228}Th	^{40}K
Soil	7.21×10^{-4}	7.56×10^{-6}	5.03×10^{-2}	4.41×10^{-5}	1.68×10^{-6}	2.53×10^{-2}	3.88×10^{-2}	5.46×10^{-2}
Gypsum	4.52×10^{-4}	—	2.31×10^{-1}	8.42×10^{-5}	—	2.08×10^{-3}	1.39×10^{-3}	—
Total	1.17×10^{-3}	7.56×10^{-6}	2.81×10^{-1}	1.28×10^{-5}	1.68×10^{-6}	2.74×10^{-2}	4.02×10^{-2}	5.46×10^{-2}

Table 6. Theoretical contribution to the external terrestrial gamma dose rate due to the ^{238}U series from the underlying phosphogypsum block which extends in deep between a and ∞.

Layer limits (a cm, ∞)	(25,∞)	(40,∞)	(50,∞)	(60,∞)	(80,∞)	(100,∞)	(160,∞)
Dose rate (mGy y^{-1})	0.230	0.086	0.047	0.025	0.0076	0.0013	2.2×10^{-8}

PG layer. The substitution in the model of the underlying PG by a similar layer of the soil used for the covering of the piles, allows the determination that PG contributes to an increase in absorbed dose rates of 0.20 mGy y^{-1}. Additionally, most of the PG contribution (about 99%) to the absorbed gamma dose rate is due to the gamma-emitters from U-series, mainly coming from the ^{226}Ra and its short half-life progeny.

Further model calculations indicated that if the covering is eliminated, the calculated dose rate produced by the PG will be 2.20 mGy y^{-1}, which is also in agreement with the experimental average value measured in uncovered PG piles. More than 95% of this dose rate also is produced by ^{226}Ra and progeny.

And, finally, it is interesting to note that in spite of the fact of the similar contributions of the PG and soil layers, the obtained total dose rate is in agreement with the values measured in Huelva town and in many parts of the world. This result is due to the fact that the radionuclide concentrations in the soils used to cover the piles have approximately the half-value of the soils of Huelva town.

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