

Research Article

Assessment of Heavy Metals in Water, Sediment, and Fishes of a Large Tropical Hydroelectric Dam in Sarawak, Malaysia

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Bakun Hydroelectric Dam in Sarawak is one of the world highest concrete rock filled dams. This paper reports the heavy metals concentrations in water, sediment, and fishes of Bakun Dam. Water and sediment samples were collected from 11 stations and 6 fish species were caught. The samples were digested with open acid digestion and the metals contents were analysed using an atomic absorption spectrophotometer and mercury analyser. The method was validated based on certified reference materials. A higher concentration of Fe and Mn was detected in downstream water with significant longitudinal variation. Cu, Zn, and Hg were present in trace amount. All elements analysed were consistently found in sediment with no risk of contamination. For fish, *Hemibagrus planiceps* was characterised by higher affinity for Hg accumulation. The concentrations detected in all fish species were within the permissible guideline of 0.5 mg/kg. The health risk assessment suggested that *Barbonymus schwanenfeldii*, *Puntiplites waandersii*, *Cylocheilichthys apogon*, and *Hemibagrus planiceps* were characterised by hazard index > 1 implying possible adverse effects. The amount of fish recommended for adults and children was in the range of 500–775 g/week and 33–135 g/week, respectively.

1. Introduction

Dams and reservoirs are mainly built for irrigation, power generation, flood control, and water supply. There are currently more than 58,000 dams built all over the world with China recording the highest number. In Malaysia, there are about 80 existing dams and the number is anticipated to increase due to the escalating demand for electricity [1]. A dam is considered a large dam if it is greater than 15 m with a storage capacity of more than 1 Mm³.

Dams and reservoirs can serve as a sink for accumulation of heavy metals. Their mobility and availability in aquatic environments are primarily controlled by water quality parameters including pH, dissolved oxygen and organic matter content. In anoxic hypolimnion, the ubiquitous Fe³⁺ and Mn⁴⁺ in sediment are readily reduced into dissolving Fe²⁺ and Mn²⁺ leading to elevated concentrations in overlying water [2]. As the elements diffuse upward into oxic level, they are oxidized and reprecipitated. Al is also an element that is naturally abundant in sediment, occurring as insoluble

silicoaluminate. When pH of the water is low, the dissolution of Al is enhanced thereby increasing its concentration in water. Dams and reservoirs also play an important role in facilitating the transportation of heavy metals. When water is released from a dam, resuspension of deposited sediments under high flow rate tends to carry heavy metals downstream. In Peru, the acid mine drainage from adjacent mining district was flushed downstream from Upamayo Dam, contributing to high concentrations of copper (Cu) and zinc (Zn) in sediment [3].

Impoundment of a dam inevitably involves large area of inundation that fosters an environment with accelerated microbial decomposition. This, in turn, may trigger the transformation of inorganic Hg to organic mercury that can be bioaccumulated and biomagnified along the food chain [4]. Fishes affected by mercury accumulation have been found to take 20–30 years to restore to the background level after impoundment [5, 6]. Considering the possible consequences, the geochemical behaviour of heavy metals in dams and reservoirs has been continuously reported worldwide:

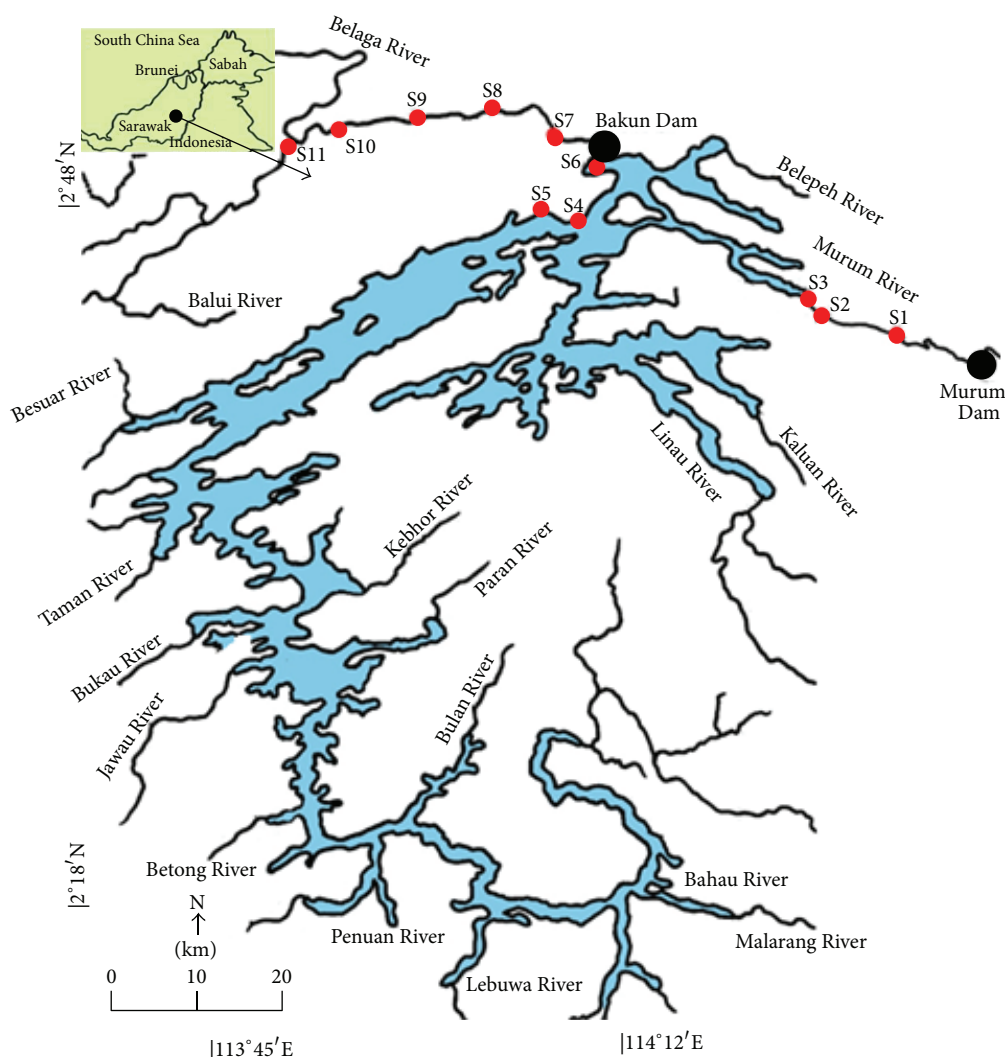


FIGURE 1: The locations of sampling stations.

Dobczycki reservoir [7]; Rybnicki reservoir [8]; Manwan, Dachaoshan, and Nuozhadu dams [9, 10]; Quiberon Bay Water [11]; Dicle and Batman dam [12]; Shahid Rajaee dam [13]; Masinga Dam [14].

Bakun Hydroelectric Dam, situated on the Balui River, Sarawak, Malaysia, is the one of the highest concrete rock filled dams (205 m) in the world with a catchment area of 14,750 km². The dam is built for power generation. It is impounded in 2010 where the flooded area is over 695 km², approximately the size of Singapore [15, 16]. Despite its substantial size and economic potential, the geochemical distribution of heavy metals in the dam is scarcely known. It is revealed that tropical climates of high temperature and high rainfall throughout the year could lead to elevated leaching rates upstream causing the accumulation of heavy metal in reservoirs [17]. Besides, tropical hydroelectric dams are also predicted to be more susceptible to mercury transformation as tropical soil is naturally rich in mercury [18, 19]. This postulation however contradicts the findings of Yingcharoen and Bodaly [20] who conclude that the transformation of Hg

is less severe in warmer areas. As a matter of fact, there are relatively fewer reports on the fate of Hg in tropical reservoirs. Therefore, the objective of this study is to determine the heavy metals concentrations in water, sediment, and fishes of Bakun Hydroelectric Dam.

2. Materials and Methods

2.1. Sampling. Water and sediment samples were collected upstream, in the reservoir, and downstream of Bakun Hydroelectric Reservoir in Nov. 2014 as indicated in Figure 1. Note that Bakun Dam is cascading with a new dam located upstream at the Murum River (Murum Hydroelectric Dam, impounded in 2013). The GPS coordinates of the sampling stations are summarised in Table 1.

Triplicate water samples (1 L) were collected from sub-surface into polyethylene bottles and acidified with 5 mL of 2 M HNO₃. The corresponding pH and dissolved oxygen content were measured *in situ* using YSI multiparameter water quality meter. Submerged sediment at least 30 cm below

TABLE 1: The corresponding GPS coordinates of respective stations.

Sampling station	GPS coordinates	Description
ST1	N02°40'16.1" E114°17'22.0"	Near to the power house of Murum Dam
ST2	N02°41'31.9" E114°11'47.9"	Approximately 10 km away from ST1
ST3	N02°42'28.8" E114°10'54.3"	Near to the reservoir (transitional zone; water begins to be stagnant)
ST4	N02°41'52.1" E114°01'36.1"	Reservoir
ST5	N02°42'22.8" E114°01'14.3"	Reservoir
ST6	N02°44'43.9" E114°02'26.5"	Reservoir
ST7	N02°46'21.7" E114°01'41.4"	Near to the power house of Bakun Dam (4.3 km from Bakun Dam)
ST8	N02°46'37.9" E113°59'23.3"	Right above Metjawah (Lubok Metjawah)
ST9	N02°46'05.2" E113°54'23.0"	Lahanan Long Semuang
ST10	N02°44'36.5" E113°50'21.8"	Above Longhouse Kejaman Neh
ST11	N02°42'02.9" E113°46'46.32"	Below Belaga Town

TABLE 2: The number of samples and corresponding length and weight of various fishes.

Fish species	Local name	Number of individuals	Range of weight (g)	Range of length (cm)
<i>Cyclocheilichthys armatus</i>	Boeng	10	15.7–230.5	9.1–28.1
<i>Barbonymus schwanenfeldii</i>	Tenggadak	16	182–520	12.4–36.5
<i>Puntioplites waandersii</i>	Mengalan	17	59.6–520	18.6–33.4
<i>Pangasius micronemus</i>		7	71.7–520	23.1–39.3
<i>Cyclocheilichthys apogon</i>	Boeng	101	22.8–135	12.5–17.2
<i>Hemibagrus planiceps</i>	Baong	20	74.5–285	23.6–38.9

the water level was collected using a grab sampler and stored in plastic bags. Fish samples were caught using the netting method from the Balui River, Linau River, and Long Baagu. The total length and weight were recorded and the species were identified according to Mohsin and Ambak [21], Kottelat and Whitten [22], Rainboth [23], and Inger and Chin [24]. Table 2 summarises the species and the range of total length and weight. All samples were stored in a cooler box at 4°C for transportation to the laboratory. At the laboratory, the samples were kept at –20°C until further analysis.

2.2. Acid Digestion and Metal Analysis. Water samples were filtered through 0.45 µm membrane filters and digested according to the standard method of APHA [25]. 5 mL of HNO₃ was added to 100 mL of sample and the sample was put to slow boil on a hotplate to 10–20 mL until the solution is clear. The sample was then left to cool, filtered through 0.45 µm membrane filters, and diluted to 100 mL. The metal content analysed is referred to as the acid extractable metals constituting dissolved and weakly sorbed metals on particulates. Sediment samples were air dried. 0.25 g of the air dried sample was digested with HNO₃/HCl (3:1 v/v) until the solution turned colorless. The sample was left to cool, filtered, and diluted to 100 mL. The fish muscle was dissected using a ceramic knife and oven-dried at 100°C for 1 hr [26]. 0.25 g of sample was digested in 6 mL of HNO₃ according to the aforementioned procedure. Note that all glassware

was acid washed before use. The digested samples were subjected to metal analysis including copper (Cu), zinc (Zn), iron (Fe), and manganese (Mn) using an atomic absorption spectrophotometer (Thermo Scientific iCE 3000). The total mercury (Hg) was determined using a mercury analyser (Perkin Elmer, FIMS 400).

2.3. Quality Control and Method Validation. For every batch of digestion, a blank was prepared to monitor contamination and the method efficiency was determined according to the certified reference materials (CRM) of stream sediment (STSD-1) and fish muscle (ERM-BB422). The recovery percentages attained for the reference materials of sediment and fish are between 97–102% and 90–120%, respectively, meeting the acceptable recovery of 80–120% recommended by the Environmental Agency [27].

In this paper, the metal contents were measured according to dry weight to ensure consistencies. Loss of Hg due to dry weight measurement was evaluated based on CRM. CRM at ambient temperature and that dried at 100°C (1 hr) were subjected to digestion and the recovery performance was determined; no statistical different was inferred at 95% significant level.

2.4. Moisture Content. The concentration in wet weight is estimated based on the moisture content for comparison with the guideline of WHO [28], FAO [29], and Malaysia Food

Act [30]. The moisture content was determined based on loss-on-drying approach. The concentration in wet weight is calculated as follows:

$$\begin{aligned} \text{Conc. in wet wt. (mg/kg)} \\ &= \text{Moisture factor} \times \text{conc. in dry wt.} \\ \text{Moisture factor} &= 1 - \frac{\text{wet wt.} - \text{dry wt.}}{\text{wet wt.}} \end{aligned} \quad (1)$$

2.5. Assessment of Contamination Status. The contamination status of sediment was evaluated based on the geoaccumulation index (I_{geo}), contamination factor (CF), and pollution load index (PLI). The contamination factor (CF) is the concentration of a given element, C_{sample} , against the average metal in the world surface rock, $C_{\text{background}}$ (Mn: 750 mg/kg; Hg: 0.4 mg/kg; Fe: 35900 mg/kg; Cu: 32 mg/kg; Zn: 127 mg/kg). The CF value < 1 indicates low level of contamination [31]. The geoaccumulation index, I_{geo} , is used to indicate the enrichment of metals above the baseline concentrations, where $I_{\text{geo}} = \log_2(C_{\text{sample}}/(1.5 \times C_{\text{background}}))$. The sediment is classified as unpolluted if I_{geo} value < 0. As the value increases, it suggests progressive contamination with ($0 < I_{\text{geo}} < 1$), ($1 < I_{\text{geo}} < 2$), ($2 < I_{\text{geo}} < 3$), and ($3 < I_{\text{geo}} < 4$) implying being unpolluted, moderately polluted, and heavily polluted, respectively [32]. The pollution load index (PLI) is calculated as $(CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n}$, where n is the number of metals. The PLI value of >1 suggests that the sediment is polluted whilst PLI < 1 infers unpolluted sediment [33].

2.6. Statistical Analysis. Analysis of Variance (ANOVA) with multiple comparisons using Tukey's test was performed to deduce the significant difference between the means at a significant level of 0.05. The relationship between the two variables was evaluated based on Pearson's correlation analysis. The statistical analysis was performed using SPSS version 20.

3. Results and Discussion

The metal concentrations in water and sediment according to sampling stations are shown in Figure 2. All elements studied are continuously found in sediment corroborating that heavy metals are naturally occurring. In water, Fe and Mn are detected at higher concentrations with an average of 1.43 and 0.15 mg/L, respectively. Fe and Mn are detected at elevated concentrations at ST1-ST2 upstream (average Fe: 5.0 mg/L; Mn: 0.42 mg/L) and ST7-ST11 below the dam (average Fe: 1.12 mg/L; Mn: 0.17 mg/L). In the reservoir at ST3-ST6, the levels of Fe and Mn are considerably low (average Fe: nd; Mn < 0.01 mg/L).

ST1-ST2 and ST7-ST11 are stations below Murum and Bakun Dam suggesting that the downstream water is enriched with Fe and Mn with the former exhibiting higher concentrations. From ST7-ST11, the extractable Fe and Mn are progressively increased with statistical different confirmed between stations ($p < 0.05$). The upsurge of Fe and

Mn in water below the dam has been commonly reported where the concentrations could escalate to above 8 mg/L. However, the levels, according to Knoll et al. [39], usually return to the background thresholds at a distance of approximately 1 km below the dam. The longitudinal variation is likewise reported by Ashby [2] where the peak concentration is detected at 6 km with Fe experiencing relatively greater fluctuations compared to Mn. At Megget Reservoir, Fe and Mn below the dam were also reported above the UK drinking water guideline of 0.20 mg/L and 0.02 mg/L, respectively [40]. The downstream increase of Fe and Mn is likewise registered in dams of T. T. Hue and Binh Dien Province, Vietnam [41]. According to studies, a newly formed reservoir is more susceptible to issues relating to Fe and Mn [42]. The suggestion appears to be well corroborated based on the results from this study; Murum Dam downstream is characterised by higher Fe and Mn (Fe: 5.0 mg/L; Mn: 0.42 mg/L) compared to Bakun Dam (Fe: 1.12 mg/L; Mn: 0.17 mg/L) and that reported in Batang Ai Dam by Sim et al. (Fe: <1.0 mg/L; Mn: <0.1 mg/L) [34].

The high Fe and Mn in downstream water body are possibly attributed to the discharge of anoxic hypolimnion water [43]. According to HDR Engineer [42], the concentration of Mn in hypolimnion could easily reach 2–20 mg/L. Chauhan et al. [44] on the other hand suggest that the considerable amount of Fe and Mn may have associated with corrosion and abrasion from hydro turbine parts underwater. A high concentration of soluble Fe (>0.3 mg/L) could lead to rusty staining while Mn (>0.05 mg/L) causes brownish black precipitates in plumbing fixtures. Fe and Mn are ubiquitous in soil and sediment, present as insoluble Fe^{3+} and Mn^{4+} . Their occurrence is primarily controlled by pH and dissolved oxygen (DO). Under anoxic and low pH environments, the dissolution of Fe and Mn is facilitated. In this study, pH and DO recorded upstream (pH: 5.65–6.12; DO: 2.36–3.61 mg/L) and downstream (pH: 6.19–6.34; DO: 2.37–3.30 mg/L) are lower than those in the reservoir (pH: 7.07–7.21; DO: 5.51–6.50 mg/L) where a significant negative correlation is deduced between pH and the concentrations of Fe and Mn.

In the reservoir, Fe and Mn in subsurface water are negligible. These elements, in contact with water consisting of high dissolved oxygen, tend to be oxidized and precipitated. Cu and Zn are inconsistently detected in water whilst Hg is found in all stations at an average 0.039 $\mu\text{g/L}$, well below the permissible level of 1.0 $\mu\text{g/L}$ [28]. Statistically, no significant difference is established in Hg between stations ($p > 0.05$). The average concentration of Hg in Bakun is relatively lower than that reported in Batang Ai (0.367 $\mu\text{g/L}$) [34].

For sediment, all elements appear to demonstrate similar pattern of distributions. At ST1 and ST7 below the dam, the metal concentrations are relatively lower. The concentration is seen to increase in the subsequent stations suggesting metal deposition as a result of inclining DO in water. The concentration soon ebbs after entering the reservoir of Bakun Dam concurring the findings of Varol [12]. Metal concentration was reported the lowest near the dam wall and gradually increased to reach its maximum at the entrance of the streams

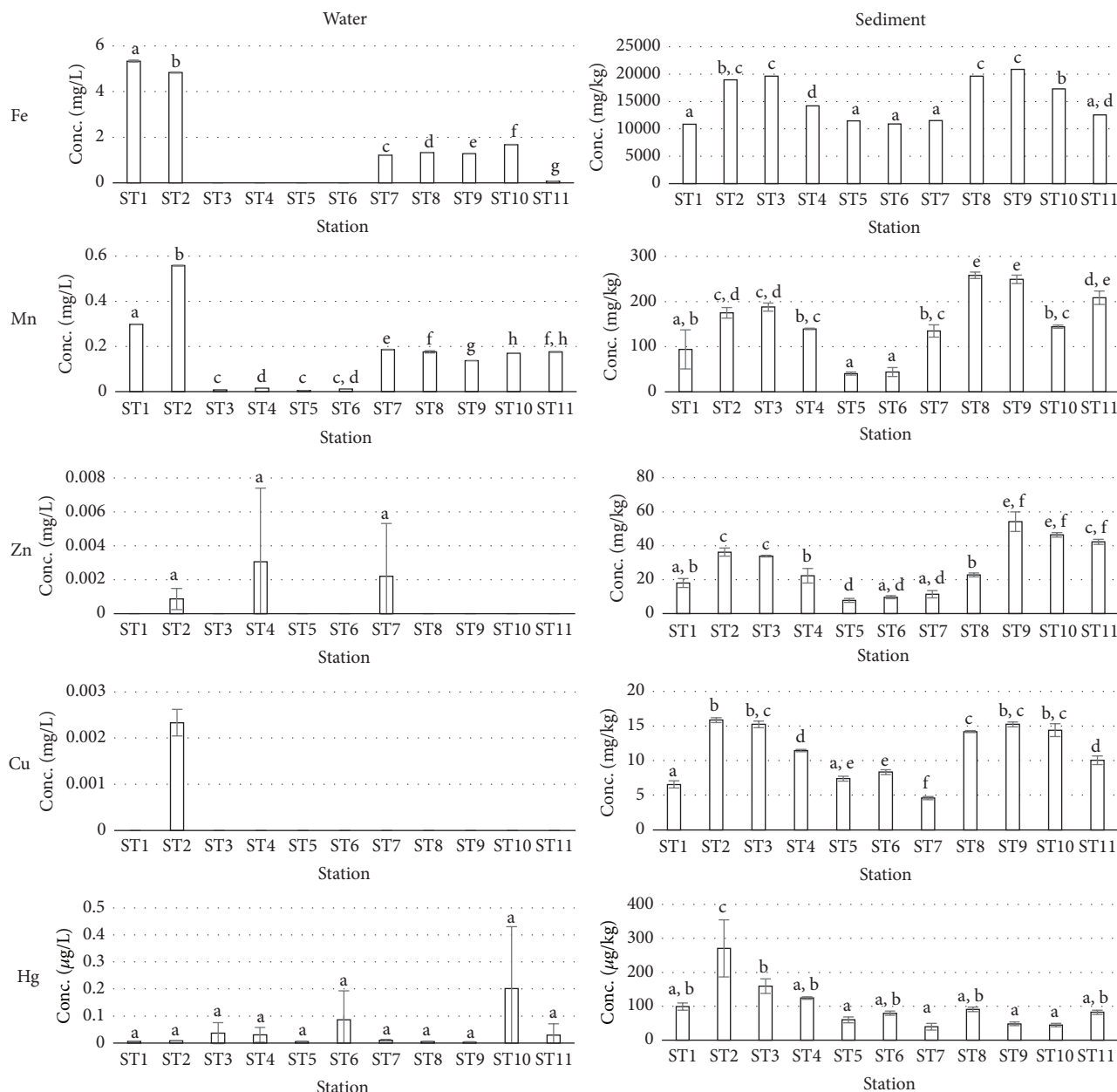


FIGURE 2: The average metal concentrations in water and sediment (different letters indicate significant difference at $p = 0.05$).

to the reservoir. The declining concentration at the dam wall is possibly associated with the reaerated environment. Sim et al. [34] reveal similar observation with all elements detected at lower concentrations at the station after Batang Ai Dam. In terms of contamination status, there is minimal risk of contamination with I_{geo} consistently < 0 , CF and PLI < 1 . In comparison to metals reported elsewhere in the dam environments (Table 3), the range of metals identified is comparable to other studies suggesting no particular concern of contamination.

The average metal concentrations in fishes are shown in Figure 3. Hg is evidently higher in *Barbonymus schwanenfeldii* (average concentration 2.158 mg/kg), *Puntioplites waandersii* (1.921 mg/kg), *Cyclocheilichthys apogon* (1.934 mg/kg),

and *Hemibagrus planiceps* (2.397 mg/kg) in dry weight. Among these, *Barbonymus schwanenfeldii*, *Cyclocheilichthys apogon*, and *Hemibagrus planiceps* could also be found in the environment of Batang Ai where the Hg level reported was relatively lower at 0.07, 0.13, and 0.34 mg/kg (in dry weight), respectively [34]. As observed, *Hemibagrus planiceps* tends to accumulate Hg corroborating the findings of Sim et al. [34] at Batang Ai, possibly due to its diet nature as a bottom feeder [34]. *C. apogon*, *B. schwanenfeldii*, and *P. waandersii* are also found to accumulate considerable Hg likely associated with their carnivorous diet. Numerous literatures have revealed elevated Hg in carnivorous fish, higher than that in omnivorous and herbivorous species [4, 45, 46].

TABLE 3: The metals content in sediment reported in various studies with regard to the dam environment.

Study site	mg/kg				
	Fe	Mn	Zn	Cu	Hg
Bakun (this study)	10342–22126	32.32–265.50	6.54–62.28	4.26–16.26	0.03–0.39
Batang Ai Dam ^a	2994–3693	16.44–820.06	nd–16.48	—	0.08–0.17
Masinga Dam ^b	—	259.12–642.3	60.04–75.84	11.38–23.67	—
Iron Gate Dam ^c	5000–32000	400–1100	49.4–389.5	17.80–57.60	0.06–0.30
Atatürk Dam ^d	12587–19265	73–514	59–60	14.5–22.7	nd
Aguamilpa Dam ^e	4740–15900	—	14.8–51.8	0.79–60.8	0.01–0.04
Avsar Dam ^f	22734–25268	—	—	23.5–30.0	—

^aSim et al., 2014 [34].^bNzeve et al., 2014 [14].^cMilenkovic et al., 2005 [35].^dKaradede and Ünlü, 2000 [36].^eRangel-Peraza et al., 2015 [37].^fÖztürk et al., 2009 [38].

In this study, the concentration in wet weight is estimated based on the moisture content and compared with the guideline of FAO/WHO [29]. The moisture content is in good agreement with the findings of Olley [47], at an average of 81% with no significant difference between species. Based on the estimation, there is no concern of Zn and Cu contamination in fishes; the levels detected are well below the guideline of 100 and 30 mg/kg (in wet weight), respectively. For Hg, all species examined exhibit

concentrations below the permissible level of 0.5 mg/kg [*Barbonymus schwanenfeldii* (0.43 mg/kg), *Puntius waandersii* (0.31 mg/kg), *Cyclocheilichthys apogon* (0.40 mg/kg), *Hemibagrus planiceps* (0.41 mg/kg), *Cyclocheilichthys armatus* (0.15 mg/kg), and *Pangasius micronemus* (0.04 mg/kg)]. The health risk of Hg is evaluated based on the hazard index (HI) calculated according to the estimated daily dose (ED (mg/kg/day)) and reference dose (RfD). The reference dose for total Hg is 0.0003 mg/kg body weight per day, defined by US EPA [48]:

$$HI = \frac{ED}{RfD}$$

$$ED = \frac{\text{Conc. of element} \times \text{fish consumption rate} \times \text{bioavailability factor}}{\text{body weight (kg)}} \quad (2)$$

The fish consumption rate, bioavailability factor, and hypothetical body weight of different groups (including children and adults) are assumed based on the guideline of US EPA. The assumed body weights for children between 0–1 years and 1–11 years are 14.4 and 26.4 kg, respectively. For adult women and men, the body weight is estimated at 65 and 73 kg, respectively. The bioavailability rate of Hg is assumed at 100% whilst the fish consumption rate often varies according to population. The fish consumption rate is estimated at an average of 0.08 kg/day whereas for children below 11 years it is 0.02 kg/day. Table 4 summarises the health risk assessment of Hg in fish. When $HI > 1$, it indicates probability of adverse health effects whilst $HI < 1$ suggests no adverse effects. Some species appear to demonstrate $HI > 1$. Exposure ratio (ER, unitless) and consumption limits (CR, g of fish/week) were calculated to suggest the consumption rate/week:

$$ER = \frac{ED}{pTDI} \quad (3)$$

The provisional tolerable daily intake of Hg (pTDI) proposed by Health Canada for adult men and women who are not

of reproductive age is 0.47 µg/kg body weight (BW)/day. For children, the value of pTDI proposed is 0.2 µg/kg BW/day. The consumption limit is calculated as follows:

$$CR = \frac{[pTDI \times BW \times 7 \text{ (day/week)}]}{\text{Total Hg conc. in fish (µg/g)}} \quad (4)$$

The suggested consumption rate for *Barbonymus schwanenfeldii*, *Puntius waandersii*, *Cyclocheilichthys apogon*, and *Hemibagrus planiceps* is ranging between 500–775 g/week for adults and 33–135 g/week for children.

4. Conclusions

Elevated Fe and Mn were detected in downstream water of Murum and Bakun Dam due to depleted dissolved oxygen and reduced pH. Cu, Zn, and Hg were found in trace amount. The metal concentrations in sediment at ST1 and ST7, nearer to the dam, are lowest. Generally, there is no contamination in sediment according to the geochemical indices. Metal contents in fishes varied according to species with the bottom

TABLE 4: The health risk assessment of Hg.

Fish species	Conc. (mg/kg in wet weight)	Estimated dose (mg/kg)				Hazard index (HI)				Exposure ratio (ER)				Consumption limits (CR), g, of fish/week			
		0-1 year old	1-11 years old	Adult women	Adult men	0-1 year old	1-11 years old	Adult women	Adult men	0-1 year old	1-11 years old	Adult women	Adult men	0-1 year old	1-11 years old	Adult women	Adult men
<i>Cylocheilichthys armatus</i>	0.15	0.0002	0.0001	0.0002	0.0002	0.69	0.38	0.62	0.55	3.47	1.89	3.08	1.17	93	280	1426	1601
<i>Barbonymus schwanefeldii</i>	0.43	0.0006	0.0003	0.0005	0.0005	1.99	1.09	1.76	1.57	9.95	5.43	8.82	3.34	33	98	497	559
<i>Puntioptiles waandersii</i>	0.31	0.0004	0.0002	0.0004	0.0003	1.44	0.78	1.27	1.13	7.18	3.91	6.36	2.41	45	135	690	775
<i>Pangasius micronemus</i>	0.04	0.0001	0.0000	0.0000	0.0000	0.19	0.10	0.16	0.15	0.93	0.51	0.82	0.31	350	1050	5346	6004
<i>Cylocheilichthys apogon</i>	0.4	0.0006	0.0003	0.0005	0.0004	1.85	1.01	1.64	1.46	9.26	5.05	8.21	3.11	35	105	535	600
<i>Hemibagrus planiceps</i>	0.41	0.0006	0.0003	0.0005	0.0004	1.90	1.04	1.68	1.50	9.49	5.18	8.41	3.19	34	102	522	586

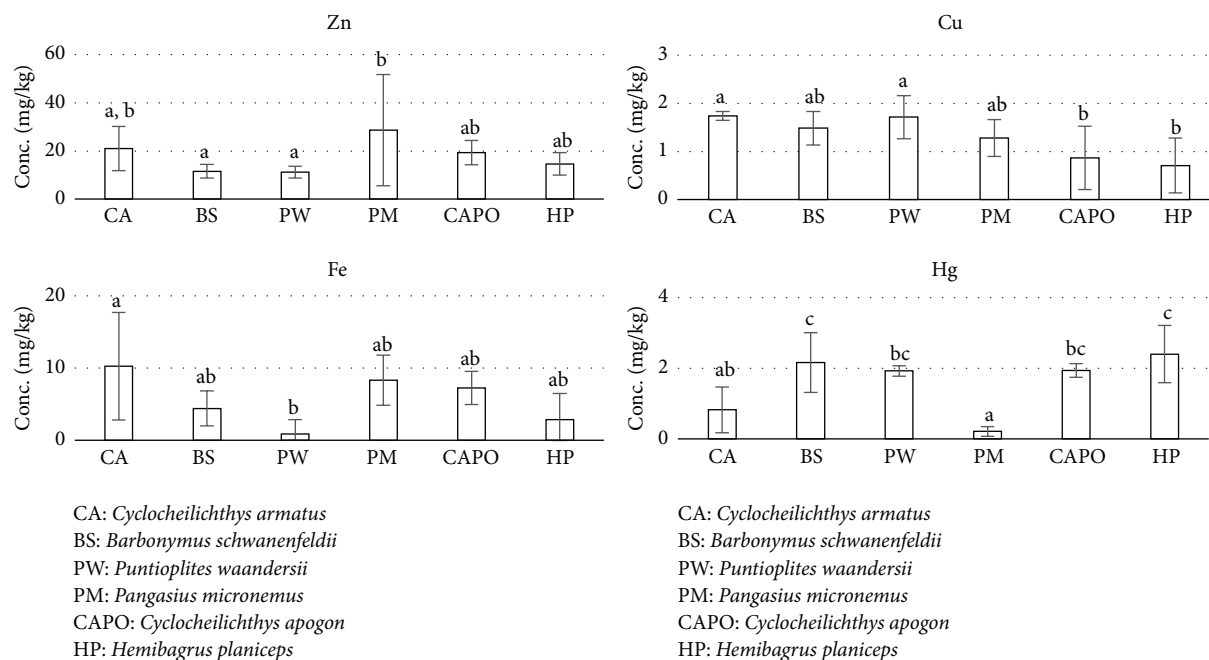


FIGURE 3: The average metal concentrations in fishes in dry weight.

feeder, *Hemibagrus planiceps*, exhibiting higher affinity for mercury accumulation. The estimated Hg concentration in wet weight for all species examined was below the permissible level of 0.5 mg/kg wet weight. *Barbonymus schwanenfeldii*, *Puntiolites waandersii*, *Cyclocheilichthys apogon*, and *Hemibagrus planiceps* were identified with hazard index > 1, implying possible adverse effects. The consumption rate was calculated and the amount recommended is in the range of 500–775 g/week for adults and 33–135 g/week for children.

Conflict of Interests

The authors have declared that no competing interest exists.

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