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# Excess Lead-210 and Plutonium-239 + 240: Two suitable radiogenic soil erosion tracers for mountain grassland sites



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### ABSTRACT

The expected growing population and challenges associated with globalisation will increase local food and feed demands and enhance the pressure on local and regional upland soil resources. In light of these potential future developments it is necessary to define sustainable land use and tolerable soil loss rates with methods applicable and adapted to mountainous areas. Fallout-radionuclides (FRNs) are proven techniques to increase our knowledge about the status and resilience of agro-ecosystems. However, the use of the Caesium-137 (<sup>137</sup>Cs) method is complicated in the European Alps due to its heterogeneous input and the timing of the Chernobyl fallout, which occurred during a few single rain events on partly snow covered ground. Other radioisotopic techniques have been proposed to overcome these limitations. The objective of this study is to evaluate the suitability of excess Lead-210 ( $^{210}Pb_{ex}$ ) and Plutonium-239 + 240 ( $^{239+240}Pu$ ) as soil erosion tracers for three different grassland management types at the steep slopes (slope angles between 35 and 38°) located in the Central Swiss Alps. All three FRNs identified pastures as having the highest mean ( ± standard deviation) net soil loss of  $-6.7\pm1.1,\ -9.8\pm6.8$  and  $-7.0\pm5.2$  Mg ha  $^1$  yr  $^1$  for  $^{137}\text{Cs},\ ^{210}\text{Pb}_{ex}$  and  $^{239+240}\text{Pu},$  respectively. A mean soil loss of  $-5.7 \pm 1.5$ ,  $-5.2 \pm 1.5$  and  $-5.6 \pm 2.1$  was assessed for hayfields and the lowest rates were established for pastures with dwarf-shrubs (  $-5.2\pm2.5,\ -4.5\pm2.5$  and  $-3.3\pm2.4$  Mg ha  $^{-1}$  yr  $^{-1}$  for  $^{137}Cs,$  $^{210}$ Pb<sub>ex</sub> and  $^{239+240}$ Pu, respectively). These rates, evaluated at sites with an elevated soil erosion risk exceed the respective soil production rates. Among the three FRN methods used, <sup>239+240</sup>Pu appears as the most promising tracer in terms of measurement uncertainty and reduced small scale variability (CV of 13%). Despite a higher level of uncertainty, <sup>210</sup>Pb<sub>ex</sub> produced comparable results, with a wide range of erosion rates sensitive to changes in grassland management.  $^{210}Pb_{ex}$  can then be as well considered as a suitable soil tracer to investigate alpine agroecosystems.

#### 1. Introduction

Land degradation and especially soil erosion are associated with the irretrievable loss of the basic soil resource and particularly in upland mountain areas have severe impacts on water storage and quality, entailing changes in water availability far beyond the mountain areas. In terms of food and feed resources, the importance of this relative unproductive marginal agricultural land decreased during the last 30 years due to the liberalization of agricultural markets that has promoted a global competition. In such a globalized market, farming in mountain regions is clearly disadvantaged because of higher production costs (Streifeneder and Rufjini, 2007) and, as a consequence, mountain farmland is abandoned in Europe (Lasanta et al., 2017). For example in Switzerland, despite subsidies, the number of farms decreased by 40% between 1985 and 2008 (BFS, 2009).

These mountain grasslands might, however, regain some of their importance because of the global demand for primary phytomass for food, which should be at least more than doubled by 2050 (Koning et al., 2008). The growing world population in combination with an unrestrained soil degradation (Borrelli et al., n.d) might increase food and feed shortage in future and trigger the expansion of cultivated land towards these grass- and woodlands (Fischer et al., 2011). In light of these future developments and the expected most pronounced effects of climate change impacting mountainous areas (Beniston, 2003, 2006; Meusburger et al., 2012), it is necessary to identify sustainable land management strategies and to characterize tolerable soil loss rates with specific techniques adapted to steep, inaccessible and rough climate conditions of mountain areas. Particularly snow and avalanches, acting

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as soil erosion agents themselves, greatly impede the use of traditional long-term soil erosion monitoring techniques such as sediment traps or erosion pins (Alewell et al., 2008; Konz et al., 2012; Meusburger et al., 2014).

Fallout radionuclides (FRN) are one of the most convenient approaches to quantify soil erosion in Alpine grasslands (Alewell et al., 2014), because they account for soil loss not only by water, but also for topsoil abrasion caused by snow movement in winter when conventional measurements are not feasible (Meusburger et al., 2014). Moreover, the FRN technique allows to complement and underpin modelled long-term soil erosion rates (Meusburger et al., 2010), although applications of FRNs in context of large scale model applications (e.g. Panagos et al., 2015) are still missing. When FRNs reach the soil surface by wet deposition, they are tightly adsorbed to fine soil particles and the subsequent lateral redistribution of adsorbed FRNs is mainly associated with soil redistribution (Zapata, 2002). The use of FRN measurements to quantify soil redistribution magnitude is commonly based upon a comparison of FRN inventories for individual sampling points to the local reference inventory, where soil erosion is indicated by lower FRN inventories, and sedimentation, by higher FRN levels as compared to the reference site (Ritchie and McHenry, 1990; Mabit et al., 2008).

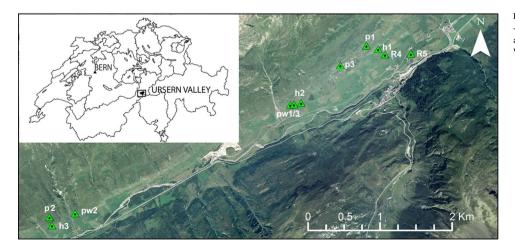
The FRN Caesium-137 (<sup>137</sup>Cs) is the most commonly used tracer for soil erosion assessment. This radioisotope was introduced into the global environment by the thermonuclear weapons testing that took place from the mid-1950s to the early 1960s and from nuclear power plant (NPP) accidents such as Chernobyl in April-May 1986. However, due to radioactive decay the concentrations of <sup>137</sup>Cs are decreasing particularly in the southern hemisphere, where <sup>137</sup>Cs fallout has been much lower than in the north hemisphere (where most of the nuclear tests took place). In addition, in the Swiss alpine areas, we are confronted with an unusually high heterogeneity of <sup>137</sup>Cs reference inventories (Alewell et al., 2014). The latter might be due to the highly uneven spatial distribution of the Chernobyl <sup>137</sup>Cs fallout and the partial presence of snow cover at the end of April-May 1986 that most likely enhanced heterogeneous redistribution during the snow melt process. This heterogeneity can complicate or even compromise the use of the <sup>137</sup>Cs method as the key assumption of homogeneity of the initial fallout is not fulfilled (Haugen, 1991; Lettner et al., 1999; Golosov et al., 2008, 2013; Mabit et al., 2013). The Chernobyl fallout entails a second limitation. To convert FRN inventories into yearly soil erosion rates, the proportion of <sup>137</sup>Cs Chernobyl fallout is required to refer the erosion rates to the correct time period.

To overcome these limitations associated with the use of  $^{137}$ Cs in mountain areas, anthropogenic Plutonium-239+240 ( $^{239+240}$ Pu) has been suggested by the research community as a new radioisotopic approach to determine soil erosion rates in mountain areas (Alewell et al., 2014; Meusburger et al., 2016). Pu isotopes (i.e.  $^{239}$ Pu [half-life =

24,110 years] and <sup>240</sup>Pu [half-life = 6561 years]) in European alpine areas originate solely from the past nuclear weapon tests, which resulted in a more homogenous input due to the longer fallout period above one decade (Alewell et al., 2014). Plutonium isotopes may also help to overcome the second limitation - the apportionment of the <sup>137</sup>Cs originating from Chernobyl fallout - by utilizing specific <sup>239+240</sup>Pu/<sup>137</sup>Cs activity ratios. Indeed, these ratios are significantly different for the two fallout sources with values of 0.029 ± 0.003 (Hardy et al., 1973; Hodge et al., 1996; Kelley et al., 1999) and 0.009 (Muramatsu et al., 2000) for the global and Chernobyl fallout, respectively.

Excess Lead-210 (<sup>210</sup>Pb<sub>ex</sub>) could also be used to overcome these restrictions in the Swiss alpine areas or more generally in areas which have been affected by heterogeneous <sup>137</sup>Cs fallout. In contrast to <sup>137</sup>Cs and  $^{239+240}$ Pu,  $^{210}$ Pb<sub>ex</sub> is a natural geogenic radioisotope (t<sub>1/2</sub> = 22.3 years), which origins from the decay of Radium-226 (<sup>226</sup>Ra).<sup>226</sup>Ra is found in most soils and rocks and produces as its daughter short-lived gaseous Radon-222 (<sup>222</sup>Rn) having a half-life of 3.8 days. Most of this <sup>222</sup>Rn decays to <sup>210</sup>Pb within the soil, producing supported <sup>210</sup>Pb that is in equilibrium with the parent <sup>226</sup>Ra. However, some of the <sup>222</sup>Rn diffuses from the soil into the atmosphere, where it rapidly decays to <sup>210</sup>Pb. This additional <sup>210</sup>Pb is deposited on the ground via precipitation and since it is not in equilibrium with the parent <sup>226</sup>Ra, it is commonly termed unsupported or excess <sup>210</sup>Pb (<sup>210</sup>Pb<sub>ex</sub>). Because of its natural origin, <sup>210</sup>Pbex fallout is essentially constant through time, although seasonal and longer-term variations in <sup>210</sup>Pb<sub>ex</sub> concentrations in the atmospheric fallout (rain, snow and dry deposition) have been reported (Preiss et al., 1996). As supported <sup>210</sup>Pb can be subtracted from total Pb in considering the <sup>226</sup>Ra to <sup>210</sup>Pb ratios, the determination of <sup>210</sup>Pb<sub>ex</sub> has been used successfully worldwide for soil erosion assessment in the last decades (Porto and Walling, 2012; Gaspar et al., 2013; Porto et al., 2013; Mabit et al., 2014). However, its suitability as soil erosion tracer in permanent alpine grasslands has not vet been tested and validated.

This study aims to fill this gap by evaluating the suitability of  $^{210}Pb_{ex}$  in addition to  $^{239+240}Pu$  for assessing soil erosion magnitude in alpine grassland areas under three different types of management (i.e. hayfield, pasture and pasture with dwarf shrubs). Further, we hypothesise that  $^{137}Cs$  based soil redistribution estimates can be improved, if information on the relative contribution of the global versus Chernobyl fallout is known. The latter will be determined by calculating  $^{239+240}Pu/^{137}Cs$  activity ratios at reference sites. The application of the three FRNs pursues the ultimate goal to assess soil redistribution rates for the alpine grassland management type hayfield, pasture and pasture with dwarf shrubs.



**Fig. 1.** Location of the 9 investigated sites (hayfields – h; pastures – p; pastures with dwarf shrubs – pw) and the two reference sites (R4, R5) in the Urseren Valley (Central Swiss Alps).

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