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Beryllium-7 in vegetation, soil, sediment and runoff on the northern Loess Plateau

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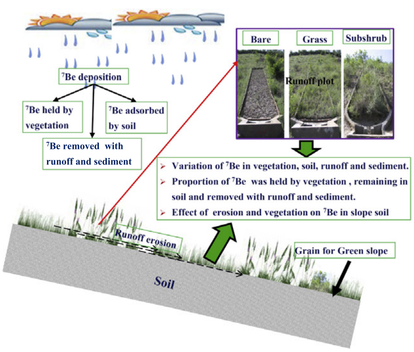
HIGHLIGHTS

- ^{7}Be activity in plant and soil varied significantly during rainy season.
- ^{7}Be inventory held by plant increased with increasing decayed cumulative rainfall.
- ^{7}Be mass activity in sediment decreased with increasing sediment amount.
- Soil erosion and vegetation affected markedly ^{7}Be inventory in the slope soil.
- Most of ^{7}Be remains in the slope soil at the end of the rainy season.

GRAPHICAL ABSTRACT



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ABSTRACT

Beryllium-7 (^{7}Be), as a potentially powerful tracer, was widely used to document soil redistribution and identify sediment sources in recent decades, but the quantity and distribution of ^{7}Be in vegetation, soil, sediment and runoff on the Loess Plateau have not been fully described. In this study, we measured ^{7}Be in vegetation, soil, sediment and runoff on the northern Loess Plateau of China and analyzed its variations during the rainy season to assess the potential of the ^{7}Be method for documenting soil redistribution and identifying sediment sources in a wide range of environments. The results indicated that vegetation, soil, and sediment samples showed higher levels and larger variations of ^{7}Be activities during the rainy season. The drying plants showed ^{7}Be mass activity that was more than three times higher than that of living and semi-decomposed plants. ^{7}Be mass activity in plants and sediment was much higher than in the soil. ^{7}Be activity in runoff water with a few submicron suspended particles varied slightly and was far lower than in plant, soil and sediment samples. The cumulative precipitation generally determined ^{7}Be inventory held by plants and soil. An inverse relationship was found between the ^{7}Be mass activity in sediment and the sediment amount. Globally, approximate 30% of the total ^{7}Be was held by plants in both the herbaceous and subshrub plots. Approximate 10% of the total ^{7}Be was lost with sediment from the bare plot. A very small proportion of ^{7}Be (1.18%–3.20%) was lost with runoff, and the vast majority of ^{7}Be was retained in the slope soil at the end of rainy season. Vegetation cover and soil erosion significantly affected the spatial distribution and variations of the ^{7}Be inventory in soil, providing a necessary condition for the development of a ^{7}Be method to document soil erosion on slopes with vegetation.

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1. Introduction

Cosmogenic ^{7}Be with a short half-life of 53.3 d is formed primarily in the stratosphere and upper troposphere as a natural product of cosmic-ray spallation of oxygen and nitrogen nuclei (Lal et al. 1958). Once produced, ^{7}Be rapidly forms BeO or $Be(OH)_2$ through ionic reactions, then attaches to sub-micrometer atmospheric aerosol particles and diffuses throughout the atmosphere until it is continuously delivered to the earth's surface by wet and dry deposition (Cho et al. 2007; Doering and Akber 2008; Ioannidou and Papastefanou 2006; Juri Ayub et al. 2012; Krmar et al. 2016; Papastefanou and Ioannidou 1995; Papastefanou et al. 1995; Wallbrink and Murray 1994; Zhang et al. 2013). Available evidence suggests that >90% of ^{7}Be deposition is delivered by wet deposition and that the magnitude of annual deposition fluxes varies primarily in response to the amount of annual precipitation and latitude (Wallbrink and Murray 1994). Upon reaching the earth's surface, ^{7}Be is rapidly and strongly adsorbed by ground cover and soil particles (Bondietti et al. 1984; Papastefanou et al. 1999; Wallbrink and Murray 1996; Zhang et al. 2012), and is commonly confined within the upper 20 mm of the soil (Blake et al. 1999; Schuller et al. 2010; Walling et al. 2009; Yang et al. 2006; Zhang et al. 2014). Due to its continual replenishment by fallout, its relatively short half-life, its restriction to a shallow surface layer and its ease of measurement by gamma spectrometry, ^{7}Be, alone or in conjunction with other radionuclides (^{137}Cs, ^{210}Pb, ^{234}Th and ^{40}K), has been successfully used as a tracer of soil particle transport processes, such as short-term or event-based soil erosion on bare slopes, sediment source identification, sediment transport rates and residence times, deposition and resuspension (Baskaran and Swarzenski 2007; Blake et al. 1999; Blake et al. 2002; Botwe et al. 2017; Le Cloarec et al. 2007; Matisoff et al., 2002; Matisoff et al. 2017; Matisoff et al. 2005; Saari et al. 2010; Walling 2013; Walling et al. 1999; Walling et al. 2009; Whiting et al. 2001; Wilson et al. 2003; Yang et al. 2013; Yang et al. 2006; Zhang et al. 2014; Zhang et al. 2018). However, there are still some uncertainties associated with the behavior of ^{7}Be in the soil and related environments, e.g., vegetation interception, root uptake, activity in different soil particle sizes, application on slopes with vegetation and loss due to runoff (Shi et al. 2013; Walling 2013). Knowledge of these behaviors of ^{7}Be in terrestrial environments is a fundamental component in extending the further use of the ^{7}Be method in a wide range of environments. Therefore, the first step in extending the use of the ^{7}Be method is to systematically study variations of ^{7}Be activity in vegetation, soil, sediment and runoff in local areas and to assess the potential of the ^{7}Be method in a wide range of environments.

^{7}Be activity in vegetation, which is mainly dependent on climatic conditions, i.e., on precipitation, and varied markedly between different growing periods, was significant, but the uptake by plant roots did not play an important role (Papastefanou et al. 1999). Wallbrink and Murray (1996) found that up to 55% of the ^{7}Be inventory was held by grass at sites with significant vegetation cover. Similar results were also reported by other researchers (Bettoli et al. 1995; Doering et al. 2006; Kaste et al. 2011). Vegetation growing in dry climates has lower ^{7}Be mass activity than vegetation growing in wet climates (Bondietti et al. 1984). Papastefanou et al. (1999) observed that ^{7}Be activity in grasses varied between 2.1 and 348.0 Bq kg^{-1} (average of 54.4 Bq kg^{-1}). A similar range of ^{7}Be mass activity in nine vegetable species from southwest Finland was reported by Lönnroth et al. (2007). Seasonal variations of ^{7}Be activity in plants were determined by precipitation and its temporal distribution (Pöschl et al. 2010; Sugihara et al., 2008). The differences in ^{7}Be mass activity between plant species were significant (Karunakara et al. 2003; Lönnroth et al. 2007; Zhang et al. 2011). Zhang et al. (2011) found that the mean ^{7}Be mass activity in plants ranked in order of highest to lowest was herbaceous, subshrubs and crop plants. In addition, previous studies also found that whole plant samples contained less ^{7}Be mass activity than leaf samples (Karunakara et al. 2003; Zhang et al. 2012).

The ^{7}Be inventory in a reference site, which is determined by the local characteristics of ^{7}Be deposition (Walling et al. 2009; Zhang et al. 2013), is greatly variable over time and space, from 200 to 1000 Bq m^2 on temperate and tropical land (Kaste et al. 2011). In general, ground cover and soil erosion significantly affected the ^{7}Be inventory in soil on the slope surface (Schuller et al. 2010; Shi et al. 2013; Wallbrink and Murray 1996). The mineral and organic particles in the surface soil strongly adsorbed ^{7}Be, and the finer particles had higher ^{7}Be activity than the coarser particles (Wallbrink and Murray 1996). ^{7}Be decreases exponentially with depth and is commonly confined within the upper 20 mm of the soil (Blake et al. 1999; Schuller et al. 2010; Walling et al. 2009; Yang et al. 2006; Zhang et al. 2014). The depth of ^{7}Be penetration may be primarily controlled by soil type, surface cover, plant roots, soil density, macrovoids, and structure (Wallbrink and Murray 1996). This distribution characteristic of ^{7}Be in the soil profile confirms its potential value as a label of surface soil to document soil redistribution and transition from sheet to rill erosion and to identify sediment source regions and land use (Matisoff et al., 2002; Whiting et al. 2001; Zhang et al. 2014).

^{7}Be mass activity in sediment decreased generally as rainfall proceeded and was consistent with the decrease in ^{7}Be mass activity in soil with depth (Wallbrink and Murray 1993; Yang et al. 2006; Zhang et al. 2014). Matisoff et al. (2002) observed that ^{7}Be mass activity was higher in suspended sediment derived from no-till sub-basins than those derived from conventionally tilled sub-basins and exhibited an inverse relationship with the suspended sediment concentration. Briefly, ^{7}Be mass activity in sediment intensively reflected the changes of erosion patterns, sediment sources, sediment transport rates, distances and residence times (Blake et al. 2009; Feng et al. 1999; Matisoff et al. 2017; Matisoff et al. 2005; Wallbrink et al. 1999; Walling et al. 1999; Whiting et al. 2001; Zhang et al. 2014).

The dissolved phase of ^{7}Be in water was analyzed. Bloom and Crecelius (1983) reported that the solubility of $^7Be^{2+}$ from submicron aerosols of air in seawater was a function of time and that ^{7}Be appeared to be strongly adsorbed on suspended matter and inorganic material at high suspended loads (>20 mg L^{-1}) and was only partially adsorbed at natural levels (~1 mg L^{-1}). Hawley et al. (1986) reported that the partitioning coefficient (K_d) of ^{7}Be in fresh water varied inversely with the solids concentrations at typical environmental values (up to 30 mg L^{-1}). At high solids concentrations (>100 mg L^{-1}), over 90% of ^{7}Be was associated with the solid phrase. Similar values were reported by Li et al. (1984). Matisoff et al. (2002) indicated that 99.9% of suspended solids removed by centrifuging accounted for 92.8% of the total ^{7}Be activity, 0.1% of the filtered solids accounted for 2.3% of the total ^{7}Be activity, and the dissolved phase of ^{7}Be in runoff contained 4.9% of the total ^{7}Be activity for the sample of 241 mg L^{-1} sediment concentration. The ^{7}Be dissolved phase in runoff and ^{7}Be in submicron suspended particles should receive more attention when using ^{7}Be as a tracer, especially for ^{7}Be in submicron suspended particles.

In conclusion, ^{7}Be activity in soil, vegetation, sediment and runoff were investigated and vary significantly. However, there is insufficient knowledge about the behavior of ^{7}Be after it enters terrestrial environments through wet and dry deposition processes. Specifically, we know little about the variations of ^{7}Be activity in vegetation, soil, sediment and runoff over time in relation to environmental conditions and how much ^{7}Be is retained in soil and vegetation or lost with sediment and runoff on slopes at a specific time. These knowledge gaps could affect using ^{7}Be as effective tracer for documenting soil redistribution and identifying sediment sources in a wide range of environments. Moreover, ^{7}Be activity in vegetation, soil, sediment and runoff varied with latitude and precipitation gradients under different climatic conditions, and more systematic analysis is required.

The Loess Plateau in Northern China is well known as one of the most severely eroded areas in the world (Li et al. 2016; Shi and Shao 2000; Zhang et al. 2017; Zhao et al. 2017). To date, although there have been several studies about using ^{7}Be as a tracer to document soil erosion on the Loess Plateau (Yang et al. 2013; Yang et al. 2006; Zhang et al. 2014; Zhang et al. 2018), few studies have been conducted to synthetically analyze ^{7}Be in vegetation, soil, sediment and runoff. There is insufficient data of ^{7}Be that can be used to assess the use of the ^{7}Be method on the Loess Plateau because ^{7}Be deposition is site specific and strongly dependent on location, particularly latitude and local meteorological conditions. Meanwhile, in order to control soil erosion, the Chinese government launched the “Grain for Green” Project in 1999 in some local areas and expanded it in 2000 to the whole Loess Plateau. In this project, cropland was normally converted into grassland, shrub, and forest, and vegetation cover significantly increased on the Loess Plateau (Wang et al. 2017; Yuan et al. 2014; Zhao et al. 2015). There is an increased need for the ^{7}Be method that quantitatively document soil redistribution on slopes with vegetation. Therefore, further studies are required to explore the behavior of ^{7}Be after its deposition on the Loess Plateau.

The purposes of this study were to describe the trends of ^{7}Be inventory in soil from a reference site and to inventory ^{7}Be held by vegetation during the rainy season, in order to analyze ^{7}Be activity in sediment and runoff during erosive rainfall events, and finally, to estimate the proportions of ^{7}Be retained in soil and vegetation and removed with sediment and runoff at the end of the rainy season on the runoff plot scale on the northern Loess Plateau. Our results are expected to provide insight into the behavior of ^{7}Be on the Loess Plateau and expand the scope and scale of utilizing ^{7}Be to trace soil particle transport processes in a wide range of environments. Meanwhile, this study provides data on the environmental behavior of ^{7}Be on the Loess Plateau to add to existing databases.

2. Methods and materials

2.1. Study site

The study site on the Loess plateau was located within the Yangou watershed near Yan'an City of northern Shannxi Province, China. The climate is warm and semi-arid. The mean annual precipitation is approximately 550 mm, >70% of which falls between June and September as intense, short-duration rainstorms that cause severe soil erosion. The mean annual temperature is 9.8 °C. The soil type is Huangmian soil (Calcaric Cambisols, Food and Agriculture Organization).

Three runoff plots, i.e., the bare, herbaceous and subshrub plots, were constructed in 2002 on a “Grain for Green” slope. They are 70 m in length and 40 m in width (Lat. 36°30′50″N, Long. 109°32′23″E). This slope has a northern aspect and is to the leeward of the prevailing winds. Each plot is 15 m in slope length and 2 m in width, with 24° slope gradient for the bare and herbaceous plots and a 29° slope gradient for the subshrub plot. The bare plot was tilled and raked each year and the weeds were removed during the growing season. Plant species in the herbaceous plot with >60% vegetation cover mainly included *Agropyron cristatum* (L.) Gaertn, *Potentilla discolor* Bunge, *Heteropappus hispidus* (Thunb.) Less, *Artemisia scoparia* Waldst. et Kit, and *Tripolium vulgare* Ness. Plant species in the subshrub runoff plot with >60% vegetation cover mainly included *Lespedeza bicolor* Turcz, *Spiraea pubescens* Turcz and *Caragana korshinskii* Kom. There are also some herbaceous plant species, such as *Heteropappus hispidus* (Thunb.) Less and *Agropyron cristatum* (L.) Gaertn in the subshrub plot. These species are the predominant species of the study area and are distributed widely over the entire Loess Plateau. All plant samples were collected from this typical slope adjacent to the runoff plots. The vegetation cover was relatively homogeneous over this slope surface.

2.2. Plant sample collection and laboratory procedures

The mixed samples of plants were collected in 2005 on May 20, June 6, July 27, September 9 and October 22 from the slope surface described above. The mixed plants were sampled within three-five quadrats (1 × 1 m) located randomly on the slope and were then mixed on each sampling date. The living, drying and semi-decomposed plants were separately collected at the first sampling time (May 20), and the living plants were only collected for the other four samplings. The plant samples were gently washed using tap water to remove the soil, dried for at least 24 h at 60–80 °C in an electric oven to obtain a constant dry weight, weighed, and then pulverized. The pulverized plant samples were prepared for analysis of the ^{7}Be activity.

2.3. Soil sample collection and laboratory procedures

On the same dates that the plants were sampled, four soil sampling sites (generally on the summit, terrace and wasteland) adjacent to the typical slope of sampling plants, which were flat, showed no apparent signs of disturbance (including erosion, deposition and human activity) during the previous 5 months and had little vegetation, were selected as reference sites for determining the ^{7}Be reference inventory in the soil. In each reference site, three soil samples were collected using a scraper-plate to a depth of 2 cm within an area of 10×10 cm^2, and these were mixed to provide a single composite soil sample. At the last sampling time, depth-incremental soil samples were collected from the upper 2 cm of the soil at a 4-mm increment at each reference site (October 22) using the scraper-plate. Six sampling points were selected at each reference site, and the total sampling area was 0.24 m^2. The samples corresponding to specific depths were mixed to provide a single composite sample for each depth. The soil depth was expressed as a mass depth (cumulative mass per unit area-kg m^{-2}) based on the measured soil mass and the sampling area. In addition, the soil samples were also collected from each runoff plot at the last sampling (October 22). The plot was equally divided into 10 sections from top to bottom. Three soil samples were collected using the scraper-plate in each section and were mixed to provide a single composite soil sample for each section. Thirty samples were collected from the three different runoff plots. All soil samples were sent back to the laboratory, air-dried, dispersed, passed through a 1-mm sieve and weighed. The samples of sieved soil were prepared for detection of the ^{7}Be activity.

2.4. Runoff and sediment collection and laboratory procedures

The cement pool constructed in the base of each plot was observed after each rainfall event. Five erosive rainfall events occurred from May to October in 2005. The water and sediment depths in the pool were measured for each erosive rainfall event. After measuring the runoff and sediment depths, the runoff and sediment were thoroughly stirred. A well-mixed subsample of approximately 10 kg was taken using the plastic container. The subsample was weighed. The sediment in the subsample was recovered after settling for >24 h, and then the clear runoff water was collected by siphoning. The volume of the siphoned water was determined volumetrically. The settled sediment was oven-dried at 105 °C and weighed to calculate the sediment concentration of the subsample. The total runoff amount and sediment yield could then be determined for each erosive rainfall event. In addition, a rain gauge was installed at the study site to collect data about the duration and amount of each rainfall event over the rainy season (Fig. 1). Considering the decay of ^{7}Be and the significant positive relationship between the ^{7}Be depositional flux and precipitation, we assumed that the cumulative precipitation had the same half-life as ^{7}Be, and the decayed cumulative precipitation was calculated according to this assumption in this study.

The dried sediment was dispersed and passed through a 1-mm sieve. The samples of sieved sediment were prepared for detection of the ^{7}Be activity. 5 L of clear runoff water without filtering, including a few submicron suspended particles, was passed through the chemic method to remove ^{7}Be. A solution of 10% HCl was added to the liquid to obtain a pH of approximately 2 to prevent the container from adsorbing the ^{7}Be. Then, 200 mg of $FeCl_3$ was added and churned. Then, after equilibrating over 2 h, NaOH was added to bring the pH to

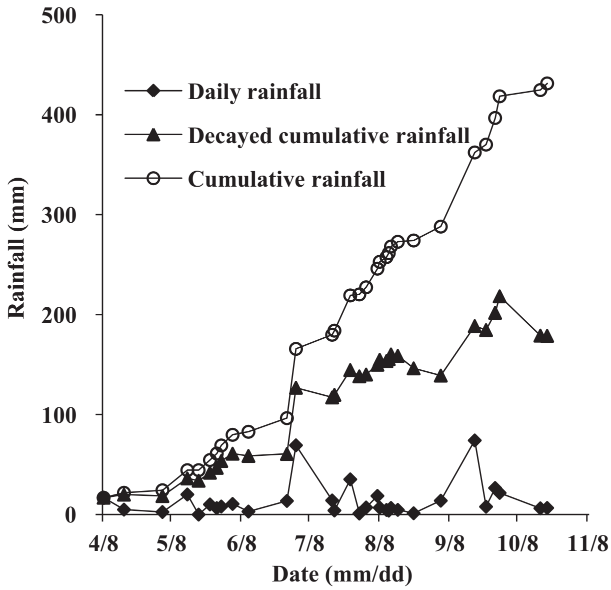

Fig. 1. Daily, decayed cumulative and cumulative rainfall at the study site in 2005.

approximately 8.5 to precipitate the dissolved iron. After 15 h, the floc was separated by a filter and then completely mixed with 400 g of quartzose particles. The mixture was prepared for detection of the ^{7}Be activity.

2.5. Measurements of the ^{7}Be activity

All prepared samples were individually packed into identical plastic cylindrical boxes prior to the detection of the ^{7}Be activity. Measurements of the ^{7}Be activity in all prepared samples were undertaken by gamma spectrometry, which is a high-resolution, low-background, low-energy, hyperpure n-type germanium coaxial r-ray detector (EG&G ORTEC, Oak Ridge, TN, USA). ^{7}Be was measured at 477.7 keV with a count time of 86,400 s. Further details of the ^{7}Be measurement procedure are provided by Yang et al. (2006) and Zhang et al. (2013). The measured ^{7}Be activity was always corrected to the sampling day using the decay constant. The mean inventory of the four soil samples in the reference sites was used to represent the ^{7}Be reference inventory on each sampling occasion.

3. Results

3.1. ^{7}Be activity in vegetation

The samples of the living, drying and semi-decomposed plants were collected on May 20, 2005 and their respective ^{7}Be activity was detected. The results are reported with respect to the dry weight of the sample. ^{7}Be mass activity values in the living, drying and semi-decomposed plants were 66.77, 224.89 and 72.99 Bq kg^{-1}, respectively. The highest ^{7}Be mass activity in the drying plants was more than three times higher than that in the living and semi-decomposed plants. ^{7}Be activity in the living plants was detected during the growing period (May 20 to October 22, 2005) to analyze its variation during the growing period. This was reported by Zhang et al. (2011). ^{7}Be mass activity in the living plants increased during the study period from 66.77 to 288.68 Bq kg^{-1}, with a mean and standard deviation (SD) of 133.91 ± 88.99 Bq kg^{-1} and a coefficient of variation (CV) of 0.66 (Table 1), but it increased at a lower rate between the first four samples and at a higher rate between the last two samples. The ^{7}Be areal activity held by the living plants, which was calculated on the basis of the ^{7}Be mass activity in the living plants and the dry weight of the living plants per square meter, also increased from 0.72 to 46.54 Bq m^{-2} during plant growth, with a mean and SD of 17.50 ± 17.67 Bq m^{-2} and a CV of 1.01. The biomass mainly determined the ^{7}Be areal activity held by vegetation. The significant ^{7}Be activity in vegetation indicated that vegetation showed a significant capacity to intercept and adsorb the ^{7}Be in rainfall.

3.2. ^{7}Be activity in soil

In this study, the ^{7}Be mass activity of soil in the reference site increased from 7.57 to 25.67 Bq kg^{-1}, and the corresponding areal activity increased from 166.61 to 566.84 Bq m^{-2} during the sampling period from May 20 to October 22, 2005 (Table 1). The coefficients of variation of ^{7}Be inventories in the reference site ranged from 7.08% to 12.97% at each sampling time. At the reference site, the depth profile samples were also collected to determine how ^{7}Be was distributed vertically in the soil profile at the last sampling time. Fig. 2 shows that the ^{7}Be activity decreased exponentially with depth and was undetectable beneath a 20-mm depth in the soil, i.e., a mass depth of approximately 24 kg m^{-2}. More than 90% of the total ^{7}Be in the soil was confined within the 10-mm depth. The ^{7}Be mass activity of the surface soil was approximately 120 Bq kg^{-1}, which was obtained by the depth distribution function of ^{7}Be in the soil profile.

^{7}Be inventories in the different types of runoff plots were measured at the last sampling time (Table 2). Soil samples collected from the bare plot showed higher levels of ^{7}Be activity than from the herbaceous and subshrub plots. The mean mass activity values of ^{7}Be in soil with SD were 21.24 ± 6.31, 16.59 ± 1.87 and 16.74 ± 3.33 Bq kg^{-1}, and the mean areal activity values of ^{7}Be in soil with SD were 467.19 ± 138.92, 364.98 ± 41.10 and 367.98 ± 73.21 Bq m^{-2} from the bare, herbaceous and subshrub plots, respectively. The coefficients of variation of the ^{7}Be inventory in the runoff plots ranked in order of highest to lowest were bare, subshrub and herbaceous. ^{7}Be inventories in the reference site were greater than in the different types of runoff plots, except for several sampling points in the bare plot, because of sediment deposition. The results indicated that, compared with the ^{7}Be inventory in the reference site, soil erosion and ground cover significantly affected ^{7}Be inventories in the runoff plots.

Table 1
^{7}Be activity in the reference site soil and in living herbaceous plants during the rainy season on the northern Loess Plateau.

Sampling date (yy/mm/dd)	Decayed cumulative precipitation (mm)	^{7}Be inventory in soil from the reference site		^{7}Be inventory in living herbaceous plants	
		Mass activity (Bq kg^{-1})	Areal activity (Bq m^{-2})	Mass activity (Bq kg^{-1})	Areal activity (Bq m^{-2})
05/05/20	33.86	7.57 ± 0.98[a]	166.6 ± 21.6	66.77 ± 5.92[b]	0.7 ± 0.1
05/06/06	59.36	10.48 ± 1.37	230.5 ± 30.1	87.52 ± 4.18	7.7 ± 0.4
05/07/27	162.53	15.59 ± 1.67	342.9 ± 36.7	102.90 ± 10.50	12.6 ± 1.28
05/09/09	130.31	22.68 ± 2.01	498.9 ± 44.2	123.69 ± 13.83	20.0 ± 2.24
05/10/22	176.53	25.67 ± 1.82	566.2 ± 40.06	288.68 ± 10.68	46.5 ± 7.72

[a] The error represents the standard deviation.
[b] The errors represent the precision of the γ spectrometry measurements at the 95% level of confidence.

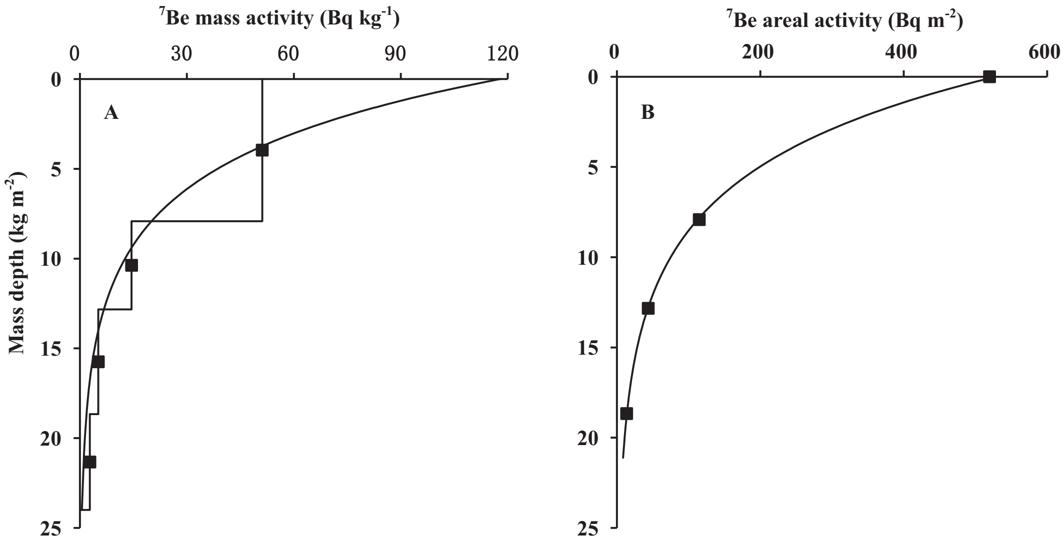

Fig. 2. The depth distribution of ^{7}Be mass activity (A) and areal activity (B) in the soil profile at the reference site.

3.3. ^{7}Be activity in runoff and sediment

Five erosive rainfall events were observed during the study period (Table 3). Runoff and sediment from the bare plot were markedly greater than those from the herbaceous and subshrub plots. ^{7}Be activity in runoff and sediment was detected for these five rainfall events. Generally, ^{7}Be activity in runoff fell into the range from 0.51 to 1.70 Bq L^{-1}, with a mean and SD of 0.92 ± 0.33 Bq L^{-1} and a CV of 0.35, while activity in sediment ranged from 20.08 to 970.64 Bq kg^{-1}, with a mean and SD of 302.94 ± 307.60 Bq kg^{-1} and a CV of 1.02, for all samples. Compared to the ^{7}Be activity in sediment, the slight variations in ^{7}Be activity in runoff were observed between different types of plots and different rainfall events. The ^{7}Be activity in sediment from the runoff plots ranked in order of lowest to highest was bare, subshrub and herbaceous plots for all erosive rainfall events, and that from the bare runoff plot was significantly lower than those from the other two runoff plots. ^{7}Be activity in sediment significantly decreased with an increasing sediment amount regardless of the plot type and rainfall events (Fig. 3).

3.4. Proportion of ^{7}Be in vegetation, soil, sediment and runoff

When considering the use of ^{7}Be as a tracer to document soil movement and identify sediment sources, it becomes important to consider the fate of depositional ^{7}Be after decay on the slope. It is necessary to know the proportion of ^{7}Be held by vegetation, transported by soil erosion and remaining on site. Based on the mass balance of ^{7}Be, the proportion of ^{7}Be held by vegetation, transported with sediment and runoff and remaining in the on-site soil at the runoff plot scale were calculated as follows:

(1) Estimating the total loss of ^{7}Be from the runoff plots:

$$L_{Be} = W_{Be} + E_{Be} \tag{1}$$

where L_{Be} is the total loss of ^{7}Be from the runoff plots (Bq), W_{Be} is the loss of ^{7}Be with runoff (Bq), and E_{Be} is the loss of ^{7}Be with sediment (Bq).

Table 2
^{7}Be inventories in soil samples from different types of runoff plots and reference sites at the last sampling time.

Sampling site (m)	Bare soil plot		Herbaceous plot		Subshrub plot	
	Mass activity (Bq kg^{-1})	Areal activity (Bq m^{-2})	Mass activity (Bq kg^{-1})	Areal activity (Bq m^{-2})	Mass activity (Bq kg^{-1})	Areal activity (Bq m^{-2})
0–1.5	15.52 ± 1.80[b]	407.38 ± 39.56	18.65 ± 1.80	410.32 ± 39.70	16.22 ± 1.68	356.74 ± 37.02
1.5–3	11.25 ± 1.40	247.48 ± 30.83	13.85 ± 1.56	304.78 ± 34.22	14.81 ± 1.61	325.75 ± 35.38
3–4.5	15.77 ± 1.66	346.84 ± 36.50	17.41 ± 1.74	382.97 ± 38.36	16.2 ± 1.68	356.39 ± 37.00
4.6–6	21.17 ± 1.92	465.84 ± 42.30	15.93 ± 1.67	350.45 ± 36.69	11.1 ± 1.39	244.16 ± 30.63
6–7.5	29.17 ± 2.26	641.69 ± 49.65	15.51 ± 1.65	341.12 ± 36.20	19.11 ± 1.83	420.52 ± 40.19
7.5–9	25.69 ± 2.12	565.23 ± 46.60	16.23 ± 1.68	357.12 ± 37.04	13.4 ± 1.53	294.72 ± 33.65
9–10.5	14.34 ± 1.58	315.55 ± 34.82	17.77 ± 1.76	390.85 ± 38.75	20.73 ± 1.90	456.04 ± 41.86
10.5–12	25.54 ± 2.11	561.84 ± 46.46	13.75 ± 1.55	302.42 ± 34.08	20.12 ± 1.87	442.73 ± 41.24
12–13.5	29.78 ± 2.28	655.18 ± 50.17	19.19 ± 1.83	422.11 ± 40.27	20.72 ± 1.90	455.76 ± 41.84
13.5–15	21.13 ± 1.92	464.87 ± 42.26	17.62 ± 1.75	387.63 ± 38.59	14.83 ± 1.61	326.95 ± 35.44
MV[a]	21.24	467.19	16.59	364.98	16.73	367.98
SD	6.31	138.92	1.87	41.10	3.33	73.21
CV	0.30	0.30	0.11	0.11	0.20	0.20
Reference sites	Mass activity (Bq kg^{-1})			Areal activity (Bq m^{-2})		
	26.84 ± 2.16[b]			590.38 ± 47.62		
	24.96 ± 2.09			549.09 ± 45.93		
	27.65 ± 2.16			608.28 ± 47.62		
	23.62 ± 2.03			519.64 ± 44.68		
MV	25.57			566.84		
SD	1.82			40.06		
CV (%)	0.07			0.07		

[a] MV: mean value; SD: standard deviation; CV: coefficient of variation.
[b] The errors represent the precision of the γ spectrometry measurements at the 95% level of confidence.

Table 3
^{7}Be activity in sediment and runoff and its loss with sediment and runoff from the runoff plot for the five erosive rainfall events during the study period.

Date (yy/mm/dd)	R[a]	RI	Plot type	Q_s	$C_{Be,s}$	$L_{Be,s}$	Q_r	$C_{Be,r}$	$L_{Be,r}$
05/07/02	69.3	6.93	Bare	13.97	20.08 ± 2.07[b]	280.51	0.34	0.94 ± 0.18	312.80
			Herbaceous	0.59	323.55 ± 5.1	190.89	0.25	0.9 ± 0.18	225.00
			Subshrub	0.51	152.48 ± 3.3	120.18	0.08	0.93 ± 0.18	74.40
05/07/26	35.24	15.32	Bare	24.5	22.42 ± 2.12	549.29	0.41	0.61 ± 0.17	250.10
			Herbaceous	0.236	607.35 ± 6.39	143.33	0.21	0.69 ± 0.17	144.90
			Subshrub	0.33	235.65 ± 3.87	77.76	0.13	0.88 ± 0.17	114.40
05/08/07–08	25.4	1.67	Bare	4.8	73.96 ± 2.21	355.01	0.13	1.35 ± 0.18	175.50
			Herbaceous	0.17	970.64 ± 8.14	165.01	0.07	0.71 ± 017	49.70
			Subshrub	0.161	643.68 ± 7.56	103.63	0.05	0.83 ± 0.17	37.40
05/09/04	13.9	11.88	Bare	7.49	124.27 ± 3.58	980.78	0.07	0.56 ± 0.17	39.80
			Herbaceous	0	–	–	0.05	0.51 ± 0.17	23.00
			Subshrub	0	–	–	0.03	0.81 ± 0.17	23.50
05/09/19–21	74.8	1.2	Bare	5.5	158.26 ± 2.84	870.43	0.28	1.23 ± 0.17	348.10
			Herbaceous	0	–	–	0.22	1.70 ± 0.17	374.00
			Subshrub	0	–	–	0.13	1.20 ± 0.17	156.00

[a] R: rainfall (mm); RI: rainfall intensity (mm h^{-1}); Q_s: sediment yield (kg); $C_{Be,s}$: ^{7}Be in sediment (Bq kg^{-1}); $L_{Be,s}$: loss of ^{7}Be with sediment (Bq); Q_r: runoff yield (m^{-3}); $C_{Be,r}$: ^{7}Be in runoff (Bq kg^{-1}); $L_{Be,r}$: loss of ^{7}Be with runoff (Bq).
[b] The errors represent the precision of the γ spectrometry measurements at the 95% level of confidence.

(2) Estimating the loss of ^{7}Be with runoff from the runoff plots:

$$W_{Be} = Q_r C_{Be,r} \quad (2)$$

where Q_r is the runoff volume from the runoff plot during rainfall (L), and $C_{Be,r}$ is the ^{7}Be concentration in runoff (Bq L^{-1}).

(3) Estimating the loss of ^{7}Be with sediment:

$$E_{Be} = Q_s C_{Be,s} \quad (3)$$

where Q_s is the sediment yield from the runoff plot during rainfall, and $C_{Be,s}$ is the ^{7}Be mass activity in sediment (Bq kg^{-1}).

(4) Estimating the ^{7}Be held by vegetation in the runoff plot:

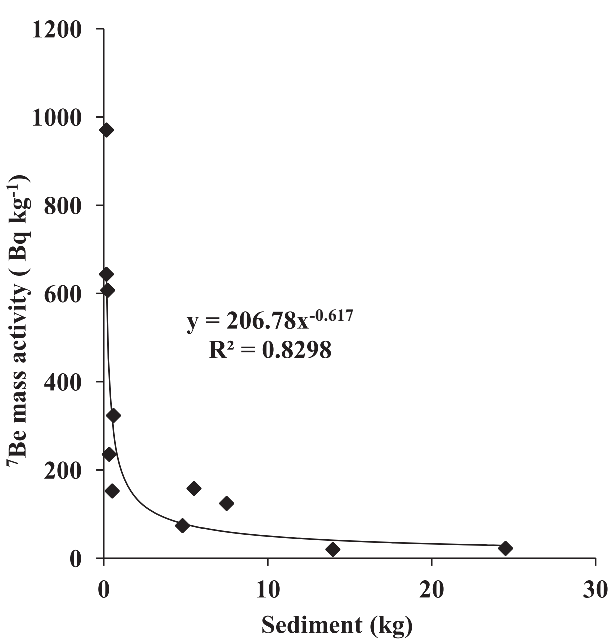

Fig. 3. The ^{7}Be activity in sediment as a function of sediment amount.

$$P_{Be} = S(A_{Be,ref} - A_{Be,av}) - L_{Be} \quad (4)$$

where P_{Be} is the ^{7}Be held by vegetation (Bq) in the runoff plot, $A_{Be,ref}$ is the ^{7}Be inventory in the reference site (Bq m^{-2}), $A_{Be,av}$ is the average ^{7}Be inventory in soil from the runoff plot (Bq m^{-2}), and S is the projective area of the runoff plot (m^2).

The soil and plant samples could not be collected from the runoff plot after each rainfall event in order to avoid the effect of sampling on the subsequent erosive rainfall event. All erosive rainfall events during the rainy season were assumed to be one rainfall event. The measured ^{7}Be activity in runoff and sediment from the different rainfall events was converted to the activity of the last sampling time (October 22, 2005) using the ^{7}Be decay constant and then used to estimate the allocation of ^{7}Be in different materials on the runoff plot scale. The percentages of ^{7}Be held by vegetation, transported with sediment and runoff and remaining in the on-site soil for the different plots are presented in Table 4. The ^{7}Be activity remaining in slope soil accounted for 85.47%, 66.68% and 66.46% of the total ^{7}Be in the bare, herbaceous and subshrub plots, respectively. The ^{7}Be held by vegetation in the bare plot was ignored because of a limited number of plants. The total loss of ^{7}Be from the bare plot accounted for 14.53% of the total ^{7}Be. With runoff, it was 3.21% of the total loss, and with sediment, it was 9.86%. The sum of the loss of ^{7}Be with runoff and sediment was slightly lower than the total loss of ^{7}Be from the bare runoff plot, which may be attributed to the limited number of plants and the uncertainty of measurement. Little soil was eroded from the herbaceous and subshrub plots. The total sediment amount from these two plots was <1 kg during the rainy season. Therefore, the loss of ^{7}Be from these two plots was mostly due to runoff. In the herbaceous plot, the loss of ^{7}Be activity with runoff was 2.39% of the total ^{7}Be activity, and the loss with sediment was 0.97%. The ^{7}Be activity held by vegetation accounted for 29.96% of the total ^{7}Be activity. In the subshrub plot, the loss of ^{7}Be

Table 4
Percentages of ^{7}Be held by vegetation, transported with sediment and runoff and that remained in slope soil at the end of rainy season for the different plots.

Plot	Percentage (%)			
	Vegetation	Soil	Sediment	Runoff
Bare	0.00	85.47	9.86	3.20
Herbaceous	29.96	66.68	0.97	2.39
Subshrub	31.83	66.46	0.53	1.18

activity with runoff was 1.18% of the total ^{7}Be activity, and the loss with sediment was 0.53%. The ^{7}Be activity held by vegetation accounted for 31.83% of the total ^{7}Be activity.

4. Discussion

The ^{7}Be mass activity values in vegetation we observed were significant and were significantly different among published studies (Bondietti et al. 1984; Karunakara et al. 2003; Lönnroth et al. 2007; Pöschl et al. 2010; Papastefanou et al. 1999; Wallbrink and Murray 1996). In general, the levels of the values measured in this study fell in the range of the published values. The mean ^{7}Be mass activity values in plants in this study were closer to the results of Bondietti et al. (1984), Lönnroth et al. (2007) and Papastefanou et al. (1999). However, the measured values in this study were markedly lower than those observed by Wallbrink and Murray (1996), Karunakara et al. (2003) and Pöschl et al. (2010). The differences among these published studies are predominately related to the deposition flux of ^{7}Be, which is mainly determined by the local precipitation. Bondietti et al. (1984) indicated that ^{7}Be mass activity in vegetation growing in a dry climate were the lower than those in a wet climate. The highest ^{7}Be value in grass among the published studies appeared in the study of Karunakara et al. (2003), with the highest annual precipitation of 3786 mm (Zhang et al. 2011). The result of this study, which indicated that the ^{7}Be inventory held by the living herbaceous plants increased generally with the decayed cumulative precipitation (Fig. 4A), was in line with the findings of the previous studies (Bondietti et al. 1984; Pöschl et al. 2010; Zhang et al. 2012).

The vegetation characteristics, such as vegetation type, plant species, vegetation cover, stem and leaf ratio and leaf area, significantly affect the ^{7}Be mass activity in vegetation. The ^{7}Be mass activity in stems was lower than in leaves (Karunakara et al. 2003; Osaki et al. 2003; Zhang et al. 2012). Zhang et al. (2011) found significant differences in ^{7}Be mass activity among plant species. The significant differences in ^{7}Be mass activity among the living, drying and semi-decomposed plants were ascribed to the differences in vegetation characteristics. The sparse density and the smaller leaves of the living herbaceous plants intercepted and adsorbed less ^{7}Be at the beginning of plant growth. The few leaves and the greater number of stems of the semi-decomposed herbaceous plants might be the reason for the lower ^{7}Be mass activity in the semi-decomposed plants than in the drying plants. In addition, the sampling time also affected the ^{7}Be mass activity in vegetation because of the temporal distribution of ^{7}Be deposition and its decay (Pöschl et al. 2010; Sugihara et al., 2008). In this study, the higher ^{7}Be activity in the drying plants was partially ascribed to the longer exposure time of the drying plants than the living plants in the air, because the living plants were just beginning to grow at the sampling time. Globally, these results indicated that the variations in ^{7}Be mass activity in vegetation were complex, and further works need to be refined.

The considerable variations in ^{7}Be inventories in reference sites through time reflect the cumulative input of ^{7}Be after decay. ^{7}Be inventories in temperate and tropical land are 200 to 1000 Bq m^{-2} (Kaste et al. 2011). The values in this study fell in this range. A positive linear relationship was obtained between ^{7}Be inventories in reference sites and the decayed cumulative precipitation during the sampling period (Fig. 4B), which is consistent with the study by Zhang et al. (2013). This relationship implied that, during the sampling period, ^{7}Be inventories and their variations in the reference site soil were predominantly determined by precipitation and its temporal distribution on the Loess Plateau. Compared to the reference sites, ^{7}Be inventories in the different runoff plots were generally lower and had larger variations. These were partially ascribed to soil erosion and vegetation cover. Some studies reported that vegetation significantly held ^{7}Be and reduced the ^{7}Be inventory in the soil (Doering et al. 2006; Kaste et al. 2011; Wallbrink and Murray 1996). ^{7}Be inventories in the soil from the runoff plots and their coefficients of variation indicated that soil erosion and vegetation cover significantly reduced the ^{7}Be inventories in slope soil and enhanced its variation.

In general, ^{7}Be mass activity values in herbaceous plants were significantly higher than in the soil, which is consistent with previous studies (Kaste et al. 2011; Pöschl et al. 2010; Wallbrink and Murray 1996). However, the ^{7}Be areal activity held by the living plants was lower compared to the ^{7}Be areal activity in soil. This was attributed to the lower dry biomass of vegetation (an initial value of 0.01 kg m^{-2} to a final value of 0.16 kg m^{-2}). Many previous studies just reported the ^{7}Be mass activity in vegetation (Kaste et al. 2011; Pöschl et al. 2010; Papastefanou et al. 1999). In fact, calculating the ^{7}Be areal activity held by vegetation was required to quantitatively assess the effects of vegetation on the ^{7}Be inventory in soil and on tracing soil particle transport processes.

^{7}Be mass activity in sediment reflected the variations in the sediment source, sediment transport rates, distances and residence times (Blake et al. 2009; Feng et al. 1999; Matisoff et al. 2017; Matisoff et al. 2005; Wallbrink et al. 1999; Walling et al. 1999; Whiting et al. 2001;

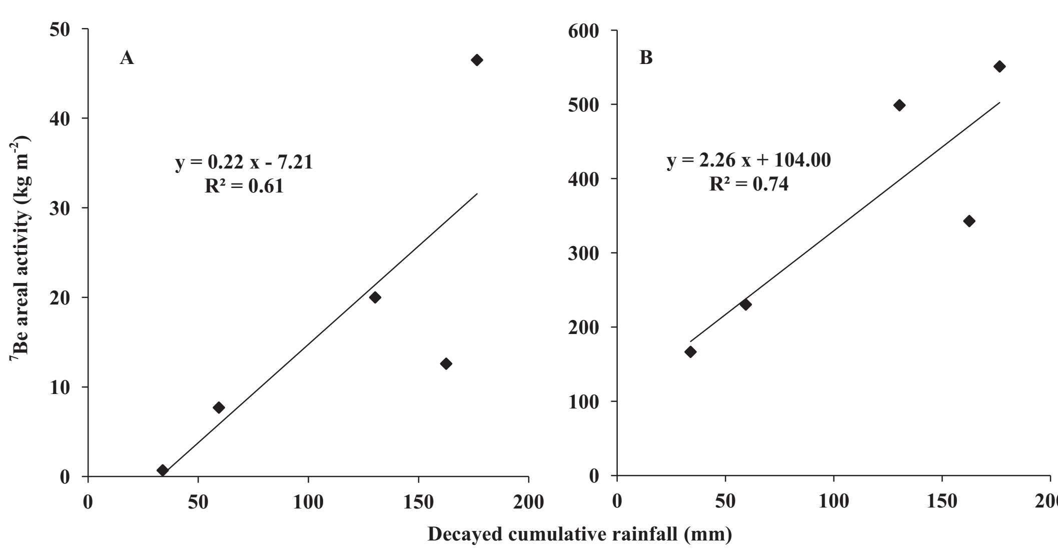

Fig. 4. Relationships between decayed cumulative rainfall and ^{7}Be areal activity held by vegetation (A) and the reference site soil (B) during the study period.

Zhang et al. 2014). Sediment samples in this work showed higher levels and larger variations in the ^{7}Be mass activity for the different plots and rainfall events during the rainy season. These were attributed to erosion intensity, vegetation cover and sample time. An inverse relationship between the ^{7}Be mass activity in sediment and sediment amount is consistent with the result reported by Matisoff et al. (2002). This result was related to the exponentially decreasing distribution of the ^{7}Be activity with depth in the soil profile (Blake et al. 1999; Walling et al. 2009; Yang et al. 2006) and the transition from sheet to rill erosion during rainfall (Whiting et al. 2001; Yang et al. 2006; Zhang et al. 2014). The ^{7}Be mass activity in sediment from the runoff plots was significantly higher than in soil from reference sites at the same sampling time, especially for sediment from the subshrub and herbaceous plots, which was tens of times higher than in soil, which indicated that transport selectivity might affect the accuracy of the ^{7}Be method during soil erosion processes. However, the ^{7}Be mass activity in sediment from the bare plot was lower or higher than in the upper 6 mm of soil, which indicated that small rill erosion occurred in the bare plot for some erosive rainfall events.

Compared with the previous results (Bloom and Crecelius 1983; Hawley et al. 1986; Matisoff et al., 2002; Olsen et al. 1986), the ^{7}Be activity in runoff in this study was higher and varied slightly. The main reason was that the clear runoff without filtering was used after settling for >24 h. However, previous studies analyzed the ^{7}Be activity in the filtered water without submicron sediment particles. The clear runoff in this study contained a few submicron suspended particles, which might adsorb much more ^{7}Be. Matisoff et al. (2002) indicated that filtered solids accounted for only 0.1% of the solids, but they contributed 2.3% of the total ^{7}Be activity and showed higher ^{7}Be mass activity. In most cases, the total loss of ^{7}Be with sediment exceeded that with runoff in this study. The loss of ^{7}Be with runoff from the bare plot ranged from 4% to 53% of the total loss of ^{7}Be for different rainfall events. This result indicated that although the concentration of submicron sediment particles in clear runoff was very low, they held higher ^{7}Be mass activity and are always carried with runoff water in practice. This may affect the estimated accuracy of using ^{7}Be to document soil erosion rates. The ^{7}Be activity in submicron sediment particles should be particularly studied in further research.

The proportions of ^{7}Be in vegetation, soil, sediment and runoff are the basis for quantitatively documenting soil particle transport processes in a wide range of environments. However, no previously published studies of the percentage of ^{7}Be held by vegetation, transported by soil erosion and remaining on site are known to exist in runoff plots. The vast majority of the ^{7}Be retained in slope soil suggested that it is possible to use the ^{7}Be method to document soil erosion on slopes with vegetation, whereas a considerable proportion of the ^{7}Be held by vegetation also suggested that it is necessary to structure a new model or a modified model that takes into consideration the impact factor of vegetation in order to estimate soil redistribution using ^{7}Be measurements. Shi et al. (2013) modified Walling's model for estimating soil redistribution (Walling et al. 1999) by subtracting the ^{7}Be inventory held by vegetation from the ^{7}Be reference inventory in the soil. In Walling's model, it was assumed that the same characteristic of the depth distribution of ^{7}Be between reference and slope sites was the basis for the bare soil. However, in the modified Walling's model, the authors did not consider the effect of vegetation on the characteristic of the depth distribution of ^{7}Be. Vegetation held the ^{7}Be and changed the depth distribution of ^{7}Be in the soil profile, which would result in a different characteristic of depth distributions of ^{7}Be between the reference site and slopes with vegetation. Therefore, the modified model for estimating soil redistribution on grassland by using ^{7}Be measurements proposed by Shi et al. (2013) needed to be reconsidered. The effect of vegetation on the depth distribution of ^{7}Be should be studied in depth. In addition, further works should also focus on the relationships among ^{7}Be deposition, the ^{7}Be inventory in the soil and the ^{7}Be inventory held by vegetation and their interaction effects during the growing period.

5. Conclusions

This study has provided data on ^{7}Be activity in vegetation, soil, sediment and runoff during the study period and the proportions of ^{7}Be held by vegetation, transported by soil erosion and remaining in slope soil. ^{7}Be activity in plants and in the reference soil showed significant increases and varied strongly during the study period. ^{7}Be inventories held by plants and the reference soil increased generally with increasing decayed cumulative rainfall. Sediment showed higher ^{7}Be mass activity than soil, and it significantly decreased with an increasing sediment amount. Slight variations in ^{7}Be activity in runoff with a few submicron suspended particles were observed among different plot types and different rainfall events. ^{7}Be activity in slope soil was significantly affected by soil erosion and vegetation cover, except for the dominant factor of the precipitation. Generally, the vast majority of ^{7}Be was retained in the slope soil and held by vegetation in the plot. A very small proportion of ^{7}Be was lost with runoff. These results indicated that the depositional ^{7}Be was redistributed on the earth surface, and this redistribution could affect utilization of the ^{7}Be method in a wide range of environments. Further work is needed to refine the relationship among ^{7}Be deposition, ^{7}Be inventories in the soil and ^{7}Be inventories held by vegetation and the effect of vegetation on the depth distribution of ^{7}Be. However, our current observations lead to the conclusion that the proportions of ^{7}Be in vegetation, soil, sediment and runoff were significant and should be considered when using the ^{7}Be method to document particle transport processes in a wide range of environments.

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