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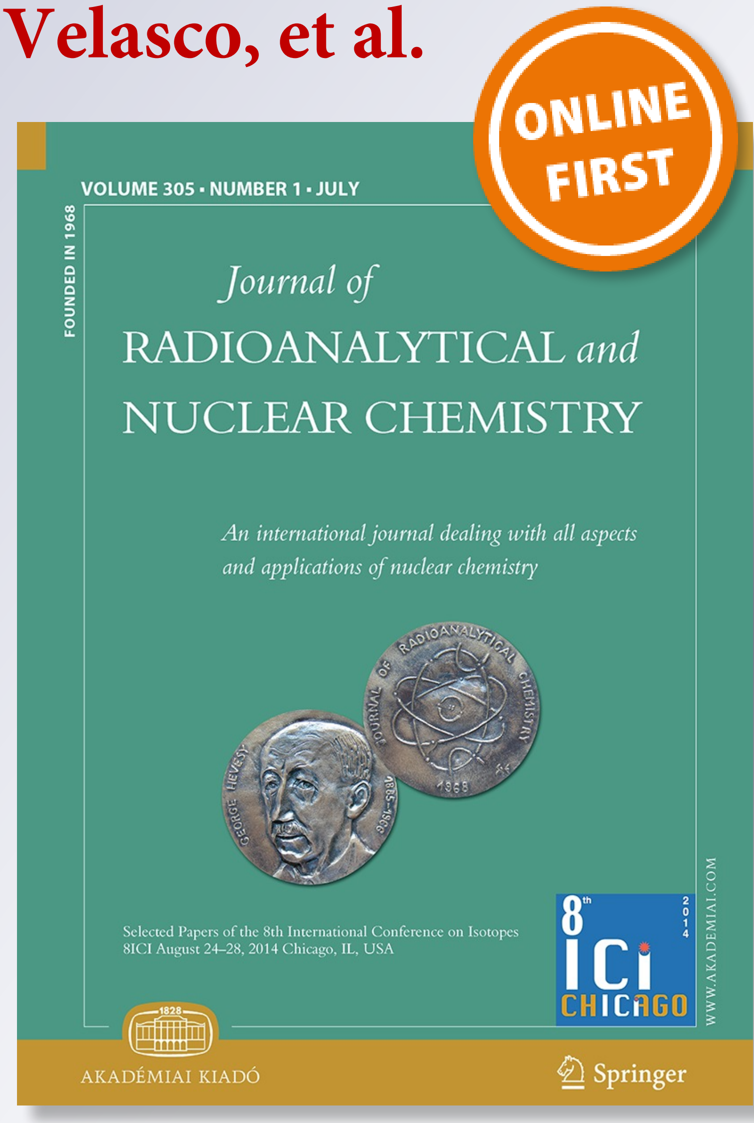

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Radon concentration in air and external gamma dose rate: is there a correlation?

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Abstract We checked the existence of correlations between experimentally determined radon concentration in indoor air and gamma dose rate, in different environments: residences, workplaces in subway stations and radiotherapies, and a gold mine. Except for the mine environment, where a linear correlation ($r^2 = 0.86$) was obtained with statistical significance, we found no correlations between those quantities. Both radiation sources are originated from natural radionuclides, nonetheless the observation of correlations depends on various conditions, as we discuss here.

Keywords Radon · Radon transport process · Gamma dose rates

Introduction

It is known that the largest contributions of natural radiation to the effective dose received by the world population come from the inhalation of radon gas and from the external irradiation of the body with penetrating gamma rays [1]. The radon gas in the indoor air is mostly Rn-222, radionuclide from the U-238 series. Due to the presence of this radioisotope in indoor air, there is an internal body irradiation (chiefly in the respiratory track) with the alpha particles emitted by its short-lived progeny. Radon gas emanates from the soil and from building materials, and due to its half-live (3.8 days), the atoms can migrate from relatively long distances till reach the inside air volume. Although the dynamics of the radon transport from the source (disintegration of Ra-226 nuclides) to the air is complex, depending on porosity of the material, humidity, temperature, presence of fissures in soil or walls [2–4], it is possible to assume, in some circumstances, a simple transport through the air in the porous of the material.

On the other hand, the natural gamma rays, which irradiate the human body, consist of a mixture of photons of various energies emitted by radionuclides of the natural series (mainly U-238 and Th-232 series) and K-40. The original spectrum of photon energies is degraded by multiple scattering of the primary photons in the soil and other materials, giving rise to a broad band of energies (from some keV to 2.6 MeV) in the geometry where people are usually irradiated. As the mean free path of energetic photons in air is usually large, the photons, which irradiate the human body, can come from large distances. In cases where the concentration of natural radionuclides in soil is uniform, and there is a large plane area (infinite slab of soil) the gamma-dose rate at 1 m from the soil is constant and determined by this radionuclide concentration, and it can be roughly estimated as half the dose rate at a position $\sim$70 cm inside the soil [5]. As well as the soil, large masonry structures can give significant contributions to the gamma-ray dose rate [5]. In the tunnels of a mine, the geometry is completely different, resembling a 4π solid-

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angle irradiation, and the dose rate in a tunnel is similar to the one inside the soil.

Although the dynamics of radon and photon dose deposition have been very different and complex, there are various attempts to correlate the doses delivered by both agents. As monitoring of gamma dose rates in air is relatively easier than monitoring radon concentrations, finding such a correlation could be very important. For instance, for addressing the best approach to determine the natural dose component when a high-radioactive area is in study, or to optimize the efforts for the determination of dose risks to a population, and even to mitigate or to remediate high radon gas concentration indoor in radon prone areas, using ambient dose rate as indicator would simplify surveys. Surveys of radon concentration in homes and at workplaces for estimation of population doses is a logistically complicated and costly effort. This is mainly true for residential surveys where also social and psychological factors related to the measurements have to be considered.

Fujimoto found good linear correlations between log of radon concentration and gamma dose rate after analyzing the data of a large survey of houses in Japan according to the type of structure of the measured houses (wooden, concrete of prefabricate houses) [6]. On the other hand, Chougaonkar and coworkers did not find a good correlation between indoor gamma and inhalation doses in a survey of 197 houses at the high-radioactive area of Kerala, India [7]. In fact, these authors found a poor and negative correlation between the quantities, and attributed the results to the type of soil (sand) in the region. Furthermore, in two areas of Taiwan, near nuclear installations, a set of data shows a weak positive relationship between the environmental gamma dose rate and the concentrations of radon and progeny [8].

García-Talavera and coworkers used national data from the whole Spanish mainland to correlate the indoor radon concentration with the external gamma dose rate and the geological characteristics of the soil. Regarding gamma dose rate and radon concentration, a good association was found when the data were regarded as categorical: for instance, the group of houses with higher radon concentration are built in areas where the gamma dose rates are higher. They concluded that the γ-dose rate has been proved to discriminate non-radon-prone municipalities excellently, but this index is not enough to classify reliably a radon-prone area [9].

In São Paulo, we determined total alpha emission concentration (radon and progeny) in air and gamma dose in 59 houses distributed throughout the city, with the objective of knowing better the irradiation conditions of the population. In addition, in an attempt to get data in places with poor ventilation conditions, we extended the work to a set of workplaces in the São Paulo subway and in a group of 16 offices located in eight radiotherapy centers. In this work, we present these data gathered in houses, subway stations and radiotherapy centers, and study the relationship between indoor radon concentrations and indoor gamma dose rates. The comparison is extended for results we have obtained recently, of radon concentration and gamma dose, in an Argentinian gold mine, where the ventilation conditions are minimal [10].

Experimental

Radon-concentration determination

The data analyzed here were obtained by our laboratory in the period from 2002 to 2010, with different objectives, and, according to the objectives, the quantity evaluated was the *total alpha-emitter concentration in air* (radon and progeny), or *radon concentration in air*. For the survey in dwellings, as the knowledge of radon and progeny is very important from the radiation protection point of view, the quantity evaluated was the total alpha-emitter concentration. For workplaces with low ventilation rates (as subway stations and underground radiotherapy centers), the goal was to verify if the working conditions were below the reference levels proposed by ICRP [11]. The same approach was used to investigate the potential occupational exposure of tourist guides who work in old mines in Argentina. The reference levels for radon exposure in working places are given by ICRP in radon concentrations.

The total alpha emission concentration in air in the 59 houses at São Paulo was measured using bare LR-115 detectors calibrated as described previously [12]. The detectors, mounted on PMMA plates were accommodated in one of the rooms (bedroom or living room) of each house. The calibration factor used to convert the track density to concentration was obtained from comparison to CR-39 detectors absolutely calibrated, and exposed simultaneously in a pilot survey in dwellings with various ventilation conditions. The uncertainty in alpha emitter concentration was calculated through a composition of the calibration factor uncertainty (7 %) and the track density uncertainty.

The radon concentrations in the subway stations and underground radiotherapy centers were determined with the use of a parallel plate monitor type: a pair of square PMMA plates (14.2 cm side), separated by a 4.0 mm air space, wrapped with PVC film to avoid the entrance of radon progeny [13, 14]. In the internal part of each plate a LR-115 type II plastic track detector, covered with 22 or 25 µm aluminum foil was fixed. The parallel plate monitor, with the plastic detectors inside, was installed in each of the measuring places at 1.5 m from the soil. These radon chambers were calibrated in low ventilation conditions, similar to the ones of the places where they were used. This kind of radon chamber has proved to give reliable results for these environmental conditions. Additionally the chambers are easy to use, giving rise to good counting statistics.

The radon concentrations in the air inside the Argentinian mine were determined with the NRPB/SSI type monitor, where a CR-39 plastic track detector of 1.7 cm^2 area and 0.9 mm thick was installed [15]. The chambers were placed in 14 points of the gold mine tunnels, at 1 m from the soil [10]. The standard relation between track density and radon concentration was used [15]. This value was previously checked using one Rn-222 source inside a 583 L chamber. The radon concentration inside the chamber was checked with a Lucas cell detector (model 110A, Pylon Electronics Inc) calibrated by the Institute for Energy and Nuclear Research of the Brazilian Nuclear Energy Commission (IPEN-CNEN) and the results obtained with the CR-39 detectors agreed with the literature.

Both types of plastic track detectors were etched (LR-115 with a 2,5 N NaOH solution, at 60 °C during 110 min, and the CR-39 detectors 30 % KOH solution, at 80 °C during 330 min) and examined with a semi-automated optical microscopy analysis system described in a previous work [16]. The evaluated track density, taking into account the exposure period, was related to the mean radon activity concentration or total alpha emitter concentration (measured in Bq kg^{-1}) through the corresponding calibration factor, as described above. The uncertainties of the concentration activities are derived from the counting statistical error.

Annual dose-rate determination

Gamma doses were measured with thermoluminescent dosimeters, accommodated in monitors, which were kept at the monitoring places, joined to the radon monitors, during long periods (3 months for the mine, 5 months for the subway and radiotherapy centers and 6 months for the houses). The readout of thermoluminescent dosimeters was performed at the Institute of Physics of the University of São Paulo, using homemade equipment. The results were converted to air doses using a group of similar detectors irradiated with known doses of gamma rays (Co-60 calibrated source). The final doses were normalized to the mean annual dose rate, assuming that it is constant during the whole year.

Monitoring places

The houses included in this study were selected according to a draw, using the Brazilian census classification, throughout the city. A drawing result was accepted only if it corresponded to residential houses, not edifices, otherwise it was repeated. The residents of the house were invited to participate, and in case of approval, they received instructions and explanations, the dwelling was registered, and a place in the house was chosen to receive the radon and the gamma monitors.

The subway stations chosen for the exposure of the monitors are located in various regions of the city of São Paulo, and the workplaces where the monitors were positioned are located in depths of 4.5–22 m from the street entrance of the station. These offices are workplaces situated away from the public circulation in the stations, and also far from the train routes.

The radiotherapy centers are also distributed in various regions of the city, and the chosen workplaces—usually the Physics calculation rooms or meeting rooms—were located far from the irradiation facilities.

La Carolina mine is one of the oldest gold mines of southern South America. It is open for touristic visits nowadays. It is located in the La Carolina District of San Luis Province, in west-central Argentina (32°48′S, 66°6′W). This mine is poorly ventilated, with no artificial resource for moving the inside air. 14 points along the tunnels of this mine were chosen for the measurements. [10].

Results and discussion

Exploratory statistics

In Fig. 1 we show the histograms of external gamma dose rates and total alpha concentrations for the 59 houses in São Paulo. The distribution of gamma dose rates is much more symmetric than that of the radon and progeny concentrations, and the last one resembles a log normal distribution. However, Chi square tests do not confirm the hypotheses of normality and lognormality respectively. Perhaps the small sample size contributed to this result.

Table 1 summarizes the statistical analysis for all the data obtained. We notice that the data taken at the metro stations have the smallest relative standard deviations (smallest *CV*—coefficients of variation) of all the samples, for both contributions to irradiation—radon and gamma. Probably the standardized building structures and the large amount of concrete in the metro stations account for the small data variation, as this makes the results more independent of the variation in the soil radioactive contents at each station. On the other hand, the sample of houses presents the largest *CV* (77 %) for the total alpha concentrations, as expected, as the type of buildings, the maintenance and the amount of ventilation vary largely from

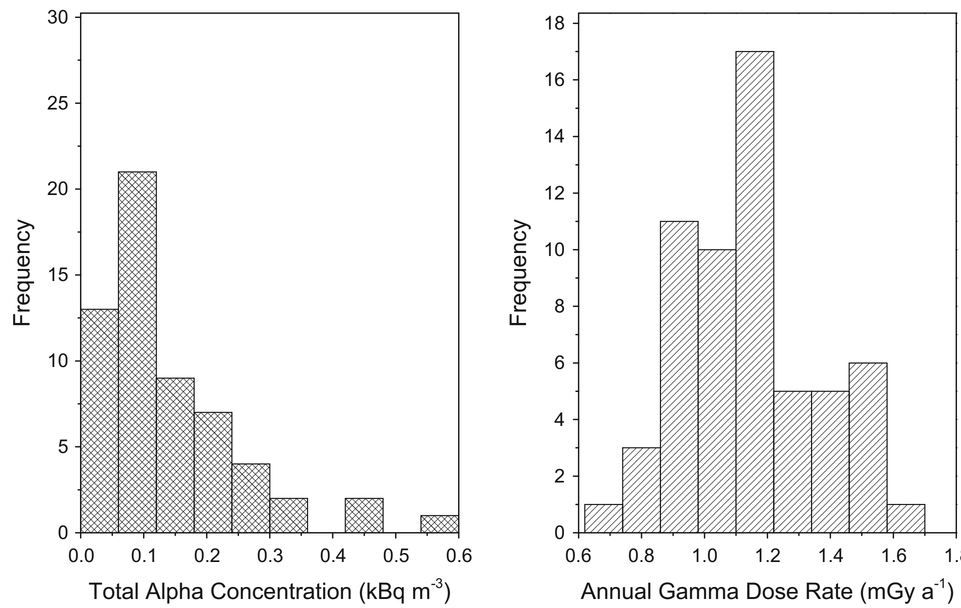

Fig. 1 Distribution of experimental values of total alpha concentrations and of annual gamma dose rates in 59 houses at São Paulo

Table 1 Statistics of the data obtained in this study for radon or total alpha concentrations in air and annual gamma dose rate in various samples

Sample (*N*)	Radon or total alpha concentration (kBq m^{-3})		Annual gamma dose rate (mGy a^{-1})	
	Mean/median	σ (*CV*)	Mean/median	σ (*CV*)
Houses (59)	0.142/0.108	0.11 (77 %)	1.14/1.15	0.22 (19 %)
Radiotherapies (16)	0.658/0.635	0.082 (12 %)	1.97/2.04	0.34 (18 %)
Subway stations (15)	0.805/0.810	0.039 (5 %)	1.89/1.90	0.150 (8 %)
La Carolina mine (14)	4.84/5.05	1.06 (22 %)	3.05/3.15	0.59 (20 %)

N is the sample size, *CV* is the coefficient of variation, σ is the standard deviation

house to house; moreover, there is no standardization on the building materials (walls, ceilings or floor). The *CV*'s for the results of the annual gamma doses, except for the metro stations, are very similar among the samples: they are circa 20 %, showing that this quantity is less affected by external variables such as ventilation rates. It is interesting that we found similar *CV*'s (from 16 to 22 %) in another survey of gamma doses for other types of locations in the city of São Paulo, using other methodology [17].

Regarding the mean values shown in Table 1, for indoor alpha emitter concentration and for annual external gamma dose, the ones obtained in the houses are much smaller than those observed at the other locations in São Paulo, as the ventilation conditions in the chosen workplaces disfavor radon transport to the exterior environment. The values obtained at La Carolina are the highest detected, the radon concentrations can be considered problematic for workers, mainly guides. This is expected in badly ventilated mines, as the tunnels are surrounded by a large quantity of soil, giving rise to a large solid angle for gamma irradiation and low air exchange and thus high Rn accumulation. In the mines, the walls have no finishing surfaces to impede radon diffusion to the inside atmosphere.

Another characteristic is the comparison of mean and median values: for the gamma dose distributions, the differences are very small, always below 4 %; whereas concerning the alpha emitter concentration in houses, there is a large discrepancy (31 %) between mean and median values. This was expected from the distribution seen in Fig. 2 and was predicted and verified by Fujimoto in a survey of Japanese houses [6]. From the radiation protection point of view, the values for the workplaces in São Paulo reported here are high if the occupation period is long. More experiments, carried out in different seasons are necessary in order to confirm the outcomes.

Correlation Rn—dose rate

The correlation of radon or alpha emitter concentrations in air and annual gamma doses, or its lack, is visualized in the

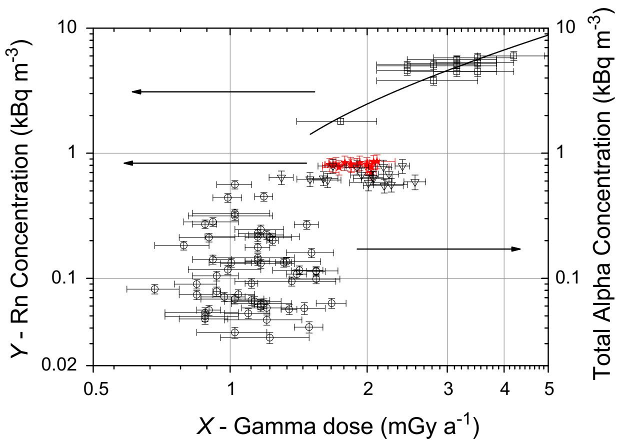

Fig. 2 Scatter plot, in log scales, of Rn (*Y*) or total alpha concentrations (kBq m^{-3}) and annual gamma dose (*X*, mGy a^{-1}) for all the samples studied here: *open circles*: houses; *open triangles*: radiotherapy workplaces; *full stars*: subway-station workplaces; *open squares*: La Carolina mine. The *line* corresponds to the linear fit ($r^2 = 0.86$) to the La Carolina data: $Y = 2.1 \times X - 1.7$

scatter plot of the quantities in Fig. 2. For the data of La Carolina mine, a linear regression line between radon concentration and annual gamma dose is also shown. It is not linear in the log scale because the intercept is not zero. A linear fitting between log of radon concentration and annual gamma dose, as the ones obtained by Fujimoto [6], was also tried to the La Carolina data, but the correlation factor ($r^2 = 0.66$) is much smaller than the one obtained with the linear fitting presented here (0.86). Additionally, to confirm the correlation, the Spearman rank test was performed. A Spearman rank regression coefficient (ρ) of 0.940 was obtained corroborating the positive correlation between radon concentration and gamma dose rate in La Carolina mine.

It is also evident from Fig. 2 that no other data correlation could be found for the other data sets: the radon concentration variations found in the radiotherapy rooms, as well as in the metro stations, are too small to recognize correlations if they exist. On the other hand, the total alpha concentrations obtained inside the houses vary so much that no relation to the gamma doses can be found. The analysis of the whole group of working places results (mine, subway and radiotherapy) resulted in a bad correlation ($r^2 = 0.3$ for either linear or log-linear fittings). Additionally, the Spearman rank regression test applied to those data resulted in a poor rank coefficient ($\rho = 0.59$), indicating absence of correlation for this group.

The fact that we found a correlation between radon concentration and annual gamma dose in one case and not in the others deserves some discussion. In a recent study, it was possible to verify that the La Carolina mine main tunnel works as a blind end system communicating with the external air only through one entrance. This occurs in summer (period when the measurements presented here were done) mainly because the temperature difference between the inside and outside temperatures is small ($\Delta T = 1$ °C) [18]. These conditions are rarely fulfilled in human living environments, where there is more ventilation, and the dynamics of change of radon concentration is more challenging.

Another notable detail is that in some circumstances the variability of indoor radon concentrations, expressed for example by the *CV*, is unusually small. The metro workplaces are such an example, which we explain with the huge quantity of building materials and the similarity of the construction in the seven stations that were monitored. Possibly this fact contributed to mask the possible role played by the natural radionuclides distribution in the soil to the radon equilibrium concentration.

Conclusions

Analyzing experimental values of indoor radon or alpha emitter concentrations and of annual gamma doses in samples of houses and low-ventilation workplaces in São Paulo, we found no correlation between these quantities, which contribute largely to the effective dose to the population. The variations obtained in the total alpha emitter concentration values measured in the houses were much higher than the ones in the workplaces. This fact is due to higher variation in the ventilation conditions in the houses, but also due to the type of construction of the workplaces in metro stations and radiotherapies. In contrast to these results, a nice correlation between the annual gamma dose values and radon concentrations inside the La Carolina mine was established. We attribute this correlation to the fact that the soil portion that contributes to the gamma irradiation is the same from which the radon atoms diffuse to the air inside the mine, and to the low ventilation conditions in this mine in summer.

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