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1    **Quantifying sediment and particulate phosphorus accumulation in restored floodplain**  
2    **wetlands using beryllium-7 as a tracer**

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## 1   **Abstract**

2   Floodplain wetlands in agricultural river basins provide critical ecosystem services such as nutrient  
3   retention, flood mitigation, carbon sequestration and ecological habitats and are a key component of  
4   a nature-based solution approach to restoration. In the context of the global challenge to reducing  
5   impact increasingly intensive food production on downstream ecosystems, restoration of wetlands  
6   in river floodplains offers a practical means for downstream retention and mitigation of P flux from  
7   upstream agricultural land. Data on short term, flood event related, accretion rates are, however,  
8   difficult to acquire via conventional monitoring yet such information is essential for restoration  
9   planning and scenario testing. Here, we evaluate a promising approach that applies naturally-  
10   occurring short-lived fallout radionuclide (FRN)  $^7\text{Be}$  as a tracer to quantify sediment and, by  
11   association, particulate phosphorus retention rates in restored floodplain wetlands. Following a  
12   series of major inundation events, a restored floodplain unit was sampled to determine  $^7\text{Be}$   
13   inventories of the floodplain relative to an undisturbed reference site. This was undertaken in  
14   conjunction with direct measurement of sedimentation as an independent check of FRN results.  
15   Accretion rates up to  $27 \text{ kg m}^{-2}$  were recorded using  $^7\text{Be}$  for recent deposition events compared to a  
16   longer term annual average rate of ca.  $6 \text{ kg m}^{-2}$  derived from  $^{137}\text{Cs}$  measurements. While accretion  
17   rates varied spatially and temporally, there was excellent coherence between FRN-based  
18   measurements and direct measurements once rigorous correction for particle size effects on tracer  
19   properties had been undertaken. The study demonstrates the important contribution that FRN  
20   technology can make to support wetland management and restoration initiatives and the essential  
21   need for a systems thinking approach across the soil-sediment continuum. Such decision support  
22   tools will become increasingly important in the 21<sup>st</sup> century with growing anthropogenic pressure  
23   on aquatic ecosystems related to upstream food production, and implementation of more nature-  
24   based solutions such as restored wetlands to counteract these pressures.

1

2 Keywords: wetland restoration; nature-based solutions; floodplain sedimentation; fallout

3 radionuclides; systems thinking

4



5

6 **Graphical abstract**

## 1. Introduction

The sources, transport and destiny of suspended sediment and associated nutrients have been intensively studied in many river basins worldwide (Walling, 1999). Riparian wetlands and floodplains have been proven to act as important sinks for river sediments when water inundates the floodplain and sedimentation of coarse and fine particles takes place (Mitch et al., 1979; Cooper et al., 1987; Walling, 1999, Owens and Walling, 2003). Thus, several studies have shown the importance of riparian wetlands and floodplains as sinks of river sediments (Kuenzler et al., 1980; Lowrance et al., 1986; Walling and He, 1997; Brunet et al., 1994; Kronvang et al., 2002).

Restoration and management of wetlands at floodplain scales can be effective practices for retention and mitigation of P downstream of agricultural land (Kröger et al., 2012; Audet et al., 2020) but there is limited information on shorter-term sedimentation dynamics related to recent restoration initiatives targeting sediment and nutrients. This represents an important knowledge gap for restoration planning in terms of both the approach taken and the extent of measures. Herein, it is essential to take a whole-of-system approach along the soil-sediment continuum.

Prior studies have included many different techniques to measure sedimentation on floodplains such as use of bulk sediment traps (Mitch et al., 1979; Kronvang et al., 2002), sedimentation captured on artificial Astroturf mats (Kronvang et al., 2009; Poulsen et al., 2014) and sediment budgets (Novitzki, 1978; Brunet et al., 1994). Investigations have also used longer-lived fallout radionuclide (FRN) tracer techniques such as  $^{210}\text{Pb}$  dating (Kadlec and Robbins, 1984) and  $^{137}\text{Cs}$  dating (Johnston et al., 1984; Walling, 1999) that provide high resolution annual average estimates from retrospective sampling over periods of decades. This permits site selection for restoration to be driven by current management questions but does not enable the benefits of recent management actions to be assessed since detail is lost within long-term averaged data.

1 Here we demonstrate evidence of the efficacy of a natural tracer-based approach to close this  
2 knowledge gap and support assessment of wetland restoration sites before and after implementation  
3 for sediment and nutrient retention. Short-lived cosmogenic beryllium-7 ( $^7\text{Be}$ ) with its half-life of  
4 53.3 days offers potential to estimate short-term rates of sediment deposition, enabling assessment  
5 of the effects of management practices. To date, limited studies have employed  $^7\text{Be}$  as a tracer to  
6 measure sedimentation rates in geomorphic sink zones (Blake et al., 2002; Neubauer et al., 2002).  
7 To the best of the authors' knowledge Blake et al. (2002) provide the only published example of its  
8 use to assess overbank sedimentation on floodplains and the opportunity to provide critical post  
9 restoration data on sediment input and storage has yet to be demonstrated. This contribution  
10 demonstrates for the first time this important use of short-lived FRNs in a wetland restoration  
11 context. Such information is becoming increasingly important with catchment restoration  
12 programmes being undertaken to meet the challenges of food production versus environmental  
13 regeneration and protection and in particular, the critical role of wetlands in management of river  
14 basin nutrient fluxes to the coastal zone.

15 Wetland restoration projects fit well into the nature-based solutions agenda (Albert et al., 2021)  
16 wherein allowing temporary floodplain inundations increases the water, sediment and nutrient  
17 storage potential of the landscape. Thus, many river and wetland restoration programmes have been  
18 implemented in countries worldwide in order to increase biodiversity values (Mitch and Gosseling,  
19 1993; Maltby et al., 1994), water storages (Verhoeven et al., 2008) and the use of wetlands and  
20 floodplains as sediment and nutrient sinks (Walling et al., 2003; Kronvang et al., 2009; Hoffmann et  
21 al., 2011; Gonzalez-Sanchis et al., 2015; Hoffmann et al., 2020). In this context, information on  
22 sediment accretion rates by restored wetland and floodplain units is required to support restoration  
23 decisions and could also assist in the development of Payment for Ecosystems Services systems  
24 (e.g. Rouquette et al., 2009; Villa and Bernal, 2018). Moreover, information on sediment accretion

1 rates is important for the restored wetland and floodplain units functioning as a phosphorus sink and  
2 for considerations concerning the life span of the restored unit.

3

4 The Water Framework Directive (EC, 2000) demands that catchment management authorities of the  
5 European Union Member States improve the ecological status of surface waters by 2027. One of the  
6 main pressures that impact the ecological state of freshwaters are phosphorus emissions from point  
7 sources and diffuse sources (Kronvang et al., 2007). Therefore, in striving to meet the Directive's  
8 targets, a key focus for catchment managers is reducing P loadings introducing both source and  
9 transport management mitigation methods (Schoumanns et al., 2014).

10

11 To quantify the effect of river basin restoration measures via nature-based solutions, reliable and  
12 cost-effective post restoration monitoring methods are required. In the context of sediment and  
13 nutrient retention by wetlands, we propose that tracer tools can overcome limitations in traditional  
14 sediment trapping technology and can assist in the construction of sediment budgets to determine  
15 sediment and particulate phosphorus storage in restored riparian wetland.

16

17 Beryllium-7 tracer technology has been proposed as a valuable decision support tool within the  
18 context of catchment management and restoration (cf Mabit and Blake, 2019). The aim of this study  
19 was to demonstrate and validate the application of  $^7\text{Be}$  as a tracer for quantification of sediment and  
20 associated phosphorus accretion rates on restored floodplain wetlands. The work also aims to add  
21 rigour to the tracer conversion process to provide a transferable protocol that can effectively deliver  
22 a new decision support tool for management and restoration of floodplain and riparian wetlands.

1

2



## 2. Study site: The restored reach and floodplain of the River Odense, Denmark

In Denmark, River Basin Management Plans under the EU Water Framework Directive included an extensive wetland restoration programme for reduction of nutrient transport to estuaries and a total of 23,275 ha had been restored in 2020 (Hoffmann et al., 2020). Sediment associated nutrients and contaminants remain a critical challenge in this region (Thodson et al., 2019). Moreover, a new greening plan for Danish agriculture adopted in October 2021 to reach the 70% CO<sub>2</sub> emission reduction goal in 2030 includes among other elements restoration of a total of 100,000 ha of wetland areas in Denmark before 2030.

This study was carried out along a 6 km restored river and floodplain site along the River Odense on the island of Funen, Denmark (55°13' N, 10°15' W) (Fig. 1) during a period of floodplain inundation over the re-wetted landscape. As well as acting to revitalise the natural hydraulic interaction between a river and its floodplain, wetland restoration programmes have been included in two Danish Action Plans for Nutrient Reduction (Kronvang et al., 2005) and this site is an innovative exemplar of this process with transferable relevance for other wetland restoration schemes worldwide. The studied floodplain was restored in autumn 2003 by re-meandering and reducing the flow capacity of the formerly straightened river channel (Fig. 1). The restored river floodplain site encompasses a total of 125 ha riparian areas that have been transformed from intensively cultivated (ploughed) land to permanently grazed meadows that are periodically inundated following restoration.

[insert Figure 1]

Figure 1: Overview map showing (a) River Odense location and (b) catchment area with (c) the study area where the river was re-meandered in autumn 2003 and (d) sample transects.

The catchment area upstream of the studied area (Fig. 1) is 254 km<sup>2</sup>. Land use in the catchment at the time of study was predominantly agriculture (ca. 65%). The area is underlain by moraine deposits from the last glaciation period (Weichsel) and primarily composed of clayey sandy (ca. 40%) and sandy clay soils (35%). Average annual long-term precipitation amounted to 727 mm and average annual long-term runoff at the restored site amounted to 316 mm during the period 1989-2011. During the same period median minimum discharge was 1.2 m<sup>3</sup> s<sup>-1</sup>, median maximum discharge was 3.3 m<sup>3</sup> s<sup>-1</sup> and absolute maximum discharge was 22 m<sup>3</sup> s<sup>-1</sup>. The baseflow (BFI) index for the River Odense is 0.67.

### **3. Experimental design and tracer method development**

#### *3.1. Direct measurement of sediment deposition and P content on the restored floodplain surface*

To permit development and validation of the <sup>7</sup>Be approach in restored floodplain wetlands, deposition of sediment on the restored floodplain was assessed directly, during a period of storm events that inundated the floodplain on several occasions (Fig. 2), using 1-3 artificial in excess of 80 pre-weighted Astroturf mats (Astroturf Textile Management Associates, Inc., Dalton, GA, USA) with a dimension of 15 x 15 cm installed at different distances along a randomly placed transect from the river channel (1, 5.6, 10.7, 16.5, 23.8, 31.1, 40.8, 52.3, 71.8 and 101 m) between 2003 and 2012. The Astroturf mats, proven as an effective sediment trap elsewhere (e.g. Owens and Walling, 2002), were deployed in autumn (prior to the main rainy period) on the floodplain and fixed to the grass meadow with four long spikes (one in each corner). After the flooding season each year, the Astroturf mats were retrieved from the meadow and brought to the laboratory in small plastic bags.

After recovery of the Astroturf mats, the amount of sediment on each mat was determined by pre-weighing of the Astroturf mats and the newly collected Astroturf mats, and sediment was dried and re-weighed at 60 °C for 24 h. The total mass of sediment on each Astroturf mat was then calculated by subtracting the pre-weighed weight of the Astroturf mat. These samples were the most recent of a longer term direct accretion measurement programme that has been in operation since floodplain restoration (Poulsen et al., 2014). The content of total P in the sediment core was analyzed by spectrophotometry according to the method described in Svendsen et al. (1993).

[Insert Figure 2]

Figure 2: flow regime of the restored river reach during the study period showing periods of floodplain inundation, where flow (daily mean Q) exceed the bankfull flow (shown by red line), and suspended sediment concentration (SCC)

### *3.2. Sampling for $^7\text{Be}$ and $^{137}\text{Cs}$ to quantify sediment deposition on the restored floodplain surface*

A key assumption for the use of  $^7\text{Be}$  as a tracer is that following fallout, sorption to particles is rapid and effectively irreversible over the timescales of interest (Taylor et al., 2013; Taylor et al., 2012a; Mabit et al., 2008). By determining the difference in  $^7\text{Be}$  areal activity (inventory) at a sampling point and comparing the results with the activity at a reference baseline, from a site that has not been inundated nor eroded, and considering the  $^7\text{Be}$  activity in transported sediment, deposition rates can be estimated (Blake et al., 2002; Neubauer et al., 2002). A possible limit of using radioisotopes for estimating sediment accretion rates on floodplains is that  $^7\text{Be}$  activity is not evenly

1 distributed across sediment size fractions (Blake et al., 2009; Taylor et al., 2014; Mabit and Blake  
2 2019), therefore, it is necessary to account for variation in  $^7\text{Be}$  activity in transported sediment size  
3 fractions with respect to total bulk sediment activity. For example,  $^7\text{Be}$  typically displays  
4 enrichment in fractions  $< 63 \mu\text{m}$  (Blake et al., 2009; Taylor et al., 2014), implying that if these  
5 fractions are representative of deposited material then failure to account for enrichment will lead to  
6 overestimation of deposition rates.

7 The  $^7\text{Be}$  approach to determine short-term accretion rates requires quantification of (i) the  $^7\text{Be}$   
8 inventory ( $\text{Bq m}^{-2}$ ) at a local reference site that has not been inundated by flood waters i.e. only  
9 received aerial fallout inputs, (ii) the  $^7\text{Be}$  inventory on the inundated floodplain surface ( $\text{Bq m}^{-2}$ )  
10 which, after accretion, will be in excess of the reference inventory, (iii) the  $^7\text{Be}$  activity  
11 concentration ( $\text{Bq kg}^{-1}$ ) of the suspended sediment transported by the river channel during  
12 inundation events (iv) the relationship between  $^7\text{Be}$  and specific surface area ( $\text{m}^2 \text{g}^{-1}$ ) of the  
13 sediment (Taylor et al., 2014) to permit effect of particle size enrichment to be accounted for and  
14 (v) the specific surface area of the above sediment samples. Alongside, in the context of the  
15 restoration history, it is also useful to establish medium-term rates of accretion which can be  
16 derived using  $^{137}\text{Cs}$  inventories as described elsewhere (see Walling and He, 1997; Soster et al.,  
17 2007).

18 Short bulk sediment core (50 mm diameter) samples for  $^7\text{Be}$  analysis were taken from the  
19 floodplain of the River Ouse during March 2011 and 2012 using simple cut drainpipe rings (30  
20 mm depth). In addition, deep (0.5 m) section core samples and bulk sediment core samples for  $^{137}\text{Cs}$   
21 analysis (cf Walling and He, 1997) were collected in March 2011 following the winter inundation  
22 season. The purpose of these latter samples was to support assessment of longer term accretion rates  
23 since restoration to contextualise the contemporary processes observed.

1 During the period leading up to March 2012, an *in situ* suspended sediment sampler (Laubel et al.,  
2 2002) was installed in the channel to characterise the bulk  $^7\text{Be}$  concentration of suspended sediment  
3 in transit during the inundation events. Note that this bulk signature of material entering the study  
4 reach encapsulates any influence of suspended ‘old’ material from the bed and represents the  $^7\text{Be}$   
5 activity concentration of material delivered to the floodplain surface. Bulk sediment core samples  
6 for  $^7\text{Be}$  analysis were retrieved from the floodplain in March 2012 to a depth of minimum 25 mm  
7 (based on Blake et al., 2002) plus the depth of any freshly deposited sediment where observed. Five  
8 multiple cores (diameter 54 mm) were taken at each site alongside the installed AstroTurf mats and  
9 bulked to create a spatially integrated and representative sediment sample for  $^7\text{Be}$  analysis. All  
10 individual cores were taken to the same depth. In total, ten bulk core sample sets were collected at  
11 different distances from the river channel reflecting the AstroTurf mat sites. Eleven bulk cores were  
12 taken at the nearby reference site on the floodplain at a point that was above the height of all  
13 inundations which occurred during the sampled winter inundation season.

14 For assessment of longer term accretion rates over the post restoration period (2003 – 2011), deeper  
15 cores for  $^{137}\text{Cs}$  analysis were collected (cf Walling and He, 1997). To determine the depth profile,  
16 one sediment core was collected 16 m from the river channel beside the AstroTurf mat site that had  
17 been monitored during all winter seasons since the restoration in autumn 2003. The sediment core  
18 was retrieved using a Kajak coring device with a diameter of 5.4 cm equipped with a steel cap to  
19 enable coring in sandy material (Svendsen and Kronvang, 1995). The sediment core was brought to  
20 the laboratory and sliced into thirty two 1 cm slices. Each slice was air dried at 40°C and weighed  
21 prior to homogenisation with a pestle and mortar and passed through a 2 mm sieve. In addition,  
22 bulk sediment samples for  $^{137}\text{Cs}$  analysis were retrieved to a depth of at least 30 cm along the full  
23 transect, including all deposit material from the river on both the floodplain experiencing  
24 inundations, where eight representative bulk sediment samples were retrieved. To determine the

1 reference inventory, 11 representative sediment bulk samples taken from a nearby level site above  
2 the height of all inundations since the 2003 river restoration.

3 All sediments were processed following the methods of Pennock and Appleby (2002). Samples  
4 were air dried ( $< 40\text{ }^{\circ}\text{C}$ ) and disaggregated by hand using a pestle and mortar. The samples were  
5 then sieved to remove large mineral debris  $> 2\text{ mm}$ , care being taken to ensure retention of organic  
6 material. A subsample of the  $< 2\text{ mm}$  fraction was then oven dried at  $105\text{ }^{\circ}\text{C}$  to provide a dry-  
7 weight correction value. The weights of both the  $> 2\text{ mm}$  and less than  $2\text{ mm}$  fractions were  
8 recorded and the material fraction  $< 2\text{ mm}$  was shipped to University of Plymouth Consolidated  
9 Radioisotope Facility for gamma spectrometry analysis within 2 weeks of sampling.

10 Air-dried  $< 2\text{ mm}$  soil samples were packed into Marinelli beakers for analysis by gamma  
11 spectrometry. All isotope analyses were carried out using a low background high purity germanium  
12 (HPGe) gamma detector (GM50-83-LB-C-SMN-S Planar, Ortec, UK). Calibration was carried out  
13 using standards of the same geometry as the experimental samples. Standards were prepared using  
14 QCY58b-mixed standard solution (G E Healthcare, Amersham, UK) distributed in a mineral soil  
15 matrix. Detector efficiency for  $^7\text{Be}$  ( $477.6\text{ keV}$ ) was determined by interpolation between the  
16 efficiency values of  $^{137}\text{Cs}$  ( $661.7\text{ keV}$ ) and  $^{113}\text{Sn}$  ( $391.7\text{ keV}$ ). Sample counts were corrected for  
17 background emission, geometry efficiency and decay. All values were decay corrected to the time  
18 of sampling and reported as activity ( $\text{Bq kg}^{-1}$ ). Laboratory analytical quality control procedures  
19 were carried out in accordance with Wallbrink et al. (2002).

20

### 21 *3.3. Converting $^7\text{Be}$ and $^{137}\text{Cs}$ measurements into short- and medium-term accretion rates*

22 The FRN inventories were converted to accretion rates based on the excess inventory (i.e. the  
23 amount of inventory greater than that measured at the non-depositional and non-eroding site)

1 method described by Walling and He (1997) for  $^{137}\text{Cs}$  and Blake et al. (2002) for  $^7\text{Be}$  wherein the  
2 latter was developed further to account for particle size selectivity and deposition across multiple  
3 events.

#### 4 *3.4 Examining the particle size association of $^7\text{Be}$*

5 A key first step in the  $^7\text{Be}$  inventory conversion process was to determine how fluvial sorting would  
6 affect the  $^7\text{Be}$  activity concentration of deposited sediment across the floodplain. Due to enrichment  
7 of  $^7\text{Be}$  in finer sediment fractions with greater specific surface area, it was necessary to i) determine  
8 the specific surface area, related to sediment size distribution, in deposited material and ii) estimate  
9  $^7\text{Be}$  enrichment factors for material at each sampled point on the floodplain surface. Estimating the  
10 relationship between  $^7\text{Be}$  and specific surface area in field material involves separating fractions  
11 from a large mass of sediment to ensure a suitable mass for gamma detection. A more practical  
12 approach for determining enrichment, using a low mass of sediment, was adopted here using stable  
13 Be ( $^9\text{Be}$ ) following Taylor et al. (2012b).

14 A subsample of reference surface floodplain sediment was selected for the experiment and assumed  
15 to be representative of suspended material. The sample was air dried, lightly disaggregated by hand  
16 and sieved ( $< 2\text{ mm}$ ) prior to being shaken with an equilibrating solution of stable  $^9\text{Be}$  based upon  
17 the method given in Taylor et al. (2012b). Stable  $^9\text{Be}$  solution was obtained by dissolving  $\text{BeCl}_2$  salt  
18 (99%) (Sigma Aldrich, UK) in ultra-pure water (Millipore Milli Q Plus 185 system), with solution  
19 pH being buffered to 5.6 (natural rainwater pH) using NaOH. A 100 g subsample of the soil was  
20 rehydrated overnight in 100 mL ultrapure water after which 100 mL Be solution ( $10\text{ mg L}^{-1}$ ) was  
21 added to obtain a  $5\text{ mg L}^{-1}$  concentration in the vessel. The sample was then placed on a reciprocal  
22 shaker for 24 hours. This concentration enabled clear detection of Be sorption above natural

1 background levels. A 100 g soil sample was used to ensure that a suitable mass of material would  
2 be retrieved for each particle size fraction during the separation procedure.

3 Following shaking, the material was centrifuged at 3500 g for 20 minutes (Sorvall Legend RT, DJB  
4 Labcare Ltd, UK) and an aliquot of the supernatant was sampled, acidified ( $\text{pH} < 2$ , HCl) and  
5 retained for analysis to determine sorption from solution ( $> 98\%$  Be sorption to soil from solution,  
6 corrected for vessel sorption). The soil was then allowed to partially air dry in the open vessel to  
7 enable the sample to be lightly disaggregated and homogenised by hand. A subsample of known  
8 mass was taken to determine total Be concentration in the soil. Ultra-pure water was then added to  
9 the remaining sample (to above surface) and the sample was left to rehydrate overnight. Following  
10 this, the vessel was placed in an ultrasonic bath (Laborette 17, Fritsch, Germany) for 20 minutes to  
11 aid particle dispersion.

12 Next, the sample was added to a known volume of ultra-pure water in a settling column and  
13 thoroughly mixed into suspension. The sample was split by a combination of sieving and settling to  
14 produce samples of decreasing particle size and hence increasing specific surface area, the key  
15 measure used in the particle size correction process. All samples were dried at  $40\text{ }^{\circ}\text{C}$ . During each  
16 stage of the separation, a subsample of solution was filtered ( $< 0.45\text{ }\mu\text{m}$ ) and retained for analysis to  
17 confirm that there was no loss of  $^9\text{Be}$  to the solution during the process. In addition to the  
18 fractionation of the experimental sample, control samples, not mixed with  $^9\text{Be}$  solution, were  
19 separated in the same manner to enable determination of background  $^9\text{Be}$  concentrations.

20 The air-dried sample fractions were then homogenised by hand and triplicate subsamples of known  
21 mass were digested using microwave-assisted digestion (MARS 5 Accelerated Reaction System,  
22 CEM Microwave Technology Ltd, UK) following Hassan et al. (2007). All samples were analysed  
23 for Be concentration using ICP-OES (Varian 725 ES, Varian, Australia). Separate subsamples were



oven dried at 105 °C to allow moisture correction to be calculated. Further subsamples were oxidised using H<sub>2</sub>O<sub>2</sub> to remove organic matter prior to particle size analysis using a Malvern Mastersizer 2000 with Hydro-G (Malvern Instruments Limited, Malvern, UK) according to ISO 13320:2009.

The concentration of <sup>9</sup>Be in the separated materials was then plotted against specific surface area (SSA) as determined by laser granulometry, and the associated power function was then used to estimate <sup>7</sup>Be activity in field samples (following Taylor et al., 2014), taking into account its grain size composition using SSA data from (i) the cores samples and (ii) surface sediment trap samples:

$$S(t) = S(m) \left[ \frac{SSA(c)}{SSA(s)} \right]^v \quad (1)$$

where  $S(t)$  = the adjusted <sup>7</sup>Be concentration in the deposited sediment fraction (mg kg<sup>-1</sup>) and  $S(m)$  = the measured <sup>7</sup>Be concentration (mg kg<sup>-1</sup>),  $SSA(c)$  = the standard specific surface area (m<sup>2</sup> g<sup>-1</sup>) to which the sample is adjusted (in this case the SSA of the suspended sediment collected from the channel during the inundation events).  $SSA(s)$  is the SSA of the sediment and  $v$  is a parameter reflecting the particle size selectivity of the sorption process (cf. He and Walling, 1996) which in this case was derived from the <sup>9</sup>Be:SSA relationship derived above.

16

### 3.5 Conversion model for <sup>7</sup>Be floodplain inventories.

Event-based deposition estimates ( $R_d$ , kg m<sup>-2</sup>) based on <sup>7</sup>Be inventory values (Bq m<sup>-2</sup>) were proposed by Blake et al. (2002) as follows:

$$R_d = \frac{I_{ref} - I_{dep}}{S} \quad (2)$$

20

1 where  $I_{ref}$  =  $^7\text{Be}$  inventory obtained from the reference cores and fallout estimates ( $\text{Bq m}^{-2}$ ),  $I_{dep}$  =  
2  $^7\text{Be}$  inventory in the deposition zone ( $\text{Bq m}^{-2}$ ) and  $S$  =  $^7\text{Be}$  the activity of the deposited sediment  
3 ( $\text{Bq kg}^{-1}$ ).

4 In this case, however, deposition resulted from a series of inundation events and hence there was a  
5 risk that inputs from events early in the study period might be underestimated due to radioactive  
6 decay of the associated  $^7\text{Be}$  signal. This issue has previously been raised in the context of erosion  
7 estimates using  $^7\text{Be}$  by Walling et al. (2009) who developed a model to apportion erosion across  
8 multiple events. In a similar fashion, a new deposition model was developed for this study which  
9 accounted for additional  $^7\text{Be}$  inventory from fallout ( $F$ ) as well as deposited sediment, while  
10 simultaneously accounting for radioactive decay across the study period to avoid underestimation of  
11 deposition rates.

12 Here, a daily value of relative sediment loading  $Sl$  was estimated across the study period using  
13 channel-suspended sediment concentration data and calculated from:

$$14 \quad Sl = \frac{Sl_d}{Sl_t} \quad (3)$$

15 where  $Sl_d$  = daily sediment loading and  $Sl_t$  = total sediment loading for the study period. Sediment  
16 loading was selected as it integrates suspended sediment concentration and water depth, via flow,  
17 both of which control potential accretion on the floodplain surface. The loading factor was only  
18 applied at times over overbank flow to ensure the correction process related to overbank events  
19 only.

20 Values of deposition rate  $R$  on each inundation day ( $t$ ) are assumed to be proportional to the relative  
21 sediment loading at times of overbank flow:

$$R(t) = Sl(t) \times C \quad (4)$$

where  $C$  represents a constant value.

Where deposition has occurred, the inventory in place at the end of each day,  $I_{dep}(t)$ , can be described as:

$$I_{dep}(t) = I_{dep}(t-1)e^{(-\lambda)} + F(t) + I_{gain}(t) \quad (5)$$

The increase in inventory at a sample point on day  $t$  ( $I_{gain}(t)$ ) will reflect the depth of deposition  $R(t)$  and the particle size corrected  $^{7}\text{Be}$  activity of the deposited sediment  $S(t)$ :

$$I_{gain}(t) = Sl(t) \times C \times S(t) \quad (6)$$

A continuous mass balance for each study point was established and solved for  $C$  using an automatic optimization procedure. By applying the value of  $C$  obtained for an individual sampling point to time series of  $Sl(t)$  for the study period, estimates of event deposition rates and the total deposition for the study period was calculated (cf Walling et al., 2009). The mass balance required quantification of reference inventory losses and gains throughout the study period which was achieved by estimating rainfall activity concentrations using an iterative process to fit the measured reference inventory. To explore the sensitivity of the conversion to particle size correction factors, the specific surface area of the recently deposited sediment (from AstroTurf mats) and the bulk cores (25 mm depth) were used independently.

### 3.6 Conversion of $^{137}\text{Cs}$ inventories.

The excess inventory approach (cf. equation 1), i.e. excess relative to reference site, adopted for conversion of  $^{137}\text{Cs}$  data followed the principles of Walling and He (1997) but in this case the

1 period to which the excess inventory applied was not from mid 1950s or 1963 but the time since  
2 restoration i.e. 2003 to 2011. This meant that the measured reference inventory represented the  
3 inventory of the floodplain surface from 1963 up until 2003, i.e. the time of restoration, and it had  
4 to be assumed that soil erosion on the drained and isolated, low gradient valley floor prior to 2003  
5 was minimal. It was also noted that the valley floor had been cultivated prior to restoration giving a  
6 uniform depth profile prior to sediment accumulation post restoration. For simplicity and given the  
7 short period of time since restoration, the surface concentration of  $^{137}\text{Cs}$  was used in equation 1 to  
8 avoid the need for particle size correction of contemporary suspended sediment activity  
9 concentrations, which, with variability in erosion and deposition dynamics over the 10 year period  
10 are potentially not representative.

11

## 12 **4. Results and discussion**

### 13 *4.1 Accretion rates since floodplain wetland restoration 2003 - 2011*

#### 14 4.1.1 Direct measurement of sediment accretion

15 The eight years of measured sediment deposition at the investigated transect with installed AstroTurf  
16 mats since restoration (Fig. 3) reflected deposition being clearly highest within the first 30 m from  
17 the river channel after which it declines rapidly. Accretion rates measured using AstroTurf mats  
18 annually from 2003/04 to 2011/12 (Fig. 4) show high temporal variability with highest accretion  
19 rate measured during the first winter period following the re-meandering of the channel in autumn  
20 2003 and no sediment accretion during the winter of 2008/09 (Fig. 4A). Sediment accretion rates  
21 were in general highest during winters with highest flow conditions and highest number of flood  
22 events such as the winter of 2006/2007 (Fig. 4B,C). The sediment accretion rates, ranging 0.1 to 100  
23  $\text{kg m}^{-2}$ , measured during the eight year period in this study (2003/04 to 2011/12 ) are comparable to

accretion rates found by Johnston et al. (1984) in a riparian forest levee ( $7.8 \text{ kg m}^{-2} \text{ yr}^{-1}$ ), Brunet et al. (1994) for a single floodplain, riparian zone, France ( $28.9 \text{ kg m}^{-2} \text{ yr}^{-1}$ ), Tockner et al. (1999) for a 10 km stretch of floodplain along the River Danube, Austria ( $25 \text{ kg m}^{-2} \text{ yr}^{-1}$ ) and Walling (1999) for 21 floodplains in UK ( $0.4\text{-}12.2 \text{ kg m}^{-2} \text{ yr}^{-1}$ ).

[insert Figure 3]

Figure 3: Annual floodplain sedimentation rates directly measured using Astroturf mats along the sample transect line for the period 2003/04 to 2011/12. Note that no floods took place during the winter of 2008/09 and that grass mats were not installed during the winter of 2010/11.

[insert Figure 4]

Figure 4: (a) average sediment accretion rates, (b) runoff during the winter period October to March (c) and number of flood events in each winter period for the period 2003/04 to 2011/12. Note that no floods took place during the winter of 2008/09 and that grass mats were not installed during the winter of 2010/11.

4.1.2 Tracer-based assessment of medium-term sediment accretion rates to contextualise the contemporary situation

1 The  $^{137}\text{Cs}$  inventory at the non-inundated reference site on the River Odense floodplain was  $1002 \pm$   
2  $129 \text{ Bq m}^{-2}$  ( $n = 10$ ). The sediment core taken at the central part of the transect showed measurable  
3  $^{137}\text{Cs}$  activities to a depth of 35 cm (Fig. 5). The lack of a clear 1963 peak in activity concentration  
4 relates to the fact that the land had been cultivated prior to restoration mixing the  $^{137}\text{Cs}$  inventory  
5 associated with direct fallout throughout the ploughed layer of soil. Inundation after 2003  
6 restoration led to a total inventory of  $2070 \pm 408 \text{ Bq m}^{-2}$ ,  $1068 \text{ Bq m}^{-2}$  in excess of the reference.  
7 Using the surface concentration of  $^{137}\text{Cs}$  to represent sediment inputs, the excess inventory equates  
8 to  $74 \text{ kg m}^{-2}$  accretion over the 8 years since restoration in line with the direct measurements  
9 (approximately 16 cm true depth given the low bulk density of the deposited sediment). Post  
10 restoration accretion is therefore represented by the upper ca  $8 \text{ g m}^{-2}$  mass depth of the section core  
11 (Figure 5) at which point a change in the character of total P loading of the deposited material is  
12 observed (see section 4.3).

13  
14 [insert Figure 5]  
15

16 Figure 5: Depth distribution of  $^{137}\text{Cs}$  activity concentration and total P concentration at 16 m from  
17 the river bank in the restored floodplain against mass depth (note there was no measurable activity  
18 concentration below  $16 \text{ g cm}^{-2}$ ).

#### 19 20 4.2. *Quantifying short-term sediment deposition on floodplains using $^7\text{Be}$ as tracer*

21 The  $^7\text{Be}$  activity concentrations of the suspended sediment samples collected on 16/11/2011,  
22 14/12/2011 and 12/01/2012 were 12.7, 65.3 and  $32.6 \text{ Bq kg}^{-1}$ , respectively. Variability in

concentration was likely related to (i) the developing catchment inventory during the study period and (ii) variability in erosion process, e.g. sheetwash versus rill erosion, during the study period (Walling et al., 1993; Wallbrink and Murray, 1993).

The importance of accounting for sorting effects when applying fallout radionuclides to quantify sedimentation rates is illustrated in the relationship between  $^9\text{Be}$  concentration and specific surface area (SSA) (Fig. 6). While close to linear in this case, it is appropriate to represent this relationship with a power function in accord with prior work (e.g. Blake et al., 2002; 2009; He and Walling, 1997) and equation 1. While the coarser fractions display similar SSA values, potentially the influence of sediment aggregation and organic matter controls on density, there is a clear increase in  $^9\text{Be}$  concentration with increasing SSA linked to the greater reactive surface area of the fine fraction.

[insert Figure 6]

Figure 6: Relationship between specific surface area (as a proxy for overall particle size) and  $^9\text{Be}$  in the floodplain sediment to derive a correction factor for  $^7\text{Be}$  inventory conversion to deposition rates.

It is also noteworthy that the  $^9\text{Be}$  concentration had a strong positive correlation with clay content ( $R^2 = 0.98$ ). This has important implications for the  $^7\text{Be}$  approach in the context of differences in SSA of suspended sediment and deposited sediment on the floodplain surface (Table 1a). In this system, the suspended sediment captured in the channel (Table 1b) had an average SSA of  $0.11 \text{ m}^2 \text{ g}^{-1}$ . This was notably lower to that of the sediment deposited on the AstroTurf mats which ranged  $0.3$  to  $0.7 \text{ m}^2 \text{ g}^{-1}$  and was linked to a greater proportion of clay sized material in the deposits. It is possible that the material captured by the suspended sediment sampler underrepresented the clay

size fraction which further emphasises the importance of particle size correction to relate excess inventory to the  $^7\text{Be}$  contributed by deposited sediment. It is also notable that the SSA of the bulk surface cores collected was lower than that of the recent deposits. This could reflect (i) greater input of coarse material in prior overbank events of higher magnitude or (ii) remobilisation of fines within recently-deposited overbank-derived sediment over subsequent inundation events (Greenwood et al., 2013). The difference in the grade of the bulk material and the surficial deposits from the most recent events has important implications for  $^7\text{Be}$  inventory conversion to accretion rates.

At the time of sampling, the mean  $^7\text{Be}$  reference inventory measured at the non-inundated site was  $640 \pm 94 \text{ Bq m}^{-2}$  reflecting inputs from rainfall and radioactive decay over the study period (Figure 7). The  $^7\text{Be}$  inventory data showed higher inventories along the floodplain transect where sediment deposition took place during winter 2011-2012 (Figure 8). The inventory conversion approaches used to estimate sedimentation rates yielded a range of results (Table 2) wherein the extended time-series model with particle size correction based on the recent surficial deposit demonstrated excellent coherence with the AstroTurf mat-derived accretion rates (note the site closest to the river showed evidence of scour leading to an estimate of sediment loss) over the study period, i.e. event scale deposition (Table 2, dataset D; Figure 8). Correction of the  $^7\text{Be}$  excess inventory for particle size effects using the bulk core sediment properties gave results that markedly overestimated sediment accretion rates (Table 2, dataset B). This emphasises the need to use sediment material representative of the specific storm events being studied for particle size correction procedures, which presents a challenge when using the method as a retrospective investigative tool.

Table 1: (a) Particle size properties of the bulk cores collected for  $^7\text{Be}$  measurements and the recently deposited material recovered from the AstroTurf mats (b) particle size properties of the suspended sediment.



1

(a)	0 - 25 mm cores				Recent AstroTurf mat deposits			
Sample location	SSA	Sand	Silt	Clay	SSA	Sand	Silt	Clay
<i>Distance from channel (m)</i>	(m <sup>2</sup> g <sup>-1</sup> )	(%)	(%)	(%)	(m <sup>2</sup> g <sup>-1</sup> )	(%)	(%)	(%)
1	0.02	92.1	7.9	0.0	n/a			
5.6	0.03	85.0	15.0	0.0	0.28	44.5	47.7	7.8
10.7	0.07	60.9	38.6	0.5	0.34	37.2	53.3	9.5
16.5	0.10	38.7	60.5	0.8	0.40	22.5	66.4	11.1
23.8	0.11	27.4	71.7	0.9	0.47	13.7	73.1	13.3
31.1	0.11	29.9	69.2	0.8	0.52	10.0	75.5	14.5
40.8	0.12	29.6	69.6	0.8	0.57	6.2	78.0	15.8
52.3	0.14	18.4	80.6	1.0	0.56	2.8	82.9	14.3
71.8	0.13	22.4	76.8	0.9	0.78	2.1	74.7	23.2
101	0.15	18.4	80.5	1.1	0.74	3.1	75.0	21.8

(b)

Suspended Sediment	SSA	Sand	Silt	Clay
<i>Date of collection</i>	(m <sup>2</sup> g <sup>-1</sup> )	(%)	(%)	(%)
16/11/2011	0.14	26.9	71.9	1.2
14/12/2011	0.15	22.4	76.3	1.3
12/01/2012	0.06	71.4	28.2	0.3

2

3

[insert Figure 7]

4

5 Figure 7: Rainfall and associated <sup>7</sup>Be inventory development and decay at the non-inundated site in  
6 months prior to overbank events studied.

7

[insert Figure 8]

8

9 Figure 8: Beryllium-7 inventory plotted against distance from the River Odense along the transect  
10 deployed with Astroturf mats during winter 2011-2012.

1 Table 2: Sediment accretion rates estimated from  $^7\text{Be}$  measurements using both bulk and surficial sediment properties for particle size  
2 correction.

3

Distance from channel (m)	Excess $^7\text{Be}$ inventory ( $\text{Bq m}^{-2}$ )	A: Deposition measured directly on AstroTurf mats ( $\text{kg m}^{-2}$ )	B: Deposition from $^7\text{Be}$ inventory with particle size correction based on bulk core properties ( $\text{kg m}^{-2}$ )	C: Deposition from $^7\text{Be}$ inventory with particle size correction based on surficial sediment properties ( $\text{kg m}^{-2}$ )	D: Deposition from $^7\text{Be}$ inventory as for A but using extended time series model ( $\text{kg m}^{-2}$ )	<i>D: lower limit*</i> ( $\text{kg m}^{-2}$ )	<i>D: upper limit*</i> ( $\text{kg m}^{-2}$ )
1	-301	-10.0	-6.1	n/a			
5.6	1285	11.7	26.2	10.0	11.0	8.5	13.8
10.7	3819	37.2	78.0	24.5	27.0	23.3	31.2
16.5	3684	15.6	75.2	19.7	21.7	18.7	25.2
23.8	2206	8.4	45.1	10.0	11.1	9.2	13.2
31.1	1634	5.9	33.4	6.8	7.5	6.1	9.2
40.8	1038	4.7	21.2	3.9	4.3	3.2	5.6
52.3	1346	3.5	27.5	5.2	5.7	4.5	7.2
71.8	481	1.4	9.8	1.3	1.4	0.8	2.2
101	489	1.0	10.0	1.4	1.5	0.9	2.3

4

5 \* where upper and lower limit uncertainty is based on 95% confidence limits of the reference inventory estimate

[insert Figure 9]

Figure 9: Comparison of directly measured and  $^7\text{Be}$ -derived sedimentation rates (see Table 2 for details).

#### *4.3 Storage of sediment associated phosphorus on restored floodplain wetlands.*

The deposition of sediment-associated phosphorus (P) was measured at each sampling point on the floodplain as restoration of wetlands in river floodplains has been shown to be an effective practice for retention and mitigation of P flux from upstream agricultural land (Kroger et al., 2013). The concentration of TP deposited on the floodplain showed a general increase with increasing distance to the channel, a pattern which opposite to the sediment deposition amount (Fig. 3) which correlates with particle size sorting effects. The deposited sediment near the channel bank edge (1 m) had a measured average annual total P concentration amounting to  $417 \pm 151 \text{ mg P kg DW}^{-1}$  which increased to  $1017 \pm 330 \text{ mg P kg DW}^{-1}$  at a distance of 5.5 m,  $2309 \pm 372 \text{ mg P kg DW}^{-1}$  at a distance of 16.5 m from the channel,  $3554 \pm 455 \text{ mg P kg DW}^{-1}$  at a distance of 52 m from the channel and  $6610 \pm 4918 \text{ mg P kg DW}^{-1}$  at a distance of 101 m from the channel during the period 2003/04-2011/12. The deposition of P varied both spatially and temporally according to the accretion patterns for sediment with an average deposition of  $6.87 \text{ g P m}^{-2} \text{ yr}^{-1}$  (range:  $0\text{-}13.5 \text{ g P m}^{-2} \text{ yr}^{-1}$ ) during the investigated period (2003/04 to 2011/12). The range is comparable to upper values seen elsewhere e.g.  $1.46 \text{ g P m}^{-2} \text{ yr}^{-1}$  in eleven floodplains in USA by Johnston (1991),  $12.7 \text{ g P m}^{-2} \text{ yr}^{-1}$  in a French floodplain by Brunet and Astin (1998),  $1.3\text{-}11.6 \text{ g P m}^{-2} \text{ yr}^{-1}$  in twenty-one floodplains in UK (Walling, 1999).

1 The depth profile of total P in the sectioned core showed an increase in mass concentration from the  
2 point where  $^{137}\text{Cs}$  data indicated post-restoration deposition to commence i.e. at ca  $7.5 \text{ g m}^{-2}$  mass  
3 depth (approximately 16 cm true depth). The upper section total P concentration reaches a steady  
4 ca.  $2.6 \text{ g kg}^{-1}$  which compares well to the mean average concentration measured in the deposited  
5 sediment amounting to  $2309 \pm 372 \text{ mg P kg DW}^{-1}$  during the period 2003/04-2011/12.

6

7

8 This study demonstrates that restored floodplains and associated wetlands act as an important sink  
9 for excess P in agricultural river basins and should be included as a part of catchment-wide  
10 initiatives to mitigate enhanced P flux due to agricultural land use. In the context of this global  
11 challenge, the Danish Nature Agency decided in 2010 to include restored floodplains and associated  
12 wetlands, so-called 'P-wetlands', as a mitigation option for reducing P loads to lakes and estuaries  
13 as part of the WFD River Basin Management Plan I (RBM I; Kronvang et al., 2011). Herein,  
14 additional environmental geochemistry factors also need careful consideration. Phosphorus-  
15 mitigation wetlands are temporarily inundated floodplains that are re-established by re-meandering  
16 the old channelized watercourse and lowering the discharge capacity. A technical guidance requires  
17 project areas to be screened for their content of iron and P before the floodplain areas are allowed to  
18 be inundated due to the risk of releases of P from former fields having a high build-up of Fe-  
19 associated P in the soils following Fe dissolution under saturated conditions (Hoffmann et al., 2009;  
20 Kronvang et al., 2011). Utilization of restored floodplains and associated wetlands as sinks for  
21 sediment P is also strongly in line with the need for development of novel technologies to capture  
22 and recycle P back to the circular economy to overcome P shortage in a world where this resource is

1 finite and to combat current dramatic losses in ecosystem biodiversity (Vaccari and Strigul, 2011;  
2 Elser and Bennett 2011; Steffen et al., 2015).

3

## 4 **5. Synthesis and significance**

5 A large range of functions and processes occur in naturally functioning floodplains that can affect  
6 hydrology, water quality and biodiversity (Maltby et al., 1996). Generally these functions and  
7 processes involve and rely on the import, transformation, export and/or storage of sediment,  
8 sediment-associated chemicals and solutes during inundations of the floodplain with river water  
9 (Malmon et al., 2002). Understanding sedimentation dynamics is essential to plan and evaluate the  
10 success of floodplain and wetland restoration projects.

11 Attempts to assemble detailed information on contemporary rates of overbank sediment deposition  
12 on floodplains have to date faced many uncertainties and difficulties related to the inherent spatial  
13 and temporal variability of such sedimentation and to the operational problems of studying an  
14 inundated floodplain. Equally, because the depths of accretion involved are likely to be small, for  
15 instance typically of the order of < 10 mm, reliable *in situ* measurements are difficult to obtain  
16 (Walling 1999). Furthermore, the amounts of deposition associated with individual events will vary  
17 according to their magnitude and duration and other characteristics, including the suspended  
18 sediment concentration in the river which vary considerably both spatially and temporally (Walling  
19 2000).

20 In this study, the labour-intensive collection of deposited material in conventional, direct  
21 measurement sediment traps permitted validation of a new tracer tool that overcomes the above  
22 critical limitations. Firstly, eight years of *in situ* measured sediment accretion rates on a floodplain  
23 along the restored River Odense, Denmark was used as a mean of testing established use of  $^{137}\text{Cs}$  as

1 a tracer for longer term sediment accumulation rates on restored floodplain wetlands. Secondly, in  
2 situ measured sediment accretion rates during one winter period could be reliably mapped using a  
3 new application of  $^7\text{Be}$  as a sediment tracer. Fallout radionuclides have been shown to be a valuable  
4 tool for assessing longer term (decadal) rates and patterns of accretion on rewetted land and the  
5 innovative  $^7\text{Be}$  methodology here offers complementary evidence on the short term i.e. event to wet  
6 season timescale. Critical methodological considerations emerged within this study. In particular,  
7 the need for careful application of particle size correction procedures using deposited material at  
8 each sampling point that is representative of the study period.

9 Our results from in situ measurements of sediment P storage on restored floodplains and associated  
10 wetlands clearly demonstrate their use as an innovative technological tool to trap P from where it  
11 may be later recovered and re-introduced into the P bio-cycle (Elser and Bennett, 2011; Steffen et  
12 al., 2015).

13 . With growing global interest in Payments for Ecosystem Services systems linked to nature-based  
14 solutions and river basin restoration programmes (Vlachopoulou et al., 2014), reliable  
15 methodologies to assess sedimentation rates and nutrient retention are required. The use of fallout  
16 radionuclides to assess sedimentation patterns on floodplains permits the benefits of wetland  
17 restoration programmes to be assessed. This in turn makes a valuable contribution to River Basin  
18 Management Plans designed to combat excess P loading to lakes and estuaries as required by  
19 environmental legislation across the world.

20 Floodplain wetland restoration is demonstrated to be an important means of mitigating excess  
21 nutrient flux in agricultural and urban catchment systems. The tracer-based approach demonstrated  
22 and tested here offers a powerful new decision support tool for floodplain and wetland restoration  
23 planning. Floodplain rivers are a key strategic global resource in terms ecosystem service provision

(Tochner and Stanford, 2002) and decision support tools to support their conservation will become increasingly important in the 21<sup>st</sup> century with growing anthropogenic pressures on aquatic ecosystems (Albert et al., 2021).

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