Characterization of Bed Sediment Behind the Lower Three Dams on the Susquehanna River: Activities of Anthropogenic Gamma Emitting Isotopes

Report to Susquehanna River Basin Commission

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Jeffrey C. Cornwell
University of Maryland Center for Environmental Science
Horn Point Laboratory
Cambridge, MD 21613-0775

Introduction

Potential management actions regarding the rapid infilling of the lower Susquehanna River dams require a characterization of the chemical properties of the underlying sediments. Dams are important sites for the retention of sediments in fluvial ecosystems, often exhibiting high rates of sediment accumulation. Organic contaminants and trace metals are generally the main concerns with regard to contamination in most reservoirs. In the Susquehanna River basin, the presence of two nuclear power plants adds a second level of concern. Nuclides come from ongoing low level inputs and from accidental releases. The high rate of sediment input has lead to the concern that sediment retention by these dams will diminish in the future; this may lead to higher inputs of sediment to the downstream Chesapeake Bay. There appears to be considerable imprecision in the time frame of complete infill (i.e. Hermann et al. 2003), as will as in the estimate of the efficiency of particulate retention (i.e. < 25% from Donoghue et al. 1989 & McClean et al. 1991; >50% from Hermann et al. 2003).

Several key studies have investigated the distribution of reactor nuclides in the lower Susquehanna. Donoghue et al. (1989) and McClean et al. (1991) both used reactor nuclides to estimate sediment accretion and the retention of fine-grained sediment in the lower three reservoirs. Donoghue et al. (1989) used a combination of surface grabs and 6 core profiles to make estimates of retention; McClean et al. (1991) used several core profiles; of 5 cores collected, only 3 were useful for reactor nuclides other than ^{137}Cs. ^{134}Cesium was largely restricted to the top 5-10 cm of sediment.

The question asked by this measurement program is quite different than that of these other studies. This study is focused on the bulk sediment profiles from a contaminant perspective, rather than as a measure of accretion or particle retention efficiency. This is the perspective used in this overall larger study of contaminants (Hill et al. 2000). Thus, we are not doing “traditional” geochronology (i.e. McClean et al. 1991; Owens and Cornwell 1995), but rather are sampling these sediments to best characterize the bulk deposit that might conceivably be dredged. The advantage of this approach is relevance to dredging; the disadvantage, as shown by the metals and organic data, is that we do not develop a sense of time.

Background on Radionuclides

Radioactive decay is a key part of the solar system and has major effects on the present day earth. Primordial nuclides, formed at the “big bang”, generate sufficient heat in the earth’s core to keep the earth’s mantle from solidifying. Substantial amounts of uranium-thorium series nuclides are found in the earth’s crust, as well as other nuclides such as the gamma-emitting nuclide ^{40}K. Human exposure to radiation includes sources from space, soils, the atmosphere, and in more recent times, man’s activities. Many radionuclides are occluded in minerals (i.e. ^{40}K) or are particle reactive (i.e. ^{137}Cs). The chemical behavior of radionuclides is identical to that of non-radioactive forms of the nuclide. In the case of radioactive metals, they often are strongly particle reactive and most of the metal is retained in particulate form. The accumulation of metals in reservoir

sediments such as in the Susquehanna is a major sink for metals in that system. Testing of nuclear weapons in the 1950's and early 1960's spread radioactive isotopes of cesium, strontium, tritium and plutonium throughout the earth's atmosphere, with a fall out onto soils and water. Nuclear power plants can also be sources of radionuclides, even without catastrophic events such as Three Mile Island and Cherbonyl.

Donoghue et al. (1989) identified the nuclides of environmental interest in his paper (Table 1). In this study, we examined our counting data for all of these nuclides, with particular emphasis on ^{134}Cs, ^{137}Cs and ^{60}Co. The short half lives of the nuclides (except ^{137}Cs) results in their rapid decrease to levels below the detection limit. In Figure 1, the releases of nuclides in the late 1970's from the reactors is plotted, with the calculated decay of the nuclides shown over time. Clearly, in the 10 year time frame (i.e. ~15% of the time of existence of these dams) most of the nuclides disappear. Thus, the average deposit in these reservoirs should only have ^{137}Cs; this nuclide first appeared in the 1950's as a consequence of atmospheric testing of fusion weapons.

Table 1. Reactor nuclides (gamma only), half life, and gamma energies. The nuclides in bold have been observed in surface sediments (Donoghue et al. 1989). We checked for all these nuclides in our counting.

Nuclide	Half Life ($T_{1/2}$, days)	Gamma Energies (Kev)
^{110}Ag	252	885
^{58}Co	71	511
^{60}Co	1924	1173
^{134}Cs	752	796
^{137}Cs	11019	662
^{54}Mn	312	835
^{65}Zn	244	1115
^{95}Zr	64	724

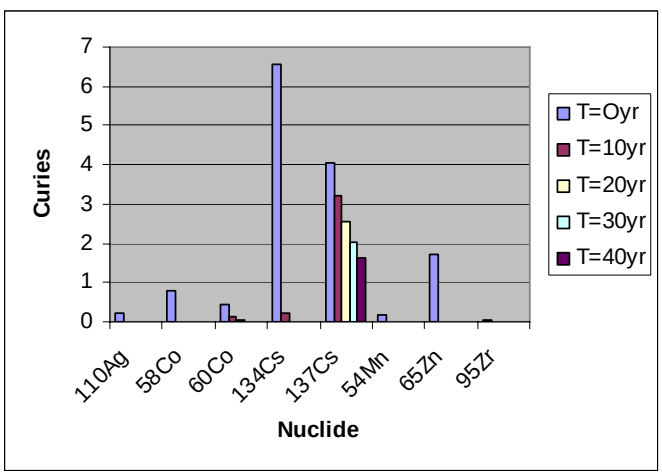
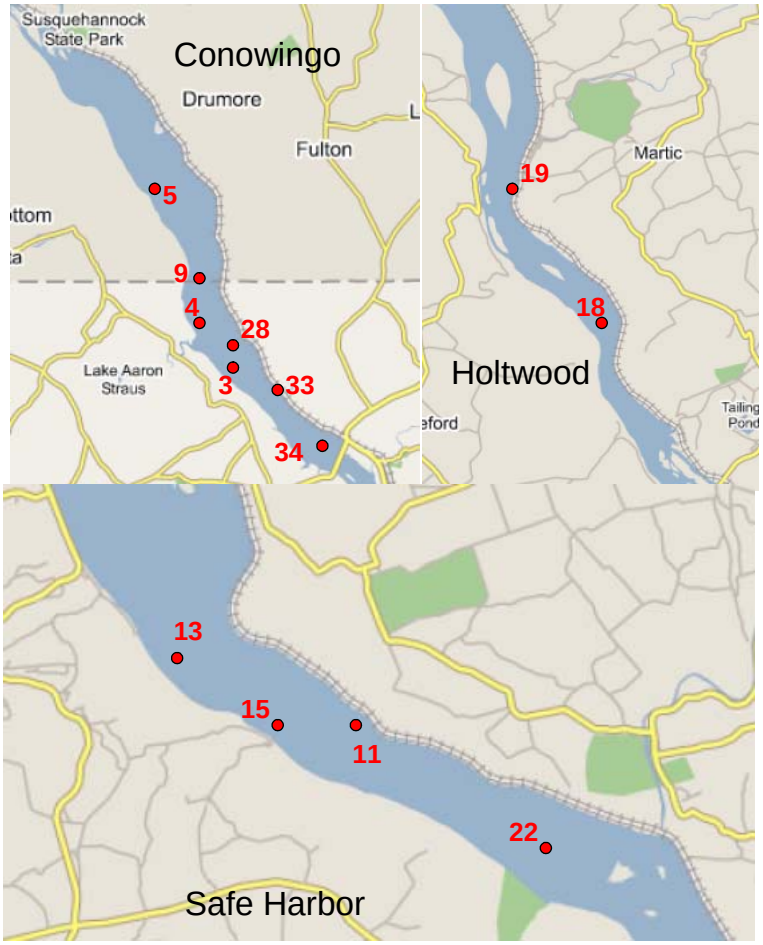


Figure 1. Inputs of reactor nuclides in the late 1990's, as well as their persistence after decay. Note that ^{137}Cs is the main persistent reactor nuclide.

Study Approach

For gamma analysis, I chose 13 sites, averaging 4 samples per site (Figure 2). Sites were chosen to cover a reasonable distribution of locations within each reservoir, with a somewhat greater density of sites in Conowingo because of the inputs from Peach Bottom Atomic Power Station, where somewhat higher activities have been found (McClean et al. 1989).



Methods

Sediment collection methods are detailed in Hill et al. (2000). Sediments were dried at 65°C and powdered using a mortar and pestle. Sediment (20 g) was placed in a plastic jar for counting. A Canberra germanium detector system (planar LeGe) was used for counting. This system was calibrated with a multi gamma source; sediment data were compared to our counts of NIST sediment SRM 4357, a low-level calibrated marine

sediment. Samples were generally counted for 24 hours. We did not observe any reactor nuclides in these samples except ^{137}Cs; each spectra was examined for all nuclides in Table 1. In addition, we calibrated ^{226}Ra using the NIST standard. While ^{226}Ra was not of environmental concern, it's relatively constant activity in fine grained muds provided a secondary check of the counting system. We expected 20-30 mBq g^{-1} activities; in our Chesapeake Bay ^{210}Pb dating (i.e. Cornwell et al. 1996), we generally found ^{226}Ra activities of ~25 mBq g^{-1}. The detector we used was optimized for lower energy nuclides and could not measure the higher energy ^{40}K; Donoghue et al. (1979) have shown a relatively constant activity of this primordial nuclide and it does not have other chemical or radiological characteristics. A sediment ^{137}Cs peak is shown in Figure 3.

Counting statistics are shown with the data. We normalized our counting to 24 hour; with shorter counts, identifying the 137Cs peak was more difficult. At non-zero activities < 10 mBq g^{-1}, the counting error averaged 35% of the activity, for activities between 10 and 20 mBq g^{-1} the average counting error was 14% and at activities > 20 mBq g^{-1}, the average counting error decreased to 10%. Two recounts of the NIST SRM 4357 sediment standard for 24 hours yielded ^{137}Cs activities of 10.4±1.6 and 13.3±1.6 mBq g^{-1}, well within the 2.5-97.5% tolerance range (10.3-15.0 mBq g^{-1}). Results for recounts of NIST SRM 4357 for ^{226}Ra were at the high end of the tolerance range (16.3, 15.5 mBq g^{-1} for counts, NIST range = 10.3-15.0), we have not adjusted this data downward.

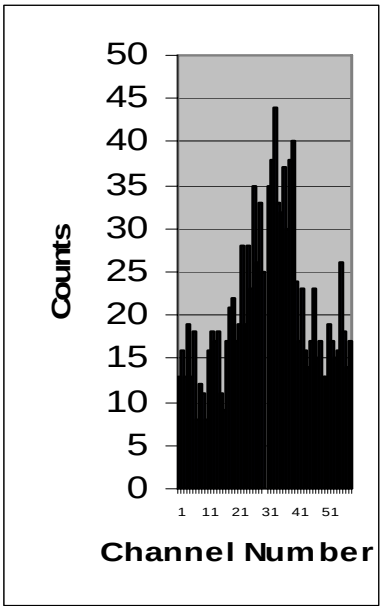


Figure 3. Spectra for ^{137}Cs; the MCA board in the gamma system has > 8,000 channels and this peak is centered at 662 Kev.

Results

The analysis showed a broad range of activities for ^{137}Cs and a limited range for ^{226}Ra (as expected). All data are shown in Table 1, with summary statistics in Table 2. Two histograms were generated from all 52 samples that were counted (Figure 4). We had no measurable 137 activity in 1/5 of the samples, with the most abundant activity range of 5-10 mBq g^{-1}; our sampling emphasized more samples near the sediment surface, so a truly random selection would have resulted in more non-detectable activities. The ^{226}Ra activities were similar in most cores sections, with the average (24±5 mBq g^{-1}) similar to the 25 mBq g^{-1} observed in fine-grained Chesapeake Bay sediments.

Our highest activities (Table 1) were found at SRC-4, near the Peach Bottom Atomic Power Station (PBAPS). This result is consistent with that of McClean et al. (1991) who found higher ^{137}Cs (and ^{134}Cs) near the reactor (their highest activities were ~ 350 mBq g^{-1} with most activities of their core A < 60 mBq g^{-1}). Our SRC-4 core location is further from Peach Bottom than their core and these ^{137}Cs activities are generally about half of the McClean et al. (1991) data. Core SRC-9 is located closer to PBAPS, and the top two sections analyzed have activities about 2/3 of that at SRC-4; the three deeper sections have no measureable activity. A plot of the ^{137}Cs activity versus depth (Figure 5) shows only modest down core change in SRC-3; core SRC-3 showed low activity in the surface and higher activity in deeper sections. Cores SRS-15 and SRH-19 had much lower activities with downcore decreases.

Table 2. Analytical results for ^{137}Cs and ^{226}Ra. The results are report as milli-bequerels per gram (NIST units); these may be converted to dpm per gram by multiplying by 0.060. To convert to pCi per gram, multiply these data by 0.00758. Standard errors are counting errors based on total net counts and background.

Core	Depth (cm)	^{137}Cs	^{137}Cs S.E.	^{226}Ra	^{226}Ra S.E.
		mBq g^{-1}			
SRC-3	0-8	4.14	1.18	24.0	2.0
	51-55	3.99	2.00	23.6	2.1
	70-80	15.77	0.08	31.3	2.1
	100-110	5.92	1.64	24.5	1.8
	125-130	9.47	2.81	33.4	2.7
SRC-4	0-10	29.9	2.9	24.0	1.1
	50-60	42.0	3.7	20.8	2.5
	158-188	36.5	2.9	18.3	1.8
	158-188	31.2	3.1	21.3	2.2
	100-110	28.8	2.5	26.6	2.4
	310-320	35.7	2.2	24.7	2.0
SRC-5	10-20	30.6	2.5	28.3	2.2
	60-73	0.0	0.0	23.7	2.2
	150-160	0.0	0.0	16.1	2.0
	203-313	0.0	0.0	26.0	2.8
SRC-9	10-20	23.18	1.90	20.0	1.6
	90-100	32.00	2.43	31.4	2.2
	170-180	0.00		23.5	1.7
	250-260	0.85	1.51	26.8	1.9
	310-320	0.00		26.8	2.0
SRS-11	10-20	7.0	1.5	24.5	2.2
	40-50	11.5	2.0	25.2	2.7
	75-85	2.7	1.3	21.7	2.0
	115-123	14.1	1.7	25.4	1.9
SRS-13	20-30	7.61	1.68	23.9	1.9
	50-60	11.87	1.85	24.2	1.8
	103-113	0.00		21.3	1.7
SRS-15	5-10	5.8	1.3	21.5	2.1
	40-50	12.2	1.9	26.2	2.8
	80-90	11.1	1.8	25.6	2.0
	120-130	8.6	1.0	22.7	1.6
	155-165	6.4	1.5	13.3	2.0
SRC-18	10-23	2.17	0.72	17.6	1.7
	40-81	0.00		20.2	2.1
SRH-19	3-13	2.1	1.8	21.9	2.7
	50-60	0.0	0.0	21.5	2.1
	90-100	0.0	0.0	24.4	1.6
	130-140	0.0	0.0	24.1	2.3
SRS-22	10-20	7.87	2.31	21.6	2.0
	40-50	6.32	1.38	22.4	1.9
	70-82	8.81	1.51	23.1	1.9
SRC-28	10-20	6.8	1.4	23.5	1.9
	60-70	8.0	1.4	22.9	1.6
	100-110	0.0	0.0	25.0	2.0
	140-150	6.6	1.4	22.2	2.0
SRC-33	10-20	14.9	3.8	44.4	4.1
	40-50	8.0	1.7	23.9	2.0
	50-60	3.5	1.7	26.1	1.9
	87-98	4.8	1.6	22.6	2.1
SRC-34	10-20	5.5	1.7	26.3	2.2
	30-40	9.7	1.9	27.2	2.7
	50-60	9.6	1.9	27.1	2.1

Table 2. Data statistics.

Nuclide	Low	High	Median	Average	Standard Deviation
	Activity (mBq g^{-1})				
^{137}Cs	0	42	7	10	11
^{226}Ra	13	44	24	24	4

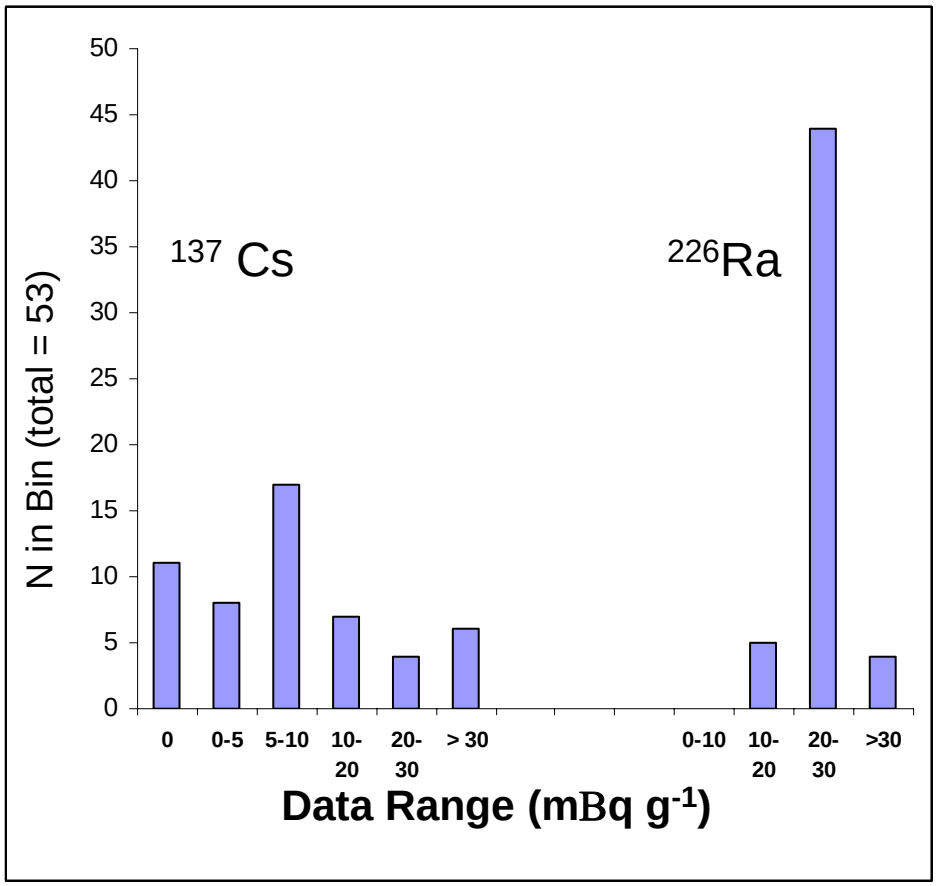


Figure 4. Histogram of ^{137}Cs and ^{226}Ra in Susquehanna reservoir samples (n=50).

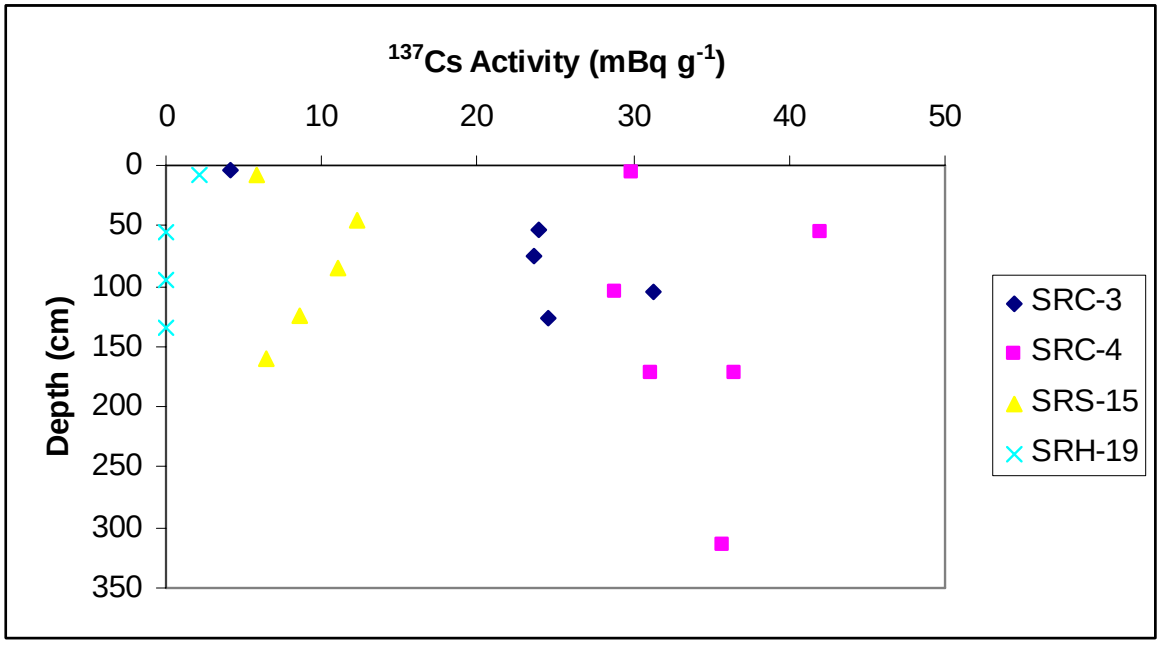


Figure 5. Representative Depth Profiles of ^{137}Cs.

Discussion

The activities of ^{137}Cs found in these reservoir sediments are generally lower than that observed in surficial grabs (Donoghue et al.1989) and lower than the core data from near Peach Bottom (McClean et al. 1991). I did not observed any hot spots of high activity of any other nuclides, suggesting that radionuclide "contamination" is not a likely issue with these sediments. Overall activities are somewhat lower than peaks found in slowly accumulating sediments (i.e. as in lakes; Robbins et al. 1978). A comparison of 137Cs activity with other aquatic systems shows that these numbers are not extraordinary. In two reservoir studies, Van Metre and Callender (1997) and Callender (1997) found maximum137Cs activity of 180 and 60 mBq g^{-1} respectively. In New England softwater lakes, Davis et al. (1984) found peak core ^{137}Cs activity of 800-5,300 mBq g^{-1}; such high activity sediment result from low particulate inputs relative to the atmospheric input of ^{137}Cs. In higher sedimentation sites such as in this study, the overall ^{137}Cs activity on a per gram basis in not especially high because of dilution by the high inputs of particulates.

It would appear that the higher activities near Peach Bottom are a consistent feature of that region (i.e. from this study and others). It is unlikely that such modest activities would have any environmental consequences for sediment placement. Any future sampling for radionuclides in conjunction with dredging assessment would do well to focus on the region near PBAPS.

The issue of mobility of ^{137}Cs in dredge sediment placement is beyond the scope of this report because of the wide range of placement options and unknown nature of the ^{137}Cs-sediment association. Unlike many metals, ^{137}Cs can be somewhat mobile in anaerobic sediments (i.e. Evans et al. 1983; Comans et al. 1989) because of displacement by NH_4^+. Leaching of ^{137}Cs into hydrological systems may be important in highly contaminated systems, but the low activity of these sediments suggest this would not be an issue. Although only of modest relevance to this situation, work on ^{137}Cs contaminated soils at the Idaho National Engineering and Environmental Laboratory have as a goal an activity of ~ 300 mBq g^{-1} before fitting into "residential onsite" criteria. Overall activity from our work, assuming about half of the reservoir sediment is uncontaminated (i.e. deposited before major ^{137}Cs inputs) would be ~5 mBq g^{-1}, a level considerably below what DOE is concerned about at their sites (http://c2d2.eml.doe.gov/ap.cfm?target=ap.cfm&medium=soil&contami=cesium-137&instl=INEL)

Conclusions

- Activities of 110Ag, ^{58}Co, ^{60}Co, ^{134}Cs, ^{54}Mn, ^{65}Zn, and ^{95}Zr were below the limit of detection due to the short half lives of these nuclides.

- The highest activities of ^{137}Cs were found near the Peach Bottom Atomic Power station. These results are consistent with other studies conducted near the reactor.

- It is unlikely that higher activities near Peach Bottom would have any environmental consequences for sediment placement.

- When compared to measured results in other studies (Donoghue et al.1989; McClean et al. 1991), the activities of ^{137}Cs found in this investigation of reservoir sediments were generally lower than activities observed in core data from near Peach Bottom and in surficial grab samples.

- The range of ^{226}Ra activities of 20-30 mBq g^{-1} were similar to ^{226}Ra activities found in other studies in the Chesapeake Bay.

- Hot spots of high activity were not observed suggesting that radionuclide contamination was not a likely issue with these sediments.

Recommendations

In future assessments of contaminants associated with dredging, it would be valuable to composite samples at 50 cm increments (or more), to capture the whole profile. The approached used in this study is useful to identify potential hot spots, but especially with hydraulic dredging, the most relevant number is that which is likely to end up in placement. Dredging will homogenize these profiles and result in a dilution of the higher ^{137}Cs activity sediment with low ^{137}Cs activity sediment. Such compositing is less important for trace metal or trace organic sampling because of the longer record of such

contamination. However, the overall low ^{137}Cs activity found in the most enriched samples in this study suggests that the conclusions from the work would be unaffected.

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