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Spatial variation of metals in bed sediments of an urban drainage basin, Hawaii

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Abstract Fish from Manoa Stream, Hawaii, have consistently shown the highest Pb concentrations and some of the highest Cu contents in nation-wide surveys by the National Contaminant Biomonitoring Program (NCBP). To investigate the source(s) of these trace metals a detailed systematic programme was instituted to collect bed sediment samples from 5.8 km of the Manoa Stream. Bed material <63 µm was examined for a variety of elements; those reported here include Al, Cu, Ni, Pb and Zn. All statistical analyses and contaminant indices point to minor anthropogenic contamination for Cu and Zn; mineralogical control for Ni; and a very strong contamination signal for Pb. Maximum Pb concentrations (up to 1080 mg kg^{-1}) were associated with anthropogenic material dumping in minor tributaries, storm sewer sediments and sediments in the "lower" section of the basin.

INTRODUCTION

Metal pollution of aquatic environments is a critical area of inquiry. By examining aquatic sediments significant advances have been made over the last decade in detecting metal pollution sources, locating metal sinks and deciphering chronologies of pollution inputs. Horowitz (1991) states that the strong association of numerous trace elements with sediment (suspended plus bed sediments) indicates that the distribution, transportation and availability of these can not be evaluated intelligently solely through the sampling and analysis of the dissolved phase. Bed sediments are not only a sink for trace metals but they are also a source of resuspended sediment and under changing environmental conditions, sediment bound pollutants may be chemically remobilized and enter the water column or food chain.

BACKGROUND

Few studies have been conducted to examine anthropogenic inputs of trace metals to the Hawaiian environment. Exceptions on the island of Oahu include studies by Ashwood *et al.* (1989), Fu *et al.* (1989), DeCarlo & Spencer (1995) and McMurtry *et al.* (1995). To date, no detailed examinations have been conducted in Hawaii to characterize the spatial distribution of trace metals in fluvial bed sediments. This is somewhat surprising given the results from nation-wide metal testing in freshwater fish in lakes and rivers in the USA. Data from the National Contaminant Biomonitoring Program (NCBP) have consistently found Manoa Stream to have the highest Pb and some of the highest Cu contents in whole fish samples (Schmitt &

Brumbaugh, 1990). Out of 315 fish samples from 109 stations in 1984, three species from Manoa ranked 1, 2 and 5 in the nation for Pb and ranked 3, 6 and 7 for Cu.

The objectives of this paper are to examine in detail the geochemical signature and spatial distribution patterns of selected trace metals in the <63 µm fraction of bed sediment from an urban river system. It is assumed that such a study will answer questions related to potential source areas of contamination and provide an explanation for the highest Pb concentrations in fish of any stream in the USA since the early 1970s.

STUDY AREA

The Manoa basin is located in southeast Oahu (Fig. 1) and is drained by a third-order perennial stream; with a planimetric area of about 15 km^2. Over a distance of less than 8 km the annual rainfall increases from about 100 cm near the basin outlet to 400 cm in the headwaters. Manoa Stream is designated as Inland Waters-Class 2 in the Hawaii Administration Rules, Title 11: waters that are to be protected for recreational purposes, propagation of fish, shellfish and aquatic life, agricultural and industrial water supplies, shipping and navigation. Conservation and undeveloped lands account for about 55–60% of the basin, commercial developments about 2–3%, education and parks about 10%, and the remainder is residential area. Based on land use, traffic intensity, anthropogenic activities, size and frequency of storm sewer outlets draining directly into the stream, Manoa was divided into four separate zones. In the headwaters an "Undisturbed" zone encompasses 15 sites (numbers 102 to 116), i.e. 5.1–5.8 km from the basin outlet. The second zone is designated "Residential" and includes sites 54 to 101 ($n = 49$, including tributary and storm drain samples) between 2.7 and 5.1 km. The next zone is termed "Commercial-Institutional" and includes 42 samples (sites 16 to 53) from 0.8 to 2.7 km; this zone is characterized by increased traffic, the most significant commercial development in the basin, and a variety of educational institutes including the University of Hawaii. The "Lower" zone includes 17 samples (sites 0 to 15) and below Dole Street bridge and the main portion of the University of Hawaii.

MATERIALS AND METHODS

Bed sediment samples were systematically collected at 50 m intervals from the stream outlet, totalling 117 sites over a distance of 5.8 km, during baseflow on three successive days (Fig. 1). Samples from three locations could not be collected due to channelization. In addition to the main stream channel sites, samples from two minor tributaries were collected as they entered the main channel, and samples from the mouth of seven storm drains entering the main channel.

A clear 10 cm internal diameter Plexiglas tube sampler was used to collect three separate 5 cm cores at each sampling site. Cores were composited, double bagged and mixed thoroughly prior to transport to the laboratory. All samples were cold stored at about 5°C prior to processing. Samples were then placed in acid washed beakers and dried for 48 h at 40°C and dry-sieved through acid washed nylon sieves. The <63 µm fraction was examined since this has been the most common size range analysed for trace metals.

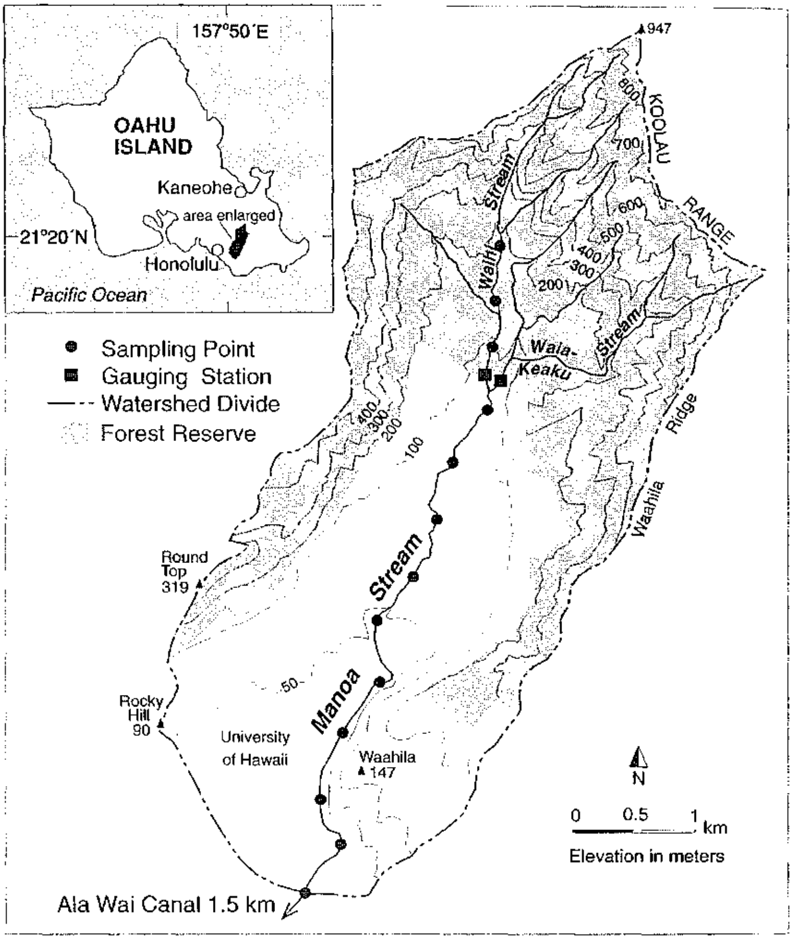

Fig. 1 Map of Manoa Stream drainage basin, Oahu, Hawaii.

Samples were ground in a mixer-mill with a tungsten carbide bowl and balls, for 5 min and then digested with HNO_3, $HClO_4$ and HF to dryness overnight on a hot plate. The residue was solubilized with HCl and diluted to volume. Elements (except Pb) were determined by inductively coupled plasma-atomic emission spectrometry and Pb by flame atomic absorption spectrometry. Quality control procedures included analysis of 17 replicates of SRM 2704 collected from Buffalo River, New York. Precision for Al, Cu, Ni, Pb and Zn were ≤5%, with accuracies between ± 5% for Al, Cu, Pb and Zn, but +14.0% for Ni.

Pollution index computations

To assess the anthropogenic contribution to each site a modified index of geoaccumulation (MI_{geo}) was calculated:

$$MI_{geo} = \log_2\left[\frac{C_n}{1.5 \cdot BE_n}\right] \quad (1)$$

where C_n is the concentration of element n; BE_n is the best estimate of element n in the Koolau Basalts or the area influenced by the Sugarloaf volcanic flow. The BE_n values for the Koolau Basalts in Table 1 were applied to bed sediment sample sites 56 to 116 (from 2.8 to 5.8 km upstream of the basin outlet), below this the Sugarloaf flow outcropped. Therefore for sites 0 to 55 (0 to 2.8 km) the BE_n values were weighted according to an assumed 50% contribution of Koolau Basalts and 50% Honolulu Volcanics. The factor 1.5 in equation (1) is used because of the possible variations of the baseline (BE_n) data due to lithogenic effects (c.f. Salomons & Forstner, 1984). The following descriptive classification is given for the index of geo-accumulation by Forstner *et al.* (1990): <0 = practically unpolluted; 0–1 = unpolluted to moderately polluted; 1–2 = moderately polluted; 2–3 = moderately to strongly polluted; 3–4 = strongly polluted; 4–5 = strong to very strongly polluted; and >5 very strongly polluted.

An enrichment ratio (ER_n) for a given element n was computed using Al as the conservative element because this was the most accurately and the most precisely measured major element; also it is the most commonly used normalizing element in the geochemical literature. The equation is defined as:

$$ER_n = \frac{\left[\dfrac{C_n Sample}{C_{Al} Sample}\right]}{\left[\dfrac{BE_n Background}{BE_{Al} Background}\right]} \tag{2}$$

where C_n is as before; C_{Al} is the aluminium concentration in the <63 μm bed sediment fraction; BE_n *Background* is as before; and BE_{Al} *Background* is the best estimate of the Al concentration in the Koolau Basalts or Honolulu Volcanics (Table 1). There is no accepted ranking or categorization of the degree of pollution based on the enrichment ratio methodology; thus a preliminary five-category system is proposed: (1) *ER* <2 = depletion to minimal enrichment, suggestive of no or minimal pollution; (2) *ER* 2–5 = moderate enrichment, suggestive of moderate pollution; (3) *ER* 5–20 = significant enrichment, suggestive of a significant pollution signal; (4) *ER* 20–40 = very highly enriched, indicating a very strong pollution signal; and (5) extremely enriched, indicating an extreme pollution signal.

Table 1 Best estimates of baseline metal concentrations (mg kg^{-1}, except Al%) for the Koolau Basalts and the Honolulu Volcanics (Sugarloaf Flow) in Manoa drainage basin.

Element	Koolau Basalts*	Honolulu Volcanics*
Al	7.72	5.83
Cu	100	100
Ni	190	240
Pb	6	7
Zn	110	180

* Sources: Taylor (1964), Heinrichs *et al.* (1980), Wilkinson & Stolz (1983), Roden *et al.* (1984), Frey *et al.* (1994).

Statistical analyses

A variety of descriptive statistics will be presented to describe concentration and pollution index data; this will allow comparisons to be made between data in this study, those previously published, and those to be conducted in the future. Spatial variation in Pb pollution indices were examined using the robust lowess technique (Cleveland & Devlin, 1988). One-way analysis of variance (ANOVA) was conducted to examine differences in metal concentrations or pollution indices on $\log_{10}$ transformed data for the four zones. If a significant F-ratio (Prob $\leq \alpha = 0.05$) was found *post-hoc* multiple comparison testing was performed using Fisher's protected least significant difference test.

RESULTS AND DISCUSSION

During the measurement period the pH of baseflow ranged from 7.5 to 7.9, with a median of 7.6. These values are similar to others previously reported for Manoa Stream (e.g. Ikeno, 1996). Metals in the solution phase were not analysed during this study because previously published studies (e.g. Yim & Dugan, 1975; Hasan, 1991) indicated low levels for Cu, Ni, Pb and Zn; typically ≤ 40 $\mu g\ l^{-1}$.

Trace element concentrations in bed sediments

Bed sediment data are plotted for metals in the four zones of Manoa Stream (Fig. 2). Nickel is the only element in Fig. 2 that exhibits the highest concentration in the "Undisturbed" section of Manoa. This may suggest that Ni is primarily controlled by basin mineralogy supporting DeCarlo and Spencer's (1995) statement for mineralogical control of Ni in the nearby Ala Wai Canal sediments. Lead and Zn exhibit a pattern of significantly lower values in the Undisturbed headwater section and highest in the Lower zone near the basin outlet (Fig. 2 and Table 2). Such a pattern

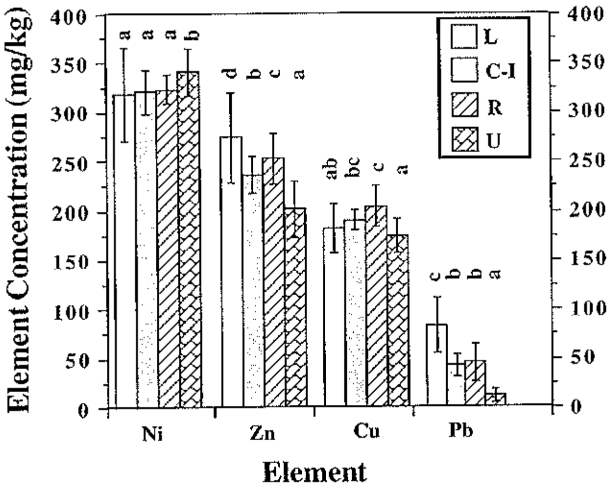

Fig. 2 Minor element geometric mean concentrations (± one standard deviation) for bed sediment (<63 μm) samples from Manoa Stream. The four zones are: L = Lower; C-I = Commercial-Institutional; R = Residential; and U = Undisturbed. For a given element bars with the same letter are **not** significantly different at $\alpha = 0.05$.

Table 2 Descriptive statistics for selected metal concentrations in Manoa Stream bed sediments for four zones (all concentrations are mg kg^{-1}, except Al which is %).

	Metal/Zone	Mean ± SD	Geo Mean	Median	25%	75%	Min	Max
Al*	Lower	8.94 ± 0.35	8.93	8.98	8.82	9.10	8.05	9.47
	Comm-Inst	9.24 ± 0.57	9.22	9.16	8.74	9.67	8.26	10.65
	Residential	9.48 ± 0.54	9.47	9.44	9.24	9.76	8.05	10.90
	Undisturbed	9.60 ± 0.44	9.59	9.56	9.42	9.86	8.72	10.65
Cu*	Lower	188 ± 29	186	181	177	198	133	258
	Comm-Inst	193 ± 20	192	191	183	199	170	300
	Residential	206 ± 22	205	199	189	220	172	267
	Undisturbed	174 ± 17	174	173	162	187	150	212
Ni*	Lower	317 ± 44	314	307	292	315	278	439
	Comm-Inst	316 ± 29	314	315	303	329	216	408
	Residential	324 ± 15	324	323	317	331	289	360
	Undisturbed	341 ± 24	340	341	320	360	302	379
Pb*	Lower	97 ± 31	93	95	72	104	59	175
	Comm-Inst	53 ± 24	49	50	45	59	10	175
	Residential	84 ± 153	56	52	40	71	17	1078
	Undisturbed	13 ± 7	11	10	10	14	5	28
Zn*	Lower	274 ± 42	271	276	242	286	210	368
	Comm-Inst	246 ± 40	243	242	226	248	180	444
	Residential	262 ± 48	258	252	240	267	208	510
	Undisturbed	202 ± 29	200	196	181	223	162	268

* The number of samples analysed per zone were: Lower = 17; Commercial-Institutional = 42; Residential = 49; and Undisturbed = 15.

may reflect increased vehicle traffic, and increased road runoff of vehicle-associated metals as we proceed from the Undisturbed headwater area to the highly impacted Lower stream reach. Several studies have found strong associations between bed sediment metal concentrations (Pb and Zn) and vehicle traffic density (e.g. Van Hassel *et al.*, 1980). McMurtry et *al.* (1995) estimated that motor vehicle sources account for at least 97% of the Pb burden of Oahu coastal sediments with the remaining sources (volcanic emissions and exogenic atmospheric transport) at most contributing 3%. Median Pb concentration in the Undisturbed zone = 10 mg kg^{-1} (Table 2), which was about 10 times less than in the Lower section of the basin. Mid-basin concentrations were intermediate between the upper and lower reaches. Lead in the Undisturbed zone was only slightly greater than the baseline of 6 mg kg^{-1} adopted for the Koolau Basalts (Table 1). This minor increase, and also that displayed by Zn, may be accounted for by one or more of the following sources: atmospheric deposition of locally derived exhaust emissions, natural deposition from volcanic emanations, or from deposition of exogenic aerosols transported from distant locations to the Hawaiian islands. Regardless of the source, Pb and Zn contributions to the Undisturbed portion of the basin are inconsequential when compared to the concentrations of these elements in sediments in the heavily impacted downstream zones.

All the available data supports a strong anthropogenic signal for Pb and to a lesser degree Zn. The spatial patterns are strongly suggestive of vehicle contributions even though Pb in gasoline has recently been phased out in the USA. It is clear that the Pb and Zn levels present in Manoa Stream sediment reflect physical remobilization through erosion of polluted soils and sediments from basin slopes, and transport via surface runoff to the stream. Once sediments enter Manoa Stream the sorbed metals undergo a circuitous path downstream as material is deposited, re-suspended and transported to eventually be deposited in the Ala Wai Canal.

Modified Index of Geoaccumulation (MI_{geo})

From Table 3 it is clear that the Pb MI_{geo} values are the most distinct for the trace metals examined in Manoa Stream. About 55% of the sample sites were classified as moderately or strongly polluted (i.e. MI_{geo} 2–3). Four of the five samples classified as strong to very strongly polluted (MI_{geo} 4–5, and >5) were associated with storm drain outlets and a tributary outlet influenced by metal pipes dumped within the stream net. These Pb MI_{geo} data combined with concentration data suggest a continued flux of Pb sorbed to sediments entering Manoa Stream from urban runoff.

The smoothed Pb $\mathrm{MI}_{\mathrm{geo}}$ pattern is shown in Fig. 3, with storm drain outlet and tributary samples identified with arrows. Median Pb MI_{geo} values for each zone were 3.3 (Lower) > 2.5 (Residential) = 2.4 (Commercial-Institutional) > 0.1 (Undisturbed), with ANOVA indicating no significant differences between the two mid-basin zones at $\alpha = 0.05$.

Enrichment ratios (ER_n)

Enrichment ratio data indicate Pb contamination of bed sediments in Manoa Stream is the most pronounced amongst the trace metals investigated (Table 4). Approximately 67% of the sites sampled were classified as having a significant degree of Pb pollution (ER_{Pb} 5–20), and three sites were considered to be extremely polluted (ER_{Pb} >40). The spatial variation in bed sediment ER_{Pb} values is shown in Fig. 4, with arrows identifying sediments associated with storm drain outlets and small ephemeral tributaries. Analysis of variance indicated that ER_{Pb} values for each of the four zones where statistically different at $\alpha = 0.05$, with the following median values: 11.4 (Lower) > 6.7 (Residential) > 5.8 (Commercial-Institutional) > 1.3 (Undisturbed). As with all other data presented there is strong support for Pb enrichment, and the anthropogenic signal is considered to be related with urban runoff carrying automobile associated pollutants.

Table 3 Modified Index of Geoaccumulation percentages in each pollution category* for trace metals in Manoa Stream bed sediments (<63 μm). (Values in parentheses represent the actual number of samples, total no. = 123.)

	<0	0–1	1–2	2–3	3–4	4–5	>5	Median†	Min	Max
Cu	1.6 (2)	97.6 (120)	0.8 (1)	-	-	-	-	0.35	–0.17	1.00
Ni	32.5 (40)	67.5 (83)	-	-	-	-	-	0.11	–0.58	0.44
Pb	3.3 (4)	9.7 (12)	12.2 (15)	55.3 (68)	15.4 (19)	3.3 (4)	0.8 (1)	2.42	–0.94	6.90
Zn	5.7 (7)	90.2 (111)	4.1 (5)	-	-	-	-	0.38	–0.27	1.63

* Modified Index of Geoaccumulation (MI_{geo}) values were determined using equation (1), with values <0 = practically unpolluted; 0–1 = unpolluted to moderately polluted; 1–2 = moderately polluted; 2–3 = moderately or strongly polluted; 3–4 = strongly polluted; 4–5 = strong to very strongly polluted; >5 = very strongly polluted (Forstner *et al.*, 1990).

† Median, minimum and maximum represent actual MI_{geo} values and not percentages.

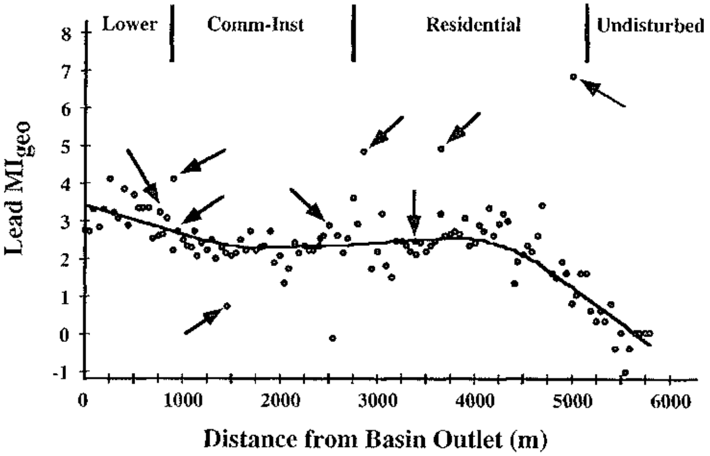

Fig. 3 Spatial variation in the lead modified index of geoaccumulation (MI_{geo}) for 123 Manoa Stream bed sediment samples. The major line through the data represents a lowess smoothed trace. Arrows identify either storm drain outlet or minor tributary outlet samples.

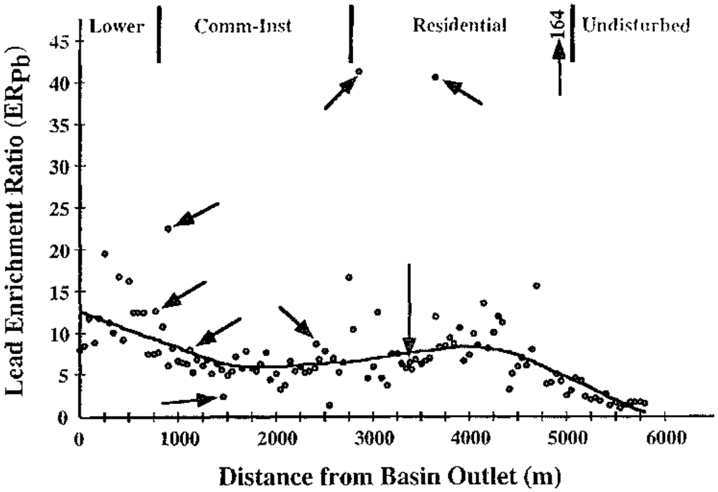

Fig. 4 Spatial variation in the lead enrichment ratio (ER_{Pb}) for 123 Manoa Stream bed sediment samples. The major line through the data represents a lowess smoothed trace. Arrows identify either storm drain outlet or minor tributary outlet samples.

Table 4 Enrichment ratio percentages in each pollution category* for trace metals in Manoa Stream bed sediments (<63 μm). (Values in parentheses represent the actual number of samples, total no. = 123.)

	ER <2	*ER* 2–5	*ER* 5–20	*ER* 20–40	*ER* >40	Median†	Min	Max
Cu	92.7 (114)	7.3 (9)	-	-	-	1.45	1.06	2.46
Ni	100.0 (123)	-	-	-	-	1.29	0.81	1.62
Pb	10.6 (13)	19.5 (24)	66.7 (82)	0.8 (1)	2.4 (3)	6.23	0.63	163.80
Zn	87.8 (108)	12.2 (15)	-	-	-	1.48	0.81	4.18

* Enrichment ratio (ER_n) values were determined using equation (2), with *ER* values <2 representing sites with no or minimal pollution; 2–5 represents sites moderately polluted; 5–20 represents sites with significant pollution; 20–40 represents sites with very strong pollution; and >40 represents those sites that are extremely polluted.

† Median, minimum and maximum represent actual ER values and not percentages.

CONCLUSIONS

Stream bed sediments provide an important archive for examining trace metals and identifying anthropogenic pollution signals when detailed spatial investigations are conducted. Concentration data, the modified index of geoaccumulation and enrichment ratios all point to Manoa Stream bed sediments (<63 μm) as being primarily contaminated by Pb. Sediments with the highest Pb concentrations were located in the Lower zone of the basin (0 to 0.8 km upstream of the basin outlet) and also associated with storm drain outlets and minor tributaries. The lowest Pb concentrations, near background levels, were measured in sediments of the Undisturbed zone in the Waihi sub-basin, 5.1–5.8 km upstream of the basin outlet. Data for Cu and Zn indicated some degree of anthropogenic enhancement for samples sites between 0 and 5.1 km upstream of the basin outlet, but none showed the magnitude of enhancement exhibited by Pb.

The primary source of Pb and Zn contamination in Manoa Stream is the automobile; including vehicle wear, tire wear, fluid output, and exhaust emissions. Many of these sources exist today in Manoa Valley, but the contemporaneous contribution of Pb from vehicle emissions is negligible. However, the high Pb vehicle fluxes associated with emissions in the 1960s–1980s, have been stored in the basin and are now being remobilized by surface erosion processes and contributed to the stream channel network. It is clear that the high Pb concentrations found in whole fish samples collected from Manoa Stream as part of the NCBP reflect ingestion of Pb-associated with bed sediment. Lead, and other trace metals, ingested via contaminated sediment will undergo desorption and bioaccumulation. To date the biological toxicity of sediments in Manoa Stream have not been assessed. Thus, future work should consider examining biological toxicity with attention paid to the synergism between anthropogenically contributed metals, and not just Pb.

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