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The use of bed sediments in water quality studies and monitoring programs

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Abstract. In most water quality monitoring programs, either filtered water (dissolved) or suspended sediment (either whole water or separated suspended sediment) are the traditional sample media of choice. This results both from regulatory requirements and a desire to maintain consistency with long-standing data collection procedures. Despite the fact that both bed sediments and/or flood plain deposits have been used to identify substantial water quality issues, they rarely are used in traditional water quality monitoring programs. The usual rationale is that bed sediment chemistry does not provide the temporal immediacy that can be obtained using more traditional sample media (e.g., suspended sediment, water). However, despite the issue of temporal immediacy, bed sediments can be used to address/identify certain types of water quality problems and could be employed more frequently for that purpose. Examples where bed sediments could be used include: (1) identifying potential long-term monitoring sites/water quality "hot spots", (2) establishing a water quality/geochemical history for a particular site/area, and (3) as a surrogate for establishing mean/median chemical values for suspended sediment.

1 Introduction

In typical environmental/water quality-monitoring programs the most common samples collected and analyzed are filtered and unfiltered (whole) water (e.g., ASTM, 2011a, b; APHA, 2012; Horowitz, 2013; USGS, various). The former are used to determine "dissolved" constituents whereas the latter are used to determine suspended sediment concentrations (SSCs)/grain-size distributions, and sediment-associated chemical constituents by subtraction. Although bed sediments have been used in large-scale (e.g., national) environmental surveys, as well as for geochemical reconnaissance (e.g., mineral exploration), they rarely are used in traditional water quality-monitoring programs (e.g., Hawkes and Webb, 1962; Webb et al., 1978; Fauth et al., 1985; Otteson et al., 2000; Gustavsson et al., 2001). However, of all the potential sample media, bed sediments probably are the easiest to collect and process, and are least likely to suffer from contamination or insufficient sample mass/analytical detection issues. Hence, their lack of use is somewhat surprising, but results from several factors. First, since the publication of the Hawkes and Webb (1962) treatise on geochemical exploration, as well as the publication of several national geochemical/environmental atlases (e.g., Webb et al., 1978; Fauth et al., 1985; Otteson et al., 2000), bed sediments have been used to detect geochemical/environmental spatial differences, but rarely are viewed as sensitive to short-term (geo)chemical variations; whereas the latter are the usual goal of most monitoring programs. Second, bed sediment surveys typically employ grain-size limited aliquots (e.g., $< 180\,\mu m$) to facilitate spatial comparisons. That traditional grain-size range has little connection with suspended sediment grain-size distributions; hence, bed sediments have never been viewed as surrogates for suspended sediment. However, recent surveys using the $< 63\,\mu m$ fraction of the upper 1 cm of bed sediments, collected after low-flow periods (e.g., Horowitz et al., 1999, 2001a, b, 2012, 2014), or surficial bed sediments collected from exposed surfaces after storms or floods (e.g., Van Metre et al., 2006; Horowitz et al., 2014), do appear to reasonably reflect suspended sed-

Table 1. Summary of Utoy Creek data from the < 63 µm fraction of bed sediment samples ($n = 47$).

Units	(mg kg^{-1})						(wt %)			
Category	Cu	Co	Ag	Cd	Pb	Zn	Al	Fe	Ti	TOC
Min	16	16	< 0.5	< 0.5	38	100	9.3	2.8	0.55	1.4
Max	120	35	0.88	0.88	200	910	12.6	6.7	0.9	5.1
Median	56.5	22	0.61	0.35	80	220	10.5	4.7	0.68	2.8
Baseline (min)*	14	8	0.1	0.2	14	71	4.9	2.2	0.25	1.3
Baseline (max)*	26	16	0.3	0.6	26	110	6.9	3.6	0.41	2.5

* From Horowitz and Stephens (2008)

iment chemical composition. Hence, grain-size limited bed sediment aliquots may represent a useful media for non-traditional monitoring programs intended to detect event-related effects, or long-term environmental/water quality-changes (e.g., decadal climate/land use-changes), provided samples are collected and analyzed every 5 to 10 years, or to determine mean/median suspended sediment associated chemical concentrations as a means of establishing first order approximations of fluvial fluxes of suspended sediment-associated chemical constituents (e.g., Horowitz et al., 2012, 2014). Additionally, the chemical analysis of cored bed sediment samples, in conjunction with absolute age dating, can be used to reconstruct the water quality/geochemical history of an impoundment (e.g., Horowitz et al., 1988, 1995; Grosbois et al., 2001; Van Metre and Horowitz, 2013; Gray et al., 2015). Three examples of water quality studies that employed bed sediments *in lieu* of more traditional sample media, as a proof of concept, are described herein.

2 Utoy Creek, Atlanta, Georgia

Utoy Creek is a small urban stream that flows through the City of Atlanta, Georgia as well as well as being a tributary of the Chattahoochee River (e.g., Horowitz and Hughes, 2006; Horowitz, 2009; West Atlanta Watershed Alliance, 2016). The creek is some 42 km long and the watershed encompasses some 89 km^2 (West Atlanta Watershed Alliance, 2016). In 2015 representatives from Fulton County contacted the authors with a request to evaluate water quality in the watershed with a view to identifying potential contaminants and to recommend long-term monitoring sites. The creek already had been listed as impaired as a result of elevated dissolved Zn levels (Andrew Mycroft, personal communication, 2015).

2.1 Methods

Bed sediment samples were manually collected during a week in late 2014 after an extended period of low-flow at roughly equidistant intervals covering the entire watershed; additionally, major tributaries to Utoy Creek also were sampled. Sampling procedures were similar to those used by Horowitz et al. (2012) and entailed collecting multiple equal volume aliquots collected from the upper 1 cm of the creek bed and/or the still wet floodplain at each site. The aliquots for each site were composited in the field, stored in clear plastic bags, and refrigerated until they could be returned to the laboratory. In total, 47 samples were collected, of which 5 were replicates to evaluate sampling precision.

All the samples were processed and analyzed following the procedures outlined in Horowitz et al. (2012). Each sample was oven-dried at 105 °C. Representative aliquots of each composite were obtained by coning and quartering the dried sediment and then wet-sieved through a 63 µm non-contaminating nylon and clear plastic sieve to obtain representative subsamples for subsequent chemical analysis. A selected group of trace elements (Ag, Al, Cd, Co, Cu, Fe, Pb, Ti, and Zn) were determined using AAS after a HF/$HClO_4$/*aqua regia* digestion in teflon beakers at 200 °C. TOC was determined by combustion using a Carlo-Erba C / N analyzer.

2.2 Results and discussion

With the exception of Cd, all the other constituents determined contained elevated individual as well as median levels that exceeded national baseline concentrations (Table 1; Horowitz and Stephens, 2008). Whilst the median concentration for Cd did not exceed national baseline levels, individual samples did.

Of particular interest to Fulton County was that whilst most of the samples contained elevated major and trace element, as well as TOC concentrations, the samples displaying the most elevated levels all clustered around one particular location in the watershed. That location is near the mouth of Utoy Creek where it discharges to the Chattahoochee River, and also is the site of a number of medium to light industries, including a galvanizing plant (Fig. 1). The survey also indicated that Utoy Creek is likely to display impaired dissolved concentrations for a number of constituents other than Zn (e.g., Cu, Co, TOC). Finally, based on this survey, potential new, long-term monitoring also sites were identified.

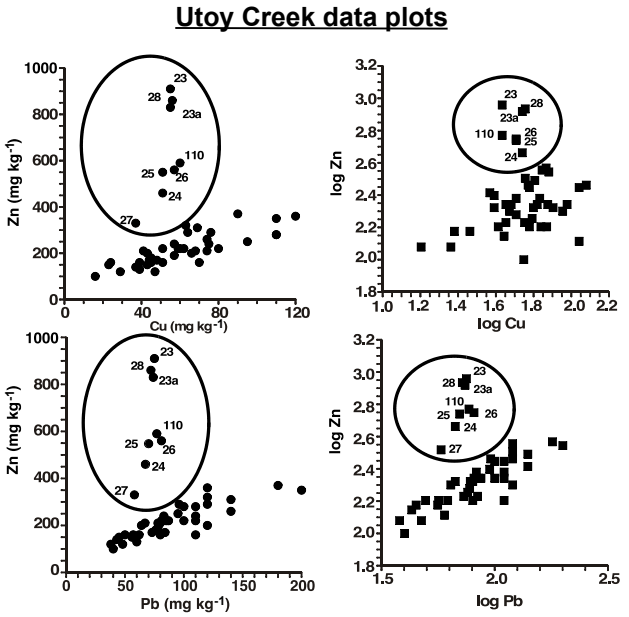

Figure 1. Plots of chemical data from the < 63 µm fraction of bed sediments from Utoy Creek, Georgia, USA. There are similar plots for Zn/Fe, Zn/TOC, Zn/Al, and Zn/Co.

3 Lake Coeur d'Alene, Idaho and the Spokane River

Lake Coeur d'Alene (CDA) is a natural (submerged river bed) lake in the northern panhandle of Idaho that formed from the outwash from Lake Missoula during the last interglacial (e.g., Hobbs et al., 1965). The lake lies between the Selkirk and the CDA Mountains and extends northward from the St. Joe River to the headwaters of the Spokane River near the city of CDA, Idaho (Meckel Engineering et al., 1983; Bender, 1991). The main body of the lake is about 3.2 km wide by 40 km long, and up to 150 m deep in the thalweg (Meckel Engineering et al., 1983). The Spokane River flows downstream from the lake, passes through a series of dams, and eventually discharges into the Columbia River, some 180 km downstream.

The South Fork of the CDA River drains a substantial part of the CDA mining district and the so-called "Silver Valley". The mining district has been in operation since the 1880's and was one of the major sources of Ag, Pb, and Zn in the US (e.g., Bender, 1991). Most of the mining and ore-processing wastes were discharged directly into the South Fork of the CDA River (e.g., Horowitz et al., 1993). As late as 1964, estimates indicated that some 2200 tonnes/day of mining and processing wastes still were entering the South Fork (Reece et al., 1978). It also has been estimated that during the course of mining, processing, and smelting operations in the area, some 115 million tonnes of mine tailings were produced and that over 60 % of this material probably entered the South Fork and the CDA River system (Javorka, 1991). These materials were highly enriched in Ag, As, Cd, Cu, Fe, Hg, Mn,

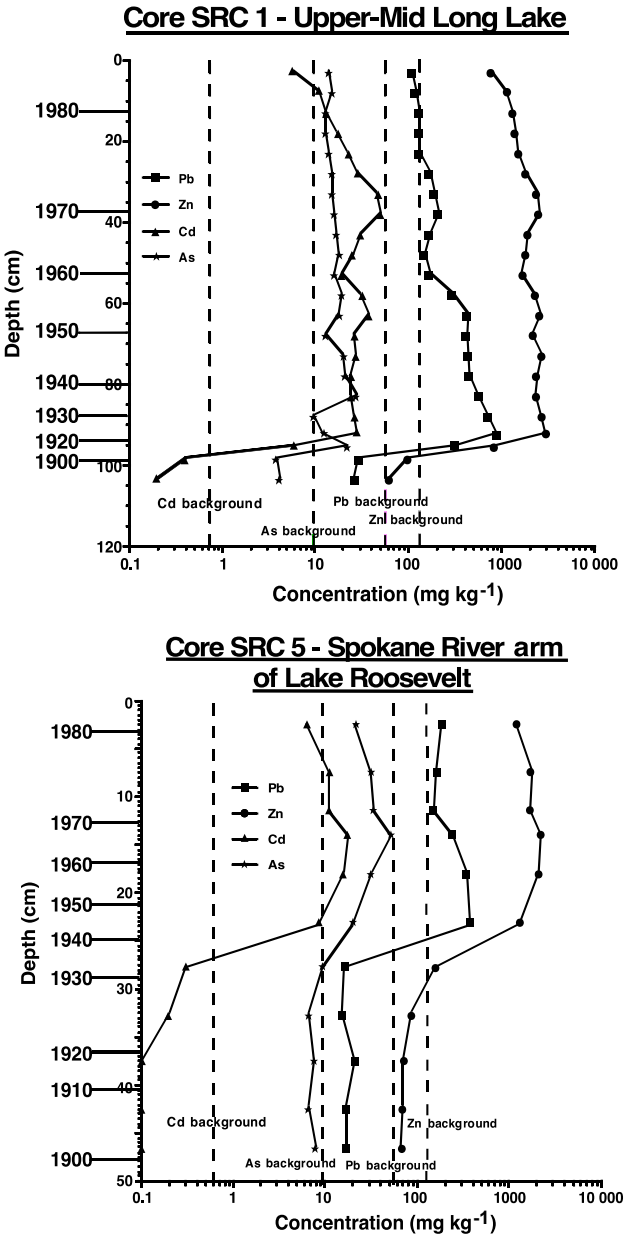

Figure 2. Plots of chemical data from two sediment cores collected in impoundments along the Spokane River, Washington, USA.

Pb, Sb, and Zn (Rabe and Bauer, 1977; Bender, 1991). In 1968, tailings ponds were established to limit sediment dispersion and downstream transport (Horowitz et al., 1993). A series of studies conducted between 1989 and 1999 indicated that Lake CDA contained some 75 million tonnes of trace element rich sediment, and that elevated sediment-associated constituents could be traced all the way to the Columbia River in the state of Washington (e.g., Horowitz et al., 1993, 1995; Grosbois et al., 2001).

The local mining companies, as well as the Northwest Mining Association, contended that the majority of the im-

pacted sediment resulted from natural weathering processes associated with exposed segments of the ore body. To address this issue, a series of gravity cores were collected in Lake CDA and in the Spokane River system (Horowitz et al., 1995; Grosbois et al., 2001). The analytical data from the cores, in conjunction with absolute age dating based on ^{137}Cs and $^{210}Pb_{ex}$, clearly indicates that the onset of sediment-associated trace element enrichment in Lake CDA and in the Spokane River began somewhere around 1910 ± 20 years, which is roughly contemporaneous with the onset of mining and smelting operations in the region (Fig. 2). Note that the onset of sediment associated-trace element enrichment of the most downstream core that was collected in the Spokane River arm of Lake Roosevelt began around 1930 (Fig. 2; Grosbois et al., 2001). That is contemporaneous with the initial closing of the Grand Coulee Dam, downstream from the Spokane River, on the Columbia River that also caused the formation of Lake Roosevelt and the Spokane River arm (e.g., Grosbois et al., 2001). Also note that the sediment-associated trace element concentrations, at least in the Spokane River, began to decline around 1970; that is contemporaneous with the construction of the tailings ponds along the South Fork of the CDA River. This would indicate that bed sediment chemistry does appear to respond fairly quickly to land use changes and or remediation measures. The CDA ore body has been dated as Proterozoic (> 500 million years BP; Hobbs et al., 1965), whereas Lake CDA has been dated as forming some 14 000 years BP (Wyman, 1993); as a result, it is highly unlikely that the vast majority of the trace element-rich sediments in Lake CDA, and in the Spokane River system, resulted from natural weathering processes.

4 The U.S. National Coastal River Survey

Between 2010 and 2011, the U.S. Geological Survey (USGS) carried out a bed sediment-associated chemical survey using material collected at or near the mouths of 132 coastal rivers in the US. The primary objective of the study was to use the chemical data generated from the samples to develop first-order approximations of the chemical fluxes emanating from the US to the coastal zone (Horowitz et al., 2012). The methods employed were essentially the same as those used in the Utoy Creek survey described earlier.

The basic premise of the study is that the chemical analyses of the < 63 µm fraction of the upper 1 cm of riverbed sediments can serve as a surrogate for the average chemical composition of recent suspended sediments transiting each site. To evaluate the validity of that premise, minimum, maximum, and median suspended sediment-associated chemical concentrations determined between 1994 and 2006, as part of the revised NASQAN Program, determined for separated suspended sediment and analyzed utilizing the same procedures used in this study (Horowitz et al., 2001a, b), were compared with the chemical concentrations generated from the < 63 µm fraction of the collected coastal river samples (Table 2; Horowitz et al., 2012). Based on that comparison, it appears that the premise is valid for the majority of the trace/major elements determined in the study. On the other hand, the data for carbon (TOC and TC), nitrogen (TN), and phosphorus (TP) are more ambiguous (Table 2). The bed sediment values for these constituents tend to fall either just below or just within the minimum levels for suspended sediment. This difference may be a reflection of the relatively small number of samples available for the comparison (especially for TN, and the Mississippi River Basin sites). However, the difference also may be the result of chemical/biological post-depositional remobilization that could reduce bed sediment-associated nutrient concentrations. As a result, regardless of the cause, the carbon and nutrient concentrations/annual fluxes determined from the < 63 µm fraction of the bed sediments collected and analyzed during this study should be viewed as minimums.

5 Conclusions

Based on the three studies cited, it is possible to draw some conclusions about the efficacy of using bed sediment-associated major/trace element and nutrient concentrations in water quality studies and monitoring programs. It does appear as if bed sediment chemical data, particularly that generated from the < 63 µm fraction of the upper 1 cm, can be used for a variety of water quality studies and monitoring programs provided that short term variations and/or temporal immediacy are not of primary concern, e.g.: for reconnaissance surveys to identify potential monitoring sites and/or "hotspots" (Utoy Creek); to establish historical reconstructions of the water quality/geochemical history of an area (Lake CDA and the Spokane River); and as surrogates for determining the mean/median concentrations of suspended sediment-associated trace/major elements and nutrients (National Coastal Survey). In the latter case, bed sediment-surrogates may provide a means of establishing long term-trends that could be associated with decadal-long processes such as climate change, without having to resort to much more resource intensive intra- and interannual sampling and analysis programs.

Data availability. Individual data are available from the author(s) on request.

Competing interests. The authors declare that they have no conflict of interest.

Table 2. Comparison between long-term suspended sediment-associated chemical levels with the < 63 µm fraction of bed sediment-associated chemical levels collected during National Coastal Survey.

	Al	Sb	As	Ba	Be	Cd	Cr	Co	Cu	Fe	Pb	Li	Mn	Hg	Mo	Ni	TP	Se	Ag	Sr	V	Zn	Ti	TOC	TC	TN
Sample Name	(%)	(mg kg^{-1})								(%)	(mg kg^{-1})												(%)			
Mississippi River @ Belle Chasse, LA*																										
n	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	1	9	9	9	9	6	9	9
Min.	2.4	0.4	5.6	310	0.8	0.5	40	6	21	1.3	13	24	800	0.01	2	30	890	0.5	<0.5	160	39	91	0.12	1.4	1.9	0.20
Med.	5.8	0.8	9.5	580	1.7	0.7	65	13	24	2.7	24	38	1200	0.05	2	34	1200	1.2	NA	210	83	120	0.35	2.0	2.9	0.45
Max.	6.7	0.9	11	620	2.1	2.2	73	18	27	3.4	31	46	1600	0.11	5	42	5000	2.4	0.7	360	98	150	0.44	3.2	13	1.9
Bed Sediment	6.1	0.8	11	620	1.7	0.9	60	15	23	3.0	28	37	1300	0.08	2	32	1000	0.7	<0.5	150	91	110	0.44	1.4	1.8	0.16
Wax Lake Outlet @ Calumet, LA*																										
n	9	9	9	9	9	9	9	9	9	9	9	9	9	7	9	9	9	9	3	9	9	9	9	9	9	9
Min.	2.2	0.3	4.1	360	0.6	0.3	25	4	11	1.1	7	17	370	0.04	1	13	800	0.4	<0.5	140	37	48	0.11	1.0	1.6	0.20
Med.	6.5	0.8	10	590	1.9	0.5	71	14	24	3.2	24	42	1300	0.07	2	36	1100	1.0	NA	200	95	120	0.38	2.2	2.8	0.40
Max.	7.1	0.9	12	620	2.2	0.6	79	16	37	3.5	26	54	1400	0.15	3	40	1500	1.4	0.9	410	110	150	0.41	3.6	12	0.61
Bed Sediment	6.4	0.8	10	630	1.9	0.5	62	16	23	3.3	26	41	1500	0.05	1	35	980	0.6	<0.5	140	97	120	0.44	1.3	1.7	0.14
Lower Atchafalaya River @ Morgan City, LA*																										
n	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	9	2	9	9	9	9	9	9	9
Min.	2.3	0.2	4.7	370	0.7	0.3	26	4	11	1.2	7	18	500	0.02	1	14	790	0.5	<0.5	150	37	54	0.11	0.9	1.6	0.20
Med.	6.5	0.8	11	580	1.9	0.4	69	14	23	3.4	22	43	1300	0.05	2	35	1100	1.1	NA	230	98	120	0.37	2.2	3.0	0.40
Max.	7.4	0.8	12	650	2.3	0.6	82	17	34	3.7	27	56	1600	0.11	3	42	1300	1.5	0.5	420	110	150	0.43	3.0	11	0.56
Bed Sediment	5.7	0.7	8.1	650	1.5	0.3	52	11	16	2.5	20	30	890	0.02	1	24	760	0.4	<0.5	140	77	81	0.42	0.8	1.1	0.08
Rio Grande nr Brownsville, TX																										
n	40	40	40	40	39	39	39	40	40	40	40	40	40	38	7	39	40	40	11	40	40	40	40	34	35	7
Min.	1.9	0.5	7.0	85	0.5	0.1	16	3	8	0.9	10	22	650	0.04	2	12	800	0.4	<0.5	340	27	49	0.09	1.6	3.1	0.21
Med.	5.0	1.6	10	400	1.4	0.4	37	7	17	2.3	25	44	1500	0.11	3	22	1200	0.7	NA	660	65	110	0.23	2.8	5.0	0.30
Max.	6.8	9.8	16	490	2.0	3.4	160	13	41	5.8	150	51	4100	0.42	21	88	1800	2.6	2.6	1500	90	250	0.33	4.1	9.7	0.65
Bed Sediment	5.7	0.9	7.5	480	1.7	0.3	37	8	14	2.7	19	37	490	0.05	1	18	640	0.3	<0.5	390	74	86	0.32	0.6	3.3	0.08
Colorado River @ N.I.B. above Morelos Dam nr Andrade, CA																										
n	25	25	25	25	25	21	25	25	25	25	25	25	25	16	7	25	25	25	2	25	25	25	25	14	14	2
Min.	2.0	0.3	4.4	160	0.6	0.1	36	3	8	1.1	8	21	490	0.02	6	20	400	0.3	<0.5	310	25	39	0.12	0.4	0.9	0.32
Med.	4.2	1.5	7.1	510	1.0	0.2	65	8	16	2.0	18	30	1400	0.04	8	38	700	0.9	NA	470	54	62	0.27	1.1	1.9	0.41
Max.	5.2	29	26	610	1.7	1.7	120	12	41	3.0	43	54	4500	0.26	11	76	1400	3.4	3.6	910	71	140	0.36	2.2	3.9	0.50
Bed Sediment	4.3	1.2	13	680	1.3	0.4	46	10	38	2.5	34	37	4100	0.09	2	25	980	1.2	0.5	490	65	91	0.31	1.6	3.8	0.25
Columbia River near Beaver Army Terminal, OR																										
n	101	101	101	101	101	99	95	101	101	101	101	101	101	93	31	95	101	101	8	101	101	101	101	89	97	33
Min.	5.4	0.2	2.6	380	1.0	0.1	21	9	34	2.8	7	15	590	0.02	1	15	860	0.1	<0.5	240	63	69	0.31	0.5	0.6	0.06
Med.	7.8	0.9	7.1	540	1.4	0.7	58	18	50	4.3	22	26	1100	0.07	2	36	1300	0.4	NA	320	110	160	0.50	2.2	2.3	0.32
Max.	9.5	50	19	670	2.0	1.5	200	22	150	5.6	85	100	2100	0.30	25	75	2600	1.1	1.4	450	150	340	0.66	6.1	6.2	1.1
Bed Sediment	8.0	1.0	9.6	690	1.8	1.2	61	14	47	3.4	66	26	450	0.06	<1	25	580	0.3	<0.5	320	140	220	0.65	1.5	1.6	0.14

NA – not available

* The small numbers of samples for these sites are due to a change in the most downstream sampling location for the Mississippi River from St. Francisville to Belle Chasse, and on the Atchafalaya

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