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Preliminary Evaluation of Heavy Metal Contamination and Source Identification in Kuala Lumpur SMART Stormwater Pond Sediments Using Pb Isotopic Signature

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Abstract: Uncontrolled urbanization and growing industrialization are major sources of pollutants that affect the urban stormwater quality and, therefore, the receiving aquatic environment. The concentrations of heavy metals (As, Cd, Cr, Cu, Fe, Ni, Pb, and Zn), and Pb isotope ratios in surface sediment samples obtained from SMART holding and storage ponds located in Kuala Lumpur were investigated using inductively coupled plasma-mass spectrometry (ICP-MS). The highest metal concentrations were found at the SMART holding pond (SHP), the first recipient of urban stormwater runoff from the SMART system catchment area. As, Cd, Pb, and Zn are the dominant metal contaminants in the sediments of both SMART ponds, with values exceeding the average shale values. According to contamination indices applied to evaluate the environmental risk caused by heavy metals, As had the highest values among the metals examined, denoting moderate contamination. Hence, it can frequently cause harmful effects on the sediment-living species. The Pb isotope ratios ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$) indicated that coal combustion was the foremost source of anthropogenic Pb in the sediments of both SMART ponds. The control of coal combustion and sites undergoing intensive human activities should be given priority in the foreseeable future.

Keywords: SMART stormwater ponds; heavy metals contamination; contamination indices; Pb isotope ratios; tracing contamination sources

1. Introduction

The expansion and densification of urban areas lead to an increase in impermeable surfaces, which, during rainfall events, lead to an increase in stormwater runoff volumes [1,2]. The stormwater management and road tunnel (SMART) catchment is located in Klang Valley which is one of the most densely populated and fastest urbanized catchments in

Malaysia, with various industries and a high level of traffic [3–5]. Thus, SMART catchment is constantly exposed to the large amounts of pollutants that accumulate on various impervious surfaces from different sources such as activity-related (e.g., vehicle emissions), land cover related (buildings and infrastructure), and atmospheric deposition [6]. Stormwater ponds have been certified as one of the best management practices in urban areas that can control flooding by temporary storage of stormwater runoff, and trapping sediment and decayed plant and animal debris in runoff water, and other contaminants associated with settleable particulates [7,8]. The treatment of urban stormwater by stormwater ponds is known to be effective for both water quality improvement and storm flow management, and it has become widely implemented over the last two decades [9]. Through three mechanisms, these ponds are capable of eliminating numerous groups of contaminants such as trace metals, pesticides, polycyclic aromatic hydrocarbons, and nutrients metal pollutants. These include binding to sediments, particulates, and soluble organics or precipitation as insoluble salts, or uptake by plants and bacteria [9,10]. These ponds open up new avenues for addressing water quality and promoting infiltration via landscape-friendly technologies and multidisciplinary collaboration [11].

However, stormwater ponds are inhabited or used by various organisms, and thus, the accumulation of these pollutants in the ponds may cause toxicity to the organisms [7,12]. Stormwater pond sediments serve as an excellent reservoir and ultimate sink for a wide variety of chemicals like heavy metals [13,14]. As it flows over different impervious surfaces, stormwater runoff can capture and carry large pollutants, such as heavy metals, nutrients, hydrocarbons, and organic chemicals, and discharges them into the local receiving rivers or ponds [15–17]. Among stormwater pollutants discharged into the ponds via stormwater runoff, heavy metals are a particular concern due to their toxic effects on living organisms, accumulation in aquatic habitats, and slow removal rate [18–20]. Once discharged into stormwater ponds, heavy metals can be strongly accumulated in the sediments and biomagnified along aquatic food chains, eventually resulting in human health issues such as liver damage, cardiovascular diseases, and renal failure [21–23] Sediment-associated metals pose a direct risk to detrital and sediment-ingesting benthic species and may also represent long-term sources of contamination to higher tropic levels [24]. Moreover, heavy metals may adversely affect various metabolic processes in developing fish (embryos or larvae), resulting in developmental retardation, hatching delay, morphological and functional deformities, or death of the most sensitive individuals [25,26].

Heavy metal released into the aquatic environment quickly binds to particles and finally settles in bottom sediments [27]. Only a small part of free metal ions remains dissolved in water due to adsorption, hydrolysis, and co-precipitation, whereas a considerable number of them are deposited in the sediment [28,29]. Therefore, heavy metal concentrations in sediments are frequently three to five orders of magnitude higher than those in the overlying water [30,31]. Sediments serve as a contamination indicator and a screening tool for detecting historical and recent pollution in the nearby environment [32]. It is therefore considered to be an appropriate indicator of heavy metal contamination in SMART ponds receiving stormwater runoff from differing industrial, commercial, residential and highway land uses. Hence, studding the sediments becomes more considerable due to its associated metals which pose a direct high risk to aquatic organisms compared to water in the ponds. However, the heavy metal contamination in stormwater and fish from SMART ponds were obviously discussed in our previous publication [6].

Metals come from a number of sources in urban settings, which can be difficult to pinpoint. Isotopic fingerprinting, based on stable isotope ratios, is a powerful tool for tracing the source of metals in various environmental media [33,34]. The isotopic ratios or signatures of different ore deposits and anthropogenic sources are distinct and do not alter during the physicochemical processes of mining, smelting, and manufacturing [35,36]. Therefore, Pb isotope ratios are increasingly being used to determine geochemical origins, classify the major sources of anthropogenic Pb, and determine the mechanisms by which Pb reaches the environment [33,37].

SMART is a unique and innovative project designed primarily to divert excess stormwater from the upper Klang/Ampang basin away from entering the Kuala Lumpur city centre [6]. During a major storm event, the SMART ponds receive a massive amount of stormwater runoff which can pick up a wide range of contaminants, including heavy metals from various impermeable surfaces throughout the catchment region. Over time, these heavy metals can accumulate in the pond sediments and become available for bio-accumulation in resident organisms, thereby posing a health hazard to the organisms concerned and the consumers.

Since the SMART Project began operations in 2007, no studies of heavy metal contamination and tracing of its sources in SMART holding and storage pond sediments have been published. Hence, to fill this gap this paper aims to: (i) determine the heavy metal contamination levels in the surface sediment of both ponds to provide preliminary baseline data for SMART ponds contamination control; (ii) assess the environmental risk associated with selected metals using certain contamination indexes such as the enrichment factor (EF), the pollution load index (PLI), the Geoaccumulation index (Igeo), and contamination factor (CF) sediment quality guidelines (SQGs); and (iii) trace the source of these metals released to the SMART ponds using the Pb isotopic fingerprinting technique.

This study is significant as the first effort to document the extent of heavy metal contamination of accumulated sediment within the stormwater SMART ponds receiving stormwater runoff from various land uses within their urban catchment. It also highlights the key source that can contribute to heavy metal pollution to the ponds in order to control heavy metal emissions from their origins. In addition, the environmental risk assessment of sediments from SMART ponds serving as aquatic habitat is essential for the conservation and restoration of biodiversity in the urban environment.

2. Materials and Methods

2.1. Study Area and the SMART System Descriptions

SMART is a major national project planned primarily to relieve frequent floods in Kuala Lumpur and minimize traffic congestion between Kuala Lumpur city center and the main southern gateway. The main components of this project are the SHP in Kampung Berembang, the SMART tunnel, and the SMART storage pond (SSP) in Taman Desa (Figure 1).

During major storm events, when the cumulative discharge reaches 70 m^3/s at the confluence of the Klang and Ampang rivers, the SMART system diverts excessive floodwater into the SHP through eight inflow gates. The SHP temporarily retains diverted floodwater until it reaches a particular permitted level before being released to the SSP through the SMART tunnel (bypass tunnel) [38]. The SHP, the head of the SMART project, is situated at 3°09′51.8″ N, 101°44′35.6″ E and has an area of 8 hectares and a capacity of 0.6 million m^3. On the other hand, the SSP is located downstream of the city center (Taman Desa) (3°06′03.5″ N 101°41′25.1″ E) and covers 23 hectares with a capacity of 1.4 million m^3. The SMART tunnel which connects the SHP and SSP, consists of a 9.7 km stormwater bypass tunnel and a 3 km dual-deck motorway in the middle. It has a total flood storage capacity of 1.0 million m^3. However, The Klang/Ampang catchment is situated in the upper area of Kuala Lumpur, Klang Valley. It covers a total area of about 121 km^2. The region receives an average of 2790.7 mm of annual precipitation, while the annual mean temperature is 27 °C, with monthly mean temperatures ranging from 23 to 32 °C [39].

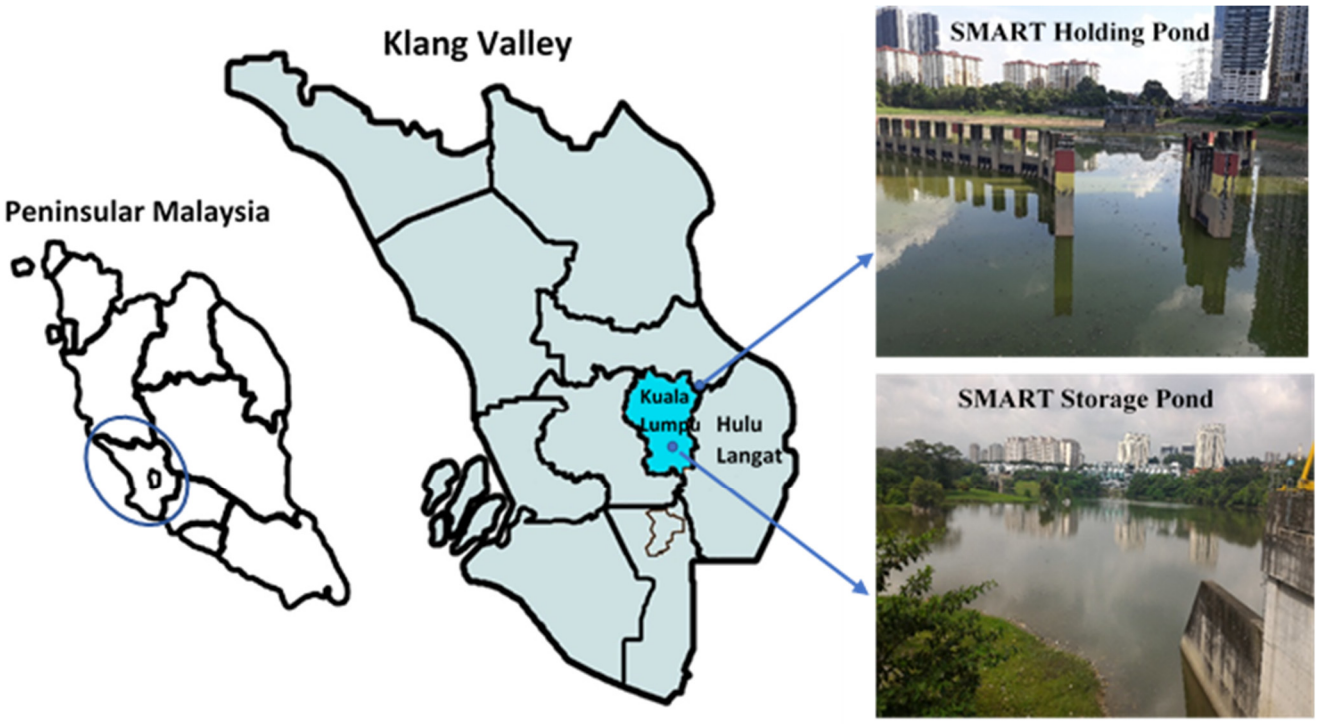

Figure 1. Map of the study area located within the Klang Valley, Malaysia.

2.2. Sediment Sampling and Analysis

Superficial sediments (0–5 cm) were sampled from each pond in triplicate using a pre-cleaned plastic scoop and an Ekman-Birge grab sampler, depending on the accessibility to the sediments. Sediment samples were obtained within 24 h after stormwater was diverted to SMART ponds on January 14, March 19 and May 5, 2017. The samples were then wrapped in acid-washed, pre-labelled polyethylene bags and put in an icebox before being transferred to the laboratory for analysis. Once arrived at the research facility, the sediment samples were naturally air-dried at ambient temperature until the sediment weight was constant and then screened through a 2 mm sieve to remove stones and other unwanted materials. Samples were then agitated, homogenized, and placed in sealed and acid-washed polyethylene bags before chemical analysis [40].

pH in sediment samples was measured in a suspension of 1:2.5 sediment to Mill-Q water ratio after shaking for 4 h using a calibrated pH meter (model 230A+) [41]. The total organic matter (TOM) of each sample was measured using the loss on ignition technique (LOI). Samples were oven-dried at 105 °C, weighed and ignited in a muffle furnace at 550 °C for 8 h [42,43]. Ignited samples were then cooled and re-weighed to determine the amount of organic matter lost.

For heavy metal analysis, approximately 0.5 g of each dried sample (fraction < 63 µg) was digested with a mixture of HNO_3–H_3ClO_4 (3/1, *v*/*v*). The sample solution was heated on a digestion block at 40 °C for 1 h and then at 140 °C for 4 h [44,45]. After cooling, the digested samples were diluted with 0.15 M HNO_3 to a final volume of 40 mL and filtered through nylon syringe filters 0.45 µm and 13 mm (Figure 2). The filtered samples were then analysed using an ICP-MS Perkin Elmer ELAN DRC-e (Sciex, Mundelein, IL, USA). Following the complete analysis protocol without samples, blanks were prepared to ensure that the samples were free of contaminants.

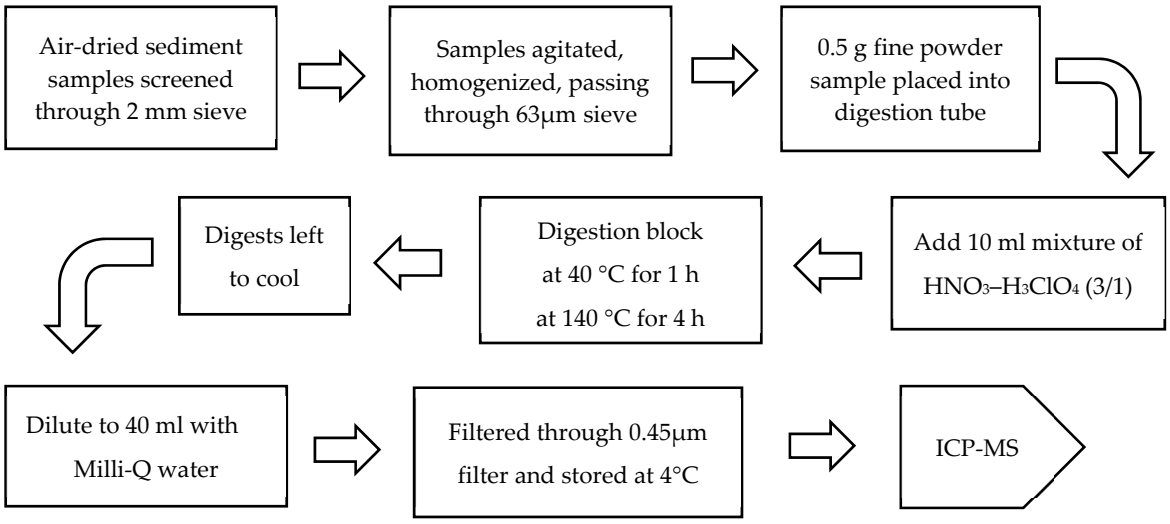

Figure 2. Graphical diagram of the preparation and determination of heavy metals in sediment samples.

2.3. Quality Assurance and Quality Control

Before the experiments, the laboratory glassware and the apparatus were adequately cleaned by immersion in HNO3 (20% v/v) for two days and then rinsed several times with Mili-Q water to avoid contamination. Ultrapure water 18 MΩ purified with a Mili-Q system (Millipore, St. Louis, MI, USA) was used to prepare standard and diluted samples and blank. To ensure the analytical method accuracy, multiple levels of calibration of standard solutions were prepared from Multi-Element Calibration Standard 3 (Pure Plus PerkinElmer, Shelton, CT, USA). Moreover, a certified reference standard SRM 1646a (estuarine sediment, NIST), was used to test the analytical and instrument accuracy of the method (Supplementary Materials Table S1).

2.4. Pb Isotopic Analysis

The air-dried and ground sediments samples were heated at 550 °C for 30 min in a muffle furnace to demolish the organic substance contained in the samples [34,46]. Approximately 0.5 g of each sediment sample (fraction < 63μg) was sequentially placed in a pre-cleaned glass tube with 4 mL suprapure HNO_3 (65%, Merck) and 12 mL suprapure HCL (30%, Merck) at room temperature overnight (~12 h) [47,48]. The tubes were then placed in a digestion block, preheated at 40 °C for 1 h, and then post heated at 140 °C for 4 h. After digestion, the solutions obtained were filtered through 0.45 μm nylon syringe filters and brought to a volume of 40 mL with Milli-Q water. After measuring the concentration of Pb in sample digests, the solutions were adjusted to a lead concentration of approximately 20 μg/L because Pb isotope ratios are affected by high lead concentrations [49].

The mass discrimination (mass bias) correction was performed using standard reference material (NIST SRM 981 common lead isotopic material). A NIST SRM 981 solution with a concentration of 20 μg/l was analyzed at the start and end of each run and after every 10 samples [50]. All the Pb isotope measurements (^{206}Pb, ^{207}Pb, and ^{208}Pb) were determined using the ICP-MS. The average measured ratios of ^{208}Pb/^{207}Pb and ^{206}Pb/^{207}Pb of the SRM 981 were 2.3710 and 1.0832, respectively. These values were entirely in agreement with certified standard values of 2.3704 and 1.0930, respectively. At the same time, each sample measure had a relative standard deviation of less than 0.5%.

2.5. Sediment Contamination Indices

2.5.1. Enrichment Factor (EF)

The enrichment factor approach can help distinguish between natural and anthropogenic metal sources in the environment [51]. It can also be used to assess the extent

of sediment contamination. The EF for each metal was calculated using the following definition [52]:

$$EF = (Cx/Fe)\ sample)/(Cx/Fe)\ background$$

where (Cx/Fe) sample is the concentration ratio of the target metal (Cx) to Fe in the examined sediments, and (Cx/Fe) background is the same ratio in the reference material value. In this study, average shale values were selected as suitable background reference values [53]. Fe was chosen as the normalizing metal in sediments as it is a major element in sediment and its background concentration does not change significantly [54]. The EF values were interpreted as proposed by Chen et al. [51] (Supplementary Material Table S2).

2.5.2. Geoaccumulation Index (Igeo)

The geoaccumulation index presented by Muller [55] was used to assess the heavy metal contamination of the sediment samples by comparing present metal concentrations to background concentrations:

$$Igeo = \log2\ [Cn/1.5Bn]$$

where Cn is the observed metal concentration in the sediment, Bn is the background value of a provided metal (n) and constant 1.5 is used to reduce potential shifts in background values due to lithological variations [56]. Igeo values are classified by Muller [55] into seven classes based on contamination levels, as illustrated in (Supplementary Material Table S2).

2.5.3. Contamination Factor (CF)

The contamination factor is calculated by dividing the metal concentration in the investigated sediment (Cx sample) by the background values (Cx background). It is expressed as follows:

$$CF = (Cx\ sample)/(Cx\ background)$$

The values of CF are classified into four classes as proposed by Hakanson [57]: very high degree of contamination (CF > 6), considerable degree of contamination (3 < CF < 6), moderate degree of contamination (1 < CF < 3) and low degree of contamination (CF < 1).

2.5.4. Pollution Load Index (PLI)

The PLI developed by Tomlinson et al. [58] was employed to evaluate the pollution extents of heavy metals in sediment based on baseline metal concentrations. It is calculated by applying the following formula:

$$PLI = (CF1 \times CF2 \times CF3 \times \ldots \times CFn)^{1/n}$$

where CF is the contamination factor CF = (Cx sample)/(Cx background) and n is the number of the investigated metals (8 heavy metals in our study). The PLI values above 1 suggest metal pollution, while PLI values below 1 suggest no metal pollution [58].

2.5.5. Sediment Quality Guidelines (SQGs)

Although chemical analysis of heavy metal content in sediments is crucial for assessing sediment quality, the SQGs assess the extent to which the status of metals associated with sediments may affect aquatic life and are intended to help understand sediment quality [59,60]. Numerous SQGs were established to correlate chemical concentrations in sediment with their possible effects on benthic organisms. Analytical results of this study were compared to the SQGs developed by MacDonald et al. [61], which include a probable effect concentration (PEC) and threshold effect concentration (TEC) in order to ascertain the true extent of sediment contamination by heavy metals and to forecast possible biological effects on aquatic life in the SMART ponds.

2.6. Statistical Analysis

Data were statistically evaluated with IBM SPSS Statistics version 25. Before parametric testing, all data were tested for both homogeneity of variance (Levene's test) and normal distribution (Shapiro–Wilk test). Analysis of variance (ANOVA) and Tukey's honestly significant difference (HSD) analysis was applied to detect the meaningful differences in heavy metal concentrations among the three sampling periods for both SMART ponds, while an independent t-test was used to identify significant differences in all metals studied between the two sites (SHP and SSP). Pearson's correlation coefficients were used to calculate the strength of the correlations among the heavy metals, pH, and TOM in the sediments of both SMART ponds.

3. Results and discussion

3.1. Heavy Metals and Sediment Characteristics

The concentrations of the heavy metals, pH, and TOM in the sediment samples detected during three different sampling periods are listed in Table 1. The average pH of the sediment samples varied from 6.56 to 6.88 at SHP and from 6.27 to 6.83 at SSP, indicating the slight acidity of the sediments in both ponds. Changes in pH contribute significantly to the solubility of metal concentrations [62]. However, the influence of pH on the solubility of observed heavy metals in both pond samples is not apparent. The TOM of the sediment samples varies between the ponds. The TOM of SHP sediment samples ranged from 5.45 to 8.30% (mean: 6.85%), while SSP sediment samples had a TOM of 3.82 to 5.77%, with an average of 4.85%. The relatively high TOM value in SHP could be due to the fact that it is the first recipient of stormwater runoff receiving high loads of organic pollutants from industrial and residential areas in its catchment. However, it is well established that TOM plays a role in distributing heavy metals in sediments [51,63].

Table 1. Heavy metal concentrations (μg g^{-1}, Fe%), pH and total organic (TOC%) contents in surface sediments of the SMART ponds.

Pond	Period		pH	TOM	As	Cd	Cr	Cu	Fe	Ni	Pb	Zn
HSP	January	Mean	6.78	6.786	47.29	0.38	20.05	48.38	2.428	5.216	40.1	204.2
		SD	0.05	0.162	3.131	0.075	1.016	4.006	0.198	0.4	3.619	15.6
	March	Mean	6.88	8.303	41.58	0.523	18.57	53.26	3.248	7.082	43.78	194.5
		SD	0.04	0.383	3.29	0.058	2.78	3.902	0.343	0.678	3.005	24.46
	May	Mean	6.56	5.445	34.69	0.345	15.56	42.99	2.689	5.452	34.13	174.3
		SD	0.07	0.306	3.518	0.026	1.525	3.822	0.339	0.561	3.259	14.29
SSP	January	Mean	6.83	4.966	42.85	0.36	23.48	30.68	2.292	3.863	37.83	183.1
		SD	0.06	0.134	2.881	0.099	1.814	2.775	0.182	0.285	3.153	15.04
	March	Mean	6.27	5.768	33.61	0.349	20.52	34.32	2.809	5.424	35.63	156.5
		SD	0.09	0.21	1.487	0.03	2.648	1.946	0.324	0.963	2.668	16.32
	May	Mean	6.53	3.822	27.63	0.239	18.21	32.95	2.324	4.964	30.77	144
		SD	0.07	0.265	3.227	0.022	1.307	2.928	0.227	0.527	1.627	17.47
	Average shale [a]		-	-	13	0.3	90	45	4.72	68	20	95
	SQGs [b]	TEC	-	-	9.79	0.99	43.4	31.6	-	22.7	35.8	121
		PEC	-	-	33	4.98	111	149	-	48.6	128	459

[a] Average shale [53]. [b] Sediment Quality Guidelines [61].

pH and TOM values vary significantly ($p < 0.01$) between the sampling periods due to the difference in organic matter content and chemical substances loads in each storm event diverted to SMART ponds. Similarly, TOM exhibited a significant difference ($p < 0.01$) between both ponds, while values of pH indicated no significant difference ($p > 0.05$) between the ponds.

The concentrations of heavy metals in SMART pond sediments ranged from 27.6–47.3 μg/g for As, 0.24–0.52 μg/g for Cd, 18.2–23.48 μg/g for Cr, 30.7–53.3 μg/g for Cu, 2.23–3.25 μg/g for Fe (%), 2.83–7.08 μg/g for Ni, 30.8–43.8 μg/g for Pb and 144–204 μg/g for Zn. The overall concentration of the heavy metals increased in the

order of Cd < Ni < Cr < Pb < As < Cu < Zn < Fe. Concentrations of all metals studied, except Cr, were lower in SSP samples than in SHP. This is most likely due to the pre-treatment occurring in the SHP, which may have removed a large portion of these metals, both particulate loads through desilting and dissolved loads by in-pond processes before the stormwater is released to the SSP. In contrast, the higher concentration of Cr observed in SSP could be attributed to dust built-up on the SMART Tunnel motorway surface, which washes off to SSP during the stormwater diversion event.

Statistically, all heavy metal concentrations showed a significant difference ($p < 0.05$) between sampling periods, possibly due to the differences in metal loading in stormwater runoff at each diversion event. A comparison between sampling sites (SHP and SSP) showed that there were significant differences in all heavy metals ($p < 0.05$), except for As, Cr and Fe ($p > 0.05$).

Except for Cr, Fe, and Ni along with Cd in SSP sediment collected in May, all sediment samples had higher concentrations of As, Cd, Pb, and Zn than the average shale values, indicating that these metals are specifically enriched in the study area, and their origins are predominantly from anthropogenic sources and deposited from the atmosphere to the watershed [64]. The sources of such metals in urban catchments could arise from various anthropogenic activities, such as fossil fuel combustion emissions, industrial activities, and traffic-related sources (e.g., brake linings, tyre wear, and auto-catalysts) [65,66]. However, the concentrations of Cu concentrations were higher than average shale values only in the SHP samples obtained in January and March, as shown in Table 1. It was reported that a significant proportion of Cu in urban stormwater runoff is related to vehicles by brake pads and wear on tires [67].

With respect to SQGs, the obtained results reveal that the values of As, Cu, Pb, and Zn were greater than the TEC values in all sediment samples, except for Pb (30.8 μg/g) at SSP. The concentrations of Cd, Cr, Cu, Pb, Ni, and Zn were all below the PEC in all samples, suggesting that harmful effects on aquatic life are rarely expected to occur. On the other hand, As concentrations exceeded the PEC limit of 33 μg/g in 100% of surface sediments, implying that adverse effects on sediment-living species are more likely to occur frequently. However, since no TEC and PEC values for Fe were available, we could not compare our findings to the SQGs.

Pearson's correlation coefficients among the measured parameters (Table 2) indicated that there were no significant relationships between TOM and pH ($p > 0.05$; r = 0.451). pH is positively correlated with Cd, Cu and Pb ($p < 0.05$), and significantly correlated with As and Zn ($p < 0.01$; r = 0.659–0.614). TOM had a significant correlation with all metals tested ($p < 0.01$), except Cr ($p > 0.05$), suggesting that TOM may raise the concentrations of these metals in the sediments of both SMART ponds.

Table 2. Pearson correlation of heavy metals concentrations with sediments properties in SMART ponds.

	pH	TOM%	As	Cd	Cr	Cu	Fe	Ni	Pb	Zn
pH	1									
TOM%	0.451	1								
As	0.659 **	0.617 **	1							
Cd	0.523 *	0.789 **	0.523 *	1						
Cr	0.209	−0.052	0.038	0.572	1					
Cu	0.472 *	0.819 **	0.441	0.568 *	−0.298	1				
Fe	0.025	0.642 **	0.057	0.549 *	−0.452	0.389	1			
Ni	0.068	0.647 **	−0.025	0.436	−0.348	0.761 **	0.565 *	1		
Pb	0.554 *	0.796 **	0.785 **	0.698 **	0.244	0.573 *	0.399	0.416	1	
Zn	0.614 **	0.671 **	0.839 **	0.538 *	0.097	0.481 *	0.234	−0.011	0.633 **	1

* Correlation is significant at the 0.05 level (2-tailed). ** Correlation is significant at the 0.01 level (2-tailed).

However, the correlation coefficients between heavy metals indicated a positive correlation of Cd ($p < 0.05$; r = 0.523), Pb ($p < 0.01$; r = 0.785), and Zn ($p < 0.01$; r = 0.839) with As, while Cu correlated positively with Cd ($p < 0.05$; r = 0.568), Ni ($p < 0.01$; r = 0.761), Pb ($p < 0.05$; r = 0.573), and Zn ($p < 0.05$; r = 0.481), indicating that they might have derived from the same sources. Among the studied metals, only Cd and Ni had a significant connection with Fe ($p < 0.05$), indicating that Fe could have originated from lithogenic sources. Cr however, had no noticeable correlation with any of other metals ($p > 0.05$) suggesting that it may derived from a different source. Cr, on the other hand, had no discernible correlation with any of the other metals ($p > 0.05$), implying that it is generated from a natural source, as evidenced by the EF analysis results in the following section. It was also found that the Cr is significantly bonded to organic matter [68].

Data on heavy metal contamination in sediments from stormwater systems in Malaysia are very limited. In this case, we have compared the findings of this study with the few such studies available in the literature (Table 3). In general, the levels of Cd, Cr, Cu, Fe, Ni, and Pb in this study were found to be lower than those reported from stormwater ponds in other countries [9,16,69–71], except for Zn at stormwater management ponds (Maryland, USA) [69]. In contrast, the extent of As contamination in sediments from both ponds was relatively higher than that in the stormwater ponds (Florida) [16], and stormwater retention ponds (Calgary, Canada) [9], probably due to increased human activities within the SMART system catchment, which can contribute to high levels of As to stormwater runoff diverted to the ponds during the heavy rain events. However, the type and intensity of land use, type of asphalt, rainfall intensity and duration, storm frequency, traffic volume, and antecedent dry periods are all factors that affect the concentration of metals in stormwater runoff [72,73]. Therefore, differences in metal concentrations between our study and others could be attributed to any combination of these factors.

Table 3. Comparison of heavy metals in sediments of different stormwater ponds of the world (μg g^{-1}).

Location	As	Cd	Cr	Cu	Fe	Ni	Pb	Zn	References
HSP	35–47	0.4–0.5	16–20	43–53	2.4–3.3	5.2–7.1	34–44	174–204	This study
SSP	28–43	0.2–0.4	18–24	31–34.	2.3–2.8	3.9–5.4	31–38	144–183	This study
Stormwater ponds, Florida	0.6–25	5.3	5.8–175	4.5–90	-	5.4–40	6–196	5–711	[16]
Stormwater management ponds, Maryland, USA	-	0.1–3.2	14–176	19–86	-	12–286	10–55	40–58	[69]
Stormwater management ponds, Toronto, Canada	-	<0.01–3.5	21–195	28–351	1.3–4	13–35	18–166	114–1260	[70]
Stormwater retention ponds, Calgary, Canada	4–9	0.5–64	13–35	12–89	-	14–38	10–106	58–1220	[9]
Retention pond, Nantes, France	-	2.5–2.7	69–89	95–123	-	50–53	178–205	781–1040	[71]
Retention pond (Wissous) Paris, France	-	4–5	260–429	288–343	-	333–648	296–346	1226–1725	[71]

3.2. Sediment Contamination Indices

The EF values of heavy metals in the surface sediments are tabulated in Table 4. EF values $0.05 \leq EF \leq 1.5$ imply that the metals are completely delivered from natural weathering processes or crustal materials [74], whereas values above 1.5 indicate an anthropogenic source of heavy metals [29]. In this study, the EF values for As, Cd, Fe, Pb, and Zn in all sediment samples were greater than 1.5, indicating anthropogenic input of these metals. However, among all analysed metals, As had the highest EF values and a moderate to moderately severe enrichment. In urban areas, anthropogenic sources of arsenic are multiple, including traffic exhaust, smelters and fuel combustion, coal-fired power station, wood combustion, the use of pesticides and fertilizers, and waste incineration [66,75–77]. One or more of these sources can contribute significantly to arsenic deposition on road surfaces in

the SMART watershed, and eventually conveyed by stormwater runoff and discharged over the pond sediments. The EF values revealed that most of the sediment samples in both ponds were moderately enriched by Pb and Zn (EF > 3), while all sediment samples were minorly enriched by Cd and Cu (EF < 3). There was no enrichment (EF < 2) of Cr and Ni in the SMART pond sediments.

The results of Igeo revealed that the SMART ponds sediments were classified as practically unpolluted (class 0, Igeo $\leq$ 0) with respect to most studied metals (Cd, Cr, Cu, Fe, and Ni), suggesting that the surface sediments are unpolluted by such metals (Table 4). Furthermore, the sediments of SMART ponds were classified as unpolluted to moderately polluted (Class 1, 0 < Igeo < 1) by Pb and Zn in all sediment samples. Otherwise, the Igeo values of As were classified as unpolluted to moderately and moderately polluted, indicating that As may present more significant risks to the ponds than other metals and should be subject to regular monitoring. However, none of the metals can be categorized under heavily to extremely polluted.

Table 4. Enrichment Factor (EF) and Geoaccumulation Index (Igeo) values in surface sediments of SMART ponds.

		As		Cd		Cr		Cu		Fe		Ni		Pb		Zn	
Pond	**Period**	**EF**	**Igeo**	**EF**	**Igeo**	**EF**	**Igeo**	**EF**	**Igeo**	**EF**	**Igeo**	**EF**	**Igeo**	**EF**	**Igeo**	**EF**	**Igeo**
HSP	January	7.07	1.28	2.46	−0.25	0.43	−2.75	2.09	−0.48	1	−1.54	0.15	−4.29	3.9	0.42	4.18	0.52
	March	4.65	1.09	2.53	0.22	0.3	−2.86	1.72	−0.34	1	−1.12	0.15	−3.86	3.18	0.55	2.98	0.45
	May	4.68	0.85	2.02	−0.39	0.30	−3.12	1.68	−0.65	1	−1.4	0.14	−4.24	3	0.19	3.22	0.29
	Total mean	5.47	1.07	2.34	−0.32	0.35	−2.91	1.83	−0.65	1	−1.35	0.15	−4.05	3.36	0.38	3.46	0.42
SSP	January	6.79	1.14	2.47	−0.32	0.54	−2.52	1.40	−1.14	1	−1.63	0.12	−4.72	3.9	0.34	3.97	0.36
	March	4.34	0.79	1.95	−0.37	0.38	−2.72	1.28	−0.98	1	−1.33	0.13	−4.24	2.99	0.25	2.77	0.14
	May	4.32	0.50	1.62	−0.91	0.41	−2.89	1.49	−1.04	1	−1.61	0.15	−4.35	3.12	0.04	3.08	0.02
	Total mean	5.15	0.81	2.02	−0.53	0.44	−2.62	1.39	−1.09	1	−1.52	0.13	−4.3	3.34	0.21	3.27	0.17

The CF value for Cu, Cr, Fe, and Ni indicates a low degree of contamination (CF > 1) (Table 5). The CF values for As in 50% of sediments sampled indicated considerable contamination (3 < CF < 6), while the remaining samples showed moderate contamination. However, for Cd, Pb, and Zn, moderate contamination was found in most samples from both ponds. Overall, the CF values for all metals decreased in the following order: As > Zn > Pb > Cd > Cu > Fe > Cr > Ni. However, the calculated PLI values for heavy metals in the sediment ranged from 0.72 to 0.90 at SHP and from 0.62 to 0.72 at SSP, as shown in Table 5. PLI values were found to be generally low (PLI < 1), suggesting that the presence of metals studied in both pond sediments had no negative impacts.

Table 5. Contamination factors (CF) and pollution load index (PLI) of heavy metals in surface sediments of the SMART ponds.

Pond	**Sampling Occasion**	**Contamination Factors (CF)**								**PLI**
		As	**Cd**	**Cr**	**Cu**	**Fe**	**Ni**	**Pb**	**Zn**	
SHP	January	3.638	1.266	0.223	1.075	0.515	0.077	2.005	2.149	0.81
	March	3.198	1.744	0.206	1.184	0.688	0.104	2.189	2.047	0.90
	May	2.669	1.149	0.173	0.955	0.570	0.080	1.707	1.835	0.72
	Total mean	3.168	1.386	0.201	1.071	0.591	0.087	1.967	2.010	0.81
SSP	January	3.296	1.201	0.261	0.682	0.486	0.057	1.891	1.927	0.72
	March	2.586	1.162	0.228	0.763	0.495	0.08	1.781	1.648	0.72
	May	2.126	0.797	0.202	0.732	0.492	0.073	1.538	1.516	0.62
	Total mean	2.669	1.054	0.230	0.726	0.491	0.07	1.737	1.697	0.69

3.3. Tracing Metal-Pollution Sources Using Pb Isotopic Signatures

Pb isotopes were analyzed for possible sources of heavy metals, mainly Pb, in sediments from SMART ponds. The Pb isotopic ratios in the SHP sediments varied from 1.178 to 1.184 for $^{206}Pb/^{207}Pb$ and 2.449 to 2.493 for $^{208}Pb/^{207}Pb$, with an average of 1.181 and 2.466, respectively. In the SSP sediments, the corresponding $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$

ratios are ranged from 1.190 to 1.193 and from 2.469 to 2.493, with an average of 1.191 and 2.485, respectively (Supplementary Material Table S3).

For specific source determination of Pb deposited in the SMART pond sediments, the detected isotope ratios $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ were compared to those of source-related materials as presented in Supplementary Material, Figure 3 and Table S3. These Pb related materials included natural sources, coal combustion, automobile exhaust (leaded and unleaded), and aerosols from regional cities in Indonesia, Vietnam, China, Thailand, India, and Australia. However, $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ isotope ratios in the studied sediments were closer to those of natural sources and coal combustion found in Indonesia, Australia and Shanghai, but differ from the corresponding ratios in aerosols and vehicle emissions. Malaysia imported over 22.2 million metric tonnes of coal in 2015, accounting for 99.2% of the coal supply utilized to generate electricity. The import of coal continues to rise each year, reaching 27 million metric tonnes in 2017 [78], 66.9% imports from Indonesia and 19.6% from Australia [79]. Hence, we anticipate that the majority of coals consumed in the region came from Indonesia and Australia, as Indonesian coals had Pb isotope ratios of $^{206}Pb/^{207}Pb = 1.184$ and $^{208}Pb/^{207}Pb = 2.477$, while Australian coals had $^{206}Pb/^{207}Pb = 1.206$ and $^{208}Pb/^{207}Pb = 2.488$ [80].

The Malaysian government took the first step in protecting the country's air quality in April 1998 by banning the sale and use of leaded gasoline. By the year 2000, leaded gasoline was phased entirely out in Malaysia [81]. Thus, the Pb isotopic ratios in this study are distinct from the range of leaded vehicle exhaust, suggesting that automobile lead is not currently the major component of Pb in the sediments of SMART ponds. It is worth noting that the Pb isotopic ratios in the SMART pond sediments sampled 16 years after Malaysia concluded phasing out leaded gasoline differ somewhat from those recorded in Kuala Lumpur aerosols when leaded gasoline was still in use [82]. It was reported that the Pb contribution from automobile exhaust was reduced with the progressive phasing-out of Pb from gasoline, resulting in a notable increase in the isotopic ratio in the aerosol [83]. The findings suggest that coal combustion-related emissions are the dominant source of the anthropogenic Pb in the sediments of both ponds. Furthermore, correlation analysis (Table 2) revealed statistically significant correlations ($p < 0.05$) between Pb and each of the studied metals, except for Cr, Fe and Ni ($p > 0.05$), suggesting that these metals might have originated from the same source with Pb.

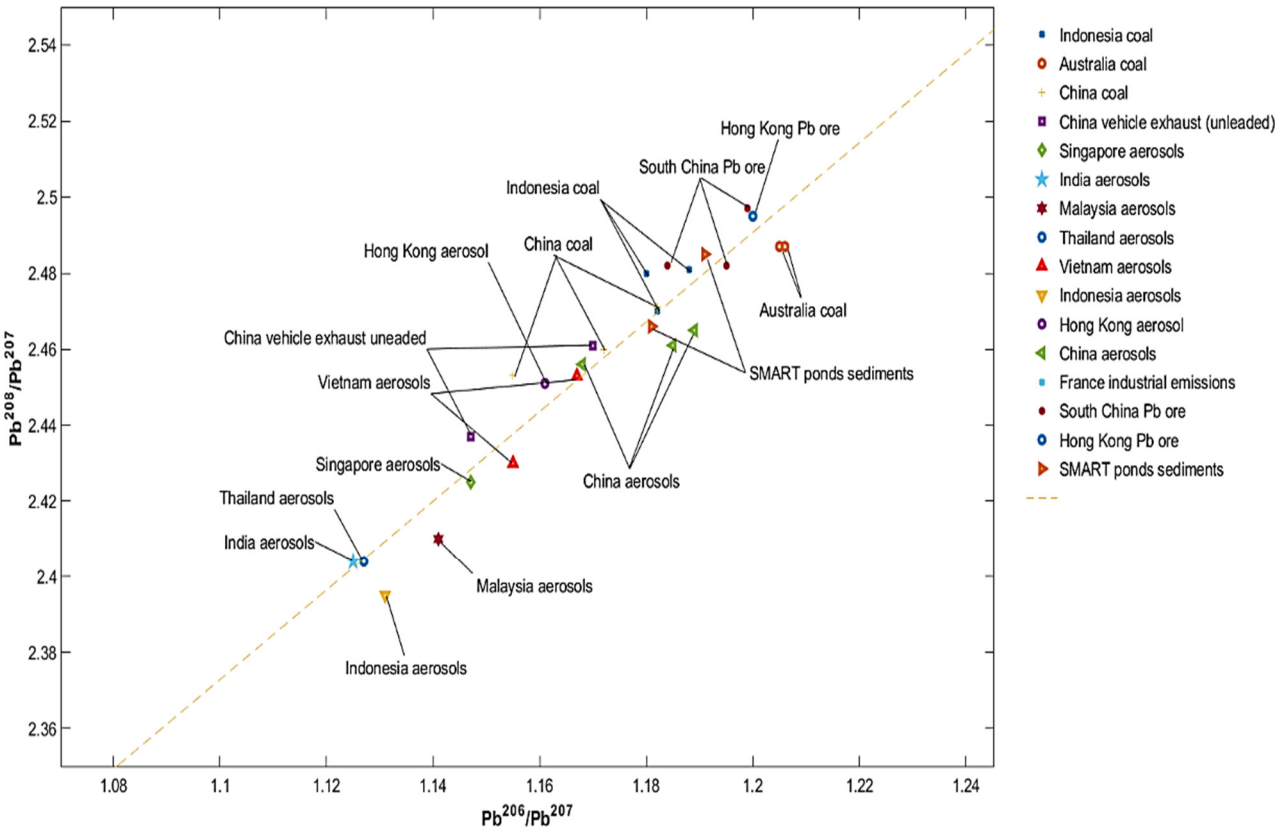

Figure 3. Isotopic composition (208Pb/207Pb vs. 206Pb/207Pb) in sediment samples investigated and relative literature. Data Sources: Coal, including coal combustion, from Díaz-Somoano et al. [80]; Zheng et al. [84]; Mukai et al. [85], Vehicle exhaust, including leaded and unleaded gasoline, Chen et al. [86]; Bi et al. [87], Aerosols, Lee et al. [88]; Kumar et al. [89]; Bollhöfer and Rosman [82]; Lee et al. [90]; Mukai et al. [85], Industrial emissions, Monna et al. [91], Background, Bing-Quan et al. [92]; Lee et al. [90].

4. Conclusions

This study provides the first data on heavy metals contamination levels in SMRT holding and storage ponds sediments, and possible sources of these metals using Pb isotopic fingerprinting. Higher concentrations of all the heavy metals in SHP sediments are ascribed to sedimentation occurring in the pond, which is considered the primary treatment mechanism for metal contamination. Heavy metals concentrations in all sediment samples decrease in the following order: Zn > As > Cu > Pb > Cr > Ni > Fe% > Cd. Comparing the metal concentrations with average shale values shows that sediments from both ponds are contaminated with As, Cd, Pb, and Zn. The SQGs indicate that As concentrations are likely to cause adverse effects on sediment-living species and are expected to occur regularly. The EF results revealed that the SMART pond sediments were moderately enriched with Pb and Zn, and moderately severely enriched with As. The Igeo values suggest that individual metal contamination in the surface sediments might be categorized as practically unpolluted to moderately polluted. CF values for the pond sediments showed moderate to considerable contamination for As, and moderate contamination for Cd, Pb, and Zn. In both ponds, PLI values were generally low (PLI < 1), suggesting a low risk of contamination. The Pb isotope ratios ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$) in the SMART pond sediments indicated that coal combustion is most likely the major source of the Pb in this region. The baseline data obtained through Pb isotopic signatures could lead to further research into identifying heavy metal sources in various environmental media in Peninsular Malaysia. It could also help policymakers in the area develop successful environmental pollution control policies. A follow-up study is suggested to evaluate the chemical fractionation of selected metals in SMART pond sediments to provide more

insights on heavy metals mobility, toxicity, and potential availability for aquatic organisms in the ponds. In addition, sampling intervals at each diverting storm event to the SMART ponds through the years are recommended to provide a comprehensive picture of heavy metal load washed off by stormwater runoff from different land use to the ponds.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/su13169020/s1, Table S1: Observed and certified values of heavy metals for SRM 1646a (μg g^{-1}, Fe%), Table S2: Descriptive classes of the geoaccumulation index (Igeo) and indication of enrichment factor (EF), Table S3: A comparison of Pb isotope ratios from different sources to those in sediment of SMART ponds.

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