# S1 Supplementary figures



Figure S1. 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 (2002) (partially unpublished).



Figure S2: 240Pu/239Pu of all samples measured (including replicates; error bars denote 1σ AMS measurement uncertainties). The weighed mean *xCE* of all ratios is 0.180 ± 0.002 (thick black line includes the standard error *σx*). The weighed mean thus plots within the range of 239Pu and 240Pu sourced from global fallout for the latitudinal band 0-30°S (0.173 ± 0.014, 1σ) as constrained by Kelley et al. (1999). Other FRN sources are characterised by different 240Pu/239Pu, e.g. when originating from the Chernobyl nuclear power plant (NPP) accident (here shown as the 1σ range of the mean; 0.396 ± 0.014) cited from Boulyga and Becker (2002), Fukushima NPP accident (1σ of the mean, i.e. 0.319 ± 0.014 of three surface soil samples taken 25-32 km away from the power plant) calculated from Zheng et al. (2012), or when originating from weapons-grade plutonium (Nevada National Security Site, GMX area, 1σ of the mean – 0.051 ± 0.000) cited from Krey et al. (1976).



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



Figure S4: Soil organic carbon (SOC) contents and 210Pbex activities in the bulk topsoil. 210Pbex data includes error-propagated 1σ measurement uncertainties.



**Figure S5:** Changes in topsoil fallout concentrations in the bulk topsoil (0-20 cm). The concentrations in cultivated soils are shown relative to those found in adjacent native grassland soils (i.e., 100% at t = 0). The black line (enveloped by grey 68% confidence interval indicated a mono-exponential regression approaching towards a concentration equilibrium level (here: after 25-45 years of cropping). By that time, about 50-60% of the initial 239+240Pu concentration has been lost. The extrapolated post-35 years cropping equilibrium level is indicated by the dashed line. 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. 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.



Figure S6: Linear correlations between N contents (Lobe et al., 2001) and 239+240Pu concentrations in the bulk topsoil. 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 S7: Silt fraction as measured by Lobe et al. (2001) and Amelung et al. (2002). Uncertainties are 1σ standard deviations of the arithmetic means (*n* >2 replicates per sample).

# S2 Supplementary tables

The supplementary tables S1 to S6 are presented in a separate .xlsx file.

# References

Amelung, W., Lobe, I., and Du Preez, C. C.: Fate of microbial residues in sandy soils of the South African Highveld as influenced by prolonged arable cropping, European Journal of Soil Science, 53, 29-35, <https://doi.org/10.1046/j.1365-2389.2002.00428.x>, 2002.

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