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Assessment of ^{7}Be content in precipitation in a South American semi-arid environment

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HIGHLIGHTS

► ^{7}Be in precipitation was measured at a semiarid location in central Argentina.
► No atmospheric washout was observed except for one high precipitation event.
► Wet deposition of ^{7}Be was linearly correlated with precipitation.
► This relationship may be applied as a tool for assessing environmental processes.
► The latter includes natural processes as well as the impacts of human disturbance.

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ABSTRACT

There are two naturally occurring radiogenic isotopes of beryllium, ^{7}Be and ^{10}Be. These are produced when cosmic radiation interacts with oxygen and nitrogen in the atmosphere. After production, these radionuclides are input to ecosystems through wet and dry deposition. In recent years ^{7}Be and ^{10}Be have proved to be powerful tools for studying dynamic processes that occur on the surface of the earth. We measured the ^{7}Be content in precipitation at a semiarid location in central Argentina. From November 2006 to March 2009, 68 precipitation events were collected. Measured ^{7}Be content ranged from 0.7 ± 0.4 Bq L^{-1} to 3.2 ± 0.7 Bq L^{-1}, with a mean of 1.7 Bq L^{-1} $\pm$ 0.6 Bq L^{-1}. Beryllium-7 content of rainfall did not show clear relationships with amount of rainfall (mm), mean intensity (mm h^{-1}) or duration (h^{-1}), or elapsed time between events (day). The general results indicate that for the typical range of precipitation there was no atmospheric washout and that the reload of the atmosphere is not a relevant factor, but when the amount of precipitation is very high washout may occur. On the other hand, when the ^{7}Be content was measured during single rain events, a high content of this radionuclide was found to be associated with very low rainfall intensity ($\approx$3 mm h^{-1}), this suggests that rain intensity could affect the ^{7}Be content. Using all data, a good linear relationship between ^{7}Be deposition and rain magnitude was obtained ($r^2 = 0.82$, $p < 0.0001$). Because of this, the slope of this linear regression equation may be applied as a tool for tracing environmental processes that affect the surface of the earth. We can do this by directly estimating erosion/sedimentation processes using ^{7}Be or by estimating the input of ^{10}Be in the environment with the aim to evaluate land degradation phenomena.

1. Introduction

Beryllium-7 (^{7}Be) 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 nuclear reaction produces BeO or $Be(OH)_2$ that diffuse through the atmosphere and adsorbs electrostatically to atmospheric aerosol. 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 and the efficiency with which it is removed from the troposphere (Feely et al., 1989 and Kaste et al., 2002). Beryllium-7 is thought to be input to ecosystems principally by wet deposition with dry deposition contributing less than 10% (Salisbury and Cartwright, 2005; Ioannidou et al., 2005 and Wallbrink and Murray, 1994).

The relatively short half-life of ^{7}Be (53 days), along with the continuous and definable production rates, and competitiveness for cation exchange sites make it a potentially powerful tool for examining

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environmental processes; such as soil redistribution, sediment source assessment, air 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). Willenbring and von Blanckenburg (2010) pointed out that knowledge of the wet deposition, seasonal variations and spatial patterns of ^{7}Be could be a useful tool along with ^{10}Be in the study of dynamic processes that occur on the surface of the earth due to the expected similar behavior of these two radionuclides.

Several studies 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), and the seasonal deposition pattern may be closely associated with rainfall variability (Caillet et al., 2001). Studies of the correlation of ^{7}Be content, measured as activity concentration (Bq L^{-1}), in wet deposition as a function of total precipitation amount (mm) and precipitation intensity (mm h^{-1}) have been extensively studied with divergent results. Wallbrink and Murray (1994) found no significant correlation between measured activities and rainfall amount for precipitation events less than 25 mm, with values ranging from 0.02 to 5.9 Bq L^{-1}. Caillet et al. (2001) found that rainfall events of a few millimeters (<20 mm) showed the highest ^{7}Be activity concentration and that the elapsed time between events affected ^{7}Be content in rain due to the need to reload of the atmosphere with beryllium after a rain event. Through measurement of ^{7}Be in monthly precipitation samples over a seven-year period, Zhu and Olsen (2009) found that the atmospheric deposition fluxes of ^{7}Be varied seasonally, with spring and summer having 1.5 times larger fluxes than autumn and winter. Ioannidou and Papastefanou (2006) showed that precipitation intensity qualitatively characterized ^{7}Be content in rainwater with the content of rainwater being greater for events with low precipitation intensity (<5 mm h^{-1}). They also showed that for short duration events (<5 h), the ^{7}Be content of rainwater was almost five times larger for low rain intensity events relative to those of high intensity.

Kaste et al. (2011) pointed out that, in order to evaluate the potential of ^{7}Be as a tracer in arid and semi-arid environments, it is necessary to know the seasonal and spatial depositional variability and quantify the relationship between precipitation and surface inventories. Walling et al. (2009) proposed a new model to enable the use of ^{7}Be measurements to evaluate soil redistribution rates on a time scale larger than a single rain event; and emphasized that for the applications of this model the ^{7}Be content in rainfall needs to be known.

In a previous work we analyzed the deposition of ^{7}Be by the precipitations, and developed a model for predicting ^{7}Be content in soil (Juri Ayub et al., 2009). This was a first step in a project investigating the use of ^{7}Be for assessing soil redistribution studies in the framework of the IAEA ARCAL RLA 5/0/51 project: "Using Environmental Radionuclides as Indicators of Land Degradation in Latin American, Caribbean and Antarctic Ecosystems". The aim of this paper is to evaluate, in a more exhaustive way, the temporal variability in ^{7}Be content of rainfall with respect to the characteristics of precipitation events using data from 68 samples collected from November 2006 to March 2009. In order to evaluate the effect of rainfall intensity on ^{7}Be content, three rain events were studied in detail to determine changes in activity concentration during the events.

2. Materials and methods

2.1. Study area

The study site was located in central Argentina (S 33°11′; W 66°18′), 15 km north of San Luis City (Province of San Luis); the altitude of the sampling site is 709 m above sea level (Supplementary material). 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). Rains in the dry season are scarce and sporadic (Fig. 1).

2.2. Sampling procedures

Rainwater was collected using two types of collectors: Type I and Type II. Type I collector consisted of a plastic collector of 0.16 cm^{2} surface area and 10 L capacity to facilitate capture of an entire precipitation event. Type II collector was constructed of galvanized steel with a surface area of 0.5 m^{2}. This collector allowed us to collect manually 500 mL of rainwater for each mm of rainfall. The two collectors were placed on a stand 1 m above the ground in order to avoid soil contamination. These were deployed when precipitation began and removed when it ended. After each event collection these were thoroughly rinsed with distilled water.

From the total amount of water collected in Type I collector, a sample of 500 mL was taken. Rainwater samples (collector Types I and II) were filtered and transferred to plastic bottles without further treatment.

The amount of precipitation was measured with a standard rain gauge (Hellmann type, TFA). When Type II collector was used, the elapsed time between collections of each millimeter of rain was recorded with a stopwatch.

2.3. ^{7}Be activity analysis

Beryllium-7 activity concentration (^{7}Be content) in rainwater was determined by measuring gamma emission at 477.6 keV. Gamma-ray measurements were performed using a 1.033 kg high-purity germanium detector built by Princeton Gamma-Tech. The cryostat, made from electroformed high-purity copper, and the related ultralow-background technology were developed by the collaboration of the Pacific Northwest Laboratory and the University of South Carolina (Brodzinski et al., 1990). Background count rate was 0.0005 counts/s/keV at the region of interest (475 to 480 keV). The detector was positioned at the center of a *castle* (≈1 m^{3}) made from lead bricks to provide shielding against radioactive background; the shield has a total mass of ≈12 t.

For gamma counting, 400 mL of each rainwater sample was placed in plastic Marinelli beakers. Counting periods were typically one day. With this geometrical configuration, an absolute photo-peak detection

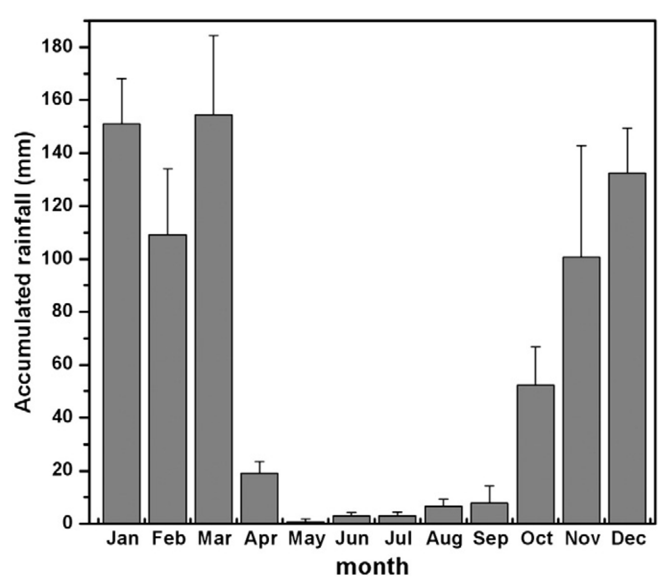

Fig. 1. Mean monthly accumulated rainfall amount in the studied zone. Mean values were obtained averaging the last four years. Bars indicate the standard error.

efficiency was determined by the method described Di Gregorio et al. (2004). The calibration source was prepared by mixing known amounts of the chemical compounds Lu_2O_3, La_2O_3 and KCl in 400 mL of deionized water. The natural occurring long-lived isotopes, ^{176}Lu, ^{138}La, and ^{40}K, emit gamma-rays with energies ranging from 88 to 1460 keV and have simple and well understood decay schemes. A preamplifier extracts the signal from the detector; this analogical output signal is further amplified and connected to an 8K ADC (analog to digital converter) multichannel analyzer. Activity concentrations of ^{7}Be were determined by measuring the 477.6 keV gamma-ray and were corrected for decay to the time of sample collection.

Error associated with calculated activity concentrations arise from the statistical uncertainty in the peak areas and the uncertainty in the absolute efficiency of the gamma detector. Over all uncertainties ranged from 20% to ~50% for activity concentrations in the order of 0.3 Bq L^{-1}. This latter value represents the limit of detection of the measurements.

2.4. Data analysis and statistical procedures

Descriptive statistical analyses, statistical tests and linear regression analyses were carried out using OriginPro® 7.0 (OriginLab, 2002). When the regression analyses between ^{7}Be wet deposition on soil and the precipitation records were performed the uncertainties in the measured values were taken into account as $(1/(\text{error})^2)$ (Bevington, 1969).

In order to investigate the effect of magnitude (mm), duration (h) and intensity (mm h^{-1}) of precipitation events and elapsed time between events (day) on ^{7}Be rainfall content, events were categorized and clustered. Clustering provided better statistical robustness. Because the data showed a normal distribution, a one-way ANOVA test was performed to determine probable differences among the clusters at a significance level of 5% ($\alpha = 0.05$). Statistical results are reported as the value of F-test ($F_{n,m}$), where n is degrees of freedom between-groups and m is degrees of freedom within-groups; and the p value, where $p < 0.05$ indicates significant differences between groups. When the ANOVA test showed differences among groups, a posteriori Tukey test was performed to determine what clusters were different.

3. Results and discussion

3.1. Beryllium-7 content in bulk samples of rainfall events

Beryllium-7 content in individual precipitation events ranged from 0.4 ± 0.2 Bq L^{-1} to 3.2 ± 0.7 Bq L^{-1} (Mean = 1.7 Bq L^{-1}, SD − 0.5 Bq L^{-1}) and the amount of precipitation ranged from 1 to 111 mm (Fig. 2). Most of the precipitation events ranged from 1 to 60 mm; however, there were two extreme events of 70 mm and 111 mm. The 70 mm rainfall showed high ^{7}Be content while the 111 mm rainfall showed low ^{7}Be content (Fig. 2).

There were limited significant differences in ^{7}Be content between individual months ($F_{51,67} = 2.84$; $p = 0.002$) (Fig. 3). Tukey test results showed significant differences only between July 2007 and December 2006, and July 2007 and February 2007. In July 2007, two precipitation events occurred as snow; mean ^{7}Be activity concentrations for these were 0.5 ± 0.2 Bq L^{-1} (Mean ± SD), the lowest values detected among all precipitation events. This is in contrast to the work of Ioannidou and Papastefanou (2006) who found that ^{7}Be content in snow to be twice the ^{7}Be content in rain. These events were not included in our data analyses because snowfall is uncommon in the study region and we cannot reliably assess the difference between rain and snow. Removing these values did not change the mean value for ^{7}Be content in rain over the three years of study of 1.7 Bq L^{-1} (SD = 0.6 Bq L^{-1}).

Kaste et al. (2011), working in an arid environment in California (USA), reported seasonal variation in ^{7}Be content in the soil due to

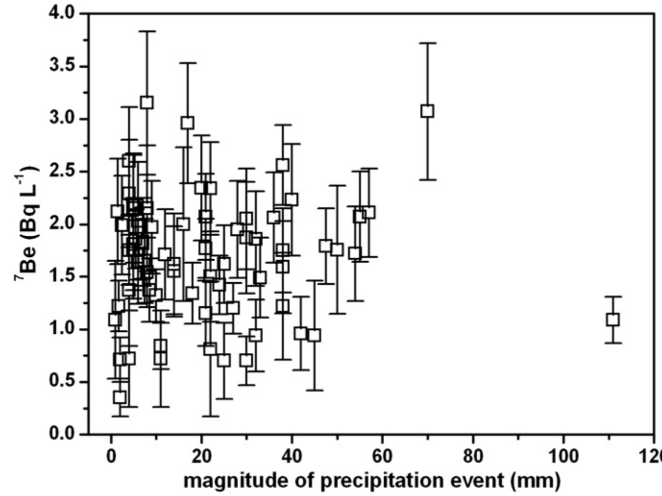

Fig. 2. ^{7}Be activity concentration in bulk sample rain versus magnitude of precipitation. Error bars show the uncertainty in activity concentration measurements.

the asymmetric distribution of rainfall through the year. Feely et al. (1989) found seasonal variations in atmospheric ^{7}Be concentrations and attributed this to a washout effect in an Andean region (Bolivia). Since the same pattern of rainfall distribution (wet and dry seasons) both in our region and in the other two regions exists, similar results might be expected. Our data do not allow the evaluation of differences in ^{7}Be content in rains throughout the year; due to the few rainfalls that occur in winter and autumn (Fig 1).

We also analyzed ^{7}Be activity concentration in rainwater for rain events clustered according to: (a) magnitude of precipitation [mm], (b) elapsed time between events [days], (c) event duration [h] and (d) mean rain intensity [mm h^{-1}] (Fig. 4). When ^{7}Be content of rainfall is compared with respect to the magnitude of precipitation (Fig. 4.a), differences among groups were not found ($F_{55,65} = 1.24$; $p = 0.29$). Caillet et al. (2001) showed that ^{7}Be content in events of a few mm (<20 mm) contained the highest ^{7}Be content. However, Wallbrink and Murray (1994) did not find significant correlation between ^{7}Be content and rainfall magnitude for precipitation events of less than 25 mm.

No differences were found for ^{7}Be content in precipitation with respect to the elapsed time between events ($F_{53,62} = 1.84$; $p = 0.08$; $n = 41$; Fig. 4.b). ^{7}Be activity concentration did not show differences with respect to event duration ($F_{33,40} = 0.98$; $p = 0.46$; Fig. 4.c) as well. No significant differences in ^{7}Be content among clusters were found with

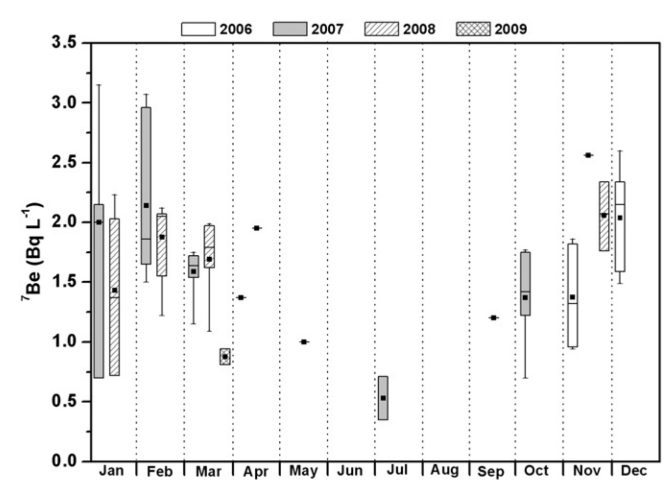

Fig. 3. Box chart representation of ^{7}Be activity concentration in rainwater for rainfall events clustered according to the month of the year. Boxes are determined by the 25th and 75th percentiles. Whiskers are determined by the 5th and 95th percentiles. The arithmetic means of ^{7}Be activity concentration values are also represented.

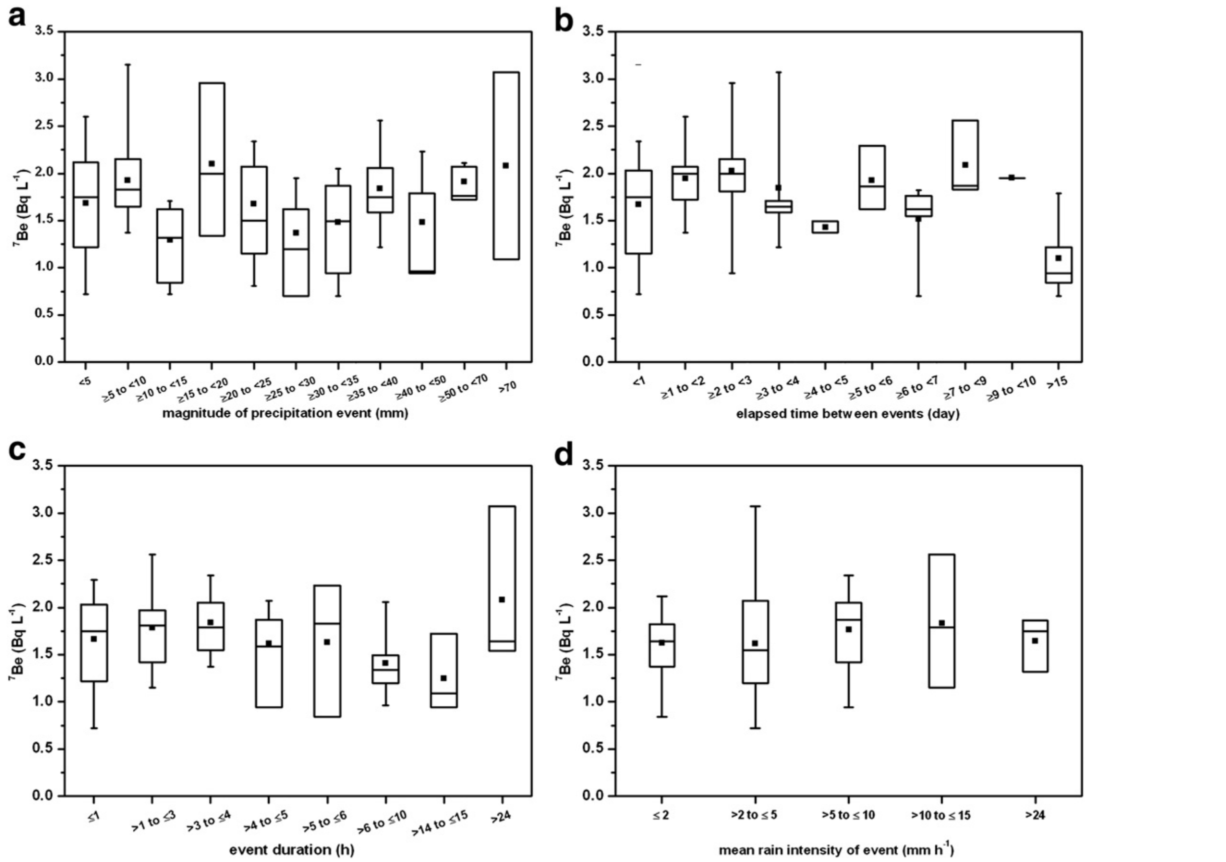

Fig. 4. Box chart representation of ^{7}Be activity concentration in rainwater for rainfall events clustered according to (a) amount of precipitation, (b) elapsed time between events, (c) event duration and (d) mean rain intensity. Boxes are determined by the 25th and 75th percentiles. Whiskers are determined by the 5th and 95th percentiles. The arithmetic means of ^{7}Be activity concentrations values are also represented.

respect to mean rainfall intensity ($F_{36,40}=0.22$, $p=0.92$, Fig. 4.d). When event duration and mean rain intensity were combined to evaluate whether any differences existed in ^{7}Be activity concentrations among rainfall of short (≤5 h) or long duration (>5 h) for rain of low (≤5 mm h^{-1}) or high intensity (>5 mm h^{-1}), no differences were found ($F_{37,40}=0.61$; $p=0.61$, Fig. 4.d).

Caillet et al. (2001) found changes in ^{7}Be content of rainfall with respect to the elapsed time between events and attributed this to the washing effects of rainfall on atmospheric beryllium. These authors estimated an atmospheric reload rate of 0.96 ± 0.10 day^{-1} (recharge time of 1 ± 0.1 day). Ioannidou and Papastefanou (2006) found differences in ^{7}Be content between rains of short (<5 h) and long (>5 h) duration. Our results show that, for our study region, and events of less than 60 mm and 14 hour duration washout of the atmosphere by rain does not occur. However, during one extreme and atypical rain event of 111 mm, a low ^{7}Be content (1.09 ± 0.22 Bq L^{-1}, Fig. 2) was recorded while a high value was reported (3.07 ± 0.65 Bq L^{-1}) for a 70 mm event. These observations do not let us rule out the possibility of a washout of ^{7}Be, as have been reported for other environments (Caillet et al., 2001 and Kaste et al., 2011).

Our results are also in contrast to the results of Ioannidou and Papastefanou (2006) who found a correlation of ^{7}Be content in rain with the intensity of the precipitation: higher for low intensities and lower for high intensities.

3.2. Beryllium-7 content in precipitation over three events

Rain intensity is a variable that can change throughout a precipitation event and the influence on ^{7}Be content in rainfall may be hidden when mean rain intensity is evaluated. Beryllium-7 content and rain intensity were measured for 3 individual rainfall events (identified as A, B and C) for each mm of fallen rain. Rain A (November 8, 2007) had a magnitude of 38 mm and lasted 2.6 h; it was a short duration and high intensity rainfall. Rain B (September 18, 2008) had a magnitude of 27 mm and lasted 9.5 h; it was a long duration and low intensity rainfall; the first 10 mm were collected as 1 mm samples and no sample collected for the last 17 mm due to deposition as hailstones. Rain

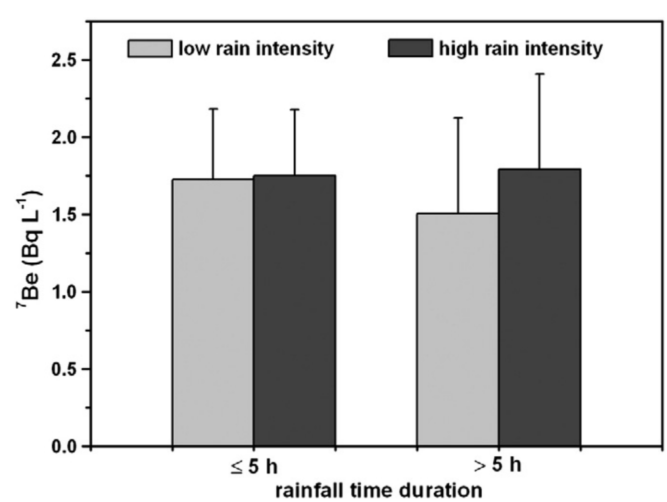

Fig. 5. ^{7}Be activity concentration in rain samples for rainfall events of different mean rain intensities and different duration.

C (November 29, 2008) had a magnitude of 21 mm and lasted 4.2 h; it was a short duration and low intensity rainfall. For this event individual samples of 1 mm of rain were collected for the first 16 mm; one sample was taken from millimeters 17 to 19 (3 mm accumulated), and one sample from millimeters 20 to 21 (2 mm accumulated). Rain intensity showed oscillations in Rains A and C (Fig. 6.a and c) while Rain B shows a constant rain intensity (Fig. 6.b). The oscillations in ^{7}Be content for Rains A and C occurred when the rain intensity changed. The largest changes in ^{7}Be content were observed in Rain A, which showed the largest fluctuations in rain intensity. Although changes in rain intensity seem to be related to changes in ^{7}Be content, low correlation between these variables was obtained ($r^2<0.35$, $p=0.02$).

Ioannidou and Papastefanou (2006) showed that for rain events of short duration, ^{7}Be content was higher in events of low intensity than in events of high intensity; and for events of low rain intensity, ^{7}Be content was higher in those with short duration than in events of long duration. Our results do not show this pattern, Rain A (short duration and high intensity) had a similar ^{7}Be content (2.6 ± 0.4 Bq L^{-1}) than Rain C (2.1 ± 0.4 Bq L^{-1}; short duration and low intensity); and Rain B (long duration and low intensity, 1.2 ± 0.3 Bq L^{-1}) shows a slightly lower ^{7}Be content than Rain C. For rains of low intensity the similar ^{7}Be content between rains of short (Rain C) and long (Rain B) duration time could be explained because in our study region the washing of ^{7}Be from the atmosphere does not seem to occur (for the typical rain magnitudes); the increment in the duration of the rain did not affect atmospheric ^{7}Be content.

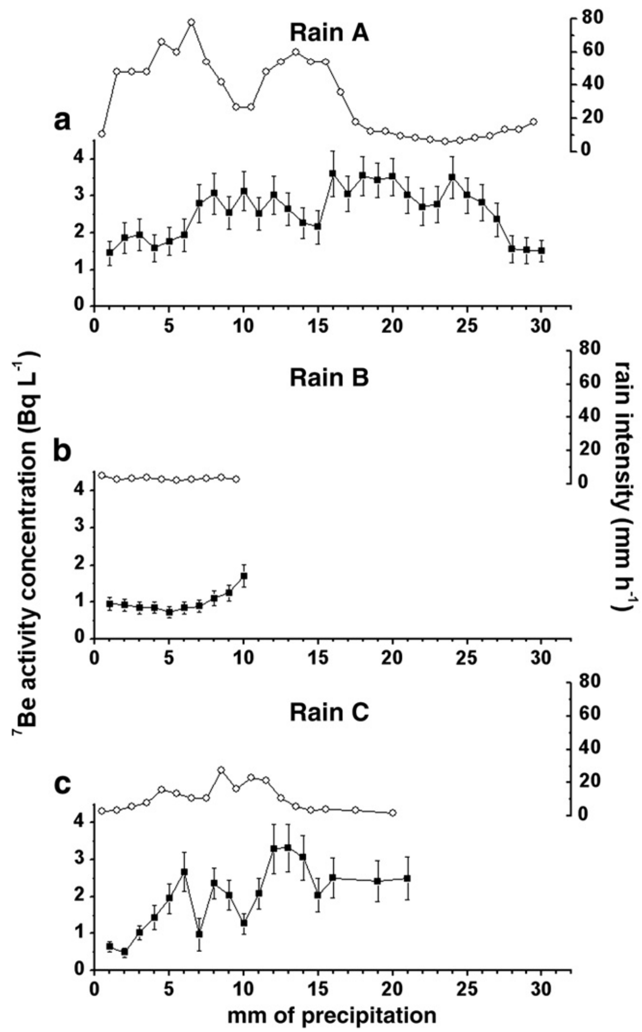

Fig. 6. ^{7}Be activity concentration (filled square) and rain intensity (open circles) during a single precipitation event for (a) Rain A, (b) Rain B and (c) Rain C.

The effect of rain intensity on ^{7}Be content of rain is not clear. Using all data, the changes in rain intensity does not seem to affect the ^{7}Be content (Fig. 5); and analysis of individual events seems to show the same results. However, for individual events when the rain intensity is very low (≈3 mm h^{-1}) the ^{7}Be content in rain is still high (Rain B and last millimeters of Rain C). These data suggest that rain intensity does not affect ^{7}Be content in rain.

3.3. Input of ^{7}Be to the ecosystem by wet deposition

Several authors have reported a linear relationship between ^{7}Be deposition and rain magnitude. Olsen et al. (1985) reported linear relationships with $r^2=0.63$ and 0.54 for two regions in the USA; Caillet et al. (2001) reported $r^2=0.66$ ($p<0.001$) for a site in Switzerland; and Zhu and Olsen (2009), measuring ^{7}Be in monthly precipitation samples in the USA, reported a positive correlation with $r^2=0.46$. Walling et al. (2009), working in Southern Chile, found a good linear relationship between monthly ^{7}Be deposition and monthly precipitations ($r^2=0.82$); similar results were found by Kaste et al. (2011) for an arid environment in the USA ($r^2=0.8$). Our data show a good linear relationship between ^{7}Be deposition and rain event magnitude, with $r^2=0.82$ ($p<0.0001$) and a mean ^{7}Be activity concentration in rainfall of 1.49 ± 0.08 Bq L^{-1} (Fig. 7). This value is of a similar magnitude to the one reported for Chile (Walling et al., 2009) and the estimated ^{7}Be deposition reported by Kaste et al. (2011) for a similar environment in the USA. While we did not find differences in ^{7}Be content when rains are clustered according to amount of precipitation (Fig 4.a.), one event of great magnitude (111 mm) exhibited a low ^{7}Be deposition (Fig. 7). This suggests that extreme rainfall could cause a washing of the atmosphere. Despite this, the slope of linear regression equation (Fig. 7) allows for predicting the expected ^{7}Be content in soil due to rain deposition for the studied region.

This relationship could be applied as a tool for estimating erosion/sedimentation of soil using ^{7}Be. The cosmogenic isotope of Beryllium, ^{10}Be, has been used to evaluate terrestrial surface processes such as soil residence time, soil transport, river sediment and dissolved fluxes, seafloor sedimentation rates and metal scavenging processes in the oceans (Kaste et al., 2002; Willenbring and von Blanckenburg, 2010). However, some analytical and technical difficulties limit its application (cf. Willenbring and von Blanckenburg, 2010). Strong correlations between cosmogenic ^{7}Be and ^{10}Be contents in the atmosphere (Kaste et al., 2002) and rain (Willenbring and von Blanckenburg, 2010) have been established; this is because the production of these two cosmogenic isotopes of beryllium is related and they have a similar chemical behavior. Knowing the linear correlations for ^{7}Be deposition as a

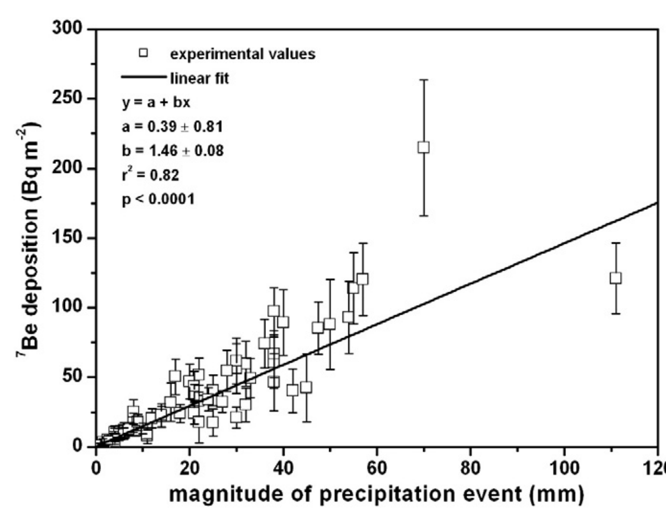

Fig. 7. ^{7}Be deposition versus rainfall amount. Linear fit was obtained with error as weight.

function of precipitation provides mechanisms to allow us to estimate the ^{10}Be content from ^{7}Be measures. This information will be useful for evaluating human impacts on land degradation phenomena as a whole.

4. Conclusions

Beryllium-7 content in rainfall events was analyzed and a mean value of 1.7 ± 0.6 Bq L^{-1} was found for precipitations ranging from 1 to 111 mm. Oscillations in ^{7}Be content in rainfall were observed throughout the year. For the studied region and the typical range of precipitations ^{7}Be content in rain has not shown dependence with the magnitude of the rain, elapsed time between events, mean rainfall intensity and duration of the event. However, data from extreme precipitation events suggest that rainfall intensity and magnitude of the rain could affect the ^{7}Be content in rainwater.

Atypical rain (111 mm) suggests that washing of the atmosphere could occur when the magnitude of the precipitation is greater than 70 mm; in this situation the reload of atmosphere with ^{7}Be might be a relevant factor in the ^{7}Be content of rains. Nevertheless, our results suggest that the reload rate is very fast in this region (<1 day), or that washing effect is not appreciable within the typical range of rainfall magnitude ($\approx$70 mm), or both things together.

When the effect of rain intensity on ^{7}Be content throughout a single rain event was evaluated no correlation was found and the increment of duration time of the rain had no significant effect on ^{7}Be content, also indicating that no washing of the atmosphere is observed. However for limited data from individual rain events was found that low rain intensities are associated with high content of ^{7}Be, indicating that the effect of rainfall intensity on ^{7}Be content should not be rejected.

Beryllium-7 deposition from the atmosphere by rains was calculated, showing a good linear relationship with the rain magnitude. This result allows us to estimate the expected ^{7}Be content in soil by wet deposition from the regional precipitation regime. This could become a useful tool for studies that directly use ^{7}Be to estimate soil redistribution (erosion/sedimentation) or indirectly use ^{7}Be to estimate the cosmogenic ^{10}Be fraction from the total ^{10}Be present in soil.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in the online version, http://dx.doi.org/10.1016/j.scitotenv.2012.09.079. These data include Google maps of the most important areas described in this article.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2012.09.079.

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