Paper

INFLUENCE OF SOIL AND BUILDINGS ON OUTDOOR GAMMA DOSE RATES IN SÃO PAULO, BRAZIL

F. H. M. Medeiros and E. M. Yoshimura*

Fn1 *Abstract*—**This work analyzes the influence of the most abundant natural gamma emitters in soil (^{226}Ra, ^{232}Th, and ^{40}K) on the total outdoor gamma dose rate in the city of Sao Paulo, Brazil. A new method is introduced to determine gamma dose rates due to soil 1 m above the ground through measurements performed deep in the soil. This allows evaluation of the soil component even in places where the measurement at 1 m height is influenced by other sources (mainly the presence of buildings). The methodology was tested in non-urbanized areas by comparing direct dose rate measurements in air with those deep in soil. In addition, high-resolution gamma ray spectrometry of soil samples collected throughout the city was used to determine the natural radionuclide concentrations, allowing the comparison with the in-situ dose rate results. Measurements deep in soil followed a log-normal distribution. The fitted geometric mean (median) and geometric standard deviation of the soil contribution to the ambient dose equivalent rate at 1 m height were, respectively, 80.9(6) and 0.642(4) nSv h^{-1}. Compared to previous data, these values show that buildings enhance about 35% the outdoor gamma dose rate expected only from soil. The specific activities of ^{226}Ra, ^{232}Th, and ^{40}K in dry soil, given by their medians, were, respectively, 41, 75, and 176 Bq kg^{-1}. These results reveal that the terrestrial gamma dose rates in Sao Paulo are higher than the world average, a fact that can be attributed to high thorium concentration. Direct measurements of dose rates were compared to the corresponding values determined from radionuclide concentrations in soil. Good agreement between methods was found.**
Health Phys. 88(1):000–000; 2005

Key words: spectrometry, gamma; soil; naturally occurring radionuclides; radioactivity, environmental

INTRODUCTION

The naturally occurring radionuclides ^{40}K, ^{232}Th, and ^{238}U in soil and building materials are the main source of human exposure to radiation. Surveys are performed worldwide in order to determine their presence in soil and correlate it to environmental dose to the population. In Brazil (the world's 5th largest population), however, there has been no systematic study in this subject. There are no data about the country in the last UNSCEAR Report (2000), except for specific areas of exceptionally high background radiation. As the city of São Paulo makes up about 10% of the Brazilian population, results obtained in the city are a first step in characterizing the whole country. Knowledge about the global background radiation is important, as there is a considerable controversy among experts about radiation risks or benefits at low doses (less than ~100 mSv y^{-1}) and low dose rates (Johansson 2003; Kaiser 2003).

Furthermore, the background level can be used as a parameter in remedial actions after environmental contamination and, if measured continuously, can give information about the trends with time and the influence of manmade actions (Maiello 1997).

A previous work compiled the results of direct measurements of total gamma dose rate performed throughout the city, either in indoor and outdoor environments (Yoshimura et al. 2004). It showed that the average values are higher than in the rest of the world. Indoor results are especially higher than the outdoor ones, and, as the city is highly urbanized, it was suspected that the presence of buildings in our city enhances rather than shields the external radiation (Muck 1996). This work uses the methods described to distinguish total outdoor dose rates in the city from those attributable to natural radionuclides in soil.

METHODS

Direct measurements of dose rates at 1 m height in parks

The gamma dose rate in typical urban outdoor environments has two main sources: soil and nearby building materials. It is not possible to identify each component separately in a direct measurement.

One possible means of evaluating the soil component is to measure the dose rate in places far away from buildings, where soil is the only source of gamma

* Departamento de Física Nuclear, Instituto de Física, Universidade de São Paulo, P. O. Box 66318, 05315-970, São Paulo, SP, Brazil.

For correspondence or reprints contact: E. Y. Yoshimura at the above address, or email at e.yoshimura@dfn.if.usp.br.

(*Manuscript received* 17 *January* 2004; *revised manuscript received* 23 *June* 2004, *accepted* 11 *September* 2004)

0017-9078/05/0

radiation, and extrapolate the results to the rest of the city. This procedure was adopted in 24 big parks in the city of São Paulo. In each location, two measuring points situated over plane geometries (when possible) were selected. The measurements were performed with a portable NaI spectrometer calibrated in the dosimetric quantity *ambient dose equivalent,* $H^*(10)$ (Yoshimura et al. 2002). The acquisition time was 10 min. Typical uncertainties, evaluated through repeatability experiments, range from 1 to 2%.

Unfortunately, the city has very few places isolated from buildings and the results obtained in them could be non-representative of the soil of the whole city. To solve this problem, an indirect procedure was used to determine the soil dose-rate at 1 m height in places where there is a non-negligible component from nearby buildings. This method is described below.

Dose rate measurements performed deep in the soil

Analytical model. Evans (1955) showed that the absorbed dose rate in a small mass in the center of a homogeneous sphere, uniformly filled with a radioactive medium having n emissions of photons with energy $h\nu$ per unit mass per unit time, could be given by the following approximate expression:

$$\dot{D}_2 = nh\nu\frac{(\mu_{ab}/\rho)_2}{(\mu/\rho)_1}\{1-e^{-\mu_1 R}+(\mu_1/\mu_{ab1}-1)[1-e^{-\mu_1 R}(1+\mu_1 R)]\}, \quad (1)$$

where R is the radius of the sphere, and μ, μ/ρ and μ_{ab}, μ_{ab}/ρ are, as usual, the linear and mass attenuation and energy-absorption coefficients of the radioactive medium (sub-index 1) and the medium in the center of the sphere, where the dose is calculated (sub-index 2).

Eqn (1) gives the geometric dependence of the dose rate. For a radius R of about $6/\mu_1$, i.e., 6 mean free paths (mfp) of the $h\nu$ photon in the radioactive medium, the sphere can be considered infinite, i.e., the dose rate saturates with the radius. Under this condition, namely $\exp(-6) \sim 0$, eqn (1) reduces to

$$\dot{D}_2 = nh\nu\frac{(\mu_{ab}/\rho)_2}{(\mu_{ab}/\rho)_1}, \quad (2)$$

which can be interpreted as a statement of energy conservation. It is possible, then, to define conversion factors (CF) between the dose rate in an infinite medium and the emission rate of the $h\nu$ photons:

$$CF_{h\nu} \equiv \frac{\dot{D}_2}{n} = h\nu\frac{(\mu_{ab}/\rho)_2}{(\mu_{ab}/\rho)_1}. \quad (3)$$

Considering soil as the infinite medium (with known atomic composition) and assuming that the natural radionuclides are uniformly distributed and in secular equilibrium, eqn (3) can be calculated for all the gamma lines of each radionuclide:

$$CF_{radionuclide} = \sum p_i CF_i, \quad (4)$$

where the sum is calculated over all the gamma lines, weighted by their relative probabilities of emission p_i.

Eqn (4) was evaluated for the three natural radionuclides, considering air as the dosimetric medium. The atomic composition of soil was the same as that used by Saito (1995). The attenuation coefficients of both soil and air were taken from NIST website (2003), as well as the gamma intensities p_i. Table 1 presents the results of the calculation and compares them to the conversion factors for the dose rate at 1 m height in a semi-infinite model (Saito 1995). AQ: A T1

It can be seen that there is an approximately constant ratio of about 0.46 between the doses at 1 m height in a semi-infinite model and inside an infinite volume, regardless of the nuclide considered. In fact, this ratio was expected to be roughly 0.5, because of the ratio between the solid angles in the 2π and 4π geometries, but the actual ratio is smaller due to the attenuation of radiation in air in the semi-infinite model.

The similarity of the calculated ratios can be understood as a consequence of the fact that the higher energy emissions of the series are the main responsible for the dose rate. In the limit of low energies, the ratio is expected to decrease as the 1 m height value is diminished through attenuation in air. On the other hand, for high energies the ratio is expected to reach 0.5 asymptotically as the attenuation and backscattering in air becomes negligible.

Monte Carlo verification. In order to verify the behavior of the dose rate with the photon energy and the sphere radius predicted by the analytical model, Monte Carlo simulations of the problem were made with the MCNP code (Briesmeister 1993). The first step was the calculation of the absorbed dose (tally F6 of MCNP) in a "detector sphere" of air with 1 cm radius inserted in the center of a soil sphere, whose radius was varied. Photon

Table 1. Comparison between conversion factors (nGy h^{-1} per Bq kg^{-1}) for the doses at 1 m height and at depth.

Radionuclide	CF at 1 m height (Saito 1995)	CF at depth (eqn 4)	Ratio
^{238}U series	0.463	0.990	0.47
^{232}Th series	0.604	1.312	0.46
^{40}K	0.0417	0.093	0.45

balt5/zhl-hp/zhl-hp/zhl00105/zhl2528-05a | browncm | S=7 | 10/23/04 | 0:22 | Art: 154273 | Input-8000

emissions were uniformly distributed in the sphere of soil with the same composition used in the other calculations. It was found, as predicted by eqn (1), that there is a saturation of the dose rate for a radius of about 6 mfp of
F1 the primary photons. Fig. 1 shows the results for 1 MeV photons, where each point corresponds to a number of histories necessary to keep the statistical uncertainty of calculations below 5%. Despite of the systematic overestimation (not more than 10%) of the analytical values in relation to the simulated ones, the predicted saturation with the radius occurs at the same point. The cause of the overestimation is probably an approximation used to derive eqn (1), which states that in an infinite medium the build-up factor increases linearly with the distance to the source of radiation (Evans 1955).

Fixing the radius of the soil sphere in 7 mfp (to guarantee the saturation), the second step was to calculate the dose in the center of the soil sphere as a function of the photon energy $h\nu$. The results were compared to those obtained from eqn (3), and a very good agreement
F2 was obtained, as shown in Fig. 2.

Conclusion. Therefore, the conclusion based on the analytical model and verified by Monte Carlo calculations is that a dose rate measurement performed deep in the soil (at a depth that the surrounding soil volume can be considered infinite) allows the determination of the 1 m height value in a semi-infinite model, using a multiplying factor of 0.46, no matter the proportion of each radionuclide in soil. The crucial point necessary for the validity of this conversion is the equivalence (in moisture, atomic composition, and radionuclide concentration) of the surrounding soil sphere that contributes to the depth value and the huge soil volume that contributes to the 1 m height value. This is particularly important because the first 20 cm layer of soil in the semi-infinite model contributes with a great fraction (around 80%) of the total dose rate at 1 m height (Beck 1972). Nevertheless, this same layer has a much smaller influence on the depth value, as shown in Fig. 1. A large variation in soil along the depth profile might cause deviations in the ratio of dose rates.

It is worthwhile to mention that the same soil depth that produces the saturation of dose is sufficient to shield the radiation coming from any natural source situated outside the soil.

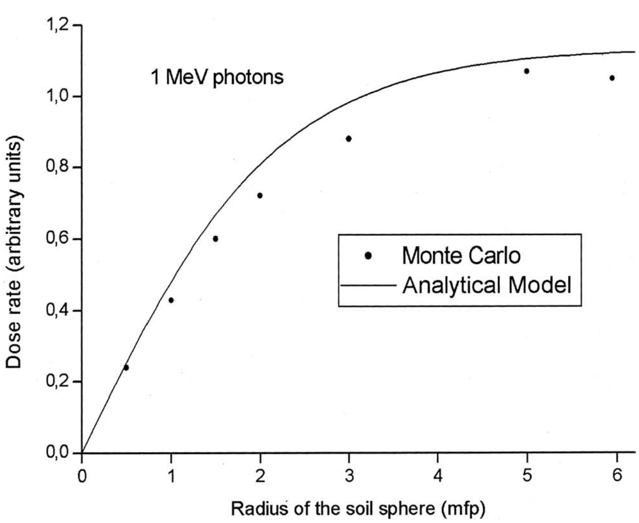

Fig. 1. Saturation of the dose rate with the radius of the soil sphere given by the analytical model and Monte Carlo calculations for 1 MeV photons.

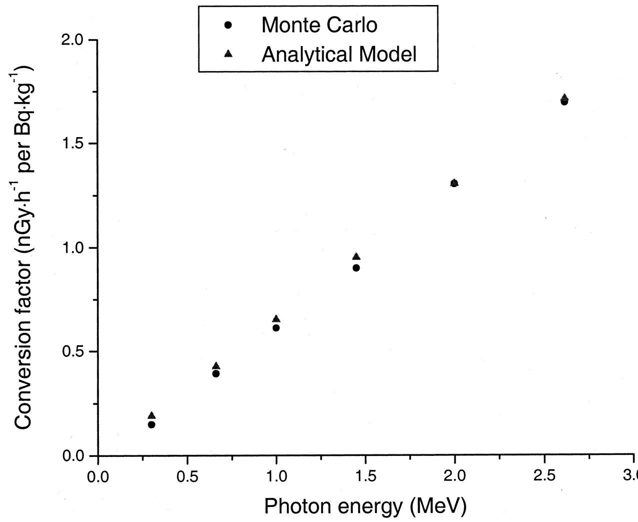

Fig. 2. Comparison between conversion factors for the dose inside an infinite volume obtained by analytical (eqn 3) and Monte Carlo calculations.

Measurements. The method described above solves the problem of determining the soil contribution to the 1 m height dose rate in places where there is an additional component due to nearby buildings. The soil component can be evaluated at any place where it is possible to dig a hole in the soil. Thus, 60 squares (*square* meaning an open green area in the city, smaller than a park, in which there is a non-negligible influence of nearby buildings to the dose rate) of the city were included in the data group, which made the global results certainly more representative than they would be taking into account only the values of the 24 parks.

The depth measurements were performed, both in parks and squares, with the same portable spectrometer, which has a probe that can be introduced in the holes. The hole depth and diameter were about 70 cm and 7 cm, respectively. In principle, the depth should be about 6 mfp of the most energetic natural gamma emission. In practice, the 70 cm were more than sufficient to guarantee the saturation of the dose rate.

Gamma spectrometry of soil samples

Forty-two soil samples were collected in the parks, in the same places where the depth measurements were performed, to be analyzed by gamma ray spectrometry. The procedure was the same adopted by Ribeiro et al. (2001), based on the comparison of photopeak areas measured in the samples and in calibrated standards. In this work, the matrix of the standards was silica, to reproduce with better accuracy the soil attenuation properties. The spectra were acquired with a HPGe detector with 30% relative efficiency, and an acquisition time of 12 h was used. Each individual value of activity had an uncertainty given mainly by the statistics of counting of the photopeaks, which varied between 2–6% for the ^{232}Th and ^{226}Ra series and 3–40% for ^{40}K.

The whole ^{238}U decay series was assumed to be in secular equilibrium with ^{226}Ra, whose activity was determined through the 609 keV photopeak of ^{214}Bi. This is not strictly correct as ^{238}U and ^{226}Ra can be (and frequently are) in radioactive disequilibrium. Nevertheless, the elements of the chain after ^{226}Ra (in equilibrium with it) are responsible for more than 98% of the dose rate of the ^{238}U series. For the ^{232}Th series, two photopeaks (583 keV from ^{208}Tl and 911 keV from ^{228}Ac) were measured in order to verify the occurrence of disequilibrium between ^{228}Ra and ^{228}Th. The concentration of ^{40}K was determined by its only emission of 1,461 keV.

RESULTS AND DISCUSSION

Dose rate measurements

A summary of the dose rate results is presented in
T2 Table 2. The mean dose rate at 1 m height is significantly higher in the squares than in the parks. On the other hand, the mean dose at depth is basically the same in both groups, taking into account the statistical fluctuation of the data. This reflects the fact that the depth value relates only to the soil radionuclide concentration, while the 1 m height value, in the squares, has an additional component from nearby buildings.

The mean ratio between the 1 m height and depth values obtained in the parks was 0.43(1), which is very close to the 0.46 ratio predicted theoretically. This is a very interesting result that shows that even in actual conditions, where the assumed hypotheses for the conversion (mainly soil homogeneity and semi-infinite geometry) certainly are not strictly satisfied, the conversion from the depth measurement to the 1 m height value is, on the average, correct.

Gathering the depth results of both data groups, it was found that they follow with good agreement a log-normal distribution ($\chi^2 = 0.41$). The histogram and the fitted function are presented in Fig. 3, and the fitted F3
parameters are in Table 3. T3

The depth value represents the soil radioactivity better than the 1 m height value because it is unaffected by the presence of buildings and by the topography. So, the expected distribution of soil dose rates at 1 m height over the city, at plane geometries, is the same as presented in Fig. 3 except for a factor 0.46. The median, for example, would result in a dose rate of $176 \times 0.46 = 81$ nSv h^{-1}. Using the conversion between $H^*(10)$ and absorbed dose in air (Yi et al. 1997), we find the median of the absorbed dose rate in air to be 66 nGy h^{-1}. This value is 15% above the world median (57 nGy h^{-1}) published in the UNSCEAR report (2000). The origin of these high values was investigated by the gamma spectrometric analysis of soil samples, shown in the next section.

An important conclusion can be made regarding the soil component to the total gamma dose rate in the city. As the total outdoor gamma dose rate was reported to be 108 nSv h^{-1} (Yoshimura et al. 2004), the manmade structures increase the expected soil dose in a semi-infinite geometry about 35%, on average.

Gamma ray spectrometry of soil samples

The mean and median values of the distributions of specific activities of ^{232}Th, ^{226}Ra and ^{40}K determined from gamma spectrometric analysis in the 42 dried soil samples are presented in Table 4. World medians are T4

Table 2. Results of direct measurements of H*(10) rate (nSv h^{-1}) throughout the city.

Dataset	Number of points	Median	Mean	Standard deviation	Standard deviation of the mean
Parks—1 m height	42	61	73	30	4.7
Parks—depth	42	164	177	86	13
Squares—1 m height	69	94	93	17	2.2
Squares—depth	69	179	186	52	6
Squares + Parks—depth	111	171	183	67	6

balt5/zhl-hp/zhl-hp/zhl00105/zhl2528-05a | browncm | S=7 | 10/23/04 | 0:22 | Art: 154273 | Input-8000

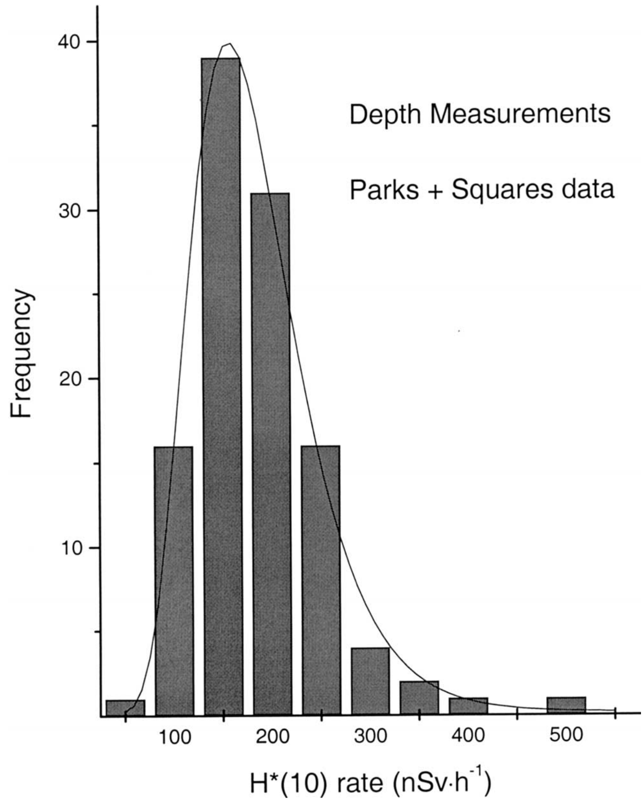

Fig. 3. Experimental and fitted (log-normal) distributions of $H^*(10)$ rates measured at depth.

Table 3. Parameters of the experimental and fitted distribution of H*(10) rates (nSv h^{-1}) measured at depth (Fig. 3). (GM = Geometric Mean, GSD = Geometric Standard Deviation).

	GM	GSD	1st quartile	Median	3rd quartile
Experimental data	172	1.42	140	171	213
Fitted log-normal	175.8 (14)	1.395 (9)	141	176	220

Table 4. Parameters of the experimental distribution of specific activities (Bq kg^{-1}) of the natural radionuclides in 42 dry soil samples. The last column shows world values from UNSCEAR Report (2000).

Radionuclide	Median	Mean	Standard deviation	World median (UNSCEAR 2000)
^{232}Th	76	88	49	49
^{226}Ra	41	44	19	19
^{40}K	176	222	223	223

also shown. The obtained distributions are also asymmetric, which can be verified comparing the median to the mean.

The ^{228}Ra and ^{228}Th activities in all samples were found to be in a condition consistent with secular equilibrium, within experimental uncertainties, so that their mean value was adopted as the activity of the whole ^{232}Th series in each sample.

The soil in São Paulo is significantly richer in ^{232}Th than the world average, while ^{226}Ra activities are similar to the ones reported in the literature (UNSCEAR 2000). The ^{40}K activity is significantly lower than worldwide values, but the values varied up to two orders of magnitude. This likely reflects the great mobility of potassium in the surface soil.

Thus, it can be concluded that ^{232}Th series is the major force responsible for the high dose rate values found in the city.

Comparison between gamma spectrometry and direct measurements of dose rates

Using the conversion factors from Saito (1995) and from eqn (4) (Table 1, columns 2 and 3), it is possible to compare the direct measurements of dose rates at 1 m height and at depth, respectively, with those determined from the measured specific activities. This was done for each of the 42 soil samples, correcting the specific activities of dry soil by a factor 0.81 as suggested by UNSCEAR (2000) to take into account the in-situ soil moisture. The comparison is shown in Figs. 4 and 5, where the $y = x$ line is shown as a visual guide. F4 F5

The correlation coefficients are 0.75 (1 m height dose) and 0.87 (depth dose). This means that spectrometric analysis of soil samples and direct measurements of dose rates are consistent methods for the whole set of measuring points. Once again it is important to emphasize that the conversion between concentration and dose rate depends on some hypotheses that can't be achieved in field measurements, but, even so, the methods are consistent on the average. The depth data, in particular,

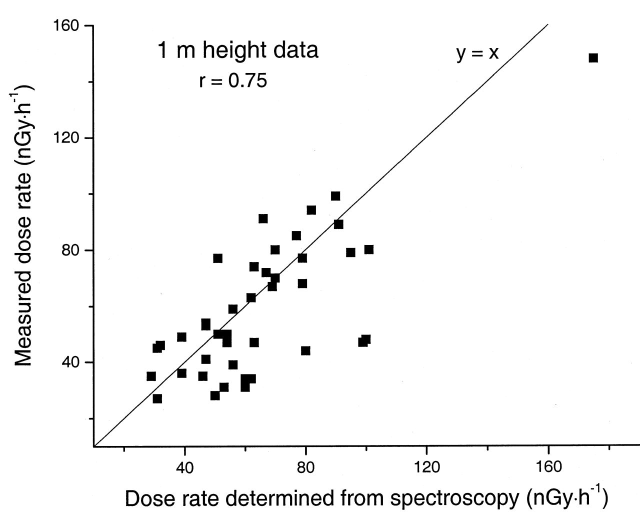

Fig. 4. Comparison between dose rates at 1 m height measured directly and determined from the radionuclide concentrations.

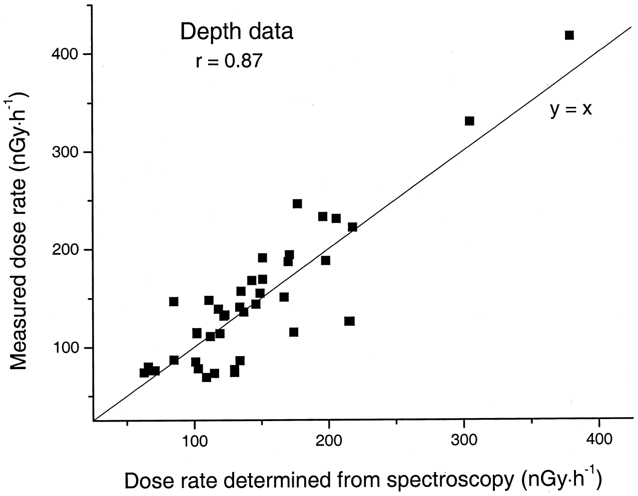

Fig. 5. Comparison between dose rates at depth measured directly and determined from the radionuclide concentrations.

present better correlation than the 1 m height data, which can be explained in terms of representativeness of the soil samples. Each sample used for the spectrometric analysis is certainly more representative of the relatively small volume of soil that contributes to the depth measurement than of the much larger volume that contributes to the 1 m height measurement, where the soil inhomogeneity is probably more significant.

CONCLUSION

In this work the influence of soil radioactivity to the total outdoor gamma dose rate in the city of São Paulo was analyzed. Direct measurements of dose rates were performed at 111 points. Forty-two soil samples were analyzed by gamma spectrometry in order to determine the specific activities of natural radionuclides. A new method was developed, which allowed the evaluation of the dose rate due to soil even in places where there is an additional component coming from nearby buildings.

The median of the dose rate values in São Paulo is above the world median. This is a consequence of high thorium concentrations in the soil, about two times greater than the world average. Comparing the soil dose rate to the total dose rate measured previously, one concludes that man-made structures increase the outdoor dose rate about 35%, on the average.

Acknowledgments—The authors thank Professors F. Y. Hyodo and N. H. Medina for lending the equipment necessary to perform some of the measurements. F. H. M. Medeiros also thanks CNPq for the financial support.

REFERENCES

Beck HL. The physics of environmental gamma radiation fields. In: Adams JAS, Lowder WM, Gesell TF, eds. The natural radiation environment II. Houston: Rice University; 1972: 101–133.

Briesmeister JF. MCNP—A general Monte Carlo N-particle transport code, version 4A. Los Alamos: Los Alamos National Laboratory; LA-12625-M; 1993.

Evans RD. The atomic nucleus. New York: McGraw-Hill; 1955.

Johansson L. Hormesis, an update of the present position. Eur J Nucl Med Mol I, 30:921–933; 2003.

Kaiser J. A healthful dab of radiation? Science 302:378–378; 2003.

Maiello ML. The variations in long term TLD measurements of environmental background radiation at locations in southeastern New York state and northern New Jersey. Health Phys 72:915–922; 1997.

Muck K. Shielding of buildings in an urban environment. Rad Prot Dosim 63:113–121; 1996.

National Institute of Standards and Technology. Tables of x-ray mass attenuation coefficients and mass energy-absorption coefficients [online]. Available at: http://www-.physics.nist.gov/PhysRefData/XrayMassCoef/cover.html. Accessed June 2003.

Ribeiro FB, Roque A, Boggiani PC, Flexor JM. Uranium and thorium series disequilibrium in quaternary carbonate deposits from the Serra da Bodoquena and Pantanal do Miranda, Mato Grosso do Sul State, Central Brazil. Appl Rad Isot 54:153–173; 2001.

United Nations Scientific Committee on Effects of Atomic Radiation. Sources and effects of ionizing radiation. Report to the General Assembly, with Scientific Annexes. New York: United Nations; 2000.

Yi CY, Jun JS, Chai HS, Oh JJ, Yun JY. Measurement of ambient dose equivalent using a NaI(Tl) scintillation detector. Radiat Prot Dosim 74:273–278; 1997.

Yoshimura EM, Umisedo NK, Okuno E. Assessment of ambient dose equivalent rate: performance of an automatic survey meter as an instrument to quantify the presence of radiation in soils. Nucl Instr Meth Phys Res A 487:457–464; 2002.

Yoshimura EM, Otsubo SM, Oliveira RER. Gamma ray contribution to the ambient dose rate in the city of São Paulo, Brazil. Radiat Meas 38:51–57; 2004.

■ ■

AUTHOR QUERIES

AUTHOR PLEASE ANSWER ALL QUERIES **1**

1—Author? Please add complete citation to ref list. Tx, Ed.