Dear Editor,

We received two very positive reviews with some excellent suggestions to improve the manuscript and the CheSS method giving it a more general relevance, since the suitability of references site is crucial, maybe even the most crucial step, in all FRN based erosion assessments.

The reviewers identified three main concerns regarding the proposed CheSS method. The first major point is that the approach might be site specific and not of general applicability. As respond to this point we included the spatial variability in a modified version of the decision tree (Figure 2) as such CheSS will be also applicable without temporal replicates.
The second point was related to the time lag needed for the resampling approach. In general time span should be of sufficient length to cause an inventory change that it larger than the uncertainty related to the inventory assessment and the small scale variability e.g. larger 35%. We added information on this point in the ms. Moreover, we modified CheSS in a way that it is not mandatory to have temporal replicates.
The final point was related to the criteria of the variability of the FRN depth profile. With the help of the example data of reviewer to we could establish a less strict data driven criteria, which in addition also allows to assess erosion and deposition processes (Figure 3).

With the updated presented manuscript, we took care to answer the reviewer's questions and comments as well as your editorial request. We hope you will consider our new improved submission for publication. Please do not hesitate if you have any further questions or concerns.

Looking forward to hearing from you.

Yours sincerely,
Dr. Katrin Meusburger on behalf of Dr. Laura Arata and the co-authors

Reply Anonymous Referee #1
In this paper the authors explored an important issue in the application of the 137Cs technique that relates to the choice of a reference site. They proposed a decision support tool (CheSS) to check the suitability of a reference site using repeated measurements of 137Cs undertaken in 2013 and 2015 on the same sites and measurements of 239+240Pu carried out in 2015. The basic assumption is that suitable reference sites are expected to present no significant temporal variation in their decay corrected 137Cs depth profiles. The authors individuated four main causes of possible variation in the inventory.
These are (1) small scale variability connected to the non-homogeneus fallout (see areas affected by Chernobyl), (2) signs of erosion and/or deposition, (3) artefacts due to sampling and measurements, (4) turbation processes. The authors screened their six reference sites in the Urseren Valley (Switzerland) based on this assumption and tried to individuate the suitable reference site. The paper seems to me very in-teresting and I think it should be published but some more details need to be added and/or discussed in this version.

Specific comments
Introduction – The authors explain briefly the basic assumptions of the 137Cs with the help of Fig. 1. I agree with the explainations reported in the text but I think Figure 1 is a little bit misleading for people that are not familiar with the technique.
In fact, what they depict as 'reference site (R)' is a valley bottom and, as is, it can be a depositional site. Also, what they depict as 'depositional site (D)' is a foot-slope and it may not necessarily be a 'depression' where deposition occurs. I suggest to redraw this figure in a more proper way (see my example below) Fig. 1 (modified)
Reply 1_1: Thanks you for this comment. The depiction of this Figure was clearly driven by a site-specific adaptation of the FRN-method to our alpine sites. In our area, we cannot sample ridges, since they are very elevated and consist of bare rock. So we selected a ridge of a moraine at the valley floor, which for simplification was not depicted in the original Figure 1. We agree that the modified version of reviewer 1 will be of more general applicability and will change Figure 1 as suggested.

2.1 Repeated sampling strategy and calculation of inventories

This is a very important point. More work must be done to establish how long the time period between two sampling campaigns should be. This depends on the 137Cs inventory of the reference site and on its spatial variability. If the inventory is low, it is difficult to understand if the difference between the two sampling campaigns should be attributed to decay or to erosion and/or deposition or to the detector efficiency. In this case, a period of at least 10-15 years could be necessary. If the inventory is high and it is affected by the Chernobyl fallout, we could expect that the small scale spatial variability and the temporal variability are of the same order of magnitude and it is difficult to distinguish the relative contributions. Something like that is suggested by the data that the authors show in their Fig. 6. The inventory provided in 2015 are all higher than those obtained two years before (in 2013) with the only exception of Ref 6. This is not unexpected because it is not possible to relocate exactly the same sampling points. Clearly, more samples are necessary in this case. But also, a time period of two years between two sampling campaigns could not be enough. The authors can add some comments here.

Reply 1_2: We will add some more discussion on the time lag needed between the repeated sampling. For sure there is no general advice and it will depend on the type of disturbance. An anthropogenic or animal disturbance can cause an immediate and significant change of the inventory. If the applicant also seeks to identify significant changes of the inventory due to erosion or deposition the time span should be of sufficient length to cause an inventory change that it larger than the uncertainty related to the inventory assessment e.g. larger 35%. For instance, in our case we have 700 Bq/m2 in the upper 3cm. We would need to loss or gain one third (1cm) of the soil to induce a significant change of the inventory. Assuming a bulk density of 0.5 g cm^{-3} this would correspond to 25 t $ha^{-1}yr^{-1}$ for the selected time lag of two years. We added a discussion on that point in line 98-101.

Node 2: No significant temporal variation of the 137Cs depth profile

I agree with the test related to the total inventory as explained in Node 1. However, I found node 2 too severe. I agree that the shape of the reference profile is important but, I think the test should be done on the entire profile not on the single layers. In many years of experience, I have never seen two profiles collected in the same site being identical. Maybe a practicle example can clarify my thoughts. Below there are 3 potential reference profiles characterised by the same total inventory (2510 Bq m-2), so they passed Node 1. They can be fitted by the same exponential model (same shape parameter h0 = 70 kg m-2 and same surface concentration A0 = 35 Bq kg-1). The values of cesium activity and mass depth for each layer are reported below.

Figure (My example)

Using the Sutherland range as a test (see suggestions in Node 2), in 5 cases out of 10 (see my values in red) the CV is greater than 35% (which is the upper limit suggested by Sutherland). If I have understood well, this result would suggest that the site where these profiles have been obtained is not suitable as reference site. I do not think I can agree with that because they show the same exponential decline with depth, and the difference between each single layer can be attributed to other factors (the authors mentioned some of the other causes later in the paper). On the contrary, if we use a t-test or other statistical tests to compare mean and variance of these three profiles, I may be wrong, but I did not find any statistical difference. I think the authors should think about it and add some comments.

There is another limitation in the application of this procedure suggested by the authors. In my example I have considered the same value of mass depth increment for the 3 profiles. This is an ideal case. In reality, due to differences in soil type, land use, presence of stones etc., it is difficult to obtain equal values of mass depth for the corresponding layers of different profiles. This makes this comparison not possible. In the end, I find more useful to check the shape of the entire profile.

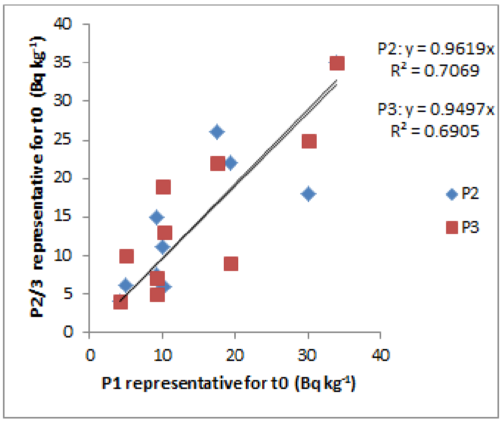
Reply 1_3: Since the shape of the FRN depth profile is decisive in many conversion models, we consider it essential to retain a node to evaluate the shape in CheSS. However, we agree with reviewer 1 that the chosen criteria was too selective. With the help of the valuable example data supplied by the reviewer we modified the criteria of node 2. Instead of using a threshold value of the CV, we suggest to plot the depth profile of t0 against t1. If the shape of the profiles remained over time, the regression between the two depth profiles should follow a 1:1 line and the R2 >0.5. Taking into account that part of the variability may be explained by small scale variability of soil properties and adsorbed FRNs as well as procedural imprecision.

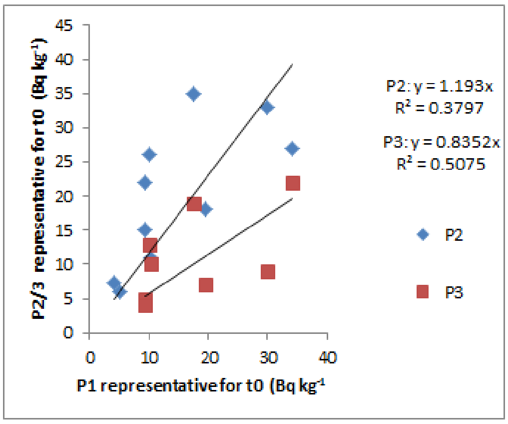
Node 4 - Signs of disturbance associated with erosion and deposition processes I agree with the explaination in the text but, Figure 3 shows only the case where the 137Cs profile is perfectly exponential. In many places, profiles obtained in reference sites show a peak below the surface due to migration

processes downward. In this case, the profile shown in figure 3b can be an undisturbed reference profile and, consequently, sheet erosion and deposition processes modify the shape accordingly. I suggest to improve their figure 3 considering both the possible cases (see my example below).
Fig. 3 (modified)
Reply 1_4: We agree with the concern of reviewer 1 in case of an none exponential depth distribution and thought about a data based criteria in addition to visual assessment to identify erosion or deposition processes between the two time steps. The regression equation established for the modified node 2 offers such a databased decision support. In case of accumulation the gradient of the trend line will be larger 1.1 and in case of erosion <0.9. We replaced Figure 3 with a new graph displaying this data driven approach (Figure 3, line 165).
Please find below how your example data performs for the modified criteria of node 2&4:

Node2		
P1	P2	P3
34	35	35
30	18	25
17.6	26	22
19.4	22	9
10	11	19
9.3	15	7
10.2	6	13
9.3	7.4	5
5	6.1	10
4.2	4	4

Node 4	deposition	erosion
P1	P2	P3
34	27	22
30	33	9
17.6	35	19
19.4	18	7
10	26	13
9.3	22	5
10.2	11	10
9.3	15	4
5	6	1
4.2	7.4	1





Page 7 – Line 21-22 – The authors say 'If information on the depth distribution of another FRN is available, this might provide a reliable confirmation'. I agree with this statement, but an example is necessary. The use of 210Pbex proved to be very effective in combination with or in alternative to 137Cs. In fact, good relationships exist between the results obtained with 137Cs and 210Pbex. I am sure the authors want to add some comments here maybe recalling some of the works done in this field (see for example Porto et al., 2006; 2013). Porto P., Walling D.E., Callegari G. and Catona F. (2006). Using fallout lead-210 measurements to estimate soil erosion in three small catchments in Southern Italy. Water, air and soil pollution: focus 6, 657-667 Porto P., Walling D.E., Callegari G. (2013). Using 137Cs and 210Pbex measurements to investigate the sediment budget of a small forested catchment in Southern Italy. Hydrological Processes 27(6), 795-806.
Reply 1_5: Indeed this is another good example, how other FRNs can underpin the selection of a suitable reference site. We included further discussion and references in line 214.

Reply Referee 2

The paper addressed a very important topic in the soil erosion evaluation using Cs-137 technique. A good orientation about the reference site choice will define a different history about the soil erosion and deposition rates in the end. In my opinion the MS give us a good understanding about this and the complexity associate. In my opinion, the discussion about the commitment about the reference site is meaningful to the scientific community to reveal the uncertainties about it and to help to establish a protocol. However, I think the protocol will be site specific.

In general terms, it is quite difficult to establish a protocol to choose the reference value. I agree with the arguments and factors explored by the authors, but I'm not sure if the protocol suggested is the main contribution of the MS or even the application of this with the study of case showed. In my point of view the main contribution is the discussion about the control factor and the uncertainty associated. Maybe the paper should be written more theoretical and with less pretension to establish a protocol applicable worldwide or to prove its application.

Reply 2_1: We can understand the concern of the reviewer, that a detailed protocol maybe too site-specific and may impede the application or adaptation of the reference site selection process for other areas. We will follow his/her suggestion in order to shift the focus of the ms away from a detailed protocol towards a theoretical concept.

Another question/doubt is about the temporal variation. It was not so clear if the authors highly recommend a temporal analysis or no. If yes, how much time it takes to researchers decide if this is a good site to be used. Is this a constrain about the methodology proposed? Maybe the author could explore the fact the temporal evaluation will take a long time and maybe people will not be able to test it.

Reply 2_2: So far, we did not provide a specific suggestion when temporal reference analysis could be beneficial and how long the time lag between the sampling should be. The time lag, as detailed in the reply R1_2 to reviewer 1, will depend on various conditions e.g. spatial variation, measurement uncertainty. In answer to the concern of reviewer 2 we will be more specific on this point and added to the existing paper in line 98-101.

In my point of view the spatial variation is more pertinent and easy to be applicable. In the abstract the temporal variability is highlighted, for example, maybe the analysis could start with spatial variability and after the authors could show some insights about the temporal analysis.

R2_3: Thanks for this very good idea. We implemented the spatial approach in the abstract and implemented the spatial approach in CheSS (Figure 2) to make the concept more general applicable.

Figure 1: Because this MS is proposing a reflexing/protocol, in my opinion the reference site should be chosen in a flat area in the top instead in the base of hillslope, for example, in a plateau without erosion/deposition possibilities.

R2_4: As explained above modifed Figure 1, displaying the reference site in a flat part of the ridge.

Beside this, Maybe we can come back to the form and structure after the discussion about the points presented above.

Best regards

R2_5: We are sorry not to have replied in time to this comment because of the maternity leave of the corresponding author.

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

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Abstract. The classical approach to use ^{137}Cs as soil erosion tracer is based on the comparison between stable reference sites and sites affected by soil redistribution processes, and enables to derive 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) ~~for systematic validation or rejection of reference sites~~. Commonly the variation among ^{137}Cs inventories of spatial replicate reference samples are taken as sole criteria to decide on the suitability of a reference inventory. Here we propose an extension of this procedure using ~~The method is based on~~ a repeated sampling approach, where 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 ~~temporal~~ variation are assessed by a decision tree. More specifically, the decision tree tests for (i) uncertainty connected to small scale variability of ^{137}Cs due to its heterogeneous initial fallout (such as in areas affected by the Chernobyl fallout), (ii) signs of erosion/deposition processes, (iii) artefacts due to the collection, preparation and measurement of the samples and (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 rock falls). CheSS was exemplarily applied ~~has been tested~~ in one Swiss alpine valley, where the apparent temporal variability ~~suitability~~ ~~of six reference sites~~ was ~~tested~~~~in~~ questioning the suitability of selected reference sites. In general we suggest the application of CheSS to implement first steps towards a comprehensible approach to test for the suitability of reference sites.

Keywords: FRN, fallout radionuclides, soil degradation, $^{210}Pb_{ex}$, $^{239+240}Pu$, comparability of gamma spectrometers, Cesium-137

1 Introduction

Soil erosion is a global threat (Lal, 2003). Recent estimated erosion rates range from low rates of 0.001–2 t ha^{-1} yr^{-1} on flat relatively undisturbed lands (Patric, 2002) to high rates under intensive agricultural use of > 50 t ha^{-1} yr^{-1}. In mountainous regions, rates ranging from 1–30 t ha^{-1} yr^{-1} have been reported (e.g. Descroix et al. 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 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 soil erosion tracer allows 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 in the atmosphere during nuclear bomb tests and as a consequence of nuclear power plant (NPP) accidents such as Chernobyl in April 1986. It reached 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 in 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 lower ^{137}Cs inventory as compared to the reference site, and sediment deposition processes at sites with a greater ^{137}Cs inventory (Figure 1, A). Specific mathematical conversion models allow then to derive from the latter comparison quantitative estimates of soil erosion and deposition rates (IAEA, 2014).

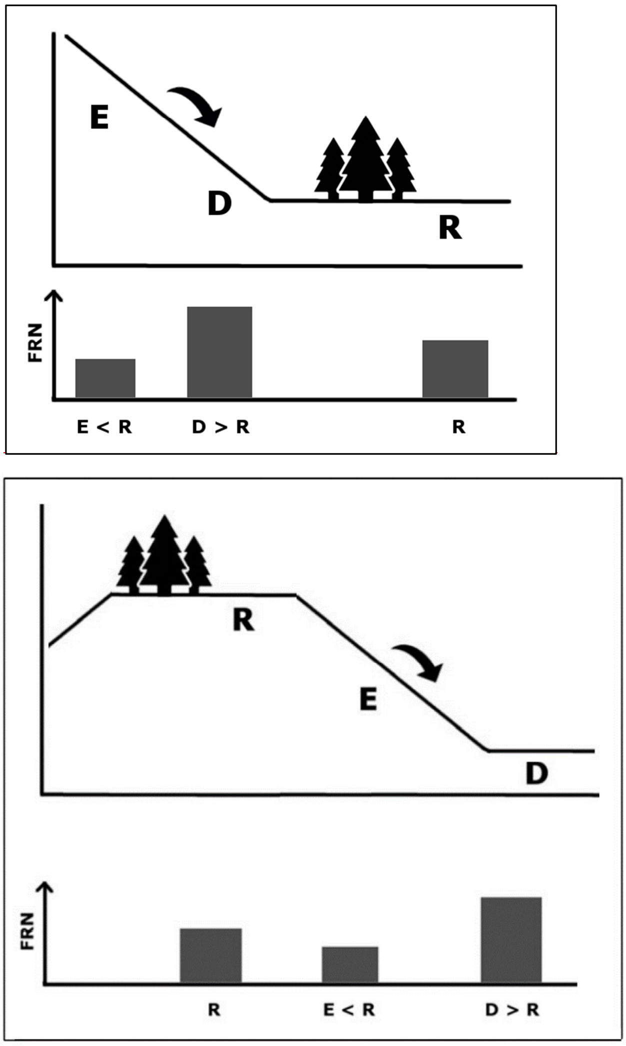

Figure 1: Concept of the fallout radionuclide (FRN) traditional method, in which the FRN content of a reference site located in a flat and undisturbed area (R) is compared to the FRN content of disturbed sites (E and D). If the FRN at the site under investigation is lower than at the reference site,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

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 changes to quantitative estimates of soil erosion rates (Walling et al., 2002). Recent studies 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).

A close proximity of a reference site to the area under investigation is required to meet the assumption that both experienced similar initial fallout. The latter is particularly important if the study area was strongly affected by Chernobyl fallout, which is, besides global fallout from nuclear weapons testing, the major input of ^{137}Cs 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 of ^{137}Cs distribution may be expected and, as already pointed out by Lettner et al.~~,~~ (1999) and Owens and Walling (1996), might impede the comparison between reference and sampling sites. To consider adequately the spatial variability of the FRN fallout, multiple reference sites should be selected and the variability within the sites properly tackled (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-, ~~-or~~ anthropogenic- ~~impact~~ 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 the gamma analysis might introduce a certain level of uncertainty, which should be carefully considered. For instance, Lettner et al. (1999) estimated that the preparation and measuring processes contribute 12.2% to the overall variability of the reference inventory. Guidance in form of independent indicators (e.g. stable isotopes as suggested by Meusburger et al., 2013) for the suitability of reference sites might help to assist with the selection of reference sites.

All in all the suitability or unsuitability of references site is crucial, maybe even the most crucial step, in all FRN based erosion assessments. The general suitability of ^{137}Cs based erosion assessment has been recently discussed very controversially (Parsons and Forster 2011, 2013; Mabit et al., 2013). We would like to propose that the FRN community needs to agree on general concepts and sampling strategies to test the suitability of reference sites in order to improve the method as well as establish trust in this useful erosion assessment method. Up to now, t~~T~~he variability among spatial replicate samples at reference sites are commonly the sole criteria to decide on the suitability of a reference value. We propose an ~~alternative~~ extended method to Check the Suitability of reference Sites (CheSS) using a repeated sampling strategy and as such an assessment of the temporal variability of reference sites. ~~Our basic assumption is that decay corrected ^{137}Cs depth profiles measured in two points in time should be identical.~~ The suitability of reference sites for an accurate application of ^{137}Cs as soil erosion tracer is tested at ~~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 ~~proper~~ application of ^{137}Cs as 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 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 it larger than the uncertainty related to the inventory assessment e.g. larger 35%. In our study site being effected by anthropogenic disturbance and snow erosion of several mm per winter already 2 years can be considered sufficient (Meusburger et al., 2014). Several spatial repetitions following the suggestion of Sutherland et al. (1996) are necessary and should be ~~analyzed~~analysed separately to investigate the small scale variability of ^{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 one), considering the half-life of ^{137}Cs (30.17 years).

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

$$Inv = act \times xm \tag{1}$$

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

$$Inv_{change} = \frac{Inv_{t0} - Inv_{t1}}{Inv_{t0}} \times 100 \tag{2}$$

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

2.2 A decision tree to ~~identify~~ assess ~~possible pitfalls for~~ the suitability of reference sites

~~Our aim is to~~We evaluated ~~assess~~ the suitability of the reference sites by analyzing ~~for~~ in addition to the spatial variability the ~~a possible~~ temporal variation of 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 of the ^{137}Cs content should be attributable to different forms of soil disturbance or to artefacts in the preparation/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~~six~~ main nodes (Figure 2).

Node 1: Spatial~~temporal~~ variation of FRN~~the ^{137}Cs~~ total inventory

Firstly, the ~~temporal~~ spatial variation of the ^{137}Cs total inventory at each reference site is tested. Ideally, ~~in both sampling campaigns~~ 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 leaves the possibility of i) increasing sampling numbers, ii) analysing the causes for the spatial variation (see CheSS A to D) and iii) moving to node 2 and 3 in CheSS.

Node 2: V~~No significant temporal v~~ariation of the ^{137}Cs depth profile

Secondly, it is tested whether there is a significant ~~temporal~~ variation between the ^{137}Cs depth profiles measured as spatial or temporal (in *t0* and *t1*) replicates. In theory, at a stable site the shape of the depth profile should not ~~have~~ change~~d~~ between ~~t0 and t1~~replicates ~~if the site remained undisturbed~~. 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). ~~If replicate samples have been collected during the two sampling campaigns, a statistical test (for example the t-test or the Wilcoxon test) is run for the ^{137}Cs inventories of each depth increment between the groups of replicate samples collected in *t0* and *t1* (Figure 2, node 2).~~ 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 of the profile, which is typically caused by anthropogenic disturbance. For the FRN application at ploughed sites ~~If there is a significant difference between the shapes of the two depth profiles,~~ the reference site ~~should not~~might still be considered appropriate if the total inventory is not affected, ~~for the ^{137}Cs method application~~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 the causes A to D might help to understand the causes for the variability. Alternative options would be to take temporal replicates to evaluate the stability and thus suitability of the reference site (node 3).~~; Kachanoski & de Jong, 1984.~~

Node 3: Temporal variation of 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 is the more reliable the yielded assessments will be. Further a suitable test for significant differences should confirm or reject the hypothesis of ^{137}Cs total inventory stability over time. If the potential causes for variation (A to D) do not apply, the site is not suitable for the traditional FRN approach. Still a repeated sampling approach could be used to assess soil redistribution rates based on FRN methods (Porto et al., 2014; Kachanoski & de Jong, 1984).

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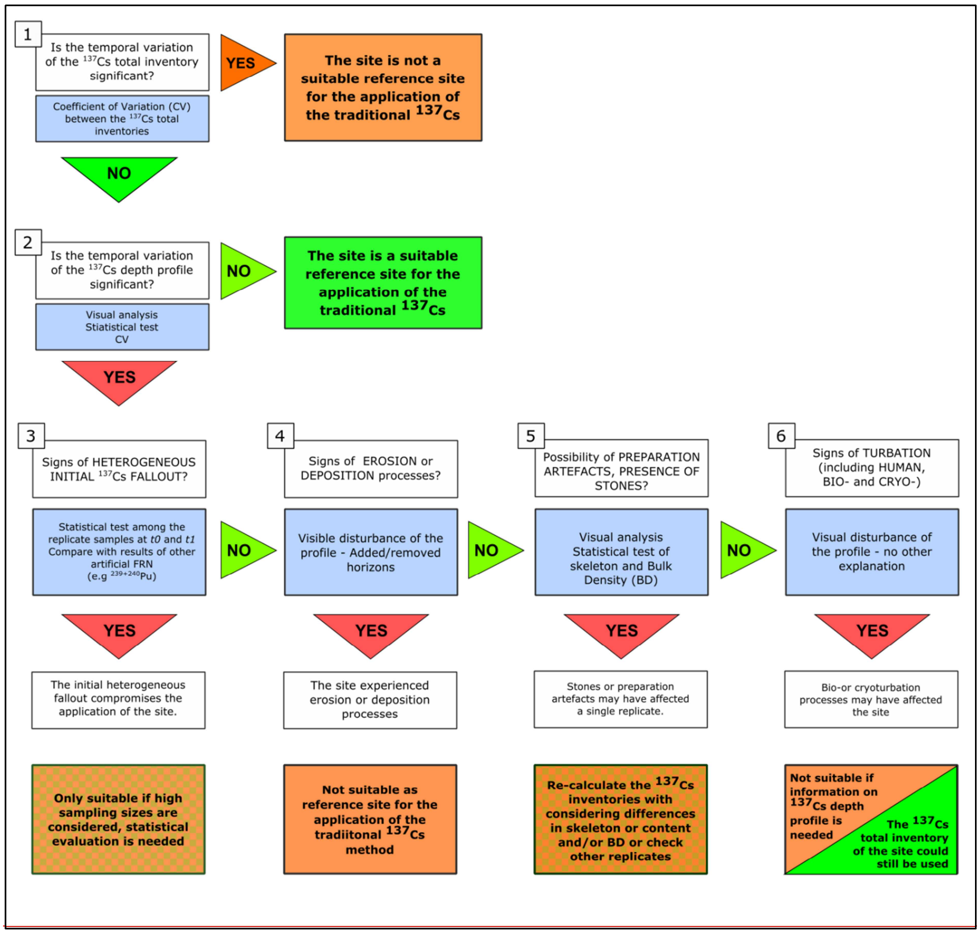


1
Is the temporal variation of the ¹³⁷Cs total inventory significant?
Coefficient of Variation (CV) between the ¹³⁷Cs total inventories
YES
The site is not a suitable reference site for the application of the traditional ¹³⁷Cs
NO
2
Is the temporal variation of the ¹³⁷Cs depth profile significant?
Visual analysis
Stiatistical test
CV
NO
The site is a suitable reference site for the application of the traditional ¹³⁷Cs
YES
3
Signs of HETEROGENEOUS INITIAL ¹³⁷Cs FALLOUT?
Statistical test among the replicate samples at t0 and t1
Compare with results of other artificial FRN
(e.g ²³⁹⁺²⁴⁰Pu)
NO
4
Signs of EROSION or DEPOSITION processes?
Visible disturbance of the profile - Added/removed horizons
NO
5
Possibility of PREPARATION ARTEFACTS, PRESENCE OF STONES?
Visual analysis
Statistical test of skeleton and Bulk Density (BD)
NO
6
Signs of TURBATION (including HUMAN, BIO- and CRYO-)
Visual disturbance of the profile - no other explanation
YES
YES
YES
YES
The initial heterogeneous fallout compromises the application of the site.
The site experienced erosion or deposition processes
Stones or preparation artefacts may have affected a single replicate.
Bio-or cryoturbation processes may have affected the site
Only suitable if high sampling sizes are considered, statistical evaluation is needed
Not suitable as reference site for the application of the tradiitonal ¹³⁷Cs method
Re-calculate the ¹³⁷Cs inventories with considering differences in skeleton and/or BD or check other replicates
Not suitable if information on ¹³⁷Cs depth profile is needed
The ¹³⁷Cs total inventory of the site could still be used

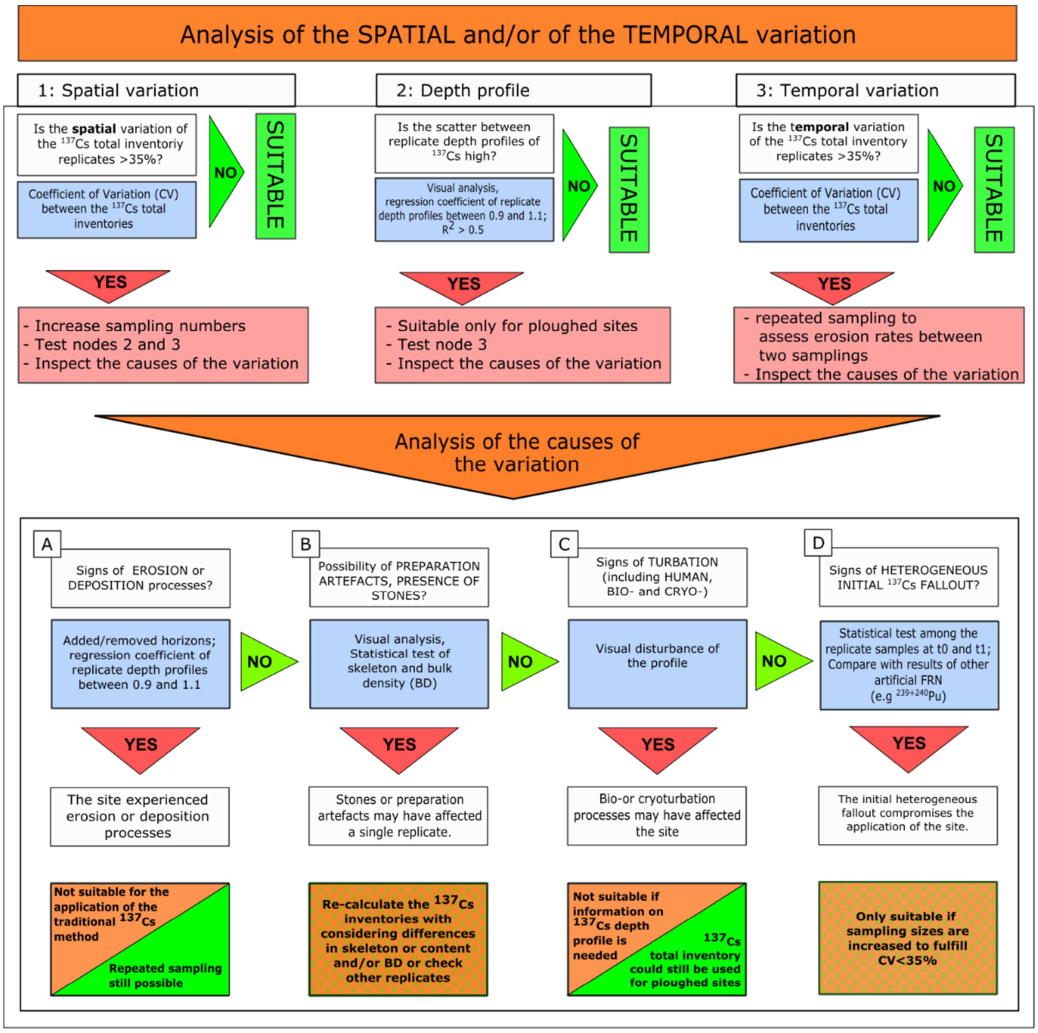


Figure 20: The CheSS decision tree to evaluatevalidate the suitability of a reference site for using ^{137}Cs as soil erosion tracer.

A: Signs of disturbance associated with erosion and deposition processes

A variation in the ^{137}Cs depth profile may have been caused by soil movement processes affecting the site (Figure 2, A). 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 (Figure 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 (Figure 3). In 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 (Figure 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 other location or a repeated FRN sampling approach to estimate erosion rates between the two sampling campaign should be considered (Kachanoski & de Jong, 1984).

reference	deposition	erosion
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

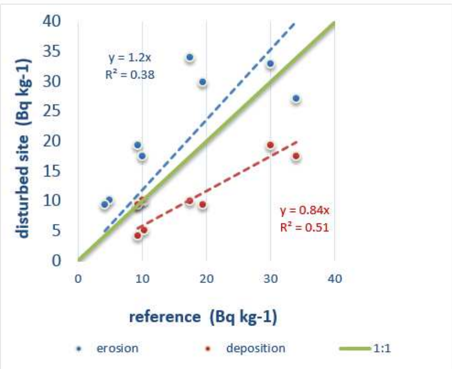


Figure 3: Signs of sheet erosion (A) and deposition (B) on a depth profile of a hypothetical reference site.

B: Sampling or preparation artefacts

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 > 2mm) (Figure 2, B). The presence of stones might determine pass ways of water as well as very fine particles and solutes in the soil and thus influences the accumulation/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 low skeleton content (Figure 4, A) 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 (Figure 4, B) or (ii) creating macro- and micro-pores favouring ^{137}Cs associated with fine particles to "migrate" to deeper layers (Figure 4, C) or causing lateral movement which will induce a lower ^{137}Cs content in our samples.

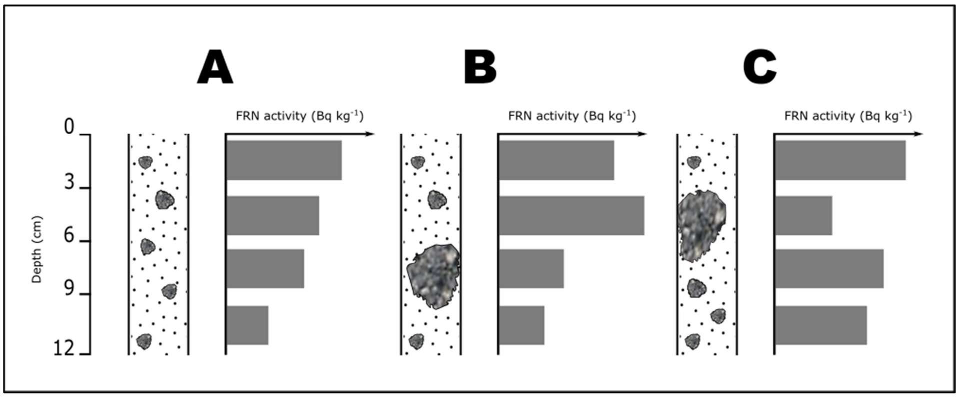


Figure 4: Possible influence of stones on the FRN depth distribution.

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 paired T test) if skeleton content and bulk densities are comparable between replicates is suggested. Finally, sampling, preparation artefacts and measuring processes may produce various sources of errors between different sites and years. The latter is especially the case, if different people prepared 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 of the results should be properly reported.

C: Signs of soil disturbance

Different forms of disturbance, such as bio-, cryoturbation or even human induced soil perturbation (e.g. tillage, seed bed preparation, digging etc.) might have influenced the ^{137}Cs depth distribution between different sites and *t0* and *t1* (Figure 2, C). 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 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.

D: Signs of a heterogeneous initial fallout of ^{137}Cs over the area

Finally a significant difference between reference replicates ^{137}Cs depth profiles measured at *t0* and at *t1* may not be necessarily due to a temporal variation because of soil disturbance, but instead be caused by a high small scale spatial variability of ^{137}Cs distribution at the site, due to heterogeneous initial fallout over the study area (Figure 2, node 3D). In Europe, significant small scale variability of ^{137}Cs distribution is known to be due to the Chernobyl fallout, which was characterized by a high ^{137}Cs deposition associated with few rain events. Compared to the nuclear bomb tests 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 between reference site replicates) measured at both or at least one sampling campaign, looking at the CV again, as presented in the previous sections, or through a statistical test (for example the Analysis of the 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). (Figure 2, node 3D). 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, 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 dataset 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.), 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, 2013). The U-formed 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 2013 for ^{137}Cs. Six reference sites (REF1 to REF6) were identified in flat and undisturbed areas along the valley. At each site 3 cores (40 cm 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 >1cm 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 analyzed separately, to investigate the small scale variability of the FRN content. All cores were air-dried (40°C for 72h), sieved (<2 mm) to remove coarse particles and the skeleton content as well as the bulk density (BD) was 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 were analysed at the Institute of Physics of the University of Basel using a coaxial, high resolution germanium lithium detector (Princeton Gammatech) with a relative efficiency of 19% (at 1.33 MeV, ^{60}Co). Counting time was set to 24 hours per sample. Samples collected in 2015 were analysed at the state laboratory Basel-City using coaxial high resolution germanium detectors having 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.

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, Prag. This solution was bound in silicon resin of 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 were performed using a Thermo X Series II quadrupole ICP-MS at the Northern Arizona University, USA. 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 equation (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 (i.e. the one located at the Institute of Physics of the University of Basel and the other located at the State Laboratory Basel-City). The results highlight a high correspondence of the measurements held by the two analytical systems ($R^2 = 0.97$; $p < 0.005$), however the detector of the State Laboratory Basel-City returns slightly lower ^{137}Cs activities (Figure 5). Thus, the ^{137}Cs activities of the samples measured in 2013 were corrected to the values of the detector of State Laboratory Basel-City (higher efficiency) to allow comparability between the different data sets.

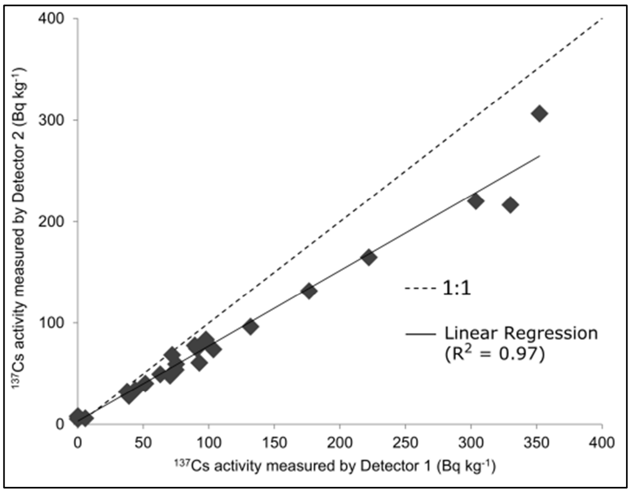


Figure 5: The comparison between the ^{137}Cs measurements of a subset of samples (n=16) performed with two different HpGe detectors, where detector 1: detector hosted at the Physics department of the University of Basel (CH) and detector 2: detector hosted at the State-Laboratory of Basel (CH).

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 to 8619 Bq m^{-2}, with a mean value of 5701 Bq m^{-2} and a SD of 1730 Bq m^{-2} (Figure 6).

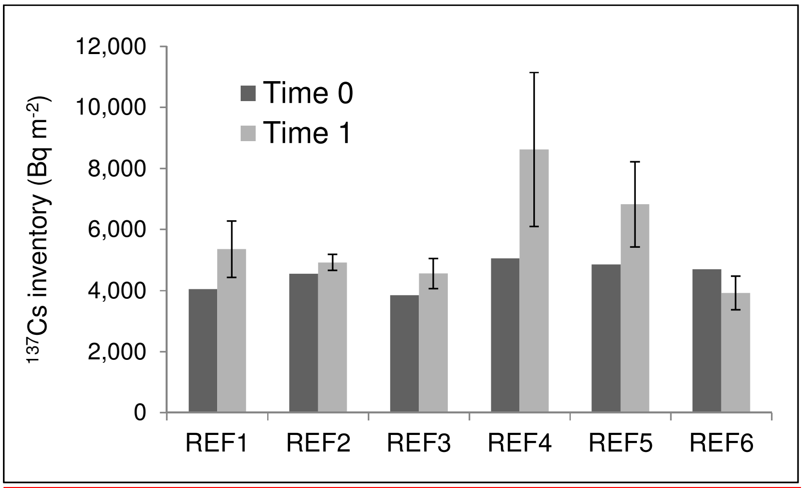


Figure 6: Temporal variation between the total ^{137}Cs inventories measured at the reference sites in the Urseren Valley, where Time 0 = 2013 and Time 1 = 2015. The errors bars indicate the standard deviations of the inventories among the replicates collected at each reference in 2015.

~~All reference sites have been investigated through the CheSS decision tree presented in section 2.2.~~ When following the CheSS decision tree~~As for the first node~~, we investigated the ~~temporal~~ variation in the ^{137}Cs total inventories at each reference site (node 1). ~~-~~The replicate samples were analyzed separately only during the second sampling campaign (t1), while during the first sampling campaign (t0) only composite samples were analysed. ~~Thus, the approach presented in section 3.2 (Node 1) was followed, and the analysis first focused on the spatial variability measured at t1.~~ Reference sites 3, 5 and 6 presented signs of high small scale variability, as expressed by CV of 48 % ~~for all cases~~ (Table 1). Such variability excluded them from any further application as reference sites without subsequent additional sampling. F~~As f~~or reference sites 1, 2 and 4, the CV~~t1~~ was between 19 – 31%.

Passing to~~at~~ ~~the second~~ node 2 of the CheSS decision tree~~ presented in section 2~~, the analysis focuses on the ~~temporal~~ variation ~~-~~of the shape of the ^{137}Cs depth profile (F~~f~~igure 7). Here we examined the regression between the reference depth profiles in t0 and t1. For the three sites ~~under investigation~~with acceptable spatial variability (i.e. reference site 1, 2 and 4) the site REF4 shows sign of deposition with a regression coefficient between t0 and t1 = 1.34. The deposition was confirmed by field observation of construction works that were conducted between the two samplings. Thus, after this disturbance the site is not a suitable reference site anymore. Among the sites with high spatial variability 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 REF4 showed a significant difference of the total ^{137}Cs inventories between t0 and t1. Thus, confirming the unsuitability of the site after the construction works.

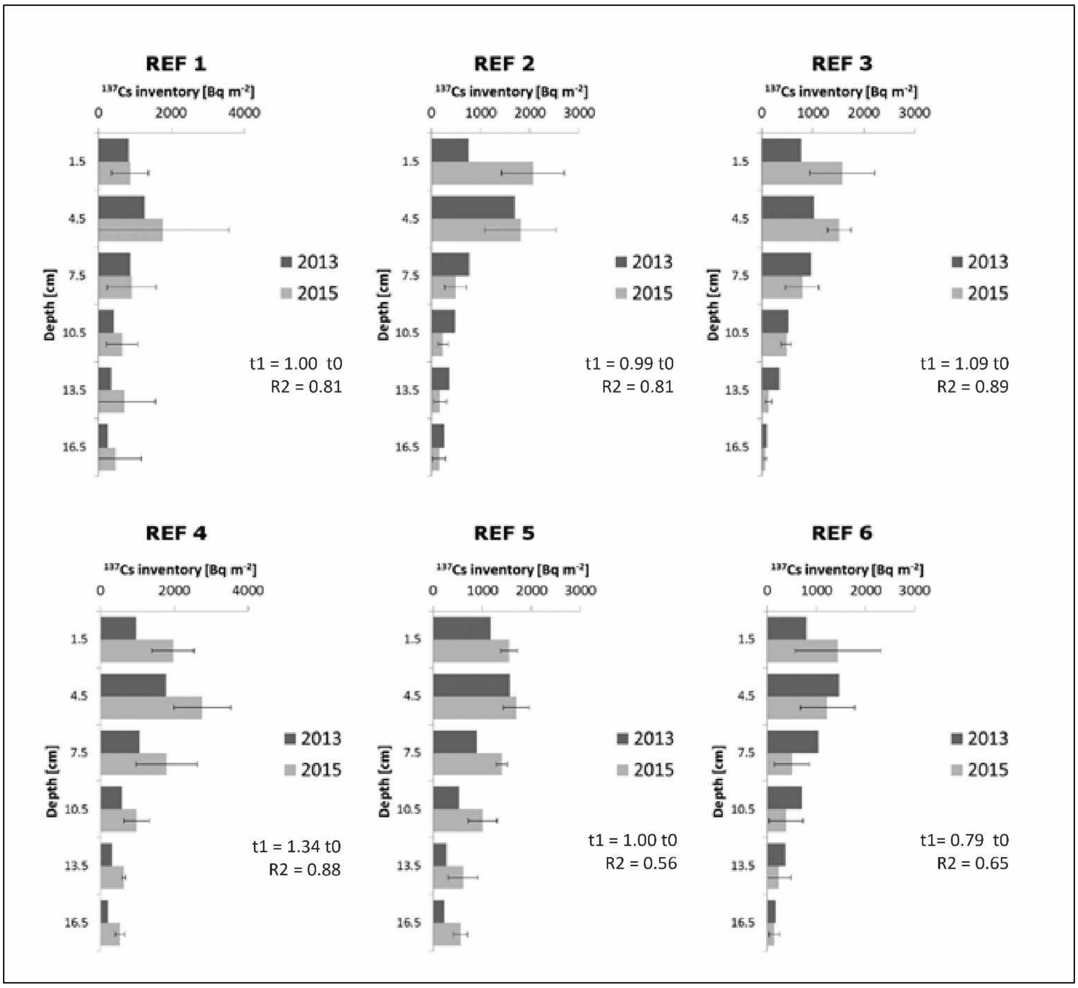


Figure 7: The ^{137}Cs depth profiles of the ~~nine~~ six investigated reference sites in the Urseren Valley for the two different sampling campaigns. The errors bars indicate the standard deviations of the inventories among the replicates collected at each reference site 2015. Further the regression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

To further investigate the causes for the spatial variation, $^{239+240}$Pu inventories measured at the three replicates of each site were analysed for t0 = 2010 and t1 = 2015 (Figure 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.

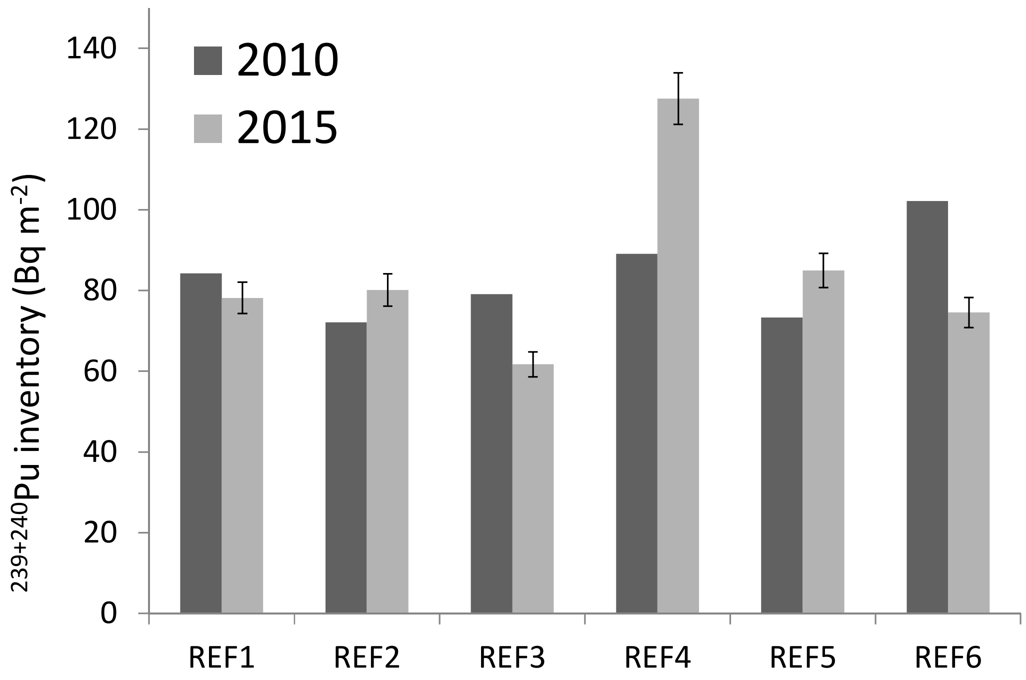
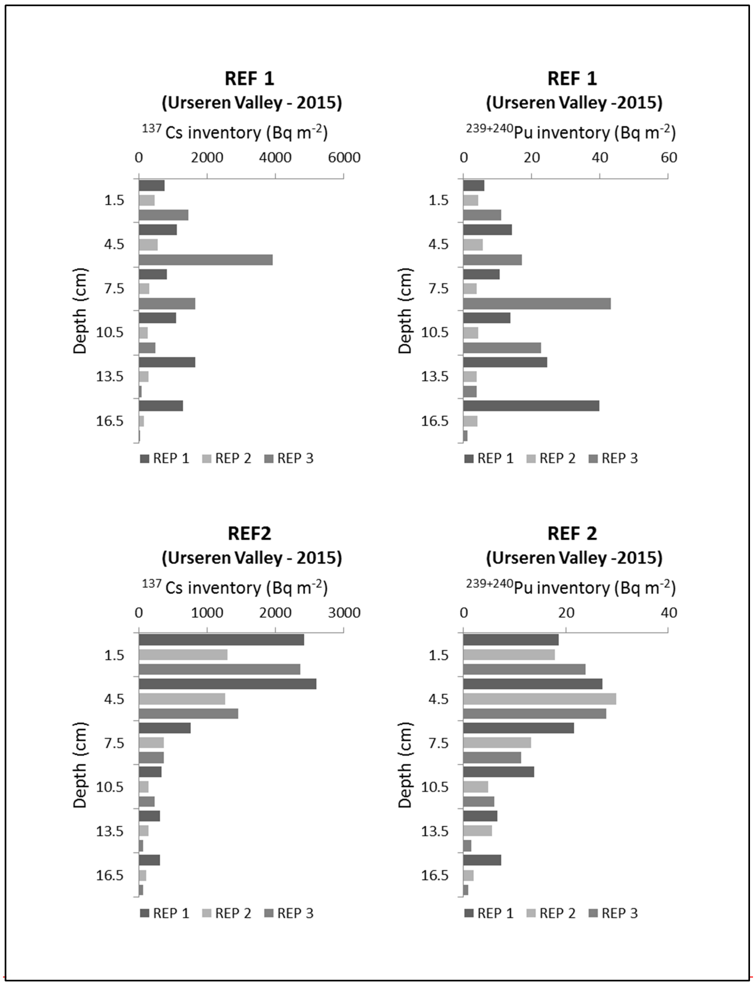


Figure 8: Temporal variation between the total $^{239+240}$Pu inventories measured at the reference sites in the Urseren Valley, where Time 0 = 2010 and Time 1 = 2015. The errors bars indicate the standard deviations of the inventories among the replicates collected at each reference in 2015.

To validate those results, $^{239+240}$Pu inventories measured at the three replicates of the site were also analysed (Figure 8). In this case, the test highlighted no significant difference between the replicates. Thus, at reference site 2, the heterogeneous distribution of ^{137}Cs might be due to the heterogeneous Chernobyl deposition on snow covered ground. FurtherIn contrast, the depth profiles of the three replicates at reference site 1 present also significant differences. We then looked at the differences in the skeleton content of the three replicates (node 5Figure 2, B). An ANOVA test showed a significant difference (p-value of 0.025), thus, a difference in the presence 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 the siteREF1 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 REF2, REF4 (before the construction works) seem most suitable for ^{137}Cs. Form visual inspection of the soil profile be could exclude the 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.

Table 2: The Coefficients of Variation (%) between the ^{137}Cs inventory of each depth increment at the reference sites measured at *t0* and *t1*.

Depth	REF1		REF2	
-	CV_{t1}	Cv_{t0-t1}	CV_{t1}	Cv_{t0-t1}
1.5	57	47	31	48
4.5	97	88	41	34
7.5	73	61	45	40
10.5	69	63	42	50
13.5	120	121	72	62
16.5	140	136	82	64



~~**Figure 8: The ^{137}Cs depth profiles (on the left) and the $^{239+240}$Pu depth profiles (on the right) of the three replicates collected at reference sites 1 and 2 in 2015 in the Urseren Valley.**~~

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 the analysis of the small scale variability of ^{137}Cs distribution in the reference areas, especially in those regions affected by ~~Chernobyl~~ nuclear accident fallout. To cope with small scale variability, sampling numbers might be increased, or the temporal variation of ^{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 ~~most~~ some reference sites. Additionally ~~At reference site 1~~ the presence of stones affected the shapes of the depth profile in at least one replicate sample at reference site 1. ~~Therefore~~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 & de Jong, 1984). It doesn't 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 to exclude those sites which present signs of uncertainty. With this we are convinced to contribute improving the reliability of the FRN based soil erosion assessments.

Authors contributions.

L. Arata, K. Meusburger, L. Mabit and C. Alewell designed the concept of the method and analysed the data. A. Bürge contributed to the collection and preparation of the soil samples, and to the analysis of the data. M. Zehringer measured the ^{137}Cs activity of the soil samples and analysed the results. M. E. Ketterer measured the $^{239+240}Pu$ activity of the soil samples. L. Arata prepared the manuscript with contributions from all co-authors.

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