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Annual variation of ^{7}Be soil inventory in a semiarid region of central Argentina

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A B S T R A C T

Reliable information on environmental radionuclides atmospheric entrance, and their distribution along the soil profile, is a necessary condition for using these soil and sediment tracers to investigate key environmental processes. To address this need, ^{7}Be content in rainwater and the wet deposition in a semiarid region at San Luis Province, Argentina, were studied. Following these researches, in the same region, we have assessed the ^{7}Be content along a soil profile, during 2.5 years from September 2009 to January 2012. As expected, the specific activity values in soil samples in the wet period (November–April) were higher than in the dry period (May–October). During the investigated period (2009 – beginning 2012) and for all sampled points, the maximum value of the ^{7}Be specific activity (Bq kg^{-1}) was measured at the surface level. A typical decreasing exponential function of ^{7}Be areal activity (Bq m^{-2}) with soil mass depth (kg m^{-2}) was found and the key distribution parameters were determined for each month. The minimum value of areal activity was 51 Bq m^{-2} in August, and the maximum was 438 Bq m^{-2} in February. The relaxation mass depth ranges from 2.9 kg m^{-2} in March to 1.3 kg m^{-2} in August. ^{7}Be wet deposition can explain in a very significant proportion the ^{7}Be inventory in soil. During the period of winds in the region (September and October), the ^{7}Be content in soil was greater than the expected contribution from wet deposition, situation that is compatible with a higher relative contribution of dry deposition at this period of the year.

1. Introduction

Fallout Radionuclides (*FRNs*) are being increasingly used as tracers of different environmental processes across the globe (Zapata, 2002; Mabit et al., 2008). Among these, ^{7}Be has been recognized as a useful tool for documenting short-term soil redistribution due to single rain event and short periods of heavy rainfall (Wallbrink and Murray, 1994; Blake et al., 1999; Walling et al., 1999; Wilson et al., 2003; Schuller et al., 2006). Additionally, Walling et al. (2009) have presented a new approach which provides a means of extending over several months the timeframe to using ^{7}Be measurements to estimate soil redistribution magnitudes.

The use of *FRNs,* particularly of ^{7}Be, could contribute significantly to provide information of both erosion and sedimentation processes, in very sensitive environments to soil degradation which are changing rapidly, such as arid and semi-arid ecosystems. Soil degradation could be accelerated in countries as Argentina, where the agricultural frontier expands rapidly from the pampas to the arid regions which is often detrimental to the sustainability of natural vegetation (FAO, 2008; Viglizzo, 2005; Viglizzo et al., 2003, 2011). Nevertheless, very limited information can be found based on environment radionuclide research in these critical areas.

In previous papers, we have analyzed in a semiarid region of the San Luis Province, in central Argentina, the variability of ^{7}Be content in rainwater and the ^{7}Be wet deposition. A simple iteration algorithm was developed to predict the ^{7}Be areal activity in soil, along an entire annual cycle, from the precipitation regime of the region (Juri Ayub et al., 2009; Juri Ayub et al., 2012).

In the present research, we have evaluated for the same region, the content of ^{7}Be in surface soil. The ^{7}Be areal activity in the vertical soil profile was measured monthly during 2.5 years.

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2. Materials and methods

2.1. Berillium-7 in the environment

Beryllium-7 (Eγ = 477.6 keV, half-life of 53.3 days) is a cosmogenic radionuclide produced in the upper atmosphere and lower stratosphere by cosmic ray spallation with nitrogen and oxygen (Lal et al., 1958). The deposition of ^{7}Be on the earth's surface depends on its production rate (cosmic-ray intensity) which varies according to latitude, altitude and solar activity (Kaste et al., 2002). Factors that will influence concentration in the atmosphere include stratosphere–troposphere mixing, circulation and advection processes within the troposphere, including the mode that it is removed from the troposphere (Feely et al., 1989; Kaste et al., 2002). The atmospheric concentration of ^{7}Be can be considered uniform over small areas (Doering et al., 2006). Beryllium-7 reaches ecosystems mainly through wet deposition, the dry deposition representing less than 10% of the total ^{7}Be fallout (Salisbury and Cartwright, 2005; Ioannidou et al., 2005; Wallbrink and Murray, 1994). It falls on the soil surface primarily in ionic form (Be^{++}), then it is extremely competitive for cation exchange sites being rapidly and strongly fixed to clay minerals, remaining in the first centimeters of the soil profile (Kaste et al., 2002; Hawley et al., 1986; Wallbrink and Murray, 1996; Blake et al., 1999; Walling et al., 1999; Doering et al., 2006).

The above mentioned characteristics make ^{7}Be a useful tool for documenting soil redistribution processes (erosion and sedimentation) and allow to quantify the magnitude of these phenomena (Blake et al., 1999; Walling et al., 1999; Schuller et al., 2006). Essentially, the ^{7}Be technique is based on the comparison of ^{7}Be areal activity (Bq m^{-2}) measurements between a sampling point and a nearby stable reference site. It is assumed that neither erosion nor deposition has occurred at reference site. In soil the reduction of the ^{7}Be content, relative to reference value, shows that erosion has occurred; and inversely, net sedimentation process is evidenced by an increase of ^{7}Be content versus the background inventory of the reference site (Zapata, 2002; Schuller et al., 2006).

2.2. Beryllium-7 content in rainfall and wet deposition

Previous investigations have shown that for different regions and environments, the flux of ^{7}Be in wet deposition is dependent on precipitation volume (Kaste et al., 2002; Juri Ayub et al., 2009; Juri Ayub et al., 2012), and the seasonal deposition pattern may be closely associated with rainfall variability (Feely et al., 1989; Caillet et al., 2001; Juri Ayub et al., 2009; Kaste et al., 2011; Juri Ayub et al., 2012).

Different authors report a linear relationship between ^{7}Be wet deposition (Bq m^{-2}) and precipitation magnitude. Olsen et al. (1985) reported linear relationships with a $r^2 = 0.63$ and 0.54 for two regions in USA; Caillet et al. (2001) reported a $r^2 = 0.66$ ($p < 0.001$) for a site in Switzerland during a complete annual period (November 1997–November 1998). Zhu and Olsen (2009), measuring ^{7}Be in monthly precipitation samples in USA, reported a positive correlation with a $r^2 = 0.46$. In Southern Chile, Walling et al. (2009), found a significant linear relationship between monthly ^{7}Be deposition and monthly precipitations ($r^2 = 0.82$); similar results were found by Kaste et al. (2011) under arid environment in the USA ($r^2 = 0.8$).

For the same semiarid region of the present research, Juri Ayub et al. (2012) found that for typical range of rainfall, ^{7}Be content in rain is independent on precipitation parameters such as the magnitude of the rain, the elapsed time between events, the mean rainfall intensity and duration of the event. The mean value of the ^{7}Be activity concentrations in rainwater was determined as 1.7 Bq l^{-1} (Juri Ayub et al., 2009; Juri Ayub et al., 2012). Beryllium-7 deposition from the atmosphere by rains shows a positive linear relationship with the rain magnitude ($r^2 = 0.85$; $p < 0.0001$), and the ^{7}Be atmospheric depositional flux has been estimated at 1140 ± 120 Bq m^{-2} y^{-1} (Juri Ayub et al., 2009).

2.3. Beryllium-7 along the soil profile

As mentioned above, when ^{7}Be reached the soil, it remains in the first centimeters of the profile. Previous field and laboratory studies suggest that the vertical distribution of the ^{7}Be specific activity (C [Bq kg^{-1}]) is characterized by an exponential decrease with depth (Walling and Woodward, 1992; Blake et al., 1999; Schuller et al., 2006; Walling et al., 2009):

$$C^m(z) = C^m(0)\exp\left(-z/h_0^m\right) \quad (1)$$

where z (kg m^{-2}) represents the mass depth of soil measured from the surface (positive downward), $C^m(0)$ is ^{7}Be specific activity at the surface soil ($z = 0$) and h_0 is the relaxation mass depth, i.e. $C^m(h_0) = 1/e\, C^m(0)$. Since the function $C(z)$ depends on the period of the year, the superscript m (with $m = 1, 2, \ldots, 12$) indicates the month of the year.

By integration of Eq. (1) with respect to z, the total ^{7}Be areal activity $A(z)$ (Bq m^{-2}) at the surface soil, $z = 0$, is (Schuller et al., 2006):

$$A^m(0) = h_0^m C^m(0) \quad (2)$$

and when $z = h_0$, the ^{7}Be areal activity is:

$$A^m\left(h_0^m\right) = 0.368 A^m(0) \quad (3)$$

Thus, 63.2% of the total ^{7}Be areal activity is found within the soil layer from 0 to h_0.

2.4. Sampling and analysis procedures

The study site is located in central Argentina (S 33°9′43″; W 66°16′17″) 17 km north of San Luis City (Province of San Luis). The altitude of the sampling site is 1047 m asl. The average annual temperature is 17 °C, while in summer (December–March) the mean temperature is 23 °C. Annual rainfall ranges from 600 mm to 800 mm. In this region rainfall varies seasonally, with a dry season from May to October and a rainy season from November to April. Rain events in the dry season are scarce and sporadic, with occasional drizzles. In the present study the monthly rainfall records in the period 2005–2011 were considered (REM, 2013; SMN, 2013).

The soil is a sandy loam with 64% of sand, 26% of silt and 10% of clay.

Soil sampling was carried out from September 2010 to January 2012. Soil samples were taken monthly using plastic tubes (10.6 cm diameter × 5 cm height). The plastic tubes were driven into the soil and then, the soil profile was extracted without disturbance. In field, for each sampling place, the procedure consisted of the extraction of three soil cores up to 5 cm deep, identical to each other. In laboratory the soil profile was sliced per 1.5–2 mm increments for the 2 cm topsoil. Soil samples from the same layer were mixed, dried in stove (90 °C) to constant weight with the standard methodology, sieved through a 2 mm mesh and packed Petri dishes (55 mm diameter × 14 mm height) for further gamma spectroscopy analysis.

Table 1
^{7}Be specific activity in soil for each month.

Month, year	^{7}Be specific activity (Bq kg^{-1})							
	Soil layer (1×10^{-3} m)							
	0.0–1.5	0.0–2.0	1.5–3.0	2.0–4.0	3.0–4.5	4.0–6.0	6.0–8.0	8.0–10.0
January, 2012	198 (27)		78 (17)					
February, 2010		139 (26)		84 (14)		42 (10)	20 (8)	11 (8)
March, 2010		98 (22)		53 (12)		26 (9)	19 (6)	
April, 2010		93 (18)		39 (10)		29 (9)	8.9 (6.4)	
May, 2010	67 (17)		38 (14)		17 (8)			
June, 2010	52 (11)		23 (6)		14 (6)			
July, 2010	45 (12)		17 (8)		13 (6)			
August, 2010	24 (10)		10 (6)					
September, 2009		24 (8)		20 (6)		13 (6)	4.2 (4.7)	
September, 2010	87 (22)		40 (14)		23 (10)			
October, 2009		24 (12)		12 (7)				
October, 2010	104 (40)		45 (14)		30 (10)			
November, 2010	73 (18)		29 (12)		3.6 (1.4)			
November, 2011	79 (22)							
December, 2010	115 (34)		62 (21)		24 (11)			
December, 2011	52 (16)							

The value in parentheses indicates the total error in the determination of the activity.

Beryllium-7 activity concentrations in soil samples were determined by measuring its gamma emission at 477.6 keV. Gamma-ray measurements were performed using two germanium detectors. The detector located at the Tandar laboratory (Comisión Nacional de Energía Atómica, Buenos Aires) is a 1.033 kg high-purity germanium crystal with an electroformed high-purity copper cryostat built by Princeton Gamma Tech. The second detector, located in GEA Laboratory (Instituto de Matemática Aplicada, San Luis) is a 'p'-type extended coaxial germanium detector (Canberra GX4018) with a 40% relative efficiency at 1.3 MeV. The energy resolution of each detection system was around 1.7 keV at 1.3 MeV. Both detectors were surrounded by lead bricks to provide shielding against radioactive background.

Data were acquired by using conventional modular electronic instruments and were accumulated in an 8 K ADC (Analog to Digital Converter) multichannel analyzer based on a personal computer. Counting periods for the samples were around 1 day. The gamma spectra were analyzed for all samples to obtain the 477.6 keV peak area. The measured activities were corrected for the decay to the time of sample collection. The background count rate was 0.0009 counts/s/keV at the region of interest (475–480 keV) for the PGT ultra low background technology detector. The errors in the activity concentrations arise from the statistical uncertainties in the peak areas and the uncertainties in the absolute efficiencies of the gamma detectors. Over all uncertainties ranged from 15% to ~50% for activity concentrations in the order of 8 Bq/kg. For specific activities of about 50 Bq/kg, the MDA is around 20 Bq/kg.

3. Results and discussion

Table 1 presents the measured ^{7}Be specific activity in each soil layer for the sampled months. In all cases, the maximum ^{7}Be specific activity was found in the uppermost layer of soil. The ^{7}Be content along the soil profile reached 1 cm depth in February 2010, a wet month. For the topsoil layer (i.e. the first 1.5–2 mm of the soil) ^{7}Be content varied among the months of the studied period (2009 – beginning of 2012). During the dry months ^{7}Be content was minimum and increased when the rainy period started (Table 1). For the uppermost soil layer, ^{7}Be specific activity ranged from 24 Bq kg^{-1} to 45 Bq kg^{-1} in the middle of dry period (July–August) and reached up to 3 or 4 times this value in the middle and ending of the wet period (January–February).

Using the mean value of ^{7}Be activity concentration in rainfalls of 1.7 ± 0.53 Bq l^{-1} estimated by Juri Ayub et al. (2009) and the mean precipitation (mm) in the last six-year period (2005–2011, Table 2) we can estimate the monthly wet deposition of ^{7}Be in Bq m^{-2} (Table 2, Column 4). The error was determined from the standard deviation of precipitation amount and the error in the measurements of rainwater activity.

From the experimental measures of ^{7}Be activity concentration (Table 1) the ^{7}Be areal activity (Bq m^{-2}) on soil profile was calculated (Table 2, Column 5). These values show seasonal variations, like ^{7}Be specific activity; this result is expected from the bimodal distribution of rainfall in the region, with a wet and dry seasons. For a similar semiarid environment in California (USA) seasonal variations in ^{7}Be content in soil due to the asymmetric distribution of rainfall throughout the year, have also been reported (Kaste et al., 2011).

For assessing the ^{7}Be areal activity that could be expected in soil from the wet depositional flux of ^{7}Be, a simple iteration model was applied (Juri Ayub et al., 2009). In this approach, for each time step of the iteration (i.e. ten days), the rainfall used is the mean rainfall value for this period, calculated on the basis of the last six years. The iteration stops when the difference between two successive values of ^{7}Be areal activity is less than 0.02%. This model assumes that: a) ^{7}Be dry deposition is negligible (Salisbury and Cartwright, 2005; Ioannidou et al., 2005; Wallbrink and Murray, 1994); b) all ^{7}Be (wet) deposited on soil is retained (Kaste et al., 2002; Hawley et al., 1986; Wallbrink and Murray, 1996; Blake et al., 1999; Walling et al., 1999; Doering et al., 2006), i.e. there are no superficial or in-depth losses, only radioactive decay occurs; and c) ^{7}Be activity concentration in rainwater remains constant (Juri Ayub et al., 2012). Table 2 (Column 4) shows the estimated values of ^{7}Be areal activity that can be expected in soil taking into account both the monthly atmospheric input by wet deposition and the residual content in soil of ^{7}Be. The relation between the ^{7}Be input from atmosphere to soil and the ^{7}Be content in soil presents a large variability during the investigated period. This difference is about 1% in June when the rain is scarce, and reaches almost 90% in November and December at the beginning of the wet period. Fig. 1 shows, for each month, the ^{7}Be wet deposition (bars), the estimated ^{7}Be areal activity obtained from the model (filled circles) and the measured ^{7}Be areal activity on soil (open squares). The measured values of ^{7}Be areal activity are very close to those estimated from the model.

Table 2
Mean rainfall, calculated ^{7}Be wet deposition, estimated and measured ^{7}Be areal activity and relaxation mass depth for every month.

Month	Mean rainfall (2005–2011) mm	^{7}Be wet deposition Bq m^{-2} calculated	^{7}Be areal activity Bq m^{-2}		Relaxation mass depth kg m^{-2}
			Estimated	Measured	
January	132 (24)	224 (41)	474 (114)	407 (76)	1.2 (0.0)
February	115 (20)	196 (35)	512 (123)	438 (114)	2.2 (0.4)
March	98 (25)	167 (43)	523 (126)	393 (110)	2.9 (0.4)
April	20 (6)	33.4 (10.8)	383 (92)	303 (86)	2.3 (0.4)
May	6.6 (4.3)	11.2 (7.6)	270 (65)	301 (100)	2.6 (0.7)
June	1.8 (0.8)	3.0 (1.6)	183 (44)	164 (47)	2.1 (0.5)
July	4.0 (2.1)	6.8 (3.9)	131 (31)	104 (39)	1.7 (0.6)
August	5.4 (1.6)	9.2 (3.1)	96.5 (23.0)	51 (25)	1.3 (0.0)
September	13 (5)	22.8 (8.3)	87.2 (21.0)	127 (44)	1.3 (0.2)
October	38 (10)	64.9 (17.8)	124 (30)	187 (73)	1.8 (0.3)
November	84 (26)	144 (45)	225 (54)	234 (103)	1.7 (1.6)
December	130 (26)	221 (45)	374 (90)	314 (110)	1.7 (0.4)

The value in parentheses indicates the error: a) in mean rainfall, the error is the standard deviation; b) in wet deposition, the error was estimated by considering rainfall and ^{7}Be rainwater activity errors; c) in estimated areal activity, the error results from ^{7}Be wet deposition error; d) in measured areal activity, the error comes from statistics and detector efficiency errors; e) in the relaxation mass depth, the error is derived from fitting the areal activity as a decreasing exponential function of mass depth.

During the wet period the measured values are systematically lower than the estimated values. Since there is no evidence of soil redistribution process in the sampling region, these differences are probably due to two factors: a) the surface runoff of rain water during heavy rains in summer, or b) the fraction of ^{7}Be retained by the natural grass that grows on the ground during these months.

For all months the ^{7}Be areal activity shows an exponential decrease with mass depth and was fitted according to Eq. (2). Table 2 shows the relaxation mass depth (h_0) obtained for each month of the year. The maximum value of h_0 (2.9 kg m^{-2}) was obtained on the end of the wet period (March); and decrease to 1.3 kg m^{-2} in the dry period (August and September). Fig. 2 displays the vertical distribution of the ^{7}Be areal activity for every month. The theoretical curves (Eq. (2)) for the extreme values of h_0 (March, full line and August, dashed line) have also been represented. These two curves limit a band that contains the measurements of ^{7}Be areal activity for the entire annual cycle.

Walling et al. (2009) and Schuller et al. (2010) report the ^{7}Be content in soil in a forest ecosystem located near Valdivia in Southern Chile during the year 2006 (El Monumento sampling site). Fig. 3 shows comparatively the results obtained at Valdivia and our results at San Luis sampling site. For Valdivia sampling site, authors report a ^{7}Be mean concentration activity in rainfall close to 1.56 Bq l^{-1}, with an annual precipitation of about of 2300mm. Juri Ayub et al. (2009), for the San Luis sampling site stated an annual precipitation around 700 mm with a ^{7}Be mean concentration activity of 1.7 Bq l^{-1}. Consequently, the annual ^{7}Be wet deposition was close to 3600 Bq m^{-2} at Valdivia and 1140 Bq m^{-2} at San Luis. While the rainy period at Valdivia encompasses the months from April to September, at San Luis this period includes the months from November to March (Fig. 3). The maximum values for the ^{7}Be areal activity in soil were respectively 1401 Bq m^{-2} at Valdivia (early August) and 438 Bq m^{-2} at San Luis (February). The ^{7}Be areal activity, in both sites, follows the regional precipitation pattern. Despite the higher ^{7}Be soil content in the Chilean soils during most of the year (from April to December), the relaxation mass depth (h_0) was significantly grater at San Luis sampling site. In this semiarid region the maximum value for h_0 was 2.9 kg m^{-2} in March; instead, at Valdivia sampling site, h_0 reaches 2.15 kg m^{-2} in November. Minimum relaxation mass depth values are very similar in both sites: 1.28 kg m^{-2} at Valdivia sampling site (June) and 1.3 kg m^{-2} at San Luis sampling site (August and September). The disparities observed in the ^{7}Be soil penetration between these sites could be

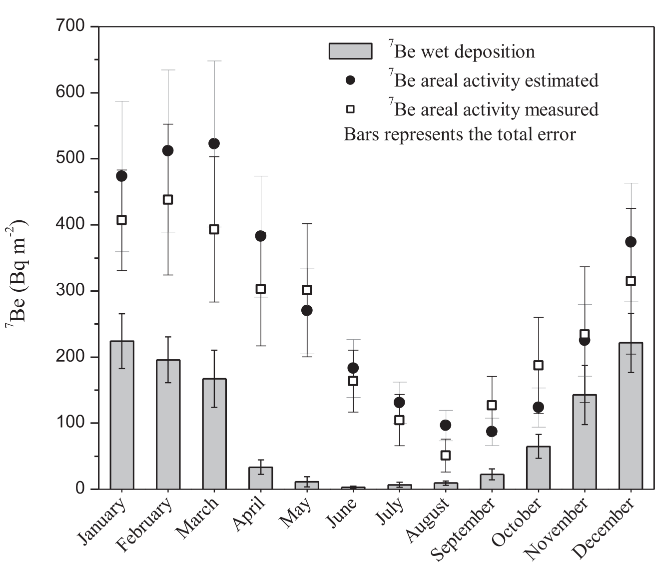

Fig. 1. ^{7}Be wet deposition, calculated and measured ^{7}Be areal activities for an entire annual cycle.

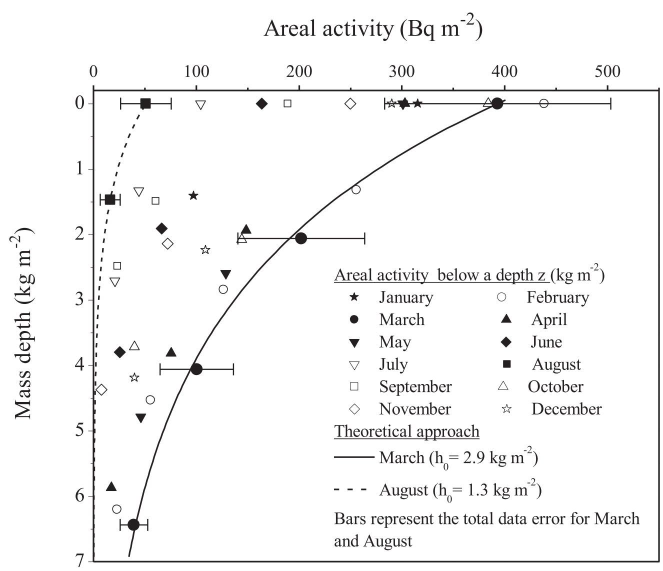

Fig. 2. ^{7}Be areal activity as a function of mass depth for each month. Curves represent the theoretical approach for March and August using Eq. (2).

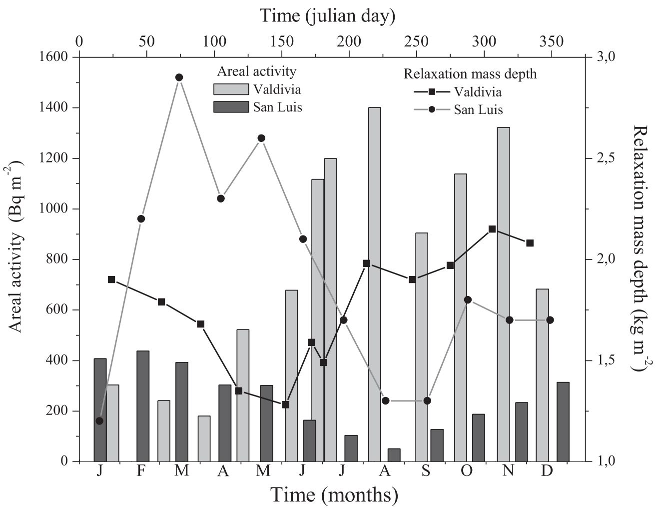

Fig. 3. Monthly ^{7}Be areal activity and relaxation mass depth in a Chilean sampling site (Walling et al., 2009) and in the present study site.

attributable to the wide differences in soil texture; for Valdivia site, the percentage of clay, silt and sand are respectively: 27 ± 2%, 62 ± 2%, 11 ± 1%, while at San Luis sampling sites these percentages are: 10 ± 4%, 26 ± 3%, 64 ± 3%. The lowest clay content and higher sand content in the San Luis sampling station could explain the higher penetration of ^{7}Be along the soil profile.

The progress achieved in this work complement previous studies (Juri Ayub et al., 2009; Juri Ayub et al., 2012) in this region of Central Argentina, recognized as very sensitive to soil degradation and where indicators of the soil conservation status are urgently needed (Viglizzo, 2005). FRNs can provide good approximations to quantify soil conservation status of spatial and temporal scale.

With this in mind, the research carried out was focused not only on knowing the entry and circulation of ^{7}Be in the ecosystem but on identifying the study area as a referential site, in order to apply the ^{7}Be technique to document soil redistribution processes in these marginal agricultural regions.

In this context, it is of importance to identify a site where ^{7}Be input by wet deposition and the ^{7}Be content along the soil profile presents differences smaller than the experimental error of the respective measurements, for most of the annual period. Particularly, in the wet period (from November to March), with the biggest storms in the region, the explored site retains all of the deposited ^{7}Be, allowing the full use of the technique to quantify erosion and sedimentation processes.

The results also show that, immediately after a rainfall, the contribution of ^{7}Be by rain in this region is, in most part of the year, comparatively lower than the pre-existing content of ^{7}Be in ground. This conclusion is particularly valid in the extended wet period (from October to May). This condition should be particularly taken into account when using the technique of ^{7}Be to document soil redistribution process. Precise measurements of ^{7}Be content in soil before and immediately after the rainfall, in the reference site and in the study site, are indispensable.

4. Conclusions

In this paper, we assessed the ^{7}Be content in surface soil in a sensible semiarid region of Central Argentina during an entire annual cycle. The ^{7}Be specific activity was analyzed in the vertical soil profile along this period.

For all the investigated months, the maximum ^{7}Be specific activity was found at the soil surface. On the other hand, the ^{7}Be specific activity in the wet period (November–April) was higher than in the dry period (May–October).

Using the assessed monthly wet deposition of ^{7}Be, the expected ^{7}Be areal activity in soil was estimated using a simple model. These estimated values were confronted with the experimental measurements of ^{7}Be areal activity in soil, showing a good agreement. Differences between estimated and measured ^{7}Be areal activities are probably due to the surface runoff of rain water during heavy rains of summer. In the months of September and October the ^{7}Be areal activity in soil was higher than the estimates from wet deposition. Accordingly, it is expected that dry deposition has a higher contribution in the ^{7}Be content in soil. This hypothesis should be confirmed with further researches in the area.

The ^{7}Be areal activity along the soil profile highlights an exponentially decreasing distribution with mass depth, with a relaxation mass depth ranging from 2.9 kg m^{-2} in March (wet period) to 1.3 kg m^{-2} in August and September (dry period).

The two parameters obtained from the theoretical approach for the ^{7}Be content along the soil profile, *A(0)* and h_0 (Eq. (2)) have been comparatively discussed with those obtained in a Chilean forest, investigated by Schuller et al. (2006, 2010). In both cases *A(0)* follows the annual rainy regime with a clear difference between dry and wet period. The total wet deposition in the Chilean soils was 3.15 times greater than in Argentine soils, while this relationship for the total annual rainfall was 3.29. The relaxation mass depth, h_0, was during the respective wet period greater at the Argentine sampling site. This higher penetration of ^{7}Be in the arid ecosystem could be attributable to the differences in the soil texture in both sampling sites.

The research carried out allows knowing the entry and circulation of ^{7}Be in the ecosystem and identifying the study area as a referential site, in order to apply the ^{7}Be technique to document soil redistribution processes.

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