Natural Radioactivity in Spanish Soils

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Luis santiago quindos poncela
Universidad de Cantabria

Javier Soto
Pfizer, Spain

Carmen Ródenas
Universidad de Cantabria

José Gómez-Arozamena
Universidad de Cantabria

Paper

NATURAL RADIOACTIVITY IN SPANISH SOILS

L. S. Quindós, P. L. Fernández, J. Soto, C. Ródenas, and J. Gómez*

Abstract—**The program of studies and surveys of natural radiation and radioactivity in Spain organized by our research group at the end of the 1980s included a 4-y national survey to determine the concentrations of natural radionuclides in soil. Results obtained from measurements of soil samples collected nationwide at >900 sampling sites are reported and discussed in this paper. Correlations between absorbed dose rates in air calculated from natural radionuclide concentrations in soil and terrestrial gamma dose rates measured experimentally outdoors are shown for the different autonomous regions of Spain. Assessment is also made of the dose to the Spanish population from outdoor exposure to terrestrial gamma rays.**

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Key words: radioactivity, natural; soil; radiation, gamma; exposure, population

INTRODUCTION

STUDIES and surveys of natural environmental radiation and radioactivity are of great importance and interest in health physics not only for many practical reasons but also for more fundamental scientific reasons. The progressive development of the nuclear industry and other contaminating technologies and the widespread and ever-increasing use of radiation and radioactive isotopes make it absolutely necessary to evaluate the background of natural radiation and radioactivity in order to detect humanly manufactured contamination, assess its impact, and implement appropriate countermeasures to protect the population and the environment. At the same time, since natural radiation is the main source of human exposure, studies of the dose from this source and its effects on health improve the understanding of radiation damage and, therefore, are of great value as a reference when standards and regulatory control actions on radiation protection are established.

The interest in this kind of study has led worldwide to many national surveys on natural radiation and radioactivity, especially in advanced countries in the last decade. Thus, in Spain, the first program of surveys on natural radiation and radioactivity, the only ones to date covering the whole country, was organized and implemented by our research group in the 5 y from 1988–1992.

A great deal of work has already been done regarding different aspects of natural radiation and radioactivity towards achieving the main objectives of these national surveys. Several papers, either published or in press, analyze and discuss the main results both from the studies of the natural exposure of the Spanish population to terrestrial gamma radiation (Quindós et al. 1991c, 1992; Fernández et al. 1993) and to radon and radon progeny in dwellings (Quindós et al. 1991a,b,e) and from the determination of natural radioactivity concentrations in building materials and water (Quindós et al. 1988; Soto et al. 1988, 1991, 1992).

To complete the program, a national survey of natural radioactivity in soil was also carried out by our research team to set up a map of naturally occurring radionuclides in soil and, thereby, to correlate the concentration values of these natural radionuclides with the external exposure rates from terrestrial gamma rays measured experimentally outdoors throughout Spain. Given the low levels of environmental radioactive contamination due to ^{137}Cs and ^{134}Cs that the Chernobyl nuclear accident caused in Spain (Gil 1986; Ferrero et al. 1987), and since terrestrial gamma rays arise essentially from ^{40}K and the radionuclides in the ^{238}U and ^{232}Th radioactive decay series, which are widely dispersed in the earth's crust, the aim of this survey was focused on determining the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil by making an extensive series of measurements on soil samples collected nationwide. This paper will briefly analyze and discuss the main results available for natural radioactivity in soil in Spain.

MATERIALS AND METHODS

Fig. 1 shows a map of Spain with the geographical distribution of the 17 different autonomous regions into which the country is administratively divided, each in turn consisting of one or more provinces.

According to the prevalent soil types, Spain can be split into roughly four large geological areas. The first is basically composed of granitic and shale formations of Paleozoic origin, which occur with other bedded formations such as quartzites. This geological area pri-

* Department of Medical Physics, Faculty of Medicine, University of Cantabria, 39011 Santander, Spain.

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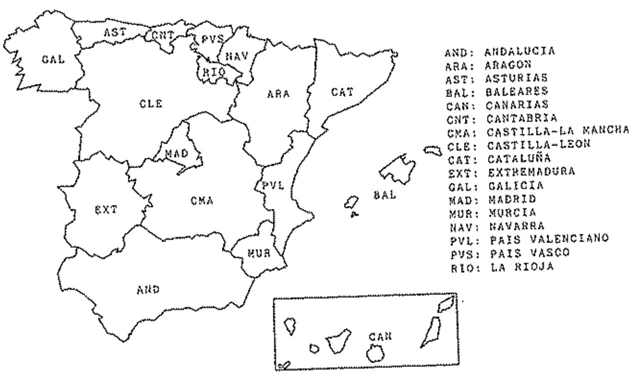

Fig. 1. Map of Spain showing the geographical distribution of its 17 different autonomous regions.

marily occupies the western part of the peninsula and extends southward to the Guadalquivir Valley; it includes the autonomous regions of Galicia, Extremadura, and Madrid, together with most of the provinces of Avila, Salamanca, Segovia, and Zamora situated in the region of Castilla-León, and the western part of the provinces of Ciudad Real and Toledo in the region of Castilla-La Mancha.

The second is mainly composed of sediments of marine origin, principally limestones, marls, and clays. This geological area essentially covers the whole of the Mediterranean coastal region, the northern part of the peninsula, and the southern part of the Aragón region.

The third area fundamentally consists of sedimentary rocks of more recent continental origin, mainly clays, sandstones, marls, and gypsum-bearing and calcareous formations. It basically covers most of the regions of Aragón, Castilla-León, Castilla-La Mancha, and La Rioja.

The fourth and final geological area is found in the Atlantic archipelago of the Canary Islands (autonomous region of Canarias). These islands are volcanic in origin and feature a large number of basaltic formations, primarily lava flows and pyroclasts, which form thick sequences.

As part of the research program, at the beginning of 1989 a national survey was designed to determine the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Spanish soils. This survey, carried out over the period 1989–1992, was performed in such a way as to obtain, as far as possible, a uniform and representative distribution of the sampling sites for soil collection throughout Spain. The selection procedure used in the survey yielded a total of 952 sampling sites chosen on bare soil areas. These sites were selected on the spot by the investigators and were open, reasonably level, and not obviously prone to flooding or other natural disturbances. Apart from these limitations, they were selected at random.

At each sampling site, five cores of topsoil were taken over an area of $\sim$1,000 m^2. Each core was taken to a depth of 5 cm at a randomly chosen point within the site area; the only constraint was that no core should be taken close to a field boundary, tree, building, or other obstruction. In order to obtain a representative sample, the soil cores collected at each site were thoroughly mixed together in the laboratory. A sample of 2 L was then weighed, air-dried for several days, and placed in an oven at 100°C for 24 h. The sample was subsequently reweighed to determine the water content, sieved to remove stones and pebbles, and crushed to pass through a 2-mm mesh sieve. Finally, the prepared sample was packed in a sealable 250-mL PVC[†] can and left for at least 4 wk before counting by gamma spectrometry in order to ensure that radioactive equilibrium was reached between ^{226}Ra, ^{222}Rn, and ^{222}Rn progeny.

Gamma spectrometry measurements of ^{226}Ra, ^{232}Th, and ^{40}K activities in soil were made using a high-purity germanium coaxial detector with an efficiency of 20%, a resolution of 1.86 keV, and surrounded with shielding material to reduce the background counting rate. The detector was calibrated using standard solutions of ^{226}Ra, ^{232}Th, and ^{40}K in the same geometry as the measured soil samples. Corrections for self-absorption in the samples were experimentally evaluated using gamma spectra of four pumice samples prepared with equal specific activities but with different densities ranging from 0.8–1.5 kg L^{-1}. To validate our results and to establish a quality control of our measurements, our laboratory coordinated and participated, with another 12 laboratories from different countries, in an international exercise of intercomparison of gamma spectrometry measurements of radioactivity in soils and building materials which was carried out in 1990 (Quindós et al. 1991d).

Considering the appropriate corrections for laboratory background, the activity of ^{226}Ra was evaluated, in all cases, from the 609-keV peak of ^{214}Bi, while the ^{232}Th activity was determined from the 911-keV peak of ^{228}Ac, and the ^{40}K activity was determined from the ^{40}K peak at 1.461 MeV. Detection limits for a counting time of 60,000 s were estimated to be $\sim$3 Bq kg^{-1} for ^{226}Ra, 2 Bq kg^{-1} for ^{232}Th, and 10 Bq kg^{-1} for ^{40}K. Random uncertainties in the measurements of these activities were typically 10%. Systematic uncertainties are estimated to have been of the same order.

Table 1 shows the number of soil samples measured in the 17 different autonomous regions of Spain, together with their respective surface areas and population density data from the 1991 national census.

Because sampling sites in the different provinces of the country were not exactly selected at the same sampling rate per unit area, it was necessary to calculate and assign appropriate sampling weights that counterbalanced the unequal selection probabilities. Accordingly, all data analyses were performed on a surface-weighted basis using properly weighted data from individual measurements of naturally occurring radio-

[†] PVC = polyvinyl chloride.

Table 1. Number of measurements, surface area, and population density data for the different autonomous regions of Spain.

Region	Number of measurements	Surface (km^2)	Population (millions)
AND	126	87,268	6.9405
ARA	65	47,650	1.1888
AST	17	10,565	1.0939
BAL[a]	11	5,014	0.7091
CAN[b]	30	2,036	0.7258
CNT	94	5,289	0.5273
CMA	119	79,230	1.6584
CLE	144	94,193	2.5459
CAT	45	31,930	6.0595
EXT	73	41,602	1.0618
GAL	71	29,434	2.7317
MAD	37	7,995	4.9475
MUR	16	11,317	1.0456
NAV	15	10,421	0.5193
PVL	32	23,305	3.8572
PVS	45	7,261	2.1040
RIO	12	5,034	0.2634

[a] Data referring only to the island of Mallorca.
[b] Data referring only to the island of Tenerife.

nuclides in soil made in the different provinces surveyed. This permitted the generation of unbiased statistical estimates [e.g., geometric mean (GM), arithmetic mean (AM)] (Shah 1984; White et al. 1992) both for the different autonomous regions of Spain and for the country as a whole.

RESULTS AND DISCUSSION

The results from individual measurements of the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil, along with the corresponding sampling weights, were combined into one data set and analyzed to derive the statistical estimates characterizing the distributions of natural radionuclide concentrations in soil for each autonomous region and the country as a whole.

Table 2 shows the estimates of the AM, GM, and geometric standard deviations (GSD), as well as the ranges, of the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil (expressed in Bq kg^{-1} dry weight) for the 17 autonomous regions of Spain and for the whole country. The AM concentrations range from 28 Bq kg^{-1} in Pais Valenciano to 94 Bq kg^{-1} in Galicia for ^{226}Ra, from 25 Bq kg^{-1} in Murcia to 68 Bq kg^{-1} again in Galicia for ^{232}Th, and from 367 Bq kg^{-1} in Baleares to 1,113 Bq kg^{-1} in Madrid for ^{40}K. With regard to the GM concentrations, the lowest values were found in Pais Valenciano for ^{226}Ra and ^{40}K, 26 and 327 Bq kg^{-1}, respectively, and in Murcia for ^{232}Th, 23 Bq kg^{-1}. The highest values of GM were found in Galicia for ^{226}Ra and ^{232}Th, 72 and 57 Bq kg^{-1}, respectively, and in Madrid for ^{40}K, 999 Bq kg^{-1}. Overall, the nationwide estimates of the respective AM and GM concentrations are 39 and 32 Bq kg^{-1} for ^{226}Ra, 41 and 35 Bq kg^{-1} for ^{232}Th, and 578 and 476 Bq kg^{-1} for ^{40}K.

The results summarized in Table 2 show that, in general, the averages and ranges of the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil in most autonomous regions and nationally are higher than the world figures reported by UNSCEAR (1988) (world average concentrations of 30, 25, and 370 Bq kg^{-1} are for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, and typical ranges are 15–70 Bq kg^{-1} for ^{226}Ra, 7–50 Bq kg^{-1} for ^{232}Th, and 100–700 Bq kg^{-1} for ^{40}K). Only the regions of Andalucia, Baleares, Murcia, Navarra, and Pais Valenciano show results comparable to the typical UNSCEAR values for the entire world, except for the range of the ^{40}K concentrations measured in the region of Andalucia, whose upper limit is more than double that given by UNSCEAR.

In relation to the geological structure of the country, it can be seen that the regions of lower average concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil match those areas (Andalucia, Baleares, Murcia, Navarra, and Pais Valenciano) that essentially consist of recent marine sediments (mainly limestones, marls, and clays) ranging in age from the Late Oligocene to the present. It can also be observed that, except for the autonomous region of Canarias, which is geologically volcanic in origin, the higher activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil generally correspond with those areas where granitic and shale formations are prevalent. As mentioned previously, these areas mainly belong to the regions of Galicia, Extremadura, and Madrid and to the provinces of Avila, Salamanca, Segovia, and Zamora in the region of Castilla-León. However, despite the high natural radionuclide concentrations measured in many of the soil samples from these four provinces, the AM and GM values of the ^{226}Ra, ^{232}Th, and ^{40}K concentrations estimated for the region of Castilla-León are comparable with the nationwide averages (Table 2). This is due to the low concentrations of these three radionuclides in soil for the other five provinces in this region (Burgos, León, Palencia, Soria, and Valladolid), whose contribution to the regional averages offsets the contribution of the high concentrations previously mentioned.

A more detailed analysis of the results from individual measurements shows that there is some degree of correlation between the concentrations of ^{226}Ra and ^{40}K in soil and between ^{232}Th and ^{40}K. Figs. 2 and 3 indicate that both correlations are weak, though significant because of the large number of measurements. Individual results for any one of the three radionuclide concentrations are not, therefore, good predictors of individual values for the other two. Although some scatter in the concentrations of ^{226}Ra and ^{232}Th vs. the ^{40}K content is noted, the activity concentrations of ^{226}Ra and ^{232}Th in soil appear to increase as the ^{40}K content increases. To achieve a better understanding of the relationships between the concentrations of these three natural radionuclides in soil shown in Figs. 2 and 3, our laboratory has now undertaken a more complete and specific study. Its main purpose is to analyze how the concentrations of ^{226}Ra, ^{232}Th, and ^{40}K vary with soil particle size for the most representative lithotypes

Table 2. Arithmetic means (AM), geometric means (GM), geometric standard deviations (GSD), and ranges of the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Spanish soils.

Region	AM (Bq kg^{-1})			GM (Bq kg^{-1})			GSD			Range (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
AND	29	30	426	26	26	347	1.58	1.83	2.08	8–82	5–79	46–1,660
ARA	36	35	485	33	32	438	1.47	1.51	1.70	48–72	16–77	141–1,120
AST	44	45	410	37	41	350	1.76	1.54	1.96	20–167	21–81	64–800
BAL[a]	35	31	367	33	28	343	1.39	1.73	1.50	23–70	10–58	153–600
CAN[b]	51	65	832	45	57	684	1.90	1.89	2.22	21–136	32–169	270–1,990
CNT	44	46	565	40	42	474	1.61	1.60	1.92	10–141	8–87	107–1,560
CMA	29	42	526	27	37	450	1.58	1.75	1.83	9–65	9–108	93–1,390
CLE	39	41	674	30	36	561	1.86	1.76	2.01	9–300	8–128	87–1,710
CAT	40	38	598	35	34	520	1.67	1.70	2.03	13–138	7–96	33–1,200
EXT	50	57	712	39	46	609	1.94	1.93	1.81	11–240	9–258	118–1,840
GAL	94	68	999	72	57	901	1.98	1.87	1.68	18–310	10–215	280–2,040
MAD	49	58	1,113	42	50	999	1.84	1.85	1.81	12–158	7–139	113–1,720
MUR	32	25	424	30	23	398	1.40	1.48	1.45	18–63	11–45	240–700
NAV	34	29	410	31	28	392	1.46	1.33	1.35	19–80	16–39	230–670
PVL	28	29	382	26	26	327	1.61	1.66	1.90	8–61	10–66	78–910
PVS	45	47	516	37	41	371	2.04	1.81	2.68	9–93	8–92	31–1,380
RIO	42	50	653	41	50	623	1.29	1.22	1.36	16–69	27–85	360–1,060
SPAIN	39	41	578	32	35	476	1.75	1.79	1.90	8–310	5–258	31–2,040

[a] Data referring only to the island of Mallorca.
[b] Data referring only to the island of Tenerife.

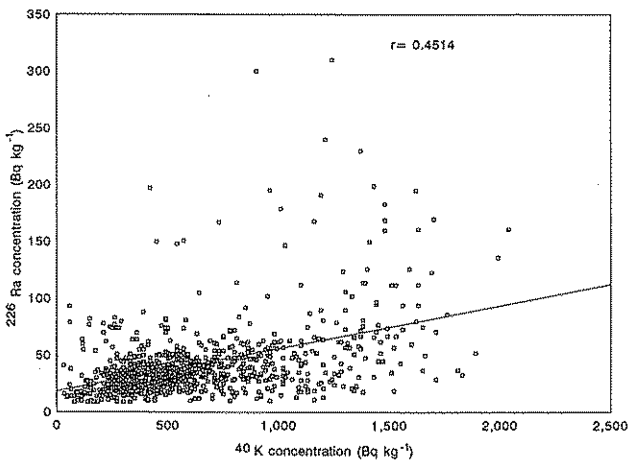

Fig. 2. ^{226}Ra vs. ^{40}K concentrations in Spanish soils.

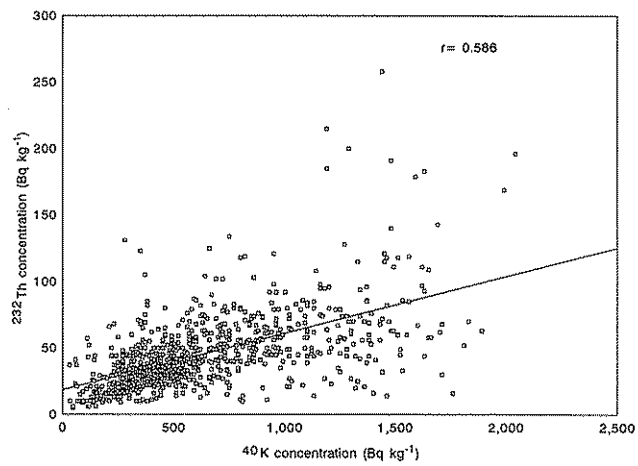

Fig. 3. ^{232}Th vs. ^{40}K concentrations in Spanish soils.

of soil in Spain and, subsequently, to investigate whether the scattering of the ^{226}Ra and ^{232}Th concentrations vs. ^{40}K content can be explained by differences in soil particle size, as suggested by other authors (Kvasnicka 1986; Megumi et al. 1986, 1988).

Though not included in Table 2, nationwide estimates of the median concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil were also derived. In all cases, the estimated median concentrations (31.6, 35.7, and 480 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively) were very similar to the corresponding nationwide GMs given in Table 2. This is an indication that the observed distributions of ^{226}Ra, ^{232}Th, and ^{40}K concentrations in Spanish soils can be approximated by a lognormal distribution.

Fig. 4 shows the observed cumulative frequency distributions for ^{226}Ra, ^{232}Th, and ^{40}K concentrations in soil in Spain, obtained with surface-weighted cumulative percentages from individual measurements made in the different provinces of the country. Solid lines represent the cumulative frequencies expected under the assumption of lognormal distribution. The parameters GM and GSD used to construct these lines are the respective nationwide GM and GSD estimates given in Table 2.

Fig. 4 shows that the observed distributions of

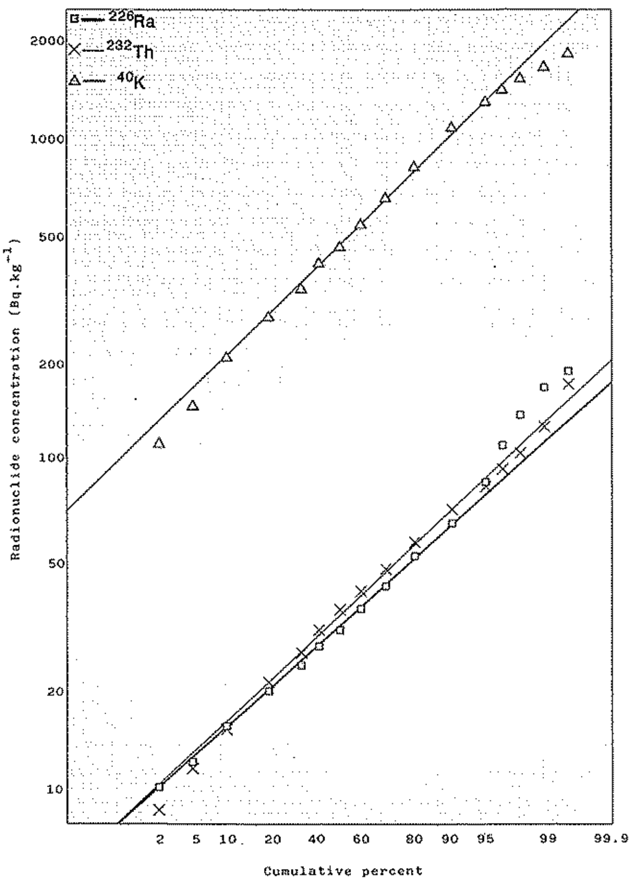

Fig. 4. Cumulative frequency distributions of ^{226}Ra, ^{232}Th, and ^{40}K concentrations in Spanish soils.

concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Spanish soils can be adequately approximated by a lognormal distribution. At a concentration of 100 Bq kg^{-1}, however, the lognormal distribution begins to underestimate the observed distribution of ^{226}Ra concentrations in soil, but the magnitude of the underestimate is quite small: 3.9% of the measured ^{226}Ra concentrations are greater than 100 Bq kg^{-1} as compared to 2.2% estimated from the lognormal distribution. At high concentrations (>1,500 Bq kg^{-1}), the lognormal distribution also seems to overestimate, by a very small margin, the observed ^{40}K distribution in soil: 2.8% > 1,500 Bq kg^{-1} for measured ^{40}K concentrations as compared to 3.9% from the lognormal distribution.

Finally, in order to correlate the natural radionuclide concentrations measured in Spanish soils with the experimental results obtained in a parallel nationwide survey of outdoor terrestrial gamma radiation (Quindós et al. 1992; Fernández et al. 1993), we calculated the outdoor dose rates due to terrestrial gamma rays at 1 m above the ground from the ^{226}Ra, ^{232}Th, and ^{40}K concentration values in soil, assuming that other radionuclides, such as ^{137}Cs, ^{90}Sr, and those present in the ^{235}U decay series, can be neglected as they contribute very little to the total dose from environmental background (Beck et al. 1972; Green et al. 1989). The conversion factors used to calculate the absorbed dose rates in air from natural radionuclides in soil were those given by Leung et al. (1990): 421, 587, and 43.1 pGy h^{-1} (Bq $kg^{-1})^{-1}$ for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. From among the different estimates reported in the literature, these values were finally chosen as they provided the best fitting between outdoor terrestrial gamma dose rates and radionuclide concentrations in soil experimentally measured in an earlier specific regional survey carried out by our research group in the autonomous region of Cantabria (Quindós et al. 1991c, 1992).

On the assumption that all decay products of ^{238}U and ^{232}Th are in radioactive equilibrium with their precursors, the conversion factors previously discussed in this paper are, in turn, derived from the dose-rate factors computed by Kocher and Sjoreen (1985) for monoenergetic photon emitters in soil of a uniform, semi-infinite source region using the point-kernel integration method and assuming a soil density of 1.4×10^3 kg m^{-3}. It should be kept in mind, however, that these dose-rate conversion factors refer to natural radionuclide activities per kilogram of *in situ* ground soil which, for uniformly distributed sources, depend on the soil moisture content (Beck 1972; Chen 1991). Accordingly, all the calculations of the outdoor terrestrial gamma dose rates from the measured ^{226}Ra, ^{232}Th, and ^{40}K concentrations in soil have been made using the appropriate density corrections from the water content in the original soil samples to convert the dry-weight activity concentrations found in the laboratory measurements to specific activities per kilogram of actual ground soil.

Weighting the mean dose rates calculated from natural radionuclide concentrations in soil for the different provinces according to their population, we estimated the population-weighted mean dose rates from outdoor terrestrial gamma radiation for the 17 autonomous regions of the country and for the whole of Spain. Fig. 5 shows the relationship between these calculated population-weighted mean dose rates and those derived from a total set of >1,000 measurements of outdoor exposure to terrestrial gamma rays made in 1990 and 1991 throughout Spain using a Mini-Instruments Environmental Monitor, Type 6–80, fitted with an MC-71‡ energy-compensated Geiger-Müller detector, calibrated and intercompared at the National Radiological Protection Board, UK. Good agreement can be seen between the calculated and measured population-weighted mean dose rates, with percentage differences <25% in all cases. These differences are reasonable since, besides the inherent uncertainties of the experimental measurements, many idealized assumptions were made to compute the absorbed dose rates in air from radionuclides in soil.

‡ Mini Instruments Ltd., 8 Station Industrial Estate, Burnham-on-Crouch, Essex, CM0 8RN, UK.

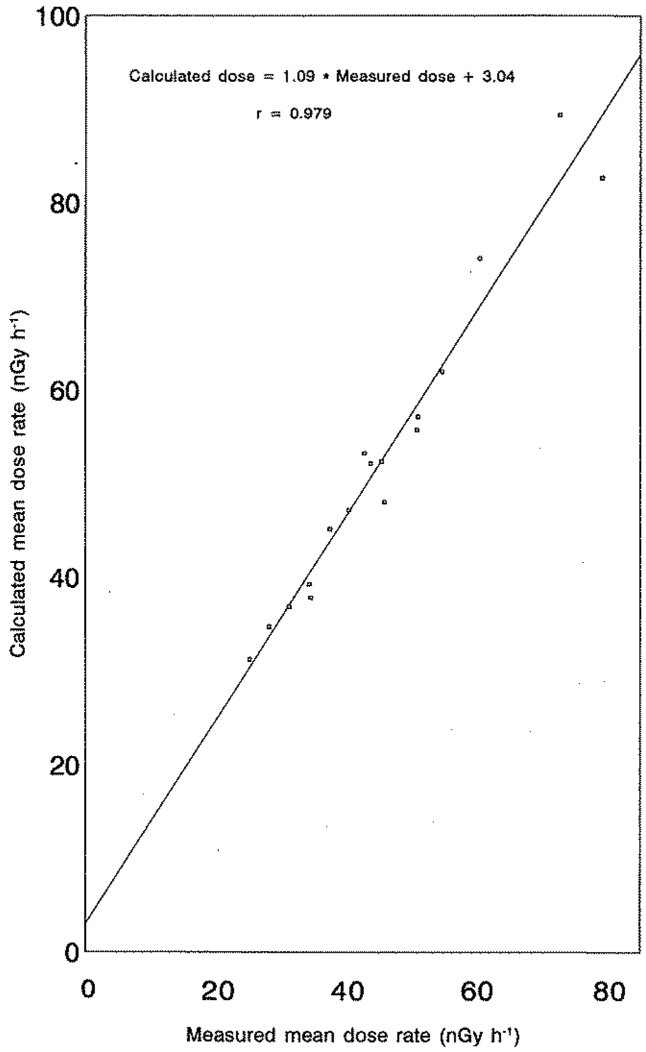

Fig. 5. Calculated vs. measured population-weighted mean outdoor dose rates from terrestrial gamma radiation in the different autonomous regions of Spain.

For the whole of Spain, an overall population-weighted mean outdoor terrestrial gamma dose rate of 53.3 nGy h^{-1} was calculated from the measurements of the ^{226}Ra, ^{232}Th, and ^{40}K concentrations in soil. This value is comparable with that of 47.1 nGy h^{-1} derived from absorbed dose rates in air measured experimentally outdoors throughout the country and is also similar to the world average of 55 nGy h^{-1} reported by UNSCEAR (1988). A degree of caution is necessary in using the previously given population-weighted mean dose rates estimated for Spain since they were derived from measurements related to essentially unaltered ground, whereas people spend some of their time outdoors in paved areas. Nevertheless, assuming that local masonry is made of local earth materials, the given values provide a good indication of the likely state of affairs.

Using the dose rate data obtained from the concentration values of natural radionuclides in soil, adopting the conversion factor of 0.7 Sv Gy^{-1} as recommended by UNSCEAR (1988), and considering that people in Spain on the average spend ~15% of their time outdoors, the mean annual dose equivalent to the Spanish population from outdoor terrestrial gamma radiation is 49 μSv, and the annual collective dose equivalent is ~1,900 person-Sv. These values are also comparable with those of 43.3 μSv and 1,683 person-Sv estimated from the results of measurements of outdoor gamma dose rates made throughout the country.

CONCLUSIONS

In the framework of a national survey conducted by our research group during the last 4 y, a map of naturally-occurring radionuclides in Spanish soils has been set up after completing an extensive series of measurements of specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples collected nationwide at a total of >900 sampling sites.

The results reported here for the national survey of natural radioactivity in soil in Spain, the only current data referring to the whole country, represent a basic data set for the assessment of dose to the Spanish population from outdoor exposure to terrestrial gamma rays. They also provide a valuable and useful reference for the design and development of specific regional surveys related to the measurement of indoor radon in those areas showing enhanced levels of natural radiation and radioactivity.

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