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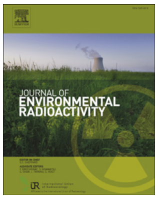

Wet deposition and soil content of Beryllium – 7 in a micro-watershed of Minas Gerais (Brazil)

Alexander D. Esquivel L [a, b], Rubens M. Moreira [b], Roberto Pellacani G. Monteiro [c], Anômora A. Rochido Dos Santos [d], Jimena Juri Ayub [e, f, *], Diego L. Valladares [e, g]

[a] *Universidad Tecnológica de Panamá, Centro de Investigaciones Hidráulicas e Hidrotécnicas - (CIHH), Vía Domingo Díaz al lado de Pazco, S.A., 0819-07289, El Dorado, Panama*

[b] *Setor de Meio Ambiente - (SEMAM), Centro de Desenvolvimento da Tecnologia Nuclear - (CDTN-CNEN), Av. Presidente Antônio Carlos 6627, Campus da UFMG, 31270-901 Belo Horizonte, MG, Brazil*

[c] *Serviço de Técnicas Analíticas - (SERTA), Centro de Desenvolvimento da Tecnologia Nuclear - (CDTN-CNEN), Av. Presidente Antônio Carlos 6627, Campus da UFMG, 31270-901 Belo Horizonte, MG, Brazil*

[d] *Pontifícia Universidade Católica de Minas Gerais - (PUC-Minas), Av. Dom José Gaspar, 500 - Coração Eucarístico, Belo Horizonte, MG CEP 30535-901, Brazil*

[e] *Grupo de Estudios Ambientales - (GEA), Instituto de Matemática Aplicada San Luis - (IMASL), Universidad Nacional de San Luis – CONICET, Ejercito de los Andes 950, D5700HHW San Luis, Argentina*

[f] *Departamento de Bioquímica y Ciencias Biológicas, Facultad de Química, Bioquímica y Farmacia, Universidad Nacional de San Luis, Ejercito de los Andes 950, D5700HHW San Luis, Argentina*

[g] *Departamento de Física, Facultad de Ciencias Físico Matemáticas y Naturales, Universidad Nacional de San Luis, Ejercito de los Andes 950, D5700HHW San Luis, Argentina*

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ABSTRACT

Beryllium-7 (^{7}Be) is a natural radionuclide of cosmogenic origin, normally used as a tracer for several environmental processes; such as soil redistribution, sediment source discrimination, atmospheric mass transport, and trace metal scavenging from the atmosphere. In this research the content of ^{7}Be in soil, its seasonal variation throughout the year and its relationship with the rainfall regime in the Mato Frio creek micro-watershed was investigated, to assess its potential use in estimating soil erosion. The ^{7}Be content in soil shows a marked variation throughout the year. Minimum ^{7}Be values were observed in the dry season (from April to September) and were between 7 and 14 times higher in the rainy season (from October to March). The seasonal oscillations in ^{7}Be soil content could be explained by the asymmetric rainfall regime. A highly linear relationship between rainfall amount and ^{7}Be deposition was observed in rain water. A good agreement between ^{7}Be soil content and ^{7}Be atmospheric deposition was noticed, mainly in wet months. ^{7}Be penetration in soil reaches a 5 cm depth, this could be explained by the soil type in the region. The soils are Acrisol type, characterized by low pH values and clay illuviation in deeper layers of the soil. In some regions of Brazil special attention should be paid if this radionuclide will be used as soil erosion tracer, taking into account the soil origin and its particular properties.

1. Introduction

Beryllium-7 (^{7}Be) is a natural radionuclide of cosmogenic origin. It is produced in the atmosphere by spallation when cosmic rays hit nitrogen and oxygen atoms (Lal et al., 1958). Once formed it diffuses through the atmosphere and is electrostatically adsorbed in atmospheric aerosol particles. It reaches the soil surface via two mechanisms: wet and dry deposition. It is assumed that the wet deposition is the main path leading to the ^{7}Be input (90%) into the soil, dry deposition being negligible (Salisbury and Cartwright, 2005; Ioannidou et al., 2005; Wallbrink and Murray, 1994).

^{7}Be is an important environmental radionuclide, its relatively short half-life of ^{7}Be (53 days), along with its continuous and assessable production rate makes it a potentially powerful tool for surveying environmental processes; such as soil redistribution,

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* Corresponding author. Grupo de Estudios Ambientales - (GEA), Instituto de Matemática Aplicada San Luis - (IMASL), Universidad Nacional de San Luis – CONICET, Ejercito de los Andes 950, D5700HHW San Luis, Argentina.

E-mail address: jjuri@unsl.edu.ar (J. Juri Ayub).

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sediment source discrimination, atmospheric mass transport, and trace metal scavenging from the atmosphere (Kaste et al., 2002; Yoshimori, 2005; Steinmann et al., 1999; Daish et al., 2005; Matissoff et al., 2002; Walling et al., 1999; Blake et al., 1999; Schuller et al., 2006; Sepulveda et al., 2008). Kaste et al. (2011) pointed out that, in order to evaluate the potential of ^{7}Be as a tracer, it is necessary to know its seasonal and spatial depositional variability as well as quantify the relationship between precipitation and surface inventories.

The Mato Frio creek is one of the main tributaries forming the Serra Azul River watershed, near Belo Horizonte, one of the main cities in the central Brazilian plateau. The importance of this watershed is related with a large water reservoir constructed at its lower course, which is the third largest drinking water supply system (2.5 m^3/s) to the metropolitan region of Belo Horizonte (about 4 million inhabitants). Besides supplying water, two other conflicting activities are practiced within the watershed area: vegetable agriculture in the central area and intensive iron ore mining at its headwaters. As regards the Mato Frio creek micro-watershed, it covers an area of about 8 km^2 and has a hilly landscape with steep slopes. Therefore the main farming activity is related with raising livestock, hence both the heavily inclined pasture lands and the constant trampling by cattle promote soil erosion. The sediment load thus added to the watercourses, compounded with those resulting from the ore mining activities, will result in increased sedimentation at the water reservoir downstream with severe volume losses and water quality impairment.

The aim of the research is to investigate the content of ^{7}Be in soil, its seasonal variation throughout the year and its relationship with the rainfall regime in the Mato Frio creek micro-watershed, to assess its potential use to estimate soil erosion.

2. Materials and methods

2.1. Study area

The study area is located within the Mato Frio creek micro-watershed (20 °04′00″ S, 44 °28′00″ W; 20 °08′00″ S, 44 °31′00″ W), about 50 km to the southwest of Belo Horizonte, in the state of Minas Gerais, Brazil (Maia et al., 2006). The micro-watershed is located 908 m asl and has slopes ranging from 6.5% to 15.5%. The climate in the region is characterized by a warm and rainy season from November to March and a cool and dry season from June to August (Neves et al., 2004). Average monthly temperatures vary between 16 °C and 23 °C. Taking into account temperature and rainfall records, the climate can be characterized as high altitude tropical according to the Köppen classification (Soares, 2010).

The soils in the region are Acrisols and Ferrasols, the most important soils in Brazil; covering 40% and 20% of the country, respectively. The Acrisol type is characterized by clay illuviation. In this soil type the clay particles are accumulated in the Bt horizon, causing sand enrichment in the upper layers of soil. This soil is rather acid ($pH = 4.5$), well-drained and has low fertility. The Ferrasols are a deep soil characterized by the sandy loam texture of the B horizon. This soil type is also acid (pH 4.5–5.0), well-drained with high porosity and has moderate to high fertility (Soares, 2010).

Two parcels, Parcel 1 (P1) and Parcel 2 (P2), on different soil types in the Mato Frio River micro-watershed were selected for this study. Parcel P1 was an Acrisol type and P2 a Ferrasol type, with the intention of exploring differences in the ^{7}Be soil content.

2.2. Rainfall

A rainfall database at the micro-watershed was obtained from the nearest available rain gauge station, located 1 km away from the study sites (www.snirh.gov.br/hidroweb/FAZENDA LARANJEIRAS-JUSANTE, Rainfall Code Station: 2044041, Responsible: ANA, Operating Agency: CPRM, State: Minas Gerais, County: Itaúna, Basin: Rio São Francisco, Micro-watershed: Rios São Francisco, Paraopeba E). The database covers daily rainfall over a period of 39 years, from 1977 to 2015. Additionally, monthly rainfall samples were collected throughout the wet season from October 2015 to May 2016. A standard gauge was used and the ^{7}Be activity concentration was measured by gamma spectrometry.

2.3. Soil

During the period from May 2014 to May 2015 monthly soil samples were taken at 1 cm depth, at both the P1 and P2 parcels. In each sampling time and for each parcel, two soil samples were taken. All soil samples were collected using a scraper plate with a 50 cm × 20 cm collection surface. The soil samples were dried at room temperature for 48 h, sieved through a 2 mm mesh and placed in a Marinelli beaker for gamma spectroscopy analysis.

In October 2015 at parcel P1, the soil profile was sampled at 6 cm depth, cutting the profile in layers of 1 cm thick, with the aim to explore the total ^{7}Be soil content. Following this in the same parcel, from November 2015 to May 2016, monthly soil samples were collected to 5 cm depths, cutting the profile into two layers: 0–2.5 cm depth and 2.5–5.0 cm depth. In each month only one soil profile were collected. The soil samples were collected and processed using the same equipment and procedures as before.

2.4. Gamma spectrometry analysis

The soil and rain water samples were submitted to gamma spectrometry analysis and the ^{7}Be emission pulses were measured at the 477.6 keV energy peak using a Hyper-pure Germanium detector (GX5019, CANBERRA) at the Nuclear Spectrometry Laboratory of the Center for Development of Nuclear Technology (CDTN). This spectrometer has a 1.9 keV resolution and a 50% relative efficiency at the 1.33 MeV gamma energy of ^{60}Co. The samples, weighting around 600 g, were placed in 700 mL Marinelli beaker, and the total counting time varied between 86,000 s and 180,000 s.

The efficiency curve of the Hyper-pure Germanium detector was obtained using the Genie 2000 CAMBERRA Monte Carlo mathematical model software. Compounding it with the detector efficiency curve, the counting efficiency (ε) of the ^{7}Be gamma ray energy in the soil samples was $\varepsilon = 3.3\%$. This methodology had to be used given that no soil reference standard was available at the laboratory. The same procedure has been used in other studies involving gamma spectrometry analysis (Díaz and Vargas, 2008; ”Vidmar et al., 1994; García, 2012; Pinto et al., 2013).

Equation (1) has been used to calculate the ^{7}Be activity in the soil and rain water samples (A).

$$A = \frac{N}{\varepsilon m_a t I_\gamma} \tag{1}$$

where $\boldsymbol{N}$ is the net number of counts corresponding to the gamma radiation (γ) per counting time interval t (s), m_a is the mass of the soil sample (kg), I_γ is the absolute transition probability for the measured gamma ray, and ε is the counting efficiency. All the activity measurements were corrected for the ^{7}Be radioactive decay. The final results were expressed in terms of activity per unit mass of soil (Bq kg^{-1}) and of activity per unit of volume of rain (Bq L^{-1}).

It is necessary to check the interference level of ^{228}Ac in ^{7}Be activity measurements by gamma spectrometry in order to establish an analytical protocol for the ^{7}Be determination. Therefore, some soil samples were counted at different time intervals, of approximately one year following sampling, in order to secure the complete decay of ^{7}Be ($t_{1/2} = 53.3$d) and detect if ^{228}Ac from the ^{232}Th the series was present. Twelve soil samples were recounted about one year after collection and showed the absence of photopeak at 478.40 keV. It was therefore concluded that the interference of ^{228}Ac was negligible in analysed samples. For rain water samples, it was not necessary to perform this analysis, since they would not have the ^{232}Th decay series. Fig. 1 (a–b) shows the gamma spectra of the same sample recorded after a time interval of approximately one year.

A further analytical check for soil samples as to count at 477.59 keV after different storage times to check if the activity decay was consistent with the ^{7}Be half-life (Fig. 1 c–d). A half-life of 51.3d was measured consistent with the value of 53.3d found in the literature for ^{7}Be (http://ie.lbl.gov/education/isotopes.htm (2000)).

The lower limit of detection (LLD) was determined (Currie, 1968) at the 95% confidence level and counting time of 180,000 s was 0.12 Bq kg^{-1}. The limit of detection calculated by Kaste et al. (2014) was 0.45 Bq kg^{-1} for sample masses in the range of 125–175 g.

3. Results and discussion

The monthly rainfall over the last 39 year period is depicted in Fig. 2. The annual precipitation is in the range of 1500 mm, with a wet season (from November to March) and a dry season (from April to October). During the period of the study, 80% of the precipitation events occurred in the wet season.

During the period between May 2014 and May 2015 the activity concentration of ^{7}Be in the top 1 cm of soil ranged from 2.0 ± 0.1 Bq kg^{-1} to 34.0 ± 6.5 Bq kg^{-1} at P1 and from 3.1 Bq kg^{-1} to 22.0 ± 0.2 Bq kg^{-1} at P2 (Table 1). The ^{7}Be contents at 1 cm depth expressed per unit area, varied from 21.1 ± 2.4 Bq m^{-2} to 295.9 ± 63.1 Bq m^{-2} at parcel P1 and from 27.7 ± 5.2 Bq m^{-2} to 162.4 ± 57.9 Bq m^{-2} at parcel P2 (Table 1). At both parcels, the ^{7}Be content in the upper soil showed a marked variation throughout the period. The minimum ^{7}Be contents were observed in the dry season (from April to September) and were 14 and 7 times higher in the rainy season (from October to March), at P1 and P2, respectively. Variations in the ^{7}Be content in soils throughout the year

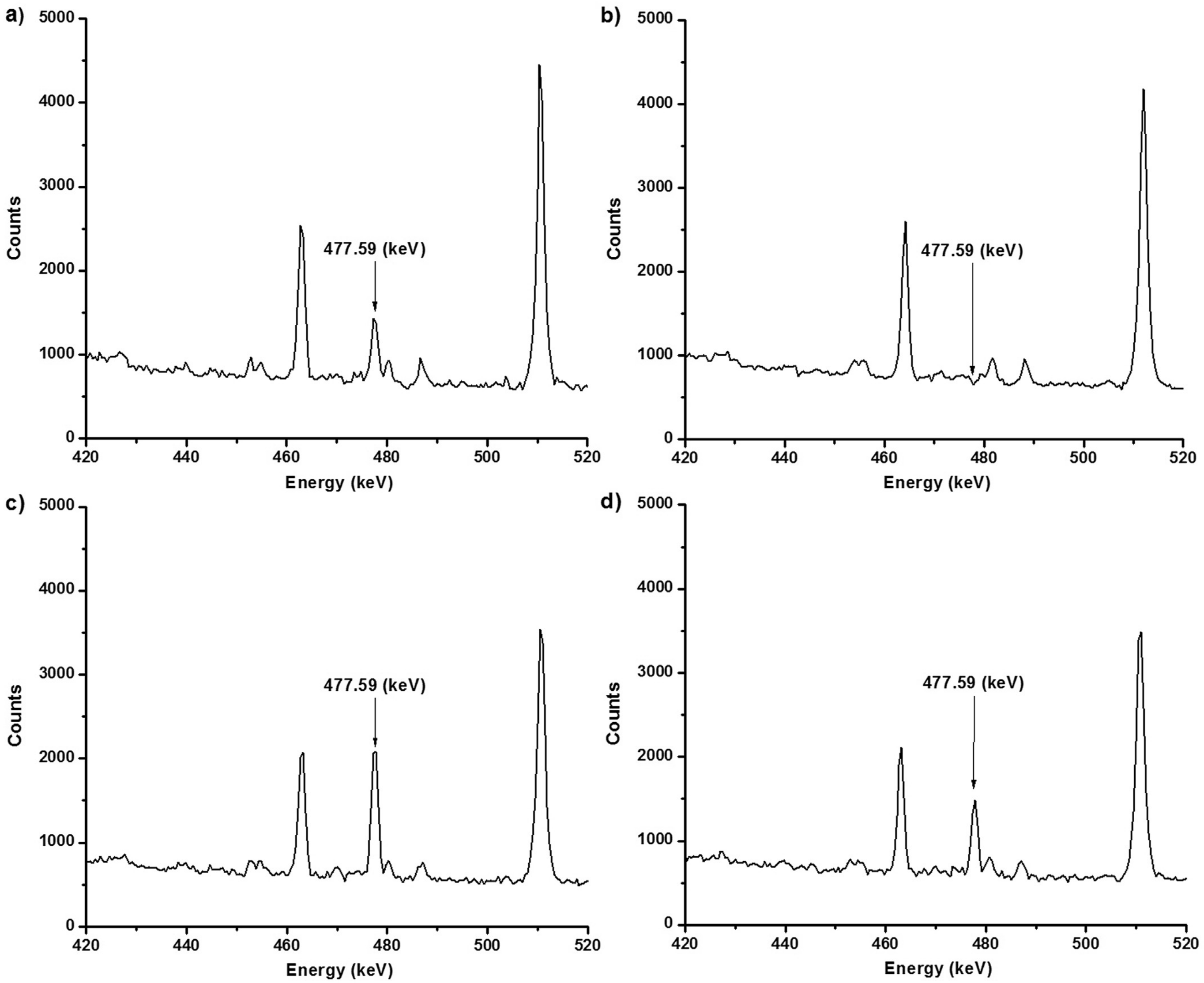

Fig. 1. Record spectra of ^{228}Ac interference evaluation and of ^{7}Be half-life measurement (a) Soil sample gamma spectrum registred in march of 2015 (counting time of 180,000 s), (b) Soil sample gamma spectrum registred in september of 2016 (counting time of 347,000 s), (c) Gamma spectrum of soil sample recorded on 04/13/2015 and (d) Gamma spectrum of soil sample after one half-life decay (recorded on 06/03/2015).

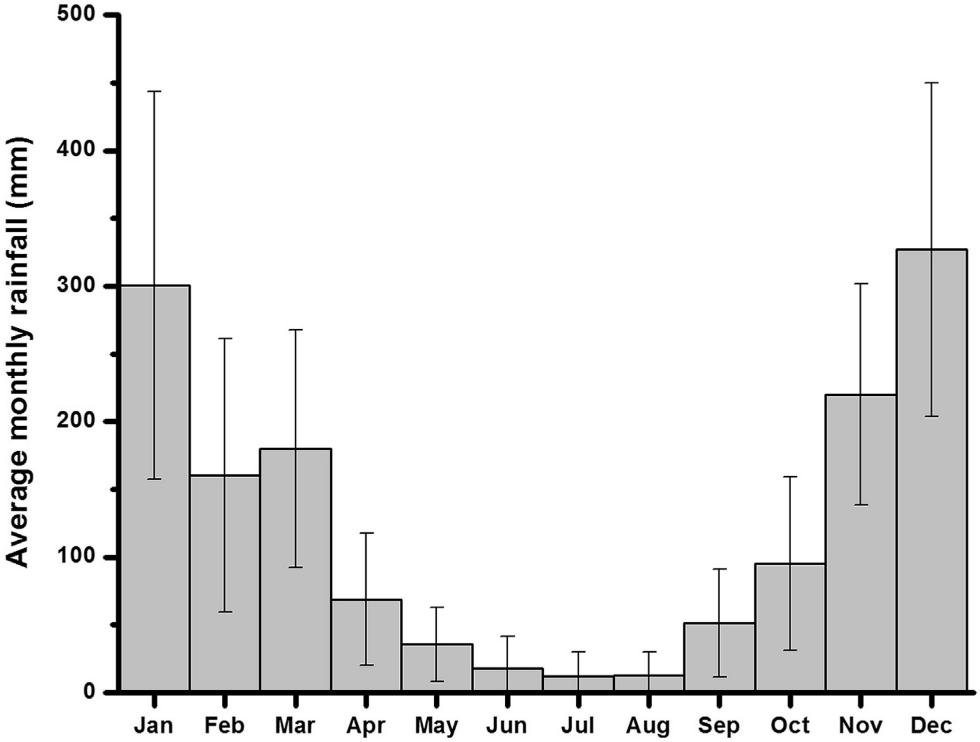

Fig. 2. Mean monthly accumulated rainfall amount in the studied area. Mean values were obtained averaging over the last thirty-nine year period. Bars indicate the standard deviation.

Table 1
^{7}Be activity concentration (C) and ^{7}Be content (A) in the first centimeter of soil, for each study parcel. The mean value of the 6 profiles and estándar deviation (SD) is reported.

		Parcel 1				Parcel 2			
		C (Bq kg^{-1})		*A* (Bq m^{-2})		*C* (Bq kg^{-1})		*A* (Bq m^{-2})	
		Mean	SD	*Mean*	SD	*Mean*	SD	*Mean*	SD
2014	May	3.2	1.1	24.7	8.2	**	**	**	**
	Jun	2.0	0.1	21.1	2.4	3.8	***	35.1	***
	Jul	*	*	*	*	*	*	*	*
	Aug	3.6	1.5	28.6	11.2	*	*	*	*
	Sep	4.2	1.1	32.6	10.3	4.4	***	40.1	***
	Oct	2.9	***	22.5	7.1	3.1	***	27.7	***
	Nov	34.0	6.5	295.9	63.1	22.0	0.2	126.3	4.6
	Dec	19.3	5.0	133.3	38.1	19.1	4.2	162.4	57.9
2015	Jan	5.5	0.4	50.5	0.8	5.4	0.4	47.8	3.6
	Feb	18.8	10.3	144.3	117.1	15.4	4.1	124.4	24.4
	Mar	21.1	5.9	266.0	119.1	14.7	4.3	150.7	47.2
	April	9.8	0.8	100.5	32.1	5.0	0.6	48.1	8.3
	May	8.2	2.9	70.0	8.2	4.2	0.5	35.5	2.7

* Below the detection limit.
** No sampling.
*** Only one profile sampled.

have been described at other regions in the world. Kaste et al. (2011) in California (USA) and Juri Ayub et al. (2009) in San Luis, Argentina found that the seasonal variation in the ^{7}Be content in the soil can be explained by the asymmetric pattern of the rainfall distribution throughout the year. The dry and wet seasons of the present study area are akin to both the California and San Luis regions.

During the wet season, from October 2015 to May 2016 monthly accumulated rainfall was sampled and ^{7}Be content measured. Fig. 3 shows the dependence of ^{7}Be deposition on the amount of rainfall. There was a strong linear relationship between these two parameters; with a slope of 1.37 ± 0.17 Bq L^{-1} ($r^2 = 0.93$; $p = 0.001$). Olsen et al. (1985) also reported linear relationships with $r^2 = 0.63$ and 0.54 at two regions in the USA, Caillet et al. (2001) reported $r^2 = 0.66$ ($p = 0.001$) at a site in Switzerland. Zhu and Olsen (2009) in the USA reported a positive correlation with $r^2 = 0.46$. Walling et al. (2009) in Southern Chile, found $r^2 = 0.82$. Similar results were found at an environment with a seasonal precipitation regime by Kaste et al. (2011) with $r^2 = 0.8$ and by Juri Ayub et al. (2012), with $r^2 = 0.82$.

If it is assumed that: a) ^{7}Be dry deposition is negligible (Salisbury and Cartwright, 2005; Ioannidou et al., 2005; Wallbrink and Murray, 1994), b) the value of ^{7}Be activity concentration in rainwater remains constant (Juri Ayub et al., 2012), and c) the only mechanism leading to ^{7}Be loss from the soil is radioactive decay, then the expected value of ^{7}Be content in the soil due to wet deposition could be estimated using both the value of the slope, 1.37 ± 0.17 Bq L-1, and the mean monthly precipitation (Fig. 2). Fig. 4 depicts the measured values of ^{7}Be content in soil (Table 1) and the predicted values of ^{7}Be content by wet deposition. Only a few cases indicate a perfect match between measured and estimated ^{7}Be content. In both parcels, most of the measured ^{7}Be inventory values in the soil (at 1 cm depth) were less than the estimate of ^{7}Be content from wet deposition. The few data with a close match corresponded to dry months.

This difference between these two values could be due to a deeper penetration of ^{7}Be in the soil profile. With the objective of evaluating this hypothesis, in October 2015, for P1 the soil profile was sampled at 6 cm depth, cutting the profile in layers of 1 cm thickness. It was found that ^{7}Be was detectable down to the 5 cm depth. The ^{7}Be content in soil shows the typical decreasing vertical distribution (Fig. 5). The estimated relaxation mass depth (h_0) is 43.4 kg m^{-2}. The magnitude of this parameter indicates a deeper

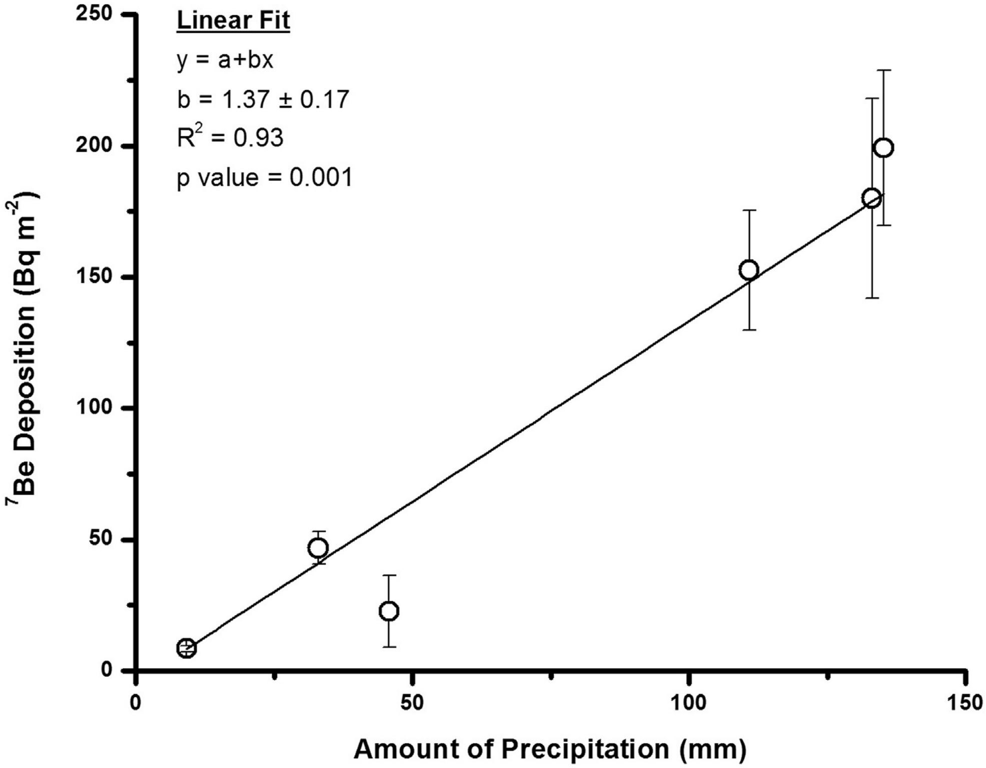

Fig. 3. ^{7}Be deposition versus rainfall amount. Linear fit was obtained with error as weight. The bar error was estimated from the statistical counting error and the error in the measurments of amount of precipitation.

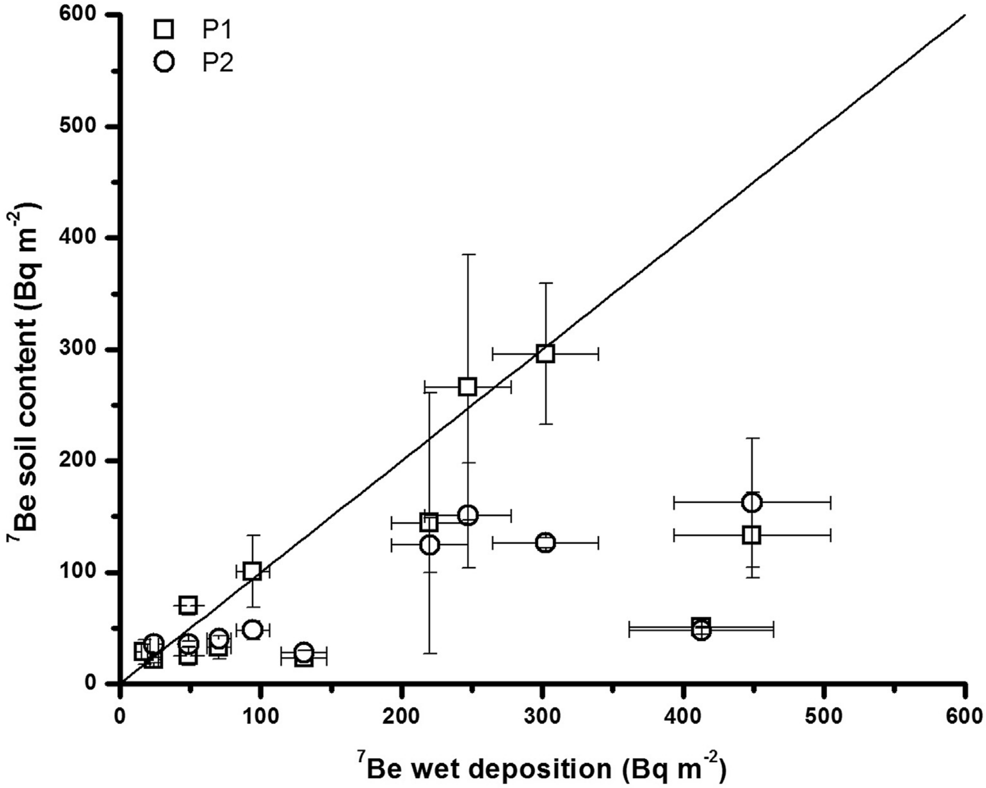

Fig. 4. Measured ^{7}Be soil content in soil vs ^{7}Be wet deposition (estimate in soil). The ^{7}Be wet deposition was estimated using the mean monthly annual rainfal (Fig. 5) and the estimated slope obtained in Fig. 6. The bar error in y axis was estimated taking into account de statistical counting error and the error in the soil sampling. The continuous line shows the hipothethical perfect match between measured and estimate ^{7}Be.

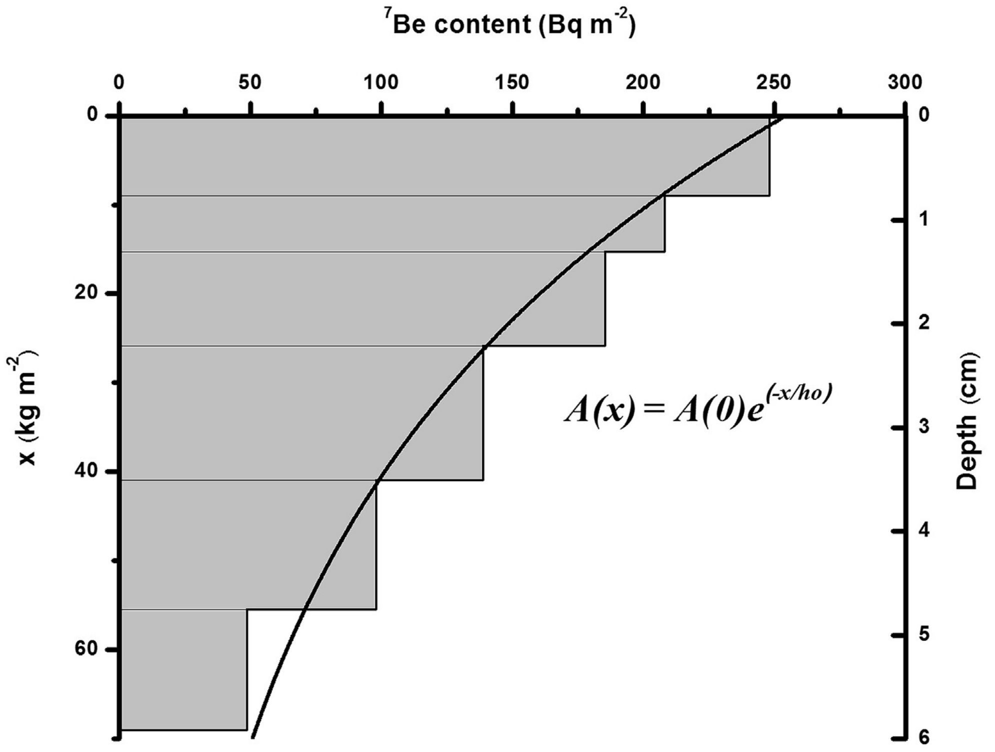

Fig. 5. Vertical soil distribution of ^{7}Be content in the soil profile for P1 in October 2015. The line correspond the fit to an exponential decreasing function.

penetration of the radionuclide into the soil; 63.2% of the radionuclide is retained in the soil layer between the surface and h_0 (Sepulveda et al., 2008).

Most of the literature describing the distribution of ^{7}Be in soil report maximum penetrations depths of about 2 cm of soil (Sepulveda et al., 2008; Lohaiza et al., 2014). The penetration depth observed in the present study is unusual, but has been observed by other authors who report penetration depths down to 8 or 10 cm in some soils (Kaste et al., 2002). These authors suggest that the drainage structure and the moisture status of the soil could be affecting the depth distribution of ^{7}Be. The soil at the P1 study site is of Acrisol type, characterized by low pH and clay illuviation. The higher sand content in the superficial layers of soil and its low pH could be the reasons for the deeper penetration of ^{7}Be at this site.

Taking into account the recorded rainfall events from the 2015/2016 biennium and the ^{7}Be activity concentration in rain, the expected annual cycle of ^{7}Be wet deposition on the soil could be quite accurately estimated (Fig. 6, upper part, lines). The bars in the lower part of this figure show the ^{7}Be input from the atmosphere as pulses which are related to the rain episodes. Based on the deeper penetration of ^{7}Be in parcel P1, for the period from November 2015 to May 2016 the soil profiles were sampled monthly to 5 cm depth (Fig. 6, circles). This figure reveals that: 1) each ^{7}Be pulse (each precipitation event) caused an increment of ^{7}Be deposition, 2) the ^{7}Be deposition exhibited oscillation cycles due to the asymmetric precipitation pattern, 3) during dry periods the expected ^{7}Be content in the soil decreased due to radioactive decay; 4) the measured ^{7}Be content were closer to the value expected from wet deposition and show the same annual cycle due to the asymmetric precipitation regime. These results confirm the deeper penetration (down to 5 cm) of ^{7}Be in these soils, 5) the seasonal changes in ^{7}Be content in the soil could be predicted from the atmospheric deposition by rainfalls, and 6) during the dry period the ^{7}Be content in the soil was lesser than the expected by atmospheric deposition. This last point suggests that the soils in the investigated site could be subject to an additional loss of ^{7}Be activity. During the dry season this hilly region is subjected to winds that may lead to soil erosion and explain the lesser value of ^{7}Be in soil.

4. Conclusions

The ^{7}Be content of soil and the corresponding input by precipitation events were monitored at a micro-watershed. The results indicated penetration of ^{7}Be reached the 5 cm depth. The observed vertical penetration is atypical for this radionuclide, but could be explained by the soil type at the site. It is characterized by low pH and clayey illuviation. ^{7}Be has been widely used to estimate soil erosion and sedimentation. An important precondition for this application is the knowledge of its vertical distribution. Our results show that in some regions and soil types the vertical ^{7}Be profile can reach deeper depths. In such cases the vertical distribution has to be carefully evaluated if ^{7}Be is to be used to estimate erosion or sedimentation taken into account the soil type and its properties (clay content, pH, etc).

The ^{7}Be content in soil had a marked seasonal variation throughout the year, explained by the precipitation pattern. The region have a marked rainy and dry seasons, at least the 80% of the precipitation occurs during the wet season. The oscillation of the ^{7}Be soil content had a similar pattern, which can be expected from the rainfall events. During the rainy season the measured and predicted values of ^{7}Be are closer, major differences were recorded in dry periods; which could be attributed to erosion process. During the dry season, winds are common in the region and loss of soil bearing adhered ^{7}Be is expect to occur.

The good agreement between the measured and expected values of ^{7}Be content in soil, mainly during the rainy season, confirms the general assumption that wet deposition is the main mechanism by which ^{7}Be reaches the soil. Furthermore, the ^{7}Be content in the soil can be accurately estimated by the ^{7}Be rain content.

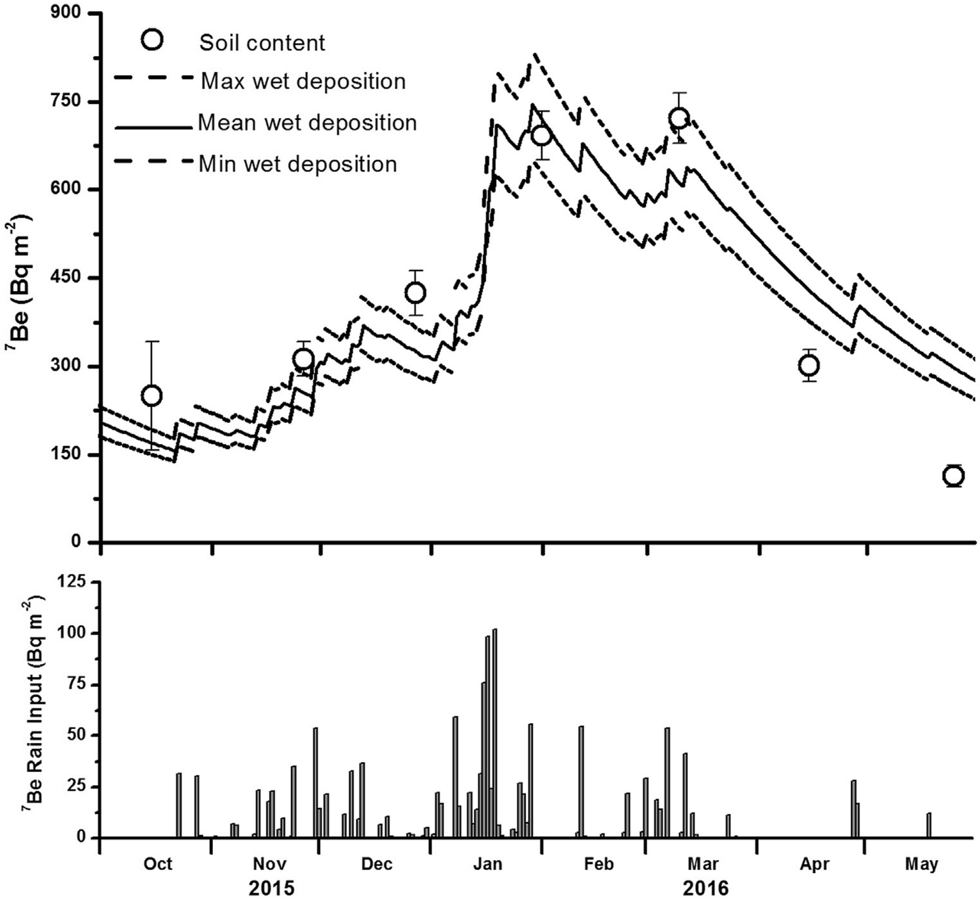

Fig. 6. ^{7}Be wet deposition and ^{7}Be soil content (upper) and ^{7}Be input by rains (lower). ^{7}Be input was estimated from the amount of daily precipitations occurred in the period October 2015 to May 2016 and the estimated slope of Fig. 2. ^{7}Be wet deposition was estimated taking into account the ^{7}Be inputs and its radioactive decay and the error in the slope parameter. The error bar for ^{7}Be soil content are the same that in Fig. 3.

The use of ^{7}Be as a tool for erosion and/or sedimentation is based on the comparison of the total content of ^{7}Be between at a study site (eroded or settled) with that at a reference site. Hence, an accurate measurement of ^{7}Be at a reference site is crucial to carry studies using the ^{7}Be technique. Assessment of the expected ^{7}Be content in soil from the rainfall could be a strong tool to confirm the correct selection of the reference site.

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