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Decision support for the selection of reference sites using ^{137}Cs as a soil erosion tracer

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Abstract. The classical approach of using ^{137}Cs as a soil erosion tracer is based on the comparison between stable reference sites and sites affected by soil redistribution processes; it enables the derivation of soil erosion and deposition rates. The method is associated with potentially large sources of uncertainty with major parts of this uncertainty being associated with the selection of the reference sites. We propose a decision support tool to Check the Suitability of reference Sites (CheSS). Commonly, the variation among ^{137}Cs inventories of spatial replicate reference samples is taken as the sole criterion to decide on the suitability of a reference inventory. Here we propose an extension of this procedure using a repeated sampling approach, in which the reference sites are resampled after a certain time period. Suitable reference sites are expected to present no significant temporal variation in their decay-corrected ^{137}Cs depth profiles. Possible causes of variation are assessed by a decision tree. More specifically, the decision tree tests for (i) uncertainty connected to small-scale variability in ^{137}Cs due to its heterogeneous initial fallout (such as in areas affected by the Chernobyl fallout), (ii) signs of erosion or deposition processes and (iii) artefacts due to the collection, preparation and measurement of the samples; (iv) finally, if none of the above can be assigned, this variation might be attributed to “turbation” processes (e.g. bioturbation, cryoturbation and mechanical turbation, such as avalanches or rockfalls). CheSS was exemplarily applied in one Swiss alpine valley where the apparent temporal variability called into question the suitability of the selected reference sites. In general we suggest the application of CheSS as a first step towards a comprehensible approach to test for the suitability of reference sites.

1 Introduction

Soil erosion is a global threat (Lal, 2003). Recently estimated erosion rates range from low rates of 0.001–2 $t\,ha^{-1}\,yr^{-1}$ on flat relatively undisturbed lands (Pimentel, 2006) to high rates under intensive agricultural use of $> 50\,t\,ha^{-1}\,yr^{-1}$. In mountainous regions, rates ranging from 1 to 30 $t\,ha^{-1}\,yr^{-1}$ have been reported (e.g. Descroix and Mathys, 2003; Frankenberg et al., 1995; Konz et al., 2012) where they often exceed the natural process of soil formation (Alewell et al., 2015). The use of the artificial radionuclide ^{137}Cs as a soil erosion tracer has been increasing during the last decades, and the method has been applied all over the world with success (e.g. Mabit et al., 2013; Zapata, 2002). The use of ^{137}Cs as a soil erosion tracer allows for an integrated temporal estimate of the total net soil redistribution rate per year since the time of the main fallout, including all erosion processes by water, wind and snow during summer and winter seasons (Meusburger et al., 2014).

^{137}Cs was released into the atmosphere during nuclear bomb tests and as a consequence of nuclear power plant (NPP) accidents, such as Chernobyl in April 1986. It reaches

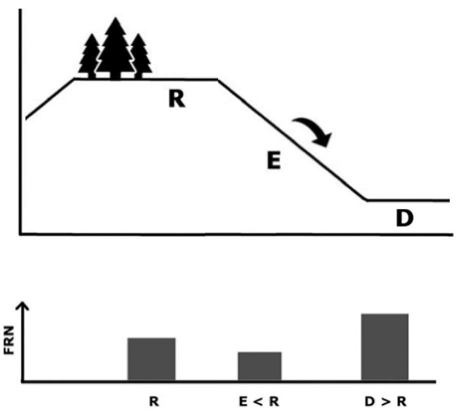


Figure 1. Concept of the fallout radionuclide (FRN) traditional method, in which the FRN content of a reference site located in a flat and undisturbed area (R) is compared to the FRN content of disturbed sites (E and D). If the FRN at the site under investigation is lower than at the reference site, the site has experienced erosion processes (E). If the FRN content is greater than at the reference site, the site has experienced deposition processes (D).

the land surface by dry and wet fallouts and once deposited on the ground, it is strongly bound to fine particles at the soil surface. Due to its low vertical migration rates, it moves predominantly in association with fine soil particles through physical processes and provides an effective track of soil and sediment redistribution processes (Mabit et al., 2008). The traditional approach to using the ^{137}Cs method is based on the comparison between the inventory (total radionuclide activity per unit area) at a given sampling site and that of a so-called reference site located in a flat and undisturbed stable area. The method indicates the occurrence of erosion processes at sites with a lower ^{137}Cs inventory compared to the reference site and sediment deposition processes at sites with a greater ^{137}Cs inventory (Fig. 1a). Specific mathematical conversion models allow for the derivation of quantitative estimates of soil erosion and deposition rates from the latter comparison (IAEA, 2014).

The efficacy of the method relies on an accurate selection of representative reference sites (Mabit et al., 2008; Owens and Walling, 1996; Sutherland, 1996). The measured total ^{137}Cs inventory at the reference sites represents the baseline fallout (i.e. reference inventory), a fundamental parameter for the qualitative and quantitative assessment of soil redistribution rates (Loughran et al., 2002). It is used for the comparison with the total ^{137}Cs inventories of the sampling sites and therefore determines if and how strongly a site is eroding or accumulating sediments. Moreover, the depth profile of the ^{137}Cs distribution in the soil at the reference site plays a very important role, as the shape of this profile is used in the conversion models to convert changes in ^{137}Cs inventory to quantitative estimates of soil erosion rates (Walling et al., 2002). Recent studies have demonstrated the sensitivity of conversion models to uncertainties or even biases in the reference inventory (e.g. Arata et al., 2016; Iurian et al., 2014; Kirchner, 2013).

The close proximity of a reference site to the area under investigation is required to meet the assumption that both have experienced similar initial fallout. The latter is particularly important if the study area was strongly affected by Chernobyl fallout, which, aside from global fallout from nuclear weapons testing, is the major ^{137}Cs input in many regions of Europe. Because of different geographical situations and meteorological conditions at the time of passage of the radioactive cloud, the contamination associated with Chernobyl fallout was very inhomogeneous (Chawla et al., 2010; Alewell et al., 2014). Therefore, in some areas a significant small-scale variability in ^{137}Cs distribution may be expected. As already pointed out by Lettner et al. (1999) and Owens and Walling (1996), this might impede the comparison between reference and sampling sites. To adequately consider the spatial variability in the FRN fallout, multiple reference sites should be selected and the variability within the sites properly addressed (Kirchner, 2013; Mabit et al., 2013; Pennock and Appleby, 2002). In addition, the reference site should not have experienced any soil erosion or deposition processes since the main ^{137}Cs fallout (which generally requires that it was under continuous vegetation cover, such as perennial grass). Different forms of turbation, including animal turbation, anthropogenic turbation and cryoturbation or snow processes, may also affect the ^{137}Cs soil depth distribution at the reference site. Finally, the collection of the samples, the preparation process and gamma analysis might introduce a certain level of uncertainty, which should be carefully considered. For instance, Lettner et al. (1999) estimated that preparation and measuring processes contribute 12.2 % to the overall variability in the reference inventory. Guidance in the form of independent indicators (e.g. stable isotopes as suggested by Meusburger et al., 2013) for the suitability of reference sites might assist with the selection of reference sites.

All in all the suitability or unsuitability of references site is crucial; it may even be the most crucial step in all FRN-based erosion assessments. The general suitability of ^{137}Cs-based erosion assessment has been recently and controversially discussed (Parsons and Forster, 2011, 2013; Mabit et al., 2013). We would like to propose that the FRN community agree on general concepts and sampling strategies to test the suitability of reference sites in order to improve the method and establish trust in this useful erosion assessment method. Up to now, the variability among spatial replicate samples at reference sites has commonly been the sole criterion to decide on the suitability of a reference value. We propose an extended method to Check the Suitability of reference Sites (CheSS) using a repeated sampling strategy and an assessment of the temporal variability of reference sites. The suitability of reference sites for an accurate application of ^{137}Cs as a soil erosion tracer is tested at Urseren Valley (Canton Uri, Swiss Central Alps).

2 CheSS (Check the Suitability of reference Sites): a concept to assess the suitability of reference sites for the application of ^{137}Cs as a soil erosion tracer

2.1 Repeated sampling strategy and calculation of inventories

The time period for the repeated sampling of reference sites needed for the application of ^{137}Cs as a soil erosion tracer will be site- and case-specific and depends on the initial small-scale spatial variability and the depth distribution of the reference inventory. The time span should be of sufficient length to cause an inventory change that is larger than the uncertainty related to the inventory assessment, e.g. larger than 35 %. In our study site, which is affected by anthropogenic disturbance and snow erosion of several millimetres per winter, 2 years can be considered sufficient (Meusburger et al., 2014). Several spatial repetitions following the suggestion of Sutherland (1996) are necessary and should be analysed separately to investigate the small-scale variability in ^{137}Cs in the area. As we detected measurement differences between different detectors (see below), all samples should ideally be measured for ^{137}Cs activity using the same analytical facilities. Finally, ^{137}Cs activity needs to be decay corrected to the same date (either the period of the first sampling campaign or the second) considering the half-life of ^{137}Cs (30.17 years).

The decay-corrected ^{137}Cs activities (act, $\mathrm{Bq\,kg^{-1}}$) of each soil layer in the depth profile are converted into inventories (Inv, $\mathrm{Bq\,m^{-2}}$) with the following equation:

$$\mathrm{Inv} = \mathrm{act} \times xm, \qquad (1)$$

where xm is the measured mass depth of fine soil material (< 2 mm fraction; $\mathrm{kg\,m^{-2}}$) in the respective soil sample. The depth profile of each reference site is then displayed as inventory ($\mathrm{Bq\,m^{-2}}$) against the depth of each layer (cm). The repeated sampling inventory change ($\mathrm{Inv_{change}}$) can then be defined as

$$\mathrm{Inv_{change}} = \frac{\mathrm{Inv}_{t0} - \mathrm{Inv}_{t1}}{\mathrm{Inv}_{t0}} \times 100, \qquad (2)$$

where t_0 and t_1 are the dates of the first and second sampling campaigns, respectively, Inv_{t1} is the ^{137}Cs inventory ($\mathrm{Bq\,m^{-2}}$) at t_1 and Inv_{t0} is the ^{137}Cs inventory at $t0$. Positive values of $\mathrm{Inv_{change}}$ indicate erosion, whereas negative values stand for deposition.

2.2 A decision tree to assess the suitability of reference sites

We evaluated the suitability of the reference sites by analysing, in addition to the spatial variability, the temporal variation in the ^{137}Cs inventory. Given the assumption that no additional deposition of ^{137}Cs occurred at the sites during the investigated time window (which is valid worldwide except for the areas affected by the Fukushima Daiichi fallout), any temporal variation in the ^{137}Cs content should be attributable to different forms of soil disturbance or artefacts in the preparation or measurement of the samples. The potential causes of the spatial and temporal variation in the ^{137}Cs total inventories and depth profiles are examined through a decision tree which includes three main nodes (Fig. 2).

2.2.1 Node 1: spatial variation in FRN total inventory

Firstly, the spatial variation in the ^{137}Cs total inventory at each reference site is tested. Ideally, several replicates have been collected. If the coefficient of variation (CV) exceeds 35 % as suggested by Sutherland (1996), this could be a sign of unsuitability of the reference site, but it leaves the possibility of (i) increasing sampling numbers, (ii) analysing the causes of the spatial variation (see CheSS A to D) and (iii) moving to nodes 2 and 3 in CheSS.

2.2.2 Node 2: variation in the ^{137}Cs depth profile

Secondly, whether there is a significant variation between the ^{137}Cs depth profiles measured as spatial or temporal (in $t0$ and $t1$) replicates is tested. In theory, at a stable site the shape of the depth profile should not change between replicates. Consequently, a regression between the FRN activity depth profiles collected as spatial or temporal replicates should follow a 1 : 1 line, and the variability should lie within the range of the observed spatial uncertainty (node 1). A deviation of the linear regression coefficient from the 1 : 1 line in combination with high residues and low R^2 values ($< 0.5\,R^2$) indicates an immediate and significant change in the profile, which is typically caused by anthropogenic disturbance. For the FRN application at ploughed sites, the reference site might still be considered appropriate if the total inventory is not affected because conversion models used for ploughed sites are less sensitive to the shape of the FRN depth distribution. For unploughed soils, again the analysis of causes A to D might help in understanding the causes of the variability. Alternative options would be to take temporal replicates to evaluate the stability and thus the suitability of the reference site (node 3).

2.2.3 Node 3: temporal variation in FRN total inventory

If the CV of all replicates taken in $t0$ and $t1$ is $< 35\,\%$, the reference site might be used for the FRN method. The longer the time period between the first and second sampling, the more reliable the yielded assessments. A suitable test for significant differences should confirm or reject the hypothesis of ^{137}Cs total inventory stability over time. If the potential causes of variation (A to D) do not apply, the site is not suitable for the traditional FRN approach, but a repeated sampling approach could still be used to assess soil redistribution rates based on FRN methods (Porto et al., 2014; Kachanoski and de Jong, 1984).

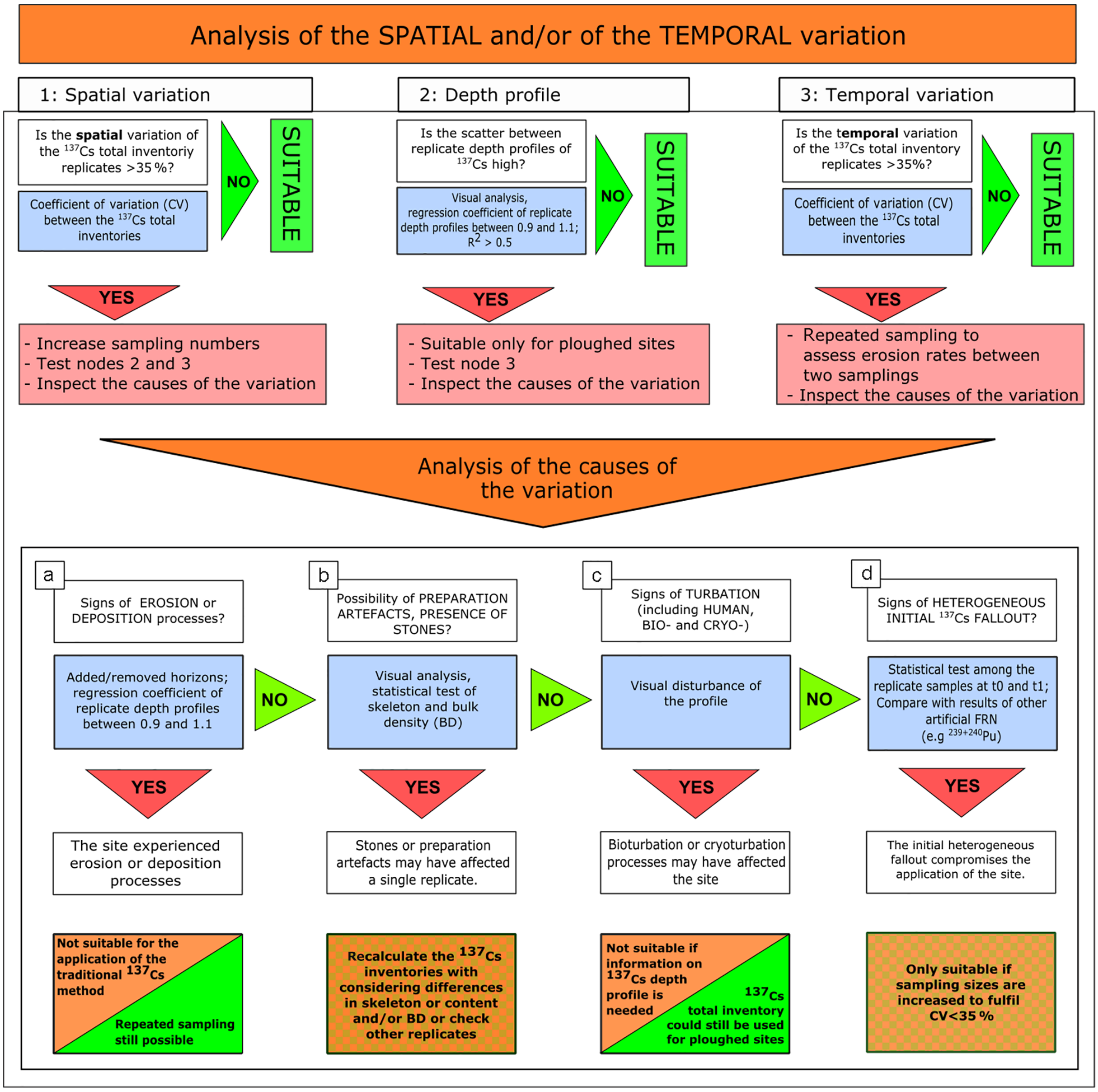


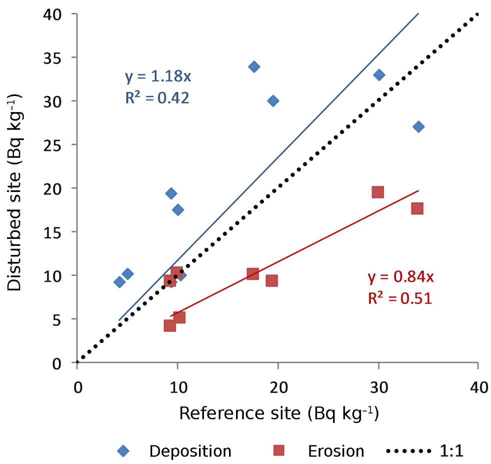
Figure 2. The CheSS decision tree to evaluate the suitability of a reference site for using ^{137}Cs as a soil erosion tracer.

2.2.4 Signs of disturbance associated with erosion and deposition processes (A)

A variation in the ^{137}Cs depth profile may have been caused by soil movement processes affecting the site (Fig. 2a). If the site experienced a loss of soil due to erosion, we expect to observe a removal of the top soil layers of the profile measured, for instance during the second sampling campaign (Fig. 3; red values below the reference profile). Further, the regression coefficient of the reference site that was affected by erosion will tend to be < 0.9 when plotted against a suitable reference profile or (for node 3) the reference profile before the disturbance (Fig. 3). In the case of deposition, a sedimentation layer should be found on the top of the reference depth profile, assuming that no ploughing operations affected the site (Fig. 3; red values above the reference profile). In this case, the regression coefficient will be > 1.1. Information on the depth distribution of another FRN might provide additional reliable confirmation. If redistribution processes are confirmed, the site is not suitable as a reference site and another location or a repeated FRN sampling approach to estimate erosion rates between the two sampling campaign should be considered (Kachanoski and de Jong, 1984).

2.2.5 Sampling or preparation artefacts (B)

One very common artefact which might bias the comparison between the samples collected at different sites or at $t0$ and $t1$ is the difference in the skeleton content (the percentage of soil fractions > 2 mm; Fig. 2b). The presence of stones might



Reference Bq kg^{-1}	Deposition Bq kg^{-1}	Erosion Bq kg^{-1}
34	27	17.6
30	33	19.4
17.6	34	10
19.4	30	9.3
10	17.6	10.2
9.3	19.4	9.3
10.2	10	5
9.3	9.3	4.2
5	10.2	1
4.2	9.3	1

Figure 3. Hypothetical signs of sheet erosion (red) and deposition (blue) on a depth profile compared to an undisturbed site.

determine passways for water and very fine particles and solutes in the soil and thus influence the accumulation and migration of ^{137}Cs through the soil layers. As ^{137}Cs reaches the soil by fallout from the atmosphere, the common shape of the ^{137}Cs distribution along the undisturbed depth profile can be described by an exponential function with the highest ^{137}Cs concentrations located in the uppermost soil layers (Mabit et al., 2008; Walling et al., 2002). This is particularly the case for soils with a low skeleton content (Fig. 4a) since the presence of stones may affect ^{137}Cs depth distribution either through (i) impeding the ^{137}Cs downward migration (^{137}Cs activity could then be concentrated in the layer above the stone; Fig. 4b) or (ii) creating macropores and micropores, favouring the ^{137}Cs associated with fine particles to "migrate" to deeper layers (Fig. 4c) or causing lateral movement which will induce a lower ^{137}Cs content in our samples.

As such, the seemingly spatial or temporal variation in the depth profile might indeed be a spatial variation induced by differences in skeleton content and/or bulk densities. Higher bulk densities will result in higher increment inventories even if ^{137}Cs activities at the layers are comparable. Thus, a thorough control (eventually through a statistical test, such as a paired t test) of whether skeleton content and bulk densities are comparable between replicates is suggested. Finally, sampling, preparation artefacts and measuring processes may produce various sources of error between different sites and years. The latter is especially the case if different people prepare the samples. An estimation of possible errors might be considered, for example through a simulation of different increment assignment along the profile. If different detectors or different calibration sources and/or geometry are used in the two sampling campaigns, a comparability check of the measurements is advisable. For instance, a subset of samples could be measured with the two different detectors, and any potential discrepancy in the results should be properly reported.

2.2.6 Signs of soil disturbance (C)

Different forms of disturbance, such as bioturbation, cryoturbation or even human-induced soil perturbation (e.g. tillage, seedbed preparation or digging), might have influenced the ^{137}Cs depth distribution between different sites and $t0$ and $t1$ (Fig. 2c). Occurrences of turbation are often difficult to identify prior to sampling but might eventually be detected by using other tracing approaches, such as the δ^{13}C depth distribution (Meusburger et al., 2013; Schaub and Alewell, 2009). In the case of turbation, the shape of the depth profile will be highly variable and should not be considered in the estimation of soil redistribution rates for unploughed soils. Nonetheless, the total inventory of ^{137}Cs at a ploughed site could still be used in combination with simple and basic mathematical conversion models, such as the proportional model (Ritchie and McHenry, 1990; IAEA, 2014), which require information only about the total reference inventory of ^{137}Cs and do not need detailed information about the ^{137}Cs depth distribution.

2.2.7 Signs of a heterogeneous initial fallout of ^{137}Cs over the area (D)

Finally, a significant difference between reference replicates may be caused by high small-scale spatial variability in ^{137}Cs distribution at the site due to heterogeneous initial fallout over the study area (Fig. 2d). In Europe, significant small-scale variability in ^{137}Cs distribution is known to be due to the Chernobyl fallout, which was characterized by high ^{137}Cs deposition associated with few rain events. Compared to nuclear bomb test fallout, the Chernobyl fallout was significantly more heterogeneous (e.g. Alewell et al., 2014). Therefore, in the areas affected by the Chernobyl fallout, sites sampled closely to each other may present very different ^{137}Cs contents. It is therefore necessary to investigate the small-scale spatial variability (e.g. the same scale as distance be-

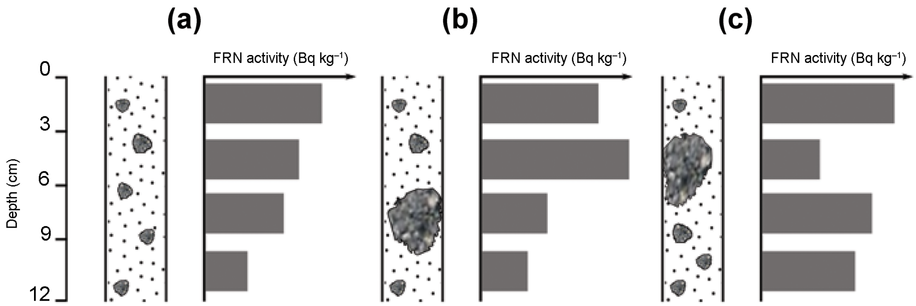


Figure 4. Possible influence of stones on the FRN depth distribution.

tween reference site replicates) measured at both or at least one sampling campaign by looking at the CV again, as presented in the previous sections, or through a statistical test (for example, the analysis of variance, ANOVA). If the spatial variability is highly significant, the site should not be envisaged as a reference site for the application of the ^{137}Cs method unless the number of samples collected for the determination of the reference baseline is large enough (at least 10) to counterweight the small-scale variability within the site (Mabit et al., 2012; Sutherland, 1996; Kirchner, 2013). A possible validation of this cause of heterogeneity might be a comparison with the spatial distribution of another FRN, such as $^{239+240}Pu$ or $^{210}Pb_{ex}$ (Porto et al., 2013; Fig. 2d). As the fallout deposition of $^{239+240}Pu$ after the Chernobyl accident was confined to a restricted area in the vicinity of the nuclear power plant (Ketterer et al., 2004), the origin of plutonium fallout in the rest of Europe is linked to the past nuclear bomb tests only. Consequently, the Pu fallout distribution was more homogeneous (Alewell et al., 2014; Ketterer et al., 2004; Zollinger et al., 2015). If the $^{239+240}Pu$ depth profiles do not vary significantly between the two sampling years, there should be no disturbance (e.g. turbation, erosion) or measurement artefacts. As such, it might be concluded that the heterogeneous deposition of ^{137}Cs at the time of the fallout prejudices the use of Cs at this site.

3 The application of the CheSS decision tree

3.1 Study area

To test the methodology described above, we used a data set from an alpine study area, the Urseren Valley (30 km^2) in Central Switzerland (Canton Uri), which has an elevation ranging from 1440 to 3200 m a.s.l. At the valley bottom (1442 m a.s.l.), the average annual air temperature for the years 1980–2012 is around 4.1 ± 0.7 °C and the mean annual precipitation is 1457 ± 290 mm with 30 % falling as snow (MeteoSwiss). The U-shaped valley is snow covered from November to April. On the slopes, pasture is the dominant land use, whereas hayfields are prevalent near the valley bottom.

3.2 Sampling design

Supportive information was provided by the local landowners to select the reference sites in both valleys. Sites used for ploughing and grazing activities were excluded. A first sampling campaign was undertaken in autumn 2010 for $^{239+240}Pu$ and in 2013 for ^{137}Cs. Six reference sites (REF1 to REF6) were identified in flat and undisturbed areas along the valley. At each site, three cores (40 cm of depth) 1 m apart from each other were sampled. The cores were cut in 3 cm increments to derive information on the ^{137}Cs depth profile. The three cores from each site were bulked to provide one composite sample per site. During the second sampling campaign in spring 2015, all six reference sites were resampled. Considering the typical and high soil redistribution dynamics of the valley of > 1 cm per year caused by snow-induced soil removal (Meusburger et al., 2014), the time span is sufficiently long to ensure the possibility to observe changes in the depth profiles if soil erosion and deposition processes affected the area. At each site, we collected three replicates, which were analysed separately, to investigate the small-scale variability in the FRN content. All cores were air-dried (40 °C for 72 h) and sieved (< 2 mm) to remove coarse particles; the skeleton content and the bulk density (BD) were determined.

3.3 Measurement of anthropogenic FRN activities and inventories

The measurements of the ^{137}Cs activity (Bq kg^{-1}) were performed with high-resolution HPGe detectors. The ^{137}Cs activity (Bq kg^{-1}) from 2013 was analysed at the Institute of Physics at the University of Basel using a coaxial high-resolution germanium lithium detector (Princeton Gamma Tech) with a relative efficiency of 19 % (at 1.33 MeV; ^{60}Co). Counting time was set to 24 h per sample. Samples collected in 2015 were analysed at the State Laboratory Basel-City using coaxial high-resolution germanium detectors with 25 to 50 % relative efficiencies (at 1.33 MeV; ^{60}Co). Counting times were set to provide a precision of less than ± 10 % for ^{137}Cs at the 95 % level of confidence.

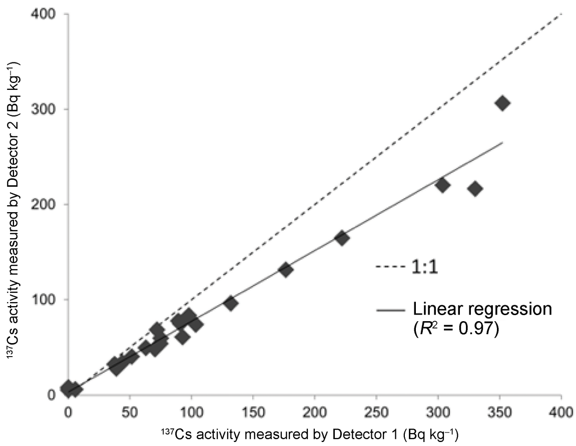


Figure 5. The comparison between the ^{137}Cs measurements of a subset of samples ($n = 16$) performed with two different HPGe detectors; detector 1 is housed by the Physics Department at the University of Basel (CH) and detector 2 is housed by the State Laboratory Basel-City (CH).

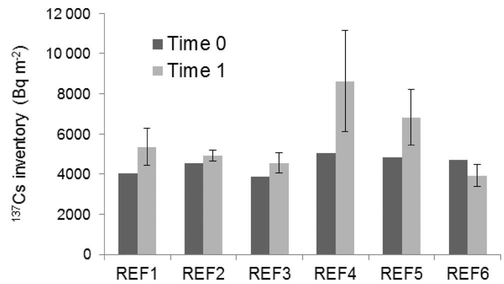


Figure 6. Temporal variation between the total ^{137}Cs inventories measured at the reference sites in the Urseren Valley; time 0 = 2013 and time 1 = 2015. The error bars indicate the standard deviations of the inventories among the replicates collected at each reference site in 2015.

All soil samples were counted in sealed discs (65 mm diameter, 12 mm height, 32 cm^3) and the measurements were corrected for sample density and potential radioactivity background. The detectors located at the State Laboratory Basel-City were calibrated with a reference solution of the same geometry. The reference contained ^{152}Eu and ^{241}Am (2.6 kBq rsp. 7.7 kBq) to calibrate the detectors from 60 to 1765 keV. It was obtained from the Czech Metrology Institute, Prague. This solution was bound in silicon resin with a density of 1.0. The efficiency functions were corrected for coincidence summing of the ^{152}Eu lines using a Monte Carlo simulation program (Gespecor). The ^{137}Cs was counted at 662 keV with an emission probability of 0.85 and a (detector) resolution of 1.3 to 1.6 keV (FWHM). All measurements and calculations were performed with the gamma software Interwinner 7. The ^{137}Cs activity measurements were all decay corrected to the year 2015.

To compare the ^{137}Cs results to another artificial FRN, all samples were also measured for $^{239+240}$Pu activity. The determination of plutonium isotopes from both valleys and for both sampling years was performed using a Thermo X Series II quadrupole ICP-MS at Northern Arizona University, USA. A detailed description of the ICP-MS specifications and sample preparation procedure can be found in Alewell et al. (2014). The activities of ^{137}Cs and $^{239+240}$Pu (act, Bq kg^{-1}) were converted into inventories (Bq m^{-2}) according to Eq. (1).

3.4 Application of the CheSS decision support tool to the reference sites

Because the ^{137}Cs activity of the samples was measured with different detectors for the two sampling years, we investigated the potential variability between the two detectors. A selected subset of samples ($n = 24$) was analysed using both detectors (one located at the Institute of Physics at the University of Basel and the other located at the State Laboratory Basel-City). The results highlight a strong correspondence between the measurements by the two analytical systems ($R^2 = 0.97$; $p < 0.005$); however, the detector at the State Laboratory Basel-City returned slightly lower ^{137}Cs activities (Fig. 5). Thus, the ^{137}Cs activities of the samples measured in 2013 were corrected to the values of the detector at the State Laboratory Basel-City (which has a higher efficiency) to allow for comparability between the different data sets.

Total ^{137}Cs inventories (decay corrected to the year 2015) of the six reference sites collected in the Urseren Valley in 2013 range from 3858 to 5057 Bq m^{-2} with a mean value of 4515 Bq m^{-2} and a standard deviation (SD) of 468 Bq m^{-2}. Data from 2015 range between 3925 and 8619 Bq m^{-2} with a mean value of 5701 Bq m^{-2} and a SD of 1730 Bq m^{-2} (Fig. 6).

When following the CheSS decision tree, we investigated the variation in the ^{137}Cs total inventories at each reference site (node 1). The replicate samples were analysed separately only during the second sampling campaign ($t1$), while during the first sampling campaign ($t0$) only composite samples were analysed. Reference sites REF3, REF5 and REF6 presented signs of high small-scale variability, as expressed by a CV of 48 %. Such variability excluded them from any further application as reference sites without subsequent additional sampling. For sites REF1, REF2 and REF4, the CV was between 19 and 31 %.

Passing to node 2 of the CheSS decision tree, the analysis focuses on the variation in the shape of the ^{137}Cs depth profile (Fig. 7). Here we examined the regression between the reference depth profiles in $t0$ and $t1$. For the three sites with acceptable spatial variability (i.e. reference sites 1, 2 and 4), the site REF4 shows signs of deposition with a regression coefficient between $t0$ and $t1 = 1.34$. The deposition was confirmed by field observations of construction work conducted between the two samplings. After this disturbance the site is

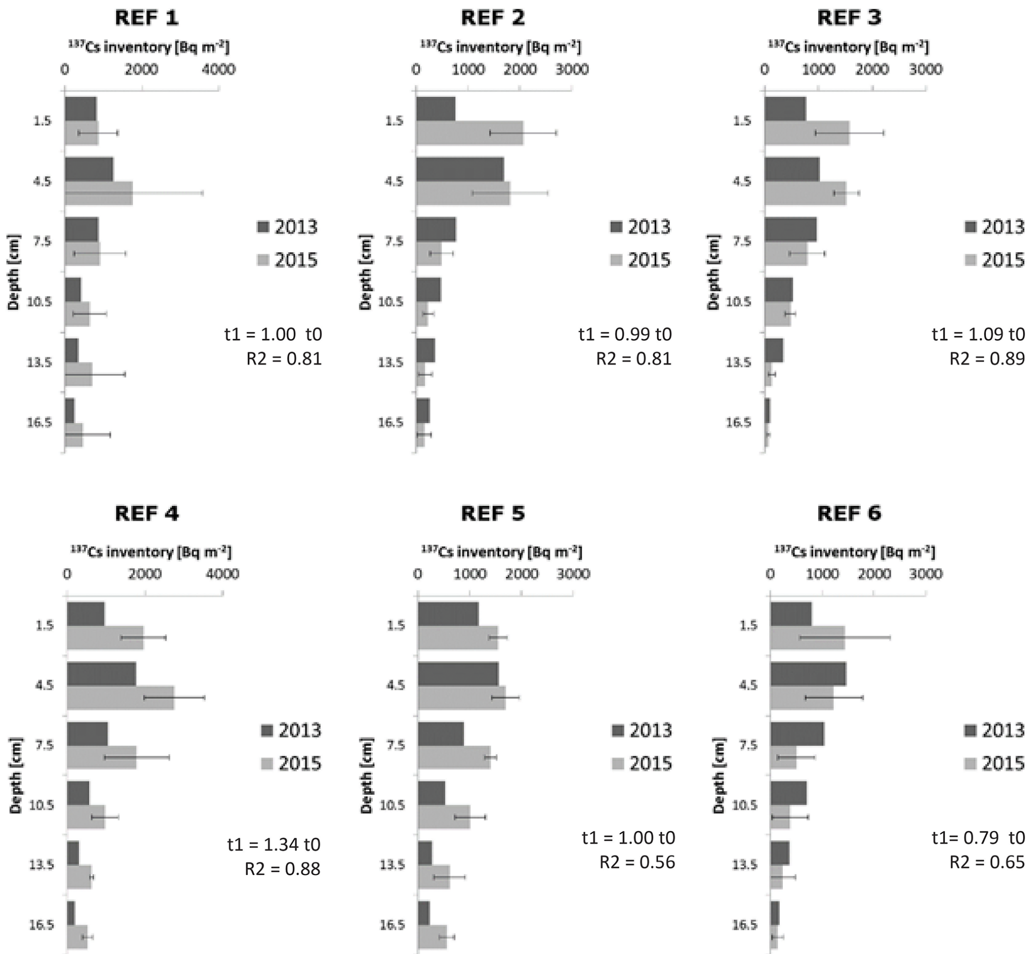


Figure 7. The ^{137}Cs depth profiles of the six investigated reference sites in the Urseren Valley for the two different sampling campaigns. The error bars indicate the standard deviations of the inventories among the replicates collected at each reference site 2015. The regression equation between the depth profile at $t0$ and $t1$ is displayed together with the R^2.

no longer a suitable reference site. Among the sites with high spatial variability, the site REF6 showed signs of erosion with a regression coefficient between $t0$ and $t1 = 0.79$.

In node 3 the temporal differences in total inventories between $t0$ and $t1$ were assessed. Here only the site REF4 showed a significant difference in the total ^{137}Cs inventories between $t0$ and $t1$, thus confirming the unsuitability of the site after the construction work.

To further investigate the causes of the spatial variation, $^{239+240}$Pu inventories measured at the three replicates of each site were analysed for $t0 = 2010$ and $t1 = 2015$ (Fig. 8). Clearly, deposition for REF4 and erosion processes for REF6 were confirmed with an increase of 46 % and a decrease of 27 % in the total $^{239+240}$Pu inventory between $t0$ and $t1$, respectively.

Further, we looked at the differences in the skeleton content of the three replicate samples collected at $t1$ (Fig. 2b). For site REF1, an ANOVA test showed a significant difference (p value of 0.025), and thus a difference in the pres-

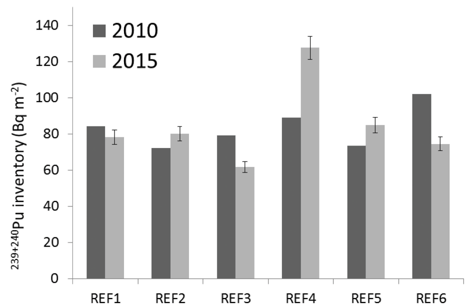


Figure 8. Temporal variation between the total $^{239+240}$Pu inventories measured at the reference sites in the Urseren Valley; time 0 = 2010 and time 1 = 2015. The error bars indicate the standard deviations of the inventories among the replicates collected at each reference site in 2015.

ence of stones in the three soil cores might have affected the FRN depth distribution. In particular, a Tukey's HSD (honest significant difference) post-hoc pairwise comparison identified the replicate number 3 at REF1 as a potential outlier. To validate the suitability of REF1, more replicates should be collected and measured in order to compare their ^{137}Cs depth profiles to the results obtained during the first sampling campaign. In summary, only the reference site REF2 appeared to be suitable for ^{137}Cs-based studies. For the site REF4, the construction work precluded its suitability for further application as a reference site. Form visual inspection of the soil profile, B could exclude cause C and consequently the final cause of heterogeneous fallout with high spatial variability (D) applies for the sites REF3 and REF5. These sites may be suitable for other FRNs or for ^{137}Cs if more samples are collected to constrain the spatial heterogeneity that was introduced by the ^{137}Cs Chernobyl fallout.

4 Conclusion

With the decision tree CheSS, a support tool to verify the suitability of reference sites for a ^{137}Cs-based soil erosion assessment is presented. Great attention has to be given to analysis of the small-scale variability in ^{137}Cs distribution in the reference areas, especially in regions affected by nuclear accident fallout. To cope with small-scale variability, sampling numbers might be increased or the temporal variation in ^{137}Cs or another radionuclide, such as $^{239+240}$Pu, might be analysed. The CheSS test in the Urseren Valley indicated that the heterogeneity and disturbance of ^{137}Cs distribution prejudiced the suitability of some reference sites. Additionally, the presence of stones affected the shapes of the depth profiles in at least one replicate sample at reference site 1. Including unsuitable reference sites, the application of the traditional ^{137}Cs approach, based on a spatial comparison between reference and sampling sites, is compromised. To derive soil redistribution rates, a ^{137}Cs repeated sampling approach should be preferred. This approach is based on a temporal comparison of the FRN inventories measured at the same site in different times (Kachanoski and de Jong, 1984). It does not require the selection of reference sites because the inventory documented by the initial sampling campaign is used as the reference inventory for that point (Porto et al., 2014).

Accurate soil erosion assessment is crucially needed to validate soil erosion modelling, which can help prevent and mitigate soil losses on larger spatial scales. In this context, FRN could play a decisive role if we are able to overcome its potential pitfalls, especially related to the selection of suitable reference sites. The decision tree CheSS provides a concept for objective and comparable reference site testing, which enables the exclusion of sites which present signs of uncertainty. We are convinced that this can contribute to improving the reliability of FRN-based soil erosion assessments.

Data availability. The data in this paper can be found in the Supplement.

The Supplement related to this article is available online at https://doi.org/10.5194/soil-3-113-2017-supplement.

Author contributions. LA, KM, LM and CA designed the concept of the method and analysed the data. AB contributed to the collection and preparation of the soil samples and to the analysis of the data. MZ measured the ^{137}Cs activity in the soil samples and analysed the results. MEK measured the $^{239+240}$Pu activity in the soil samples. LA prepared the paper with contributions from all coauthors.

Competing interests. The authors declare that they have no conflict of interest.

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