Mohren et al. Biogeosciences manuscript – Reply to Anonymous Referee #3

Dear Anonymous Referee #3, thank you for reviewing our manuscript and for providing your detailed assessment as well as the additional references. We address the points you raised below.

AR3 #1: In the manuscript, the authors recognise (Line 465) the occurrence of deposition from wind erosion and dust emission elsewhere. The source of that deposition could be from nearby and therefore include a wide range of particle sizes (coarse material will not be preferentially removed with distance). Alternatively, that deposited material may be distal and therefore associated with fine material very likely enriched in fallout radionuclides. In their approach to establishing a reference site, the authors have neglected to consider that their chosen site may be influenced by deposited aeolian material. A larger reference inventory would change the magnitude of losses and gains identified at sites made relative to that site. The results are therefore uncertain depending on the amount of deposited aeolian material at the native grassland site by contrast to deposition at the other sites.

The corollary is that deposited aeolian material is likely to be occurring across the region as the authors suggest (Line 465). If different sources of that deposited material are proximal and distal, then the deposited particle size distribution will change and consequently the FRN concentration per unit mass will change. These changes in concentrations do not conform to the expected behaviour of the approach and its underlying assumptions (Chappell, 1999). The uncertainty is further dependent on the mixing of deposited material from different sources.

Reply to AR3 #1: We agree and add and/or change text as follows:

l. 168: *Furthermore, sampling focused on upland agricultural areas with level surfaces to minimise the possibility of fluvial erosion and aeolian influx affecting the SOM content (and FRN concentrations).*

l. 193: *Likewise, we cannot assess effects of soil particle fluxes that may alter inventories in the composite reference samples (cf. Chappell, 1999; Sect. 4.4).* (see also AR3 #2)

l. 325: *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 or soil particle influx.*

l. 474: *If such regional patterns of sediment redistribution caused significant influx of soil particles to reference sites from both local and regional sources after global fallout (cf. Wiggs and Holmes, 2010), it is possible that FRN inventories have been subject to alterations. As our methodological approach relies on undisturbed reference sites, significant influx to the reference sites would violate that most important precondition. Chappell (1999) showed that influx of soil particles can significantly alter ^{137}Cs specific activities at a reference plateau site located in semi-arid bushland. Influx of distal dust particles may increase FRN concentrations (Chappell, 1999), more proximal influx of coarser grains could dilute them (Funk et al., 2011). Hence, post-fallout accumulation of soil particles on our reference sites could have different effects on the overall FRN concentrations, depending on the concentration of FRNs in the deposited soil particles (generally linked to soil particle grain size and source). However, visual inspection of the reference sites before sampling suggested that significant coarse-grained influx from local sources can be ruled out. In addition, Funk et al. (2011) demonstrated that ^{137}Cs reference sites rather unaffected by aeolian deposition could be identified in their study region, which resembles our study setting (grassland plateau site in Mongolia with significant wind erosion). We also note that soil bulk densities are generally homogenous across the individual agroecosystems and that the ^{137}Cs and $^{239+240}Pu$ concentrations we obtained are strongly correlated (Fig. 3). Given that ^{137}Cs and $^{239+240}Pu$ are suspected to show somewhat different grain size and SOM-dependent adsorption patterns (Sect. 1.3),* *the finding could imply that influx to the reference sites was limited. Furthermore, given that reference sites were located directly adjacent to the eroding sites, alterations of the relative inventories due to soil particle influx should decrease in significance at decreasing YOCs.*

AR3 #2: The authors have provided a very basic description of the assumptions upon which the fallout radionuclides are used to estimate soil redistribution (around Line 125). I think a much clearer description of the assumptions is required. I think this description needs to be updated with the alternative approach using resampling (Li et al., 2011). Most importantly, I think it is essential that this description in the manuscript is improved by including a more critical evaluation of the approach including the work by Foster and Parsons (2011), the Comment by Mabit et al. (2011) and other arising commentary since then.

<u>Reply to AR3 #2:</u> We add and/or change text as follows:

<u>l. 124:</u> *The method 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; a critical assessment of the technique and a reply to the critical view are provided by Parsons and Foster, 2011, and Mabit et al., 2013, respectively). One precondition of the widely used traditional sampling approach (cf. Li et al., 2011) is that of a homogeneous distribution of the target FRN over the limited area covering the undisturbed reference site and the nearby eroding sites.*

l. 130: *A certain variance attached to reference inventories may be inevitable but can be reduced by applying the repeated-sampling-approach, which relies on on-site point-specific reference inventories (Li et al., 2011; Kachanoski and De Jong, 1984). Such a sampling strategy, however, would require a resampling campaign and hence be difficult to implement in our case given possible changes in land use and cropping practices since 1998 as well as individual permits required.*

A reasonable application of the traditional approach relies on reference sites that ideally are vegetated with perennial grass or low herb cover (Pennock and Appleby, 2002) and shielded from sediment deposition, such as likely achieved on level upland sites (Funk et al., 2011).

<u>l. 135:</u> *While ^{137}Cs sorption has been found to be generally dependent on the availability of cation exchange sites in soils and hence on clay mineralogy (Mabit et al., 2013; Parsons and Foster, 2011), it might bind more selectively to the clay fraction compared to plutonium, implying that ^{137}Cs could be more sensitive to preferential transport (Xu et al., 2017).*

<u>l. 182:</u> *Up to nine different agricultural plots were sampled per agroecosystem, with the requirement that the cultivation history (up to 98 yrs) could be precisely ascertained and a reference site could be sampled adjacent to the eroding site (cf. Lobe, 2003).*

<u>l. 185:</u> *The latter were included in order to test whether the topsoil sampling approach captured most of the plutonium stored within the soil column (cf. Parsons and Foster, 2011).*

<u>L. 190:</u> *The sampling scheme, which originally did not focus on FRN analyses, has some disadvantages potentially biasing FRN data interpretation (cf. Sect. 1.3). Firstly, the lack of high-resolution depth profile samples means that we are unable to present FRN mass depth profile data. Consequently, we 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). Likewise, we cannot assess effects of soil particle fluxes that may alter inventories in the composite reference samples (cf. Chappell, 1999; Sect. 4.4), although visual inspection suggests that significant coarse-grained influx from local sources can be ruled out. Finally, amalgamation of the reference site samples (n = 7-9 samples per agroecosystem; with n = 5 subsamples per site) implies that we cannot provide statistical measures to evaluate the accuracy of fallout inventories in the reference samples.*

l. 303: *In order to investigate whether plutonium could have migrated below this soil layer (e.g. Parsons and Foster, 2011), samples spanning the depth interval 20-40 cm were analysed for selected sites from the Tweespruit (n = 4) and Harrismith (n = 2) agroecosystems.*

AR3 #3: The abstract does not adequately represent the issue that the identification of wind erosion needs to be without the presence of water erosion. I think this topic is reasonably well described in the main text, perhaps with the inclusion of Van Pelt et al. (2017).

Reply to AR3 #3: We rephrase as sentence in the abstract and add the reference (l. 149 & 152).

l. 26-28: *Wind erosion has previously been shown to play a dominant role in soil particle loss from agricultural sites in the Highveld, and the level plots we investigate here did not show any evidence of fluvial erosion. Hence, we interpret the fate of soil fines, including SOM, to be governed by wind erosion.*

AR3 #4: The word 'flat' is used throughout the manuscript to incorrectly describe how level the land surface is. The word flat is a description of the land surface roughness and should be replaced with the word level, as appropriate.

Reply to AR3 #4: We follow your advice and change the terms accordingly (l. 81, 97, 130, 169).

AR3 #5: Line 165 "Since our samples were already taken in 1998 and have been characterised in numerous studies (Amelung et al., 2002; Lobe..." This sentence is ambiguous in the description of the characterisation. Please rephrase.

Reply to AR3 #5: We rephrase:

l. 165: *The samples analysed in this study were taken in 1998 and splits from these samples have been measured in previous studies to investigate a variety of soil components and patterns of soil degradation over time (Sect. 1.2). In the following, we give a brief overview of the sampling strategy that was applied.*

Other changes made to the manuscript

Words or letters added/removed:

l. 18: losses → *loss*

l. 22: + *during the*

l. 87: cropping → *cultivation (YOC)*

l. 137: regime → *regimes*

l. 305: + *indicate*

l. 360, 365, 368, 404, 421, 424: years of cropping → *YOC*

l. 408: + *1.2 and*

l. 469: are likely to arise from the → *could arise from*

l. 517: + *We thank three anonymous referees, whose comments have significantly improved the quality of this paper.*

Section titles changed:

l. 164: 2.1 Sampling strategy and sample processing → *2.1 Sampling strategy*

l. 194: + *2.2 Sample processing*

l. 223: 2.2 FRN measurements → *2.3 FRN measurements*

l. 246: 2.3 Interpretation of $^{239+240}$Pu results → *2.4 Interpretation of $^{239+240}$Pu results*

l. 374: 4.3 Temporal limitation of $^{239+240}$Pu topsoil inventories → *4.3 Factors that may influence the interpretation of $^{239+240}$Pu topsoil inventories*

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