



Research Article

On the challenges of dating alluvial sediments with radiocesium: a caveat from the Wurm River, Central Europe

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Abstract

The activity of radiocesium in alluvial sediment is a widely used proxy for sediment age determination for the period after its first occurrence in the environment in 1952. In a continuous and undisturbed sediment archive, the results of this method are reliable as shown by various studies. However, depending on the specific characteristics of an archive provided by alluvial sediment, the results might be unreliable or biased. This article describes the challenges and ambiguous results obtained by the measurement of radiocesium in a soil profile that provides a time marker with a concrete structure of known age. The applicability of radiocesium for the study area was successfully tested on a floodplain profile. The results showed radiocesium peaks up to 22.5 Bq/kg that correspond to the years 1963 and 1986. As a result of different hydromorphological conditions and sedimentation rates at the sampling locations, the peaks occurred in different depths. In the floodplain, the ¹³⁷Cs-peaks are significantly more distinguished than in the profile at the concrete structure. There, the radiocesium content is scattered, which queries the function of the structure as a chronological marker. The scattering might be caused by various syn-sedimentary and post-sedimentary processes and factors, such as scouring and redeposition of contaminated sediment, topsoil recycling by vegetation uptake and dieback, or input of eroded sediments from the hinterland forest soils. Thus, the concrete structure creates and at the same time destroys the possibility of using it as a chronological marker.

Keywords ¹³⁷Cs · Chronological marker · Alluvial sediment · Stratigraphy · Sediment chronology · Sediment age determination

1 Introduction

1.1 Radiocesium activity for sediment age determination

The activity of ¹³⁷Cs in sediment and soil is widely used as a chronological marker for the past decades. It has been applied for sediment chronologies and determination

of sediment accumulation rates in a high variety of sediments (e.g. [1–7 and references therein]). According to [8], radiocesium is a useful marker for assessing rates of sediment accumulation in sediment sinks and serves as a casual tracer for environmental processes [9].

The anthropogenic short-lived radioactive isotope ¹³⁷Cs with a half-life of approximately 30 years was released in the atmosphere during three major phases: (1) the

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beginning of nuclear weapon testing programs by the USA and USSR in 1952 [10], and the first significant atmospheric increase in the northern hemisphere in 1954 [11], (2) the peak of the atmospheric weapon tests in 1963, and (3) the Chernobyl accident in 1986 [3, 12]. Subsequent to the input to the pedosphere via fallout (dry and wet deposition), the radiocesium is adsorbed quickly and irreversibly to clay minerals and organic matter [9, 13, 14]. Age determination with radiocesium can be an alternative to more common techniques such as radiocarbon dating. Dating with ^{137}Cs is often combined with the ^{210}Pb dating technique. In soils and sediments, ^{210}Pb originates from in situ decay of ^{226}Ra as well as from atmospheric deposition of this radionuclide formed in the decay of gaseous ^{222}Rn . Decreasing activity of the latter, excess or unsupported ^{210}Pb with increasing depth is a basic observation allowing calculation of the ages of individual layers as well as of the sedimentation rate [e.g. 15]. However, quantification of sedimentation rates does not only depend on radioactive decay of excess ^{210}Pb and initial fallout radionuclide activity related to atmospheric deposition. In dynamic sedimentary systems, such as floodplain deposits characterized by episodic sedimentation, this also depends on input, removal or mixing (physical or biological) of excess ^{210}Pb adsorbed to detrital particles, possibly even derived from different sources [16, 17]. Consequently, determination of sedimentation rates on the basis of excess ^{210}Pb activity requires modelling to account for these different processes.

In contrast to the ^{210}Pb dating method, application of ^{137}Cs yields independent chronological information.

Provided that element mobility can be neglected, the presence of marked peaks in the depth distribution of this radionuclide can be attributed to the above-mentioned emission events representing time marks independent on modelling and uncertainty inherent in determination of the input parameters—particularly in the case of highly variable flood-related sedimentation [6, 18, 19]. Thus, ^{210}Pb dating was not considered appropriate for this study.

This decision is based on the morphological position of the sampled profiles, which are within the riparian area of the river and are, therefore, subject to frequent floods. However, if radiocesium dating is applied in alluvial sediments, there are crucial factors to be considered when interpreting the results of a gamma spectrometry analysis.

1.2 Background

The so-called Westwall (also known as 'Siegfried Line') is a defence structure from World War II that was built in the period of 1938 to 1940 [20]. Although partly destroyed, large parts of the Westwall still exist. In the area of the city of Aachen, a concrete tank trap was built in the floodplain of the Wurm River (Lower Rhine Embayment, Germany/Netherlands; see [21]). At some point over the decades, the concrete structure began to submerge into the floodplain sediments to complete coverage in the river bank (Figs. 1, 2b–d). As the construction year of the particular tank trap is known (= 1939), this field observation suggested the research question whether it is possible to use the concrete structure as a landmark for evaluating subsidence that occurred after the decline of coal mining in the

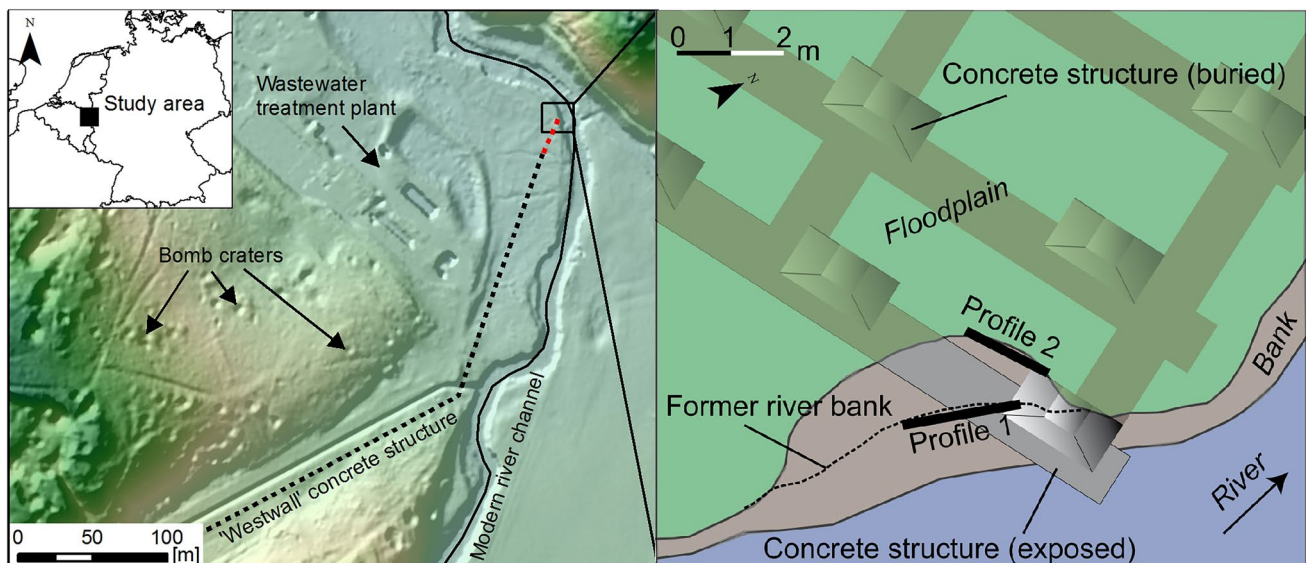
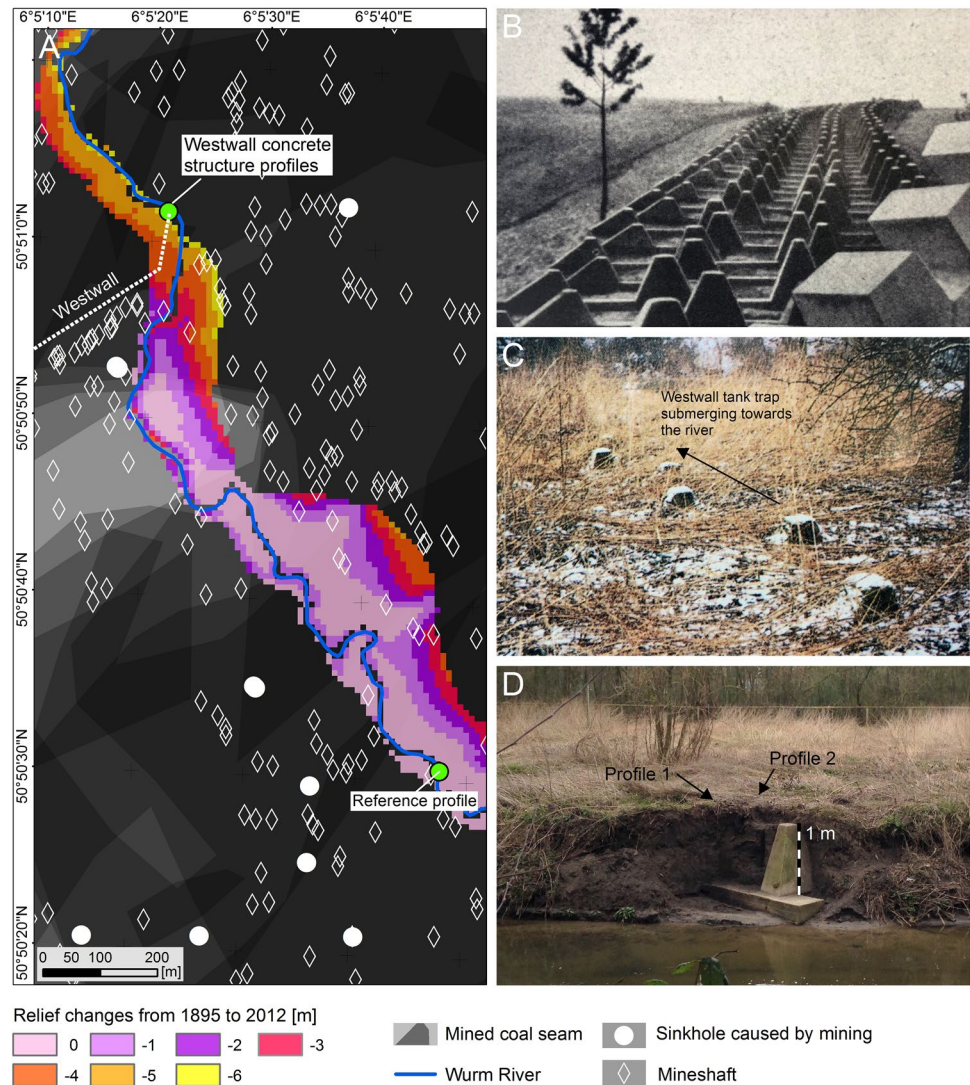


Fig. 1 Study site overview (left) and draft of the field situation (right) with the concrete structure profiles at the Westwall site sampled in 2013 and 2018 (plan view)

Fig. 2 **a** Relief changes 1895–2012 in m and the location of the sampling sites, **b** Westwall original appearance (off-site) [24], **c** Westwall submerging into sediment (on-site; view direction towards the river) [24], **d** bank profile; view from the opposite bank; the Westwall runs towards the left of the picture



twentieth century in the study area [see 22, 23]. With this period and the environmental and morphological setting of the study site, age determination with radiocesium was the most promising method to determinate the sedimentation sequence. Historical maps suggest that at the time of the fallout events described above, the concrete structure was exposed on a point-bar surface. With ongoing accumulation of point-bar sediments and a subsequent change from lateral accumulation to overbank deposition, the surface elevation rose, causing a decrease in flooding recurrence interval.

1.3 Objective

According to the above considerations, this article aims at estimating sedimentation rates, or subsidence rates, respectively, and discusses the issues and complexity that emerge with radiocesium dating in highly dynamic

riparian environments. This case study is related to other publications concerning the Wurm River that were part of a research project funded by the Deutsche Forschungsgemeinschaft. The research project aimed at investigating the pre-industrial to post-industrial human impact on small river catchments in Central Europe by the example of the Wurm River; see [21, 23, 25, 26]. In this context, riparian profiles were sampled and measured for radiocesium dating. On the basis of a floodplain profile, the applicability of radiocesium dating in the study area was verified. The actual target profile in between the concrete tank trap was sampled to assess the subsidence of the structure. In an unpublished master's thesis (Krauser 2012), subsidence rates were derived from a comparison of historical maps and the recent digital elevation model (Fig. 2a). The verification was unsuccessful in the first attempt, as the measurement results were not suitable for an interpretation. Therefore, the profile was sampled a second time later on.

The unsatisfactory data situation raised the question of the extent to which the methodology is applicable here and in comparable alluvial sediments.

2 Methods

To verify whether dating with ^{137}Cs is applicable in the study area, a riparian profile was created in the vicinity of the actual target site of the study, which is the Westwall concrete structure. This reference profile was created 1.4 km upstream; the site was chosen for being the closest to the Westwall profile available for sampling, and, furthermore, including sharply defined natural levee sediments with a presumed age consistent with the period of investigation. The profile at the Westwall was created in between the pyramid-shaped tank trap concrete structure that was laterally exposed in the river bank [see 26]. The floodplain and Westwall profiles were then sampled in 5-cm intervals for grain size and elemental analysis. For ^{137}Cs activity measurement via gamma spectrometry, the Westwall profile was sampled from 22 to 87 cm depth, and the floodplain profile from 10 to 65 cm depth; sampling resolution was 1 cm in both profiles.

Because of the riparian environment that is characterized by episodic and highly variable flood-related deposition, there were no models used for calculating sedimentation rates. Instead, the rates were calculated by dividing the sediment thickness above radiocesium peaks in mm by the time between the radiocesium emission year and the sampling year [see 27].

Because of the insufficient data lacking a zero-content of radiocesium at the bottom of the sample track, a second profile was sampled later on in a similar position but rotated approx. 35 to 45° and shifted 50 to 60 cm backwards, as the riverbank has been further eroded in the meantime (Fig. 1). The morphological position with regard to the river, however, remained the same. The second sampling was done to the maximum possible depth of 135 cm from the topsoil to the gaps between the tank trap footings. Sampling interval was 2.5 to 5 cm.

The tank trap was laterally exposed in a river bank that was 140 cm high; in the floodplain, the concrete structure was fully covered with alluvial sediment (Fig. 1). According to the flood risk map, the area is inundated in case of a 20-year flood [28]. The floodplain was covered with herbaceous perennials and shrubs. The profile revealed anthropogenically undisturbed natural alluvial structures, mainly point-bar sediments in the lower part and overbank deposits in the upper part; fine sand content was between 50 and 80%, silt content between 15 and 55%, and clay content between 3 and 8%. The reference floodplain site is inundated in case of a 50-year

flood [21] and covered with grass. A sharply delineated natural levee is underlain by buried topsoil and unstructured fine sand subsoil. The proportions of fine sand, silt, and clay are comparable to the Westwall profiles. Anthropogenic disturbances are absent. The measurement of the ^{137}Cs activity was performed by gamma spectrometry. The sediment samples were homogenized; roots and stones were removed. Samples were dried at 105° C for a minimum of 24 h and then weighed. For the analysis, high-purity germanium detectors (HPGe, from ORTEC and Canberra) in a low-level shielding with relative efficiencies of 30–70% were used; counting time was 15 h. Calibration of the detector efficiencies was done using a certified geometry reference source containing the nuclides ^{133}Ba , ^{57}Co , ^{139}Ce , ^{85}Sr , ^{137}Cs , ^{54}Mn , ^{65}Zn , and ^{88}Y . The average uncertainty was 0.03 Bq/kg for gamma activity below 0.5 Bq/kg, 0.04 Bq/kg for gamma activity between 0.5 and 0.7 Bq/kg, and 0.05 Bq/kg for gamma activity above 0.7 Bq/kg.

3 Results

The floodplain profile (Fig. 3, left) was characterized by two peaks (21.5 and 22.3 ± 0.05 Bq/kg) at depths of 27 and 41 cm that corresponded to the major radiocesium emission phases (1963 = max. emission by nuclear weapons testing; 1986 = emission by Chernobyl disaster). At 52 cm and downwards to the lowest sampled depth of 63 cm below the floodplain surface, the ^{137}Cs signal decreased to zero. In contrast, the results for the ^{137}Cs activity in the two concrete structure profiles (Fig. 3, middle and right) showed an ambiguous and unclear pattern, scattered, with peaks up to 14.3 ± 0.05 Bq/kg (concrete structure profile 1, 82.5 cm depth) and 22.0 ± 0.05 Bq/kg (concrete structure profile 2, 100 cm depth); a decrease to zero radiocesium activity cannot be observed. The depth gradient of the radiocesium signal was comparable in both concrete structure profiles with differences in the range of 2–3 cm in depth due to the slightly different positions of the profiles (see Fig. 1). For example, the peak at 85 cm depth in profile 1 corresponds to the peak in profile 2 in a depth of 82.5 cm. Differences in the gamma activity curves are caused by the different sampling resolutions: the more detailed measurement in profile 1 is smoothed by the coarser resolution in profile 2. In profile 1, four slightly established peaks could be recognized at 23, 33, 61, and 85 cm depths. In profile 2, slightly elevated contents were present in a curved shape between 12 and 30 cm depth. At 82.5 and 100 cm depth, the radiocesium contents showed Cs-peaks up to 22.0 Bq/kg, exceeding the average gamma activity by the factor of two.

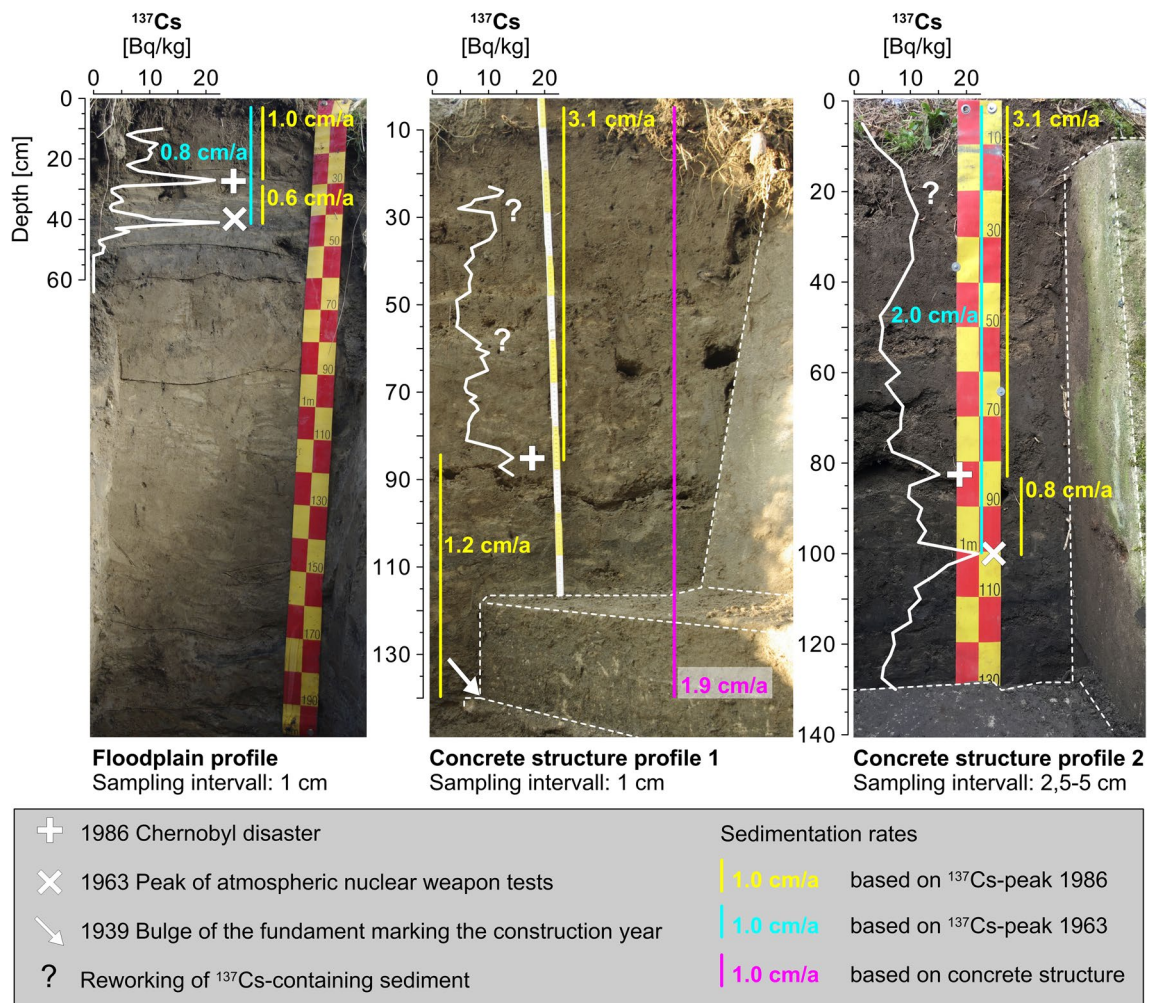


Fig. 3 Results of the radiocesium activity measurement for the reference floodplain profile (left) and for the two concrete structure profiles (middle and right). The signal in both Westwall profiles is ambiguous, and there is no decrease to zero-content at the base of

the sample line. In comparison, the floodplain profile exhibits two clear peaks and a zero-signal in the lower sample line. The dashed line marks the Westwall concrete structure

4 Discussion

From the results, the following estimations for sedimentation rates can be derived:

- In the floodplain profile (Fig. 3, left), the gamma activity shows a clear pattern with two distinct peaks of radiocesium. Therefore, the undisturbed sediment with a sharply delineated natural levee provides a valuable archive for gamma activity and hence provides reliable age information. When the radiocesium time markers are converted into sedimentation rates, these rates are 0.6 cm/a between 1963 and 1986, and 1.0 cm/a between 1986 and 2013. Compared to studies at the adjacent catchment of the Geul River by [29, 30], these rates are realistic for the given environmental setting.

- In the concrete structure profile 1 (Fig. 3, middle), the gamma activity shows an ambiguous pattern, characterized by poorly developed peaks ≥ 10 Bq/kg and oscillating values between 4 and 8 Bq/kg. This pattern might be caused by the remobilization and redeposition of contaminated sediments, either by scouring and filling of ex situ sediments, or by in situ mixing—or by a combination of both. The peak ≥ 14 Bq/kg in the lower part of the profile can be assigned to the Chernobyl disaster in 1986, as it corresponds to the same peak in concrete structure profile 2. Therefore, the following sedimentation rates can be assumed: from the fundament of the concrete structure built in 1939 to the gamma activity peak assigned to 1986, the rate is 1.2 cm/a. From the activity peak to the floodplain surface from 2013, the rate is 3.1 cm/a. From the concrete fundament to the ground surface without referring to

the measurement data, the rate is 1.9 cm/a—this rate largely corresponds to the average rate of 2.15 Bq/kg derived from the radiocesium content.

- In the concrete structure profile 2, the gamma activity curve is similar to profile 1 but smoothed. A second peak of 22 Bq/kg that might correspond to the atmospheric nuclear weapon tests in 1963 is present in 100 cm depth. The gamma activity does not decrease to zero in the bottom part in between the fundament. According to the gamma activity, the sedimentation rate is 0.8 cm/a from 1963 to 1986, and 3.1 cm/a from 1986 to 2013. The average sedimentation rate is 2.0 Bq/kg, which is in accordance with the average rate of profile 1.
- With average rates of 0.8 Bq/kg at the reference floodplain site and an average for both Westwall profiles of 2.1 Bq/kg, the sites differ by a factor of approximately 2.5. Therefore, we conclude that the sedimentation at the Westwall site was about twice as high as at the reference site. As both sites are part of the same river segment characterized by the same environmental conditions, sediment input, and discharge, we conclude that the doubled sedimentation is caused by a subsidence of the Westwall site. Given the circumstances that—according to historical maps—the concrete structure was in a point-bar position during the radiocesium emission phases, a scenario where 3.1 cm/a sediment deposition takes place without subsidence is unlikely. Although the flooding interval is twice as high as at the reference site (50 years at the reference floodplain compared to 20 years at the Westwall site, see methods section), an abrupt increase in sedimentation from 1963 onwards by a factor of almost 4 is implausible.

In the following, factors and circumstances that might have an influence on the vertical radiocesium distribution at the study site are explained. Because every study site is different for a variety of reasons, the observations are not necessarily comparable to every similar floodplain, but they are generally transferable.

- Détriché et al. [27] found that at the River Loire, sandy particles can lead to a substantial retention of radiocesium. However, in our study, a relation between the texture of the sediment and the gamma activity cannot be observed. The proportion of sand in the sampled profiles varies between 40 and 80%, but the gamma activity is independent from these variations.
- At the Westwall profile, alluvial deposition took place within a geometric concrete structure with perpendicular shapes and cavities in the footing. Therefore, the aggradation that filled up the space in the structure was most likely discontinuous, as the flow conditions

are highly variable, creating scours, in situ resuspension and subsequent sedimentation. Therefore, one or more hiatus is to be expected, and the sedimentological evidence might be biased and unreliable.

- In freshly accumulated alluvial sediments, the majority of the ^{137}Cs is adsorbed to amorphous Fe-oxyhydroxides or organic matter [31]. Therefore, the maturity of the soil is a driver for the capacity to adsorb radiocesium. In young sediments with weak pedogenesis, a higher amount of radiocesium is adsorbed, as the amount of Fe-oxyhydroxides such as ferrihydrite is higher. Soils with organic-rich O, Ah, or Bh horizons, organic-rich muds or peat are likely to adsorb high amounts of radiocesium as well. According to [32], bog-like alluvial soils tend to incorporate significant higher amounts of radiocesium. Therefore, the alluvial sediment in the Westwall profile, in which pedogenetic processes play a minor role, and the interbedded organic-rich layers might influence the vertical radiocesium distribution.
- As described by [15], soil processes such as bioturbation, leaching, diffusion, and translocation might contribute significantly to a redistribution of radiocesium. As the Westwall profiles are in a lower hydromorphological position than the reference floodplain profile, alternations of the groundwater table might cause particle redistribution.
- According to [33], forest soils are hot spots of radiocesium contamination. Hence, since the valley slopes are forested, there might appear a pulsed input of contaminated sediment through erosion during heavy rainfalls, flushing sediment enriched in radiocesium into the river system. This might result in a biased radiocesium depth profile, when contaminated sediment is deposited above the layers that correspond to the fallout period. This effect might not apply to the control profile because of a higher distance to the valley slope.
- Horizontal distribution and vertical redistribution of radiocesium seems to be related with seasonal microbial activity (Kostyuk and Bunnenberg 1999 in [14]). Kagan and Kadatsky [34] showed that after 10 years of a fallout event, up to 90% of the initial radiocesium input remains in the topsoil. In sandy soils, plants tend to an increased uptake of radiocesium [14]. Accordingly, the densely vegetated study site and the sandy substrate might lead to a 'topsoil recycling', in which the radiocesium input is circulated between plant uptake and input from vegetation dieback. This process would cause a continuing ascendant redistribution of the ^{137}Cs in the rooted soil, changing the vertical position of the ^{137}Cs -peaks and spreading the distribution into a broader, undefined signal. In comparison, the reference site is used for grazing; therefore, the principle

described above does not apply and the radiocesium peaks are more defined.

- In case of a fallout event on fresh overbank deposits, the rainwater might percolate through the uppermost sediment layer to an underlying organic-rich or clay mineral-rich topsoil, where the radiocesium is adsorbed [see 15]. Accordingly, the ^{137}Cs -peak might underestimate the sediment age, as the peak appears in a lower layer than the corresponding surface at the time of the fallout event. Based on a visual assessment of the Westwall profile, the radiocesium signal is elevated in slightly darker layers. However, the analytical results show no evidence for higher organic carbon contents in those layers.

5 Conclusions

The idea to use a concrete structure with a known construction date as a landmark for sedimentation history seems to be a promising approach. However, the results from the radiocesium activity age determination are surprising and not truly satisfactory. Compared to the reference profile, which shows clear radiocesium peaks and thus confirms the applicability of radiocesium dating in the study area, the results of the Westwall profiles are significantly less differentiated. The secondary effects of the interplay between the concrete structure and the alluvial aggradation, or the modification of flow conditions and aggradation processes, respectively, seems to destroy the continuous radiocesium signal and creates evidence that is ambiguous. The ^{137}Cs signal in the investigated sediment might have been altered by a variety of abiotic and biotic factors that are dependent and independent from the concrete structure and affected by the specific parameters of the study site. Some aspects that apply to a wider range of future studies using radiocesium can be pointed out:

- Alluvial sediments with embedded concrete structures are not suitable for sediment age determination with radiocesium activity measurement. The concrete structure itself is both giving the opportunity to reconstruct the sedimentation history and hindering the sediment age determination via radiocesium activity as a result of its impact on sedimentation processes.
- Studies that used radiocesium for age determination in alluvial sediments rarely provide discussions about the syn-sedimentary conditions of the deposition during the fallout occurrences, the detailed sediment parameters, or post-depositional mobility. In the authors' opinion, such information must be considered when interpreting radiocesium results.

- The sediment age determination with radiocesium in alluvial sediment is a well-functioning method if applied on 'reliable' sediment such as natural levee sediment. If the method is to be applied in sediment with multi-factor influence, the results have to be interpreted with caution. However, the usage in highly dynamic environments should be avoided. If the sampled sediment might be eroded infrequently and irregularly, this impedes the ability to reconstruct the sedimentation history. Therefore, the subrecent topography and morphology of the riverscape should be investigated to the most possible extent.

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Authors' contributions MBL wrote the manuscript and conducted the field work. EK conducted the gamma spectroscopy measurements and was responsible for the methodology section. SS wrote the discussion of applicability of ^{210}Pb . FL, JS, and SS gave advice for data interpretation, verified the scientific validity of the results, and reviewed the manuscript.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflicts of interests.

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