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Impact of soil hydrological properties on the ^{7}Be depth distribution and the spatial variation of ^{7}Be inventories across a small catchment

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A R T I C L E I N F O

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A B S T R A C T

The natural fallout radionuclide ^{7}Be is used as a tracer allowing estimates of soil redistribution on an event-based time scale. The observed ^{7}Be inventory in the soil is converted to soil redistribution by comparing the ^{7}Be inventory at a stable reference site to the location of interest and taking the ^{7}Be depth distribution in the surface soil into account. The relaxation mass depth (h_0), describing this depth distribution, is assumed to be uniform across the study area, whereas the ^{7}Be inventory at the reference site represents the balance of atmospheric ^{7}Be input and radioactive decay. The ^{7}Be reference inventory should, therefore, not be influenced by soil redistribution or by any variation in physico-chemical characteristics of the soil within the entire study area. Most studies to date use ^{7}Be to monitor soil redistribution on the spatial scale of a hillslope. However, if the assumptions of spatially uniform fallout and rapid and irreversible sorption of ^{7}Be to soil particles can be extended over a larger area, ^{7}Be could be used to monitor soil redistribution at the catchment scale. In this paper, the variability in ^{7}Be distribution in the soil at hillslope and catchment scales is explored and possible sources of this variability are identified. To assess the impact of variability in soil hydraulic conductivity on the depth penetration of ^{7}Be in surface soil, a rainfall simulation experiment with ^{9}Be spiked rainfall was performed on artificially compacted soil cores. These rainfall simulation experiments indicated a significant positive correlation between the saturated hydraulic conductivity (K_{sat}) and the relaxation mass depth and, thus, demonstrated that the assumption of a spatially constant relaxation mass depth is likely to be invalid. An empirical correction factor is proposed to circumvent this problem. This work demonstrates the importance of assessing variability in soil hydrological properties across a study area and is also relevant to studies concerning the vertical transport of fallout contaminants in surface soil. To assess spatial variability in fallout across a catchment and across soil types, soil trays containing different soil types were placed at three reference locations in a small catchment of 8 km^2 across a nine month period. The spatial variation in ^{7}Be reference inventory between the sites in the catchment was not larger than the variation within one reference site (37% and 36% respectively), indicating that the uncertainty on the reference inventory will be similar over the small catchment. However, the different soil types displayed diverse ^{7}Be depth profiles and total ^{7}Be inventories, suggesting that a clear understanding of sorption behavior across the soil types present in a catchment is needed prior to the use of ^{7}Be as a catchment-scale sediment tracer.

1. Introduction

Fallout radionuclides (FRNs) can provide high resolution data to assess spatial patterns and rates of soil redistribution in a retrospective manner, although there are still numerous challenges underlying soil erosion modeling (Walling, 2006). The most commonly used FRNs, caesium-137 (^{137}Cs, $t_{1/2}$ = 30.2 years) and to a lesser extent excess lead-210 ($^{210}Pb_{ex}$, $t_{1/2}$ = 22.1 years), are applied to assess soil redistribution on a medium-term time scale (up to 50 years) (Dercon et al., 2012; Gaspar et al., 2013; Mabit et al., 2008; Porto et al., 2012; Zapata, 2002). The upscaling and mechanization of agriculture increased the sensitivity of soils towards large erosion events (Morgan, 1995), which occur primarily during highly intense, short duration rainfall events. Such events can lead to undesirable offsite effects including mudflows, siltation of downstream ecosystems, flooding and transport of sediment-fixed pollutants (García-Ruiz, 2010; Nagasaka et al., 2005; Wei et al., 2007; Xiao et al., 2013). ^{137}Cs and $^{210}Pb_{ex}$ are not suited to accurately estimate the erosion rates during these short, intense rainfall events

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and, yet, with growing pressure on landscapes through population expansion and climate change, the short-term perspective is becoming increasingly relevant to policy makers. Beryllium-7 (^{7}Be, $t_{1/2}$ = 53.3 days) can be used to estimate erosion rates on a short-term time scale (event-based up to a few months) and can, therefore, be complementary to the application of other FRNs as a tool to monitor event scale soil erosion (Benmansour et al., 2013; Blake et al., 1999, 2002; Dercon et al., 2012; Schuller et al., 2006; Walling, 2012; Wilson et al., 2003). In addition, the ratio of ^{7}Be to $^{210}Pb_{ex}$ can be used to evaluate the residence time of sediments, to track the fate of contaminants, or to distinguish surface versus sub-surface origin of suspended sediment in rivers (Evrard et al., 2016; Landis et al., 2016; Le Cloarec et al., 2007; Matisoff et al., 2005; Wilson et al., 2007).

Despite its potential, to date, the use of ^{7}Be as a sediment tracer has not become common practice (Walling, 2012). Practical issues relating to sampling and ^{7}Be analysis, and knowledge gaps relating to the uncertainty of the method most likely account for this. First, the half-life of 53.3 days in combination with the long measurement time per sample (typically around 86.000 s) limits the total number of samples that can be analyzed before the ^{7}Be has decayed (Ryken et al., 2016; Taylor et al., 2012, 2013; Walling, 2012). Several authors try to circumvent this issue by taking composite samples (Ryken et al., 2016; Schuller et al., 2006, 2010; Sepulveda et al., 2008), wherein the trade-off is reduced quantification of spatial variability. Moreover, the presence of ^{7}Be is generally restricted to the upper two centimeters of the soil. This implies the need for specific sampling equipment operated by an experienced sampler (Baumgart et al., 2016; Mabit et al., 2014; Ryken et al., 2016). Secondly, several knowledge gaps pertain to uncertainty about the validity of the key assumptions underpinning the approach: i) ^{7}Be fallout is spatially uniform across the study area, ii) upon fallout there is rapid sorption to soil particles and iii) sorption to particles is irreversible (Taylor et al., 2013). Finally, analytical problems arise during the measurement of ^{7}Be. A low signal-to-noise ratio in the gamma spectrum due to the low natural abundance of ^{7}Be results in uncertainties during the measurement (Landis et al., 2012). Furthermore, interferences from ^{238}U- and ^{232}Th-series radionuclides complicates accurate ^{7}Be quantification (Landis et al., 2012).

Most current soil redistribution estimations are based on the comparison of the ^{7}Be inventory at the sampling points to the ^{7}Be inventory at a reference site, where no erosion or deposition has occurred during the study period. The observed differences are converted into soil redistribution rates based on the ^{7}Be depth distribution at that reference site and the ^{7}Be inventory variations at the sampling locations compared to the reference inventory. The model is described in detail by Blake et al. (1999) and Walling et al. (2009). The ^{7}Be depth distribution is represented by the relaxation mass depth (h_0), which is defined as the depth above which 63.2% of the total ^{7}Be inventory can be found, directly reflecting ^{7}Be penetration depth. The eroded mass depth, h ($kg\ m^{-2}$) can be calculated as:

$$h = h_0 \ln\left(\frac{A_{ref}}{A}\right) \tag{1}$$

where h_0 is the relaxation mass depth, A ($Bq\ m^{-2}$) is the total ^{7}Be inventory at the sampling location and A_{ref} ($Bq\ m^{-2}$) the total ^{7}Be inventory at a stable reference site.

With this approach the assumption is made that the ^{7}Be inventory across the study site was spatially uniform prior to the erosion event and equal to that at the reference site. This is a major limitation on the use of ^{7}Be as a sediment tracer at locations where these conditions are only rarely met. Only non-erosive rainfall can occur in the preceding few months of the study event or the inventory has to be set at zero due to dilution below the detection limit by tillage. It is crucial that this assumption is met prior to using ^{7}Be as a soil erosion tracer. However several authors have tried to circumvent this problem, e.g. Wilson et al. (2003) calculated erosion rates based on a ^{7}Be inventory balance by measuring the ^{7}Be profile prior and post the event; Walling et al. (2009) used a mass balance model to extend the time scale on which ^{7}Be can be used to monitor erosion rates. This model takes the temporal distribution of the ^{7}Be fallout and erosion during the study period into account and makes use of the evolution of the ^{7}Be depth distribution during the study period. Porto and Walling (2014) also extended the time scale by using a mass balance model to assess the soil loss by resampling the inventories at the sampling points after each rainfall event. Inherent within all of these approaches is the use of the ^{7}Be depth distribution to convert ^{7}Be inventories into soil redistribution rates. It is, therefore, of crucial importance to accurately measure the ^{7}Be depth distribution and to gain a better understanding of the possible spatial variability in the ^{7}Be depth distribution across a study area.

Commonly, the ^{7}Be distribution is observed to decrease exponentially with depth and, with logistical restrictions on the amount of depth profiles that can sampled and analyzed, the measured value of h_0 is assumed to be representative for the depth distribution of ^{7}Be across the study area before erosion occurs (Blake et al., 1999; Schuller et al., 2010). This h_0 is estimated by an exponential fit on the ^{7}Be depth distribution, which can introduce a large uncertainty when the depth distribution deviates from the exponential decrease with depth. Recent research, however, has indicated that the ^{7}Be depth penetration and depth profile are influenced by soil infiltration capacity, which is closely correlated with the saturated hydraulic conductivity (K_{sat}) of the soil (Ryken et al., 2016). In addition, older studies also indicated variation in ^{7}Be depth distribution in relation to textural characteristics of the soil (Wallbrink and Murray, 1996a). The physical characteristics of the soil can vary significantly across a hillslope, resulting in a large variability in hydraulic conductivity across that hillslope (Strudley et al., 2008). This could have a major implication on the modeled erosion rates since variability in h_0 is propagated linearly in the erosion rate estimations. A better understanding of the depth distribution behavior will not only improve erosion rate estimations, but will also lead to a better understanding of tracer behavior of other fallout radionuclides and other atmospheric metals (Landis et al., 2016).

In the first part of this paper the correlation between the hydraulic conductivity and the ^{7}Be depth distribution is investigated. If these are well correlated the hydraulic conductivity can be used as a proxy for the variability in h_0 permitting this uncertainty to be accounted for in the conversion model.

In the second part of this paper an exploratory study was performed to assess the variability in ^{7}Be reference inventory and depth distribution across a small catchment. Rainfall patterns are known to vary across catchments leading to a high variation in ^{7}Be input across a catchment area (Pinto et al., 2013). Several factors such as pH, complexation with dissolved organic compounds, presence of hydrous oxides, mineralogy, presence of competing cations and humic acid have been shown to influence sorption behavior of ^{7}Be in the soil (Armiento et al., 2013; Gil-Garcia et al., 2008; Baskaran et al., 1997; Aldahan et al., 1999; Takahashi et al., 1999), making incomplete ^{7}Be sorption possible and variations in the ^{7}Be depth distribution likely. To date, the use of ^{7}Be to estimate erosion rates is mostly limited to the hillslope scale. It is of interest to explore the possibility of applying ^{7}Be as an erosion indicator at a small catchment scale. Recent research by Porto et al. (2016) evaluated erosion rates in a small catchment of 1.38 ha by applying a mass balance approach. This approaches avoids the need to assess the ^{7}Be reference inventory, but still makes use of the ^{7}Be depth profile to convert the measured ^{7}Be inventories to soil losses. However, physico-chemical characteristics of the soil can influence this depth distribution and significant variations in depth distribution can occur at a catchment scale (Ryken et al., 2016). The variation in ^{7}Be inventory and ^{7}Be depth distribution was evaluated by installing nine small trays at three locations in a 8 km^2 catchment filled with three representative soil types and measuring their ^{7}Be inventory and ^{7}Be depth distribution after 8 months in the catchment.

The aim of is this paper is, therefore, to explore the variability in ^{7}Be distribution in the soil at hillslope and catchment scale, and to identify