Pu manuscript – Reply to Anonymous Referee #2

Content

1. List of changes made to the manuscript due to a change in the soil fraction considered

While discussing a response to a comment of Referee #1 (AR1 #5), our team discovered a significant reporting error in the physical processing of the samples. At some point in the past (the samples were already processed in 2012; see l. 134), a typo shifted the measured soil fraction from <2 mm to <20 µm. This mistake was then carried further, since it appeared a logical step to focus on this fraction (higher concentration of FRNs in that fraction). However, it turned out that there was not enough original sample material left to separate the required amount of the <20 µm fraction (sandy soils). As a result, all measurements (^{137}Cs, ^{210}Pbex, $^{239+240}$Pu) were actually conducted on the <2 mm fraction, i.e. the measurements include the bulk soil.

This discovery highlights the importance proper sample processing documentation and brings a few changes to the data interpretation. It is worth noting that the numbers as presented in the manuscript do either not change at all (measurements) or do not change significantly (correlations). An advantage is that we can now report bulk soil nuclide inventories instead of concentrations. In the following section, we track the adjustments made to the manuscript as a result of interpreting nuclide activities in the bulk soil rather than in the <20 µm fraction:

ll. 29-30: Updated for inventories: *Specifically, the original inventories of both ^{137}Cs and $^{239+240}$Pu are approximately halved after ~20-40 years of cropping.*

l. 102: Updated to: *… possibly as a consequence of selective removal in this fraction and a relatively higher input of organic matter from crops, …*

l. 171: Added "chemical": *"The chemical sample preparation for plutonium …"*

l. 173: Rephrased to: *The physical preparation of the samples was conducted at the Institute of Crop Science and Resource Conservation, Bonn (sieving), and at the Commonwealth Scientific and Industrial Research Organisation, Land and Water Laboratories (CSIRO), Canberra (homogenisation).*

l. 174: Rephrased to: *In short, samples were sieved to obtain the <2 mm fraction and afterwards homogenised using a planetary mill.*

ll. 174-175: Sentence deleted, since we did not focus on the fraction <20 µm.

ll. 175-176: Rephrased to: *For AMS, about 20 g per sample were dried at 105°C to constant weight.*

ll. 176-177: Sentence deleted, since we did not focus on the fraction <20 µm.

ll. 195-197: Rephrased to: *To measure ^{137}Cs, 50-70 g of the same homogenised material used for AMS were pressed into cylindrical counting discs to ensure a well-defined geometry.*

ll. 197-198: Rephrased to: *These sample measurements were conducted at CSIRO.*

ll. 225-227: Rephrased to: *From these $^{239+240}$Pu activities per mass (here also termed "specific activities") we derived inventories, i.e. activities per area, by including sampling depth and bulk density data (Table S1).*

ll. 240-241: Updated for inventories.

l. 258: Updated for bulk soil.

ll. 259-260: Updated for inventories: *The measured inventories in the top 20 cm of soil span a wide range between 0.43 ± 0.01 mBq cm^2 (KR98/0-20) and 1.95 ± 0.06 mBq cm^2 (TW0/0-20).*

ll. 261-263: Updated for inventories: *Similarly, the other samples from the uncultivated plots in the other two agroecosystems also have the largest inventories in their respective agroecosystems (HS0/0-20 1.44 ± 0.08 mBq cm^2; KR0/0-20 0.98 ± 0.03 mBq cm^2).*

ll. 264-270: Updated for inventories *(R^2 = 0.76).*

l. 270: Sentence added: *Sample KR2.5/0-20 shows an elevated relative inventory of 103.84 ± 4.22% (relative concentration 99.93 ± 3.00%) but does overlap within uncertainties with the defined initial activity. Hence, the sample was excluded from the fit.*

ll. 270-271: Updated for inventories: *From the fit, I_{eq} equals 56.03 ± 6.01% ($1\sigma_x$), and τ equals 6.86 ± 3.03 years.*

l. 276: Deleted: *... in the <20 µm fraction ...*

l. 284: Updated for inventories.

ll. 287-291: Updated for inventories: *The results indicate that inventories are generally much lower than in the top 20 cm, ranging from ~5 to 36% of what is measured in the corresponding topsoil sample (Table 2; Fig. 5). Sample HS45/20-40 is, however, a conspicuous exception with a surprisingly high inventory of 1.01 ± 0.03 mBq cm^2 in the 20-40 cm interval, which is even higher than the 0.70 ± 0.03 mBq cm^2 measured in the uppermost 20 cm of the soil (HS45/0-20).*

l. 294: Updated for bulk soil.

l. 301: Sentence added: *In line with this argumentation, the $^{239+240}$Pu inventories obtained from the native grassland composite samples are in the range expected for surface samples located within 20-30°S, which has been constrained to be 1.44 ± 0.59 mBq cm^{-2} (Hardy et al. 1973).*

Ll. 305-307: Updated for bulk soil: *Equation (2) predicts a minor excess of $^{239+240}$Pu activities (5.4 ± 1.9 mBq kg^{-1}) as compared to ^{137}Cs activity in the soils. Exceeding $^{239+240}$Pu has been proposed to reflect grain-size dependent preferential adsorption patterns (e.g. Everett et al., 2008, Xu et al., 2017), and such a pattern could become important in case of selective erosion.*

l. 316: Updated for bulk soil.

ll. 338-341: Updated for inventories: *Equation (1) predicts a decline in the $^{239+240}$Pu inventory of ~6% to ~2% per year during the first 10 years of cropping. After ~20-40 years, the measured inventories approach the equilibrium level at ~56% of the initial reference values, here constrained by a drop in the decline rate below 0.1% per year. Hence, Eq. (1) predicts that about half of the initial aerial activities in the soil is retained over the long term.*

l. 347: Updated for inventories.

l. 350: Updated for inventories and bulk soil.

l. 354: Updated for inventories.

l. 355: Updated for bulk soils.

ll. 361-363: Updated for inventories: *The relative $^{239+240}$Pu inventories obtained from arable land with a cultivation history exceeding 35 years mostly plot below the equilibrium asymptote defined by Eq. (1) in Fig. 2A, i.e. the weighted mean of these data points is 39.2 ± 3.7% (arithmetic mean 46.2 ± 11.0%).*

ll. 381-391: Updated and rephrased to: *Bulk SOC has been shown to approach an equilibrium concentration of 39.4 ± 2.0% of the initial values after 33.6 ± 8.0 years of cropping, with τ = 6.7 ± 1.7 years (Fig. 2B; Lobe et al., 2001; their exponential model). The SOC equilibrium concentration differs from the Pu inventory equilibrium (and, likewise, the Pu concentration equilibrium, Fig. S5). This deviation may to a certain extent be explained by the cut-off of Pu data included in the fit after 35 years of cropping (Sect. 4.3). On the other hand, the similarity in time constants might indicate that the plutonium was similarly distributed as SOC in the upper 20 cm of the soil soon after initial ploughing, as homogenisation appears to be generally achieved rapidly (Sect. 2.1). Consequently, SOM contents and Pu activities both reflect largest rates of decline during the first years after native grasslands were converted to arable land. This similar behaviour of $^{239+240}$Pu activities and SOM content over time indicates a strong linkage between both variables. The relationship is underscored by high correlation coefficients (R^2 = 0.56 to 0.99; Fig. 2C) between the plutonium concentrations (1963-1998) and bulk soil SOC contents as measured by Lobe et al. (2001) (Table S5). Similar R^2 values (0.63 – 0.99) are obtained when $^{239+240}$Pu is correlated to total N contents (Fig. S1).*

ll. 399-401: Updated and rephrased to: *Here, we indeed observe a strong correlation between plutonium activities and SOM contents, and the majority of SOM (80-90%; Lobe et al. 2001) is bound in the <20 µm fraction of the soils. Likewise, Plutonium isotopes are generally considered to be predominantly bound in that soil fraction (Xu et al. 2017).*

ll. 404-405: Sentence deleted, redundant.

ll. 406-410: Updated and rephrased to: *It should be noted that the close correlation between bulk SOM loss and Pu activity decrease includes SOM losses due to particulate organic matter (POM). Due to its lower density, a certain degree of POM loss by wind erosion is more likely than for similar-sized clastics. Besides, the fate of mineral associated organic matter and POM can be closely coupled (Lobe et al., 2001; Lavallee et al., 2020; Sokol et al., 2019), and the formation of new SOM from the crops was found to be limited at the sites we investigate (Lobe et al., 2005).*

l. 417: Updated for bulk soil.

l. 422: Clause deleted since we did not focus on the >20 µm fraction.

l. 429: Only the silt fraction considered now.

Captions of Figs. 2 and 5: Updated accordingly.

Caption of Tab. 2: Updated accordingly.

2. Updated Figures and Tables

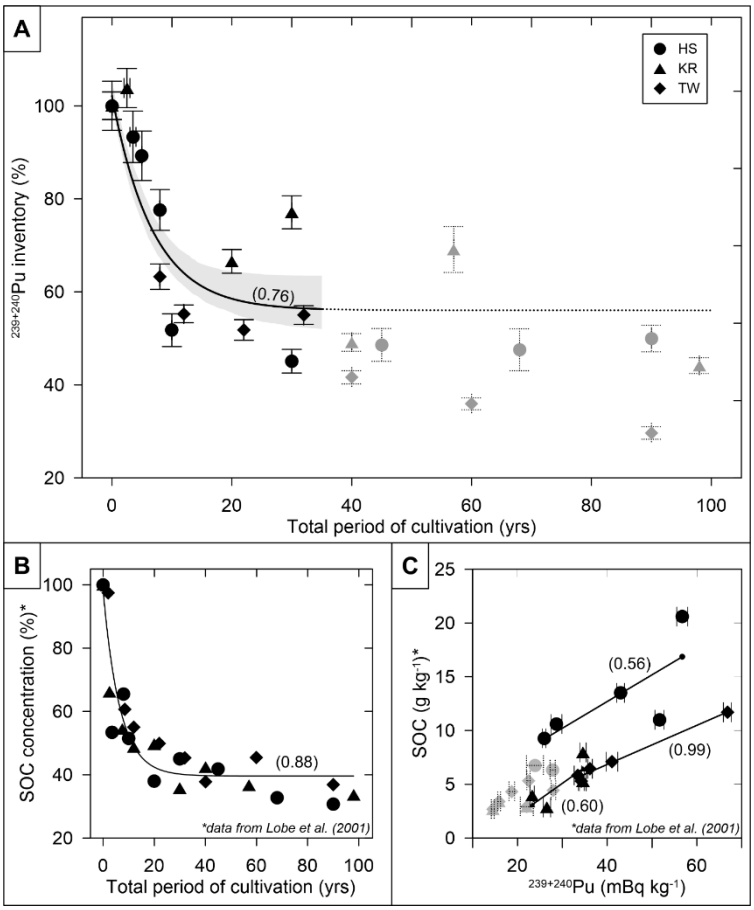
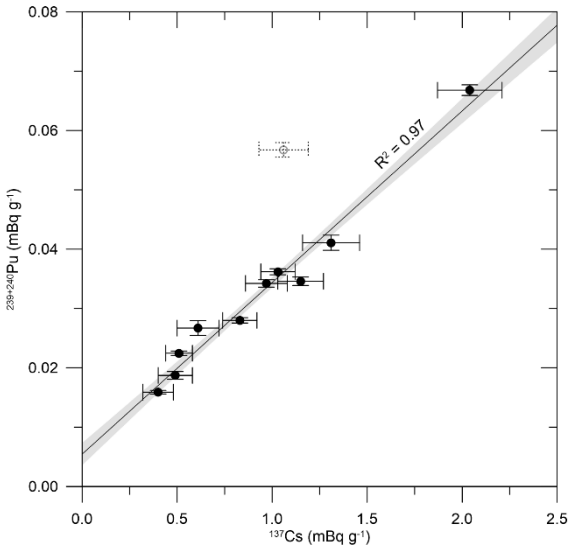
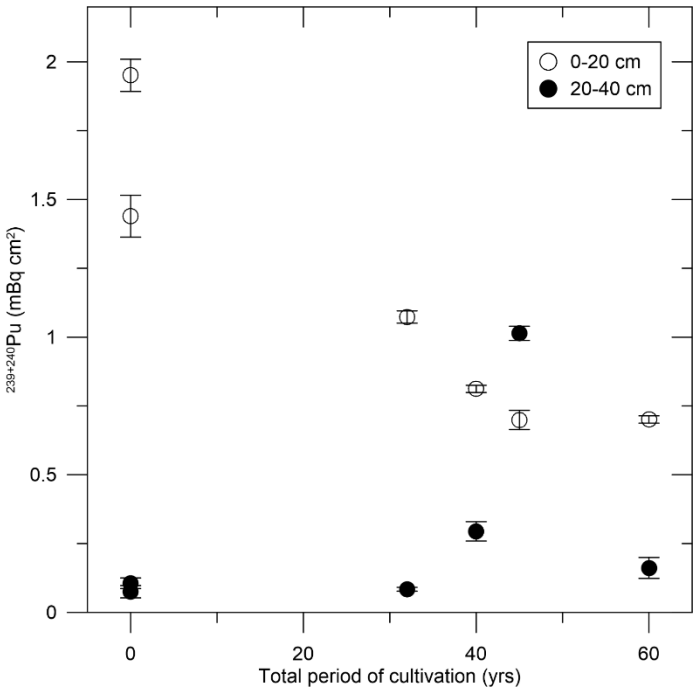


Figure 2: Changes in topsoil fallout inventories (A) and bulk soil organic carbon (SOC) content (B; Lobe et al., 2001) over time, and correlation of the SOC and $^{239+240}$Pu concentrations (C). The inventories in cultivated soils are shown relative to those found in adjacent native grassland soils (i.e., 100% at t = 0). The mono-exponential regression (thin black line, enveloped by grey 68% confidence interval) in panel (A) indicates the approach towards a concentration equilibrium level after about 20-40 years of cropping. By that time, about 50-60% of the initial $^{239+240}$Pu inventory has been lost. The extrapolated post-35 years cropping equilibrium level is indicated by the dashed line. The relationship between $^{239+240}$Pu and SOC indicates that the decrease of SOC can be traced by measuring $^{239+240}$Pu in bulk soil (C). Most plutonium samples depict replicate measurements; the corresponding activities are weighted means and the uncertainties either dominated by AMS counting statistics (weighted mean error) or external sources of uncertainty (standard error). For single measurements, the 1σ measurement uncertainty provided by the AMS facilities dominates the final uncertainty. All greyed out data points with dashed error bars denote those samples that were taken from plots with more than 32 years of cultivation history (for discussion see text). Filled circles denote samples from the Harrismith (HS) agroecosystem; filled triangle those from the Kroonstad (KR) agroecosystem and filled diamonds those from the Tweespruit (TW) agroecosystem. Numbers in brackets denote correlation coefficients (R^2).



144

145 **Figure 4: Correlation of ^{137}Cs and $^{239+240}$Pu topsoil activities. ^{137}Cs data are shown with 1σ uncertainties (which equal**
146 **the estimated measurement errors). $^{239+240}$Pu activities were mostly measured in replicate, and the corresponding**
147 **concentrations are weighted means and the uncertainties either dominated by AMS counting statistics (weighted mean**
148 **error) or external sources of uncertainty (standard error). For single measurements, the 1σ measurement uncertainty**
149 **provided by the AMS facilities dominates the final uncertainty. The majority of samples overlap with the linear**
150 **regression (black line) and its 68% confidence interval (in grey). Sample HS0/0-20 has been excluded from the**
151 **regression (greyed out; for discussion see text). The extrapolated regression intersects the ordinate at about 0.0050 mBq**
152 **g^{-1} (0.50 mBq kg^{-1}; unit conversion to mBq g^{-1} due to lower level precision achieved by γ spectrometry). ^{137}Cs data have**
153 **been decay-corrected to February 2012 (the time of measurement).**



154

155 **Figure 5: $^{239+240}$Pu inventories at depth (20-40 cm) as compared to corresponding topsoil activities (0-20 cm). The**
156 **analysis of plutonium activities at depth has been conducted for *n* = 6 samples, belonging to the agroecosystems**
157 **Tweespruit (0, 32, 40, and 60 years of cultivation) and Harrismith (0 and 45 years of cultivation). At all sites but HS45,**
158 **the nuclide concentration is significantly lower at depth than close to the surface. Error bars are 1σ uncertainties (see**
159 **Fig. 2 for details).**

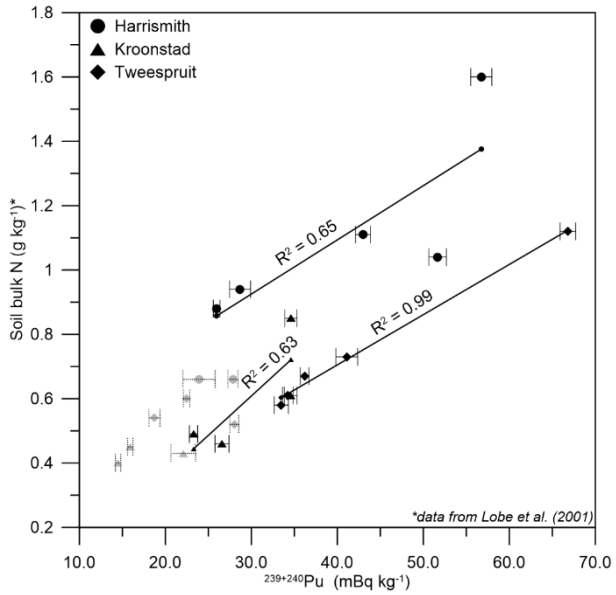


Figure S1: Linear correlations between N contents (Lobe et al., 2001) and $^{239+240}$Pu concentrations in the bulk soil. Most plutonium samples depict replicate measurements; the corresponding concentrations are weighted means and the uncertainties either dominated by AMS counting statistics (weighted mean error) or external sources of uncertainty (standard error). For single measurements, the 1σ measurement uncertainty provided by the AMS facilities dominates the final uncertainty. (vertical error bars either represent 1σ from the mean of several replicates or a 1σ error-propagated uncertainty dominated by the AMS measurement uncertainty). Samples from sites that have been cropped before 1963 were excluded from the regression (greyed out data points).

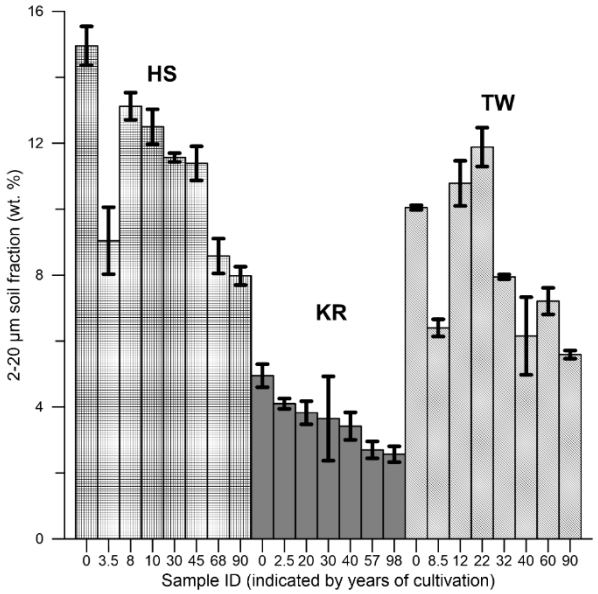


Figure S2: Silt fraction as measured by Lobe et al. (2001) and Amelung et al. (2002). Uncertainties are 1σ standard deviations of the arithmetic means ($n \geq 2$ replicates per sample).

176 **Table. 2: FRN inventories.**

Ecotope	YOC (yr)	Depth (cm)	n	$^{239+240}$Pu (mBq cm^2)	$^{239+240}$Pu (%)	^{137}Cs (mBq cm^2)
HS	0	0-20	1	1.44 ± 0.08	100.0 ± 5.3	27.14 ± 4.24
HS	3.5	0-20	1	1.34 ± 0.04	93.3 ± 5.5	-
HS	5	0-20	2	1.28 ± 0.04	89.3 ± 5.3	-
HS	8	0-20	2	1.12 ± 0.02	77.6 ± 4.4	-
HS	10	0-20	2	0.75 ± 0.03	51.8 ± 3.5	-
HS	30	0-20	2	0.65 ± 0.01	45.1 ± 2.5	-
HS	45	0-20	1	0.70 - 0.03	48.6 ± 3.5	15.98 ± 2.91
HS	68	0-20	2	0.68 ± 0.05	47.5 ± 4.5	-
HS	90	0-20	2	0.72 ± 0.01	49.9 ± 2.8	-
KR	0	0-20	1	0.98 ± 0.03	100.0 ± 2.9	32.66 ± 3.62
KR	2.5	0-20	1	1.02 ± 0.03	103.8 ± 4.2	-
KR	20	0-20	2	0.65 ± 0.02	66.6 ± 2.5	-
KR	30	0-20	1	0.75 ± 0.03	77.1 ± 3.6	-
KR	40	0-20	1	0.48 ± 0.01	49.1 ± 1.9	12.08 ± 2.43
KR	57	0-20	2	0.68 ± 0.04	69.1 ± 4.9	-
KR	98	0-20	2	0.43 ± 0.01	44.1 ± 1.7	-
TW	0	0-20	3	1.95 ± 0.06	100.0 ± 3.0	59.57 ± 6.12
TW	8.0	0-20	3	1.23 ± 0.04	63.3 ± 2.7	39.30 ± 4.61
TW	12	0-20	3	1.08 ± 0.02	55.3 ± 1.9	30.69 ± 2.79
TW	22	0-20	1	1.01 ± 0.03	51.8 ± 2.2	-
TW	32	0-20	2	1.07 ± 0.02	55.0 ± 2.0	30.46 ± 3.54
TW	40	0-20	3	0.81 ± 0.01	41.6 ± 1.4	24.07 ± 2.68
TW	60	0-20	3	0.70 ± 0.01	35.9 ± 1.3	15.91 ± 2.22
TW	90	0-20	2	0.58 ± 0.02	29.7 ± 1.4	15.09 ± 2.80
HS	0	20-40	1	0.08 ± 0.02	5.2 ± 1.6	-
HS	45	20-40	-	1.01 ± 0.03	145.0 ± 8.1	-
TW	0	20-40	2	0.11 ± 0.02	5.4 ± 1.0	-
TW	32	20-40	2	0.08 ± 0.01	7.8 ± 0.7	-
TW	40	20-40	-	0.29 ± 0.04	36.3 ± 4.4	-
TW	60	20-40	-	0.16 ± 0.04	23.0 ± 5.5	*below detection limit*

177
178 **Sample labelling as presented in the main text includes the abbreviation of the sample agroecosystem, years of**
179 **cultivation (YOC) and sampling depth interval: HS - Harrismith, KR - Kroonstad, TW - Tweespruit. The number *n* of**

^{239,240}Pu replicate measurements includes both CologneAMS and ANU AMS measurement replicates. The quoted plutonium activities from replicate measurements are weighted means and the uncertainties either dominated by AMS counting statistics (weighted mean error) or external sources of uncertainty (standard error). For single measurements, the 1σ measurement uncertainty provided by the AMS facilities dominates the final uncertainty. Percentual activities of the topsoil samples are relative to the undisturbed reference sample for each agroecosystem. For the depth samples, the percentage values denote the difference against the corresponding topsoil samples. ^{137}Cs data uncertainties equal 1σ measurement errors arising from μ spectrometry conducted at CSIRO. All ^{137}Cs has been corrected for decay to February 2012.

3. Reply to Anonymous Referee #2

Dear Anonymous Referee #2, thank you for your detailed review. In the following, we address the points you raised.

AR2 #1: One minor point is that the introduction is to much on climate change, CO2 release etc. which is not the topic of this paper.

Reply to AR2 #1: We have deleted ll. 50-52 and ll. 55-56.

AR2 #2: On the other hand, nearly nothing is said on FRN use to assess wind erosion, methods to accomplish this or how to quantify erosion. The loss in SOC and FRN is attributed to wind erosion, just from visual assessment of the site. Could you please give exact slopes in the table for all sites? I find it hard to believe that at these altitudes you have so many sites completely flat. If this is true, this needs at least be identified. If it is not true, please consider that even small slopes will induce significant water erosion even on grasslands whenever you have rain events.

Reply to AR2 #2: We refer to our replies to AR2 #10 and AR2 #11.

AR2 #3: In some parts it is not clear to me, what the authors did with the data. They talk of „normalisation" but report data in percentages. Or was there really any normalisation done? Why not presenting the SOC concentrations and the FRN inventories over time? Much more interesting to soil scientists.

Reply to AR2 #3: See reply to AR2 #21; we did not perform any normalisation but present just relative values, i.e. relative against the pooled grassland sample. Since the (maximum) absolute concentrations and inventories do vary in between the different agroecosystems, we present the data in this way. We also do so to facilitate the reader the comparison of our data with the data from the numerous previous studies published on our samples, which were presented in a similar fashion.

ll. 264: Rephrased for clarification: *We calculate the measured $^{239+240}$Pu inventories for all three agroecosystems to reflect relative inventory concentrations against the relevant pooled samples from the uncultivated plots. As a function of the duration of cultivation, a trend of initially decreasing activity with increasing cropping time is evident, although the rate of decline slows as time goes on (Fig. 2).*

AR2 #4: To apply the FRN approach to assess soil erosion you need at least 3 if not more reference sites. You cannot assume, just because SOC and FRN is highest in your "natural" site, this would be a site without soil erosion. What you could do, however, is compare your arable sites to the one natural grassland and discuss if you have higher or lower erosion. Can you really guarantee that the natural sites were never ploughed since 1950s?

Reply to AR2 #4: Our (sub-)samples taken from the reference sites are obtained from uncultivated grassland sites adjacent to the cultivated sites. For each agroecosystem, the reference sites were pooled to obtain a single composite sample. Hence, our three reference samples provide an average of all reference sites within an agroecosystem. As long as you accept interviews with the farmers and visual assessment of the samples taken with a steel cylinder as proof, we can guarantee that the natural sites were never ploughed.

AR2 #5: 26 the sampled plots did not show signs of fluvial erosion? How do you assess this? And even if they do not show this today, how do you know for 100 years back?

Reply to AR2 #5: We focused on the flat upland sites; the assessment was performed by visual interpretation and interviews with the farmers. From these interviews we also gathered that the older plots would even be less prone to erosion, because they usually are located closer to where the farmers settled, i.e. build their farms. Since their assessment of the landscape was crucial for their survival, we consider it as of even greater significance than our own assessment when we sample the sites.

AR2 #6: 31 how do you know that 6% of the FRN inventory is lost in the first year?

Reply to AR2 #6: From the exponential fit (eqn. 1).

AR2 #7: 37 subtitles is misleading.... this is not about the release of CO2 (which is not the focus of your manuscript) but about the role of SOM in soils and how it is connected to erosion processes

Reply to AR2 #7: We agree and have changed the title to:

l. 37: Soil organic matter and its degradation

AR2 #8: 79-83 the discussion on the potential CO2 of African soils is not very convincing. You already have strong arguments why SOC loss is important: because of general soil degradation. I would suggest to leave that out.

Reply to AR2 #8: We have deleted the text (ll. 79-83).

AR2 #9: 93-96 I cannot follow your rational, why the molecular compound analysis will indicate SOM loss with increasing periods of cultivation

Reply to AR2 #9: We agree and rephrase to:

l. 95: *A key finding of the study published by Lobe et al. (2001) was that SOM contents decreased exponentially with increasing periods of cultivation.*

AR2 #10: Section 1.3. literature of how caesium or plutonium is used to estimate wind erosion is lacking. Web of science lists over 80 studies for caesium and 14 studies for plutonium. Also, no literature is discussed, how inventories are converted to soil erosion rates. As you obviously had a transition from natural grasslands (e.g., distinct depth profiles with FRN declining with depth) to ploughed arable soils (mixed plough layer) this is not a trivial task.

Reply to AR2 #10: We have re-written introduction section 1.3. See also our reply to AR2 #13.

l. 106: Renamed to: Using fallout radionuclides to investigate the contribution of wind erosion to SOM decline in the South African Highveld grasslands ecoregion

ll. 143-146: Shifted to l. 161.

ll. 107-143: Rewritten to: *As noted above, long-term quantitative information on the impact of wind erosion on the loss of SOM in southern Africa due to cropping is lacking to date. The means to overcome this problem is provided by fallout radionuclides (FRNs) from the atmospheric nuclear weapons testing*

in the 1950s and early 1960s. In particular, plutonium isotopes (^{239}Pu and ^{240}Pu) and caesium-137 (^{137}Cs) were distributed world-wide. Concentrations of plutonium isotopes in soils can be measured with very high sensitivity using accelerator mass spectrometry (AMS; e.g. Fifield, 2008), and concentrations of ^{137}Cs are determined by low-background γ-ray spectrometry (e.g. Wallbrink et al., 2003). In order to assess soil redistribution, FRN concentrations in undisturbed reference sites are compared with those in adjacent eroding sites (e.g. Zapata, 2002; Schimmack et al., 2002; Van Pelt, 2013). In the northern hemisphere, the use of ^{137}Cs in this context has been seriously compromised by additional input from the Chernobyl accident in 1986 [see Meusburger et al. (2020) for a detailed study of $^{239+240}$Pu vs ^{137}Cs inventories in Europe] and the Fukushima accident in 2011. The southern hemisphere was not affected, so ^{137}Cs could still be used to complement the measurements of ^{239,240}Pu in the present work. Compared to the plutonium isotopes, ^{137}Cs has a rather short half-life of 30.08 yr [all decay values obtained from the U.S. National Nuclear Data Center (NNDC)]. At the time of measuring (2012), about two thirds of the ^{137}Cs deposited during the atmospheric nuclear weapon tests conducted until the early 1960s had already decayed. Furthermore, the deposition in the southern hemisphere was less than one third of that in the northern hemisphere (UNSCEAR 2000). The concentrations of ^{137}Cs were therefore approaching the detection limit of the γ-counting method, especially in heavily eroded soils and samples from depth. The plutonium isotopes, on the other hand, have much longer half-lives (^{239}Pu: 24,110 yr; ^{240}Pu: 6561 yr), so losses due to decay are minimal. Consequently, plutonium is increasingly supplanting ^{137}Cs as a tracer of soil redistribution (e.g. Alewell et al., 2017; Van Pelt and Ketterer, 2013).

The approach to assess soil redistribution by using FRN concentrations relies on several assumptions which should be met (for an overview, see e.g. Van Pelt, 2013; Zapata, 2002). One precondition is that of a homogeneous distribution of the target FRN over the limited area covering the undisturbed reference site and the nearby eroding sites. However, variability in wet and dry fallout deposition as well as microtopography even on the local scale needs to considered. Thus, a rather short distance between the reference site and the eroding site and a large number of subsamples to characterise the reference site FRN inventory (i.e. n >10; Sutherland, 1996) are considered as crucial (Sutherland, 1996; He and Walling, 1996; Van Pelt, 2013). Reference sites should be shielded from sediment deposition, such as likely achieved on flat upland sites (Funk et al., 2011). Once deposited on the soil surface, the migration behaviour of the fallout isotopes becomes important. In general, ^{137}Cs and plutonium behave similarly in soils, as both are strongly adsorbed on soil fines, including SOM (e.g. Schimmack et al., 2001; Xu et al., 2013). However, evidence is growing that plutonium could have a greater sorption capacity to SOM than ^{137}Cs (e.g. Schimmack et al., 2001; Alewell et al., 2017; Xu et al., 2017). ^{137}Cs has been found to bind more selectively to the clay fraction of soils than does plutonium, implying that ^{137}Cs could be more sensitive to preferential transport (Xu et al., 2017). The potential migration pathway of plutonium as a solute is dependent on its oxidation state, with Pu(III) and Pu(IV) being considered the least mobile (Alewell et al., 2017; Meusburger et al., 2020). Apart from the soil type, rainfall regime appear to affect the advection of plutonium isotopes, with sandy soils in arid environments showing potentially increased mobilisation as compared to clayey soils in the tropics (Cook et al., 2022). Vertical migration in the soil column can also be achieved due to physical processes, such as bioturbation or tillage. The latter, which is of particular importance to this study, has been shown to homogenise FRN concentrations throughout the A_p horizon rapidly, e.g. after ~1-4 times of soil inversion (Schimmack et al., 1994; Hoshino et al., 2015). Similar as for ^{137}Cs (Van Pelt, 2013), plant uptake has been found to be insignificant for plutonium in natural settings (Harper and Tinnacher, 2008; Coughtrey et al., 1984), including grasslands (Little, 1980).

Given that the abovementioned conditions are met, soil redistribution rates may be derived from comparing the FRN inventories in eroding versus those found in reference sites. Most commonly, soil redistribution models, such as the linear proportional model or mass balance models, are applied (for a comprehensive overview, see Van Pelt, 2013). Usually, such models rely on high-resolution depth profiles from undisturbed soils (e.g. Meusburger et al., 2018; Lal et al., 2013; Alewell et al., 2014). The majority of such studies has focused on fluvial erosion, with less applications of ^{137}Cs to quantify wind

erosion and a substantial lack of wind erosion studies applying $^{239+240}$Pu (Alewell et al., 2017; Van Pelt, 2013; Van Pelt and Ketterer, 2013). Most of the studies falling in the latter category identify wind as an erosional mode alongside fluvial erosion (e.g. Zhao et al., 2020; Liu and Hou, 2022), while very few address wind erosion as the main factor of soil redistribution (Little, 1980; Van Pelt and Ketterer, 2013; Michelotti et al., 2013).

In this study, we use both ^{137}Cs and $^{239+240}$Pu to reveal the proposed contribution of wind erosion to SOM loss in the three agroecosystems in the South African Highveld initially studied by Lobe et al. (2001) based on splits from original sample material taken in 1998. The samples encompass a wide range of cultivation histories, ranging from zero (i.e., native grassland) to 98 years. Our approach allows us to investigate the time evolution of SOM loss after native grassland is converted to cropland. Our study represents one of the first attempts to link plutonium activities to SOM loss by wind in arable lands. Furthermore, we are able to introduce a certain temporal resolution of process rates by analysing arable land with different cultivation histories.

AR2 #11: Table 1: could you please give exact slopes in the table for all sites? I find it hard to believe that at these altitudes you have so many sites completely flat. If this is true, this needs at least be identified.

Reply to AR2 #11: It is difficult to estimate the slope of the terrain, because the GPS location of the sampling sites is not as accurate as it could be achieved today. We kindly point out that the samples were taken in the late 1990s, and GPS selective availability was turned off in 2000; i.e. we face accuracy uncertainties on the order of 100 m at least. It is true that not the whole landscape is entirely flat; for example, the Highveld plateau is dissected by canyons. However, samples were taken from upland sites only (information added to l. 152 and l. 377). As an exercise, we calculated the surface slope for a 100 m buffer (100 m radius) around the GPS locations (i.e., ~30000 m²), as presented below. The data is based on a 30 m DEM, the highest resolution freely available for our sites. We note that the minimum slopes would relate to the upland sites:

ID	Mean (°)	Min (°)	Max (°)
HS3.5	2.0	1.0	3.8
HS8	1.7	1.4	1.9
HS10	2.4	1.9	2.9
HS30	2.1	1.8	2.2
HS45	3.9	3.6	4.6
HS68	3.0	1.1	3.9
HS90	3.4	2.2	3.9
KR2.5	0.5	0.0	1.1
KR20	0.9	0.4	1.1
KR30	0.7	0.0	1.1
KR40	0.8	0.2	1.1
KR57	0.7	0.0	1.3
KR98	0.6	0.0	1.1
TW8.5	1.5	1.0	1.9
TW12	1.1	0.7	1.5
TW22	1.1	0.3	1.7
TW32	1.7	0.8	2.1

TW40	1.8	1.1	1.9
TW60	1.6	1.0	2.1
TW90	1.6	1.4	2.1

AR2 #12: 127-129 this is a crude oversimplification. As FRN is deposited with wet and dry deposition, you have substantial heterogeneity. This needs to be considered in taking a adequate number of reference cores. These reference cores should have a CV < 30%. See Sutherland et al.....

Reply to AR2 #12: We agree. Hence, we have focused on individual agroecosystems, and the reference values are obtained from pooled samples providing an average characterisation of each agroecosystem (n = 5 subsamples per individual site within each agroecosystem). The heterogeneity is a further argument why we show inventories relative to the pooled reference sites in Figure 2. We deleted the sentence in ll. 128-129 and added instead (please also see reply to AR2 #10):

l. 128: *However, variability in wet and dry fallout deposition as well as microtopography even on the local scale needs to considered. Thus, a rather short distance between the reference site and the eroding site and a large number of subsamples to characterise the reference site FRN inventory (i.e. n >10; Sutherland, 1996) are considered as crucial (Sutherland, 1996; He and Walling, 1996; Van Pelt, 2013).*

l. 155: Rephrased and added for clarification: *Each of the three datasets includes one composite sample (HS0, KR0, TW0) taken from native grassland sites located directly adjacent to the respective cultivated sites. These reference samples represent the amalgamated sample material from all grassland sites within a common agroecosystem.*

ll. 428-429: Changed to: *For FRNs, such differences are likely to arise from the spatially variable deposition patterns, given the distances between the different agroecosystems of about 100-300 km. However, grain size data also indicate an increase in the silt fraction towards the south-east (Fig. S1).*

AR2 #13: Section 2.1 belongs to introduction. Together with a discussion on the use of FRN to estimate wind erosion.

Reply to AR2 #13: We have shifted the section to the introduction (section 1.3); the combined piece of text is presented in our reply to AR2 #10.

AR2 #14: 153 how flat is flat? Already very slight slopes will induce water erosion in African soils. If you have heavy rain events after dry periods, slopes of <2° might already induce water erosion.

Reply to AR2 #14: See our reply to AR1 #11.

AR2 #15: 174 why <20 µm? In Africa, you can expect to have winds which blow out larger grains.....?

Reply to AR2 #15: In fact, we did measure the bulk soil (see above). We apologise for the confusion caused.

AR2 #16: Did you not do any decay correction for the caesium to a reference year? Why not? How do you then relate to the year of deposition?

Reply to AR2 #16: All caesium data is decay-corrected to February 2012 (see Fig. 4 caption). To clarify, we add:

l. 198: All ^{137}Cs data presented in this publication have been decay-corrected to February 2012 (the time of measurement).

AR2 #17: Table 2: this table looks like a working table from the lab. Could you please make it reader friendly with a column for site name, sampling depth etc???

Reply to AR2 #17: We have updated the table accordingly (see above).

AR2 #18: 240 – 255 most of this is redundant if you format Table 2 properly, make suitable headings and explain some of this in the methods. This has nothing to do with results.

Reply to AR2 #18: We agree. We deleted ll. 240-245, ll. 247-249, and incorporated ll. 245-246 in l. 234. Furthermore, we shifted ll. 249-255 to l.259.

l. 234: Content from ll. 245-246 added: *The latter are reflected by the standard error, i.e. the standard deviation of the set of measurements divided by the square root of the number of measurements $\sigma/\sqrt{n}$ (the larger uncertainty value was chosen for each sample).*

AR2 #19: 260 -263 sentences incomprehensible

Reply to AR2 #19: We assume your comment arises from our insufficient description of the reference site samples. See our reply to AR2 #12.

AR2 #20: 264 sentences like "Figure 2A shows…" are unnecessary… please give adequate figure headings and delete these kind of sentences

Reply to AR2 #20: We have rephrased the sentence, see our reply to AR2 #3.

AR2 #21: Figure 2: what do you mean by "normalised"? What you did is, setting the site you defined as natural to 100% and calculated percentages from that. But this is not normalised? Why do you give percentages and not the original concentrations? Or did you any other normalisation with the data?

Reply to AR2 #21: Absolutely correct. We did not do any other normalisation with the data. Hence, we changed the title of the ordinate. See updated Fig. 2 above and our reply to AR2 #3.

AR2 #22: 275 what do you mean by "internally consistent"?

Reply to AR2 #22: The data of both ANU and CologneAMS is consistent. We deleted "internally" (l. 275).

AR2 #23: 290 even higher

Reply to AR2 #23: Corrected; thank you.

AR2 #24: Section 3.3 It is not unusual that Pu migrates down to 20-30 cm or even 35. But this might also indicate deposition of soil material. As such, you need reference soil profiles to compare you original FRN deposition to erosional or depositional sites.

Reply to AR2 #24: We agree (AR1 raised similar concerns). Here is how we adjusted our text:

l. 170: *A clear disadvantage of the applied sampling scheme is the lack of high-resolution depth profile samples, which was not required for the originally intended sample analyses. As a consequence, we are unable to present FRN mass depth profile data, and thus cannot reasonably infer mass redistribution rates as typically presented in FRN studies (e.g. Alewell et al. 2014, Lal et al. 2013, Meusburger et al. 2018).*

l. 236: peak plutonium deposition added and rephrased: *Since our sampling strategy included a spatial averaging of sampling material from each plot investigated (Sect. 2.2), the best explanation for the elevated plutonium activity in HS45/20-40 may be related to a former ploughing to 40 cm near in time to peak plutonium deposition that was not recorded during farmers' interviews or to sample contamination. Elevated inventories measured in two further depth samples might point to a certain degree of leakage of plutonium towards greater depths, but not necessarily in the pre-fallout soils (Sect. 4.3). We also note that in a reasonably comparable setting (Bsh climate; Big Spring, USA),* ^{137}*Cs and* $^{239+240}$*Pu concentrations dropped sharply below the* A_p *horizon in an Aridic Paleustalf that had been cultivated since 1915 (Van Pelt et al., 2007; Van Pelt and Ketterer, 2013).*

l. 314: *However, any leakage could equally affect the isotope concentrations and may thus not be reflected by the ratio.*

l. 371: leakage added: *Likewise, a possible incorporation of Pu-marked plant material into the soil column after harvesting might have contributed to elevated inventories found in the three depth samples with cropping histories exceeding 35 years (HS45/20-40, TW40/20-40, and TW60/20-40). In case of wet fallout deposition, such an enhanced downward migration could also have been promoted by the physical disturbance of the ploughed soil (cf. Das Gupta et al., 2006). These factors appear not to have been significant for the plot that was converted after the peak episode of global fallout (TW32/20-40). However, the topsoil samples TW40/0-20 and TW60/0-20 had the strongest inventory losses of all samples (Fig. 2a; Table 2), and show the highest negative residues against the exponential model. Thus, and given the general low scatter of the post-35 YOC data points, we may argue that if significant migration of Pu-marked soil particles below 0-20 cm has occurred, the two samples in question could represent the cases of maximum leakage in our dataset.*

l. 418: leakage added: *Furthermore, a certain degree of leakage of plutonium-marked particles to greater depths could contribute to a lowering of* $^{239+240}$*Pu activities in the topsoil over time.*

l. 423: rephrased to: *To resolve this issue, longer timescales need to be monitored and high-resolution depth profiles sampling implemented in future studies.*

AR2 #25: 323 to apply the FRN approach to assess soil erosion you need at least 3 if not more reference sites. You can not assume, just because SOC and FRN is highest, this would be a site without soil erosion. What you could do, however, is compare your arable sites to the one natural grassland and discuss if you have higher or lower erosion.

Reply to AR2 #25: We kindly refer to our replies AR2 #4 and AR2 #12.

AR2 #26: 336 I can not follow this assumption nor the conclusion

Reply to AR2 #26: We agree, there is no reasonable relationship between the two clauses. We have deleted the first part of the sentence (ll. 336-337).

AR2 #27: 341 do not understand why exponential decline indicates higher adsorption and the conclusion to aggregation seems far fetched (or explain and constrain it better)

Reply to AR2 #27: The exponential model predicts the retention of Pu over the long term. However, we state that it may not be the most accurate model to chose (ll. 420-423). Note that the original sentence was modified according to the reviewer's comments (see below). As for the aggregation, we find it reasonable to present our ideas.

ll. 420-423: *Instead, a bi-exponential model as suggested to predict SOM decrease over time (e.g. Amelung et al., 2002; Lobe et al., 2001; Lobe et al., 2011) could reflect the long-term fate of $^{239+240}$Pu inventories in the topsoils more accurately. To resolve this issue, longer timescales need to be monitored and high-resolution depth profiles sampling implemented in future studies.*

AR2 #28: Section 4.3 I do not agree with this discussion. Of course, using FRN in Southern Hemisphere means you can only assess the period from 1950ties to now. However, soils which were already ploughed during that time, are the only ones where you could quantify soil erosion, as you do not need any reference site depth profile but only a total inventory of the reference site and can then apply the proportional model. For all other sites, e.g. the sites changing from natural to ploughed in between, you would have to assume an (unknown) depth profile first and then a mixed plough layer after to quantify erosion.

Reply to AR2 #28: We have measured composite depth samples (20-40 cm), that clearly to indicate that there was – if any – very limited migration of Pu below 20 cm in the pooled reference sites (Fig. 5). We take that as a sufficient indication for assuming that the Plutonium was predominantly stored in the 0-20 cm soil column.

AR2 #29: 402 What do you mean "deflation processes rather than turnover rates"?

Reply to AR2 #29: Lobe et al. (2001) identified the turnover (predominantly mineralisation) of SOM in the silt and clay fractions as the main factor controlling the time-dependent decrease of SOM stocks in the investigated arable soils. Here we postulate that deflation processes may play a more important role for the decrease in SOM contents (see Fig. 2C).

AR2 #30: Figure 5: I am generally puzzled by this Figure. If you see this strong decline in Pu concentrations over time and attribute this to erosion process, this means that what you measure today as 0-20 cm depth was 20-40 cm depth 100 years ago, right? But why don't you see any changes in 20-40 cm depth? This should for sure decline to zero 60 years after cultivation. I think there is something else going on and you should for sure calculate your inventories considering your mass depth of soil and may be even assess erosion rates comparing it to you natural site. These simulated erosion rates (which, strictly speaking would not be absolute erosion rates but rates above the natural site values) would then give you some confidence about possible processes going on. However, you clearly need depth profiles of FRN from your natural site and the time of conversion from natural to arable land.

Reply to AR2 #30: As noted above, we have calculated inventories now; however, we cannot supply depth profiles. A certain degree of vertical migration may be affecting the FRN inventories in the 0-20 cm soil column (see our replies to AR2 #24). Concerning the erosion of soil, we kindly point out that a significant fraction of Pu loss may be due to the deflation of SOM, and not the clastics (see Fig. 2C).

AR2 #31: Conclusion – Sorry to say, but from the above, I can not really see that these conclusions are justified by your data.

Reply to AR2 #31: No problem, that is what a good reviewing process is for. We thank you again for your comments, apologise for the wrong grain size stated and interpreted in the initially submitted manuscript, and hope that you are happier with our rewritten conclusion. Furthermore, we invite you to read our reply to AR1, which has also brought significant change to the manuscript. For the new version of our conclusion, we have shifted bits of text from section 4.5 to the conclusion and deleted section 4.5 afterwards. The new version of the conclusion:

l. 452: *We have measured fallout radionuclides (^{137}Cs, $^{239+240}Pu$) to quantitatively investigate the linkages between SOM decline and wind erosion in plain arable land of South Africa's Highveld grassland ecoregion. Wind erosion, a physical process, appears to be a dominant factor removing SOM particles from the plots we investigated. Here, wheat and maize yields have been reported to be more than halved after about 30 years of cropping (Lobe et al., 2005). The severity of wind erosion can be promoted by the cropping practices that are commonly observed in the Free State province (Wiggs and Holmes, 2010; Eckardt et al., 2020)and that were applied at the sites we investigated at least until the time of sampling. These cropping practices include the clearance of arable land from any vegetation for up to 6 months per crop rotation cycle (1-2 years) to minimise soil water loss by plant uptake during the dry season. Consequently, dust emissions peak during the winter months, when arable soils remain largely unprotected in South Africa's rainfed agriculture (Eckardt et al., 2020). Under the impression of anthropogenic climate change, which is in turn predicted to increase both drought and storm event probabilities southern Africa (Arias et al., 2021), the data we present provides further evidence that these cropping practices cannot be termed sustainable. We find similar patterns of relative SOM decline in our investigated sites which are located in different agroecosystems at distances of hundreds of kilometres between each other. Based on the observations of Eckardt et al. (2020) and the grain size data obtained by Lobe et al. (2001) and Amelung et al. (2020), it appears reasonable to assume that SOM particles are conveyed to the Pacific Ocean. It is still a matter of debate whether oceans can be generally considered as sinks for organic carbon (for an overview see Chappell et al., 2019), but it seems likely that SOC particles' exposition to decay is enhanced during transport in the atmosphere (e.g. Lal, 2006), contributing to negatively balance the worldwide carbon budgets. However, the effects may be less severe than they would have been if mineralisation controlled the release of CO_2, given the chance of carbon fixation in the ocean.*

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