Pu manuscript – Reply to Anonymous Referee #1

Content

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

While discussing a reply to a comment of Referee #1 (AR1 #5), our team encountered a significant reporting mistake in how the physical processing of the samples was conducted. 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. The truth is, however, that there was not enough original sample material left to separate the needed amount of <20 µm material (sandy soils). Thus, all measurements (^{137}Cs, ^{210}Pbex, $^{239+240}$Pu) were conducted on the <2 mm fraction, i.e. the measurements include the bulk soil.

While this is a good example for the need of proper sample processing documentation, the issue brings a few changes to the data interpretation. Note, however, 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 conveniently state bulk soil nuclide inventories instead of concentrations. In the following lines, we track the changes made to the manuscript arising from interpreting nuclide activities in the bulk soil, instead of 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

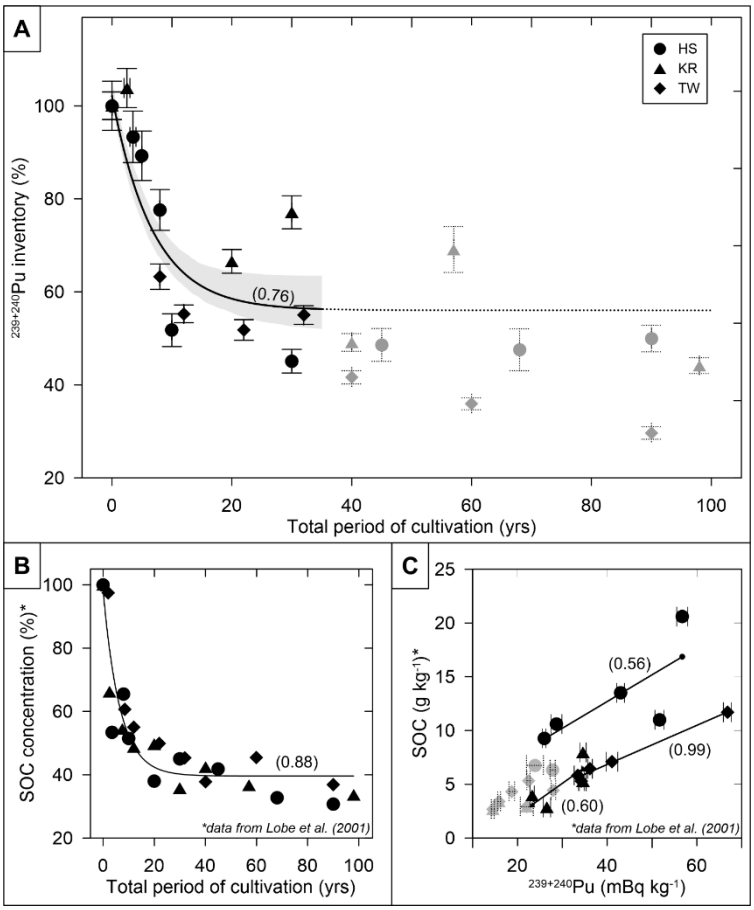


Figure 2: Changes in topsoil fallout inventories (A) and bulk soil organic carbon (SOC) content (B; Lobe et al., 2001) over time, and correlation of the SOC and $^{239+240}$Pu concentrations (C). The inventories in cultivated soils are shown relative to those found in adjacent native grassland soils (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).

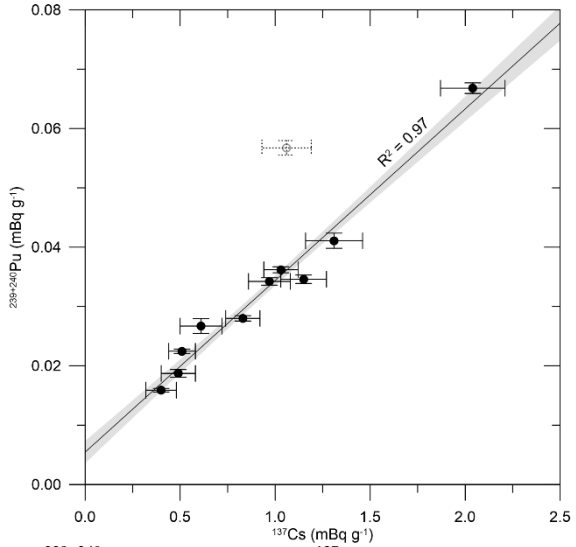


Figure 4: Correlation of ^{137}Cs and $^{239+240}$Pu topsoil activities. ^{137}Cs data are shown with 1σ uncertainties (which equal the estimated measurement errors). $^{239+240}$Pu activities were mostly measured in replicate, and the corresponding concentrations are weighted means and the uncertainties 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. The majority of samples overlap with the linear regression (black line) and its 68% confidence interval (in grey). Sample HS0/0-20 has been excluded from the regression (greyed out; for discussion see text). The extrapolated regression intersects the ordinate at about 0.0050 mBq 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 been decay-corrected to February 2012 (the time of measurement).

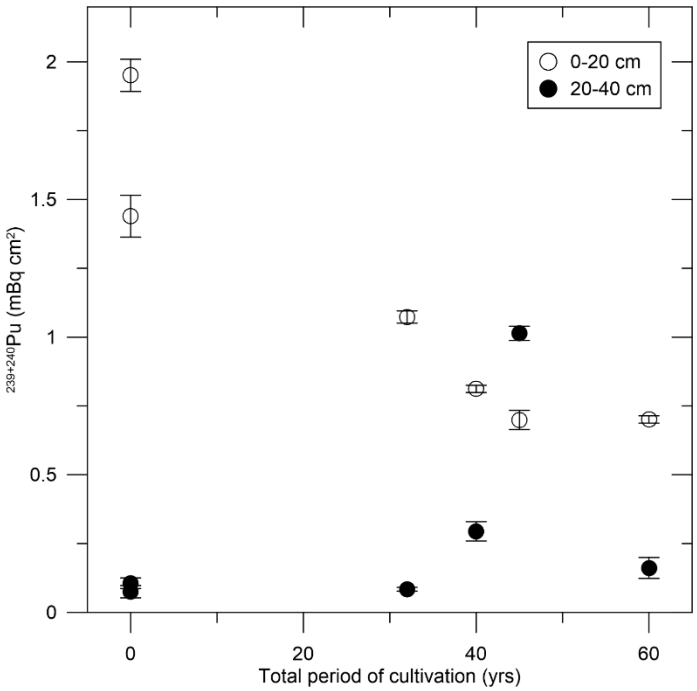


Figure 5: $^{239+240}$Pu inventories at depth (20-40 cm) as compared to corresponding topsoil activities (0-20 cm). The analysis of plutonium activities at depth has been conducted for *n* = 6 samples, belonging to the agroecosystems Tweespruit (0, 32, 40, and 60 years of cultivation) and Harrismith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

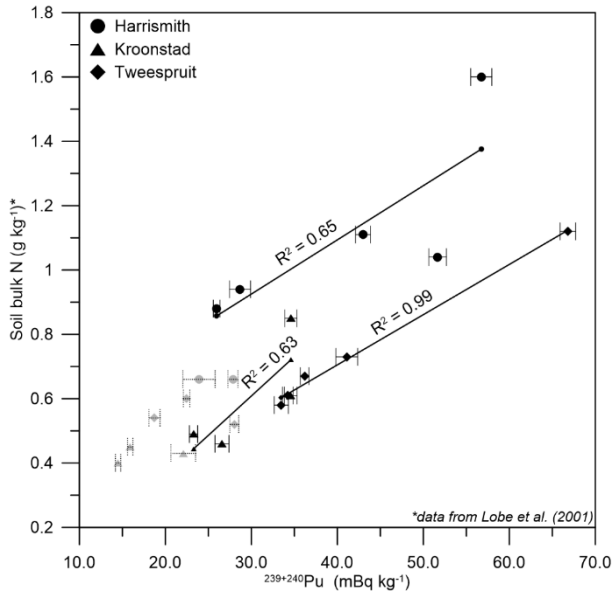


Figure S1: Linear correlations between N contents (Lobe et al., 2001) and $^{239+240}$Pu concentrations in the bulk soil. Most plutonium samples depict replicate measurements; the corresponding concentrations are weighted means and the uncertainties either dominated by AMS counting statistics (weighted 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).

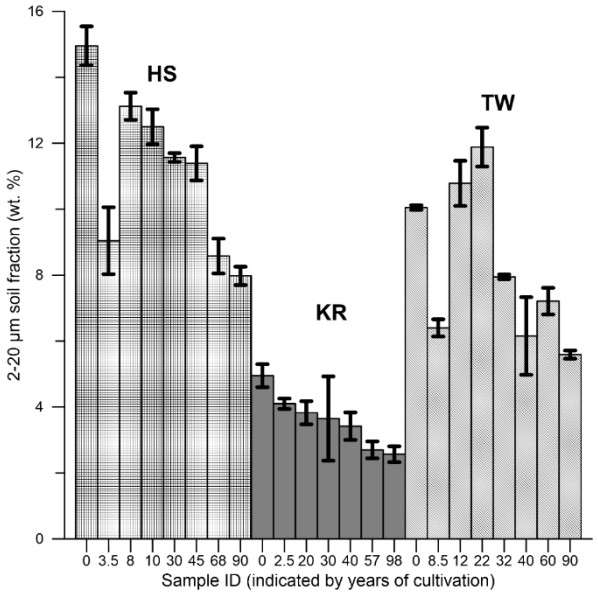


Figure S2: Silt fraction as measured by Lobe et al. (2001) and Amelung et al. (2002). Uncertainties are 1σ standard deviations of the arithmetic means ($n \geq 2$ replicates per sample).

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*

Sample labelling as presented in the main text includes the abbreviation of the sample agroecosystem, years of 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 #1

Dear Anonymous Referee #1, thank you for reviewing our manuscript and for providing your detailed assessment. We address the points you raised below.

AR1 #1: The serious scientific challenge to this study is the use of Pu and SOM concentrations rather than mass balance, since the latter is required to demonstrate mass redistribution as typically laid out by erosion studies based on fallout radionuclides (e.g. authors He, Walling, Wallbrink, Mabit, Alewell, Meusberger etc.). I recognize that the present authors are limited by the work of their predecessors but nonetheless this issue should require more direct and explicit treatment here. Else it sounds as though the attribution of declines in Pu and SOM concentrations with cultivation history to wind erosion is fait accompli, while there is otherwise no direct evidence of the process of wind erosion per se presented here.

Reply AR1 #1: We agree and add the following information and statements:

l. 105: *About 100 km to the northwest of the Tweespruit sites, Wiggs and Holmes (2011) measured dust fluxes on a flat (<2°) ploughed field belonging to the Grasslands farm near Bloemfontain. The authors reported a total dust deposition of 48.19 g cm^{-2} (0.48 g m^{-2} day^{-1}) from the local, wind-eroding sandy soils for a timespan ranging 99 days between August and November 2007. For the sites we investigate in our study, a re-assessment of the silt fraction content [partially unpublished, measured by Lobe et al. (2001) and Amelung et al. (2002); Tab. S1] reveals a linear increase in south-eastern direction (R2 = 0.73; Fig. S1), which follows the general trajectories of dust plumes in South Africa (Eckard et al. 2020).*

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

l. 426: updated to: *In our study, we measure the lowest activities in the upwind sites. The composite grassland sample HS0/0-20 has an activity twice as high as the most north-west located sites (KR0/0-20), generally coinciding with SOM patterns as published by Lobe et al. (2001). This pattern is further reflected by grain size data, indicating an increase in the silt fraction towards the south-east (Fig. S1). Consequently, KR soils had the lowest silt fraction of all soils (Fig. S2) by the time of sampling.*

AR1 #2: There are alternative explanations for a change in SOM and Pu concentrations that may be challenged more directly by the authors. First, the foremost influence on SOM and Pu concentrations in soil upon first tilling will be the tilling itself, since in native soils the highest concentrations of both are at the soil surface. If the tilling process is anything but perfectly homogenizing in the 0-20 cm soil, and there were any bias in sampling depth relative to tilling depth (say, 20 cm and 30 cm respectively), would the appearance of the concentrations over time not be exactly what we see in Figure 2? I wonder what assurance the authors provide that the observed patterns are simply not an artifact of tilling and sampling?

Reply AR1 #2: We interpret your comment as follows (please correct us, if we misunderstood): In native grassland, SOM and Pu will be stored in the uppermost part of the soil column. Then, the soil is ploughed, relocating SOM/Pu and shifting the peak concentrations down the profile, i.e. not completely homogenising it. If we then take our sample, and the sampling depth does not capture the ploughing

horizon entirely, we would miss a certain fraction of Pu/SOM. This fraction is than brought up again after more ploughing and incorporated to the range that we sample. By always missing the lower part of the inhomogeneous ploughing horizon we would face a continuous in- or outflux of Pu/SOM over time. So, basically, there are three issues to be addressed: homogenisation, ploughing depth and sampling depth. While we cannot rule out ploughing depth as a source of uncertainty, which we stated in l. 159, we focus on sampling depth and, more importantly, homogenisation.

Information on homogenisation added to rewritten Section 1.3 (see reply to AR2 #10): *Vertical migration in the soil column can also be achieved due to physical processes, such as bioturbation or tillage. The latter, which is of special 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).*

l. 162: Information about sampling added: *Five subsamples, taken by using a steel cylinder, were amalgamated per plot to obtain the final sample.*

l. 321: Error propagation (which will of course not affect the depth vs. homogenisation issue) and information about (accurate) sampling added: *However, the slight variation in soil densities resulted in larger uncertainties of the pooled reference samples due to error propagation, when compared to the ploughed plots. An accurate sampling depth was achieved by using a 20-cm long steel cylinder for sampling, but we propagate a depth error of 0.5 cm for calculating the inventories.*

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).*

AR1 #3: Next, the authors should acknowledge that the lower boundary of the 0-20 cm soil depth is not closed. They do point this out with respect to one location that actually as higher Pu in 20-40 cm than 0-20 cm, but this is attributed to deeper tilling. While this actually speaks to my point (1) above, I also highlight the potential for leakage over time of both SOM and Pu below the 20 cm boundary. The normal migration of organometallic complex at just 0.2 mm y-1 could export 1% of Pu and SOC to deeper soil, for example. Compounded with time this could easily explain the scale of Pu and SOC loss over decades.

However, it is interesting to observe that the two pulses have different time zeroes. Any SOC pulse from inherited O-horizon is necessarily timed with onset of cultivation, whereas the Pu pulse is independent of that. In fact the Pu pulse is centered at a cultivation time of approximately 40 years ... and there appears no obvious pattern in the data related to this (excepting the noted site with higher Pu >20 cm which I surmised is related to deep tilling near in time to peak Pu deposition). That the SOC and Pu patterns both reflect time since cultivation would seem therefore to be good

corroboration that it is some extant property of the soil that is regulating retention of SOC and Pu that are introduced at the soil surface rather than any artifact of tilling.

Reply to AR1 #3: We tried to address the leakage issue by measuring Plutonium in a selection of depth samples (Fig. 5). However, we did not put much emphasis on the two samples that indicate elevated inventories (> 10% w.r.t. the topsoil sample) so far.

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. 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.*

AR1 #4: It should also be noted that SOC and Pu differ in that there are continuous SOC inputs at the soil surface through ongoing plant growth, whereas there are no ongoing Pu inputs with possible exception of relatively minor remobilization through erosive process. I wonder if this difference explains the different long-term trajectories of SOC and Pu in Figure 2 ... Pu continues to decline due possibly to leakage out the bottom (point above), whereas SOC is at some steady state with respect to ongoing inputs.

Reply to AR1 #4: We agree that there are different inputs, but emphasise that previous studies also used biexponential models to capture a continuous decrease in SOM contents in the soils. Lobe et al. (2005) address the SOC in- and outputs. While there is a replacement in old SOC with new SOC (65% of SOC is crop-derived after 90 years, on average), in- and output rates are very similar with slightly higher rates for the outputs (equilibria after ~20 yrs). We find your thoughts exciting but point to the lack of causality for the topsoil Pu data exceeding 35 years. However, we include the leakage hypothesis as presented for AR1 #3 (l. 418). We also refer to our reply AR1 #7.

AR1 #5: Finally, the authors should make clear in this paper (and not through references), the handling of the samples to produce the <20 um fraction, and especially what % this represents of the whole sample. Otherwise it remains unclear how the <20 um fraction might relate to soil mass balance, and whether a decrease in SOC and Pu concentrations in this fraction is truly attributable

to mass balance, or alternatively to a change in texture or other soil properties that regulate carbon and fallout metal retention. More details on the <20 um fraction are especially important since this fraction may now be interpreted as the 'mineral associated organic carbon' or MOAC fraction, which the author touch on tangentially as possibly related to changes in carbon sequestration over time.

Reply to AR1 #5: As detailed above, this comment revealed our wrong sample processing recordings (or, to be more precise, how these records were interpreted after a long time of hibernation). We thank you very much for pushing us there!

AR1 #6: In arguing for wind erosion to the authors do conclude with some observations on a possible gradient in Pu concentrations along wind fetch. If this were the case, could this also be correlated with changes in soil texture or % fines due to deflation? Such a correlation would make a fine figure and would provide some independent evidence for wind erosion that is otherwise lacking.

Reply to AR1 #6: True! We have calculated the distance of our sampling sites along a NW-SE transect (distance = 1 km at the most northwesternmost sampling site) to obtain a correlation (see also new lines of text as presented in our reply to AR1 #1):

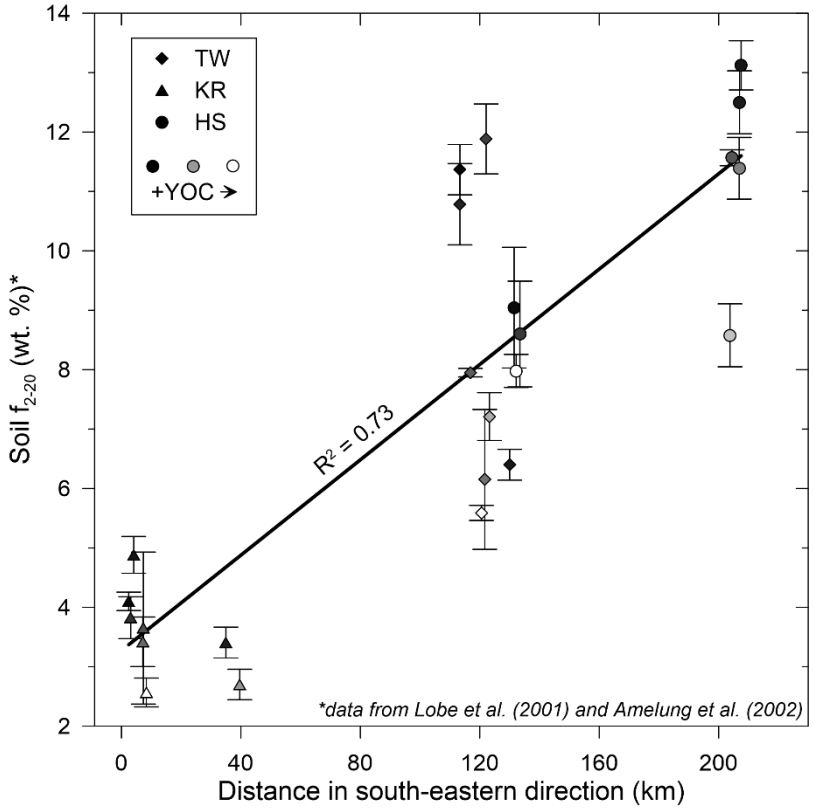


Figure S2: Soil fraction 2-20 µm and distance in south-eastern direction of individual samples (pooled samples excluded). The grain size data has been measured by Lobe et al. (2001) and Amelung et al. (2002) (partially unpublished).

AR1 #7: Finally, the peculiar problem of a Pu point source some 40 years prior to soil sampling might be countered if the authors were able to use Pb-210 which was likely measured concurrently with their Cs-137 measurements. Similar to Cs it may be that the Pb-210 half-life precluded robust measurement from old archive samples, but even in this case it would be worth including a

statement to this effect since otherwise Pb-210 could be quite valuable to the study. It would powerful to show for example that Pb-210 more precisely mirrors SOC due to continuous input to both through the history of the experiment ... or not!

Reply to AR1 #7: Indeed, $^{210}Pb_{ex}$ has been measured. We did not include it before because of large error ranges, but include it now:

l. 477: *An approach to overcome the problem of a point source in time determined by anthropogenic global fallout involves the quantification of excess ^{210}Pb, hereafter referred to as $^{210}Pb_{ex}$. This naturally occurring FRN has been widely used for erosion studies elsewhere (e.g. Matisoff, 2014; Mabit et al., 2008; Hu and Zhang, 2019; Meusburger et al., 2018), and was measured at CSIRO alongside ^{137}Cs (Tab. S6). From this data, general trends of concurrently decreasing $^{210}Pb_{ex}$ activities with decreasing ^{137}Cs activities (Fig. S3) and SOC content (Fig. S4) might be inferred. However, the short half-life (~22.2 yrs) of the nuclide plus >10 years of sample storage and generally low environmental concentrations cause propagated measurement errors of 16-30% of the respective means. Hence, we refrain from providing a detailed interpretation of $^{210}Pb_{ex}$ activities in our samples.*

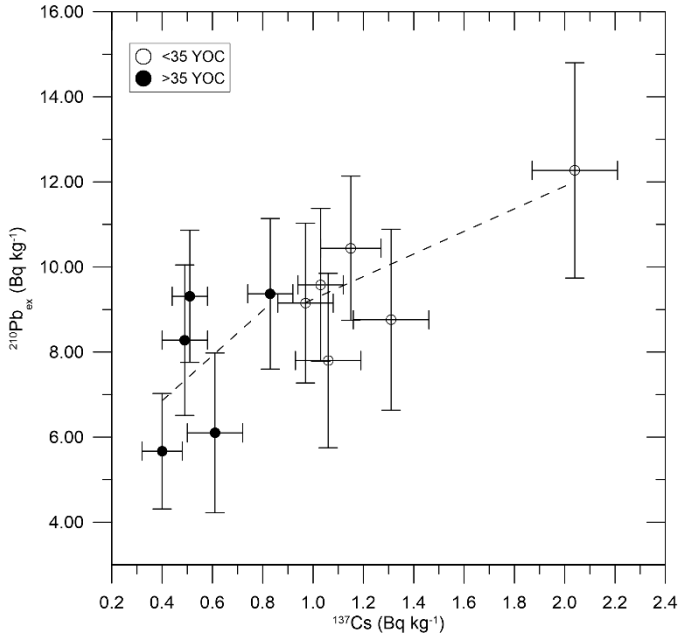
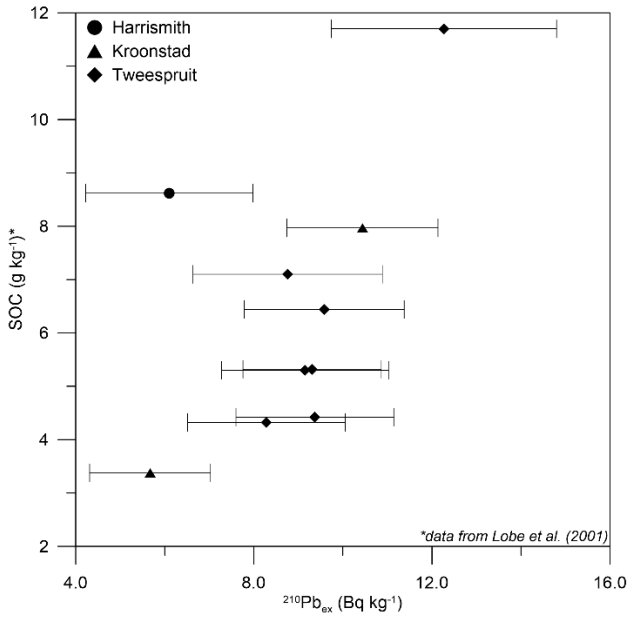


Figure S3: ^{137}Cs and $^{210}Pb_{ex}$ topsoil activities. All data shown with 1σ uncertainties, which equal the estimated measurement errors for ^{137}Cs and include error propagation for the calculation of $^{210}Pb_{ex}$. Linear fits are shown for visual comparison of the <35 YOC (years of cultivation) and >35 YOC samples, respectively (dashed lines).



404

405 **Figure S2: Soil organic carbon (SOC) contents and $^{210}Pb_{ex}$ activities in the bulk topsoil. $^{210}Pb_{ex}$ data includes error-**
406 **propagated 1σ measurement uncertainties.**

407

408 **Table S6: $^{210}Pb_{ex}$ specific activies.**

Ecotope	YOC (yr)	Depth (cm)	$^{210}Pb_{ex}$ (Bq kg^{-1})
HS	0	0-20	7.80 ± 2.05
HS	45	0-20	6.10 ± 1.88
KR	0	0-20	10.44 ± 1.70
KR	40	0-20	5.67 ± 1.36
TW	0	0-20	12.27 ± 2.53
TW	8.0	0-20	8.76 ± 2.13
TW	12	0-20	9.58 ± 1.80
TW	32	0-20	9.15 ± 1.88
TW	40	0-20	9.37 ± 1.77
TW	60	0-20	9.31 ± 1.55
TW	90	0-20	8.28 ± 1.77
TW	60	20-40	*below detection limit*

409

410 **$^{210}Pb_{ex}$ was measured alongside ^{137}Cs. Measurement procedure as e.g. detailed in Swarzenski (2014). Equilibrium ^{222}Rn**
411 **was calculated from the weighted average of ^{214}Pb (295 and 352 keV) and ^{214}Bi (609 keV). To calculate final $^{210}Pb_{ex}$**
412 **activities, ^{222}Rn activities were subtracted from ^{210}Pb (46.5 keV) activities.**

413 **YOC: Years of cultivation.**

414 **AR1 #8: c.r. line 426: for additional evidence for wind erosion, examine residuals from exponential**
415 **fits of SOM and Pu along wind direction ... do concentrations (residuals) for both SOM and Pu**
416 **covary tightly?**

Reply to AR1 #8: Assessing the residuals reveals no strong covariation – SOM has a minor tendency to positive residuals (measured value – modelled value) with increasing distance, while Pu shows a stronger tendency to decreasing values (the latter for the <35 years data only; linear R2 = 0.47, 0 YOC excluded). However, as we point out in ll. 419-422, there is no real certainty about the model type, and we feel that trying more exact fitting would be an over-elaboration given the limited dataset.

AR1 #9: line 346: please clarify, aggregate size increases with time since cultivation?

Reply to AR 1 #9: Indeed, the aggregate mass >250 increases (relative to total soil mass) with time since cultivation.

AR1 #10: line 384: excellent point

Reply to AR 1#10: Very sad to see that the reasoning you liked had to be changed due to the wrong soil fraction used by us. See our new line of argumentation for ll. 381-391 above.

AR1 #11: Figure 2: both Pu (panel a) and SOM (panel b) show exponential declines with cultivation time, but the dropoff appears much more steep for Pu than for SOC. I wonder about the significance of this, or is it simply a result of intersite variability? It may be due to different pulse input histories?

Reply to AR 1#11: Similar to reply AR 1#10 – Time constants have changed now. We apologise for the extra efforts.

AR1 #12: Figure 3: this is a methods figure and I would recommend placing in supplemental materials, or simply omitting it while replacing with relevant summary statistics in the methods section.

Reply to AR1 #12: We have moved the figure to the supplement.

AR1 #13: Figure 4: it would be nice to indicate the Pu:Cs fallout ratio decay-corrected to 2012. The correlation here is impressive, but this does not necessarily indicate that both Pu and Cs are retained to same degree, only that the fraction that IS retained is retained to same degree. This occurs in lake sediments for example where Pu and Cs are similarly strongly correlated with depth, but by mass balance as much as half of Cs depositional flux is missing presumably due to higher solubility in the water column.

Reply to AR1 #13: We have added the Cs/Pu fallout ratio decay-corrected to 2012 (26.69 ± 0.97) to Figure 4/3. We added the following piece of text:

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

AR1 #14: Figure 5: high Pu in 40-50 year cultivated sites... did first plowing quickly follow period of peak Pu deposition, and mix Pu into subsurface 20-40 cm soils thereby minimizing susceptibility to erosion? What would this mean for mass balance and assumption that Pu is lost to wind erosion at the soil surface?

Reply to AR1 #14: We might speculate that the incorporation of Pu-marked plant material into the soil and/or the physical disruption of the soil during fallout could contribute to this effect. However, we also need to acknowledge that the two other sites that show elevated Pu inventories at depth have the highest decrease of all topsoil samples (happy coincidence). See our reply to AR1 #3.

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