



Natural and anthropogenic gamma-ray emitters and iron-bearing compounds distribution in peatlands of the southern Espinhaço mountain chain, Brazil

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Abstract

To help understand the dynamics of peatlands forming on the Southern Serra do Espinhaço, Brazil, the occurring natural (^{226}Ra , ^{232}Th , ^{40}K) and anthropogenic (^{137}Cs) activity of gamma-ray emitters located at three sites, namely the localities of São João da Chapada (CH), Pinheiro (PIN) and Pau de Fruta (PDF), was studied. The iron compounds were characterized by ^{57}Fe Mössbauer spectroscopy. Differences on natural radionuclide levels and distributions were found among the peatland sites, indicating differences on mineral composition, hydric regimes and living. Significant correlation was found only for ^{232}Th – ^{226}Ra activity values (Pearson correlation coefficient of -0.5), which is different from other South American soils, where significant positive correlations among all radionuclides were determined. In PDF and CH, the dose rate results are lower than the worldwide mean value, but in PIN, they are higher because of the relatively high ^{226}Ra concentration. The only anthropogenic radionuclide is ^{137}Cs . The convection–diffusion model fits the CH and PDF activity profiles, although the PIN profile is likely affected by other soil mechanisms as well. Mössbauer spectra reveal Fe^{3+} in two different local environments and the occurrence of hematite in the upper samples of the CH site. The iron concentration decreases with depth. Contrary to soils of other regions, no significant correlation was found between iron species and the natural radioactivity. The studied peatlands exhibit clear differences between their natural and anthropogenic radionuclide distributions to those of mineral soils.

Keywords Peatlands · Radioactivity · Fe · Profiles

Introduction

Peatlands are distinctive environments capable of storing carbon, fresh water and radioactive materials as well as atmospheric deposits and pollutants (Mróz et al. 2017;

Mercader et al. 2014; Mihalík et al. 2014). The peat formation is favored by acid and anoxic conditions, along with high water content, low redox potential and the inhibitory effect of organic acids (Horák et al. 2011). Around 4.2% of the world land surface, mainly in the northern Hemisphere, is covered with peatlands (Gorham, 1991). Between 30.5 and 45.9 million hectares lie in tropical regions, which include the Southern Espinhaço mountain chain or, in the Portuguese denomination, *Serra do Espinhaço Meridional* (SdEM), with about 14,000 ha (da Silva et al. 2013). UNESCO assumes this region as a Reserve of the Terrestrial Biosphere due to its important environmental role. These peatlands exhibit heterogeneous substrates with different physical and chemical properties, which affect the water storage capacity (Mercader et al. 2014) and very likely also the radionuclide distribution. The genesis of these peatlands is actually a very complex process, which is influenced by many parameters. The knowledge of the natural (^{226}Ra , ^{232}Th , ^{40}K) and anthropogenic (^{137}Cs) gamma ray emitters supports understanding the dynamics of their components.

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Several scientific reports have dealt with the spatial ^{137}Cs distribution in soils, whose origin in South American is the radioactive fallout from atmospheric nuclear weapons tests carried out between 1955 and 1974 in the South Atlantic and Pacific Oceans (UNSCEAR 2000, 2008). Several works have found independently that in soils with high clay content, there is low bioavailability and low vertical migration rate of ^{137}Cs because cesium binds to specific clay minerals (Cremers et al. 1988; Cornell 1993; Wauters et al. 1996). Particularly, in organic matter-rich soils, as peatlands, some papers have also reported that the vertical migration rate of ^{137}Cs is also low, despite the relatively low clay content, probably due to the relatively high bioavailability, particularly due to the mycorrhizal fungi activity (Olsen et al. 1990; Steiner et al. 2002; Vinichuk and Johanson 2003; Vinichuk et al. 2004; Rosén et al. 2009).

Fijałkiewicz-Kozieł et al. (2014) studied chronologies of peat sequences by measuring the ^{14}C , ^{210}Pb and ^{137}Cs profile activities (0–20-cm depth) in a Polish peat bog. They found that most ^{137}Cs activity is related to living moss plants and proposed that the knowledge of ^{137}Cs mobility and distribution in bog profiles could be a good proxy for developing theoretical transport models and could be used to reconstruct the fallout history.

Gaca et al. (2006) determined the radioactivity profile of $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Am , ^{90}Sr , ^{137}Cs , ^{40}K and ^{228}Ac in a Tatra Mountains peatland covered by *Sphagna* sp. moss, at the border between Poland and Slovakia. ^{137}Cs activity shows the typical gaussian profile found in clay soils, with the maximum at 15–17 cm, but the authors claim that the dating method based on radionuclides, often used for sediments, seems to be limited in that peat. The ^{40}K activities decrease with the soil depth, contrary to that reported for mineral soils (Montes et al. 2012). Besides, Rosén et al. (2009) analyzed the ^{137}Cs soil profiles in peats from Sweden, where they found that vascular plants tend to accumulate ^{137}Cs , which was also observed by Mróz et al. (2017), who also found that the low soil pH favors the uptake of cesium.

Previous studies (Fijałkiewicz-Kozieł et al. 2014; Mietelski et al. 2008) observed higher activity concentrations of ^{137}Cs in the upper layers of the peat bog profiles and interpreted this fact as an upward transport of ^{137}Cs through the plants growing in the peat bog. The transfer of ^{137}Cs from the peat mass to plants varies depending principally on the species and the physical and chemical interactions.

Reported data on natural radioactivity in peatlands are scarce and indicate that several factors and features govern the ^{137}Cs activity profiles in soils; specific studies must be developed in the peatlands around the world.

Toward understanding SdEM peatlands dynamics, this work presents ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs activity profiles in three peatlands belonging to SdEM, correlating them with

the iron existing chemical species non-destructively characterized by Mössbauer spectroscopy.

Materials and methods

Studied area

The SdEM altitudes vary from 1000 to 2000 m; the mean annual temperature is 18.7 °C, with cold and dry winters and mild wet summers. The average annual rainfall is 1473 mm (da Silva and Silva 2016). The peatland substrates are heterogeneous and variable in their physical and chemical properties, which depend on the botanical characteristics of the original covering vegetation, the intensity of decomposition and the content of inorganic elements. All these factors may also affect its water storage capacity (Mercader et al. 2014).

In this study, three undisturbed SdEM peatlands were chosen: (1) Pau-de-Fruta (PDF), (2) Pinheiro (PIN) and (3) São João da Chapada (CH).

The Pau-de-Fruta peatland is localized at 1350-m altitude, 4 km away from the city of Diamantina (18°16'17,05"S, 43°41'01,15"W); the Pau-de-Fruta peatland is an ecosystem formed by typical Hemic Haplosaprists (Soil Survey staff et al. 2010), and the profile is ~90 cm thick. These soils are particularly rich in organic matter in an intermediate stage of decomposition and contain a large amount of fibrous material, which produce a pedo-environment of poor drainage. The soils are acidic (3.8 < pH < 5.3) and exhibit a relatively high cation exchange capacity, related to their high amount of organic matter. Humic is the prevailing organic matter species. According to data reported by da Rocha Campos et al., the average volume of water retained in the whole Pau-de-Fruta bog is 629,782 m³ and its genesis started about 20,000 years ago (da Rocha Campos 2009).

The peatlands at Pinheiro have a mean altitude of 1247 m above sea level. The sampling site is located at 18°03'48"S 43°39'41"W in the state of Minas Gerais, Brazil. These soils have characteristically high water contents, high acidity, low oxygen, increasingly high chemical reducing potential and high proportions of organic acids. According to Horak-Terra et al. (2014), the genesis of Pinheiro peatland started about 60,000 years ago.

The peatland at São João da Chapada (geographical coordinates, 18°05'43.6"S 43°47'6.4"W) is sited at an altitude of 1330 m above sea level, in the municipality of Diamantina, state of Minas Gerais, Brazil. Saadi (1995) has described the site as a hydromorphic depression containing peatlands overlying riverine or colluvial sands and gravel, surrounded by quartzitic rock outcrops associated with hematite phylites. According to Horak-Terra et al., the genesis of Chapada peatland started about 43,000 years ago (Horák-Terra et al. 2014).

Soil sampling and sample characterization

Soil samples, collected at a single point in each peatland, were taken from the surface down to a depth of 40 cm. Surface and subsurface samples were taken from fabric organic (0–10 cm deep) and sapric organic horizons (10–18 cm and 18–40 cm deep). The sampling was carried out using a shovel 15-cm wide and 50-cm long.

For the activity determinations, soil samples were dried, ground and sieved (2 mm mesh). Next, they were properly placed into a 2-L Marinelli-type container, sealed and kept at least for 3 weeks before measurements to ensure secular equilibrium of ^{238}U and ^{232}Th chains. The gamma-ray spectra were taken using a GMX10 gamma EG&G Ortec detector inside an EG&G Ortec low-background chamber and 8192 multichannel PHA analyzer. ^{60}Co , ^{133}Ba , ^{137}Cs and ^{152}Eu sources were used to perform the energy calibration. The detector efficiency calibration curve was acquired using an admixture of known amounts of naturally occurring ^{176}Lu and ^{138}La dispersed in a peatland sample (Perillo Isaac et al. 1997). The peaks in all the spectra were corrected after the determination of the laboratory background. The activity of ^{226}Ra was calculated by the activities of the photo-peaks emitted by ^{214}Pb and ^{214}Bi . The ^{232}Th chain activity was determined from the activity of ^{208}Tl , ^{212}Bi , ^{212}Pb and ^{228}Ac . The ^{40}K and ^{137}Cs activities were determined using the well-known 1460.75 keV and 661.62 keV lines, respectively.

Room temperature Mössbauer spectra in transmission geometry were taken in a 512-channel spectrometer with a $^{57}\text{CoRh}$ source. Calibration was performed with a 12- μm -thick $\alpha\text{-Fe}$ foil to which isomer shift values are referred. The Mössbauer spectra were numerically analyzed using a computer program that takes into account hyperfine magnetic fields and quadrupole splitting distributions (Lagarec and Rancourt 1998).

Results and discussion

Radioactivity

Figure 1 shows the activity profiles of natural radionuclides (^{232}Th , ^{226}Ra and ^{40}K). ^{232}Th is the radionuclide with the lowest activity values in all the profiles. Resulting values are lower than the mean worldwide value of 45 Bq/kg reported by UNSCEAR (2008). In Pinheiro (4–40 Bq/kg) and Chapada (26–44 Bq/kg) peats, the activity tends to increase with soil depth. Instead, in the Pau-de-Fruta (15–20 Bq/kg), it remains virtually constant. According to (Gaca et al. 2006), thorium in peatland is principally originated from mineral additions to the peat and, therefore, the ^{232}Th variation in Pinheiro and Chapada indicates different mineral composition across the profile. In these peatlands, silt and

clay percentages are negligible (only sand and organic matter content are noticeable) and therefore, X-ray diffraction measurements cannot detect or quantify the existing clay minerals. Since Fe is part of the structure of clay minerals, the ^{57}Fe hyperfine parameters revealed by Mössbauer spectra might provide some information about the distribution of clay minerals, as discussed below. Variations of the Fe environments phase abundance are indeed observed, especially for the Chapada profile and in the topmost layer of Pinheiro profiles, indicating possible variations on the mineral content of the profiles.

^{226}Ra activity in Pinheiro Peatland (82–765 Bq/kg) is considerably higher than the activity values found in Chapada (10–53 Bq/kg) and Pau-de-Fruta (17–119 Bq/kg). The significant differences of the ^{226}Ra activity values may indicate differences in the parent material or of pedogenetic processes for each of these peatlands. The different ^{226}Ra distributions can be associated with different hydric regimes, as Ra tends to form soluble chemical complexes. ^{226}Ra activity values resulted in some cases lower than the average value, 32 Bq/kg (UNSCEAR 2008), and higher in others.

All ^{40}K determined activity values resulted lower than the worldwide mean value, 412 Bq/kg (UNSCEAR 2008). The relatively lower activity for ^{40}K is in agreement with the K values reported by da Rocha Campos for the Pau-de-Fruta peatland, who attributed this fact to its low capacity to retain monovalent ions and the low content of K in the basement rock (da Rocha Campos et al. 2009). In Pinheiro the ^{40}K activity decreases down from 40 to 20 Bq/kg at 20 cm and then increases to values around those observed for the topsoil. ^{40}K activity distributions in Pau-de-Fruta (22–104 Bq/kg) and Chapada (17–30 Bq/kg) tend to decrease with depth, contrary to mineral soils (Montes et al. 2012).

In Sweden, Poland and Poland–Slovakia peatlands (Fiałkiewicz-Kozieł et al. 2014; Gaca et al. 2006; Mihalík et al. 2014), a decrease of ^{40}K activity values with depth was also observed. This behavior was interpreted as being due to the occurrence of living *Sphagnum* sp. that uptakes K as nutrient, inducing a relatively lower concentration of K in deeper layers of this soil (Mihalík et al. 2014). In SdEM, there is no living sphagnum, indicating that the living species in SdEM, such as *Lagenocarpus rigidus* Nees, *Rhynchospora speciosa* (Kuntze) Boeckeler and others (Silva et al. 2019) can also take K and concentrate it at the surface layer. Radioactivity analysis on these species could be carried out to confirm the ^{40}K incorporation. Besides, the ^{40}K activity of Pau-de-Fruta in the topsoil is notably higher than the values determined in the other two peatlands, achieving similar activity values to the other ones at soil layers deeper than 20 cm.

The Pearson correlation coefficients between activities of the three radionuclides have been determined (0.05 confidence level). Only the correlation between Th and Ra

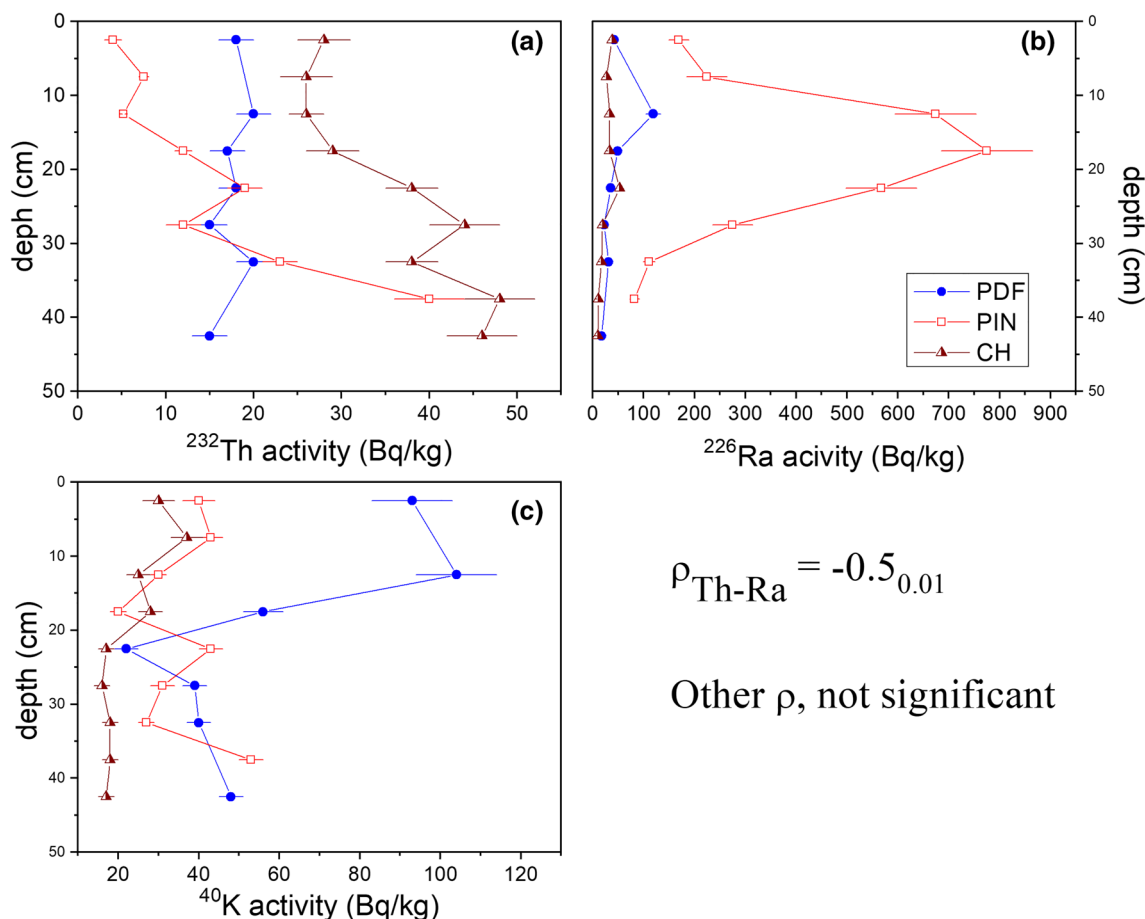


Fig. 1 Activity profiles of natural radionuclides: **a** ²³²Th, **b** ²²⁶Ra and **c** ⁴⁰K. The correlation coefficients ρ between activity values are also included

activities is significant with a Pearson correlation coefficient (ρ) of -0.5 . This is a singular result, as significant and positive correlations have been reported for activity values in other soils (Montes et al. 2012, 2016a, b; Navas et al. 2002). This result reflects the different behavior of radionuclides in peatlands compared to mineral soils.

The dose rate D at 1 m above the ground was evaluated (Table 1) taking into account the activity values and the corresponding conversion factors (UNSCEAR 2000, 2008):

$$D(\text{nGy/h}) = 0.462 A_{238\text{U}} + 0.604 A_{232\text{Th}} + 0.0417 A_{40\text{K}}$$

The highest dose rate is found at Pinheiro, because of the relatively high ²²⁶Ra activity, which is also the main

contributor to the dose rate in Pau-de-Fruta despite its intermediate value. In the Chapada peatland, which has the lowest D value, ²²⁶Ra and ²³²Th contribute to D in similar proportions. For the three peatlands, ⁴⁰K is the radionuclide with the lowest contribution to the dose rate. The dose rate values of Pau-de-Fruta and Chapada are lower than the values reported for Rio Grande do Norte and Sertão, Brazil, and lower than the worldwide value, 58 nGy/h (UNSCEAR 2000).

Figure 2 shows the ¹³⁷Cs activity profiles, which has been fitted to the convection–dispersion equation. The inventories in PDF and CH are similar while in PIN are relatively low. This difference can be associated with the peatland

Table 1 Dose rate values for the considered Peatlands

Peatland	D (nGy/h)	Locality	D (nGy/h)	References
PDF	53 ± 5	Rio Grande do Norte	72	Malanca et al. (1996)
CH	33 ± 3	Sertão	148	Malanca and Gaidolfi (1996)
PIN	96 ± 5	Worldwide mean value	58	UNSCEAR (2000, 2008)

Reference sites in Brazil and the worldwide mean value are also included

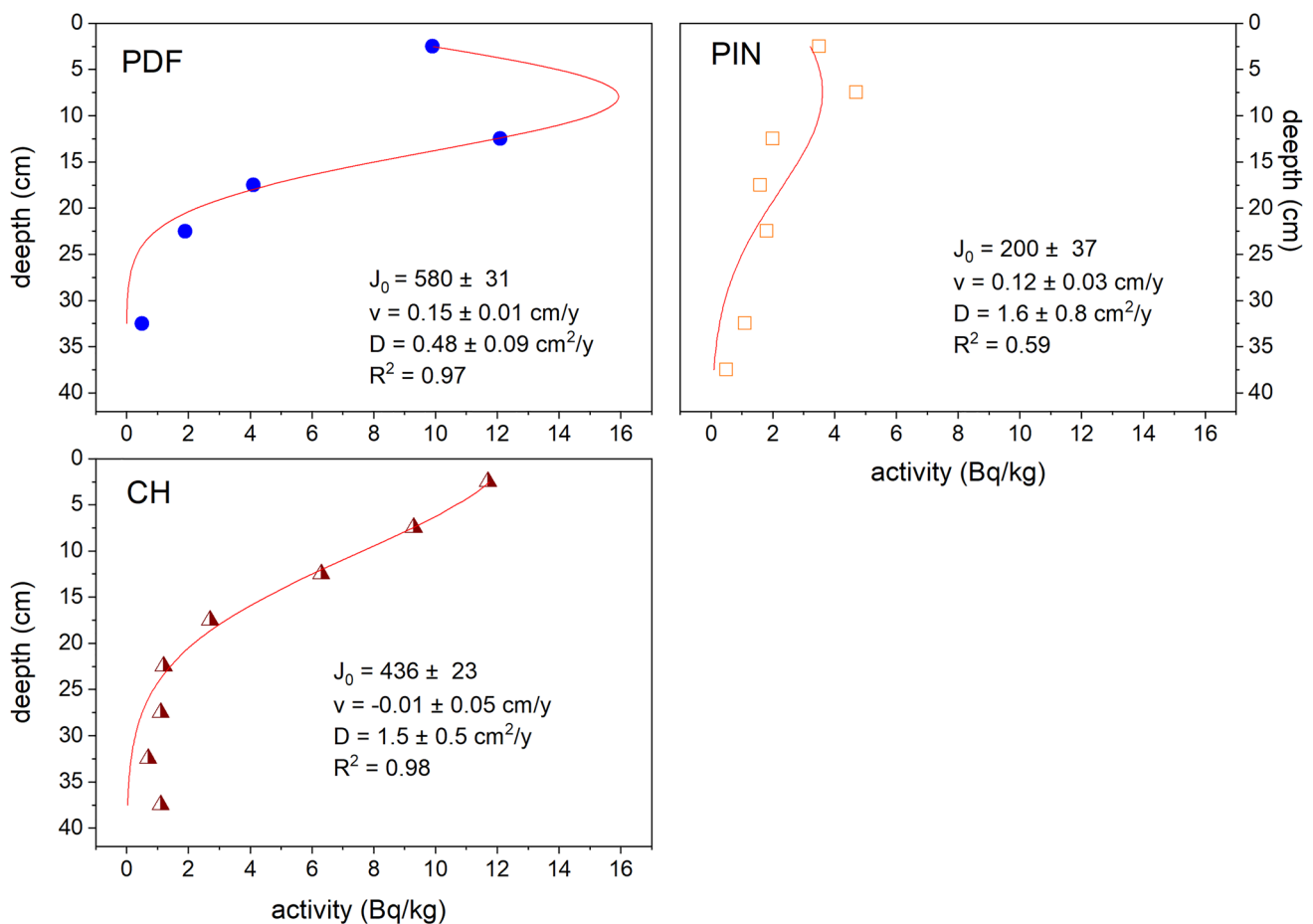


Fig. 2 ¹³⁷Cs distribution through the peatland profiles. The fit by the convection–dispersion model is also presented

altitudes. CH and PDF peatlands have similar altitudes (around 1340 m above sea level), while PIN is at a lower site (1247 m) and therefore, a lower ¹³⁷Cs deposit should be expected.

The ¹³⁷Cs activity distributions are also quite different. In PDF and CH, the ¹³⁷Cs profiles can be successfully fitted to the convection–dispersion equation (R^2 around 0.97). But R^2 of 0.59 is obtained in PIN profile, indicating that other phenomena might be affecting the ¹³⁷Cs distribution. At PIN and PDF, the maximum ¹³⁷Cs activity is observed at around 10-cm depth; while in CH peatland, it occurs at the soil surface.

The absence of clay minerals like illite do not allow associating this last behavior to irreversible ¹³⁷Cs retention with the consequent inhibition of Cs migration, as reported for mineral soils (Montes et al. 2013). Probably, this peatland exhibits extremely poor water drainage, which is also in agreement with the uniformity of the ²²⁶Ra activity profile. PDF and PIN convection velocities resulted similar to those found in other South American soils (Montes et al. 2013;

Schuller et al. 1997, 2004). The diffusion process seems to be more noteworthy in CH ($D=1.5$ cm²/years) and PIN ($D=1.6$ cm²/years) than in PDF ($D=0.48$ cm²/years).

In these soils, the relatively slow ¹³⁷Cs migration through the soil profile can be associated not with the clay minerals presence but with the higher ¹³⁷Cs bioavailability due to metal–ligand complexation with organic matter, which increases uptake by plants, and thus reduces the amount of Cs that could be downward transported. This process is also favored by the relatively low potassium concentration (Coughtrey et al. 1989; Kubo et al. 2017) in peatlands, since it is also absorbed by plants.

Fe species and concentrations

Figures 3, 4 and 5 show the room temperature Mössbauer spectra of PDF, PIN and CH peatlands, respectively. Tables 2 and 3 display the hyperfine parameters with their relative spectral area (RSA). Unlike the previous work on PIN that showed a Fe²⁺ signal (Paesano et al. 2014), all Mössbauer

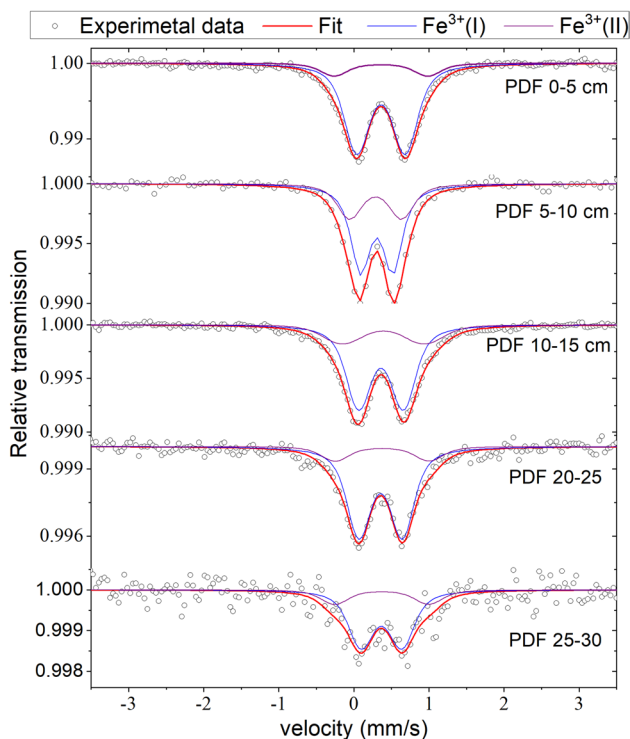


Fig. 3 Mössbauer spectra of Pau du Fruta samples. Blue and violet lines correspond to Fe^{3+} sites. The red lines represent the resultant spectra fit

spectra were fitted only considering two paramagnetic sites corresponding to Fe^{3+} environments, except for CH 0–5 cm and CH 5–10 cm samples that also reveal the hematite presence with relative spectra areas around 5% (Table 3).

Tables 2 and 3 also show the total Fe concentration, in mg/kg, determined after Montes et al. (2016). Chapada peatland is the soil with the highest Fe content, with a maximum Fe concentration of 122 mg/kg at the surface. In Pinheiro and Pau-de-Fruta, the maximum Fe concentration also is observed at the surface, with values of 37 mg/kg and 8 mg/kg, respectively. Fe concentration decreases with depth for the three peatlands, but the maximum depth at which the Mössbauer signal is noticeable above the statistical noise differs for each one: 25–30 cm for CH, 15–20 cm for PIN and 10–15 cm for PDF. There are also variations on Fe concentrations and their distributions, probably revealing differences in the concentration of soil minerals.

The relative spectral area of each Fe phase has been converted to the percentage of each Fe phase pondering the RSA by the total Fe soil concentration. The results are shown in Fig. 6. Fe^{3+} (I) mostly exhibits higher concentration than Fe^{3+} (II) and both Fe phases decrease with soil depth.

Fe phases are generally related to mineral presence. The Pearson correlation coefficients between the Fe phase concentration and ^{232}Th , ^{226}Ra and ^{40}K activity values have been determined having this in mind. No significant correlations

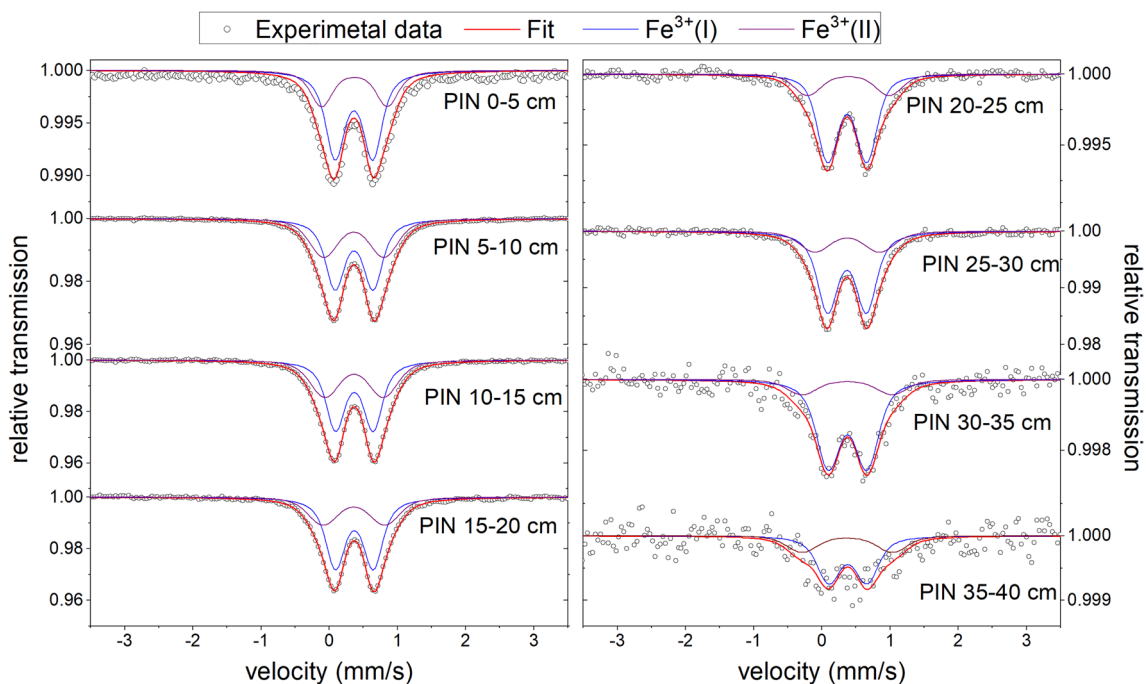


Fig. 4 Mössbauer spectra of Pinheiro samples. Blue and violet lines correspond to Fe^{3+} sites. The red lines represent the resultant spectra fit

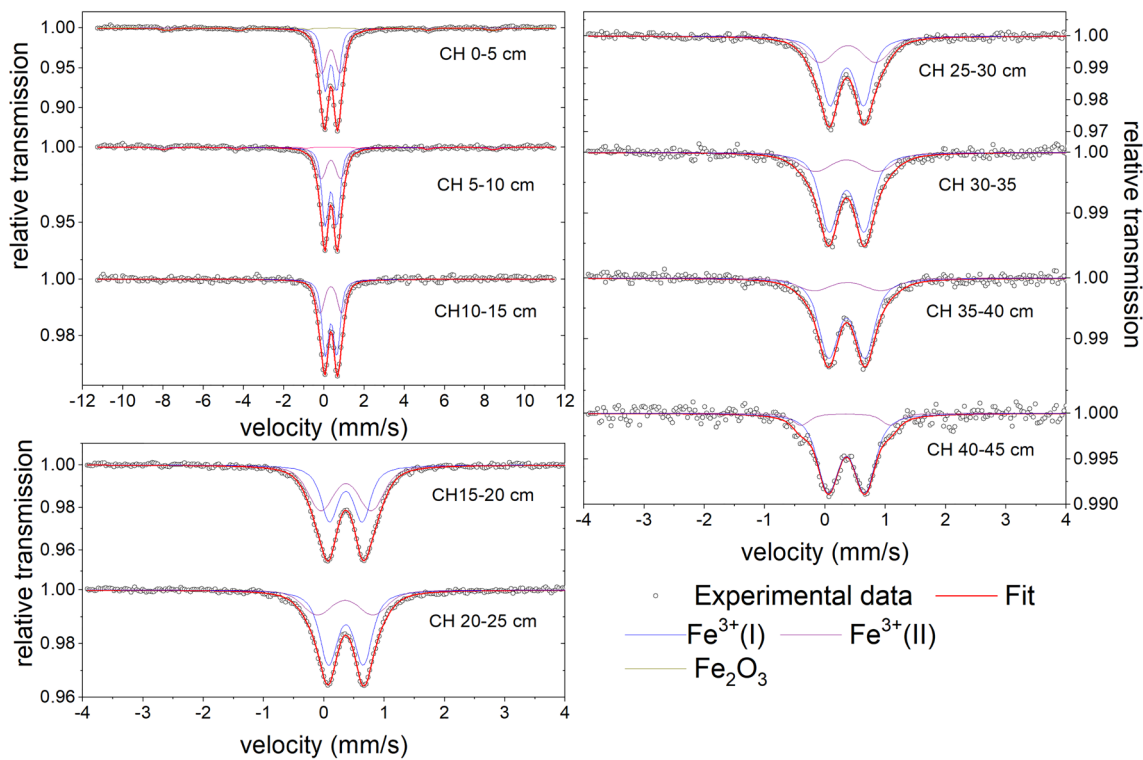


Fig. 5 Mössbauer spectra of Chapada peatland. Blue and violet lines correspond to Fe³⁺ sites, while brown line represents hematite environment. The red lines represent the resultant spectra fit

Table 2 Hyperfine parameters (isomer shift δ , quadrupole splitting Δ and relative spectral area RSA) for the Fe environments revealed by Mössbauer spectra of PDF and PIN samples

Sample	Fe ³⁺ (I)			Fe ³⁺ (II)			Fe (mg/g)
	δ (mm/s)	Δ (mm/s)	RSA (%)	δ (mm/s)	Δ (mm/s)	RSA (%)	
Pau-de-Fruta							
PDF0–5	0.36	0.64	85 ± 6	0.36	1.21	15 ± 3	8 ± 1
PDF5–10	0.35	0.51	64 ± 7	0.36	1.00	36 ± 4	3 ± 2
PDF10–15	0.36	0.60	75 ± 6	0.39	1.09	25 ± 2	1 ± 1
PDF 20–25	0.36	0.57	81 ± 5	0.38	1.16	19 ± 3	<
PDF25–30	0.37	0.53	76 ± 8	0.38	1.20	24 ± 4	<
Pinheiro							
PIN 0–5	0.36	0.55	66 ± 6	0.37	0.96	34 ± 2	37 ± 4
PIN 5–10	0.36	0.56	57 ± 4	0.36	0.88	43 ± 4	12 ± 2
PIN 10–15	0.37	0.55	58 ± 4	0.36	0.86	42 ± 4	16 ± 2
PIN15–20	0.37	0.56	65 ± 5	0.36	0.91	35 ± 4	14 ± 2
PIN20–25	0.37	0.56	69 ± 8	0.38	1.03	31 ± 2	<
PIN25–30	0.37	0.57	74 ± 7	0.37	0.95	26 ± 3	<
PIN 30–35	0.38	0.58	87 ± 14	0.37	1.42	13 ± 2	<
PIN35–40	Signal too low to perform a reliable fit						<

could be established between the radionuclide activities and the Fe phases. These correlations exist in mineral soils (Montes et al. 2016), which again is evidence of the special characteristics of peatlands.

Unlike mineral soils, the lack of correlation between Fe phases and radionuclides might indicate that in peatlands they are not being governed by the presence of clay minerals.

Table 3 Isomer shift δ , quadrupole splitting Δ , quadrupole shift ϵ in mm/s, magnetic field (H) and relative spectral area (RSA in %) for the Fe environments in Chapada

Sample	Fe ³⁺ (I)			Fe ³⁺ (II)			Fe ₂ O ₃				Fe (mg/kg)
	δ	Δ	RSA	δ	Δ	RSA	δ	ϵ	H	RSA	
CH0–5	0.36	0.58	47 ± 5	0.35	0.92	47 ± 5	0.40	−0.097	50.4	6 ± 1	122 ± 17
CH5–10	0.36	0.58	61 ± 5	0.35	0.97	34 ± 4	0.32	−0.084	50.9	5 ± 1	48 ± 6
CH10–15	0.36	0.57	65 ± 6	0.35	1.87	35 ± 4	–	–	–	–	16 ± 2
CH15–20	0.36	0.54	67 ± 5	0.37	0.84	33 ± 6	–	–	–	–	21 ± 3
CH20–25	0.37	0.58	67 ± 5	0.35	0.93	33 ± 4	–	–	–	–	14 ± 2
CH25–30	0.36	0.56	64 ± 5	0.38	0.93	36 ± 5	–	–	–	–	9 ± 2
CH30–35	0.36	0.58	72 ± 6	0.36	1.04	28 ± 3	–	–	–	–	<
CH35–40	0.36	0.60	80 ± 7	0.37	1.11	20 ± 2	–	–	–	–	<
CH40–45	0.36	0.61	88 ± 8	0.34	1.48	12 ± 2	–	–	–	–	<

Total Fe concentration is also included

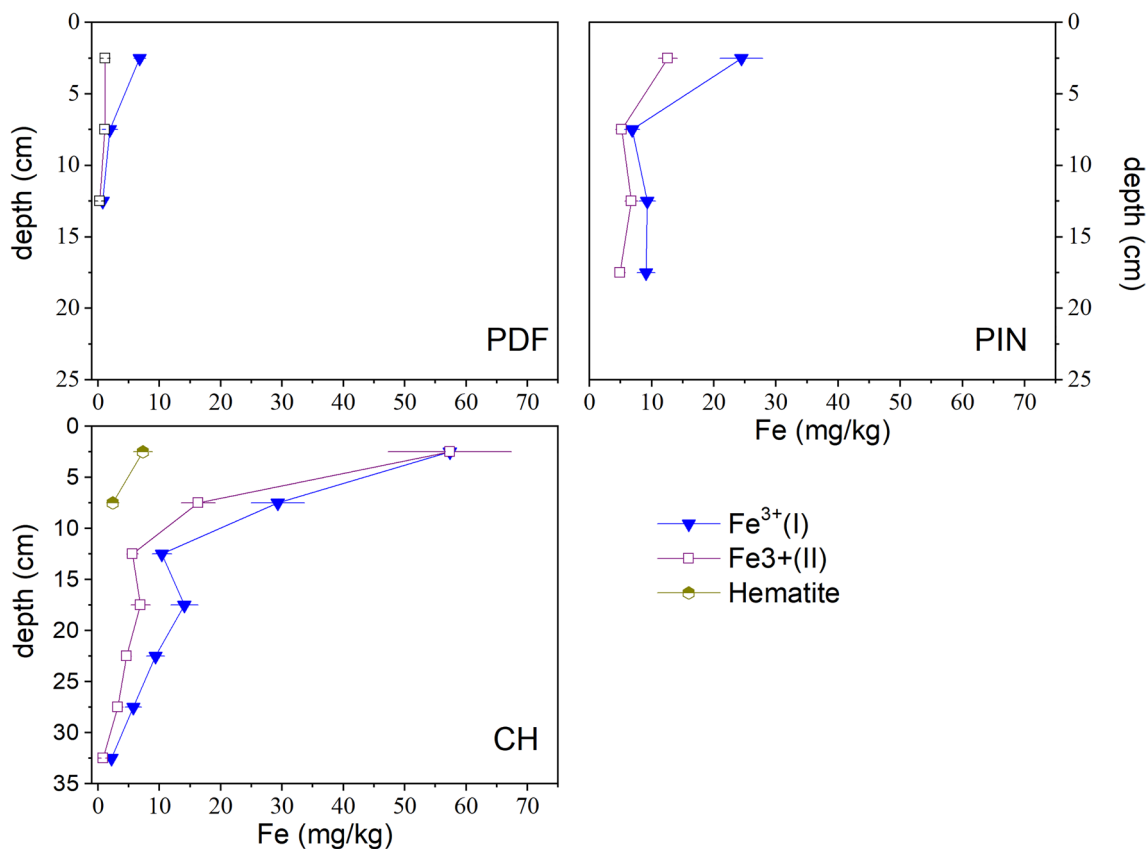


Fig. 6 Fe phase concentration profiles

Conclusions

Activity profiles of ²³²Th, ²²⁶Ra, ⁴⁰K and ¹³⁷Cs have been determined in three peatlands of *Serra do Espinhaço Meridional*, Brazil. Differences in natural and anthropogenic activity profiles reveal differences in altitude, parental material and hydric regime and expose the environment features, which are peculiarly different from mineral soils. The Pearson correlation coefficients for natural radionuclide activities

result significant only for ²³²Th and ²²⁶Ra. The Fe phases and Fe environment concentration profiles show that Fe is only in a Fe³⁺ oxidation state. Chapada is the peatland with the highest Fe amount, followed by Pinheiro. The total Fe concentration and the concentration of each Fe phase decrease with depth, but down to different depths. The correlations between the natural radionuclide activities and the Fe phase proportions yield in all cases statistically not significant results, thus revealing the extremely complex systems that peatlands are.

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