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2 **Active overbank deposition during the last century, South River, Virginia**

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Abstract

We quantify rates of overbank deposition over decadal to centennial timescales along the South River in Virginia using four independent methods. Detailed mercury profiles sampled adjacent to the stream channel preserve the peak historic mercury concentration on suspended sediment dating from 1955 to 1961 and suggest sedimentation rates of 8 to 50 cm/100 years. Sediment accumulation over the roots of trees suggest rates of 0 to 100 cm/100 years, with significantly higher values on levees and lower values on floodplains farther from the channel. Profiles of ^{137}Cs and ^{210}Pb from two eroding streambanks are fit with an advection-diffusion model calibrated at an upland reference site; these methods suggest sedimentation rates of 44 to 73 cm/100 years. Mercury inventories from 107 floodplain cores, combined with a previously published reconstruction of the history of mercury concentration on suspended sediment, provide spatially comprehensive estimates of floodplain sedimentation: median sedimentation rates are 3.8 cm/100 years for the <0.3-year floodplain, 1.37 cm/100 years for the 0.3- to 2-year floodplain, 0.4 cm/100 years for the 2- to 5-year floodplain, and 0.1 cm/100 years for the 5- to 62-year floodplain. While these sedimentation rates are relatively low, the total mass of sediment stored from 1930 to 2007 is 4.9 ± 1.7 (95% confidence interval) $\times 10^7$ kg, corresponding to an average thickness of 2.5 cm (3.2 cm/100 years). These results demonstrate that floodplains of our 4.5-km-long study reach have stored 8 to 12% of the total suspended sediment supplied to the study reach of the South River. Hydrologic Engineering Center-River Analysis System (HEC-RAS) modeling demonstrates that the floodplain of the South River remains hydraulically connected to the channel: 56% of the 100-year floodplain is inundated every two years, and 83% of the floodplain is inundated every

43 five years. These results, combined with previously published data, provide the basis for
44 a regional synthesis of floodplain deposition rates since European settlement. Floodplain
45 sedimentation rates were high following European settlement, with published estimates
46 ranging from 50 to 200 cm/100 years. Sedimentation rates decreased by 1 to 2 orders of
47 magnitude during the twentieth and twenty-first centuries; but despite these lower
48 sedimentation rates, floodplains continue to store a significant fraction of total suspended
49 sediment load. Many floodplains of the mid-Atlantic region are active landforms fully
50 connected to the rivers that flow within them and should not be considered terraces
51 isolated from contemporary fluvial processes by post-settlement aggradation.

52 *Keywords:* floodplain sedimentation; dendrochronology; fallout radionuclides; floods;
53 mercury contamination; alluviation

54

1. Introduction

Deforestation, poor agricultural practices (Happ et al., 1940; Trimble, 1969), and the construction of mill dams (Walter and Merritts, 2008) resulted in widespread valley alluviation following European settlement of the eastern and midwestern United States (Phillips, 1991). Documented by sediment budgets (Costa, 1975; Trimble, 1983; Jackson et al., 2005), stratigraphic studies (Jacobson and Coleman, 1986; Knox, 2006), geomorphic mapping (Happ et al., 1940), and accumulation of metals derived from mining (Bain and Brush, 2005; Lecce et al., 2008), post-settlement floodplain sedimentation has been described as ‘the most important geomorphic process in terms of the erosion and deposition of sediment that is currently shaping the landscape of Earth’ (Wilkinson and McElroy, 2007).

Many geomorphologists have suggested that post-settlement alluviation waned by the early twentieth century (Costa, 1975; Jacobson and Coleman, 1986), creating post-settlement *legacy* alluvial terraces (Walter and Merritts, 2008). While this hypothesis has been verified by detailed studies in the Driftless Area of Wisconsin (Knox, 2006), fewer measurements of overbank deposition rates have been available for upland watersheds of the eastern U.S. Some of the available data are restricted to small suburban watersheds (Bain and Brush, 2005; Allmendinger et al., 2007), while other studies include only indirect inferences of floodplain accretion (Jacobson and Coleman, 1986; Jackson et al., 2005; Walter and Merritts, 2008; Smith and Wilcock, 2015). Recent studies in Virginia and Maryland have greatly expanded our knowledge of mid-Atlantic floodplain sedimentation rates (Schenk and Hupp, 2009; Hupp et al., 2013; Schenk et al., 2013), but

these studies rely on direct observations of floodplain accumulation over only a few years, results that may not provide an accurate assessment of longer-term trends.

Documenting recent sedimentation rates on floodplains is important to advance scientific knowledge and to improve strategies for watershed management. Fine-grained sediment is often considered to move rapidly through watersheds, a consequence of the wash load concept of sediment transport, which posits that suspended silt and clay are carried downstream without deposition regardless of the hydraulics of flow, implying that wash load sediment travels at the same velocity downstream as the river's current. Recent studies in the mid-Atlantic region, however, suggest that suspended sediment may be stored on floodplains after being transported only 10 km (Pizzuto, 2014). Furthermore, once deposited on floodplains, fine-grained sediment is likely to remain in storage for hundreds or even thousands of years (Pizzuto et al., 2014). Under these conditions, the *virtual* velocity of sediment movement (Martin and Church, 2004) (i.e., the velocity that includes time spent in storage) may be up to 5 orders of magnitudes slower than the rate of water flow, implying that delivery timescales of sediment supplied from upland sources to estuarine depocenters may approach thousands of years (Pizzuto et al., 2014). Watershed restoration schemes to reduce loading of sediment and particle-borne contaminants (e.g., phosphorus) to estuaries such as the Chesapeake Bay (Shenk and Linker, 2013) cannot be properly evaluated or implemented if these processes are not accounted for.

In this study, we quantify floodplain sedimentation rates for a stream located in a predominantly agricultural setting in the Chesapeake Bay watershed with a drainage basin area of 385 km². We use dendrochronology, analysis of the fallout radionuclides

^{210}Pb and ^{137}Cs , and the known history of mercury contamination to determine floodplain sedimentation rates since the early 1900s. These methods quantify sedimentation rates on spatial scales that include the entire floodplain, allowing us to assess the importance of floodplain sedimentation for the sediment budget of the South River. Our results indicate that floodplain sedimentation has been an active process in our study area throughout the twentieth century, storing 8 to 12% of the annual suspended sediment load supplied to the upstream boundary of the study area.

2. Study area

The study area is the South River riparian corridor a few kilometers downstream of Waynesboro, Virginia (Fig. 1). The South River lies in the Blue Ridge and in the Valley and Ridge physiographic provinces. Bedrock consists of Paleozoic folded and faulted clastic carbonate sedimentary rocks characteristic of the Valley and Ridge, and the Paleozoic metamorphic rocks of the Blue Ridge (Bingham, 1991). The study reach is a fifth-order, gravel-bed, bedrock-influenced river with frequent pools and riffles and a well-developed, cohesive floodplain composed primarily of sand, silt, and clay. The channel is single-thread, but sinuous rather than meandering, with an average sinuosity of 1.4 (Narinesingh, 2010). The mean slope of the channel bed in the study area is 0.0013, while the bankfull depth and width of the channel are 1.5 and 40 m, respectively (Skalak and Pizzuto, 2010). The riparian zone consists of a mixture of forested, pasture, and agricultural land uses.

Fig. 1. Location map showing sampling sites along the South River (lower image) and an *upland site* used to document organic matter accumulation and processes affecting fallout radionuclides in the absence of sedimentation (upper image). *Hg data* are sites of detailed mercury profiles, *Dendro sedimentation analyses* indicates the location of dendrochronological estimates of overbank sedimentation rates, and *FRN data* refers to sites where fallout radionuclides ^{137}Cs and ^{210}Pb were analyzed. A meander cutoff created in 1974 is also indicated.

The U.S. Geological Survey (USGS) maintains stream gauging stations near the study area at Waynesboro (drainage basin area (DA) is 330 km^2) and at Doods (DA - 385 km^2 ; Fig. 1). The mean annual discharge at Waynesboro is $4.2 \text{ m}^3/\text{s}$, and the mean annual flood is $117 \text{ m}^3/\text{s}$ (period of record 1952 to 2011). At Doods, the mean annual discharge is $5.9 \text{ m}^3/\text{s}$, and the mean annual flood is $157 \text{ m}^3/\text{s}$ (period of record 1974 to 2011).

Direct observations of suspended sediment are insufficient to document the river's suspended sediment load. Eggleston (2009) suggested that the annual suspended sediment load is $5.1 \times 10^3 \text{ Mg}$ based on results from a calibrated hydrologic model. Skalak (2009) developed a regional sediment rating curve and used it to estimate the annual suspended load of the South River at $7.7 \times 10^3 \text{ Mg}$ based on 51 years of discharge data from the Waynesboro gauging station.

Mercury contamination of the South River was discovered in 1976 (Carter, 1977), and a manufacturing plant in Waynesboro has been implicated as the ultimate source of the mercury to the river (Stahl et al., 2014). Many detailed studies have been completed to guide remediation of this environmental problem. These studies document the geochemistry and fluvial transport of mercury in the river (and close associations between mercury and suspended sediment in the water column; Eggleston, 2009; Flanders et al., 2010), the ongoing supply of mercury to the river through erosion of mercury-contaminated floodplain deposits exposed in the river's banks (Rhoades et al.,

2009; O’Neal and Pizzuto, 2010), and the close connection between the accumulation of mercury on the floodplain and floodplain sedimentation (Pizzuto, 2012). In particular, previous studies (described in detail by Pizzuto, 2014) documented the geochemistry of mercury in floodplain soils and the relative immobility of sediment-associated mercury in floodplain deposits, minimizing the potential for post-depositional migration of mercury in floodplains of the South River.

Pizzuto (2014) demonstrates that mercury associated with suspended sediment only traveled an average distance of about 10 km downstream of the plant in Waynesboro before being stored on floodplains. Skalak and Pizzuto (2010, 2014) reconstruct the history of mercury concentration on suspended sediment of the South River from 1930 to the present (Fig. 2) from dated core samples and modeling. Their reconstructed history suggests that mercury concentrations increased rapidly after 1930, reaching maximum values of around 900 mg/kg between 1955 and 1961 and decreasing gradually thereafter to values of ~10 mg/kg obtained from contemporary monitoring (Eggleston, 2009; Flanders et al., 2010).

Fig. 2. Reconstruction of mercury contamination history of the South River in the study area (after Skalak and Pizzuto, 2014). The width of the gray line indicates the uncertainty in the reconstructed mercury concentrations.

Pizzuto and O'Neal (2009) identified eight colonial mill dams in a 30-km reach of the South River between Waynesboro and Grottoes, VA, that were in place in 1937. One of these, the ~1-m high run-of-river Dooks Dam, is located at the downstream end of the reach studied here (Fig. 1) and was breached in 1976. Hydraulic modeling results presented by Pizzuto and O'Neal (2009) indicated that the backwater effects from the 5-year flood extended no farther upstream than RRkm 5 before 1976 (locations along the South River are referenced according to the downstream distance (km) measured from a footbridge across the South River that connects a parking lot and the Invista nylon manufacturing plant in Waynesboro). The potential influence of Dooks Dam on the sedimentation rates presented in this paper is evaluated in the Discussion.

3. Methods

178 *3.1. Quantifying floodplain deposition rates using mercury profiles*

179 Profiles of metal concentrations in floodplain deposits can often be related to the
180 known history of the release of metals into the environment, thereby providing
181 chronostratigraphic markers that can be used to determine the history of floodplain
182 sedimentation and rates of sediment accumulation (Marron, 1992; Hudson-Edwards et
183 al., 1999; Middlekoop, 2002; Bain and Brush, 2005; Lecce et al., 2008). In our study
184 area, mercury concentrations were very low before the early 1940s and peaked
185 dramatically from 1955 to 1961 (Fig. 2). Because mercury is tightly bound to particles
186 along the South River (Flanders et al., 2010) and also because rates of floodplain
187 reworking are very low (Rhoades et al., 2009; Pizzuto, 2012, 2014), we hypothesized that
188 the historical record of mercury concentrations illustrated in Fig. 2 should be well
189 preserved in overbank deposits of the South River, providing a useful means to quantify
190 rates of floodplain rates.

191 Three floodplain sites adjacent to the river were sampled in August 2005 (Fig. 1).
192 Historical aerial photographs indicate that these sites have not been reworked by lateral
193 migration of the channel since 1937. One of the sites, located at RRkm 2.86, was located
194 more than 500 m from the channel before an artificial cutoff channel was created in 1974
195 to reduce flooding in Waynesboro upstream.

196 At each site, a trench was dug into the floodplain deposits. Samples were obtained
197 at vertical intervals of a few centimeters to depths of 130 cm (RRkm 2.86), 180 cm
198 (RRkm 3.51), and 90 cm (RRkm 4.18). For each sample, organic carbon, grain size, and
199 the concentration of inorganic mercury (THg) were measured. Organic matter context
200 was approximated by analyzing on loss-on-ignition using American Society for Testing

and Materials (ASTM) method #D2974-07 (ASTM, v 04.08, 2007), while grain size was analyzed following ASTM method D422-63 (ASTM, v. 0408, 2007). Inorganic mercury concentrations measurements were performed by Studio Geochemica, Inc. Samples were digested with aqua regia and diluted with ultrapure reagent water. Mercury was quantified using SnCl_2 reduction and purge and trap dual amalgamation cold vapor atomic fluorescence.

3.2. Dendrochronology

Hupp and Bazemore (1993) and Allmendinger et al. (2007) described a dendrochronological approach for estimating floodplain sedimentation rates. The thickness of sediment overlying a tree's basal roots is measured an arbitrarily selected standard distance away from the trunk (here taken as two times the diameter of the tree's trunk measured at breast height). The age of the tree is determined by counting tree rings. The average sedimentation rate is then determined as the thickness of sediment deposited over the lifetime of the tree.

Extensive forests are rare in the study area, but a suitable site was located (Fig. 1). Historical aerial imagery indicates that this area was not forested in 1951, while trees were present in 1976. Field measurements were made in the winter of 2006, so we expected to measure sedimentation rates averaged over maximum timescales of several decades.

We surveyed a topographic transect perpendicular to the channel using a stadia rod and automatic level and selected a total of 42 trees at varying distances along this transect. We identified the species of each tree and recorded their GPS location. The diameter at breast height (DBH) and thickness of sediment over the tree's basal roots

224 were measured. Twenty-four trees were cored with an increment borer to determine their
225 age. Tree cores were sanded, polished, and stained, and the number of rings counted by
226 two investigators.

227 To account for basal tree root burial by organic material and other detritus not
228 related to river sedimentation, we measured deposition rates at a control area where river
229 sedimentation was not expected. We selected a site along U.S. 340 between Dooms and
230 Crimora, VA, several kilometers away from the river channel (Fig. 1). Historical aerial
231 imagery demonstrates that this area has not been anthropogenically disturbed since 1937.
232 We identified 25 trees at this site and performed the same measurements as described
233 above.

234 We used simple statistical tests to compare our measurements of the basal
235 sediment accumulation thickness at the floodplain and reference sites. First, data from the
236 floodplain site was divided into two categories: levees sites within 20 m of the river
237 channel and floodplain sites located 20 to 100 m from the river. We computed descriptive
238 statistics for basal accumulation in each of the three groups of data (i.e., levee, floodplain,
239 and reference sites). We then used the nonparametric Mann-Whitney test to determine if
240 the median accumulation at the levee and floodplain sites was significantly different from
241 the median accumulation at the reference site. To estimate sedimentation rates, the
242 average basal accumulation from the reference site was subtracted from basal
243 accumulation values at the levee and floodplain sites.

244 3.3. *Fallout radionuclides*

245 The fallout radionuclides ^{137}Cs and ^{210}Pb can be used to estimate sedimentation
246 rates on floodplains (He and Walling, 1996a, 1996b; Goodbred and Kuehl, 1998; Aalto

and Nitttrouer, 2012; Du and Walling, 2012). Cesium-137 was produced by atmospheric nuclear weapons testing; ^{137}Cs activities in the atmosphere were negligible before about 1955, increased to a maximum around 1963, and rapidly decreased thereafter (He and Walling, 1996a). Lead-210 in the atmosphere is derived from radioactive decay of the gas ^{222}Rn , which is produced from the decay of ^{226}Ra in soils. Some of the ^{222}Rn escapes from soils into the atmosphere, subsequently decays to ^{210}Pb , which is then returned to the Earth's surface, primarily through wet deposition (Appleby and Oldfield, 1992). Cesium-137 and ^{210}Pb adhere strongly to sediment particles and therefore provide useful tracers for sediment transport, erosion, and deposition (Matisoff, 2014).

We selected two sites along the South River to estimate sedimentation rates using ^{137}Cs and ^{210}Pb (Fig. 1). Both sites are located on the outside banks of migrating bends. Bank migration rates at these sites have averaged <2 cm/y since 1937, based on studies of historical aerial photographs (Rhoades et al., 2009), so we can be certain that the geomorphic setting of these sites has not changed since the early twentieth century. Both sites have a narrow forested riparian zone immediately adjacent to the channel, with pasture beyond.

Sediment samples were obtained in April and July 2014. Before sampling at each site, a trench at least 50 cm deep was cut into each bank. Samples were obtained at intervals of 2 to 3 cm over the upper 30 cm. Below this depth, samples were obtained at 5-cm intervals to a maximum depth of 65 cm. To measure the bulk density, 3-cm lengths of 3-cm diameter pipe were pounded horizontally into the soil. These short cores were excavated without any loss of material and capped at both ends. Samples were weighed, dried to evaporate moisture, and reweighed to calculate bulk density.

The vertical distribution of ^{137}Cs and ^{210}Pb is influenced by a variety of processes other than sedimentation (discussed in detail below). To assess these processes, samples were also obtained in June 2014 from a single location at the reference site along SR 340 where only organic matter is accumulated over tree roots since 1937 (Fig. 1). Samples were obtained at 2-cm intervals to a depth of 32 cm from a soil pit. Bulk density samples were obtained using methods described above.

In the laboratory, we determined the grain size distribution, activities of relevant radionuclides (described further below), and bulk density. Clay ($<4\ \mu\text{m}$), silt ($4\text{--}64\ \mu\text{m}$), and sand ($>64\ \mu\text{m}$) concentrations were calculated using a Beckman Coulter LS 13 320 Laser Diffraction Particle Size Analyzer. Organic content analyses (loss-on-ignition) were available from previous studies at the two floodplain sites using methods described above.

3.3.1. Gamma spectroscopy to measure radionuclide activities

In preparation for gamma spectroscopy, samples were dried overnight at $60\ ^\circ\text{C}$. The few particles larger than 2 mm were removed by hand, and the remaining material was ground using a mortar and pestle. From each floodplain sample, 70 g was selected for analysis. Samples from the upper 10 cm of the reference site consisted of 50 to 70 g, while all the deeper samples consisted of 70 g. Samples were placed into plastic jars with lids that were sealed with electrical tape. A minimum of 21 days elapsed before analysis to enable the daughters of ^{222}Rn (^{214}Bi and ^{214}Pb) to achieve secular equilibrium, allowing the *supported* ^{210}Pb in each sample to be determined (Joshi, 1987; Schelske et al., 1994).

Gamma ray emission rates were measured for 48 hours on two high efficiency Canberra low energy Germanium detectors (model # GL2020R) run by Canberra's

GENIE2000 software. Efficiencies for ^{137}Cs were determined using a sealed standard with activities certified by Eckart and Ziegler, Inc. (this standard also has a certified activity for ^{210}Pb that was used to assess sample absorption, as noted below). For U series radionuclides, efficiencies were determined by making standards composed of material from the study sites. A single standard was created for the floodplain sites, and two standards were created for the stable site: one of the upper 4 cm (the O and A horizons) of this soil and another for samples from 4 to 32 cm (A, B, and C horizons). To create the standard, samples were oven-dried at 60 °C overnight and placed in jars without lids. Samples were counted for 48 hours to determine background activities. Then, ~0.2 g of the uranium ore BL-5 (<https://www.nrcan.gc.ca/mining-materials/certified-reference-materials/certificate-price-list/8115>, accessed 28 May 2015) was added to each standard. Because the U series radionuclides of BL-5 are in secular equilibrium, activities of the standard can be computed for ^{226}Ra , ^{234}Th , ^{214}Bi , ^{214}Pb , and ^{210}Pb , allowing efficiencies to be determined for all these isotopes (Murray et al., 1987). The standards had activities of ~2 to 3 Bq/g. For the reference site, standards were made of masses of 50, 60, and 70 g, and efficiency calibration curves as a function of mass fit to the results. These were used to determine efficiencies for samples of varying mass at this site. The efficiencies determined using these methods are presented as Table 1 of the supplementary material.

The Eckart and Ziegler standard was used to assess the ^{210}Pb absorption coefficient for each sample using the method of Cutshall et al. (1983). For each sampling interval encompassed by the constructed standards, the measured absorption was essentially a constant. Because the efficiencies determined from the standards implicitly

include the effects of absorption, ^{210}Pb activities could be computed without using the absorption coefficients determined using the Cutshall et al. (1983) method.

3.3.2. Extracting sedimentation rates from fallout radionuclide data

To extract sedimentation rates from fallout radionuclide measurements, data must be fitted to a model that accounts for the relevant processes that control the measured activities. The most commonly used models are derived from studies of lakes, where certain simplifying assumptions are frequently justifiable. Two of the most commonly used models assume that the activity at the top of the accumulating sediment column is constant (i.e., the constant initial concentration (CIC) model) or that the activity flux to the accumulating column is constant through time (i.e., the constant rate of supply (CRS) model; Appleby and Oldfield, 1992). Floodplains, however, are subject to approximately constant atmospheric deposition, while sedimentation, which typically supplies additional activity along with the sediment, is highly episodic. Accepting either of these models as being generally applicable to floodplains is difficult. He and Walling (1996a, b) and Du and Walling (2012) proposed models based on the inventory of ^{137}Cs and ^{210}Pb for quantifying sedimentation rates on floodplains. These, however, require accounting for the activity directly supplied from atmospheric deposition to the floodplain surface, in addition to the activity that is supplied by deposited sediment. If little or no activity results from sedimentation, or if the atmospheric deposition rate is poorly constrained, then these methods cannot be used. Aalto et al. (2008) and Aalto and Nitttrouer (2012) used precise alpha spectrometry and a variety of supporting measurements (and assumptions) to identify and date individual depositional events along large tropical rivers; under these conditions, fitting data to a model is not required. However, this

approach can only be used where individual depositional events can be clearly distinguished, which is unlikely where sedimentation rates are low and bioturbation processes are significant.

Because of the limitations of existing methods, we developed a numerical model to describe the evolution of ^{210}Pb and ^{137}Cs activity at our study sites. The model accounts for changes in fallout radionuclide (FRN) activity related to atmospheric deposition, bioturbation within the soil column, radioactive decay, and sedimentation. We determine most of the parameters of the model using data from the reference site (where sedimentation does not occur), and then determine sedimentation rates at the two floodplain sites by fitting the model to measured activity profiles in these settings. This approach follows a tradition of using models to determine the effects of bioturbation, sedimentation rates, and other processes on radionuclide profiles in sediments (Officer, 1982; Robbins, 1986; Nie et al., 2001).

Changes in activity, A (units of Bq/kg) with time, t (units of years), are governed by an advection/diffusion equation:

$$\frac{\partial A}{\partial t} = -V \frac{\partial A}{\partial \xi} + B \frac{\partial^2 A}{\partial \xi^2} - kA \quad (1)$$

where ξ is a coordinate of cumulative sediment mass (kg/cm^2), V (units of $\text{kg}/\text{cm}^2/\text{y}$) is a downward advection velocity that affects ^{137}Cs but not ^{210}Pb (He and Walling, 1996b; Du and Walling, 2012), B is a spatially uniform bioturbation coefficient ($\text{kg}^2/\text{cm}^4/\text{y}$), and k is the decay constant (0.03114 y^{-1} for ^{210}Pb and 0.0230 y^{-1} for ^{137}Cs). The model is formulated in units of cumulative mass rather than depth to avoid complications related to varying soil bulk density.

Initial and boundary conditions are needed to solve Eq. (1). The initial conditions specify that $A = 0$ at $t = 0$ at all values of ξ . At depth, where $\xi = \infty$, $A = 0$ for all t . At the soil surface, the boundary condition specifies that activity supplied to the ground surface through atmospheric deposition, P_0 (Bq/cm²/year) is continually transported into the soil by advection (for ¹³⁷Cs only) and bioturbation:

$$P_0 = VA_{\xi=0} - B \frac{\partial A}{\partial \xi}_{\xi=0} \quad (2)$$

For ²¹⁰Pb, P_0 is a constant, while for ¹³⁷Cs, P_0 varies with time (the specific form of $P_0(t)$ for ¹³⁷Cs is presented in the results section).

Equation (1) is solved on a uniform computational grid using an explicit finite-difference algorithm. The diffusion term is discretized using central differences, while the advection term is discretized using an upwinding scheme (Slingerland and Kump, 2011). The accuracy of the computational grid is verified using analytical solutions to Eqs (1) and (2). For ²¹⁰Pb, the computations are run until a steady state profile is achieved, typically about 8 half-lives (e.g., ~180 years). For ¹³⁷Cs, computations begin in 1950 (as no ¹³⁷Cs was supplied before atmospheric nuclear weapons testing) and end at the time of the field sampling.

To account for sedimentation at the floodplain sites, a constant mass of material $\Delta \xi$ is added to the ground surface at a specified time interval Δt , representing discrete, episodic sedimentation events with a time-averaged sedimentation rate, w , of $\Delta \xi / \Delta t$. The newly deposited sediment has a constant activity of A_0 (presumably from a mixture of diverse sources upstream of the site of deposition that could include eroding banks, soil surfaces, etc.). The mass sedimentation rate, w , can be converted to a linear sedimentation, w_L , by dividing by the bulk density of the surface sediment, ρ_{b0} :

$$w_L = \frac{w}{\rho_{b0}} \quad (3)$$

The model parameters were determined by fitting the model computations to the observed radionuclide activity profiles from the reference and floodplain sites. Model errors were assessed quantitatively using the root-mean-square (rms) where appropriate. The rms error is defined by:

$$rms\ error = \left(\frac{1}{n} \sum_{i=1}^n (x_{mi} - x_{oi})^2 \right)^{1/2} \quad (4)$$

where n is the number of observations, x represents a variable of interest (such as radionuclide activities predicted by our model), and the subscripts m and o indicate predicted and observed values, respectively. Visual error estimates of model fit were also used where quantitative measures lead to unreasonable or undesirable results (described in detail below). Values of P_0 for ^{210}Pb were obtained from the inventory at the reference site. The parameter B was constrained by fitting the model to the ^{210}Pb data at the reference site (which only depends on B once P_0 is known), while the value of V was constrained by fitting the model to the ^{137}Cs data at the reference site (which only depends on V once B and P_0 are known). Values of A_0 , $\Delta\xi$, and Δt were then determined for both floodplain sites by fitting the model to measured ^{210}Pb and ^{137}Cs profiles.

3.4. Floodplain mercury inventories

The methods described above are applied to relatively few samples, and results must be extrapolated over large areas to draw any conclusions regarding the importance of floodplain sedimentation for the sediment budget of the South River. Fortunately, many cores have been obtained in the study area to document the extent of mercury contamination in floodplain deposits. In 2008, five cutbanks and five deposits adjacent to

the channel were sampled by the authors. At each location, the uppermost 15 cm (0.5 ft) was analyzed, while deeper deposits were analyzed at intervals of 30 cm (1 ft). The total thickness sampled varied from 1.2 to 2.4 m. Another 17 cores were obtained by the Environmental Protection Agency (Bruzzezi et al., 2007), all were 0.75 m (2.5 ft) deep and sampled at 15-cm (0.5 ft) intervals. Another 80 floodplain cores, 0.75 m (2.5 ft) in depth, were obtained by URS Corp. in 2008. Only two intervals of these cores were analyzed for mercury content: the uppermost 15 cm (0.5 ft) and the remaining 60 cm.

Analyses were performed by two commercial laboratories. Samples obtained by the authors were sent to Lancaster Laboratories, who measured mercury concentrations using SW-846 method 7471A of the U.S. Environmental Protection Agency. Samples obtained under the auspices of the Environmental Protection Agency (Bruzzezi et al., 2007) were sent to Accutest, a U.S. Army Corps of Engineers Missouri River Division and National Environmental laboratory Accreditation Commission validated laboratory. Accutest also measured the total mercury concentration of soils using EPA SW-846 method 7471A. The percentages of sand, silt, and clay and organic carbon (loss-on-ignition) were also measured using the methods described above.

While the poor vertical resolution of these samples does not allow the timing of peak mercury contamination to be identified in the cores, the total amount of mercury in each core (the Hg *inventory*) can be used to estimate the amount of sediment that has accumulated since 1930 (when mercury was first introduced into the South River) using a method first described by Swanson et al. (2008). For each core, we first compute the inventory of mercury, I_{Hg} , by multiplying the mercury concentration in each soil interval (C_{soilHg_i}) by the bulk density ρ_b and sampling interval, and sum the results over all n

sampled intervals. The I_{Hg} has units of kg/m^2 ; it represents the total mass of mercury deposited during overbank flows since 1930 per unit surface area of floodplain. To obtain the corresponding mass of sediment, M_s , per unit area of floodplain deposited during the same time period, the mercury inventory is divided by the concentration of mercury on suspended sediment in the water column C_{Hgs} (units of $\text{kg Hg}/\text{kg sediment}$):

$$M_s = I_{Hg} \left(\frac{1}{C_{Hgs}} \right) = \frac{\sum_{i=1}^n \rho_b C_{soilHg_i} T_i}{C_{Hgs}} \quad (5)$$

Once the mass of sediment is known, its thickness T_t can be obtained by dividing by ρ_b :

$$T_t = \frac{M_s}{\rho_b} = \frac{\sum_{i=1}^n \rho_b C_{soilHg_i} T_i}{C_{Hgs} \rho_b} = \frac{\sum_{i=1}^n C_{soilHg_i} T_i}{C_{Hgs}} \quad (6)$$

The terms to the right in Eq. (6) illustrate the computation, where Eq. (5) is substituted for M_s and the bulk density is assumed to be constant (so it actually cancels from Eq. 6).

To apply Eq. (5), C_{Hgs} must be specified in addition to I_{Hg} . We rely on the reconstructed history of mercury concentration on suspended particles provided by Skalak and Pizzuto (2010, 2014; Fig. 2), and estimate C_{Hgs} by averaging the data of Fig. 2, which yields a value of C_{Hgs} of 239 ± 5 (95% confidence interval) mg/kg .

The method summarized by Eqs. (5) and (6) requires mercury to be approximately conservative, spatially invariant within the study area, and unaffected by differences in grain size (and other factors that could effectively partition mercury). We have discussed these assumptions in detail in previous publications (Pizzuto, 2012, 2014; Skalak and Pizzuto, 2010, 2014).

3.5. One-dimensional modeling of flood inundation and the total mass of floodplain deposition

As part of the effort to document and remediate floodplain mercury contamination, contemporary floodplain inundation patterns were evaluated with Hydrologic Engineering Center-River Analysis System (HEC-RAS) modeling by URS, Inc. Topographic data for HEC-RAS modeling was provided by a 0.6-m contour interval topographic map developed from aerial LiDAR data (the resolution of the commercially acquired LiDAR is unknown), supplemented by surveyed cross sections. Spatially averaged roughness coefficients (one each for the floodplain and channel) were determined by calibrating the model to rating curves from the USGS gaging stations and also from temporary gaging stations established at bridge crossings (Flanders et al., 2010). To report the results of the HEC-RAS modeling, URS Inc. selected flows with return periods 0.3, 2, 5, and 62 years. Results of the modeling are available from the South River Science Team (www.southeriverscienceteam.org; accessed 1 June 2015).

The hydraulic modeling determines the floodplain areas inundated with different return periods, and the mercury inventory analysis determines the amount of sediment deposited. Taken together, these results provide the basis for estimating the total mass of sediment deposited on the floodplain since 1930. We first averaged the sediment inventories from Eq. (1) for each of the 0.3-, 2-, 5-, and 62-year floodplains. The mass deposited on each floodplain was obtained by multiplying the average sediment inventory by floodplain area, and the individual results were added. Errors (represented by 95% confidence intervals) were propagated through all calculations.

4. Results

4.1. Mercury profiles

Detailed mercury concentration profiles for RRkm 2.86, 3.51, and 4.18 all have a significant peak concentration below the sediment surface (Fig. 3). Peak concentrations are about 10 mg/kg at RRkm 2.86, 60 mg/kg at RRkm 3.51, and 90 mg/kg at RRkm 4.18. The lower mercury content at RRkm 2.86 compared with the other two sites may be explained by the observation that this site was located more than 500 m from the river before the cutoff channel was constructed in 1971, while the other two sites have been adjacent to the river channel since at least 1937 (Rhoades et al., 2009). Mercury concentration profiles at RRkm 2.86 and 3.51 are not systematically related to variations in silt-clay content or organic matter content (loss-on-ignition), suggesting that these profiles preserve the history of mercury concentration on sediment carried overbank by flooding (as summarized in Fig. 2). If the peak concentrations in these profiles are assigned to 1955 to 1961 based on the mercury concentration history of Skalak and Pizzuto (2010, 2014), linear sedimentation rates of 0.08-0.2 cm/y (RRkm 2.86), 0.3-0.5 cm/y (RRkm 3.51), and 0.2–0.3 cm/y (RRkm 4.18) are obtained (Table 1).

Table 1
Depth intervals for the peak mercury concentration at RRkm 2.86, 3.51, and 4.18, and linear sedimentation rates based on dating the peak contamination from 1955-1961 (Skalak and Pizzuto, 2010, 2014); samples were obtained in 2005.

Site	Depth interval of peak Hg concentration (cm)	Linear sedimentation rate (cm/year)
RRkm 2.86	5-7	0.08-0.2
RRkm 3.51	17-22.5	0.3-0.5
RRkm 4.18	9-15	0.2-0.3

Fig. 3. Detailed mercury profiles for sites located at RRkm 2.86, 3.51, and 4.18. Organic matter (loss-on-ignition) and % silt-clay are presented for RRkm 2.86 and 3.51; these analyses were not done at RRkm 4.18.

4.2. Dendrochronology

At the upland reference site, 25 trees had mean and median accumulation over basal roots of 2.3 and 1 cm, respectively (supplementary material, Table 2). Eleven trees (44%) had no measureable accumulation. The maximum accumulation was 10 cm.

The forest along the river consisted predominately of *Juglans nigra* (black walnut), with a few *Acer saccharinum* (silver maple), *Platanus occidentalis* (American sycamore), and six trees we could not identify (supplementary material, Table 2). Along the levee, the mean and median thickness of accumulation over the tree roots was 19.3

and 17 cm, respectively (Table 2). The minimum accumulation was 7 cm, and the maximum was 37 cm (supplementary material, Table 2). On the floodplain (>20 m from the channel), the mean and median thickness of accumulation was 3.7 and 3 cm. Thirteen trees had no accumulation (45%), and the maximum accumulation was 26 cm. According to the Mann-Whitney test, the median accumulation on the levee is significantly different from that measured at the upland reference site (the probability of these samples being drawn from the same population is <0.001), while that on the floodplain is not significantly different from the upland reference site (significance of 0.513; Table 2).

The ages of the trees along the river range from 20 to 52 years (supplementary material, Table 2). To compute sedimentation rates, we subtracted 2 cm (approximately the mean thickness measured from the upland reference site) to correct our measurements for deposition of locally derived organic matter. Sedimentation rates on the levee vary from 0.1 to 1.0 cm/y, while those on the floodplain vary from 0 to 0.6 cm/y (Fig. 4). Given the significant overlap in thicknesses measured at the floodplain with those from the upland reference site and the low probability of differences between median values (Table 2), sedimentation rates from the floodplain may not be significantly different from 0. This result suggests that dendrogeomorphic methods may not be particularly useful for estimating sedimentation rates when only a few centimeters of sediment have accumulated over the lifespan of the tree.

Table 2

Depth of material over tree roots in selected settings and results of Mann-Whitney test to evaluate significance of differences in median accumulation over basal roots between the reference site and the levee and floodplain sites

Area sampled	# of samples	Mean thickness (cm)	Median thickness (cm)	% of trees with 0 thickness	Range of values (cm)	Mann-Whitney statistic – comparison with Ref. Site
Reference Site	25.0	2.3	1.0	44.0	0-10	NA
Levee (<20 m from channel)	13.0	19.3	17.0	0.0	7-37	<0.001
Floodplain (>20 m from channel)	29.0	3.7	3.0	45.0	0-26	0.513

537

538

543 Fig. 4. Floodplain and levee sedimentation rates determined from dendrochronology (12
544 values with 0 sedimentation on the floodplain are not plotted). The cross section
545 illustrates the topographic setting. Sedimentation rates determined using mercury
546 inventories from two cores are also shown, as is the range of sedimentation rate estimated
547 from the detailed mercury profile at RRkm 4.18, located just downstream (Figs. 1 and 3).

544

545 *4.3. Reference site fallout radionuclide data*

552 Depth profiles of ^{137}Cs and *excess* ^{210}Pb ($^{210}\text{Pbex}$) from the reference site indicate

553 that activities are negligible in the lower sections of the soil (Fig. 5). Cesium-137

554 increases from the surface downwards with a primary peak from 4 to 6 cm and a lower,

555 secondary peak from 8 to 10 cm. Cesium-137 activities approach 0 at a depth of about 20

556 cm. Lead-210ex profiles follow a near exponential decline from the surface, reaching

557 negligible activities at a depth of about 14 cm. The upper 4 cm of the profile consist

558 primarily of sand, while lower layers consist primarily of silt (with a sandy interval from

20-28 cm). Bulk densities are $\sim 0.6 \text{ g/cm}^3$ in the upper 8 cm, but increase to 1.1 g/cm^3 below.

556

Fig. 5. Cesium-137 and $^{210}\text{Pbex}$ activities, percentages of clay, silt, and sand, and bulk density data from the reference site. The vertical extent of ^{137}Cs and $^{210}\text{Pbex}$ data indicate the interval sampled, while the horizontal extent represents the mean activity plus and minus one standard deviation.

561

The inventories of ^{137}Cs and ^{210}Pb at the reference site are $0.125 \pm 0.0022 \text{ Bq/cm}^2$

and $0.480 \pm 0.022 \text{ Bq/cm}^2$ (Table 3). If we assume that an equilibrium has been reached

between the rates of atmospheric deposition of $^{210}\text{Pbex}$ and radioactive decay, the ^{210}Pb

inventory can be used to estimate the atmospheric deposition flux of ^{210}Pb . The result,

$0.015 \text{ Bq/cm}^2/\text{y}$ is slightly higher than measured atmospheric fluxes of $0.012 \text{ Bq/cm}^2/\text{y}$

reported from Lewes, Delaware (Hartman, 1987), $0.013 \text{ Bq/cm}^2/\text{y}$ from the Chesapeake

Bay (Kim et al., 2000), and $0.013 \text{ Bq/cm}^2/\text{y}$ (1983) and $0.014 \text{ Bq/cm}^2/\text{y}$ (1984) from

Norfolk, VA (Todd et al., 1989).

Table 3

Fallout radionuclide inventories for the 3 study sites

Site	Cs137 inventory (Bq/cm ²)	Pb210 inventory (Bq/cm ²)
Reference	0.125 +/- 0.0022	0.480 +/- 0.022
RRkm 4.75	0.086 +/- 0.0034	0.409 +/- 0.045
RRkm 5.58	0.169 +/- 0.039	0.355 +/- 0.063

4.4. Fallout radionuclide data from floodplain sites RRkm 4.75 and 5.58

Cesium-137 and ²¹⁰Pb_{2ex} activities and other supporting data from eroding floodplain banks at RRkm 4.75 and 5.58 are generally similar to one another (Figs. 6 and 7). At RRkm 4.75, ¹³⁷Cs activity concentrations increase from 1 Bq/kg at the surface to a peak of about 3 Bq/kg at a depth of 20 to 25 cm. At RRkm 5.58, vertical distribution of ¹³⁷Cs activity concentration is very similar, though the absolute values of activity concentration are slightly higher. At both sites, ²¹⁰Pb_{ex} activity concentrations in surface sediments are ~25 to 30 Bq/kg, with an approximately exponential decrease with increasing depth. Lead-210_{ex} activity concentrations approach zero values at a depth of about 40 cm.

Fig. 6. Data from RRkm 4.75, including (from left to right), ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations, percentages of clay, silt, and sand, bulk density, mercury concentration, and organic content (loss-on-ignition). The vertical extent of ^{137}Cs and $^{210}\text{Pb}_{210}$ data points indicate the interval sampled, while the horizontal extent represents the mean activity plus and minus one standard deviation.

The sediment at RRkm 4.75 is composed of ~12% clay, 38% silt, and 50 % sand, with little systematic variation with depth (Fig. 6). At RRkm 5.58, sediments have a similar texture at the surface but become significantly finer with depth, such that at a depth of 80 cm, the sediment consists of about 20% clay, 78% silt, and only 2% sand (Fig. 7). Several sand-rich layers are evident at depths of 45 to 50 cm and 60 to 65 cm. The bulk density varies from 0.87 g/cm^3 (at the surface at RRkm 4.75) to 1.1 g/cm^3 . Bulk density values at RRkm 5.58 are relatively constant with depth (Fig. 7). Loss-on-ignition

varies from about 2 to 5% at both sites. Loss-on-ignition values decrease with increasing depth at RRkm 4.75, but do not vary systematically with depth at RRkm 5.58.

Mercury profiles at RRkm 4.75 and 5.58 both contain peak concentrations from 15 to 46 cm, suggesting that the period of peak mercury contamination from 1955 to 1961 occurs somewhere within this interval at both sites (Figs. 6 and 7). The peak ^{137}Cs activity (representing sediment deposited in 1963) also occurs within this interval at both sites, though modeling results (reported below) will suggest that this peak has moved downward in the profile a few centimeters since these sediments were deposited.

Fig. 7. Data from RRkm 5.58, including (from left to right), ^{137}Cs and $^{210}\text{Pbex}$ activity concentrations, percentages of clay, silt, and sand, bulk density, mercury concentration, and organic carbon (loss-on-ignition). The vertical extent of ^{137}Cs and $^{210}\text{Pbex}$ data points indicate the interval sampled, while the horizontal extent represents the mean activity plus and minus one standard deviation.

4.5. Fallout radionuclide modeling results

The model of Eqs. 1 and 2 was first fit to the $^{210}\text{Pbex}$ data from the reference site because P_0 was determined from the data (Table 4), so that the only unknown parameter was B , the bioturbation coefficient. Radionuclide data were normalized by the percentage of silt-clay, and the coordinate of cumulative mass ξ was computed for silt-clay only. The root-mean-square (rms) error between activities predicted using the model and the measured activities was computed. Acceptable simulations produced rms errors of 20% or less, yielding excellent fit of the model to the data (assessed visually in Fig. 8A). The resulting values of B range from 3.5 to $4.5 \times 10^{-8} \text{ kg silt-clay}^2/\text{cm}^4/\text{y}$ (Table 4). These somewhat unusual units can be converted to more familiar units of cm^2/y using Eq. (7)

$$B = \frac{B}{(\rho_b F_{sc})^2} \quad (7)$$

which yields B values ranging from 0.42 to $0.54 \text{ cm}^2/\text{y}$ with a bulk density estimate of 0.00058 kg/cm^3 and F_{sc} (fraction silt-clay) of 0.50 (Fig. 5).

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629

Table 4
Parameters obtained from modeling the vertical distribution of ²¹⁰Pb_{ex} and ¹³⁷Cs

Site - radionuclide	<i>B</i> (kg silt- clay ² /cm ⁴ /y)	<i>P</i> ₀ + <i>A</i> ₀ <i>w</i> (Bq/cm ² /y)	<i>V</i> (kg silt- clay /cm ² / y)	Sed. interval (y)	Mass sed. rate (kg silt- clay/cm ² /y)	Linear sed. rate (cm/y)	Calibration approach	# Runs (within error/ total)
Reference – Pb210 _{ex}	3.5- 4.5x10 ⁻⁸	0.015	NA	NA	NA	NA	< 20% rms error	3/11
Reference – Cs137	3.5- 5.5x10 ⁻⁸	5.0-5.5x10 ⁻³	2.25- 2.5x10 ⁻⁵	NA	NA	NA	Fit peak visually	8/19
RRkm4.75 – Pb210 _{ex}	2.0- 4.5x10 ⁻⁸	0.01-0.013	NA	0.75-4.5	2.3-3.0x10 ⁻⁴	0.50-0.65	< 30% rms error	11/62
RRkm 4.75 – Cs137	4.5x10 ⁻⁸	0.0013- 0.0015	2.25x10 ⁻⁵	0.75-4.0	2.3-2.7x10 ⁻⁴	0.50-0.58	Fit peak visually	6/43
RRkm5.58 – Pb210 _{ex}	1.0- 4.5x10 ⁻⁸	0.010-0.014	NA	1.5-10.0	2.3-2.9x10 ⁻⁴	0.44-0.62	< 30% rms error	10/81
RRkm5.58 – Cs137	4.5x10 ⁻⁸	0.0027- 0.0032	2.25- 2.5x10 ⁻⁵	0.5-6.0	2.7-3.3x10 ⁻⁴	0.58-0.73	Fit peak visually	11/43

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Fig. 8. Modeled radionuclide profiles of the reference site. (A) Distribution of ^{210}Pb obtained from three model runs with rms errors of $<20\%$. (B) Distribution of ^{137}Cs obtained from eight model runs; model fit is assessed visually. (C inset) The history of ^{137}Cs atmospheric deposition (in relative units) assumed by the model computations.

Fig. 9. Modeled radionuclide profiles of the eroding floodplain at RRkm 4.75. (A) Distribution of ²¹⁰Pbex obtained from eleven model runs with rms errors of <30%. (B) Distribution of ¹³⁷Cs obtained from six model runs; fit is assessed visually.

661

664 Figure 10. Modeled radionuclide profiles of the eroding floodplain at RRkm 5.58. (A)
 665 Distribution of $^{210}\text{Pb}_{\text{ex}}$ obtained from ten model runs with rms errors of $<30\%$. (B)
 666 Distribution of ^{137}Cs obtained from eleven model runs; fit is assessed visually.

665

676 The model was parameterized for the ^{137}Cs data from the reference site using a

677 somewhat different approach. Although the temporal variability of atmospheric

678 deposition of ^{137}Cs can be estimated (Fig. 8C), the absolute values are unknown, so the

679 average rate of atmospheric deposition, P_0 , was used to calibrate the model to the

680 measured values (again normalized by the percentage of silt-clay). Because the

681 bioturbation coefficient B was determined from the $^{210}\text{Pb}_{\text{ex}}$ calibration, the remaining

682 unknown parameter is the downward advection velocity, V . For ^{137}Cs , the calibration was

683 assessed visually rather than using the rms error because the location of the peak activity

684 and the elevated activities in the lower part of the profile could not be fit simultaneously.

685 Low rms errors are associated with good fits to the lower activities, but poor fits of the

686 peak. The peak seems more important to reproduce than activities lower in the profile

(since the peak corresponds to a well-defined date of 1963), so the visual fit of the model to the peak was used to evaluate the model predictions.

Eight of 19 model runs were accepted to parameterize the model (Table 4). The time-averaged rate of atmospheric deposition of ^{137}Cs ranges from 5.0 to 5.5×10^{-3} Bq/cm²/y, and the advection velocity, V , ranges from 2.20 to 2.25×10^{-5} kg silt-clay/cm²/y. The units of V can be converted to cm/y using Eq. (8),

$$V(\text{cm y}^{-1}) = \frac{V(\text{kg silt-clay cm}^{-2} \text{y}^{-1})}{\rho_b F_{sc}} \quad (8)$$

yielding a range of values of 0.078 to 0.086 cm/y, (using the bulk density and fraction silt-clay values quoted above), equivalent to a net downward migration of the peak activity of about 4 cm since 1963.

Next, the model was parameterized at the two floodplain sites (RRkm 5 and 5.58) to estimate floodplain sedimentation rates. Following the approach used at the reference site, the ^{210}Pb ex calibrations were evaluated using the rms error (though here a threshold of 30% was used rather than 20%, because the data could not be fit as closely) and the ^{137}Cs calibrations were evaluated visually to ensure that the location of the peak ^{137}Cs activity was well-represented. Values of B and V estimated from the reference site parameterization were followed closely, though some variations were allowed (Table 4) if these were needed to better fit the data. In addition to the sedimentation rate, w , the initial activity on newly deposited sediment (A_0) is an additional unknown parameter. It turns out that supply of radionuclides from atmospheric deposition is mathematically indistinguishable from that supplied with newly deposited sediment, so the total supply $P_0 + wA_0$ is reported together. The final unknown parameter in the modeling approach is

the interval of time between deposition events (which was varied independently from the time-averaged deposition rate, w).

The ^{210}Pb profiles are followed more closely than the ^{137}Cs profiles (Figs. 9 and 10). The location of the peak activity of the ^{137}Cs profiles can be well reproduced, but the model does not adequately explain elevated ^{137}Cs activities deeper in the profile.

Sedimentation rates (units of mass silt-clay deposited/ cm^2/year) range from 2.3 to 3.0 $\times 10^{-4}$ (equivalent to 0.50 to 0.65 cm/y) at RRkm 4.75, and from 2.3 to 3.3 $\times 10^{-4}$ (0.44 to 0.73 cm/y) at RRkm 5.58. Intervals of deposition range from 0.75 to 4.5 years at RRkm 5, while those for RRkm 5.58 are poorly constrained, varying from 0.5 to 10.0 years at RRkm 5.58. Results in Table 4, combined with Eq. (7) and appropriate values for ρ_b (0.0009 kg/cm^3 – RRkm 4.75; 0.0012 kg/cm^3 – RRkm 5.8) and F_{sc} (0.6 – RRkm 4.75; 0.55 – RRkm 5.58; Figs. 6 and 7), suggest that the ^{137}Cs peak has moved downward 2.1 and 1.9 cm , respectively, at sites RRkm 4.75 and 5.58 since 1963. The lower mobility of the ^{137}Cs at the floodplain sites compared to the reference site is interesting and could be a result of the higher silt-clay content of the floodplain sites, or possibly a consequence of the ongoing sedimentation on the floodplain.

The approximately exponential decline in ^{210}Pb activity with depth at the floodplain sites suggests that simple models could also be used to extract sedimentation rates from these data (though the assumptions of these models may be difficult to justify in floodplain settings). The sedimentation rate obtained using the CIC (constant initial concentration; Appleby and Oldfield, 1992) model for RRkm 4.75 is 2.6 (2.0 to 3.5, 95% C.I.) $\times 10^{-4}$ $\text{kg silt-clay}/\text{cm}^2/\text{y}$, while the corresponding estimate for RRkm 5.85 is 2.4 (1.9 to 3.3, 95% C.I.) $\times 10^{-4}$ $\text{kg silt-clay}/\text{cm}^2/\text{y}$. Both of these estimates agree well with the

modeling results presented in Table 4. Because the inventories of ^{210}Pb at the floodplain sites are less than the inventory at the reference site (Table 3), the CRS (constant rate of supply; Appleby and Oldfield, 1992) and CICC (constant initial concentration and constant sedimentation rate; Du and Walling, 2012) models cannot provide useful estimates of sedimentation rates.

4.6. HEC-RAS modeling and sedimentation from mercury inventories

The valley of the South River is frequently inundated by overbank flows (Fig. 11). Fifty-six percent of the 100-year floodplain is inundated every two years, and 83% of the floodplain is inundated every five years.

Mercury contamination of the floodplain is pervasive (Fig. 11): 320 analyses of floodplain sediments have an average concentration of 10 mg/kg, with a standard deviation of 20 mg/kg, a minimum of 0 mg/kg, and a maximum of 160 mg/kg. Mercury inventories vary from $<4 \times 10^{-5} \text{ kg/m}^2$ to more than $4 \times 10^{-1} \text{ kg/m}^2$ (Fig. 11; supplementary materials, Table 3). While mercury concentrations are high over most of the floodplain, an area near Sawmill Run has anomalously low concentrations (Fig. 11), suggesting that the alluvial fan created by Sawmill Run has diverted floodwaters from this area. Accordingly, we omit this area in our computations of the total mass of sediment (and mercury) that has accumulated on the floodplain since 1930.

Median centennial accumulation rates are 3.8 cm/100 years for the <0.3 -year floodplain, 1.37 cm/100 years for the 0.3- to 2-year floodplain, 0.4 cm/100 years for the 2- to 5-year floodplain, and 0.1 cm/100 years for the 5- to 62-year floodplain (Fig. 12).

Fig. 11. Maps showing the study reach, mercury inventories, and extent and frequency of flood inundation.

The total mass of sediment deposited on the floodplain from 1930 to 2007 is 4.9 ± 1.7 (95% confidence interval) $\times 10^7$ kg, corresponding to an average thickness of 2.5 cm (3.2 cm/100 years). Skalak's (2009) rating curve estimate for the total suspended sediment transported by the South River at Waynesboro from 1930 to 2007 was 4×10^8 kg, while the estimate based on Eggleston's (2009) result for the annual suspended sediment load was 6×10^8 kg. These figures suggest that floodplain deposition has captured 8 to 12% of the suspended sediment carried from 1930 to 2007 into our 4.5-km-long study reach.

756

768 Fig. 12. Comparison of post-settlement and mid-Atlantic *Recent* floodplain sedimentation
 769 rates with rates for the South River (adjusted to represent 100 years' accumulation). Post-
 770 settlement aggradation rates are from Bain and Brush (2005; MD), Jackson et al. (2005;
 771 GA), Lecce et al. (2008; NC), and Wilkinson and McElroy (2007; W&M). *Recent*
 772 sedimentation rates are from Allmendinger et al. (2007; indicated as A above), Schenk et
 773 al. (2013), and Smith and Wilcock (S&W; 2015). Sedimentation rates for the South River
 774 include those determined using dendrochronology (SR dendro), detailed mercury profiles
 775 (Hg), ^{210}Pb and ^{137}Cs data (FRN), and mercury. Accumulation rates are reported for
 776 South River floodplain surfaces inundated every 0 to 0.3 years, 0.3 to 2 years, 2 to 5
 777 years, and 5 to 62 years. Boxes indicate the 5th, 25th, 50th, 75th, and 95th percentile
 778 inventories (Hg Inventories; the latter are separated into classes based on the frequency of
 779 floodplain inundation).

5. Discussion

All four independent methods used in this paper demonstrate that floodplain sedimentation has been an active process along the South River during the last 75 years or so. Three detailed mercury profiles appear to reflect the chronology of mercury concentration on suspended sediment carried by the South River from 1930 to the present, with peak concentrations occurring well below the floodplain surface, suggesting that sedimentation has continued since 1955 to 1961 (the years of peak mercury concentration on suspended sediment carried by the South River). Dendrochronology provides sedimentation rates averaged over time periods ranging from 20 to 52 years. Sedimentation rates determined from dendrochronology are lower than those obtained from the detailed mercury profiles (Fig. 12), but the mercury data were obtained immediately adjacent to the channel (though one of these sites has only been adjacent to the channel since 1974), while many of the sites used for dendrochronology are located a considerable distance from the channel (Fig. 4). Sedimentation rates estimated from ^{137}Cs and ^{210}Pb data are consistent with those estimated from the detailed mercury profiles. Sedimentation rates from mercury inventories appear to be significantly lower than those estimated using the other three methods (Fig. 12); but cores for these analyses are taken from the entire valley, and our results may simply reflect low sedimentation rates that occur far from the channel.

A rather involved modeling approach has been adopted here to estimate sedimentation rates from FRN data. We could have obtained similar results by simply analyzing our ^{210}Pb data using the CIC method and by assigning the peak ^{137}Cs concentration in our profiles to 1963, the year of peak ^{137}Cs atmospheric activity from

nuclear weapons testing. However, these two methods require assumptions that our model allows us to test (if only approximately). Exponential ^{210}Pb profiles can be created by bioturbation and other processes, leading to errors in estimating sedimentation rates from the CIC method. Furthermore, the potential migration of ^{137}Cs peaks creates considerable uncertainty in assigning dates to ^{137}Cs profiles. While our modeling efforts clearly fail to account for many details of the processes affecting radionuclide profiles, they do allow us to assess the potential influences of bioturbation, migration of ^{137}Cs (which is not very significant in our floodplain sites), and episodic sedimentation.

Each method used here to determine floodplain sedimentation rates has strengths and weaknesses, and all necessitate simplifying assumptions that are difficult to verify. We do not claim that any method is better (or worse) than the others. However, the use of four independent methods, each based on different assumptions, should provide confidence in our conclusion that sedimentation has been an active process on the floodplain of the South River from 1930 to the present. This interpretation is also supported by HEC-RAS modeling, which demonstrates that overbank flows occur frequently in our study area.

Dooms Dam probably has probably exerted little influence on the results presented here. Backwater processes could only have been important before 1976; and even then, the detailed mercury profiles, dendrochronology, and one of the FRN bank profiles are all located upstream of any backwater influence of the dam (according to the model predictions of Pizzuto and O'Neal, 2009). Sedimentation rates at the FRN site located farther downstream (RRkm 5.58) are barely within the potential hydraulic influence of the dam before 1976 and not significantly higher than at the sites farther

upstream. Some of the floodplain cores in Fig. 11 are located relatively close to Dooms Dam. Pizzuto (2014) analyzed these and other floodplain mercury inventories upstream and downstream of dams along the South River, and no influence of the dams could be detected. The hydraulic model computations of Fig. 11 entirely ignore Dooms Dam, suggesting that Dooms Dam has little influence on contemporary flood inundation patterns. These arguments all provide important evidence supporting the interpretation that floodplain sedimentation in our study area is not controlled by Dooms Dam. However, additional study of the influence of mill dams on floodplain sedimentation would be helpful.

Figure 12 compares floodplain sedimentation rates from the literature with rates presented in this paper. Following European settlement of the eastern and midwestern U.S., sediment accumulated on floodplains at rates that are approximately an order of magnitude higher (or more) than the rates reported for the South River from 1930 to the present. However, direct measurements of floodplain sedimentation rates using clay pads over the last decade in three Piedmont watersheds of Maryland and Virginia (Schenk et al., 2013) document rates that are only slightly lower than post-settlement floodplain accumulation rates. Sediment budget estimates of Smith and Wilcock (2015) yield sedimentation rates that are similar to those of Schenk et al. (2013). Sedimentation rates reported by Allmendinger et al. (2007) determined using dendrochronology are similar to those reported here for the South River, as are those reported by Renshaw et al. (2013) using ^{210}Pb geochronology at a site in the northeastern U.S.

The compilation of Fig. 12 suggests that floodplain sedimentation was high following European settlement and that rates have declined in the twentieth and twenty-

first century (with significant variations that could be at least partially explained by watershed size, extent of urbanization, and other variables). However, Fig. 12 also demonstrates that floodplain sedimentation continues to be an important process, one that has sequestered 8 to 12% of the total suspended sediment load delivered to our study area in the South River. Even though these floodplains continue to store *legacy* sediment mostly deposited soon after European settlement, they are not fill terraces isolated from contemporary fluvial processes by past aggradation. Rather, they continue to store suspended sediment at rates comparable to other active floodplains (Lambert and Walling, 1987; Walling and Owens, 2003; Bourgoïn et al., 2007; Day et al., 2008).

Our results and discussion do not indicate how the floodplains of the South River are evolving through time, and we have not placed our observations into a broader context that would indicate how the valley of the South River is responding to contemporary (and future) fluvial processes. This is an important topic, but one that requires additional data and observations that are beyond the scope of this manuscript.

Our conclusions have important implications for watershed management. Pizzuto et al. (2014) presented sediment budget data from mid-Atlantic watersheds suggesting that 1.6 to 44% of the annual sediment load can be exchanged per kilometer of downstream transport between channels and floodplains. They further suggest that these exchange rates imply travel times of 100 to 1000 years between upland source areas and the outlets of larger drainage basins. Floodplain storage along our 4.5-km study reach is equivalent to ~2% of the annual load per kilometer, a value that is consistent with results obtained by Pizzuto et al. (2014), supporting their claim that floodplain storage should be considered in watershed management schemes designed to control the downstream

860 delivery of sediment and other particulate pollutants to estuarine receiving waters. Our
861 results also imply that channel-floodplain connectivity in the mid-Atlantic region is not
862 necessarily impaired by post-settlement alluviation, obviating the need for widespread
863 restoration designed to improve channel-floodplain connectivity.

864 **6. Conclusions**

865 Four independent methods have been used to quantify rates of overbank
866 deposition over multi-decadal to centennial timescales along the South River in Virginia.
867 Detailed mercury profiles sampled adjacent to the stream channel preserve the peak
868 mercury concentration on suspended sediment dating from 1955 to 1961 and suggest
869 sedimentation rates of 8 to 50 cm/100 years since that time. Sediment accumulation over
870 the roots of trees, corrected for in situ accumulation of organic material, suggest rates of
871 0 to 100 cm/100 years, with significantly higher values on levees and lower values on
872 floodplains farther from the channel. Cesium-137 and ²¹⁰Pb profiles from two eroding
873 streambanks are fit with an advection-diffusion model calibrated at an upland reference
874 site; these methods suggest sedimentation rates of 44 to 73 cm/100 years. Mercury
875 inventories from 107 floodplain cores, combined with a previously published
876 reconstruction of the history of mercury concentration on suspended sediment, provide
877 spatially comprehensive estimates of floodplain sedimentation along the South River.
878 Median sedimentation rates are 3.8 cm/100 years for the <0.3-year floodplain, 1.37
879 cm/100 years for the 0.3- to 2-year floodplain, 0.4 cm/100 years for the 2- to 5-year
880 floodplain, and 0.1 cm/100 years for the 5- to 62-year floodplain. While these
881 sedimentation rates are relatively low, the total mass of sediment stored from 1930 to
882 2007 is 4.9 ± 1.7 (95% confidence interval) $\times 10^7$ kg, corresponding to an average

883 thickness of 2.5 cm (3.2 cm/100 years). Floodplains have therefore stored 8 to 12% of the
884 total suspended sediment supplied to the study reach of the South River. HEC-RAS
885 modeling demonstrates that the floodplain of the South River remains connected to the
886 channel: 56% of the 100-year floodplain is inundated every two years, and 83% of the
887 floodplain is inundated every five years.

888 These results, combined with previously published data, provide the basis for a
889 regional synthesis of floodplain deposition rates since European settlement. Floodplain
890 sedimentation rates were high following European settlement, with published estimates
891 ranging from 50 to 200 cm/100 years. Sedimentation rates decreased by 1 to 2 orders of
892 magnitude during the twentieth and twenty-first centuries, but despite these lower
893 sedimentation rates, floodplains apparently continue to store a significant fraction of total
894 suspended sediment load. Many floodplains of the mid-Atlantic region should therefore
895 be considered to be active landforms fully connected to the rivers that flow within them,
896 rather than as terraces isolated from contemporary fluvial processes.

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898

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