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Analysis of photon interaction parameters as function of soil composition

M. E. Medhat · L. F. Pires · R. C. J. Arthur

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Abstract In the present work the total mass attenuation coefficients (μ/ρ) for some soils collected from the Southeast and South of Brazil were measured at 59.5 (^{241}Am) and 661.6 keV (^{137}Cs) photon energies. The experimental values of the soils μ/ρ were compared with XCOM program calculations and GEANT4 Monte Carlo simulations. Total atomic and electronic cross-sections, effective atomic and electron numbers of all soil samples were calculated in a wide energy range (1 keV–100 GeV). The values of these parameters have been found to vary with photon energy and chemical composition of the soil. The variations of these parameters with energy are shown graphically for total photon interactions. The results showed that loamy soils have low photon attenuation parameters than clayey ones for the region of 59.5 keV.

Keywords Soil composition · Photon interaction · Attenuation coefficients · Monte Carlo simulations

M. E. Medhat (✉)
Experimental Nuclear Physics Department, Nuclear Research Centre, P.O. 13759, Cairo, Egypt
e-mail: medhatme@ymail.com

M. E. Medhat
Institute of High Energy Physics, CAS, Beijing 100049, China

L. F. Pires
Laboratory of Soil Physics and Environmental Sciences, Department of Physics, State University of Ponta Grossa, UEPG, Ponta Grossa, PR CEP 84.030-900, Brazil

R. C. J. Arthur
Center for Nuclear Energy in Agriculture, CENA, Piracicaba, SP CEP 13.400-970, Brazil

Introduction

Soils have chemical compositions characterized by the presence of major compounds, such as SiO_2 (40–90 %), Al_2O_3 (5–20 %), Fe_2O_3 (2–20 %) and minor compounds such as CaO, K_2O, MnO, TiO_2, and so on. The major compounds in soils present concentrations that exceeds 100 mg kg^{-1} [1, 2]. The most important quantity characterizing the penetration and diffusion of gamma radiation through soil is the mass attenuation coefficient (μ/ρ). It is the fundamental parameter to derive many other parameters of dosimetric interest such as molecular, atomic (σ_a) and electronic (σ_e) cross sections, effective atomic number (Z_{eff}) and electron density (N_{el}). These quantities can be evaluated theoretically and experimentally. In literature, a variety of works relevant to μ/ρ and Z_{eff} estimations for different compound materials has been published by several authors in different categories. Recently, a significant number of papers about experimental and theoretical determinations of these parameters in various elements, compounds and mixtures were published [3–11]. Attenuation of photons has been reported as the most accurate and convenient technique for non destructive measurements of soil parameters such as: water retention curve, water content, bulk density and porosity [12–20].

In heterogeneous media such as soil the atomic number cannot be represented by a single number, as in the case of elements. This number in such materials is called effective atomic number. Z_{eff} is considered the most basic and important quantity among the parameters determining the constitutive structure of composite materials. It gives basic information about the characteristics of multi elemental materials. Attenuation (due to scattering or absorption) of photons in composite materials depends basically on two factors, Z_{eff} and N_{el}. Thus, Z_{eff} has proved to be a convenient parameter for interpreting the γ-ray attenuation by soil. Moreover, the study of Z_{eff} provides important information on the composition of the soil. For example, higher values of Z_{eff}, soils tend to contain more inorganic compounds and metals than the smaller values [11].

The objective of this study deals with the theoretical and experimental determination of mass attenuation coefficients of soils with different textures. Other parameters such as molecular, atomic and electronic cross sections, effective atomic number and electron density were also calculated for photon energies in the range 1 keV–100 GeV. Theoretical results were compared with the measurements obtained with photon energies of 59.5 keV (^{241}Am) and 661.6 keV (^{137}Cs). The soils under consideration were collected from Southeast and South of Brazil.

Theoretical basis and simulations

Mathematical basis

For materials composed of several elements, it is assumed that the contribution of each element to total photon attenuation is additive. In such cases, μ/ρ of any material with density ρ is related to $(\mu/\rho)_{\mathrm{i}}$ values of its constituents by the mixture rule, $\sum_i c_i \left(\frac{\mu}{\rho}\right)_i$, where c_{i} is the proportion by weight of the i-th constituent element. The total molecular cross-section (σ_{m}) can be calculated from the knowledge of μ/ρ by using the following relation [9]:

$$\sigma_{\mathrm{m}} = \left(\frac{\mu}{\rho}\right)\frac{M}{N_A}, \quad (1)$$

where $\mathrm{M} = \sum_i n_i A_i$, is the molecular weight of the compound, N_A is the Avogadro number, A_i is the atomic weight of the i-th element and n_i is the number of formula units in the molecule. The average atomic cross-section can be obtained by dividing σ_{m} by the total number of formula units as follows:

$$\sigma_{\mathrm{a}} = \sigma_{\mathrm{m}}\frac{1}{\sum_i n_i}. \quad (2)$$

Similarly, the average electronic cross-section is given by

$$\sigma_{\mathrm{e}} = \frac{1}{N_{\mathrm{A}}}\sum_i \frac{f_i A_i}{Z_i}\left(\frac{\mu}{\rho}\right)_i, \quad (3)$$

where $f_i = \mathrm{n}_i/\sum_j n_j$ and Z_i are fractional abundance and atomic number of the constituent element, n_j is the number of atoms of the constituent element and $\sum_j n_j = n$ is the total number of atoms present in the molecular formula. Therefore, the effective atomic number can be now defined as:

$$Z_{\mathrm{eff}} = \frac{\sigma_{\mathrm{a}}}{\sigma_{\mathrm{e}}}. \quad (4)$$

Finally, the effective electron number or electron density (number of electrons per unit mass) of the material can be derived from:

$$N_{\mathrm{el}} = \frac{\left(\frac{\mu}{\rho}\right)}{\sigma_{\mathrm{e}}} = \left(\frac{Z_{\mathrm{eff}}}{M}\right)N_{\mathrm{A}}\sum_i n_i. \quad (5)$$

XCOM software

Calculations of μ/ρ of samples were carried out by the XCOM program [21]. The software can generate cross-sections and attenuation coefficients for elements, compounds or mixtures in the energy range 1 keV–100 GeV. The calculated data can be given in the form of total cross-sections and attenuation coefficients as well as partial cross-sections of the following processes: incoherent and coherent scatterings, photoelectric absorption and pair production in the field of the atomic nucleus and electrons. The program possesses a comprehensive database for all elements over a wide range of energies, constructed through the combination of photoelectric absorption, incoherent and coherent scatterings and pair production (nuclear and electric field) cross-sections.

GEANT4 simulation code

The GEANT4 code is based on object-oriented programming and allows user to derive classes to describe the detector geometry, primary particle generator and physics processes models along electromagnetic, hadronic and decay physics. These simulations are based on theory, materials and elements, experimental data or parameterizations [22].

Table 1 Some physical properties of the soils studied

Soil	Sand (%)	Silt (%)	Clay (%)	Classification (USDA)	ρ_{p} (g cm^{-3})
1	66	6	28	Sandy clay loam	2.55
2	26	26	48	Clay	2.54
3	24	33	43	Clay	2.68
4	64	18	18	Sandy loam	2.53
5	29	35	36	Clay loam	2.41
6	25	20	55	Clay	2.77
7	17	22	61	Clay	2.50
8	18	20	62	Clay	ND

ρ_{p} refers to soil particle density

ND not determined, *USDA* United State Department of Agriculture

Table 2 EDXRF analyses results of the dry soil samples

Compounds	Chemical components (%)							
	Soil 1	Soil 2	Soil 3	Soil 4	Soil 5	Soil 6	Soil 7	Soil 8
Al_2O_3	30.032	32.839	42.669	21.063	22.099	35.802	52.041	35.419
BaO	–	–	–	–	–	1.344	–	–
CaO	0.227	0.155	0.155	0.166	–	–	0.179	0.251
Cr_2O_3	–	–	0.026	–	–	0.048	0.032	0.034
CuO	–	0.022	0.015	–	–	0.051	0.015	0.051
Fe_2O_3	3.469	16.737	14.126	3.008	4.832	26.128	14.346	27.515
Ga_2O_3	0.007	–	–	–	–	–	–	–
K_2O	0.073	0.100	0.222	2.038	2.178	–	0.552	0.132
MnO	0.031	0.323	0.071	0.053	0.097	0.399	0.044	0.330
NbO	–	0.005	–	–	–	0.007	0.005	–
P_2O_5	–	0.222	–	–	–	–	–	–
Rb_2O	–	–	–	0.011	0.019	–	–	–
SO_3	1.385	1.712	1.239	1.247	1.450	1.210	1.023	1.042
SiO_2	62.795	44.187	38.334	71.511	68.418	31.612	29.457	30.680
SrO	–	–	–	0.006	0.005	–	0.005	–
TiO_2	1.925	3.486	3.046	0.840	0.831	3.297	2.132	4.294
V_2O_5	–	0.115	–	–	–	–	0.080	0.161
Y_2O_3	–	0.006	–	0.004	–	0.011	0.007	–
ZnO	0.005	0.012	–	–	0.009	0.013	0.010	0.016
ZrO_2	0.053	0.084	0.096	0.053	0.063	0.079	0.073	0.075

The proposed simulation model depends on narrow beam geometry with the various photon energies. The model consists of a mono-energetic photon beam impinging on a slab of one of the selected materials. Soil mass attenuation of investigated materials are determined by the transmission method according to Beer Lambert's law ($I = I_0 e^{-\mu x}$), where I_0 and I are the incident and attenuated photon intensity, respectively, μ (cm^{-1}) is the linear attenuation coefficient and x is the thickness of the slab. The thickness x is optimized according to I_0, in order to prevent the complete photon absorption by the slab or its transmission without interaction. The primary photons emerging unperturbed from the slab are counted. The energy range of incident photons varies from 1 keV to 100 GeV. Attenuation of photons is calculated by simulating all relevant physical processes and interactions before and after inserting the investigated sample [23–25].

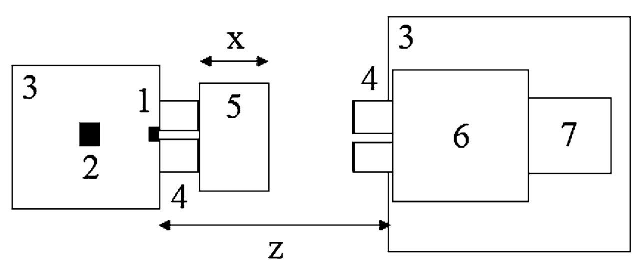

Fig. 1 Schematic diagram of the set up used to the experimental measurements of the soil mass attenuation coefficient. *1* ^{241}Am γ-ray source; *2* ^{137}Cs γ-ray source; *3* Pb shields; *4* Pb circular collimators; *5* Soil sample inside an acrylic box; *6* NaI(Tl) detector and *7* photomultiplier tube. *z* and *x* represent the distance between the radioactive source and detector and the soil sample thickness

Materials and methods

Samples collection and preparation

In the investigated study, soil samples were collected from regions located in Southeast (soils 1–3) and South (soils 4–8) of Brazil (tropical and sub-tropical climate). All samples were taken from the surface layer (0–10 cm) of every region. The physical properties of the investigated soil samples are shown in Table 1. Soil samples (1–3) were collected from an area covered by grasses and weeds cultivated with orange trees (Soil 1), coffee field (Soil 2) and mixed forest cropped with sugarcane (Soil 3). Soils (4–8) were collected from an area used for pasture (Soil 4), cultivated by tobacco field (Soil 5), maize field (Soil 6), soybean field (7) and cultivated with maize and soybean (Soil 8). Collected soil samples were dried in oven at 105 °C for 2 days and sieved through a

Table 3 Comparison among GEANT4, XCOM and experimental mass attenuation coefficients of the investigated soil samples at 59.5 and 661.6 keV

Soil type	Mass attenuation coefficients (cm^2 g^{-1})					
	Am-241			Cs-137		
	GEANT4	XCOM	Exp.	GEANT4	XCOM	Exp.
Soil 1	0.2770	0.2810	0.2734	0.0768	0.0767	0.0765
Soil 2	0.3810	0.3769	0.3829	0.0765	0.0763	0.0768
Soil 3	0.3498	0.3549	0.3472	0.0759	0.0762	0.0756
Soil 4	0.2678	0.2815	0.2546	0.0773	0.0769	0.0779
Soil 5	0.2910	0.2941	0.2891	0.0783	0.0780	0.0785
Soil 6	0.4781	0.5398	0.4258	0.0769	0.0756	0.0772
Soil 7	0.3252	0.3533	0.3164	0.0773	0.0761	0.0782
Soil 8	0.4430	0.4508	0.4336	0.0771	0.0759	0.0783

1 mm mesh sieve to obtain a homogeneous sample. Samples were kept in containers with silica-gel throughout the experimental analyses.

Elemental analysis of samples

Elemental analysis of soil samples was carried out using energy dispersive X-ray fluorescence (EDXRF) spectrometer (Shimadzu, EDX-720) equipped with Rh tube (5–50 kV; 1–1,000 μA). Samples were ground in mortar and put into proper containers supplied by the equipment manufacturer and sealed with mylar (6 μm thick). The spectra was obtained for a time of 100 s in the energy bands of Na–Sc (15 kV) and Ti–U (50 kV). All measurements were carried out with pressure under 30 Pa. EDXRF analyses of the investigated samples are presented in Table 2.

Experimental setup

A NaI(Tl) solid scintillation detector of plain type (7.62 × 7.62 cm) was used to detect gamma photons emitted from γ-ray sources of ^{137}Cs (11.1 GBq) and ^{241}Am (7.4 GBq). Circular lead collimators (2–4 and 4.5 mm),

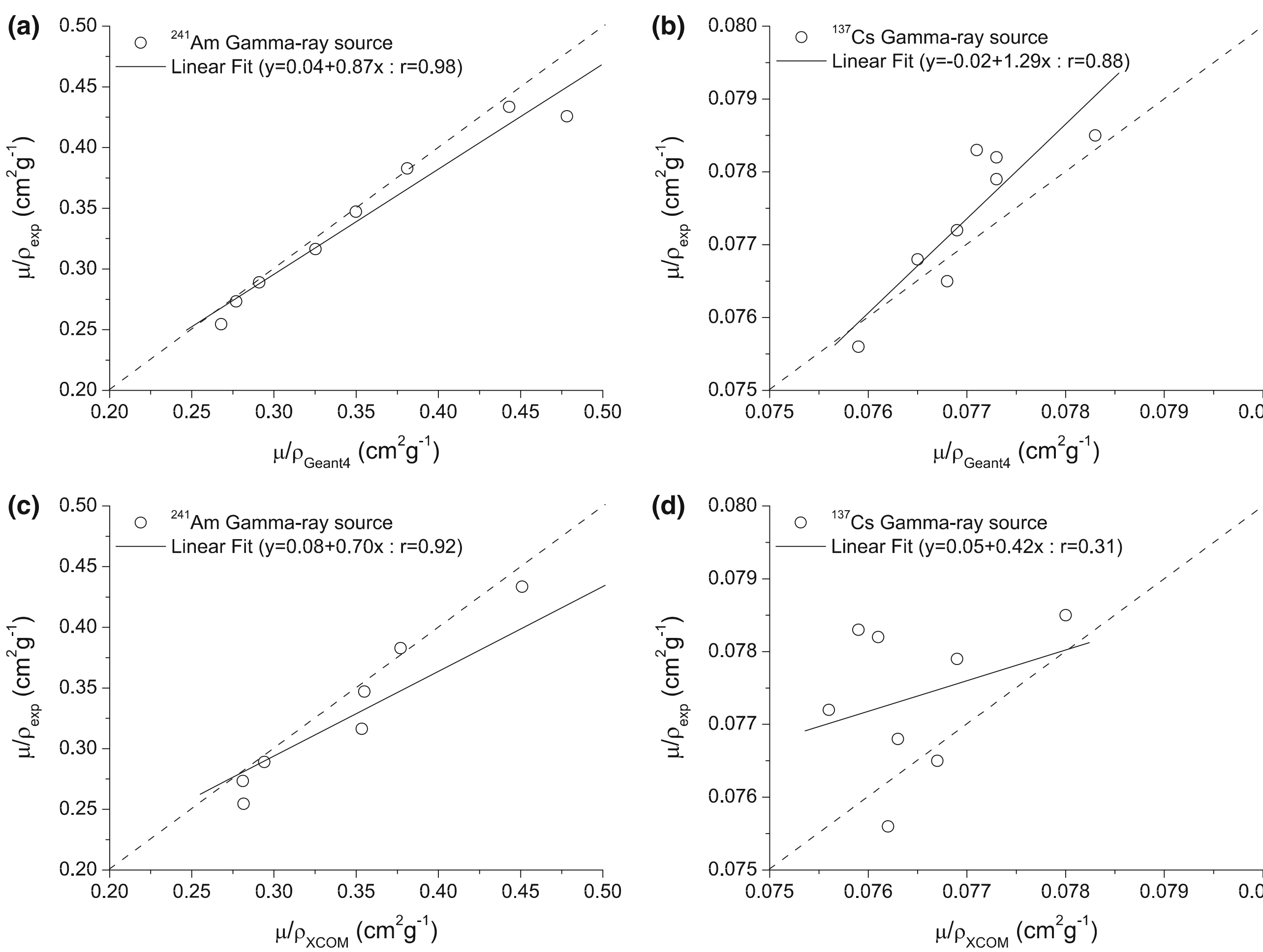

Fig. 2 Analysis of correlation among the experimental mass attenuation coefficient (μ/ρ) and theoretical μ/ρ values simulated through XCOM and GEANT4 codes for the ^{241}Am (**a–c**) and ^{137}Cs (**b–d**) γ-ray sources

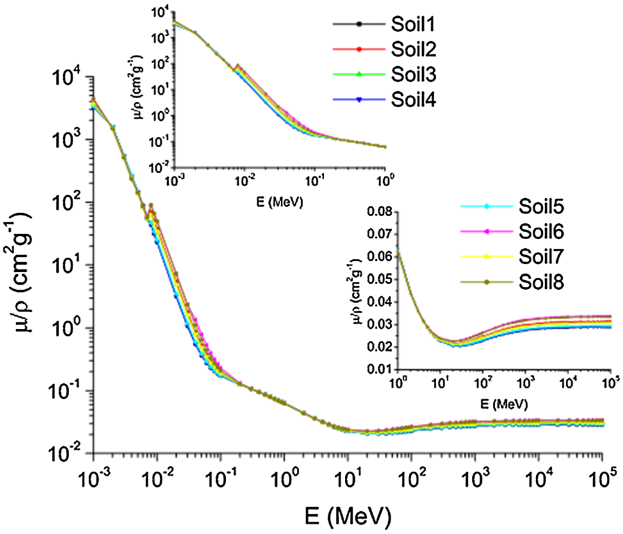

Fig. 3 Variation of photon mass attenuation coefficient of the investigated soil samples with photon energy for total photon interaction calculated by XCOM in the energy range 1 keV–100 GeV

mounted between source and detector, were utilized to collimate the radiation beam. The source and detector distance were kept constant (23 cm) and the acrylic container used to assembly the sample is 4.0 cm thick. The schematic arrangement of the experimental setup in the present investigation is shown in Fig. 1. Incident and transmitted photons for each sample were measured for sufficiently large fixed preset time to reduce statistical uncertainty. The counting time adopted in μ experimental measurements was 600 s for both of ^{137}Cs and ^{241}Am sources. The detected spectra can be translated to ASCII and processed with custom-made programs based on ROOT.

Results and discussion

The obtained results of μ/ρ for different types of soil were compared with XCOM and GEANT4 calculations as tabulated in Table 3. There is a notable deviation between the experimental and theoretical values of XCOM for most soil samples at low photon energy (59.5 keV). It is clear that the representative GEANT4 Monte Carlo generated data for μ/ρ of all samples are in agreement with experimental values. Discrepancy of the calculated and the experimental μ/ρ values could be due to deviations from narrow beam geometry in the source-detector arrangements [23].

It is notable from Fig. 2 the correlations among experimental and theoretical (XCOM and GEANT4) methods. The best correlations were obtained for μ/ρ simulated through GEANT4 in comparison to the experimental values (Fig. 2a, b). A strong correlation was also found for μ/ρ simulated through XCOM for the ^{241}Am γ-ray source (Fig. 2c), however, only a weak correlation was observed for the ^{137}Cs γ-ray source (Fig. 2d). This is due to the uncertainty results of XCOM in low energy (less than 50 keV) is smaller. These results show the importance of Monte Carlo simulations to predict γ-ray attenuation coefficients of soils [26].

The result of total μ/ρ of the studied soils as function of photon energy is presented in Fig. 3. Figure 3 shows the

Table 4 The experimental and theoretical values of atomic cross-section (σ_a), electronic cross-section (σ_e), effective atomic number (Z_{eff}) and effective electron number (N_e) for the investigated soil samples

Soil type	Am-241			Cs-137		
	GEANT4	XCOM	Exp.	GEANT4	XCOM	Exp.
σ_a (b/atom)						
Soil 1	9.48	9.62	9.36	2.63	2.62	2.62
Soil 2	13.80	13.51	13.90	2.78	2.70	2.79
Soil 3	12.60	12.60	12.50	2.73	2.68	2.72
Soil 4	9.20	9.57	8.75	2.66	2.57	2.68
Soil 5	11.01	11.01	10.8	2.71	2.59	2.72
Soil 6	18.30	20.30	16.3	2.94	2.83	2.95
Soil 7	11.80	12.60	11.4	2.80	2.68	2.83
Soil 8	16.90	17.00	16.6	2.95	2.83	3.00
σ_e (b/atom)						
Soil 1	0.994	0.995	0.993	0.253	0.251	0.252
Soil 2	1.32	1.31	1.33	0.253	0.252	0.253
Soil 3	1.22	1.23	1.24	0.254	0.253	0.254
Soil 4	0.95	0.95	0.95	0.252	0.253	0.253
Soil 5	0.98	0.99	0.98	0.251	0.251	0.252
Soil 6	1.88	1.86	1.88	0.252	0.251	0.252
Soil 7	1.23	1.22	1.21	0.252	0.252	0.251
Soil 8	1.54	1.54	1.55	0.253	0.251	0.252
Z_{eff}						
Soil 1	9.53	9.67	9.41	10.43	10.42	10.39
Soil 2	10.55	10.28	10.60	10.95	10.64	11.00
Soil 3	10.19	10.18	10.11	10.53	10.52	10.71
Soil 4	9.66	10.04	9.19	10.28	10.18	10.59
Soil 5	10.19	10.18	10.17	10.79	10.31	10.32
Soil 6	9.72	10.77	10.77	11.24	11.24	11.24
Soil 7	9.69	10.11	9.43	11.12	10.07	11.25
Soil 8	10.93	10.94	10.92	11.70	11.23	11.23
N_e ($\times 10^{23}$ electron/g)						
Soil 1	2.90	2.83	2.75	3.05	3.05	3.04
Soil 2	2.91	2.83	2.92	3.02	2.93	3.03
Soil 3	2.83	2.83	2.81	2.99	2.92	2.97
Soil 4	2.81	2.92	2.92	3.06	2.96	2.96
Soil 5	2.94	2.94	2.92	2.98	2.98	3.12
Soil 6	2.82	2.82	2.82	2.94	2.94	2.94
Soil 7	2.68	2.87	2.87	3.08	2.95	2.95
Soil 8	2.86	2.86	2.80	3.06	2.94	3.11

strong dependence of this parameter with photon energy. It can be easily seen that there are three energy ranges, where photoelectric absorption, Compton scattering and pair production, respectively, are the dominating attenuation processes. Experimental as well as theoretical values of other attenuation parameters (σ_a, σ_e, Z_{eff} and N_e) are presented in Table 4. Good agreement has been achieved between measured and calculated values.

The strongest energy dependence was observed in low photon energy region (1–100 keV). In this range, μ/ρ have the highest values, where the photoelectric absorption is significant and its cross-section is proportional to $Z^{4,5}$. In the intermediate energy region, the Compton scattering is significant, the linear Z-dependence of incoherent scattering is demonstrated and μ/ρ is found to be constant. In the high energy region, μ/ρ increases again, where the pair production is significant and μ/ρ is proportional to Z^2. The present theoretical results were in similar to the observations of Medhat [9] and Manohara and Hanagodimath [11].

For total photon interaction process, the variations of Z_{eff} and N_{el} with photon energy are shown in Figs. 4 and 5. From Fig. 4, it is clear that Z_{eff} values of the soils change with photon energy and soil chemical structures. In low energy range (1–10 keV), soils 2, 5, 6, 7 and 8 have almost constant values of Z_{eff}, while the other soils are slightly lower in their Z_{eff} values. The almost constant values for soils 2, 5, 6, 7 and 8 can be associated with the similarities between silt and sand contents among them.

In the energy range (10–200 keV), Z_{eff} values are decreased reaching their minimum values between 2 and 4 keV among soils. From the obtained Z_{eff} values, it is possible to observe that the clayey soils presented the same range of variation. Regions of Z_{eff} stabilization can be observed for all the soils studied.

It is interesting to observe the differences of Z_{eff} values among soils which can be associated sometimes with their textures. However, in the case of the loamy soils (1–4) it is hard to establish some kind of correlation due to the large variety of combinations of silt, sand and clay contents to form these types of soils. However, soils (4–5) presented similarity in Z_{eff} behavior with energy and these loamy soils have in common almost the very same values of Al_2O_3, Fe_2O_3, K_2O and SiO_2.

The variations of N_{el} with photon energy for total interaction processes (Fig. 5) are similar to that of Z_{eff} and can be explained on the similar manner. The N_{el} values show photon–energy dependence similar to that observed for Z_{eff}. This result is confirmed due to the positive correlations between Z_{eff} and N_{el} values obtained from the theoretical calculation carried out by Kucuk and collaborators [3]. These authors found small differences in Z_{eff} and N_{el} among soils independent from the texture. In the present work the discrepancies among soils for these two

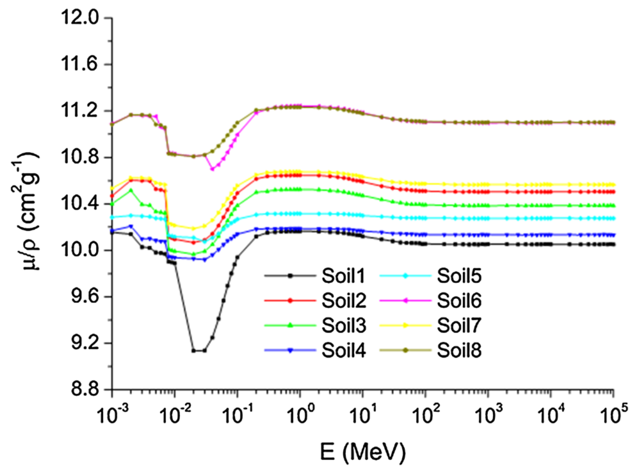

Fig. 4 Variation of Z_{eff} with photon energy of the selected soils for total photon interaction (with coherent) calculated by XCOM in the energy range 1 keV–100 GeV

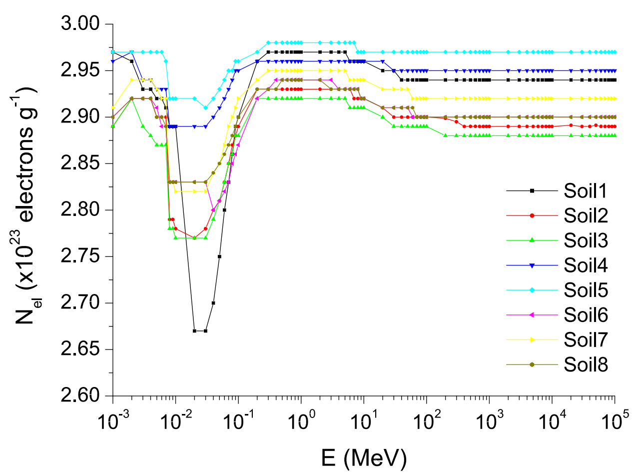

Fig. 5 Variation of N_{el} with photon energy of the selected soil for total photon interaction (with coherent) calculated by XCOM in the energy range 1 keV–100 GeV

physical parameters can be associated with differences in the nature of clays and management systems of the Brazilian soils [27].

It is important to have in mind that different proportions of sand, silt and clay will result in different soil textures. Soil texture is an important physical property related to several dynamic phenomena that take place in this porous media. As the soil is characterized by a broad distribution of particle sizes, soils with different textures can also attenuate the radiation in a different manner. For example, the loamy soils (1, 4 and 5) presented the smallest values of μ/ρ and clayey soils were characterized by the largest ones. These results are mainly related to the Fe_2O_3 content.

Considering the clay content and the soil major components it is possible to observe medium to strong positive

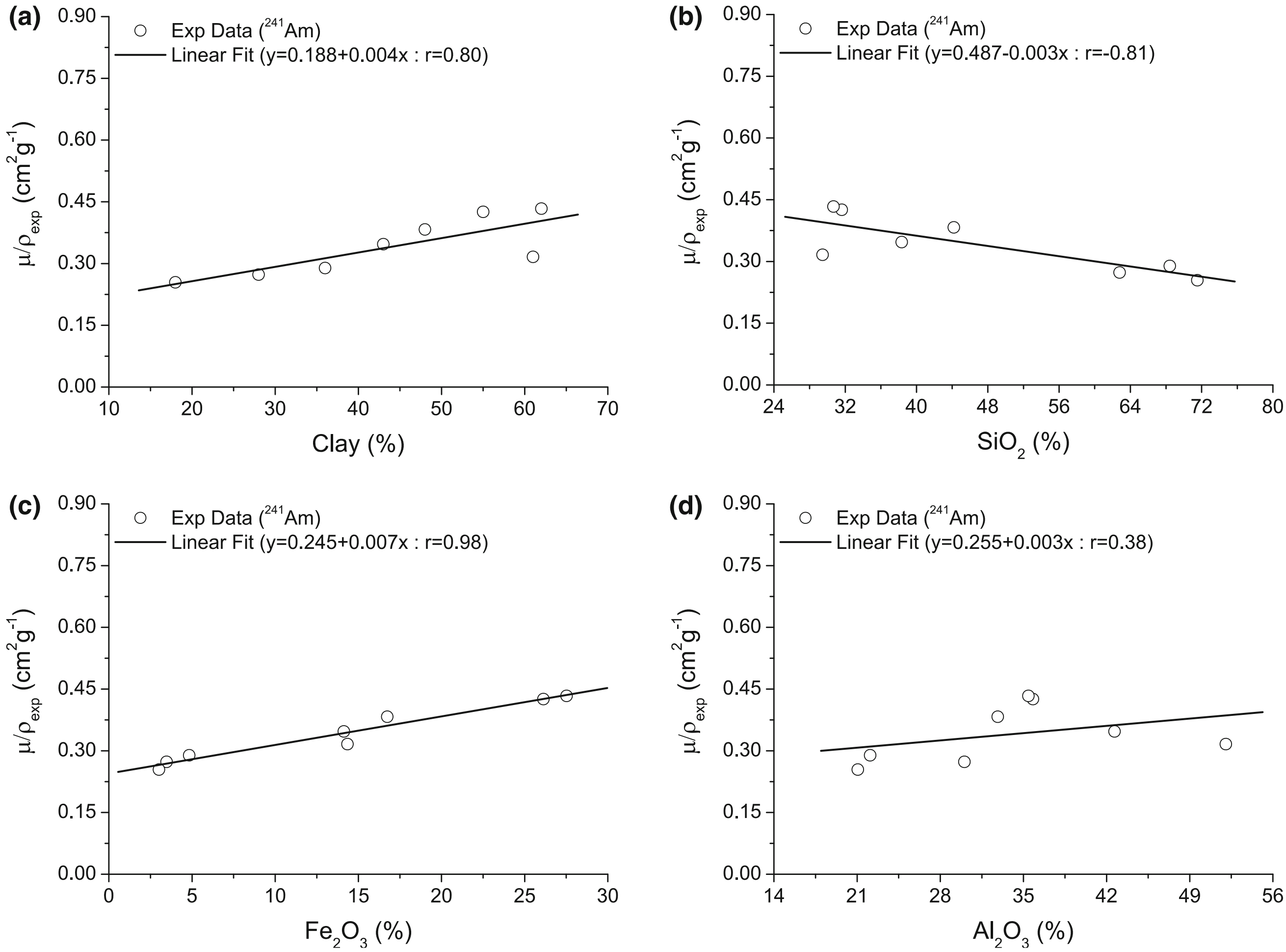

Fig. 6 Analysis of correlation between the experimental mass attenuation coefficient (μ/ρ), clay (**a**) and the major soil compounds (**b–d**) for the ^{241}Am γ-ray source

correlations between experimental μ/ρ values, clay and Fe_2O_3 contents (Fig. 6a, c). This strong positive correlation between μ/ρ and Fe_2O_3 is important because the Brazilian soils are rich in Fe. On the other hand, a medium negative correlation between μ/ρ and SiO_2 was observed (Fig. 6b). Similar results were measured by AkarTarim and collaborators [26] for the same γ-ray energy (^{241}Am). Only a weak positive correlation was found between μ/ρ and Al_2O_3 (Fig. 6d). For the case of the ^{137}Cs γ-ray source, only weak positive and negative correlations were observed between μ/ρ, clay and the major soil compounds. The results were also similar to those observed by Akar Tarim et al. [26].

Conclusion

The present study has been undertaken to get some information about mass attenuation coefficients and related parameters, effective atomic numbers and electron density for different types of soils used in agriculture purposes. The electron density and effective atomic number are closely related and they are qualitative energy dependent. A good agreement was observed between the theoretical calculations and experimental results of μ/ρ. The low incident energy region represents the one with the most remarkable differences in μ/ρ among soils. This fact is related to the Z dependence for the photoelectric absorption mechanism that governs the photon interaction in this region.

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