

Point-by-point response Referee #1

Understanding the role of water and tillage erosion from ²³⁹⁺²⁴⁰Pu tracer measurements using inverse modelling

5 Florian Wilken, Michael Ketterer, Sylvia Koszinski, Michael Sommer and Peter Fiener

We appreciate that the reviewer acknowledges the study design and supports the publication. The reviewer raised questions with regards to the modelling part of the study. We appreciate the advices and revised the manuscript accordingly. Please see the detailed answers (in italics) to the comments below:

10

L 82: I don't think parameter space is the term you are looking for here. Consider changing this to "parsimonious parameter set".

Thanks, we follow the suggestion!

15 Equation 3: This formulation of WaTEM/SEDEM looks a little different to what I usually see (e.g. $T_c = K_{tc} R K$ (LS-aSIR)). Is this correct?

*Our version of SPEROS is not identical to the WaTEM/SEDEM version of van Rompaey et al. 2001. Our SPEROS version does not include a rill coefficient as for instance used in the SPEROS version by Bouchoms et al. 2017. This was done as (i) rill erosion is not a dominant process in the study area and (ii) the rill/interrill ratio is typically used for longer slopes that cannot be found in the study area. For instance, van Rompaey et al. 2001 use observations from Govers and Poesen 1998 that on a 0.06 m m^{-1} slope after a distance of **65 m** the interrill erosion rate equals the rill erosion rate. To avoid confusion, we remove the reference to WaTEM/SEDEM from the text.*

20

Equation 5: Is k_{til} calculated by this equation? I thought it was being sampled from a pre-defined range during the inverse modelling.

25

Indeed, k_{til} was not calculated but implemented for a pre-defined range that covers low to extreme tillage erosion rates. The range was defined with respect to literature values and own experiments. As the k_{til} is one of the three basic parameters of the inverse modelling approach, we would like to provide the formula of the k_{til} to assist the reader understanding how this

parameter is derived. To avoid confusion, we mention that the *k_{til}* formula is just shown for illustration: “(as given for illustration in Eq. 5)” in the implementation section.

Are the raster outputs from equations 3 and 5 summed? A flow-chart explaining the modelling might be helpful to guide the readers. Yes, the output of the different parameter combinations was summed up and subsequently statistically analysed (e.g. Fig. 5 & 6).

We had an extensive discussion among the authors if a flow-chart would improve the readability of the manuscript. The result of this discussion was that we probably would need a quite detailed flow chart to substantially add a value for the reader. As the model was already described in detail in earlier papers, we would prefer keeping the focus on the model results rather the model description. So, we would prefer not adding an additional figure. However, if the editor also thinks that a flow-chart would be helpful, we would follow his advice.

L246: Why qualitatively?

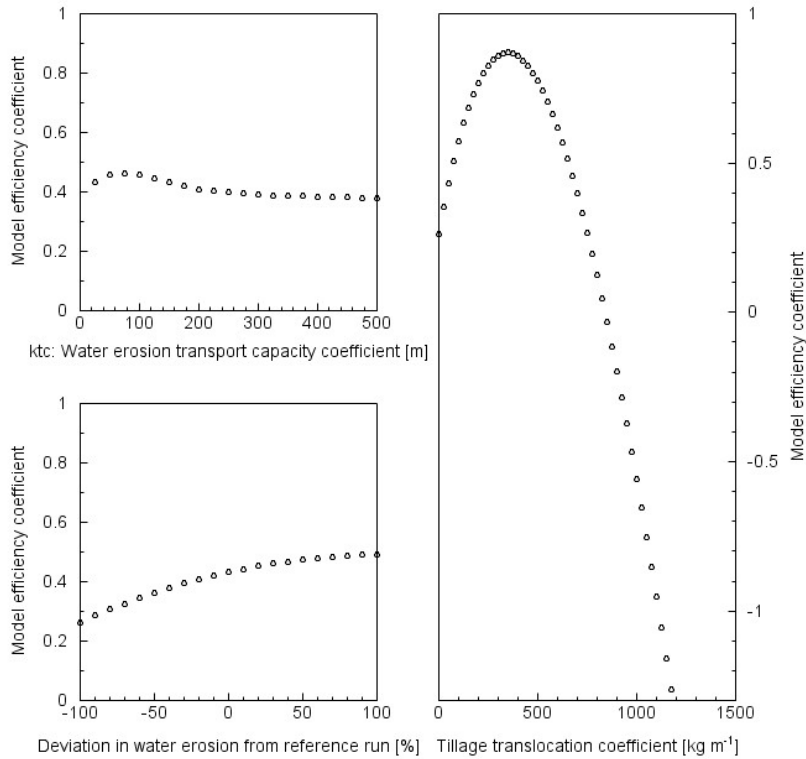
Thank you for pointing at this! We changed the text to “quantitatively”

L247-253: Are these correlations calculated based on the best-fit model realisation? Please inform this in the text.

These calculations are based on the best model fit realisation of the *k_{til}* and the reference for water erosion. We did not explicitly stress this in the text as the correlation results are only affected by changes in the *k_{tc}* value (*k_{til}* and *RUSLE* product do not influence spatial soil redistribution patterns) that follow the reference parameterisation. To avoid confusion, we reformulate the sentence: “The spatial correlation between the ²³⁹⁺²⁴⁰Pu derived patterns and the modelled best knowledge soil redistribution, including both water and tillage erosion, is only moderate (R^2 : 0.45) on a raster by raster comparison (n : 1699, 5 m x 5 m grid points; Fig. 5a).”

L256-257: By looking at figure 6c I wouldn’t say the predictions showed “hardly any sensitivity to erosion strength”. The MEF ranges to -0.2 to 0.6, with higher values clearly associated to the positive deviations in water erosion from reference run. Of course, the effect of the tillage parameter is much more pronounced. Nevertheless, it would be a good idea to show univariate dot plots of the sampled parameter space with their associated goodness-of-fit measure.

We agree, there is minor sensitivity that, however, does not lead to any indication of an optimal parameterisation. The requested dot plots underline that statement (see below), however, we think the 3D plot nicely shows the interplay between the parameterisations. Hence, we would prefer to stay with the 3D Figure.



L260: I think it would be nice to look at the parameter space which produced acceptable model realizations (for instance, within the kriging variance or above a given MEF threshold), instead of focusing on the best-fit. That would make your approach more robust. There are multiple solutions to an inverse problem, and perhaps that should be more explicitly recognized. I mean, if you consider the error in the observed data (interpolated Pu map), there might acceptable model realizations produced with the contribution of the water erosion component. This also relates to the discussion in lines 307-310.

In general, we agree that there are multiple solutions for an inverse modelling and this might result in a substantial equifinality problem. However, as we did only vary three parameters where the parameter for the tillage erosion clearly dominates soil redistribution processes, there is no typical equifinality problem resulting from our approach. We follow the idea to provide a range of the k_{til} parameterisation that achieves a MEF better than 0.8 and demonstrate the limited sensitivity of the water erosion parameterisation on the best-fit model run. The following was added to the text: “A MEF better than 0.8 and RMSE below 6.5 Bq m^{-2} were found for a k_{til} range from 225 to 475 kg m^{-1} , while the best model fit was found for a k_{til} of 350 kg m^{-1} achieving a MEF of 0.87 and a corresponding RMSE of 5.2 Bq m^{-2} . The best model fit was found without the contribution of water erosion. The highest impact on the best-fit model run was a 0.31 MEF-reduction by an extreme water erosion parameterisation ($k_{tc} = 500$, water erosion strength = 200%).”

L309-310: Not sure I understand this. If none of the sampled parameter values produced adequate system representations, why would we need to assume the reference parameterisation is the most appropriate?

Thanks for pointing at this. As the inverse modelling did not indicate any optimal value for the water erosion parameterisation, we follow the input data recommendations that has been officially published by the administration of the state Brandenburg,

5 *Germany. We changed this in the text: “Therefore, we assume the reference parameterisation for the region given by the state of Brandenburg as the most appropriate (k_{tc}: 150 m and RUSLE parameters according to Tab. 1).”*

Technical corrections:

L209: “Results” is repeated.

10 *Thanks, we changed the text as follows: “[...]raster map against the results calculated by the inverse modelling approach.”*

L307-308: “Inverse modelling” is repetitive at the end of the sentence.

Thanks, we changed the text as follows: “This sediment delivery into the kettle hole cannot be explained by the inverse modelling of water erosion applying a reasonable parameter range.”

Point-by-point response Referee #2

Understanding the role of water and tillage erosion from ^{239}Pu and ^{240}Pu tracer measurements using inverse modelling

Florian Wilken, Michael Ketterer, Sylvia Koszinski, Michael Sommer and Peter Fiener

5

We highly appreciate the comprehensive and very valuable review carried out by Olivier Evrard, thank you! The reviewer positively highlights the study design and efforts made but has concerns about the conclusions on the contribution of soil degradation before the 1960th. We revised the manuscript accordingly. Please see the detailed answers (in italics) to the comments below:

10

Abstract:

L14: Something wrong here; *we rephrased the text as follows, “The majority of soil redistribution studies focus on water erosion,[...]”*

L17: why 'whereas'?; *we changed whereas to “where”*

15 L19: Suggestion, types; *thanks, we follow the suggestion*

L19: Suggestion, individual; *thanks, we follow the suggestion*

L20: Suggestion, soil redistribution patterns; *we would like to keep this phrase. The effect of lateral soil translocations on the (regular) soil distribution (soil pattern) in landscapes is meant here, not the process dynamics/rates.*

20 L23: Suggestion, this; *we changed the text to: “[...]the study catchment[...]”*

L24: Unclear why you refer to this 'kettle hole' for the 1st time here; *we introduce the kettle hole in the study area description: “The aim of this study is to understand the contribution of water and tillage*

erosion leading to soil patterns found in a small hummocky ground moraine kettle hole catchment under intensive agricultural use.”

L26: I would clarify this sentence to make it more explicit... Do you mean that tillage erosion started before the mechanisation in the 1960s?; *We follow the recommendation and reworded the sentence as follows: “Hence, tillage erosion already started before the onset of intense mechanisation since the 1960s.”*

L28: could be more explicit what you mean here; *we know from other studies that tillage erosion is the dominant driver of soil properties (SOC, SIC, N, texture, micro- & macro aggregates) and finally crop yields in the region. However, within this study we focus on erosion patterns derived from FRN distributions that does not explicitly deal with fertility or other soil properties. Hence, we would like to avoid statements that are not covered by our analysis. We reduced the sentence as follows: “In general, the study stresses the urgent need to consider tillage erosion as a major soil degradation process that can be the dominant soil redistribution process in sloped arable landscapes.”*

15 **Introduction:**

L34: I agree with you but I think that this is more general than related with EU WFD... in many countries and regions, soils/agricultural land are managed by different authorities than waterbodies, which complicates the design of integrated approaches at the catchment scale where the connection between cultivated hillslopes and river systems would be taken into account...; *thanks for this comment. We agree that an integrated catchment management is also limited due to administrative barriers. We changed the text as follows: “Within the European Union, the focus on off-site erosion effects is partly caused by the definition of the goals of the EU Water Framework Directive (EU 2000/60/ES) that focuses mainly on water bodies and floodplains but not on a fully integrated catchment management that would call for complex shared responsibilities between different administrative units.”*

25 L36: not very clear; *We reformulate the text as follows: “Thereby, other soil erosion drivers like tillage and wind are somewhat out of the scope of most studies.”*

L39: Unclear; *We changed the text as follows: “Particularly areas of a hummocky topography with short summit-footslope distances, such as young morainic areas, [...]”*

L41: Suggestion, associated; *thanks, done*

L50: Internal redistribution within the catchment?; *To be more specific, we changed the text as follows:*

5 *“[...]address catchment internal redistribution.”*

L53: Suggested change: “cannot provide a reconstruction of soil redistribution changes throughout time?”; *we changed the text as follows: “However, these beforehand applied tracers and change monitoring methods cannot provide a reconstruction of soil redistribution of the past.”*

L53: Unclear what you mean here?; *we agree and removed that part of the sentence: “Natural or*
10 *anthropogenic tracers in soils can be used to understand soil redistribution.”*

L55: Parsons and Foster 2011, not sure this is the most appropriate article to support your statement here!;
thanks, you are right. We remove this reference.

L56: Could be rephrased here to clarify; *we rephrase the sentence as follows: “The force of atmospheric nuclear weapon tests transported radioisotopes outside the troposphere, where circulation led to a*
15 *globally almost homogeneous spatial distribution and subsequent fallout on soils by rain (Alewell et al. , 2017).”*

L57: Why ‘limited’ test ban treaty and suggestion to write it in capitals; *To our knowledge , the Limited or Partial Test Ban Treaty still allowed underground detonations. In 1996 all kind of nuclear weapon tests were banned by the Comprehensive Test Ban Treaty. However, as atmospheric tests and the year is*
20 *named, we think we can only refer to the Test Ban Treaty to avoid confusion by the word “limited”. We changed the text as follows: “[...]while the Test Ban Treaty caused[...]*”

L59: Of note, there was (much less) but still significant fallout after 1963, until the early 1980s. We've recently figured out that a lot of radionuclide atmospheric monitoring data is lacking for documenting this fallout, mainly for the periods between 1954-1959 and 1967-1976 (see Evrard et al., 2000, Fig. 1-2
25 and associated text; <https://doi.org/10.1016/j.geomorph.2020.107103>).; *Thanks, we moved from rapid end to “rapid decrease” and refer to the paper.*

L60: [...]which enables the use of radioisotopes as redistribution tracers in soils. Reference missing here.;
Thanks, we added Meusburger et al. 2016

L61: Suggestion to restrict the reference list to three items, (e.g. Van Oost and Govers, 2006; Porto and Walling, 2012; Zhang, 2015; Greenwood and Meusburger, 2019; Srivastava et al., 2019); *we followed the*
5 *suggestion: (e.g. Porto and Walling, 2012; Chartin et al., 2013; Evrard et al., 2020)*

L66: I disagree with you unless you add 'in those areas that received significant Chernobyl fallout' (is this what you mean?); *the fallout of the Chernobyl disaster shows a high spatial variability but the majority of regions in Europe were to some extent affected. As the unmixing of the 1960th and Chernobyl ¹³⁷Cs is complex, we think the use of ¹³⁷Cs is in Europe subject to uncertainties. However, it is not our intention*
10 *to say that the use of ¹³⁷Cs in Europe is generally impossible, however, requires special care. To indicate this, we changed the text as follows: “Hence, the use of ¹³⁷Cs as a soil redistribution tracer in Europe is associated with uncertainties and requires special attention concerning a potential Chernobyl contamination (Evangelidou et al., 2016).”*

L67: Could you quantify this in %? (if you calculate the decay between 1963 and 2000); *thanks, for this*
15 *good idea. We added in brackets that about 70% of the global fallout has been reduced.*

L69: Does not make sense to refer to the half-life of a sum of radionuclides, does it?; *thanks for this hint, we changed the text as follows: “[...]the half-life of ²³⁹Pu and ²⁴⁰Pu is long [...]”*

L70: Could you be more explicit here?; *we provide the radius of pronounced ²³⁹Pu and ²⁴⁰Pu contamination surrounding Prypiat, Chernobyl: “(<100 km; Kashparov et al., 2004; Matsunaga and*
20 *Nagao, 2009)”*

L74: Unclear, please rephrase; *we rephrased the text as follows: “However, the use of radioisotope tracers integrates all types of soil redistribution processes and does not provide information on the relative contribution of the driving processes at play (e.g. water, tillage, wind).”*

L75: Suggestion, Unravelling the respective contributions of these different processes [...]; *thanks, done*

25 L77: Unclear; *we rephrased the text as follows: “There are only few models that take both water and tillage erosion processes into account. Physically oriented models like MCST-C (Wilken et al., 2017b)*

and LandSoil (Ciampalini et al., 2012) simulate individual erosion events and are developed to enhance process understanding, while conceptual USLE based models (WaTEM/SEDEM: Van Oost et al., 2000; Van Rompaey et al., 2001) aim at a robust prediction of long-term soil erosion rates (Alewell et al., 2019). ”

5 L82: Range/variation? is this what you mean?; *we want to point out that a model to unravel the role of water and tillage erosion over more than 50 years should be based on an easy to understand and available parameter set (space). For instance, within this study a single tillage erosion and two water erosion parameters were iterated. We changed the text as follows to avoid confusion: “[...]with a limited parameter space covered by available input data[...]*”

10 L85: Suggestion, objective; *thanks, done*

Methods:

L92: Unclear what you mean here, as I would say that this is the case for any catchment; to indicate that the small catchment shows high morphological variability; *we changed the text as follows: “The study*
15 *area is part of a kettle hole catchment (4.2 ha; Fig. 1) showing a high morphological variability covering convex hilltops, steep slopes and flat areas. ”*

L97: Suggestion, are explained by land consolidation programmes implemented in the 1960s[...]; *Thanks, we followed the suggestion.*

L98: How large were the farming structures?; *We provide the following information in the text: “In 1939,*
20 *large farms that manage more than 100 ha of arable land cultivated 7% of the total arable area of Germany. In the present-day federal states of Mecklenburg-Pomerania, Brandenburg and Saxony-Anhalt, large farms cultivated 30% of corresponding arable land (Wolz 2013). ”*

L100: We would almost need a sketch with the location of the different soil types along the catena: *we follow the recommendation and add an idealised catena to the study area description (Fig. 1b).*

25 L108: Suggestion, take place each year?; *thanks, we followed the suggestion*

L111: Do you mean 'per transect line'? maybe add that it is to avoid 'boundary effects'?; *we changed the text as follows: “The soil sampling design was organised according to a regular 20 m x 20 m grid with at least one sampling point of the transect line exceeding the spatial extent of the catchment under study (Fig. 1) to avoid boundary effects.”*

5 L112: This is cool!! *Thanks for appreciation!*

L118: Suggestion, ranging; *thanks, we follow the suggestion*

L122: Sorry if I missed something, but I think that you shouldn't use the term 'replicate' if you collected one single core at each of these 10 different locations, right?; *We took at 209 locations 219 cores. The replicate cores were taken at ten selected sites in direct proximity to each other (max. 50 cm apart) and*
10 *were supposed to be representative for the same location. This was basically done to achieve more soil material for additional analysis like the high resolution 5 cm increment vertical resolution measurements. The replicate cores are not critically relevant for the analysis. Hence, we decide to reduce the information to avoid confusion: “Closed soil cores, using a steel drill containing a plastic liner (4.6 cm inner diameter), were driven by a percussion corer (Cobra TTe; Atlas Copco Power Techniques GmbH,*
15 *Stockholm, Sweden) into the ground down to a depth of 50 cm at 209 sampling points.”*

L127: Suggestion, n=3; *thanks, we follow the suggestion*

L129: Suggestion, air-dried; *thanks, we follow the suggestion*

L133: Suggestion, measured for their; *thanks, we follow the suggestion*

L133: I guess that you did not want to reduce the inventory of samples, but instead the number of samples
20 *for which the analysis results would be under the detection limits, right?; Yes, sure, thanks for pointing at this error. We reformulated the text as follows: “This was done to reduce the number of samples with Pu activities below the detection limit.”*

L134: Suggestion, higher-resolution; *thanks, we follow the suggestion*

L154: Suggestion, equations; *thanks, we follow the suggestion*

L158: What about the undisturbed sites? You did not refer to these sites when described the sampling plan?; *We are using a potential range for the reference concentration based on reference measurements of a study that was carried out nearby. We explain this in the implementation section. To indicate this, we point at the corresponding section in the text: “undisturbed sites in Bq m⁻² (see implementation section 2.5)”*

L168: SPEROS-C and/or SPEROS-Pu?; *Both SPEROS-C and SPEROS-Pu use the same tillage erosion module. However, as the description is for SPEROS-Pu, we only name this version in the text. Thanks, for pointing at this.*

L175: Suggestion, 10-cm depth layers?; *thanks, we follow the suggestion*

L176: Unclear what you mean here?; *Thanks! The word “same average” might have been caused the confusion. We remove the word “same” to indicate that the plough layers have the average ²³⁹⁺²⁴⁰Pu activity of the two topsoil layers: “[...]have the average ²³⁹⁺²⁴⁰Pu activity of the upper[...]”*

L179: Something missing here; *thanks, we changes the text as follows: “Soil erosion processes lead to a reduction of the ²³⁹⁺²⁴⁰Pu inventory per m² due to soil and associated ²³⁹⁺²⁴⁰Pu loss, which causes mixing in of non-contaminated subsoil.”*

L183: Suggestion, topsoil?; *thanks, we follow the suggestion*

L183: Suggestion, was?; *thanks, we follow the suggestion*

L185: Ok, based on how many measurements; *Calitri et al. 2019 determined the reference value based on four sites that showed no geochemical and soil profile morphological (e.g. soil profile truncation) indications for erosion or deposition. We changed the text accordingly: “The reference inventory of undisturbed sites follows the value determined by Calitri et al. (2019) who found a ²³⁹⁺²⁴⁰Pu inventory of 43±3 Bq m² based on four sites that did not show profile morphological or geochemical indication for soil redistribution at a location 8.5 km apart from the study area.”*

L186: Please rephrase; *we changed the text as follows: “To address the uncertainty inherent to the reference measurements, a reference range from 40 to 46 Bq m² was accounted for in the simulations.”*

L191: Suggestion, a decametre scale?; *thanks, we follow the suggestion*

L191: Something wrong here in the sentence? & missing word here?; *We rephrase the sentence as follows: “Different block sizes were tested for the kriging approach. A block size of 20 m was selected that matches the sampling resolution and did not to cause over-smoothing of the interpolation result.”*

5 L208: Suggestion, goodness-of-fit?; *thanks, we follow the suggestion*

L217: Please provide the manuscript section number instead; *thanks, we follow the suggestion*

Results:

L222: Important, do they correspond to those points having the lowest Pu inventories in topsoil layers?;

10 *Yes, the subsoil locations that fall below the detection limit show low inventories below 40 Bq m². Please note that we excluded 30% of the subsoil samples that showed the lowest topsoil activities. Hence, for locations of the lowest inventories, no subsoil measurements were carried out. However, to indicate this relationship we add the following sentence: “Those seven samples are all located at positions with ²³⁹⁺²⁴⁰Pu inventories below the lower reference boundary (40 Bq m²).”*

15 L225: unclear why you use 'in turn' here?; *we changed the text as follows: “All high-resolution depth profiles (5 cm increments) at erosional sites show a sharp reduction of the ²³⁹⁺²⁴⁰Pu activity below the plough layer (Fig. 3), while depositional sites show more complex depth distributions.”*

L227: Suggestion, reflect; *we changed the text as follows: “[...]with depth that is potentially caused by Pu enrichment processes[...]*”

20 L232: Suggestion, water and tillage soil redistribution processes?; *thanks, we follow the suggestion*

L232: Sorry; maybe again I miss something here, but isn't there a contradiction between the first and the second sentences here?; *we fully agree that these two sentences are confusing. We simplified the text as follows: “At 14 (of 209) sampling locations, a higher subsoil than topsoil ²³⁹⁺²⁴⁰Pu activity was found, which points at deposition of ²³⁹⁺²⁴⁰Pu depleted sediments. The majority of these locations show enriched*
25 *²³⁹⁺²⁴⁰Pu activities in the subsoil (11 of 14).*

L236: Suggestion, induce?; *we changed the word to “cause”*

L239: Unclear what you mean here... Furthermore, I had the impression that you assimilated the tillage depth to 20 cm (2 layers of 10 cm depth) in the above-mentioned text?; *we determined the tillage depth at each sampling location, which was found at an average depth of 23.5 cm. As the model takes 10 cm layers into account, the tillage depth was set to 20 cm in the implementation. We think the information is not critically important why we removed it to avoid confusion.*

L239: Isn't it a bit short here? Why would the enrichment be worse at these 5 locations compared to others? Including those highly enriched locations exceed the methodological maximum of detectable deposition; *As these locations are clearly subject to enrichment processes that cannot be adequately addressed, we think it is better to remove them and interpolate them from the surrounding information. Furthermore, these points were exclusively located near the kettle-hole border which does only exclude small areas. To make this clearer, we changed the text accordingly: “As the enrichment processes inherent to these five locations cannot be corrected, the locations were excluded from the analysis.”*

L247: Suggestion, soil redistribution; *thanks, we follow the suggestion*

L248: Seems to be part of the discussion?; *Our motivation to introduce this before the discussion section was to support the reader to understand the step of analysing classes. However, from a manuscript structural perspective this is indeed not correct, which is why we remove the sentence.*

L252: Suggestion, illustrates the good agreement...?; *thanks, we follow the suggestion*

L255: Description of the MEF in the methods; *we think it helps the reader to see the description of the MEF in direct proximity to the numbers. Furthermore, the MEF is rather a standard parameter that might be familiar to the majority of readers. Hence, we would prefer to keep the short description in the results section.*

L261: could you clarify what you mean here?; *we changed the text throughout as follows: “[...]geomorphological dynamics[...]”*

L262: Suggestion, reach; *thanks, we follow the suggestion*

L263: ?; we changed the text as follows: “Soil erosion at hilltop locations is shown to reach up to 14.9 cm (43 Bq m⁻² reference; 40 Bq m⁻² reference: 14.1 cm; 46 Bq m⁻² reference: 15.6 cm), while deposition can build a colluvium layer with a maximum thickness of 21.5 cm (43 Bq m⁻² reference; 40 Bq m⁻² reference: 24.9 cm; 46 Bq m⁻² reference: 18.6 cm) over the past 53 yr.”

5

Discussions:

L271: this remains questionable in my opinion (see my general comment), general reviewer comment: Importantly, the advantages of using Pu-239+240 inventories (compared to Cs-137 inventories) for reconstructing soil redistribution between 1964-2016 should be better justified in the text, in my opinion.

10 Of note, analysing Pu-239 and Pu-240 requires time-consuming chemical sample preparation steps that are not required for analysing Cs-137 (‘simple’ physical measurement). A reason for using Pu isotopes could be that the study area received significant Chernobyl fallout in 1986 (in addition to the global fallout with a peak in 1963-64), which would complicate the temporal reconstruction. However, this is not specifically addressed by the authors (nor supported by their measurement of both Pu-239 and Pu-240, 15 the ratio of which should directly provide the answer?) Of course, there could also be other (good) reasons to use Pu isotopes instead of Cs-137, but their clarification in the text would be appreciated.; *Thanks for the comment and suggested discussion points. We include a section at the beginning of the discussions:*

“4.1 ²³⁹⁺²⁴⁰Pu methodological benefits and limitations

The use of fallout radionuclides to determine soil redistribution patterns and rates over the past decades 20 has been used in many studies in various study areas around the world (see reviews: Mabit et al., 2014; Alewell et al., 2017; Evrard et al., 2020) and contributed substantially to understand soil degradation processes. However, the most frequently used fallout radionuclide ¹³⁷Cs faces upcoming limitations (Chernobyl fallout that adds on the global fallout over large parts of Europe and ongoing decay below detection limit of standard measuring devices; also see section 1) in the use as a soil redistribution tracer 25 (Evrard et al., 2020). The fallout radionuclide ²³⁹⁺²⁴⁰Pu has demonstrated its suitability to determine the recent soil redistribution history (since the 1960s; see review Alewell et al., 2017 and is a potential

alternative for ^{137}Cs as a soil redistribution tracer (Mabit et al., 2013; Alewell et al., 2017). In Europe, where large parts were re-contaminated by ^{137}Cs fallout of the Chernobyl accident (Evangelidou et al., 2016), additional information on the spatial change on the inventory is needed to derive accurate soil redistribution rates. Particularly in the area of the former GDR, almost no information that can be used
5 for a correction on the ^{137}Cs Chernobyl re-contamination are available (Evangelidou et al., 2016). The $^{239+240}\text{Pu}$ fallout caused by the Chernobyl disaster was very local (approximate radius of 100 km) and has a distinct fingerprint based on the $^{239}\text{Pu}/^{240}\text{Pu}$ ratio. While the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio of global fallout in the Northern Hemisphere is 0.180 ± 0.014 (Kelley et al., 1999), the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio soils that received high Chernobyl fallout is about twice as high (0.408 ± 0.003 , determined for soils within the 30
10 km exclusion zone of the Chernobyl reactor; Muramatsu et al., 2000; Boulyga and Becker, 2002). The 95% interval of confidence and average of the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio found in the soil samples of this study were 0.281 and 0.199, respectively. Hence, a relevant $^{239+240}\text{Pu}$ re-contamination by Chernobyl fallout can be ruled out for the study area. Another limitation for the use of ^{137}Cs as a soil redistribution tracer is the ongoing decay due to short half-life times that has already caused a substantial reduction of the
15 inventory. Due to lower activities, measuring devices of much higher complexity are needed in the future (Evrard et al., 2020). Decay is not an issue for ^{239}Pu and ^{240}Pu as both nuclides have long half-life times that allow for a quasi-unlimited use, however, it needs to be mentioned that sample preparation for $^{239+240}\text{Pu}$ ICP-MS measurements is much more laborious compared to the standard procedure of physical measurement ^{137}Cs measurements. “

20 L276: OK, I agree with you, but in the real landscapes, maybe the situation is a bit more nuanced, with the redistribution of organo-mineral complexes across landscapes?, we fully agree and added the following information to the text: “However, it needs to be mentioned that radionuclide associated particles are typically not transported as primary particles but in soil aggregate complexes (Hu and Kuhn, 2014; Hu et al., 2016), which has a pronounced effect on enrichment processes (Wilken et al.,
25 2017b).”

L281: Suggestion, = enrichment in fine particles containing Pu-239+240?; *we slightly modified the suggestion: “enrichment in fine particles of relatively high ²³⁹⁺²⁴⁰Pu activity”*

L286: OK but is this reasonable if the max erosion depth is 14.9 cm (previous section) and that radionuclide have been homogenized in the entire tilled layer (~25 cm)?; *Yes, we think this is very likely*
5 *as these highly eroded sites have a reduction of their inventory down to 12 Bq m⁻², which means 28% of their reference inventory (43 Bq m⁻²).*

L291: Suggestion, flow across?; *thanks done*

L294: Suggestion, was exceeded at 4 sampling locations?; *thanks done*

L299: Suggestion, managed; *thanks done*

10 L302: Suggestion, both water and tillage erosion processes; *thanks done*

L303: Suggestion, zone/area; *we changed the text to: “zone”*

L304: Could you define this concept at some point in the text?; *We changed the wording to: “geomorphological dynamics”*

L304: Do you mean the limited spatial extent where this process takes place?, *Thanks, we follow the*
15 *suggestion: “As a result of the small spatial extent where this process takes place, [...]”*

L306: Suggestion; an active process?, *we changed the text to “important”*

L308: Suggestion, in?; *thanks, done*

L310: Doesn't it seem a bit low compared to the erosion depths that you estimated at hilltop locations? Which 'peat' (in terms of spatial extent) are you referring to here?; *We refer to the inner kettle hole that*
20 *is not under arable use. Hence, 1.7 cm (53yr)⁻¹ of colluvium is just based on sediments that were exported from the arable land, therefore, solely transported by water. To make this clearer we changed the text as follows: “According to the model run using the reference parameterisation for water erosion, a colluvial layer of 1.7 cm (53 yr)⁻¹ would have been developed on top of the peat that has been exported from the arable part of the catchment (see Fig. 1) due to water transport over the past decades. Furthermore, we*
25 *highlight the inner peat area of the kettle hole in the study area Figure 1.*

- L313: In turn, ?; *we changed the text to “In contrast, “*
- L314: Suggestion, have been; *thanks, done*
- L320: Suggestion, cover; *thanks, done*
- L323: Suggestion, is partially mixed; *thanks, done*
- 5 L325: Suggestion, the most eroded sites; *thanks done*
- L326: Suggestion, is found at 102 cm depth on average?; *thanks, done*
- L329: Suggestion, have caused extensive soil redistribution over long periods?; *thanks, done*
- L332: Suggestion, reconsidered across a range of contrasted agricultural environments?; *thanks, done*
- L337: Suggestion, unfrequent extreme events; *thanks, we followed the suggestion but used the word*
 10 *“infrequent” instead*
- L338: Do you mean to connect hillslopes with the kettle hole?; *we change the text as follows: “Only infrequent extreme events exceed the critical runoff quantity to connect the arable hillslopes with the inner peat area of the kettle hole (Fig. 1),[...]”*
- L338: Suggestion, lead to; *thanks, done*
- 15 L339: Why using plural here (you investigated one catchment?); *thanks, you are right. We changed the text as follows: “Therefore, the study catchment shows a very limited hydrological and sedimentological connectivity between the cultivated area and kettle hole.”*
- L340: This was already written earlier in the text (although slightly differently), maybe avoid repetitions in the text at this stage; *we restructured the text and removed the redundant information as follows: “This*
 20 *statement is supported by surface runoff and sediment delivery monitoring in the study catchment (2015-2019) that has demonstrated that only very few rainfall events caused runoff and associated sediment delivery in the kettle hole (data not shown). Therefore, the study catchment shows a very limited hydrological and sedimentological connectivity between the cultivated area and kettle hole.”*

L343: could you be more specific here?; *we intentionally kept this statement loose to remain the interpretation of potential processes to the reader as we did not specifically investigate on these effects. Therefore, we would prefer to keep the statement as is but provide the reader additional literature:*

5 L344: Suggestion, impact? Extent? Magnitude?; *thanks, we changed the section title to “4.4 Impact of tillage erosion and scientific attention”*

L345: Do you mean above the tolerable soil loss rates?; *thanks, we reformulated the text as follows: “soil erosion in the study area exceeds the tolerable soil loss rates (according to Schwertmann et al., 1990: 6 Mg ha⁻¹ yr⁻¹ in the study region) and is mainly attributed to tillage erosion (Fig. 6 & 7).”*

L346: Suggestion, efficient or productivist?; *thanks, we changed the text to “productivist”*

10 L347: Suggestion, consolidation; *thanks, done*

L350: Suggestion, is rather large?; *thanks, done*

L350: Maybe rephrase here to facilitate reading?; *we reformulated the text as follows: “The average field size in the region (Quillow catchment: 22 ha) is rather large that has favoured big farming structures that utilises powerful machinery.”*

15 L351: Suggestion; likely does not receive...?; *we would like to keep the statement more a bit more drastic to underline the need and prefer to keep the wording as is.*

L351: Suggestion, its; *thanks, done*

L353: Wide spread?; *we removed the word “wide spread”*

20 L356: Maybe rephrase to clarify here; *we restructured the text to improve the reading flow: “Globally, tillage erosion has been recognized as an environmental threat in the hummocky young morainic regions that have shallow soils that are subject to dropping yields at hilltop locations (Canada: Pennock, 2003; Tiessen et al., 2007a; Tiessen et al., 2007b, North America: Li et al., 2007, 2008, Russia: Olson et al., 2002; Belyaev et al., 2005 and Northern Europe: Quine et al., 1994; Heckrath et al., 2005; Wysocka-Czubaszek and Czubaszek, 2014).”*

L358: Of note, this issue was investigated using radionuclide inventories and the SPEROS model in agricultural regions of central France (Chartin et al., 2013; 10.1016/j.catena.2013.06.006). Of note, these authors also demonstrated the dominance of tillage erosion processes (>90% of the total erosion) in these agricultural landscapes...; *thanks, for naming this very interesting paper! We included the reference here*
5 *and mention the results of the study in the introduction.*

L358: Suggestion, indeed?; *thanks, we removed the filler*

L361: Suggestion, the impacts of tillage erosion may not be as visible as those caused by water erosion?;
thanks, done

L363: Unclear what you mean here?; *we reformulated the text as follows: “[...]needs scientific*
10 *consideration and implementation in soil conservation management by policy makers.”*

L370: I didn't understand this when reading the previous text... do you mean that there would be some kind of 'sapping' process along the borders of the kettle hole? If so, this should be clarified in the text (here and above) and better underlined when describing Fig.7c/d...; *no there is no kind of 'sapping' process. Tillage forms a geomorphological flat area, what we call the 'kettle hole surrounding flat'.*
15 *Within this area, hydrological sinks are formed by tillage translocation that provide water retention capacity, which leads to a reduction of the flow velocity and therefore deposition of sediments that were transported by water. To clarify this, we reformulated the text as follows: “Furthermore, tillage erosion has a substantial impact on surface runoff. Tillage forms hydrological depressions at the downslope border between the cultivated field and the kettle hole that limits the hydrological and sedimentological*
20 *connectivity into the kettle hole and causes deposition of sediments that are transported by water.”*

L372: Suggestion, on the catchment hillslopes?; *thanks, done*

L375: Maybe you go too far in terms of extrapolation based on your results here? More than 1 m, I don't remember to have seen that above (you show a max of 17% of the profile truncation, if I understood it well?); *yes, the maximum erosion that was determined by using $^{239+240}\text{Pu}$ is 0.17 m. However, extreme*
25 *erosion and deposition of more than 1 m has taken place in the study area, which was detected based on lab analysis (e.g. CaCO_3) and soil prospection that clearly show buried fossil topsoil layers deeper than*

1 m. Our main argument is that we detected high erosion rates over the past decades (up to 0.17 m), however, those rates can still not explain the current soil degradation stage (>1 m soil profile truncation and burial). To make it clearer that 1 m of erosion or deposition was not determined using FRNs but soil prospection and lab analysis, we clarify this in the text as follows: “[...]soil degradation patterns
5 determined from soil prospection and chemical analysis that show[...]”

L376: Still, your study area can be described as a highly mechanised agro-ecosystem, right? Maybe you go again a bit too far here?; we see the point but are still convinced that tillage erosion is not just an issue for mechanised agro-ecosystems. Our results indicate an onset of tillage erosion before the main period of agricultural mechanisation (somewhere around the 1960th). We try to rephrase the statement more
10 nuanced: “This indicates that tillage erosion might not be a process that exclusively takes place in highly mechanised agro-ecosystems but is potentially causing pronounced soil degradation in smallholder farming structures.”

L378: Again, I would be more nuanced here as well...; we agree and changed the text accordingly: “Our results clearly underline that tillage erosion is a critically underrepresented soil degradation process that
15 can be the main soil redistribution driver on catchment scale.”

Understanding the role of water and tillage erosion from $^{239+240}\text{Pu}$ tracer measurements using inverse modelling

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Abstract

Soil redistribution on arable land is a major threat for a sustainable use of soil resources. ~~The soil redistribution process most~~
15 ~~The majority of soil redistribution~~ studies focus on ~~is~~-water erosion, while wind and tillage erosion also induce pronounced redistribution of soil materials. Especially, tillage erosion is understudied, as it does not lead to visible off-site damages. The analysis of on-site / in-field soil redistribution is mostly based on tracer studies, where~~as~~ radionuclide tracers (e.g. ^{137}Cs , $^{239+240}\text{Pu}$) from nuclear weapon tests are commonly used to derive the erosion history over the past 50-60 yr. Tracer studies allow to determine soil redistribution patterns, but integrate all ~~kinds-types~~ of soil redistribution processes and hence do not
20 allow to unravel the contribution of ~~different-individual~~ erosion processes. The aim of this study is to understand the contribution of water and tillage erosion leading to soil patterns found in a small hummocky ground moraine kettle hole catchment under intensive agricultural use. Therefore, $^{239+240}\text{Pu}$ derived soil redistribution patterns were analysed using an inverse modelling approach accounting for water and tillage erosion processes. The results of this analysis clearly point out that tillage erosion is the dominant process of soil redistribution in the ~~smallstudy~~ catchment, which also affects the
25 hydrological and sedimentological connectivity between arable land and the kettle hole. A topographic change up to 17 cm $(53\text{ yr})^{-1}$ in the eroded parts of the catchment is not able to explain the current soil profile truncation that exceeds the $^{239+240}\text{Pu}$ derived topographic change substantially. Hence, tillage erosion ~~already started before the is not limited to the time since the~~ onset of intense mechanisation since the 1960s. ~~In general, the study stresses the urgent need to consider tillage erosion as a major soil degradation process that can be the dominant soil redistribution process in sloped arable landscapes.~~
30 ~~In general, the study stresses the urgent need to consider tillage erosion as a very important soil degradation process that drives patterns of soil properties in our arable landscapes.~~

1. Introduction

Soil erosion is a major threat for the supply of soil related ecosystem services (Montanarella et al., 2016). Over the past decades, ~~particularly~~ the off-site effects associated with water erosion, like nutrient inputs from arable lands into inland waters (Pimentel and Burgess, 2013) or siltation of reservoirs (Krasa et al., 2019), were in the scientific and political focus. Within the European Union, the focus on off-site erosion effects is partly caused by the definition of the goals of the EU Water Framework Directive (EU 2000/60/ES) that focuses mainly on water bodies and floodplains but not on a fully integrated catchment management that would call for complex shared responsibilities between different administrative units. Thereby, other soil erosion drivers ~~than waterlike tillage and wind~~ are somewhat out of the scope of most studies. Tillage erosion is a mostly ignored soil erosion process (Fiener et al., 2018) that, however, substantially contributes to on-site effects on soil properties and hence agricultural productivity (Winnige, 2004; Nie et al., 2019). Van Oost et al. (2006) pointed out that tillage erosion rates are globally at least in the same order of magnitude as water erosion rates. In the Parisian Basin of France, Chartin et al. (2013) demonstrated the dominating role of tillage erosion. Particularly areas of a hilly-hummocky topography with short summit-footslope distances, such as young morainic areas, can be subject to pronounced in-field soil degradation patterns caused by tillage erosion (Winnige, 2004; Deumlich et al., 2017). Young morainic areas that are under intense arable cultivation and ~~corresponding associated~~ tillage erosion are widespread in northern Europe, Canada, northern USA, Russia and eastern Argentina. Measuring or monitoring water and tillage erosion is challenging as both processes are interlinked and ~~correspond to topography~~ are partly strongly controlled by topography (Van Oost et al., 2005b; Van Oost et al., 2006). ~~Water erosion calls for for~~ The quantification of water erosion requires a sufficiently long monitoring time (typically decades) to cover a statistically representative variation of rainfall events occurring on different land cover conditions (Fiener et al., 2019). Therefore, thousands of plot experiments, either driven by natural or artificial rainfall simulations, were carried out in different environments and different land cover conditions (Cerdan et al., 2010; Auerswald et al., 2014). Furthermore, a large number of small catchment studies were performed to ~~include-quantify~~ both erosion and depositional processes (for overview see; Fiener et al., 2019). However, soil erosion monitoring is mostly based on sediment delivery ~~outlet~~ monitoring, which cannot address catchment internal redistribution. In contrast, tillage erosion can only be measured based on the movement of beforehand applied tracers or by morphological change monitoring (for an overview of tillage erosion measuring techniques see; Fiener et al., 2018). However, these beforehand applied tracers and change monitoring methods cannot ~~assess the soil redistribution history of sites~~ provide a reconstruction of soil redistribution of the past. Natural or anthropogenic tracers in soils ~~that are inherent to specific soil layer characteristics~~ can be used to understand ~~the~~ soil redistribution (Fiener et al., 2018). Especially anthropogenic radionuclides (e.g. ^{137}Cs , $^{239+240}\text{Pu}$, ^{210}Pb , ^7Be) have demonstrated their ability to determine changes in topography (Mabit et al., 2014; Alewell et al., 2017; Deumlich et al., 2017). The force of atmospheric n ~~Nuclear weapon tests lifted-transported~~ radioisotopes up to outside the stratosphere troposphere (8–20 km m.a.s.l.; Aguado and Burt, 2015), where circulation mixture led to a n-almost-(regionally) homogeneous spatial distribution and ~~subsequent-corresponding~~ fallout on soils by rain via precipitation (Meusburger et al., 2016). The main period of atmospheric nuclear weapon tests is from 1953 to

1964 (Schimmack et al., 2001), while the ~~limited T~~test ~~B~~an ~~T~~treaty caused a rapid ~~end-decrease~~ of atmospheric bomb tests in 1963-1964 (Wallbrink and Murray, 1993; Evrard et al., 2020). This rapid ~~end-decrease~~ leads to a distinct peak in the activity of radioisotopes in soils, which enables the use of radioisotopes as redistribution tracers in soils (Alewell et al., 2017). The radioisotope ^{137}Cs has been used as a soil redistribution tracer in a large number of studies (e.g. Porto and Walling, 2012; Chartin et al., 2013; Evrard et al., 2020) and has become a widely used method in soil erosion science. However, the Chernobyl disaster ~~in (1986)~~ supplied additional radioactive fallout to soils across large areas of Europe ~~re-contaminated large areas of Europe with ^{137}Cs~~ (Evangelidou et al., 2016). For some years after the Chernobyl disaster, ~~a~~An unmixing of the Chernobyl fallout from the original ~~(1960s)~~ bomb peak signal was ~~able-possible~~ by the use of the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio (Lust and Realo, 2012). However, due to the short ^{134}Cs half-life of 2 yr (Schimmack et al., 2001), this method cannot be applied anymore. Hence, the use of ^{137}Cs as a soil redistribution tracer in Europe is associated with ~~large-uncertainties~~ and requires special attention concerning a potential Chernobyl contamination (Evangelidou et al., 2016). Furthermore, due to the ^{137}Cs half-life of about 30 yr, decay has already been led to a pronounced reduction (73% in 2020) of the activity until today (Alewell et al., 2017). Over the past decade, $^{239+240}\text{Pu}$ has been discussed and tested as an alternative radioisotopic tracer for soil erosion studies. Decay is not an issue as the half-life of ^{239}Pu and ^{240}Pu is long (^{239}Pu = 24000 yr; ^{240}Pu = 6563 yr-) and the $^{239+240}\text{Pu}$ contamination by the Chernobyl accident was spatially very limited (<100 km; Kashparov et al., 2004; Matsunaga and Nagao, 2009) and can be determined by the $^{239}\text{Pu}/^{240}\text{Pu}$ ratio (Alewell et al., 2014; Alewell et al., 2017).

Radionuclide tracers integrate soil erosion processes over time (e.g. since the bomb peak of the 1960s in case of ^{137}Cs and $^{239+240}\text{Pu}$), which somewhat averages out the large temporal variability of water (episodic nature) and tillage (mechanisation) erosion. However, the use of radioisotop~~iee~~ tracers integrates all ~~kinds-types~~ of soil redistribution processes and ~~lacks~~ insightsdoes not provide information on the relative contribution of the driving processes at play (e.g. water, tillage, wind). Unravelling the respective contributions of these different processes~~To unravel these different processes calls for~~ requires the use of an inverse modelling approach carrying out model runs ~~of-with~~ different parameterisations to alter the contribution and mechanisms of different soil redistribution drivers. There are only few models that take ~~-fusing~~ both water and tillage erosion processes into account. Physically oriented models like MCST-C (Wilken et al., 2017b) and LandSoil (Ciampalini et al., 2012) ~~work event-based and~~ simulate individual erosion events and are developed ~~for-to enhance~~ process understanding, while conceptual USLE based models (WaTEM/SEDEM: Van Oost et al., 2000; Van Rompaey et al., 2001) aim at a robust prediction of long-term soil erosion rates (Alewell et al., 2019). For an inverse modelling approach to unravel tillage and water erosion based on radionuclide tracer, it is necessary to use a parsimonious approach with a limited parameter space covered by~~and~~ available input data over five to six decades, which suggests the use of conceptual models.

In this study, we will determine the soil redistribution patterns in a small 4.2 ha catchment based on high-resolution $^{239+240}\text{Pu}$ measurements and analyse the contribution of water and tillage erosion processes based on an inverse modelling approach using a combined water and tillage erosion model. The general ~~aim-objective~~ is to unravel the importance of water and tillage erosion driving the~~for~~ current variability of soil properties in an intensively used arable landscape of north-eastern Germany.

2. Methods

2.1 Study area

The study area (53°21'2 N, 13°39'5 E) is situated in the hummocky ground morainic landscape of the Weichselian glacial belt ('young morainic area') of [north-eastern](#) Germany (Fig. 1). Characteristic for these landscapes are widespread closed depressions, so-called kettle holes, which result from a delayed melting of dead ice blocks. They are nowadays filled with mineral soil, (degraded) peat or water. The study area is part of a kettle hole catchment (4.2 ha; Fig. 1) ~~and consists of different~~ [showing a high topographical morphological variability covering convex hilltops, steep slopes and flat areas](#). The recent crop rotation is rape (*Brassica napus* L.)-winter wheat (*Triticum aestivum* L.)-winter barley (*Hordeum vulgare* L.)-winter barley, cultivated without cover crops, which is a typical conventional crop rotation that is adapted for the highly fertile soils of the Uckermark region. The mean arable land of a farm in the region is 352 ha, which is much larger compared to the mean of the State of Brandenburg (250 ha) and Germany (60 ha; Troegel and Schulz, 2018). These larger field sizes are ~~explained by mainly caused by~~ [land consolidation programmes implemented](#) in the 1960s during the socialist period. However, also before, agriculture in the region was already characterised by large scale farming structures ~~(East Germany in 1939: 44% of farms manage more than 5 ha of agricultural land; Wolz, 2013)~~ [and corresponding high agricultural mechanisation. In 1939, large farms that manage more than 100 ha of arable land cultivated 7% of the total arable area of Germany. In contrast, within the present-day federal states of Mecklenburg-Pomerania, Brandenburg \(study area location\) and Saxony-Anhalt, large farms](#) ~~cultivated 30% of corresponding arable land (Wolz, 2013).~~ The catchment is part of a single large field (54 ha), which is a size that can be frequently found in the region. The soils are developed from glacial till and vary with respect to their location in the landscape. Convex hilltops and steep slopes are dominated by extremely eroded A-C profiles (Calcaric Regosols, soil classification according to: IUSS, 2015), while Luvisols showing different degrees of erosion ~~and that~~ are typically situated at the up and mid slopes, the footslopes and depressions ~~show a sequence of Gleyic Colluvic Regosols, Mollie Gleysols and (buried) Terrie Histosols are dominated by Gleyic-Colluvic Regosols~~ (Fig. 1b; Sommer et al., 2008; Gerke et al., 2010). [These soils regularly reveal fossil surface horizons below 1 m depth \(fAh, fH\).](#) The closed kettle hole depression itself is built up by degraded Histosols and covered by a thin colluvial layer of mineral soil (40±8 cm mean, 25% and 75% quantile, $n=20$; based on soil auger prospection). The study area has a continental climate (Köppen: Dfb) with low annual precipitation (500 mm; Fiener et al., 2018) and high temperature amplitude (Jul $\pm 18^{\circ}\text{C}$, Jan $\pm -0^{\circ}\text{C}$; mean 8.9°C WMO-CLINO 1981-2010 for the meteorological stations Gruenow and Angermuende). Between 7 and 11 erosive rainfall events ~~per year~~ take place [each year](#) (Deumlich, 1999). In the region, maximum intensities up to 162 mm h^{-1} (10-min interval) were recorded during an extreme event in 2016 (Wilken et al., 2018).

2.2 Soil sampling design and preparation

The soil sampling design was organised ~~according to~~ [in](#) a regular 20 m x 20 m grid with at least one sampling point ~~of the~~ [transect line](#) exceeding the spatial extent of the catchment under study (Fig. 1) [to avoid boundary effects](#). To assess small scale

spatial variability (for distances of sampling points between 5 m and 20 m) a nested sampling approach was applied (Fig. 1; Hengl and MacMillan, 2019). Therefore, five densified sub-grids, located at different topographical locations (hilltop, ridge shoulder, moderately steep mid slope, steep mid slope, foot slope/valley; Fig. 1) were selected. In total, 209 locations were included in the sampling design.

135 The sampling points were located using a differential GPS (AgGPS™ 132; Trimble Navigation Ltd, Sunnyvale CA, USA) applying the SAPOS (LGB) correction signal. With respect to drill penetrability, soil sampling was carried out under moderately wet soil moisture conditions (range from 18% to 32% and 16% to 33% for topsoil and subsoil, respectively) in December 2015. Closed soil cores, using a steel drill containing a plastic liner (4.6 cm inner diameter), were driven by a percussion corer (Cobra TTe; Atlas Copco Power Techniques GmbH, Stockholm, Sweden) into the ground. ~~Soil cores driven down to a depth of 50 cm were taken at all 209 sampling points. To account for the vertical distribution of $^{239+240}\text{Pu}$ in the soil profile, ten replicate soil cores were taken at five up-hill and five foot slope locations (Fig. 1; P1-P10). Thus, overall 219 soil cores at 209 locations were taken (Fig. 1).~~

140 To minimize physical and biogeochemical disturbance, the soil cores were stored in a freezer until sample preparation. After complete thawing, the soil cores were separated into topsoil (Ap horizon) and subsoil. The separation was done by visual interpretation of soil horizon characteristics (colour, structural and density differences), showing a variation in topsoil thickness between 16 and 30 cm (mean of 23.5 cm) depending on the topographic position. Aliquot ($n=3$) samples for each topsoil and subsoil location were taken for gravimetric water content (weighted before and after drying at 105°C) and dry bulk density (known sample volume) measurements. The soil samples were air dried and subsequently sieved with a 2 mm mesh to separate stones from the fine soil.

150 2.3 $^{239+240}\text{Pu}$ measurements

All topsoil samples ($n=209$) were analysed, while 145 (~70 %) of the subsoil samples of highest topsoil activities were measured on their $^{239+240}\text{Pu}$ activity. This was done to reduce the inventory number of samples ~~that have with~~ Pu activities below the detection limit. Furthermore, at 10 locations higher-resolution (5 cm) depth increments were measured to assess the depth distribution of $^{239+240}\text{Pu}$ activity below the mixed plough layer.

155 The fallout radionuclides $^{239+240}\text{Pu}$ were used to estimate effective soil redistribution since the 1960s. Plutonium isotopes measurements were conducted following Calitri et al. (2019) based on the procedure of Ketterer et al. (2004). Before the mass spectrometry analysis, 10 g of milled fine earth were dry-ashed for at least 8 hours at 600°C to remove organic matter. Subsequently, the samples were spiked using 30 pg (c. 0.0044 Bq) of a ^{242}Pu tracer solution (NIST 4334). The sample leaching was applied using 16 M nitric acid (HNO_3) overnight at 80°C and subsequently filtered and adjusted to a concentration of 8 M HNO_3 . Plutonium species were adjusted to the Pu (IV) oxidation state using first an acidified $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ solution (2 mg ml^{-1} of leached solution) and subsequently a sodium nitrite (NaNO_2) solution (20 mg ml^{-1} of leached solution). The samples were heated at 75°C for two hours. Tetravalent Pu was separated from the leached solution using a Pu-selective TEVA resin (2 mg of TEVA per millilitre of leached solution). Following occasional agitation for two hours, the resin was collected in a

165 pipette tip equipped with a glass wool plug. This disposable column was rinsed with 2 M aqueous HNO₃ to remove unretained matrix elements [e.g. uranium (U)], then rinsed with 8 M HCl to elute thorium (Th) and finally rinsed again with 2 M aqueous HNO₃ (rinse volume $\frac{3}{4}$ ml per 30 mg of TEVA). Plutonium was eluted using 0.05 M aqueous ammonium oxalate. Data quality was evaluated through the analysis of blanks (soils or rocks devoid of Pu), duplicates and control samples of known ²³⁹⁺²⁴⁰Pu activities (Standard Reference material 4350b – River sediment for radioactivity measurements from NIST). Activities of ²³⁹⁺²⁴⁰Pu were measured using a Thermo X Series II quadrupole ICPMS, located at Northern Arizona University. 170 The ICPMS instrument is equipped with an APEX HF high-efficiency sample introduction system. The masses of ²³⁹Pu and ²⁴⁰Pu in the samples were converted into the summed activity ²³⁹⁺²⁴⁰Pu.

2.4 ²³⁹⁺²⁴⁰Pu based soil erosion assessment

We applied the proportional conversion approach of Walling et al. (2011). Erosion is calculated following equations 1 and 2:

$$SL_i = 10 * BD_i * TD_i * RR_i * T^{-1} \quad (1)$$

175 where SL_i is the mean annual soil loss in Mg ha⁻¹ yr⁻¹, BD_i is the soil bulk density in kg m⁻³, TD_i is the vertical depth of the Ap horizon in m (tillage depth), RR_i is the relative reduction of the reference inventory of the ²³⁹⁺²⁴⁰Pu inventory and T is the years that are elapsed since the end of atmospheric nuclear weapon tests (mainly since 1964).

$$RR_i = \frac{(Pu_{ref} - Pu_i)}{Pu_{ref}} \quad (2)$$

where Pu_i is the inventory at sampling point i and Pu_{ref} is the reference inventory of undisturbed sites in Bq m⁻² ([see implementation section 2.5](#)).

180 For a three-dimensional representation of the ²³⁹⁺²⁴⁰Pu redistribution by water and tillage erosion, SPEROS-Pu was developed that is based on a modified version of the SPEROS-C model (Van Oost et al., 2005a; Fiener et al., 2015; Nadeu et al., 2015). ~~We utilized the water and tillage erosion modules of the SPEROS model that are based on WaTEM/SEDEM (Van Oost et al., 2000; Van Rompaey et al., 2001).~~ SPEROS-Pu is a spatially explicit water and tillage erosion model. Water erosion is simulated based on a gridded application of the Revised Universal Soil Loss Equation (RUSLE: Renard et al., 1996) coupled with a sediment transport and deposition approach. Erosion is calculated according to a slightly modified RUSLE approach, 185 transport and deposition is based on the grid cell specific local transport capacity TC (kg m⁻¹ yr⁻¹), which multiplies RUSLE factors with a transport capacity coefficient (k_{tc} ; in m)

$$TC = k_{tc} * R * C * K * L * S * P \quad (3)$$

where R , C , K , L , S and P are the RUSLE factors (see Renard et al., 1996).

The tillage erosion module of SPEROS-Pu follows a diffusion-type equation adopted from Govers et al. (1994) that derives tillage erosion based on change in topography and management specific coefficients

$$Q_{til} = -k_{til} \frac{\Delta h}{\Delta x} \quad (4)$$

190 where Q_{til} is the soil flux in $\text{kg m}^{-2} \text{yr}^{-1}$, Δh is the elevation difference in m, Δx is the horizontal distance in m while k_{til} is the tillage transport coefficient in $\text{kg m}^{-1} \text{yr}^{-1}$

$$k_{til} = BD_i * TD_i * x_{til} \quad (5)$$

where x_{til} is the tillage translocation distance in m (for BD_i and TD_i see description of eq. 1).

The representation of the $^{239+240}\text{Pu}$ redistribution in the SPEROS-Pu model is three dimensional and accounts for $^{239+240}\text{Pu}$ source area depletion and corresponding redistribution of depleted sediments. The horizontal distribution of $^{239+240}\text{Pu}$ is grid
195 based, while the vertical distribution is represented by ten 10-cm depth layers. The two uppermost layers are assumed to be homogeneously mixed by tillage operations and have the ~~same~~ average $^{239+240}\text{Pu}$ activity of the upper two soil layers. At the beginning of the simulation, the $^{239+240}\text{Pu}$ reference activity is homogeneously distributed within the mixed plough layer and over the entire catchment. Subsequently, the local $^{239+240}\text{Pu}$ inventory is altered by soil redistribution processes. Soil erosion processes lead to a reduction of the $^{239+240}\text{Pu}$ inventory per m^2 due to ~~a~~ soil and ~~corresponding associated~~ $^{239+240}\text{Pu}$ loss, ~~which~~
200 ~~causes-and~~ mixing in of non-contaminated subsoil. Deposition adds contaminated material on top and increases the $^{239+240}\text{Pu}$ inventory.

2.5 Implementation and inverse modelling

Soil redistribution based on $^{239+240}\text{Pu}$ measurements

Spatially distributed top~~soil~~ and subsoil bulk density and tillage depth information for each individual sampling location ~~were~~
205 ~~was~~ applied to the proportional conversion approach. The reference inventory of undisturbed sites follows the value determined by Calitri et al. (2019) who found a $^{239+240}\text{Pu}$ inventory of $43 \pm 3 \text{ Bq m}^2$ based on four sites that did not show profile morphological or geochemical indication for soil redistribution at a location 8.5 km apart from the study area. To ~~account~~ ~~address for~~ the uncertainty inherent to the reference ~~measurements determined in close proximity but not at site~~, a reference range from 40 to 46 Bq m^2 was accounted for in the simulations.

210 The point data of the $^{239+240}\text{Pu}$ inventory for the depth 0-50 cm was geostatistically (block kriging) interpolated for a gridded spatial representation that matches the spatial resolution of the soil redistribution model (5 m x 5 m). Block kriging was used to reduce small-scale scattering that is naturally inherent to soil cores of 4.6 cm diameter that are supposed to represent a decametre scale. Different block sizes were tested ~~forin~~ the kriging approach. A block size of-and-finally 20 m was selected that matches the sampling resolution and ~~has-shown-did~~ not to ~~lead-causein~~ over-smoothing of the interpolation result. The
215 theoretical semivariogram model (Fig. 2) was fitted using all 209 sampling points, including the nested samples to account for variations over short distances. However, the input data of the interpolation itself solely uses the regular 20 m x 20 m grid points. The interpolation and geostatistical analysis were carried out using the statistical software GNU R (R-Core-Team, 2015), version 3.5.3 and the add-on package gstat (Pebesma, 2004).

Inverse modelling of water and tillage erosion

220 An inverse modelling approach was used to understand the proportion of water and tillage erosion that is inherent to the $^{239+240}\text{Pu}$ based soil erosion map. The inverse modelling iterates three parameter sets from low to extreme settings over the 53 yr modelling period (1964-2016): (i) the k_{til} tillage translocation coefficient (as given for illustration in Eq. 5), (ii) the product of all RUSLE factors (as given in Eq. 3; in the following referred to water erosion strength) and (iii) the water erosion transport capacity coefficient k_{tc} (see Table 1) that controls the transport distance and is the standard calibration parameter of the model.

225 While changes of the tillage translocation coefficient and water erosion strength only alter the quantity of soil redistribution, the k_{tc} has a pronounced impact on spatial patterns of modelled soil redistribution. The parameter range covers very low to extreme soil redistribution rates ($k_{til} \text{ max} \div \equiv 1000 \text{ kg m}^{-1}$; $k_{tc} \text{ max} \div \equiv 500 \text{ m}$; RUSLE-factors product deviation $\equiv 100\%$; see Table 1). The water erosion reference parameterisation for k_{tc} is 150 m (Van Oost et al., 2003). The interplay of parameter combinations was assessed in 35722 different model runs (Table 1).

230 To determine the model match, different goodness-of-fit parameters were calculated that compare the interpolated $^{239+240}\text{Pu}$ raster map against the results ~~of the raster results~~ calculated by the inverse modelling approach. To address the high spatial variability of the $5 \text{ m} \times 5 \text{ m}$ raster by raster comparison, a classification of results was carried out. Therefore, mean values were calculated based on the cells that fall into a specific class of the interpolated map of $^{239+240}\text{Pu}$ inventories. The classification covers 20 classes with 2.5 Bq m^{-2} steps from 17.5 to 65 (and a class >65) Bq m^{-2} of $^{239+240}\text{Pu}$. First, the spatial

235 correlation was calculated for both the raster by raster comparison and the classified results. Second, the classified results of the inverse modelling were tested using goodness-of-fit parameters that take absolute differences between observed and predicted values into account (RMSE, model efficiency coefficient: MEF according to Nash and Sutcliffe, 1970). As a last step, the results of the measured and modelled $^{239+240}\text{Pu}$ inventories were transferred into tillage and water erosion maps (given in Mg ha^{-1} and topographical change in cm) applying the proportional conversion approach (see chapter 2.4 $^{239+240}\text{Pu}$ based soil

240 erosion assessment).

3 Results

3.1 $^{239+240}\text{Pu}$ activities and inventories

The topsoil and subsoil $^{239+240}\text{Pu}$ activities differ substantially from each other and show distinct spatial patterns according to their topographic position. All topsoil samples ($n \div \equiv 209$) showed a $^{239+240}\text{Pu}$ activity above the detection limit, while 7 of the

245 145 subsoil samples fall below the detection limit ($< 0.002 \text{ Bq kg}^{-1}$). Those 7 samples are all located at positions with $^{239+240}\text{Pu}$ inventories below the lower reference boundary (40 Bq m^{-2}). The average $^{239+240}\text{Pu}$ activity is $0.078 \pm 0.016 \text{ Bq kg}^{-1}$ and $0.035 \pm 0.038 \text{ Bq kg}^{-1}$ for topsoil and subsoil, respectively. All The high-resolution depth profiles (5 cm increments) at erosional sites ~~distinctively~~ show a sharp reduction of the $^{239+240}\text{Pu}$ activity below the plough layer ~~at erosional sites~~ (Fig. 3). In turn, while depositional sites show more complex depth distributions. Location P4 (Fig. 3) does not show a drop in activity until a

250 depth of 0.5 m, while P1 (and partly P3) shows an increase of the $^{239+240}\text{Pu}$ activity with depth that is potentially ~~goes back~~
~~to~~caused by Pu enrichment processes during lateral transport or the deposition of already Pu depleted source material into the
topsoil. The topsoil $^{239+240}\text{Pu}$ /reference (43 Bq m⁻²) ratio indicates soil erosion related $^{239+240}\text{Pu}$ depletion or enrichment
according to a ratio lower and higher than one, respectively. The highest depletion (min $\underline{=}$ -0.28; 5% quantile $\underline{=}$ 0.37) can be
255 to the flat surroundings of the kettle hole (Fig. 1), where topsoil material was potentially deposited by both water and tillage
soil redistribution processes. At 14 (of 209) sampling locations, a higher subsoil than topsoil $^{239+240}\text{Pu}$ activity was found,
which points at deposition of $^{239+240}\text{Pu}$ depleted sediments. The majority of these locations show enriched $^{239+240}\text{Pu}$ activities
in the subsoil (119 of 14), ~~that goes in line with higher subsoil activities compared to the topsoil (13 of 14), which is caused~~
~~by deposition of $^{239+240}\text{Pu}$ depleted sediments~~. These locations are all, except for one, located at the kettle hole surrounding
260 plateau where both water and tillage erosion show-cause deposition. The $^{239+240}\text{Pu}$ depletion of sampling points (including
locations outside the study catchment) goes down to 12 Bq m⁻² while the highest five locations exceed 86 Bq m⁻², which means
that the $^{239+240}\text{Pu}$ inventory has more than doubled compared to the reference inventory of 43 Bq m⁻² ~~and therefore a tillage~~
~~depth of 25 cm~~, which has to be attributed to enrichment processes. ~~Therefore, these~~ As the enrichment processes inherent to
these -five locations cannot be corrected, the locations were excluded from the analysis.

265 The distribution of the interpolated maps covering the study catchment shows that substantially more locations fall below the
reference inventory than exceeding it (Fig. 4). This indicates that a larger area is subject to erosion processes compared to
depositional processes.

3.2 $^{239+240}\text{Pu}$ measurements vs. inverse water and tillage erosion modelling

To understand the drivers of current $^{239+240}\text{Pu}$ and associated soil degradation patterns, an inverse modelling was carried out
270 that was qualitatively-quantitatively analysed by goodness-of-fit parameters. The spatial correlation between the $^{239+240}\text{Pu}$
derived patterns and the modelled best knowledge soil ~~erosion~~redistribution, including both water and tillage erosion, is only
moderate (R^2 - $\underline{=}$ 0.45, Rho = 0.73) on a raster by raster comparison (n- $\underline{=}$ 1699, 5 m x 5 m grid points; Fig. 5a). ~~This is not~~
~~surprising due to short term and small scale topographic dynamics (e.g. roughness, wheel tracks, vegetation artefacts) that~~
~~transfer into model results as a static digital elevation model is used~~. To reduce ~~this~~ small-scale variability and understand the
275 average goodness-of-fit, the inverse modelling results were classified according to the measured $^{239+240}\text{Pu}$ activity. The
classified results average out the spatio-temporal dynamics and show a very high correlation (R^2 - $\underline{=}$ 0.95, Rho $\underline{=}$ -0.99; Fig.
5b), which pinpoints-illustrates the great agreement of the spatial soil redistribution patterns between the $^{239+240}\text{Pu}$
measurements and the model results.

While the analysis of the spatial correlation is a relative comparison, the absolute deviation is taken into accountconsidered
280 according to the MEF (model efficiency coefficient; Nash and Sutcliffe, 1970; 1 = perfect prediction, 0 = as good as mean of
all measurements, < 0 = worse than mean). The quality of model predictions shows hardly any sensitivity to water erosion
related parameterisations (k_{tc} and erosion strength; Figure 6c). In contrast, the tillage erosion strength, represented by k_{til}

parameter iterations, showed a substantial impact on the MEF (Fig. 6a & b). A MEF better than 0.8 and RMSE below 6.5 Bq m⁻² were found for a k_{til} range from 225 to 475 kg m⁻¹, while the best model fit was found for a k_{til} of 350 kg m⁻¹ achieving a MEF of 0.87 and, while the a corresponding RMSE of 5.2 Bq m⁻². The best model fit was found without the contribution of water erosion. The highest impact on the best-fit model run was a 0.31 MEF-reduction by an extreme water erosion parameterisation ($k_{tc} = 500$, water erosion strength = 200%).

Soil redistribution determined by the proportional conversion approach using ²³⁹⁺²⁴⁰Pu measurements, indicates substantial ~~morphodynamics~~ geomorphological dynamics in the study catchment over the past decades. Soil erosion at hilltop locations is shown to ~~be reach~~ up to 14.9 cm (43 Bq m⁻² reference; 40 Bq m⁻² reference \Rightarrow 14.1 cm; 46 Bq m⁻² reference \Rightarrow 15.6 cm), while deposition ~~partly can~~ builds a colluvium layer up with a maximum thickness of 21.5 cm (43 Bq m⁻² reference; 40 Bq m⁻² reference \Rightarrow 24.9 cm; 46 Bq m⁻² reference \Rightarrow 18.6 cm) ~~colluvium~~ over the past 53 yr. The inverse modelling stresses that substantial soil erosion, which takes place over large areas, is almost exclusively attributed to tillage translocation (modelled max. water erosion \Rightarrow 3.8 cm (53 yr)⁻¹ vs. max. tillage erosion \Rightarrow 13.5 cm (53 yr)⁻¹; Fig. 7c, d). In turn, both processes contribute to deposition in the kettle hole surrounding flats (max. water deposition \Rightarrow 27.1 cm (53 yr)⁻¹ vs. max. tillage deposition \Rightarrow 22.4 cm (53 yr)⁻¹; Fig. 7c, d).

4 Discussions

4.1 ²³⁹⁺²⁴⁰Pu methodological benefits and limitations

The use of fallout radionuclides to determine soil redistribution patterns and rates over the past decades has been used in many studies in various study areas around the world (see reviews: Mabit et al., 2014; Alewell et al., 2017; Evrard et al., 2020) and contributed substantially to understand soil degradation processes. However, the most frequently used fallout radionuclide ¹³⁷Cs faces upcoming limitations (Chernobyl fallout that adds on the global fallout over large parts of Europe and ongoing decay below detection limit of standard measuring devices; also see section 1) in the use as a soil redistribution tracer (Evrard et al., 2020). The fallout radionuclide ²³⁹⁺²⁴⁰Pu has demonstrated its suitability to determine the recent soil redistribution history (since the 1960s; see review Alewell et al., 2017) and is a potential alternative for fills the gap of upcoming limitations in the use of ¹³⁷Cs as a soil redistribution tracer (Mabit et al., 2013; Alewell et al., 2017). In Europe, where large parts were re-contaminated by ¹³⁷Cs fallout of the Chernobyl accident (Evangelidou et al., 2016), additional information on the spatial change on the inventory is needed to derive accurate soil redistribution rates. Particularly in the area of the former GDR, almost no information that can be used for a correction on the ¹³⁷Cs Chernobyl re-contamination are available (Evangelidou et al., 2016). The ²³⁹⁺²⁴⁰Pu fallout caused by the Chernobyl disaster was very local (radius <100 km) and has a distinct fingerprint based on the ²³⁹Pu/²⁴⁰Pu ratio. While the ²⁴⁰Pu/²³⁹Pu ratio of global fallout in the Northern Hemisphere is 0.180±0.014 (Kelley et al., 1999), the ²⁴⁰Pu/²³⁹Pu ratio soils that received high Chernobyl fallout is about twice as high (0.408±0.003, determined for soils within the 30 km exclusion zone of the Chernobyl reactor; Muramatsu et al., 2000; Boulyga and Becker, 2002). The 95% interval of confidence and average of the ²⁴⁰Pu/²³⁹Pu ratio found in the soil samples of this study were 0.281 and 0.199,

315 respectively. Hence, a relevant $^{239+240}\text{Pu}$ re-contamination by Chernobyl fallout can be ruled out for the study area. Another
limitation for the use of ^{137}Cs as a soil redistribution tracer is the ongoing decay due to short half-life times that has already
caused a substantial reduction of the inventory. Due to lower activities, measuring devices of much higher complexity are
needed in the future (Evrard et al., 2020). Decay is not an issue for ^{239}Pu and ^{240}Pu as both nuclides have long half-life times
320 that allow for a quasi-unlimited use, however, it needs to be mentioned that sample preparation for $^{239+240}\text{Pu}$ ICP-MS
measurements is much more laborious compared to the standard procedure of physical measurement ^{137}Cs measurements.
However, some methodological limitations persist that need to be taken into account.

Enrichment processes, due to selective transport of soil constituents that fallout radionuclides are preferentially associated with, are a critical issue for the use of most (e.g., $^{239+240}\text{Pu}$, ^{137}Cs , ^{210}Pb) radionuclide tracers (Parsons and Foster, 2011; Mabit et al., 2014; Alewell et al., 2017). While ^{137}Cs is mainly associated with clay particles, $^{239+240}\text{Pu}$ binds to soil organic matter
325 and oxides (Alewell et al., 2017) that are less affected by selective water transport and corresponding $^{239+240}\text{Pu}$ enrichment (Meusburger et al., 2016; Xu et al., 2017). However, it needs to be mentioned that radionuclide associated particles are typically
not transported as primary particles but in soil aggregate complexes (Hu and Kuhn, 2014; Hu et al., 2016), which has a
pronounced effect on enrichment processes (Wilken et al., 2017b). Nevertheless, the $^{239+240}\text{Pu}$ activity at depositional sites, that are redistributed by water (transport by tillage is typically assumed to be non-grain size selective; Fiener et al., 2018), can be
330 higher in relation to the activity of the source material. A soil profile that shows a distinct indicator of enrichment processes in this study is sampling profile P1 (25-45 cm; Fig. 3) that is situated in the kettle hole surrounding flat. Hence, enrichment in
fine particles of relatively high $^{239+240}\text{Pu}$ activity is to some extent also an issue within this study that causes an overestimation of deposition. A particle size correction factor was not applied as topsoil enrichment (topsoil $\text{Bq m}^{-2} > \text{ref. } 43 \text{ Bq m}^{-2}$) was exclusively found at very few sampling locations (<6%) in the kettle hole surrounding flats. Furthermore, the mean topsoil
335 ratio of enriched sediments is moderate (1.2) and supports the general assumption that $^{239+240}\text{Pu}$ is less affected by selective transport compared to ^{137}Cs (Alewell et al., 2017) and that transport by tillage is non-grain size specific. The counteracting process of enrichment is the deposition of $^{239+240}\text{Pu}$ depleted sediments that are transported from highly eroded locations. Such highly depleted locations can be found at the hilltops of the study area (Fig. 7b). Hence, the hilltops are the main source of highly depleted sediments that are deposited in kettle hole surrounding flats. However, the minimum horizontal distance from
340 the hilltops to the kettle hole surrounding flat is roughly about 70 m and the approximate tillage translocation distance 0.5 to 1 m per pass (Fiener et al., 2018). Hence, deposition of depleted $^{239+240}\text{Pu}$ material has to be mainly attributed to surface runoff that can bridge-flow across longer transport distances. SPEROS-Pu takes depletion of deposited sediments into account but does not address enrichment processes. Furthermore, a maximum soil sampling depth down to 50 cm was carried out within this study that technically allows to derive a maximum depositional depth of 25 cm using the proportional conversion approach
345 of Walling et al. (2011), which is at four sampling locations exceeded was exceeded at four sampling locations. Nevertheless, also with a deeper soil sampling, it would be arguable if these potentially enriched or depleted sampling locations should be excluded from the statistical analysis, like it was done for extreme depositional locations (4 sampling locations) within this study.

4.2 Using $^{239+240}\text{Pu}$ and inverse modelling to understand the recent soil erosion history

350 Within the intensively ~~used-managed~~ study catchment, substantial $^{239+240}\text{Pu}$ derived soil redistribution was found with soil loss up to $45 \text{ Mg ha}^{-1} \text{ yr}^{-1}$ (ref. 43 Bq m^{-2} ; ref. 40 and $46 \text{ Bq m}^{-2} \Rightarrow 43$ and $47 \text{ Mg ha}^{-1} \text{ yr}^{-1}$) and sediment deposition up to $65 \text{ Mg ha}^{-1} \text{ yr}^{-1}$ (ref. 43 Bq m^{-2} ; ref. 40 and $46 \text{ Bq m}^{-2} \Rightarrow 75$ and $56 \text{ Mg ha}^{-1} \text{ yr}^{-1}$). Very high deposition can only be found in the spatially narrow area of the kettle hole surrounding flat where both water and tillage erosion processes ~~water and tillage erosion~~ lead to deposition (Fig. 1). The kettle hole surrounding flat is a spatially narrow area, but the only region-zone where water

355 erosion substantially contributes to pronounced ~~morphodynamics-geomorphological dynamics~~ (Fig. 7d). As a result of the small spatial ~~contribution extent where this process takes place~~, the inverse modelling shows hardly any sensitivity on goodness-of-fit changes in reaction to the variation in model parameterisations (Fig. 6c). Nevertheless, sediment deposition and delivery by surface runoff is an relevant important process in the study area. Evidence for ~~runoff-based runoff-based~~ sediment delivery is a colluvial layer covering the peat in the kettle hole with an average depth of 40 cm. This sediment delivery

360 into the kettle hole cannot be explained by the inverse modelling of water erosion applying a reasonable parameter range ~~of the inverse modelling~~. Hence/Therefore, we assume the reference parameterisation for the region given by the state of Brandenburg of the region needs to be assumed as the most appropriate ($k_{tc} \Rightarrow 150 \text{ m}$ and RUSLE parameters according to Tab. 1). According to the model run using the reference parameterisation for water erosion, a colluvial layer of 1.7 cm (53 yr^{-1}) would have been developed on top of the peat that has been exported from the arable part of the catchment (see Fig. 1)

365 due to water transport over the past decades. This indicates a long water erosion history before the 1960s. This is not surprising as bare soil conditions and erosive rainfall events have taken place since the onset of arable use ~~approx approximately- 1_k-yr-~~ before present (Van der Meij et al., 2017; Kappler et al., 2018). In ~~turn contrast~~, tillage erosion is typically assumed to be a process that is linked to recent developments of increasing mechanical forces that ~~are-have been~~ applied to soils over the past century (Sommer et al., 2008; Calitri et al., 2019). Within this study, a maximum topographical change by hilltop erosion up

370 to 17 cm (53 yr^{-1}) was determined. In a review on tillage erosion by Van Oost and Govers (2006), tillage translocation coefficients of 44 experiments were reported for different tillage practices. This resulted in a mean k_{til} a mean of 234 kg m^{-1} ($\pm 5\%$ percentile $\Rightarrow 30 \text{ kg m}^{-1}$; 95% percentile $\Rightarrow 640 \text{ kg m}^{-1}$) for mouldboard and chisel plough. Within this study, a tillage translocation coefficient of 350 kg m^{-1} per year was determined. The tillage translocation coefficients, determined by the inverse modelling approach, are rather high compared to other studies considering that fallout radioisotopic tracer approaches

375 ~~integrate-cover~~ a phase of high mechanical development from low to high power farming machines (Sommer et al., 2008; Keller et al., 2019). Although recent tillage translocation rates are rather high they cannot explain the soil depth patterns that are visible by augerings. In the study region, it can be observed that tillage erosion mainly affected hilltops. Calcaric glacial till is approaching the surface by soil profile truncation and is partially mixed into the plough layer. Within the study catchment, this is the case for 20 sampling locations ($\text{CaCO}_3 > 0.5\%$) that are also indicated as strongest-the most eroded sites by the

380 $^{239+240}\text{Pu}$ measurements and the inverse modelling. Non-eroded reference profiles ($n \Rightarrow 210$) in the region show the parent material (calcaric glacial till) is found at 102 cm depth on average (van der Meij et al., 2019). Hence, less than 17% of soil

depth reduction can be attributed to most recent process dynamics. This suggests that traditional hand or cattle based tillage systems, which are used since the beginning of arable agriculture in the region (1k yr BP; Kappler et al., 2018), must ~~have~~ caused ~~d severe extensive~~ soil redistribution ~~in the over~~ long ~~run periods~~. This suggests that tillage erosion might be the dominant process even without mechanized soil tillage, which is the common practice in most developing countries that also partly cultivate very steep slopes. Therefore, the general assumption of tillage erosion being only an issue for highly mechanised agricultural systems (Van Oost et al., 2006) might need to be ~~re-evaluated~~ reconsidered across a range of contrasted agricultural environments.

4.3 Interplay of sediment redistribution by water and tillage

The inverse modelling has shown that soil redistribution by water has only a minor impact on erosion processes in the study area. However, sediment deposition by water has a complex interplay with tillage translocation (kettle hole surrounding flat; Fig. 7). Very high deposition by tillage translocation towards the field-kettle hole edge (typically >1 m known from soil augering; Kappler et al., 2018) builds up local hydrological depressions (Fig. 1 & 7). Only ~~large and rare~~ infrequent extreme events exceed the critical runoff quantity to connect the arable hillslopes with the inner peat area of ~~enter~~ the kettle hole (Fig. 1), while the majority of events ~~force~~ lead to deposition of sediments in the kettle hole surrounding flats (see Fig. 7d; Fig. 3, P1 and P4). ~~Therefore, these catchments show a very limited hydrological and sedimentological connectivity between the arable land and kettle holes. This statement is supported by~~ Surface runoff and sediment delivery monitoring in the study catchment (2015-2019) ~~have that has~~ demonstrated that only very few rainfall events ~~produced sufficient runoff to exceed the retention capacity of the kettle hole surrounding flats and~~ caused runoff and associated sediment delivery into the kettle hole (data not shown). ~~Therefore, the study catchment shows a very limited hydrological and sedimentological connectivity between the cultivated area and kettle hole.~~ Hence, tillage translocation in hummocky young morainic regions does also have a pronounced impact on hydrology and biogeochemistry.

4.4 Relevance of tillage erosion and scientific attention

Our results clearly indicate that soil erosion in the study area ~~is high~~ exceeds the tolerable soil loss rates (according to Schwertmann et al., 1990 = 6 Mg ha⁻¹ yr⁻¹ in the study region) and is mainly attributed to tillage erosion (Fig. 6 & 7). During the socialist era (1949-1990), ~~efficient-productivist~~ agricultural management strategies were implemented that included land ~~reformation-consolidation~~ to merge large fields and the use of heavy farming machines (Forstner and Isermeyer, 2000; Wolz, 2013). For instance, annual ploughing was combined with a recommended practice of episodically using a paraploUGH (tillage depth ~0.6 m; Fachbereichsstandard-DDR, 1985) to break the plough pan. The average field size in the region (Quillow catchment \approx 22 ha) ~~still consists of large field sizes~~ is rather large, this that has ~~established-favoured~~ conventional big farming structures ~~that utilises that is based on~~ powerful machinery. However, tillage erosion does not receive reasonable scientific attention (Fiener et al., 2018), even if ~~the its~~ effects on yields (Oettl et al., 2020), nutrient and carbon cycling (Wilken et al., 2017a; Zhao et al., 2018; Nie et al., 2019) and soil hydrology (Herbrich et al., 2017) are widely known. Globally, tillage erosion

has been recognized as an environmental threat in the ~~wide spread~~ hummocky young morainic regions ~~that have shallow soils~~
415 ~~that are subject to dropping yields at hilltop locations of Canada: , North America , Russia and Northern Europe (Canada:~~
~~Pennock, 2003; Tiessen et al., 2007a; Tiessen et al., 2007b, North America: Li et al., 2007, 2008, Russia: Olson et al., 2002;~~
~~Belyaev et al., 2005 and Northern Europe: Quine et al., 1994; Heckrath et al., 2005; Wysocka-Czubaszek and Czubaszek,~~
~~2014) that have shallow soils that are subject to dropping yields at hilltop locations.~~ Most arable regions are subject to
pronounced tillage erosion ~~(e.g. illustrated in the landscape by tillage banks along downslope field borders)~~(e.g. illustrated in
420 the landscape by tillage banks along downslope field borders; Chartin et al., 2013)), but may not show a pronounced impact
on yields (Lal et al., 2000). ~~For instance, IL~~oess derived soils with a homogeneous grain size distribution for several meters of
depth do not show major differences in soil structure (Blume et al., 2016), while nutrient losses are compensated by fertilizer
applications. Another reason for not being a prominent soil degradation mechanism might be that ~~the impacts of~~ tillage erosion
~~is are simply~~ not as visually ~~visible as those caused by~~ ~~present as~~ water erosion, which leads to rapid topographical dynamics (rills
425 and gullies) and off-site damages (muddy floods, siltation). However, tillage erosion is a highly important soil redistribution
process, taking place on the majority of sloped arable fields that urgently needs ~~to be (re)drawn on the map~~scientific
consideration of the scientific community and implementation in soil conservation management by policy makers.

5 Conclusions

~~I~~Within this study, ²³⁹⁺²⁴⁰Pu was used as a tracer to ~~determine-reconstruct~~ soil redistribution in a hummocky young morainic
430 study catchment under intense arable use. To understand the role of water and tillage erosion on soil degradation patterns, an
inverse modelling approach was carried out in the study catchment. The results clearly show that recent soil degradation in the
study area is dominated by tillage translocation. Furthermore, tillage erosion has a substantial impact on surface hydrology
runoff. Tillage forms hydrological depressions ~~at the~~ ~~deposition at the~~ downslope border between the cultivated field and
the kettle hole that limits the ~~edge between the field border and the kettle hole builds up hydrological depressions. These~~
435 ~~hydrological depressions limit the surface runoff~~hydrological and sedimentological connectivity into the kettle hole and ~~force~~
causes deposition of sediments that are transported by water. Soil redistribution by water has no major contribution to soil loss
~~at-on~~ the catchment hillslopes ~~of the catchment~~, but causes pronounced deposition in the spatially narrow area of the kettle
hole surrounding flat. Within this study, soil erosion up to 17 cm (53 yr)⁻¹ and deposition exceeding 25 cm (53 yr)⁻¹ of recent
~~morphodynamics-geomorphological dynamics~~ (since 1960s) were found. However, even these relatively high erosion rates
440 cannot explain the current soil degradation patterns determined from soil prospection and chemical analysis that show both
profile soil truncation and colluviation larger than one meter. This indicates that tillage erosion ~~might is not be~~ a process that
exclusively takes place in highly mechanised agro-ecosystems but is potentially causing pronounced soil degradation in
smallholder farming structures. Our results clearly underline that tillage erosion is a critically underrepresented soil degradation
process that can be the main soil redistribution driver on catchment scale ~~that takes place on the majority of sloped cultivated~~
445 land.

Data availability

The data will be made available on request.

Author contribution

450 This paper represents a result of collegial teamwork. FW, PF and MS designed the sampling scheme. FW and SK carried out the field campaign. SK and FW prepared the soil samples for $^{239+240}\text{Pu}$ activity analysis that were carried out by MK. Data processing and analysis was done by FW. FW and PF prepared the manuscript. All authors read and approved the final manuscript.

Competing interest

The authors declare that they have no conflict of interest.

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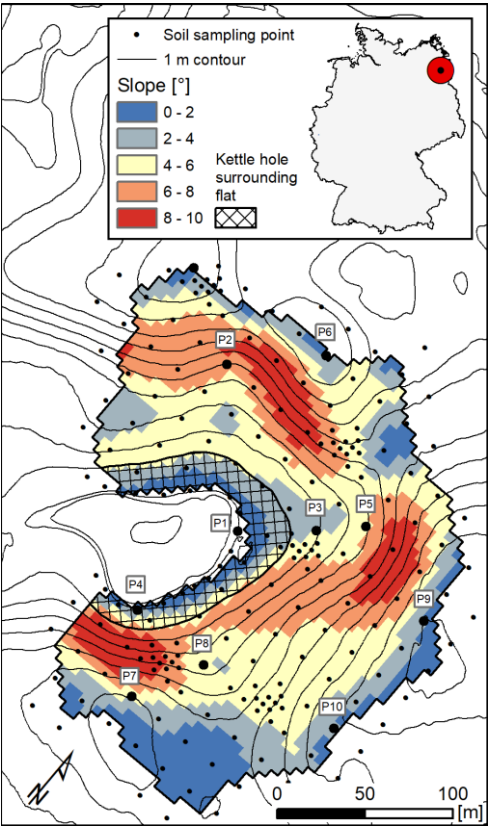
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Tables

Table 1: Parametrisation of the inverse modelling approach.

Parameter	Standard value or range in inverse modelling	Iteration step	Unit	Source
USLE factors				
P	1.0		-	Standard value for soil management without specific soil conservation, e.g. contour ploughing
C	0.081		-	Calculated from crop rotation following the procedure of Schwertmann et al. (1990)
R	45		$\text{kJ m}^{-2} \text{mm h}^{-1}$	From erosivity map of BGR (2014a)
K	25		$\text{kg m}^2 \text{h m}^{-2} \text{MJ}^{-1} \text{mm}^{-1}$	From soil map of BGR (2014b)
LS	variable		-	Calculated using 5 m DEM provided by the state of Brandenburg, Germany
Parameters varied during inverse modelling				
Factor changing the product of all USLE factors (water erosion strength)	0.1...2	0.1	-	
k_{til}	25...1000	25	kg m^{-1}	
k_{tc}	25...500	25	m	

Figures



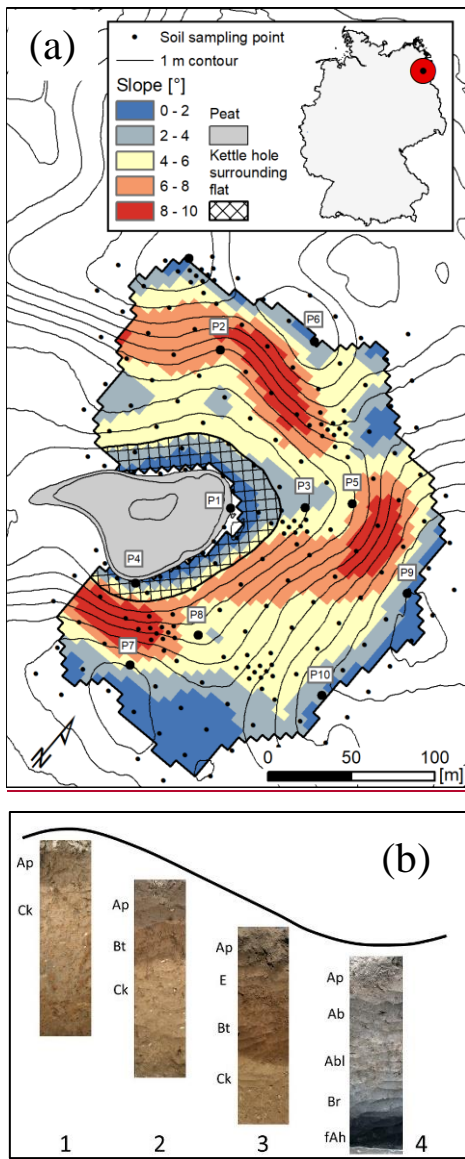


Figure 1: (a) Topography and nested soil sampling scheme in the young morainic study area in northeast Germany.

P1 to P10 indicate the locations for high-resolution depth profile sampling (see Fig. 3). (b) Idealized catena for soil landscapes in hummocky ground moraines; 1: Calcaric Regosol (extremely eroded), 2: Nudiargic Luvisol (strongly eroded), 3: Calcic Luvisol (non-eroded), 4: Gleyic-Colluvic Regosol (colluvial).

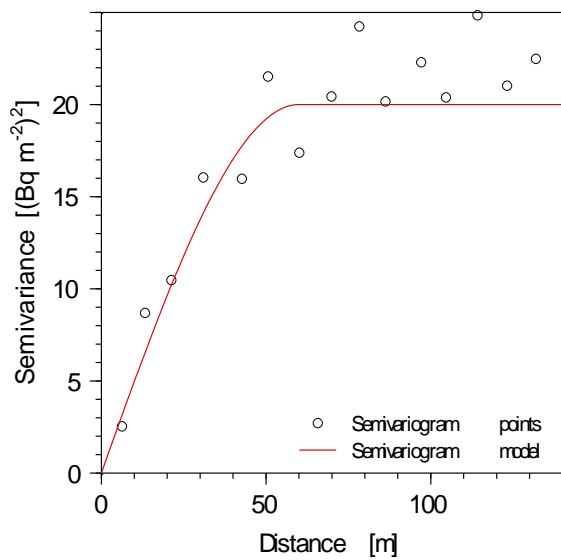


Figure 2: Semivariogram and semivariogram model fit of the $^{239+240}\text{Pu}$ block kriging interpolation.

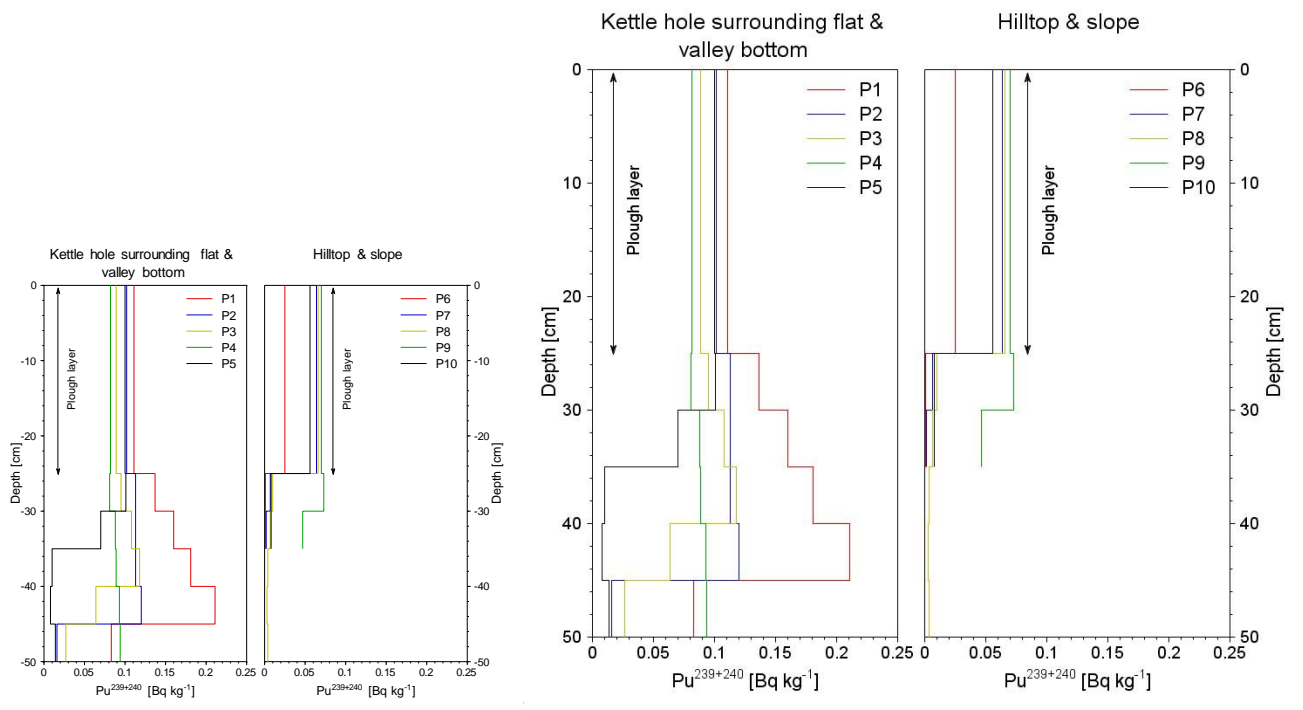


Figure 3: Depth distribution of $^{239+240}\text{Pu}$ at different geomorphological positions. Locations P1 to P10 are given in

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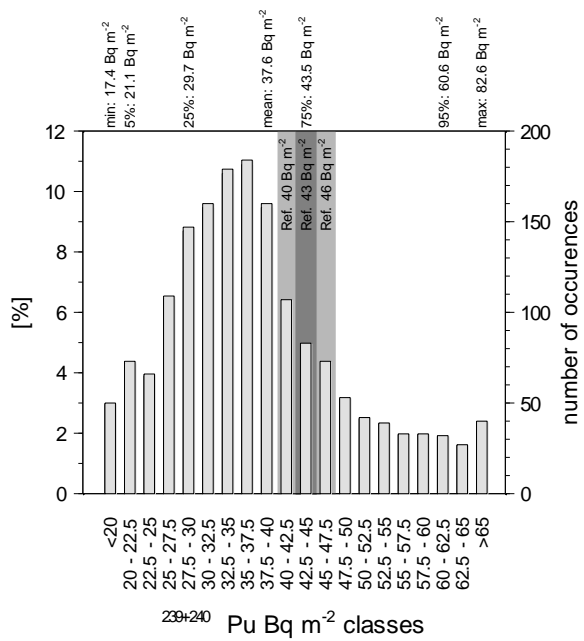
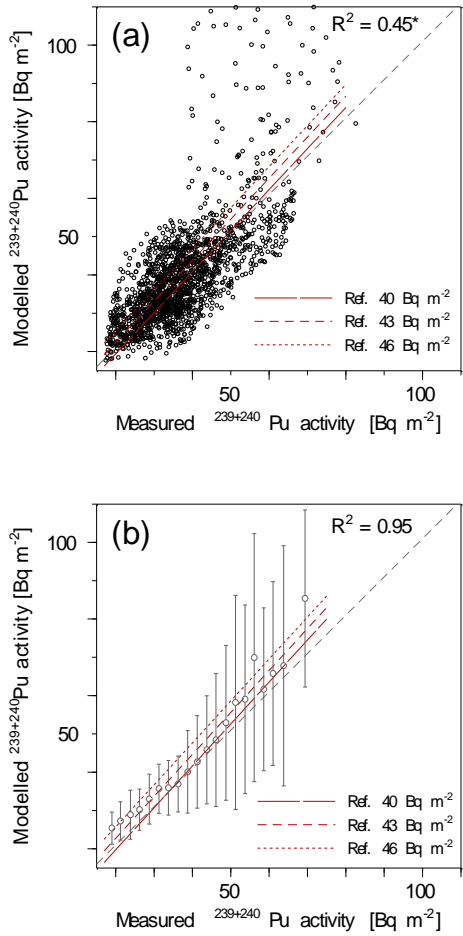


Figure 4: Distribution histogram of 5 m x 5 m interpolated ²³⁹⁺²⁴⁰Pu measurements in 20 classes with descriptive statistics.



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Figure 5: Linear correlation between measured and modelled $^{239+240}\text{Pu}$ inventories redistributed by water (k_{tc} : 150, P-factor: 1) and tillage erosion (k_{til} : 350 kg m^{-1} ; * = p-value < 0.001). (a) Point by point correlation on 5 m x 5 m resolution (n: 1699); (b) class aggregation according to $^{239+240}\text{Pu}$ derived soil redistribution. Minimum and maximum class n is 27 and 184, respectively. While the points and classes are calculated for a reference of 43 Bq m^{-2} , the trend lines display the offset sensitivity of different reference $^{239+240}\text{Pu}$ activities.

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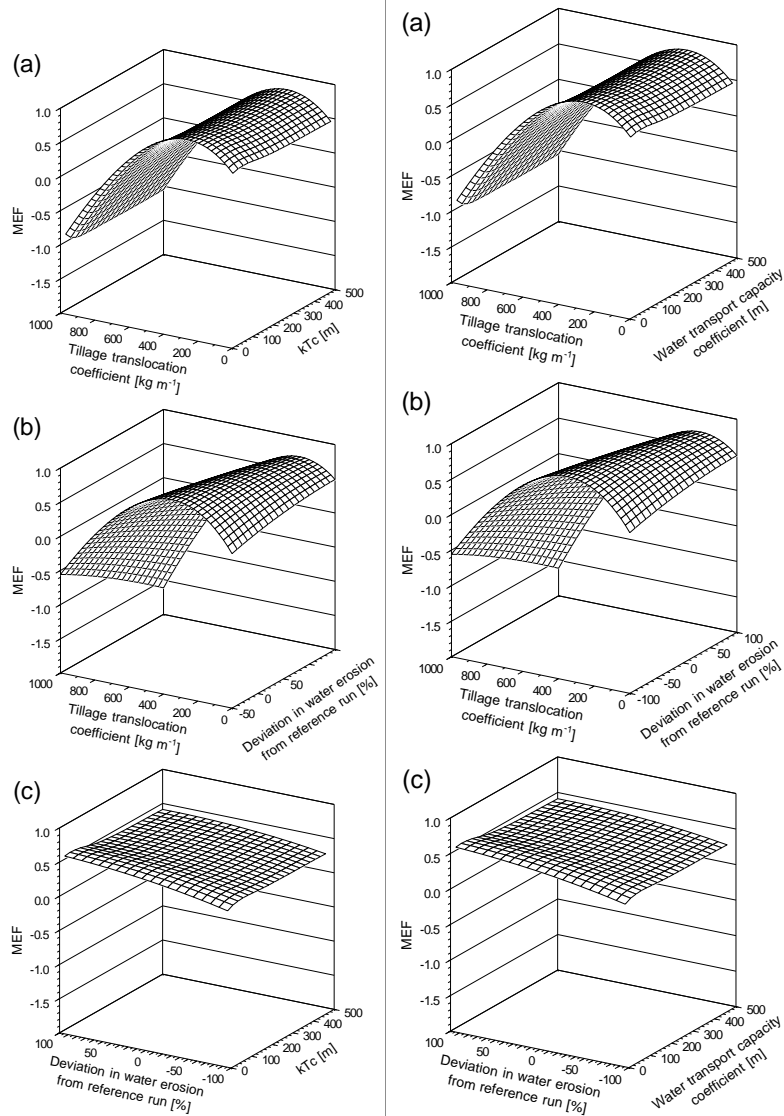


Figure 6: Inverse modelling of tillage and water erosion compared to $^{239+240}\text{Pu}$ derived soil redistribution. Three parameter combinations (tillage transport coefficient, k_{til} ; water transport capacity coefficient, k_{tc} ; deviation in water erosion strength compared to reference parameterisation) are tested on their effect on the goodness-of-fit, represented by the MEF (model efficiency coefficient: perfect model fit = 1; model prediction as good as the mean = 0; model prediction worse than mean = <0).

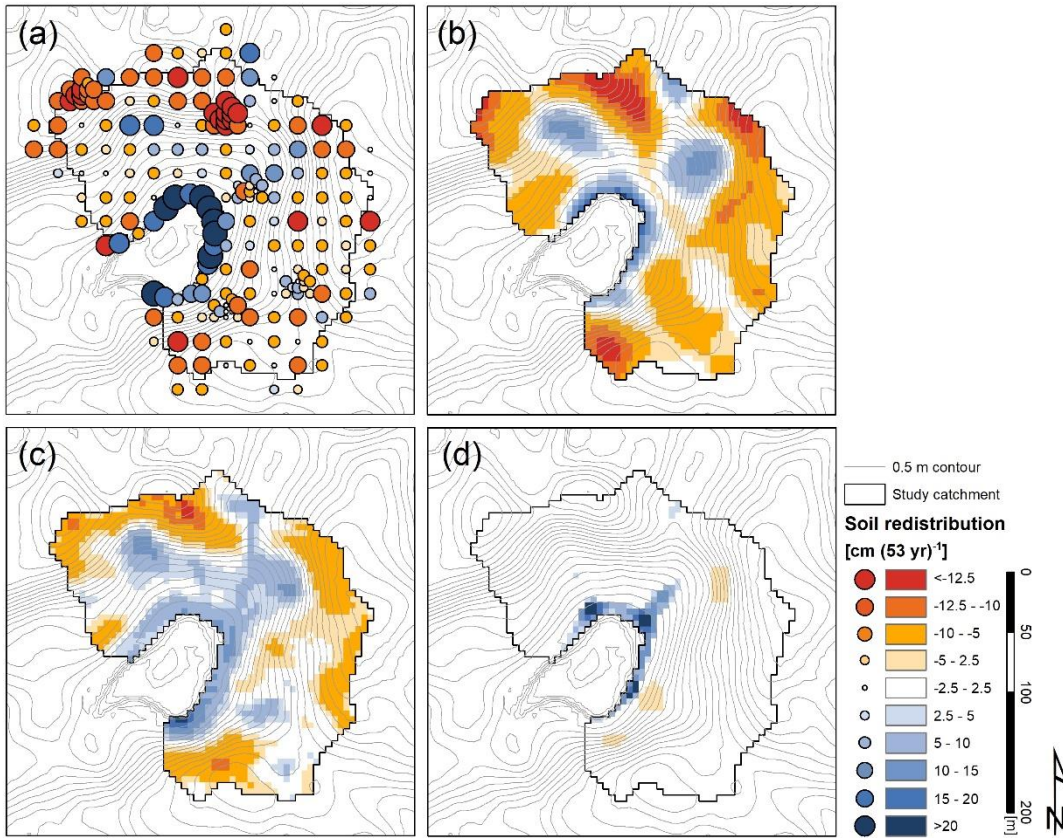


Figure 7: The Figure consists of four parts: (a) Soil redistribution derived from $^{239+240}\text{Pu}$ top and subsoil measurements using 43 Bq m^{-2} as the reference inventory; (b) geostatistically interpolated soil redistribution based on $^{239+240}\text{Pu}$ point measurements; (c) modelled tillage erosion with a tillage transport coefficient (k_{til}) of 350 kg m^{-1} ; (d) modelled water erosion according to reference parameterization (k_{tc} : 150; also see Table 1).