Natural gamma radiation in air *versus* soil nature in Portugal

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INTRODUTION

Since 1985 the portuguese population exposure to natural gamma radiation is the object of a on-going study, in order to assess the natural gamma radiation doses all over the country. For the primary surveys a G-M based portable dose rate meter was used, in order to have a general picture of natural gamma dose rates outdoors in different regions, mainly those corresponding to highly populated areas (1) The program was then extended all over the country and these data was further associated to data of an indoors survey on natural gamma dose rates, made by termoluminescent dosimetry, based on LiF square rods (TLD-100) in order to evaluate the effective dose to portuguese population due to natural gamma radiation (2).

A correction for the cosmic radiation component for different latitudes and altitudes was introduced after a cooperation for the Radiation Atlas – Natural Sources of Ionizing Radiation in Europe (3). So, the first Radiological Map for portuguese population exposure to *natural terrestrial gamma radiation* was established and the effective dose for every district (administrative division) was evaluated resulting 0.61mSv.y $^{-1}$ for the whole country. It should be noticed that for some regions indoors values were obtained by calculation upon the proposed (4) average relation 1.2 indoors /outdoors dose.

The analysis of outdoors survey results showed that northern regions had higher dose rate levels than southern ones and a quite different geological nature For instance, districts of Viana do Castelo, Braga, Porto, Viseu and Guarda, corresponding to a granitic area, present much higher values although distributed in a large interval, than those measured in Faro, Santarém and Setúbal, where soil nature is sedimentary, mainly aluvion sands and limestone, or in Beja, where soil originated in gabbro and diorite.

As this survey was complete, it was important to understand the relation of natural gamma radiation doses in air and soil nature and to see why similar dose levels might occur at different soil typpes, and it was decided to complement the study, mainly analising different types of soils among the predominant in Portugal.

We intend to caracterize qualitative and quantitatively soil nature in what concerns primordial radionuclides like ^{40}K and ^{238}U and ^{232}Th natural series. The importance of these nuclides in each type of soil is a consequence of soil formation from the different original rocks, and their concentration may be conditioned by mechanical, chemical and biochemical processes. Several points were selected from previous surveys for the present study, representing high, intermediate and low dose rate regions (5).

METHODS

In this study *in situ* gamma spectrometry was used to identify and quantify the contributions of primordial nuclides to dose rate in air in order to correlate natural terrestrial gamma radiation doses to the type of soils in Portugal. The system is based on a HpGe spectrometer Canberra Gx2020, multiattitude cryostat type 7935,7, Dewar vessel 7l ln2, know as “Canberra Big Mac”. The multychanel analyser whith 8192 channels, internal battery providing energy both for MCA and spectrometer, is a”EG&G ORTEC 7500B portable MCA.”

Each spectrum was analysed after a calibration factor $\mathbf{N_f/A}$, containing angular correction factor $\mathbf{N_f/N_0}$, for a particular energy and a given distribution; response conversion factor $\mathbf{N_0/\phi}$ for peack count rate per unit flux **(**cpm per γ cm^{-2}.s^{-1}) and geometry factor $\mathbf{\phi/A}$, **(**total uncollided flux (γ cm^{-2}.s^{-}) per unit concentration in soil). This calibration factor

$$Nf/A= \mathbf{N_0/\phi} \times \mathbf{N_f/N_0} \times \mathbf{\phi/A} \qquad \text{(equation 1)}$$

assessed in agreement with the procedure of Environmental Measurements Laboratory U.S. DOE (6) allows to establish a relation between absortion peak total counting rate for each radionuclide transition energy with its concentration in the soil.

⇨ Response factor $\mathbf{N_0/\phi}$ was obtained for each emission energy *E* by adjust equation:

$$\ln\left(\frac{N_0}{\phi}\right) = a - b\ln E \qquad \text{(equation 2)}$$

and *a* and *b* are calculated after relative efficency ε by equations *a'* and *b'*:

$$a' = 2,689 + 0,4996\ln\varepsilon + 0,0969(\ln\varepsilon)^2 \qquad \text{(equation 3.)}$$

$$b' = 1,315 - 0,02044\varepsilon + 0,00012\varepsilon^2 \qquad \text{(equation 4.)}$$

The response correction factor $\mathbf{N_0/\phi}$ for a particular nuclide energy is referred in Table 1., considering detector efficiency (ε =21,1%).

Table1.- Detector Response N_0/ϕ for the specified radionuclide energies

Nuclide	Energy (MeV)	N_0/ϕ=exp(5,113-0,937lnE) (cpm per $\gamma.s^{-1}$ cm^{-2})
^{212}Pb	0,238	638
^{214}Pb	0,352	442
^{208}Tl	0,583	275
^{214}Bi	0,609	264
^{212}Bi	0,727	224
^{228}Ac	0,911	181
^{40}K	1,460	116

⇨ Angular correction factor $\mathbf{N_f/N_0}$ for each emission energy was obtained from a table (6) relating length *v.s* diameter of E.M.L. detector types, by interpolation and for a soil facing detector window. The angular correction factor for a uniform source distribution in the soil is referred in Table 2.

Table 2.-Angular correction factor for uniform source distribution in the soil

Energy (MeV)	N_f/N_0 for L/D=1,102
0,3	1,082
0,5	1,062
0,7	1,051
1,0	1,031
1,5	1,021
2,0	1,001
2,5	0,991

⇨ Geometry factor $\mathbf{\phi/A}$ for each emmiter incident flux, for natural nuclide distribution was used (7) and considered ^{40}K and gamma emmiters from ^{238}U and ^{232}Th natural series (8). An interpolation was done on account of each nuclide energy and yield. The uncollided flux per unit concentration in soil for the studied radionuclides is referred in table 3.

Table 3.-Total uncollided flux per unit activity concentration in the soil 1m above ground

Nuclide	Energy (MeV)	Yield γ. s^{-1}/disintegration	Flux per unit concentration γ. $s^{-1}.cm^{-2}$/Bq.Kg^{-1}
^{212}Pb	0,238	0,436	1,74E-03
^{214}Pb	0,352	0,370	1,72E-03
^{208}Tl	0,583	0,860	1,73E-03
^{214}Bi	0,609	0,461	2,73E-03
^{212}Bi	0,727	0,066	4,23E-04
^{228}Ac	0,911	0,290	2,04E-03
^{40}K	1,460	0,107	9,76E-04

Calibration factor $\mathbf{N_f/A}$ for each of those radionuclides is then quoted on Table 4 and allows the conversion of peak count rate to each emmiter soil activity, after correction of Compton continuum and acquisition time.

Table 4. Calibration factors for analysed radionuclides.

Nuclide	Energy (MeV)	No/ϕ (cpm perγ.$s^{-1}cm^{-2}$)	Nf/No	ϕ/A (γ.$s^{-1}cm^{-2}$ por Bq.kg^{-1})	Nf/A (cpm por Bq.kg^{-1})
^{212}Pb	0,238	638	1,082	1,74E-03	1,204
^{214}Pb	0,352	442	1,082	1,72E-03	0,806
^{208}Tl	0,583	276	1,062	1,73E-03	0,505
^{214}Bi	0,609	265	1,051	2,73E-03	0,759
^{212}Bi	0,727	224	1,051	4,23E-04	0,100
^{228}Ac	0,911	181	1,031	2,04E-03	0,382
^{40}K	1,460	117	1,021	9,76E-04	0,116

Another factor is used to evaluate dose rate in air per unit activity concentration in the soil. This factor (dose rate/soil unit activity) was formerly calculated (7)) for ^{40}K and ^{238}U and ^{232}Th natural series. Admiting that each series is in equilibrium, it is enough to know the dose rate for one element, (for instance ^{214} Bi at ^{238}U series and ^{228}Ac at ^{232}Th series).

Table 5. -Dose rate per soil activity concentration unit

	Dose rate (nGy.h^{-1} per Bq.kg^{-1})
Uranium series	0,4500
Thorium series	0,6680
potassium	0,0424

In this work calculation was done for every identified element and the average value for each series was considered. A uniform profile of soil distribution is assumed. In Figure 1. a spectrum collected for this work, at Vila Nova de Paiva-Viseu is presented as an exemple.

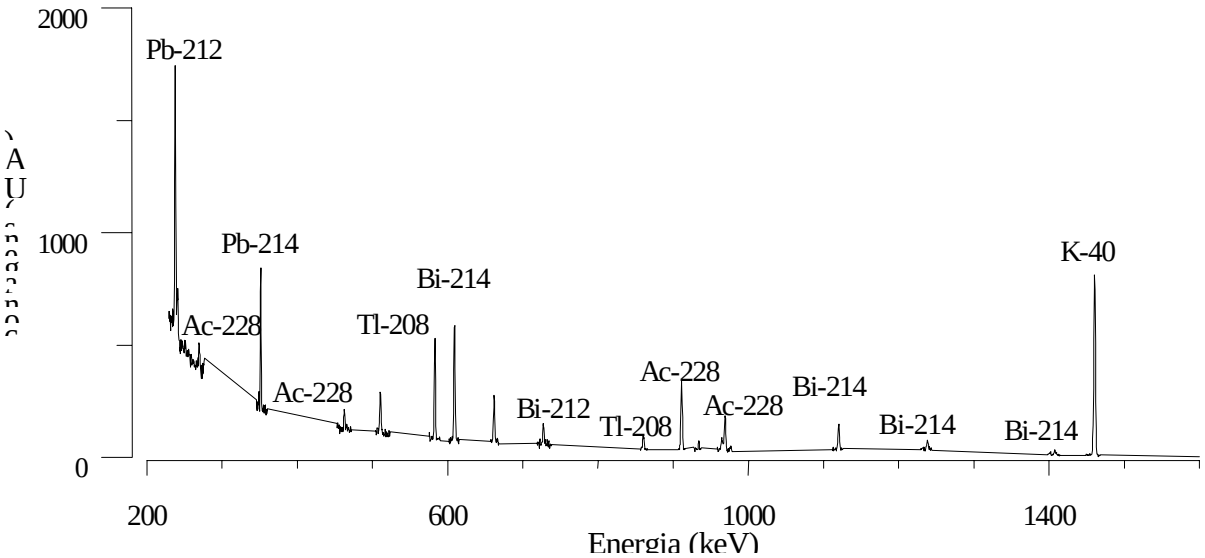

Figure 1.- *in situ* colected and laboratory analised spectrum(V. Nova de Paiva)

On Table 6. the dose assessement for ^{40}K, ^{238}U series and ^{232}Th series at the same location is detailed. Calibration factors from Table 4 were applied to peak areas corresponding to each nuclide caractheristic emission and averaged dose rates were obtained after Table 5.

Table 6.- Dose assessement after *in situ* spectrum analysis (V. Nova de Paiva)

	Dose rate factor ($nGy.h^{-1}$ per $Bq.kg^{-1}$)	Nuclide	peak area (cpm)	calibration f. (cpm per $Bq.kg^{-1}$)		dose ($nGy.h^{-1}$)	
Thorium series	0,6680	^{212}Pb-238keV	163	1,204	91		
		^{208}Tl-583keV	57	0,505	75		
		^{212}Bi-727keV	13	0,100	90		
		^{228}Ac-911keV	43	0,382	75	**média**	**83**
Uranium series	0,4500	^{214}Pb-352keV	59	0,806	33		
		^{214}Bi-609keV	64	0,759	38	**média**	**35**
potassium	0,0424	^{40}K-1460keV	143	0,116	52	**média**	**52**
					Total dose		**170**

RESULTS

The portuguese radiological map presented in Figure.2 organized by “councils”, the smaller administrative divisions in the country, shows in the northern regions higher dose rates than those registered in the south and the geological charts confirm different underlying geological formations (8).

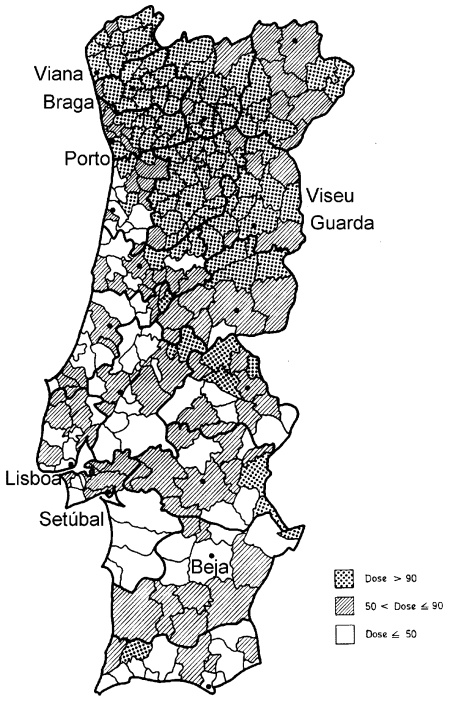

Figure 2.- Natural terrestrial gamma radiation –mean doses by council (nGy.h^{-1})

To establish the ***soil nature-natural gamma dose rate in air relation*** spectra were collected in several locations, representing the predominant types of soils in Portugal,of high, intermediate and low dose levels.

After the analysis of the data obtained by *in situ* gamma spectrometry for soils of **intrusive** origin, a predominant contribution of the thorium series elements was found, mainly at districts of Braga, Porto and Viana. Other locations at Guarda and Viseu show a similar contribution of both series and potassium, but some places where mining was done for years have a higher contribution of uranium. The Geological Charts (9) allowed the caractherization of these soils, most of them silica oversaturated mostly *granite-monzonite*, *granodiorite*, and *tonalite*. Another group of saturated soils, a *diorite-gabbro* complex at Beja, shows low dose levels and almost equivalent contributions.

For soils of **sedimentary** origin, a predominant contribution of potassium was found, followed by the thorium series. Both chemical nature rocks like *limestones*, *dolostones*, or terrigenous sediments like *conglomerates* and *sandstones (biocalcoarenites*), *arkoses* and *fossiliferous sands* were considered, although the chemical and mechanical weathering of original rocks difficults it’s association.

In the group of soils with **metamorphic** origin, both types of metamorphism are included. Some contact metamorphic rocks like *hornfels* and *quartzites*, *metagrauwakes* and *metadolostones*, or regional metamorphic rocks like *andalusite-schist*, *greenschist, garnet-schist* and *granit-gneiss*, are in the origin of the selected soils. As metamorphic rocks derive from preexisting ones the relative contibutions to dose rate differ: for northern councils the thorium series elements represent the higher contribution to dose rate while in most southern points potassium is the main contributor.(10)

On Table 7 average relative values and respective standard deviation are referred for each soil type.

Table 7.-Relative contributions to dose (%)

soil origin	thorium series	uranium series	potassium
intrusive	45%±7%	25%±6%	30%±5%
sedimentayr	32%±5%	22%±4%	46%±5%
metamorphic	42%±8%	22%±9%	36%±13%

Soils of intrusive and metamorphic origin present a major contribution to dose by thorium series elements, more relevant than those due to uranium series and potassium, which often have similar relative wheight in intrusive soils. For sedimentary origin soils ^{40}K potassium contribution is the more important followed by thorium and uranium series.

The ranges of the measured data and the contributions to the total dose rate for each type of soil are resumed on Table 8.. For intrusive soils a large spread for the values of the three contributions should be noticed.

Table 8. -Contributions for dose per soil type (nGy.h^{-1})

	thorium	uranium	potassium	total
intrusive origin				
average	67	37	43	147
s.deviation	29	14	14	50
range	4-132	4-64	3-69	11-241
sedimentary origin				
average	12	8	17	38
s.deviation	5	2	6	11
range	5-19	6-11	10-25	21-52
metamorphic origin				
average	46	25	31	102
s.deviation	36	20	11	64
range	12-102	4-56	14-43	30-196

DISCUSSION

After this survey by *in situ* spectrometry global doses were assessed by adding the three contributions at each studyied point.

- For soils of intrusive origin, assessed doses are dispersed by a large interval which is the result of different chemical composition of intrusive rocks, silica oversaturated like *granite, granodiorite, tonalite,*or saturated like *diorite* and *gabbro*. The reason for these differences is that magma cristallization begins by high melting point ferromagnesian silicates and calcic plagioclases, poor in silica and so undersaturated rocks are formed. The residual magma gets silica enriched and then minerals like monazite, zircon are formed, containing the natural radioactive elements.

❖ For soils of sedimentary origine a short dose range for every contribution is shown and the average dose rate is the lowest among the three studied soil types. The mechanical processes normally keep major constituents and those of minor size and concentration may represent a smaller importance in the sedimentary rock. So, at Campo Maior, near Beja, thorium and uranium series both, represent the same as potassium contribution. Chemical processes may cause mineral dissociation and then adsorption of acessory minerals containing uranium and thorium by clay minerals, on *sandstones* and *shales*, as the soil constitution at Vendas Novas.

➢ For soils of metamorphic origin the doses have a range similar to the intrusive soils. Most of studied places are in the north of the country and contact metamorphism was a consequence of *granite* instalation, producing *hornfels.* The *andalusite-schists* and *hornfels* in other regions result from actions by *granodiorites* over *schists*. In the southern regions doses are lower because *gneiss*es metamophised *limestones* and *dolostones*.

CONCLUSIONS

This paper intends to find a relation between terrestrial gamma radiation doses and the nature of the rock that should be on the soil origin, by the study of dose contributions of each natural series, ^{238}U and ^{232}Th and ^{40}potassium, that occur with different concentrations on rock composition.

- For soils of intrusive origine the highest dose rate is 241 nGy.h^{-1} and was registered at Guimarães, near Braga, in the northern region and soil nature is a *-monzonitic granite* were the relative wheight for the dose is 55% for thorium series, 17% uranium series and 28% for potassium.
- the lowest dose rate registered is 11 nGy.h^{-1} at Beja, were soils are based on a *diorite-gabbro complex* and the relative wheight is 36% for thorium series, 36% for uranium series and 28% for potassium.

❖ For soils of sedimentary origine the highest dose rate is 52 nGy.h^{-1} and was registered at Vendas Novas, near Beja, in the south, were soil is based on *alluvium sands* and *sandstones.*The relative wheight for the dose is 30% for thorium series, 21% uranium series and 49% for potassium.

❖ the lowest dose rate registered is 21nGy.h^{-1} at Campo Maior, were soils are based on potassic feldspar rich *arkoses* and the relative wheight is 24% for thorium series, 28% for uranium series and 48% for potassium.

➢ For soils of metamorphic origin the highest dose rate is 196 nGy.h^{-1} and was registered at Barcelos, near

Braga, in the northern region where soil is based on *hornfels* and *andalusite-schist* and where the relative wheight for the dose is 52% for thorium series, 29% uranium series and 19% for potassium

- the lowest dose rate registered is 30nGy.h^{-1} at Portel, in the southern region where were soils are based on *metadolostone* and the relative wheight is 39% for thorium series, 14% for uranium series and 47% for potassium.

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