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## Effective Additional Gamma Dose for General Population and Workers from a Mexican Radioactive Waste Site

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**Abstract:** The environmental effective gamma-dose rate has been determined at the Mexican Storage Centre for Radioactive Waste (SCRW) and surrounding communities. Soil samples were analysed for <sup>226</sup>Ra, <sup>235</sup>U, <sup>137</sup>Cs and <sup>40</sup>K concentration activities. The outdoor effective gamma dose rate was higher at the SCRW where industrial radioactive sources and uranium ore piles were stored in the past. Some nearby locations, where the action of the rain on the piles promoted the transport and surface migration of <sup>226</sup>Ra, <sup>235</sup>U and <sup>137</sup>Cs, also showed an increased gamma dose rate. The gamma dose rate at distant communities (local background) was evaluated with data obtained along 10 years measurements. The additional effective external dose for the general public at the 200 m neighbouring zone of the site, was two times higher than that from the local background. The outdoor average external effective dose for workers due to the SCRW operation was 20 times than for the local background.

**Key words:** Gamma dose rate, radioactive waste, soil

### INTRODUCTION

External irradiation from radionuclides naturally present in the environment or released from man-made practices or events is usually an important component of the exposure of human populations. The measurement of environmental gamma dose rate can give important information for the evaluation of general population and workers external irradiation. Several industries have been identified to produce high levels of Technologically Enhanced Naturally Occurring Radionuclide Materials (TENORM). Such is the case of uranium mining and milling, metal mining and smelting and phosphate industry<sup>[1]</sup>. Other industries with potential radiological impact on man and environment are: coal mining, oil and gas drilling, zirconium and ceramics' industry, pulp and paper mills and other applications of natural radium and thorium<sup>[2]</sup>.

Natural world median concentration value of <sup>40</sup>K in the soil is 400 Bq kg<sup>-1</sup> and for <sup>238</sup>U, <sup>226</sup>Ra and <sup>232</sup>Th 35, 35 and 30 Bq kg<sup>-1</sup>, respectively<sup>[3]</sup>. <sup>235</sup>U, is much less abundant than <sup>238</sup>U, the isotopic abundance (by weight) and the activity ratio of natural uranium (<sup>235</sup>U/<sup>238</sup>U) are 0.0072 and 0.046, respectively<sup>[4]</sup>.

After the uranium extraction from the ore, the waste residues and the unextracted uranium fraction are deposited in tailings. These tailings contain natural

radionuclides such as <sup>238</sup>U, <sup>226</sup>Ra and <sup>210</sup>Pb, which may provide a significant source of environmental and food chain contamination. Ore extraction and processing may lead to TENORM in products, by-products and waste, in facilities and surroundings. Since the early 1990's, several international teams have studied the potential danger for the population living in the immediate vicinity of tailings and the urgent and long-term actions needed to improve the situation created by former uranium mining and milling operations. Where people are employed in mineral processing or when waste from mining, processing and construction are disposed off, they also may be exposed to radiation burden. Taking into account the international recommendations<sup>[3]</sup>, each country can adopt secondary levels of dose and derived limits, including registration, investigation, intervention and remedial action levels.

Activity limits by which states with TENORM regulations have exempted solid materials include 185 Bq kg<sup>-1</sup> for <sup>226</sup>Ra<sup>[2]</sup>. Secondary levels for the specific activity in soil are derived with 200 Bq kg<sup>-1</sup> for unrestricted use and 1000 Bq kg<sup>-1</sup> for restricted use, where the radionuclide of the <sup>238</sup>U-series with the highest specific activity is decisive<sup>[5]</sup>.

At the Mexican Storage Centre for Radioactive Waste Site (SCRW), solid and liquid radioactive waste have been stored during four decades and uranium ore tailing piles also stood for some time before being buried in trenches

and specific containers. A contamination in the soil with  $^{137}\text{Cs}$  also occurred at the SCRW, in the decade of 1970, as a consequence of a broken industrial source. The present study is a review of external annual doses from sources of natural and artificial origin at SCRW and surrounding communities, where an environmental monitoring program has been systematically conducted during more than 10 years<sup>[6]</sup>. Intervention and decontamination actions, with removal of the upper soil layer, were performed at the site from 1993 to 2000. Several kinds of samples have been collected and analysed in this period and changes in concentrations of  $^{226}\text{Ra}$ ,  $^{235}\text{U}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in the soil, as well as in bioindicator samples, were reported<sup>[7-9]</sup>. In the present study the effective gamma dose rates in air were determined in order to estimate the effective additional gamma doses received by the general public and workers due to the mining past activities and SCRW operation.

## MATERIALS AND METHODS

**The site:** The SCRW site ( $19^{\circ} 47' 21'' \text{N}$ ;  $98^{\circ} 50' 15'' \text{W}$ ) is located at the State of Mexico, Mexico, at altitudes between 2470 and 2490 m, in the middle part of the Mexican Neovolcanic belt (Fig. 1a). The climate is temperate sub-humid with an average yearly temperature of  $14^{\circ}\text{C}$ , ranging from 6 to  $32^{\circ}\text{C}$ . The mean annual precipitation is 638.5 mm, occurring mainly during the rainy season, from June to October. The local hydrogeology shows superficial tuffs and basalts and no evidence exists of aquifers 30 km around the site.

The gamma-absorbed dose rates in air were determined based on topographic location and exposure rate magnitudes measured outdoors. The sampling period considered in the present study include 1991 to 2002. Figure 1b indicates the sampling zone around the SCRW, denominated Zone VI, where four distant communities, Teacalco, Maquixco, Colhuacan and Aztecameca, in a 10 km radius around the site, are found. Gamma dose rates at these communities correspond to the local background. In Fig. 1c, the sampling points from the SCRW are shown: Zone I, having an area of  $7372 \text{ m}^2$ , correspond to the region where the ore was piled and the  $^{137}\text{Cs}$  contamination occurred; Zone II ( $4500 \text{ m}^2$ ), at a lower altitude than Zone I was a private property until 1993, but the transport of contaminated soil due to rain water flowing through the natural slopes of the terrain, urged decommission and payment of the zone and its inclusion as part of the SCRW. Zone III ( $3000 \text{ m}^2$ ), at the bottom of the slope, corresponds to the place where rain water accumulates before leaving the SCRW and finally Zone IV ( $155\,600 \text{ m}^2$ ) that include all other sampling points inside the SCRW. Five sampling points are also located in

a 200 m zone neighbouring of the site (Zone V,  $14\,000 \text{ m}^2$ ); this external land is a private property used for local agricultural practices.

**External gamma dose rates in air obtained by direct measurements:** One of the methods for evaluating external exposures from naturally occurring radionuclides consists to summarize directly gamma dose rates in air and to subtract the dose rate due to cosmic rays. Outdoor direct measurements of external gamma exposure and absorbed dose rates in air were performed using passive integrating Thermoluminescent Dosimeters (TLD's) of  $\text{CaSO}_4: \text{Dy} + \text{PTFE}$ , developed locally by Azorin and Gutierrez<sup>[10]</sup>. This TLD's fulfill the ANSI-N-545 code for environmental monitoring. Each dosimetric-plastic package contains two pellets, that were placed 1 m above the earth surface and exposed during three periods of 4 months each year. TLD's were read-out in a Harshaw analyzer Model 4000 coupled to a PC; nitrogen gas was allowed to flow into the reader during read-out to avoid any spurious signal. The outdoor exposure rates measured include the contribution from cosmic and terrestrial radiation. The outdoor gamma dose rate ( $D\gamma$ ) in  $\text{nGy h}^{-1}$  was calculated from gamma exposure rate measurements (X) as:  $D\gamma = X \times 0.0087 \times 10^{-9} \text{ Gy R}^{-1}$ ; a coefficient of  $0.7 \text{ Sv Gy}^{-1}$  to convert absorbed dose in air to effective dose equivalent and effective dose was used in accordance to UNSCEAR<sup>[3]</sup>. To compare the different methods of measurement (passive devices and electronic detectors), some sporadic determinations were also taken with a High Pressure Ionization Chamber (HPIC) Reuter-Stokes type (model RSS-112).

**External gamma dose rates in air from concentration of radionuclides in soil measurements:** The other method to evaluate the external exposure is to calculate the external gamma dose rates in air from measurements of the concentrations of the relevant radionuclides in soil. Several soil samples have been collected and analyzed for  $^{235}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$ . The soil samples were dried at  $105^{\circ}\text{C}$  and sieved through a 50-60 mesh sieve before being analyzed in the laboratory. The (dry/wet) weight ratio was recorded, obtaining an average conversion factor for dry to wet basis=0.8. The samples were analyzed for  $^{235}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  specific activities, with a HPGe detector having 29.4% relative efficiency. Aliquots of 300-350 g were kept in a 500 mL Marinelli beaker for a period of more than 21 days to allow radium and radon to reach the equilibrium. The measurement time was from 1000 to 60 000 s and appropriate standard mixtures of  $\gamma$ -rays emitting isotopes were used to calibrate the detectors.  $^{235}\text{U}$  was measured through its very

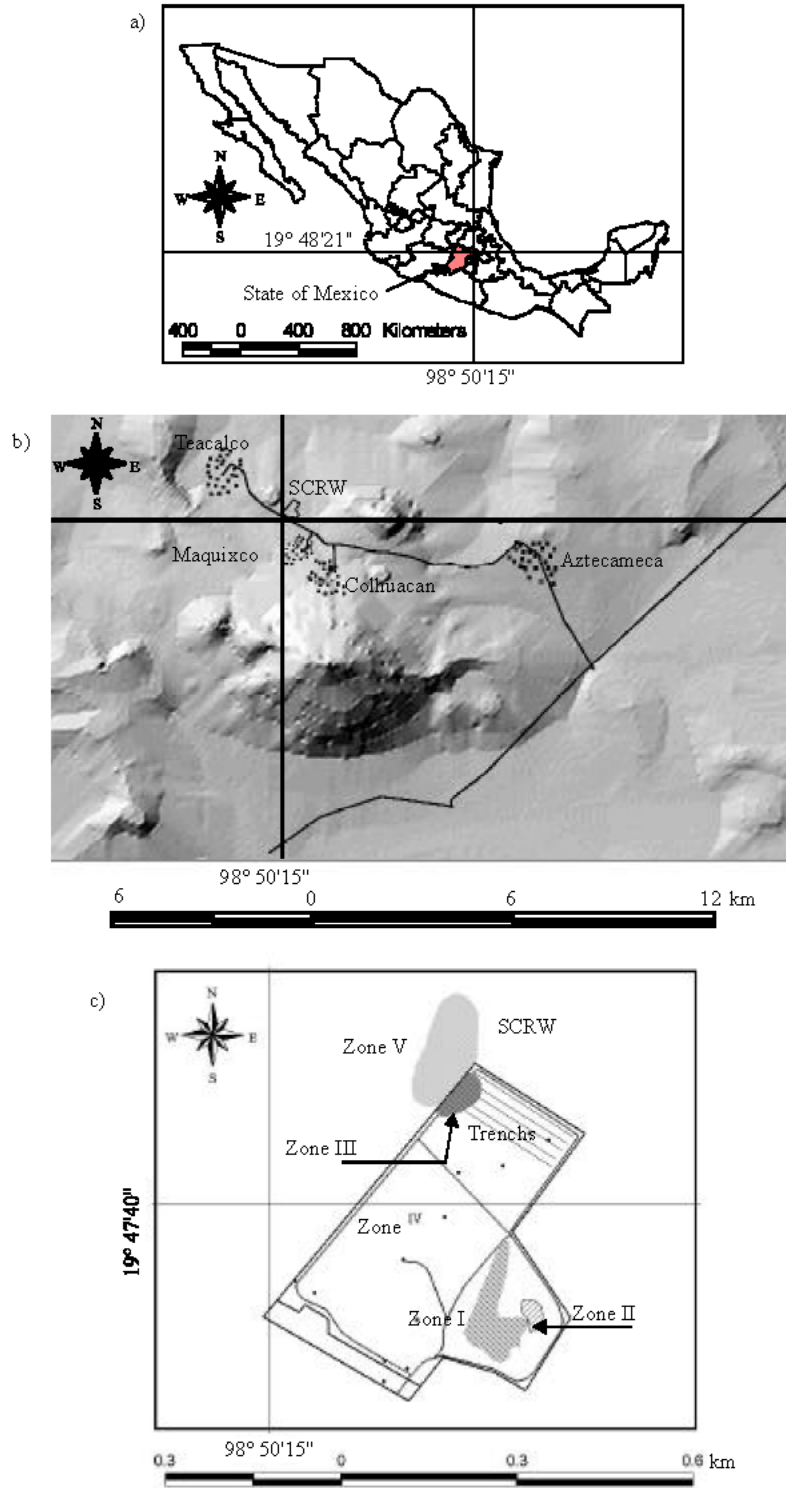


Fig. 1a): Location of the Mexican Storage Centre for Radioactive Waste (SCRW) at the state of Mexico, Mexico; b) Zone VI sampling points at distant communities (10 km radius around the SCRW); c) Zone I, II, III and IV sampling points inside the SCRW; Zone V at the border line to the SCRW

abundant 186 keV gamma-ray line, calculating the radium contribution to the radiation, using the observed gamma rays of the 214 chain and subtracting that contribution from the total, to obtain the part due to  $^{235}\text{U}$ <sup>[11]</sup>.

The absorbed gamma dose rate in air from concentrations of terrestrial radionuclides in the soil has been calculated with the equation from UNSCEAR<sup>[3]</sup> and Tosheva *et al.*<sup>[12]</sup>.

$$D\gamma = 0.0417A_{40\text{K}} + 0.604A_{232\text{Th}} + 0.462A_{226\text{Ra}} + 0.001A_{137\text{Cs}} \quad (1)$$

where:

$D\gamma$  is the absorbed gamma dose rate in air ( $\text{nGy h}^{-1}$ ) and  $A_i$  is the activity concentration of the  $i$ th radionuclide in soil samples ( $\text{Bq kg}^{-1}$ ).

## RESULTS

Results showed that the distant communities (Zone VI) and some sampling points located inside of the SCRW, can be considered as background zones with normal radionuclide concentrations, confirming a normal local outdoor gamma dose rate in air that arises from terrestrial radionuclides present at trace levels in the soil (Table 1). At SCRW the average gamma dose rate value was  $145 \pm 8 \text{ nGy h}^{-1}$ , (range:  $60\text{-}517 \text{ nGy h}^{-1}$ ), twice as compared with the local background ( $84 \pm 5 \text{ nGy h}^{-1}$ ) and five times higher if the maximum values are considered. For TLD the maximum value obtained at Zone I in 1991 ( $517 \text{ nGy/h}$ ) was lower, by a factor of 0.67, than the value measured with the Reuter-Stokes pressurized ionization chamber (HPIC). A similar behaviour was observed at other SCRW measurement stations and at Zone V, so that a correction factor of 0.7 can be considered between the two measurement methods.

Exposure to cosmic rays is strongly dependent on altitude and weakly dependent on latitude. At sea level, a representative value of  $32 \text{ nGy}^{-1}$  for the absorbed dose rate in free air is reported by UNSCEAR<sup>[3]</sup>. From the direct ionizing and photon component of cosmic radiation, an absorbed dose rate value of  $74.88 \text{ nGy}^{-1}$  (2.34 times higher than that at sea level) is obtained at Zone VI, using the method for the altitude correction<sup>[3]</sup>. The geomagnetic latitude effect is about 10%, so that a value of  $30 \text{ nGy}^{-1}$  is appropriate for latitudes below  $30^\circ$ . Applying the latitude correction factor the local background gamma dose rate outdoors is  $67 \text{ nGy}^{-1}$ .

At Zone VI, the directly measured external gamma dose rate in air, applying the correction factor of 0.7, is  $120 \text{ nGy}^{-1}$ . Subtracting the estimated dose rate due to cosmic rays ( $67 \text{ nGy h}^{-1}$ ), the external exposure from terrestrial radionuclides would be  $53 \text{ nGy h}^{-1}$ .

The  $^{226}\text{Ra}$  average value from 1991 to 2002 was  $27 \pm 3 \text{ Bq kg}^{-1}$  (d.w.) but considering the maximum values the average value was  $40 \pm 9 \text{ Bq kg}^{-1}$  (d.w.) (Table 2).  $^{137}\text{Cs}$  average value was  $3 \pm 1$  (range from 1 to 10)  $\text{Bq kg}^{-1}$  (d.w.), indicating a slight decrease along the time, from long term fallout.  $^{40}\text{K}$  average value was  $303 \pm 25$  (range from 187 to 435)  $\text{Bq kg}^{-1}$  (d.w.), in agreement with the North America mean value<sup>[3]</sup>.  $^{232}\text{Th}$  appears to be essentially in secular equilibrium with its entire decay chain and gamma radiation from  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  may all be used in its determination<sup>[11]</sup>. The average values of those radionuclides, measured during 1991 were  $22 \pm 3$ ,  $21 \pm 2$  and  $9 \pm 1 \text{ Bq kg}^{-1}$  (d.w.), respectively.  $^{232}\text{Th}$  average value of  $22 \text{ Bq kg}^{-1}$  (d.w.), was considered to calculate the gamma dose rate in air inferred from concentrations of radionuclides in soil by equation (1). The activity concentration of  $^{235}\text{U}$  was below the limit of detection but a concentration of  $1.2 \text{ Bq kg}^{-1}$  (d.w.) could be considered from the median natural  $^{235}\text{U}/^{226}\text{Ra}$  isotopic ratio of 0.046.

At Zone VI, the average value of external gamma dose rates in air from measurements of the concentration of radionuclides in the soil was  $38 \pm 2 \text{ nGy h}^{-1}$  ranging from 27 to  $54 \text{ nGy h}^{-1}$ .

$^{226}\text{Ra}$  average activity concentrations diminished from  $3738 \pm 230 \text{ Bq kg}^{-1}$  (d.w.) in 1991-1995 to  $916 \pm 117 \text{ Bq kg}^{-1}$  (d.w.) in 2002 due to the intervention and decontamination actions started in 1993 and due to soil removing during 2000 and 2001 (Table 3).  $^{137}\text{Cs}$  decreased from  $3273 \pm 179 \text{ Bq kg}^{-1}$  (d.w.) in 1991-1995 to  $61 \pm 18 \text{ Bq kg}^{-1}$  (d.w.) in 2002.  $^{40}\text{K}$  average values were similar to the regional background values and constant along the time. The average values of  $^{235}\text{U}$  was  $70 \pm 8 \text{ Bq kg}^{-1}$  (d.w.) ranging from 15 to  $168 \text{ Bq kg}^{-1}$  (d.w.) in 1991-1995. An average value of  $24 \pm 6 \text{ Bq kg}^{-1}$  (d.w.) ranging from  $6\text{-}65 \text{ Bq kg}^{-1}$  was measured in 1996-2002 period. The  $^{235}\text{U}/^{226}\text{Ra}$  isotopic ratio ranged between 0.012 and 0.043. The lowest isotopic ratio value obtained (0.012) was lower than that expected from natural uranium. However, the average values of  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$  and  $^{208}\text{Tl}$  were  $27 \pm 5$ ,  $19 \pm 2$  and  $11 \pm 1 \text{ Bq kg}^{-1}$  (d.w.), respectively similar to the regional background measurements. The average gamma dose rates to workers diminished substantially (from 1759 to  $499 \text{ nGy}^{-1}$ ) since 1991-1995 to 1996-2002 periods (Table 3).

At Zones III ( $N=48$ ) and IV ( $N=92$ ), the  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$  average activity concentration values also diminished from 1991-1995 to 1996-2002. At present time the average concentration values for  $^{226}\text{Ra}$  are slowly higher than from the regional background, while  $^{137}\text{Cs}$  concentration values still significantly higher than the regional background. At Zone III,  $^{226}\text{Ra}$  average and range concentration values from 1991-1995 were  $122 \pm 17$  ( $36\text{-}218$ )  $\text{Bq kg}^{-1}$  (d.w.) while from 1996-2002 were  $53 \pm 14$  ( $20\text{-}115$ )

Table 1: Outdoor gamma dose rates and annual effective dose estimated from direct measurements with TLD's at Zone VI (local background) and at SCRW (Zones I to IV)

Site	Sample size (N)	Latitude	Altitude (m)	Gamma dose rate (nGy h <sup>-1</sup> )		Effective dose (mSv y <sup>-1</sup> )	
				Average	Range	Average	Range
Zone VI	34	19° 46'-19° 48'	2400-2480	84±5	66-96	0.52± 0.04	0.32-0.59
Zone I	40	19° 47'31''	2491	229±11	80-517	1.40±0.08	0.39-3.17
Zone II	15	19° 47'34''	2488	187±9	168-205	1.15±0.07	0.82-1.26
Zone III	20	19° 47'45''	2472	83±5	76-91	0.51±0.03	0.37-0.56
Zone IV	79	19° 47'30''-19° 47'42''	2476-2490	82±5	60-115	0.50±0.03	0.29-0.71
Average	154	19° 47'30''-19° 47'45''	2476-2491	145±8	60-517	0.89±0.08	0.29-3.17

Table 2: Average activity concentrations of radionuclides in soil (Bq kg<sup>-1</sup> (d.w.)) and outdoor terrestrial gamma dose rate (nGy h<sup>-1</sup>) at Zone VI (local background)

Year	Sample size N	Activity concentrations in soil (average± error) in Bq kg <sup>-1</sup> (d.w.)							
		<sup>226</sup> Ra		<sup>137</sup> Cs		<sup>40</sup> K		Dose rate nGy h <sup>-1</sup>	
		Average	Range	Average	Range	Average	Range	Average	Range
1991-1995	10	25±4	17-32	5±0.5	2-8	288±15	252-347	36.76	32-43
1996	7	30±12	13-47	5±0.7	1-10	347±20	318-387	41.56	33-51
1997	5	33±8	20-57	2±0.4	1-5	298±15	257-332	41.09	33-54
1998	5	27±8	20-33	3±0.8	1-5	300±17	244-360	38.43	33-44
1999	5	25±9	16-29	2±0.6	1-3	333±29	318-345	38.82	34-42
2000	6	26±11	19-41	3±0.9	1-7	272±35	219-358	36.82	31-47
2001	10	22±11	12-39	3±0.4	1-3	282±44	189-419	35.42	27-49
2002	10	27±11	20-40	2±0.5	1-5	303±35	187-435	38.59	30-50
1996-2002	48	27±10	12-57	3±0.6	1-10	305±30	187-435	38.67	27-54

Table 3: Average activity concentrations of radionuclides in soil (Bq kg<sup>-1</sup> (d.w.)) and outdoor terrestrial gamma dose rate (nGy h<sup>-1</sup>) at SCRW (Zones I+II)

Year	Sample size N	Activity concentrations in soil (average± error) in Bq kg <sup>-1</sup> (d.w.)							
		<sup>226</sup> Ra		<sup>137</sup> Cs		<sup>40</sup> K		Dose rate nGy h <sup>-1</sup>	
		Average	Range	Average	Range	Average	Range	Average	Range
1991-1995	36	3738±230	182-19979	3273±179	13-18148	286±55	198-560	1759	1099288
1996	19	2405±132	144-7871	122±10	15-380	305±33	242-403	1140	93-3670
1997	20	1030±49	228-3576	84±8	9-652	298±22	230-430	505	131-687
1998	17	1248±77	91-3370	116±11	11-1035	293±25	257-386	605	69-1590
1999	20	1035±71	92-2571	121±15	12-739	293±28	252-449	507	69-1224
2000	24	885±69	174-2295	110±25	11-1373	300±37	237-396	438	107-1094
2001	23	899±135	115-2160	127±30	9-1952	285±57	223-378	444	79-1032
2002	24	916±117	101-2568	61±18	15-476	300±44	165-388	452	70-1219
1996-2002	147	1019±109	91-7871	152±40	9-1952	286±37	165-449	499	65-3673

Table 4: Average activity concentrations of radionuclides in soil (Bq kg<sup>-1</sup> (d.w.)) and outdoor terrestrial gamma dose rate (nGy h<sup>-1</sup>) at Zone V (neighbouring zone of the SCRW)

Year	Sample size N	Activity concentrations in soil (average± error) in Bq kg <sup>-1</sup> (d.w.)							
		<sup>226</sup> Ra		<sup>137</sup> Cs		<sup>40</sup> K		Dose rate nGy h <sup>-1</sup>	
		Average	Range	Average	Range	Average	Range	Average	Range
1991-1995	10	109±10	38-237	70±3.7	16-160	251±16	179-276	74.04	38-135
1996	4	108±17	77-142	95±5	74-117	238±15	229-250	73.34	59-89
1997	4	117±16	88-146	97±5	76-117	223±15	221-225	76.74	63-90
1998	4	153±18	109-182	115±6	94-148	261±16	230-298	94.93	73-110
1999	4	151±20	134-170	106±7	82-117	250±20	222-302	93.47	85-105
2000	8	93±27	36-154	69±7	9-113	240±26	222-276	66.48	39-96
2001	12	119±28	42-172	62±9	5-116	243±44	198-290	78.41	41-105
2002	9	97±31	39-160	45±6	2-105	234±33	203-279	68.04	40-99
1996-2002	45	120±23	36-182	84±6.6	2-148	241±26	198-302	78.77	38-110

Bq kg<sup>-1</sup> (d.w.). The <sup>137</sup>Cs concentration values were 97±6 (40-158) Bq kg<sup>-1</sup> (d.w.) and 51±7 (6-188) Bq kg<sup>-1</sup> (d.w.) from 1991-1995 and 1996-2002, respectively. At Zone IV <sup>226</sup>Ra average and range concentration values from 1991-1995 were 109±15 (21-258) Bq kg<sup>-1</sup> (d.w.) while from

1996-2002 were 32±10 (12-66) Bq kg<sup>-1</sup> (d.w.). The <sup>137</sup>Cs concentration values were 86±1 (14-136) Bq kg<sup>-1</sup> (d.w.) and 22±3 (2-126) Bq kg<sup>-1</sup> (d.w.) from 1991-1995 and 1996-2002, respectively. <sup>40</sup>K concentrations values were similar at all the monitoring points inside of SCRW.

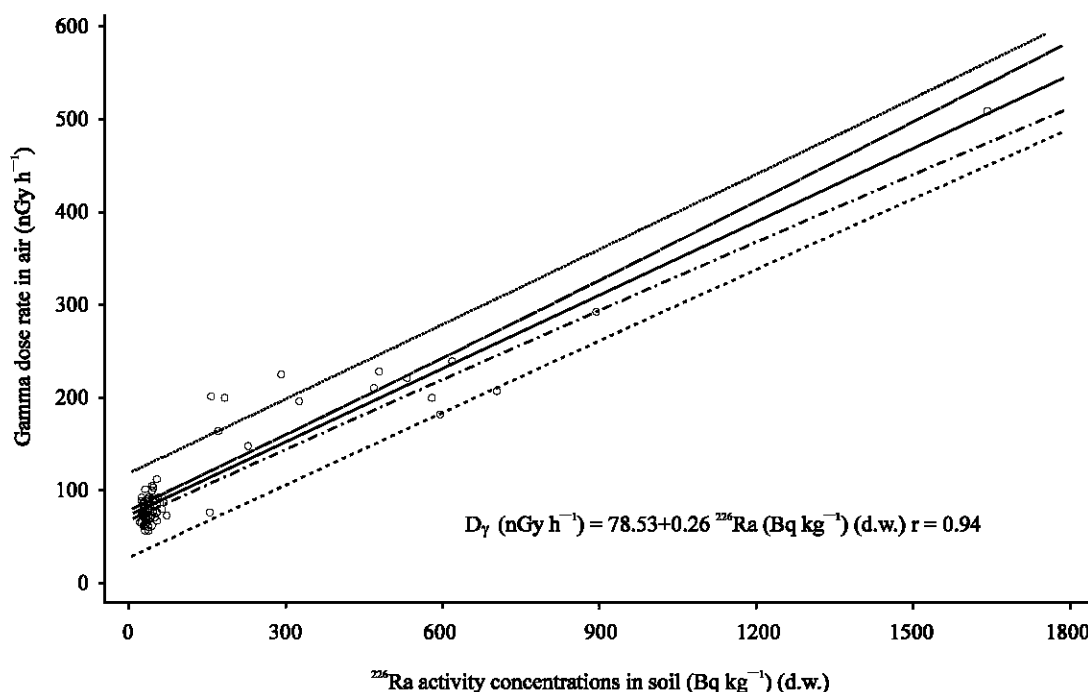


Fig. 2: Comparison of gamma dose rate in air ( $\text{nGy h}^{-1}$ ) obtained with TLD's and  $^{226}\text{Ra}$  activity concentrations in the soil (gamma-ray spectrometry). The regression curve equation and the correlation coefficient are also shown

Determinations of the average activity concentrations of  $^{226}\text{Ra}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in the surface soil of Zone V (200 m zone neighbouring of the site) are shown in Table 4. The results indicate that the soils have a significant content of  $^{226}\text{Ra}$  and  $^{137}\text{Cs}$ . The concentration values have been stable along the years due essentially to the fact that no decontamination activities have been performed at this zone. A small increase was observed during 1998-1999 due probably to superficial streams during the heavy rains from 1998. Derived intervention level adopted by the National Regulatory Commission (CNSNS) at the SCRW is  $130 \text{ Bq kg}^{-1}$ . At Zone V, the maximum value ( $237 \text{ Bq kg}^{-1}$ ) obtained in the period 1991-1995, was higher than the limits adopted internationally to exempt solid materials or as screening level for the unrestricted use ( $185$  and  $200 \text{ Bq kg}^{-1}$ , respectively). The activity concentrations of  $^{137}\text{Cs}$  in the surface soil were higher than for the regional background, while an average value of  $246 \pm 22 \text{ Bq kg}^{-1}$  (d.w.) range ( $179$ - $302 \text{ Bq kg}^{-1}$  (d.w.)) was obtained for  $^{40}\text{K}$ , similar to the local background and lower than the world median concentration value.

The correlation of the gamma dose-rate ( $\text{nGy h}^{-1}$ ) performed with TLD's from 1991-2002 with the same number ( $N=79$ ) for  $^{226}\text{Ra}$  activity concentration values measured in the soil samples ( $\text{Bq kg}^{-1}$  (d.w.)) is depicted in Fig. 2. All the Zones (I to VI) are considered. The regression analysis shows a linear model to describe the

relationship between the outdoor gamma dose rate ( $\text{nGy h}^{-1}$ ) and the  $^{226}\text{Ra}$  concentration values in the soil. The equation of the fitted model is:

$$D\gamma (\text{nGy h}^{-1}) = (78.53 \pm 2.93) + (0.263 \pm 0.01) ^{226}\text{Ra} (\text{Bq kg}^{-1}) (\text{d.w.}) \quad (2)$$

At the 99% confidence level, the correlation coefficient ( $r=0.94$ ) indicate a relatively strong relationship between the variables, since the p-value in the analysis of variance is less than 0.01.

Applying Eq. 2, a value of  $86 \text{ nGy h}^{-1}$  corresponding with the average concentration value of  $^{226}\text{Ra}$  ( $27 \text{ Bq kg}^{-1}$  (d.w.)) measured at the local background was obtained, while at Zone V, an outdoor gamma dose rate of  $141 \text{ nGy h}^{-1}$  was obtained for the maximum value of  $^{226}\text{Ra}$  in soil ( $237 \text{ Bq kg}^{-1}$  (d.w.)) measured in the 1991-1995 period.

## DISCUSSION

The local background (Zone VI) indicates that the soil/direct measurements ratio values was  $54/53 \text{ nGy h}^{-1}=1$  if we consider the maximum value of outdoor absorbed dose rate from radionuclides in soil and the direct measurements of absorbed dose rates in air. Exposures inferred from the soil concentration results generally show reasonable agreement with the measured

outdoor absorbed dose rate in air in many countries and the soil/direct measurements ratio values range from 1.5 to 0.6<sup>[3]</sup>. The difference observed between the measurements carried out with TLD's and with the Reuter-Stokes ionization chamber could be due to the different energy responses, which, for the HPIC, is larger than that for the TLD dosimeters in the energy range from 60 to 300 keV. The results are in agreement with those reported by Losana *et al.*<sup>[13]</sup>, who obtained a factor of 0.77 for the dose rate measurements performed with TLD's GR-200 of LiF: Mg, Cu, P and with a Reuter-Stokes HPIC in Turin, Italy.

In the present study we found a <sup>226</sup>Ra concentration four times higher and a <sup>137</sup>Cs concentration 19 times higher at the neighbouring zone of the SCRW (Zone V) than the local background (Zone VI). At present time there is no population inhabiting Zone V permanently and adverse health effects from increased natural background radioactivity may seem unlikely for the near term. However for a future scenario, where people would live at this zone, adverse health effects for the long-term accumulation of radioactive materials from the SCRW could occur.

To estimate annual effective doses from terrestrial gamma radiation, the corresponding indoor and outdoor occupancy factor must be considered. From the present data and considering the indoor/outdoor ratio of 1.3, reported by UNSCEAR<sup>[3]</sup> from measurements in different countries, the annual effective dose estimated at the local background (Zone VI) are determined as follows:

$$\text{Outdoors: } 38 \text{ nGy h}^{-1} \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} = 0.05 \text{ mSv}$$

$$\text{Indoors: } 50 \text{ nGy h}^{-1} \times 8760 \text{ h} \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} = 0.25 \text{ mSv}$$

The resulting total effective dose average value (0.30 mSv y<sup>-1</sup>) from terrestrial gamma radiation ranged from 0.25 to 0.44 mSv y<sup>-1</sup>, being within the 0.3-0.6 mSv y<sup>-1</sup> range reported by for different countries<sup>[3]</sup>.

The direct ionizing and photon component of cosmic radiation calculated at the local background was 67 nGy h<sup>-1</sup> (46.9 nSv h<sup>-1</sup>). If we consider the neutron component of cosmic radiation (14.1 nSv h<sup>-1</sup>) measured by Vega Carrillo<sup>[14]</sup> in Mexico at similar altitude and latitude (2420 m and 23° N, respectively) and applying an indoor shielding factor of 0.8 (assuming indoor occupancy to be 80% of time)<sup>[3]</sup>, the total annual effective dose from the cosmic radiation estimated at Zone VI was: 61 nSv h<sup>-1</sup> (1760 h+7000 h x 0.8) = 0.448 mSv y<sup>-1</sup>. The worldwide average annual effective dose from cosmic radiation source is 0.38 mSv y<sup>-1</sup><sup>[3]</sup>.

The total (outdoors+indoors) average annual external dose from terrestrial gamma exposure at Zone V (0.6 mSv y<sup>-1</sup>), is two times higher than that from the local background (0.30 mSv y<sup>-1</sup>). The additional effective external dose obtained for the general public is 0.3 mSv y<sup>-1</sup>. If the maximum values (135 nGy h<sup>-1</sup> at Zone V and 54 nGy h<sup>-1</sup> at Zone VI) are considered, the additional effective external dose would be 0.62 mSv y<sup>-1</sup>.

At SCRW (Zones I+II) the outdoor average external effective dose 1991-2002 from terrestrial gamma field estimated for workers is: 731 nGy h<sup>-1</sup> x 2000h x 0.7 Sv Gy<sup>-1</sup> = 1.02 mSv y<sup>-1</sup>, 20 times higher than for the local background (0.05 mSv y<sup>-1</sup>). If the maximum values (1991-1995) are considered, the additional effective dose would reach 2.5 mSv y<sup>-1</sup>.

The external gamma dose rate in air and the dose evaluation based on the regression equation by site-specific modeling is particularly useful for Zone V, since it is a private property closed to systematic monitoring. Restriction of land use, with special emphasis on the control of <sup>226</sup>Ra, or even remediation would be advisable. <sup>235</sup>U and <sup>238</sup>U alpha spectrometry or thermal ionization mass spectrometry measurements are necessary to determine the presence of depleted uranium (UD) at Zone V. <sup>232</sup>Th measurements by conventional neutron activation analysis is recommendable.

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