JOURNAL OF SUSTAINABLE MINING

Volume 23 | Issue 1 Article 7

2024

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Recommended Citation

Maidie, Asfie; Almadi, Ismail Fahmy; Efendi, Muchlis; Nilawardhani, Rekha Yusdha; Sukarti, Komsanah; and Pagoray, Henny (2024) "Analysis of the impact of water treatment by liming sedimentation and dredging on the content of heavy metals in fish intended for consumption," *Journal of Sustainable Mining*: Vol. 23 : Iss. 1 , Article 7.
Available at: https://doi.org/10.46873/2300-3960.1406

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Analysis of the impact of water treatment by liming sedimentation and dredging on the content of heavy metals in fish intended for consumption

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1. Introduction

The coal mining industry is an important sector of the Indonesian economy; Indonesia is the fifth-largest exporter of coal in the world [1]. Most of the country's mining companies engage in open pit mining, which offers a reserve of more than 85% compared with only 50% from underground mining and is more attractive due to geological conditions and available techniques. However, removing the cover material before extracting the coal is costly, and this cost rises even more when the open hole of the pit must be filled after mining is complete [2]. Some coal mining companies cannot fill in these ex-coal mining pits because of the cost of filling and obtaining suitable soil for filling material. This has led to environmental problems related to the large open (unfilled) holes in the ground that remain after the coal mining operation has finished. These holes are then inundated by river outflow, runoff, and rainwater and become large reservoirs. They may be used by local people for washing, bathing, agriculture, and even drinking water, and the reservoir itself may be used for fish culture with floating nets or game fishing for wild fish. These ex-mining reservoirs may, therefore, create important opportunities for former mining employees [3,4].

Past research investigated heavy metal levels present among fish living in water affected by mining activities. A study in North China of 16 freshwater sites showed that cultured cyprinid species including crucian, bighead, and grass carp had lower levels of the heavy metals copper (Cu), chromium (Cr), zinc (Zn), lead (Pb), arsenic (As), cadmium (Cd), manganese (Mn), and nickel (Ni) than wild fish, but both cultured and wild fish had levels considered

Received 19 January 2023; revised 11 August 2023; accepted 17 August 2023.
Available online 15 January 2024

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https://doi.org/10.46873/2300-3960.1406

safe for consumption [5]. In Iraq's seemingly polluted Tigris River, cadmium, chromium, lead, zinc, and copper had accumulated in the muscles of two species of cyprinid at detectable concentration levels [6]. Coal mining is expected to affect lakes of standing water and the fish living in it. In China's Gaotang Lake, concentration levels in fish of arsenic, cadmium, cobalt (Co), copper, mercury, lead, and antimony (Sb) were below the maximum permitted by the Chinese government [7].

Other active reservoirs or sedimentation pools may exist in addition to those inactive ones originating from the abandoned open holes from coal mining activities. Both types are habitats for wild and cultured fish, as well as cultured fish that become wild after their introduction to the environment. Both types of reservoirs in coal mining areas are treated with lime to increase water pH. This is combined with dredging for sediment in operating reservoirs. Past studies showed no difference ($P > 0.05$) in metal and metalloid concentration levels in water between sites with or without coal mining activities [8]. However, even if there is no apparent difference, fish have the potential to bio-magnify and accumulate even the lowest concentrations of heavy metals and metalloids in water. People consume these fish, obtained either directly from coal mining-affected waters or from local markets and restaurants. Therefore, this study aimed to determine the health risks related to toxic heavy metals and arsenic in fish in the explored coal mining area.

2. Materials and methods

The main eight-year study was divided into four two-year parts and monitored nine heavy metals and one metalloid within the fish population of two sedimentation ponds, two tailing sedimentation ponds, and one abandoned open-pit reservoir from an enterprise in East Kutai Regency. An accompanying study was carried out in abandoned open-pit reservoirs from a different enterprise that were used for wild and cultured fish in Berau Regency and Kutai Kartanegara Regency; that effort studied the levels of four heavy metals at one collection point in time. All of the abandoned open pit reservoirs were treated with lime to increase water pH, but the sedimentation ponds were treated with lime combined with regular dredging because they were active reservoirs as part of mining activities. The main study locations and coordinates of the sampling points (confirmed with a GARMIN® GPS 12 device) are described below:

1. ***19B Pond*** (latitude 00° 32′ 16.3″, longitude 117° 38′ 17.5″) is an active, primary water collection pond for tailing water. The area of the square-shaped pond is about 5 ha; it is filled by water to about 50% of its depth after a downpour, and its main bottom substrate is pine, coarse coal, and mud. The water pH is controlled by the company by adding calcium oxide (CaO) at a volume according to the pH value of the water. The pond bottom is dredged when the sediment reaches 75% of the pond volume. Nile tilapia (*Oreochromis niloticus*) and Mozambique tilapia (*Oreochromis mossambicus*) were released into the pond as biological indicators of the water quality and wild fish from nearby aquatic environments. The following water quality characteristics (mean value ±standard deviation) were determined for this pond: turbidity 16.8 ±17.73 NTU; total dissolved solids 591.8 ±354.4 mg/L; total suspended solids 22.8 ±17.59 mg/L; dissolved oxygen 4.1 ±0.68 mg/L; and pH 6.90 ±0.51. The water quality of the 19B pond met the East Kalimantan Province government's standardized regulations for coal mining tailing wastewater (total suspended solids, 200 mg/L; pH, 6–9; and Fe and Mn, 7 and 4 mg/L, respectively) [9].
2. ***19A Pond*** (latitude 00° 32′ 06.0″, longitude 117° 38′ 20.2″) is an active pond for collecting water from 19B Pond (as a secondary tailing water pond). The area of the square-shaped pond is about 5 ha; it is filled with water to 50% of its depth after a downpour, and its bottom is mud/clay with a small amount of pine and coarse coal. Bottom dredging and water pH in this pond are controlled the same way as in 19B. Nile tilapia (*O. niloticus*) and Mozambique tilapia (*O. mossambicus*) were released into this pond for the purpose of monitoring water quality; they joined wild fish that were already present. Similar to 19B, no feeding was provided for the fish. This pond had the following water quality characteristics: turbidity 27.35 ±22.25 NTU; total dissolved solids 618.19 ±280.43 mg/L; total suspended solids 39.38 ±26.95 mg/L; dissolved oxygen 4.99 ±0.61 mg/L; and pH 6.99 ±0.85. The water quality of the 19A pond met the regulations for coal mining tailing wastewater.
3. ***KJ Pond*** (latitude 00° 32′ 56.4″, longitude 117° 31′ 23.4″) is an active pond for collecting surface runoff, rainwater, inundated water from surrounding aquatic environments, and acid mine drainage (excluding tailing wastewater). The square-shaped pond has an area of about 20 ha; it is filled with water to only 30% of its depth and

about 1% of its total volume daily but is full if heavy rain occurs. The bottom of the reservoir is fine mud with a depth of less than 1 m. The company controls the water pH to neutral or basic by adding 250 kg of calcium oxide (CaO) per year for liming and water depth by dredging the mud sediment. The water quality of this pond was as follows: turbidity 35.13 ±30.28 NTU; total dissolved solids 1479.55 ±818.08 mg/L; total suspended solids 41.55 ±18.35 mg/L; dissolved oxygen 5.75 ±0.97 mg/L; and pH 8.01 ±1.21. The water quality met the standards set by the government of East Kalimantan Province for wastewater from coal mining activities, except for tailing wastewater (total suspended solids, 300 mg/L; pH 6–9; Fe 7 mg/L; and Mn 4 mg/L).

4. ***SD Pond*** (latitude 00° 32′ 56.4″, longitude 117° 34′ 11.3″) is an irregularly shaped active reservoir or pond located near public housing. It is one of the ponds used to settle the sediment from water (runoff, water from pits, and tailing water) from the coal mining area before the water is released into the Sangatta River. The pond has an area of about 1 ha; it is filled by water to 30% of its volume daily but is full if there is heavy rain. The deepest area is over 3 m, with bottom sediment mainly comprising fine coarse coal and mud having a depth of about 0.5 m. Sampling was done once at one sampling point in a sampling period. The water quality of the pond was as follows: turbidity 63.26 ±55.34 NTU; total dissolved solids 1441.67 ±126.3 mg/L; total suspended solids 43.3 ±33.43 mg/L; dissolved oxygen 3.60 ±1.11 mg/L; and pH 8.03 ±1.86. The water quality met the standards set by the government of East Kalimantan Province, which were the same as for KJ Pond.
5. ***TBA Lake*** (latitude 00° 33′ 19.17″, longitude 117° 28′ 24.64″) is a non-active reservoir that originates from an open pit, giving it an irregular shape. Termed a "lake" by the company, it should more appropriately be considered a "man-made lake." After years of pH and sediment treatment to stabilize water quality to safe levels for raw water drinking, the area is now also utilized as an area for recreation and fish culture in floating cages, and the city uses it for tap water and household use. Only fish, not water, were sampled in this body of water, which has an area of about 20 ha. Previous work showed that fish cultured in a common culture period of three months showed no significant concentration of heavy metals; thus, we tried to analyze older cultured fish with the hope of discovering the amount of heavy metals that could have accumulated over three years in this body of water.
6. ***Sangatta River*** (latitude from 00° 27′ to 00° 35′, longitude from 117° 17′ to 117° 34′) is a 92-km-long river measuring 30 m at its greatest width and nearly 10 m at its greatest depth. Three main human activities, which also produce wastewater, are present in this river: the small town of Sangatta City, huge palm plantations, and other coal mining enterprises. At least six sampling points for water quality were located in the river: two points at the upper part of the river before the coal mining area (SRU 1 and SRU 2), where the area has paddy fields, palm plantations, and forest; two points in the coal mining area, where the drainage canal (SRD 1 and SRD 2) mixes with water from the Sangatta River; and two points near the river mouth, after passing through a dense human population on the river bank (SRM 1 and SRM 2). The water quality of this river was as follows: turbidity 154.82 ±63.98 NTU; total dissolved solids 198.5 ±76.65 mg/L; total suspended solids 62 ±25.76 mg/L; dissolved oxygen 5.01 ±1.36 mg/L; and pH 6.20 ±0.27. Except for the total suspended solids, which exceeded the government limit of 50 mg/L, the parameters met the national standard for raw tap water [10].

The following water quality parameters were measured in the field using a YSI556 MPS water meter: dissolved oxygen, turbidity, water temperature, conductivity, and total dissolved solids. Along with the main project, another brief study was carried out with funding from another source to determine the quality of water treated with liming in abandoned open pit reservoirs of coal mining enterprises used for fish in Berau Regency and Kutai Kartanegara Regency. These reservoirs had areas of 20.31 ha and 30 ha and maximum depths of 10 m and 20 m, respectively, and their sediment mainly comprised a mix of mud and clay. Water quality was sampled twice at each site. Cadmium, lead, and mercury were not detected, and copper was first not detected and then detected at 0.024 ppm for the Kutai Kartanegara Regency samples. The water samples of Berau Regency revealed lead at 0.01 ppm but no detectable cadmium, copper, or mercury. The main sampling locations in East Kutai Regency are shown in Figure 1.

Sampling of fish in the studied area was conducted for 12–24 h using the following fishing gear:

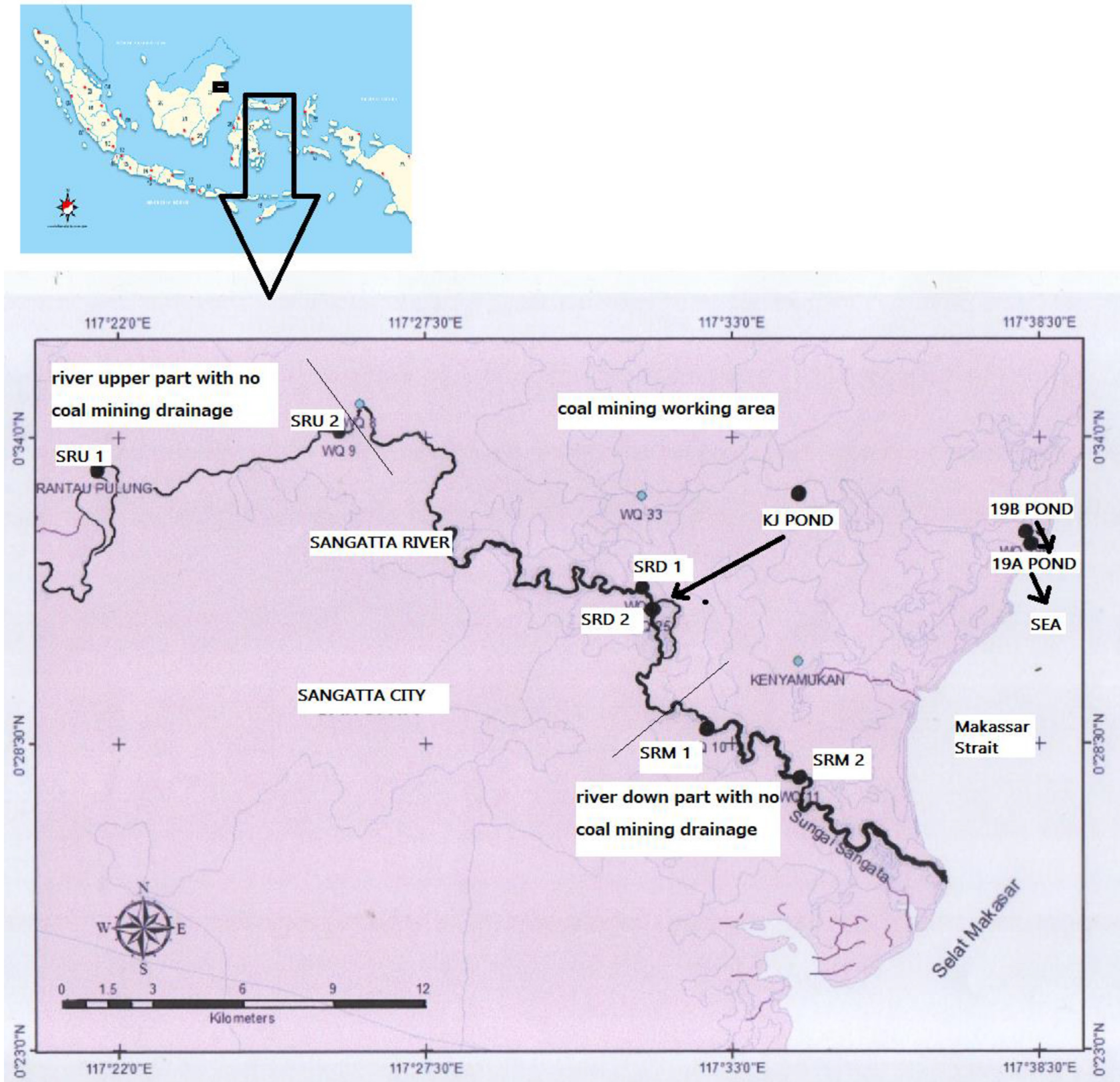

Fig. 1. The sampling area of the major locations in East Kutai Regency is the same as in the previous report [8]. KJ Pond and SD Pond (WQ 33) drain to the Sangatta River, while 19B and 19A ponds drain into Makassar Strait. SRU 1 and SRU 2 had no activities of coal mining at the river's upper part; SRD 1 and SRD 2 were in a zone of coal mining; and SRM 1 and SRM 2 were in the outer zones of coal mining activities through to the river mouth.

trammel net (based on manufacturer's instructions: twine 210 D/2, mesh size 1 5/8″, depth 52 MD, length 70 YDS, and outer net 6 MD.10″), used for 24 h; pool and line (numbers 5 to 8 hook sizes), used 12 h daily; cast net (3-m length or 6-m diameter and 1-inch mesh size), used daily; gill net (30-m length, 1-inch mesh size, and 1.5-m depth), used for 24 h; scoop net (1-m widest measure, 1.5-m length, and 0.5-m depth, with 0.01-inch mesh size), used daily; and a trap (elongated-type bamboo trap: 1.5-m length and 0.02-m diameter). The various types of fishing gear were used to minimize the bias of data that might arise if only a single gear was used [11].

Samples of fish were separately wrapped in transparent plastic according to fishing gear and cooled in ice crumble to below 4°C in the field, followed by transport to the wet laboratory for continued freezing to below 0°C for fish identification and metal analyses. Preparation of fish aliquots for metals measurement in the wet laboratory included cutting the fish with a non-metal knife for wet samples of fish with water content over 40% (standard water content of dried fish in Standard National Indonesia number 8273 2016 is below 40%), weighing them with an analytical balance, and then digesting them in borosilicate glass destructor tubes with nitric acid pro analysis (pa) (Merck®). A blank sample without fish underwent the same procedure to control the digestion quality of the samples. Heavy metals and metalloid measurements were performed by a commercial laboratory (certified ISO 9000, ISO 9002, ISO 27025 and with quality control by the National Standard of Indonesia (KAN) (https://www.sucofindo.co.id) for selenium, arsenic, cadmium, copper, lead, antimony, and mercury. Meanwhile, iron, manganese, and zinc were analyzed in our laboratory using a Shimadzu Atomic Absorption Spectrophotometer with two replicates and standard metal solution products by Merck®. Our analytical laboratory operates with guidance from Nihon University and Japanese experts and controls the quality of measurement results by testing the same samples as measured in other laboratories. The results were expressed as micrograms of heavy metal or metalloid concentration per gram of wet sample and noted as parts per million (ppm). Both our laboratory and the commercial laboratory used the following detection limits: iron (Fe, percent recovery: 108–112%), selenium (Se, percent recovery: 98–104%), arsenic (percent recovery: 97–107%), and cadmium (percent recovery: 99–115%), detected with the lowest detection limit of 0.001 ppm; copper (percent recovery: 97–101%) and manganese (Mn, percent recovery: 97–107%), with the lowest detection limit 0.02 ppm; lead (percent recovery: 99–101%) and antimony (percent recovery: 97–106%), with the lowest detection limit 0.01 ppm; zinc (percent recovery 98–172%), with the lowest detection limit 0.05 ppm; and mercury (Hg, percent recovery: 100–107%), with the lowest detetction limit 0.0005 ppm. A concentration below the detection limit was noted as not detected (nd), not as zero or 0 ppm.

Sampling for consumed fish was performed at the local markets and restaurants of Kutai Timur Regency. Marketed live fish were handled in the same way as fish caught in the reservoirs and river, while dry salted fish from the local market and cooked fish from restaurants were kept in transparent plastic wrap and refrigerated to below 10°C before digestion and analysis, the same as for the fresh fish. The concentration of heavy metals and metalloids in fresh fish was noted as micrograms per gram wet weight or ppm, while dry salted fish was noted in micrograms per gram dry weight (ppm) and compared to the safety level set by the Indonesian Government and FAO-WHO. The detectable samples containing heavy metal $x(n)$ within a total number of samples for heavy metal x (t) were expressed as percentage detectable (PD) $= n/t \times 100\%$, n or $t = 1, 2, 3 \ldots$ of sample number [12], together with the bioaccumulation factor (BAF) calculated with the concentration of x metals in fish (b) divided to the mean value of detected concentration x metal in water (p). The accumulation should be decided to present if the value of $b/p > 1$ and also exceed the expected value for the species or BAF > expected BAF value for freshwater fish for each studied metal according to species [13,14]. The accumulation factor of BAF was noted in the table as a percentage of accumulation present in detectable concentration divided by the total sample (t) of each metal studied. Because all data sets were contain or not detected, the statistical analysis was performed after the nd data was withdrawn. The analyzed data were on heavy metals in fish from the coal mining area and river, while the heavy metals in marketed fish were not statistically analyzed because the fish was originally from the studied river and also handled, prepared, and cooked in different ways. The normal distribution of the data sets (sample number > 4 of Fe, Cu, Pb, Zn, Mn, and Se) was analyzed by the Lilliefors Test [15]. The non-normal distribution data set was analyzed by non-parametric statistic [16] and calculated using the Mann-Whitney U Test Calculator (https://www.socscistatistics.com/tests/mannwhitney/default2.aspx) for data regarding Fe, Cu, Pb, Zn, and Mn, while the normal distribution of the Se data set was analyzed by parametric statistic [17], with the variance tested by F Test Calculator for equality of variances (https://www.statskingdom.com/220VarF2.html) and the equality of the mean value of two samples using the Two Samples T-Test Calculator (https://www. statskingdom.com /150MeanT2uneq.html).

3. Results and discussion

3.1. Metals and the arsenic in fish

As noted in the methodology discussion, the water quality of the studied coal mining reservoirs and the connected river, which are vital for fish life, was relatively good. The pH was over six, and dissolved oxygen was over four ppm, but although the water is turbid, suspended solid and dissolved solid exceeded the optimum, as noted in the literature

[18]. This water quality naturally occurred in this main study area of coal mining [8] and in the Sangatta River [19]. Therefore, there is no need to pay much attention to water quality for fish life in coal mining reservoirs and connected rivers. Levels of studied metals in wild fish varied markedly, from not detected to relatively high concentrations, as shown in Table 1. Lead, though detected in only small proportions of samples (12% in the Sangatta River and 13% in marketed fish), returned high readings of 0.842 ppm and 0.456 ppm, respectively, which was above the levels permitted by the WHO and the Indonesian government. The lead levels in fish, therefore, require attention. Arsenic was detected at concentrations as high as 0.467 ppm in fish sampled from the Sangatta River, although only in 14% of the samples, but it was not detected in the fish in markets. Cadmium, copper, lead, and mercury were not detected in wild fish samples from Berau Regency and Kutai Kartanegara Regency, which are therefore not noted in Table 1.

In the present study, data were obtained four times (every other year for eight years). Fish were sampled in the upper area of the Sangatta River as the experiment control, as they would not have been affected by the coal mining activities. The following results were obtained only for sampled *B. schwanefeldii*, *Labeobarbus fasciatus*, and *Rasbora caudimaculatas*: Fe, nd to 0.429 ppm (75%); Pb, nd to 0.13 ppm (25%); Cu, nd to 0.038 ppm (25%); and seven other heavy metals and metalloid, nd. However, as these fish are known to migrate between areas both unaffected and affected by coal mining, these results were considered unsuitable for use as a control for fish unaffected by coal mining activities.

The use of calcium base liming to control water acidity in reservoirs in coal mining areas is widespread. Liming not only increases water pH to neutral or basic, as recommended by the government for environmental management, but also provides conditions suitable for fish survival. In this study, the cyprinid fish, which are the major fish species living in this area, need a neutral or slightly basic pH, but the *Channidae* and *Anabantidae*, two important consumed swamp fish, require low pH levels or acidic water [22].

Studies have shown correlations for certain heavy metals between the concentration in water and that in the organs of fish, especially the gills, kidneys, and livers [23,24]. However, no pattern was identified with regard to the concentrations of heavy metals and arsenic in the present study. The metals Fe, Cu, Pb, Zn, and Mn are expected to have a higher probability of being found in fish living in

Table 1. Heavy metals and metalloids in fish.

Parameters	Unit	Sampling location and sample number (n)					
		19B Pond $n = 11$	19A Pond $n = 9$	KJ Pond $n = 10$	SD Pond $n = 7$	Sangatta River $n = 94$	Marketed fish $n = 23$
Fe	ppm	nd–1.16; 63 (45)[a]	nd–1.286; 55 (55)	nd–2.52; 55 (10)	nd–8.16; 80 (71)	nd–18.785; 80 (19)	nd–1.68; 23
Cd	ppm	nd	nd	nd	nd	nd–0.006; 2 (na)	nd
Cu	ppm	nd–15.85; 44 (36)	nd–15.659; 33(Nd)	nd–0.107; 25 (10)	nd–0.872; 60(Nd)	nd–1.538; 21 (6)	nd–0.026; 4
Pb	ppm	nd	nd–0.384; 22 (na)	nd–0.237; 22 (na)	nd	nd–0.842; 12 (2)	nd–0.456; 13
Zn	ppm	nd–0.866; 45(Nd)	nd–0.647; 44(Nd)	nd–1.356; 5 (60)	nd–0.872; 60 (57)	nd–1.299; 37 (20)	nd–0.088; 35
Mn	ppm	nd–0.058; 36 (27)	nd–0.072; 33 (22)	nd–0.050; 55 (na)	nd	nd–0.514; 30 (na)	nd–0.089; 22
As	ppm	nd–0.043; 9 (9)	nd–0.178; 11 (11)	nd–0.156; 11(Nd)	nd	nd–0.487; 14 (12)	nd–0.057; 17
Hg	ppm	nd	nd	nd	nd	nd–0.016; 6 (na)	nd
Se	ppm	nd–0.4; 18(Nd)	nd–0.385; 22(Nd)	nd–0.391; 22(Na)	nd	nd–0.936; 24(Nd)	nd–0.457; 4
Sb	ppm	nd	nd	nd	nd	nd	nd

Note: Standard set for heavy metals and metalloid in consumed fresh fish is 0.1–0.3 ppm (BPOM-RI) and 2 ppm (FAO-WHO) for Cd; 0.2–0.4 ppm (BPOM-RI) and 0.3 ppm (FAO-WHO) for Pb; 0.25 ppm (BPOM-RI) for As; and 0.5–1 ppm (BPOM-RI and FAO-WHO) for Hg in the form of methyl mercury [20,21].

[a] Concentration range; percentage detectable (%); and percentage (%) of samples has tended to bioaccumulation; **nd** = not detected or concentration below detection limit of instrument. **nd** = not detected in water, but some detected in fish (thus, seemingly bioaccumulations occur through water and food in very small concentrations). **na** (no accumulation) = the concentration of some metal was detected both in water and fish, but the concentration in fish was lowest than in water.

coal mining reservoirs than in the river, but studies show that these five heavy metals' concentrations do not differ ($P > 0.05$) in regard to both fish living in coal mining reservoirs and a connected river. In this study, Fe was detected in all samples for water quality and was expected to show higher concentrations among the fish than other detectable heavy metals, but only 80% of the sampled fish had detectable Fe levels (Fig. 2).

Iron was the most abundant metal, which is usual in freshwater rich in organic matter [25], but no specific pattern was observed with regard to concentration levels; they were higher, lower, or the same in different sampling periods. The fluctuation in heavy metals and metalloids in water samples in the present study suggests that no source constantly introduces high concentrations of these substances into the aquatic environment in this coal mining area. The concentration of iron in fish (both wild and marketed) in the present study ranged from being not detected (nd) to being as high as 18.785 ppm from a sample of wild *C. striata*, a predator fish in the Sangatta River. This appears to be a normal finding. Iron is an important component of hemoglobin, accounting for 20-90% of the total iron in fish (0.55–14.43 mg/100 g weight of fresh fish) [26]. However, cleaning fish before they are marketed would remove the blood as a source of iron, resulting in low iron (Fe) levels in marketed fish (Table 1).

Notably, metal concentrations were similar in cultured and wild fish. Samples of *Pangasius suthci* and *Clarias gariepinus*, cultured for long periods of time (i.e., over three years) in an abandoned open pit reservoir (TBA Lake) in the main study area, displayed iron concentrations of only 0.02 and 0.03 ppm, and zinc concentrations of only 0.033 and 0.102 ppm, respectively, while the wild fish *Channa striata* and *O. niloticus* were found to have concentrations of iron of 0.01 and 0.04 ppm, and zinc of 0.032 and 0.044 ppm, respectively. In addition, fish sampled from floating nets in Berau Regency and Kutai Kartanegara Regency showed no detectable levels of cadmium, copper, lead, and mercury, the same as for cultured Nile tilapia (*O. niloticus*), common carp (*Cyprinus carpio*), and a catfish (*P. sutchi*). Of note, these results differ from those of our previous work [27] regarding another sedimentation pond with insufficient liming treatment, not included in the present study, which showed that fish cultured for three months have detectable concentrations of antimony (100% detectable), selenium (57.14%), mercury (50%), manganese (14.29%), iron (7.14%), and zinc (100%), all at safe levels for human food. The previous study also showed that only chromium levels were significantly different between wild fish and cultured fish, while the other heavy metals of silver, aluminum, cadmium, cobalt, chromium, copper, iron, manganese, nickel, lead, and zinc were not statistically different [28].

3.2. Fish metals according to family

In this study, the sampled fish are common fish species in Indonesia's freshwater habitats that are also commonly consumed by people. Some are also common staple foods in other Southeast Asian countries. However, among these species (Table 2), the sea catfish from the Ariidae family is not as popular for consumption as others, even though it is

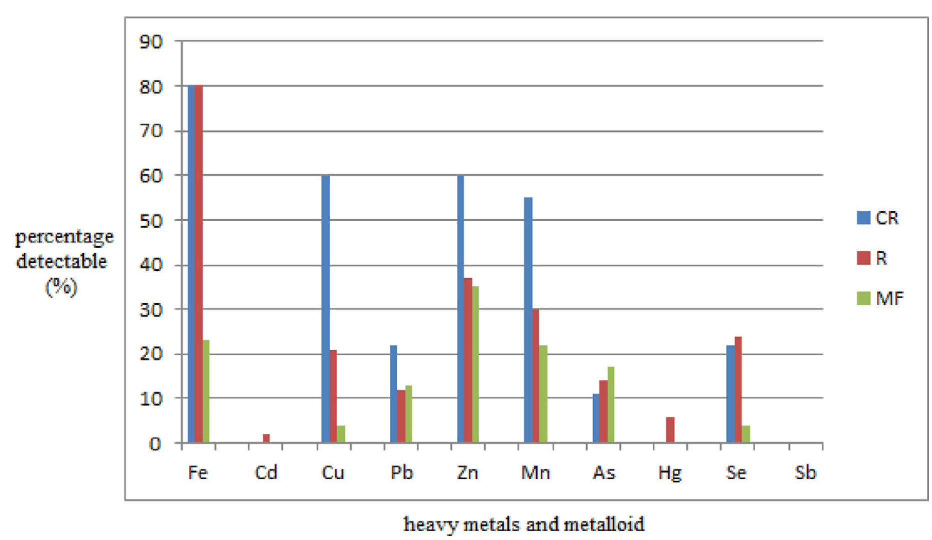

Fig. 2. Maximum percentage of detectable heavy metals and metalloids in fish from coal mining reservoirs (CR), the Sangatta River (R), and the market (MF).

Table 2. Heavy metals and the metalloid (arsenic), according to the fish family.

Fish family[a]	Heavy metals and metalloid (ppm; percentage detectable %)									
	Fe	Cd	Cu	Pb	Zn	Mn	As	Hg	Se	Sb
Ariidae: sea catfish but swimming into freshwater part of river, carnivore, swimming near bottom	nd–4.79; 58	nd–0.006; 3	nd–0.06; 20	nd–0.842; 13	nd–0.89; 48	nd–0.156; 58	nd–0.2; 10	nd-0.016; 7	nd–0.827; 6	nd
Cyprinidae: omnivores, fast swimming in water column	nd–6.281; 91	nd–0.006; 17	nd–0.079; 15	nd–0.162; 8	nd–1.72; 82	nd–0.514; 71	nd–0.447; 28	nd–0.012; 6	nd–0.936; 48	nd
Cichlidae: omnivores but primarily feed on spirogyra, slow swimming in water column	nd–2.517; 73	nd	nd–15.85; 21	nd	nd–1.356; 47	nd–0.237; 63	nd–0.487; 10	nd–0.006; 10	nd–0.702; 26	nd
Channidae: carnivores, air-breathing fish, slow swimming near surface	nd–18.785; 63	nd	nd–15.659; 36	nd–0.021; 8	nd–0.647; 81	nd–0.647; 45	nd–0.178; 9	nd	nd–0.850; 81	nd
Anabantidae: omnivores, air-breathing fish, swimming near surface	0.318–7.346; 100	nd	nd–0.051; 66	nd	nd–1.299; 33	nd–0.113; 66	nd	nd	nd	nd

[a] **Ariidae**: *Mystus gulio* and *Arius caelatus*; **Cyprinidae**: *Barbonymus scwhanenfeldii, Hampala macrolepidota, Nematabramis everetti, Rasbora einthoveni, Parambasis apogonoides, Puntius belinka, Rasbora argyrotania, Osteochilus borneensis, Parambasis punctualata, Barbodes gonionotus, Labeobarbus fasciatus,* and *Rasbora caudimaculata*; **Cichlidae**: *Oreochromis niloticus*; **Channidae**: *Channa striata*; **Anabantidae**: *Anabas testudineus*.

caught by fishermen in large quantities. When the present study began, we assumed that the content of the metals would vary according family of fish because of the behavior associated with each; for example, carnivores would be expected to have higher metal content in the flesh than herbivores or omnivores, as their position at the top of the feeding pyramid would lead to more accumulated metals from eating small fish or other aquatic biota. In turn, these fish would affect humans more profoundly. However, the data did not reflect this (Table 2).

The species composition of fish should affect the type and concentration of heavy metals. A study of lake fish showed that their heavy metal content was dependent on age, body weight, and total body length, and the bioaccumulation of heavy metals was correlated negatively or positively to different species [29,30]. The present study showed no difference or particular pattern in the concentration or detection of heavy metals and the metalloid (arsenic) studied in all groups of fish families and feeding types (herbivorous, omnivorous, and carnivorous). We assumed that because the coal mining activities and human habitation on the river bank affect only parts of the river and fish can move freely between affected and non-affected areas, the concentration of metals and metalloids would vary markedly on individual, species, and family levels. Liming is also expected to affect the metals dissolved in the water [31], and no difference was found in the concentration of metals in water between reservoirs of coal mining areas that underwent liming combined with dredging and the adjoining river water [8]. A previous study showed low levels of heavy metals and metalloids in the water of reservoirs or sedimentation ponds in coal mining areas that underwent periodic liming and dredging, which ought to affect the available metals and metalloids in water for uptake by fish in reservoirs of coal mining areas. Along the same reasoning, even if the calculated BAF > 1 is present in some fish, it will be noted that there is no detectable bioaccumulation because the calculated BAF was less than the expected BAF value for freshwater fish for each studied metal according to species [14].

Selenium (Fig. 1) was detected in fish at concentrations ranging from not detected (nd) to as high as 0.457 ppm (nd to 24% detectable) for both the coal mining reservoirs and the connected river. While it was more detectable in the fish river, it also turned out to be different in variation and mean value ($P < 0.01$) that appeared to be higher in the river fish than in fish from coal mining waters. That means the source and uptake of selenium in coal mining reservoir fish is quite different from fish in the river. The higher concentrations of heavy metal in fish occurred in water that contained insufficient quantities of selenium metal to be detected; thus, we assumed that the fish contained selenium as a result of bio-magnification accumulation of a very small amount of the metal in the water, sediment, and feed [25]. Selenium is usually present in sediment and should also be present in the water column by a bacterial route; it is an important metal for the human body [32] and also for improving defenses against disease in fish, but it does not promote growth [33].

3.3. Fish in markets

The study also sampled fish destined for human food through markets and restaurants, caught by fishers with poles and lines, cast nets, or bamboo traps in the long Sangatta River. Of note, no high concentrations of metals were found (Table 3).

Fish in restricted coal mining operation areas such as tailing sedimentation ponds 19B, 19A, and KJ may be caught and consumed by coal mining workers, but SD Pond and the Sangatta River may be fished by common fishers. During the study, there were only two people involved in fishing for economic benefit in this area, and most of the fish they caught are noted in Table 3. These fishers are only permitted to catch a total of 30 kg of fish a day, and sell it for consumption by the city people (populations of over 130,000); this does not represent a very important source of fish for consumption by local people, and no person in this area is known to exhibit diseases caused by toxic heavy metals [34]. However, the fishers and their families who consume fish from this area each day are of concern.

According to the Office of Marine and Fisheries of East Kutai Regency [35], in 2019, people of this area consumed only 32–42 kg of fish and fish products per capita per year, much less than the rest of Indonesia at 50.49 kg per capita per year. If the fishers and their relatives, as well as other consumers, consume local fish on a daily basis, then each individual should only consume about 115 g of fish a day (based on 42 kg per capita per year). If they consume fish containing higher concentrations of heavy metals (Table 1), their calculated weekly intake is much lower than the PTWI [36], with the PTWI of arsenic being 0.015 ppm body weight, cadmium 0.007 ppm body weight, lead 0.025 ppm body weight, and mercury 0.0016 ppm body weight. Because the Indonesian government has not yet regulated the PTWI, Indonesians should use the PTWI regulated by the WHO-FAO Joint

Table 3. Heavy metals and metalloids in fish collected from local markets and restaurants in the main studied area.

Fish	Heavy metals and metalloid (ppm; percentage detectable %)									
	Fe	Cd	Cu	Pb	Zn	Mn	As	Hg	Se	Sb
Cyprinid:										
Dried salted *B. schwanenfeldii*	nd	nd	nd	nd	0.255	nd	0.023	nd	0.367	nd
Fresh *L. leptocheilus*	0.114 & 0.332[a]	nd	nd	nd	nd	nd	nd	nd	nd	nd
Cichlidae:										
Dried salted *O. mossambicus*	0.057	nd	nd	nd	0.255	0.089	0.057	nd	0.457	nd
Channidae:										
Dried salted *C. striata*	nd	nd	nd	nd	0.088	nd	0.009	nd	0.348	nd
Live *C. striata*	0.148 & 2.811	nd	nd & 0.01	nd	nd	0.071 & 0.086	nd	nd	nd	nd
Anabantidae:										
Live *A. testudineus*	0.180	nd	nd	nd	nd	nd	nd	nd	nd	nd
Helostomidae:										
Dried salted *H. temminckii*	nd	nd	nd	nd	0.163	nd	nd	nd	0.006	nd
Belontiidae:										
Dried salted *Trichopodus pectoralis*	nd	nd	nd	nd	0.163	0.084	0.064	Nd	0.396	Nd
Fresh *T. pectoralis*	0.127 & 11.74	nd	nd	nd	nd	nd	nd	nd	nd	nd

[a] No value for percentage detectable is shown for number of observations below three samples.

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Commission on Food Additives or European standards (https://sib3pop.menlhk.go.id/). If they consumed fish solely from the Sangatta River (115 g of fish a day), the daily intake related to health risk should be predicted for the heavy metals contaminant using EDI. If the EDI values are less than or equal to one to five Reference Doses (RFD) via ingestion of metal, that should be a low risk to health. It is a moderate health risk if the EDI value is greater than five to ten RFD and a heavy risk if the EDI values are greater than ten RFD [37]. We should estimate the health risks of adults in East Kutai Regency according to EDI. For an Indonesian adult, the ideal body, as suggested by the Indonesian Health Ministry, is 166 cm at 19–64 years (adult), with a weight of 60–62 kg (https://www.sehatq.com). The result of health risk ingestion for adults with a 60 kg body weight with the highest concentration of heavy metals in fish ever detected in the present study revealed a low level of health risk, with the highest concentration of iron 18.785 ppm, cadmium 0.006 ppm, copper 15.85 ppm, lead 0.842 ppm, and zinc 1.299 ppm. The EDI then became 0.036; 0.0115; 0.75; 0.461; and 0.0083 mg/kg/day, respectively. These EDI values were below Reference Doses (https://www.clinmedjournals.org/articles/ijtra/ijtra-6-033-table2.html). Regarding this EDI estimation that low risk of health, and the concentration have low heavy metals, and the percentage detectable is below 50%, and only small portions of local fish are consumed and intake is not on a daily basis, the fish should be safe to consume in daily life [12,37], but we should take precaution to manage the environmental for not become toxic for organism life.

4. Conclusions

As shown by our investigation, there is no particular pattern of metals and metalloid concentration in fish. Cadmium and mercury were not detected in the coal mining area fish, and lead was not discovered among fish for sale at the market. Among metals and metalloids studied, only selenium, which has a normal distribution ($P < 0.01$) in both river fish and coal mining reservoir fish after nd data was withdrawn, showed differences in variances and mean values ($P < 0.01$). This meant that there was a particular source of Se in the river or coal mining reservoirs, but levels were higher in the river than in the reservoirs. Copper, manganese, lead, and zinc showed non-normal distributions ($P < 0.01$), and all data of these metals in the river fish and reservoir fish showed no difference ($P > 0.05$), which means that there was no special pattern of metals detected among fish living in water media treated with liming and dredging the sediment for reservoirs in coal mining area and the adjoining river. Low concentrations of metals and metalloids are apparently present in fish (calculated BAF was lower than expected BAF), this was apparently not related to the fish family or feeding behavior, so the probability of contamination that risks to human health is also low (PTWI of As, Cd, Pb, and Hg; and EDI values of Fe, Cd, Cu, Pb, and Zn were lower than standard); therefore, fish in direct and indirect contact with water from coal mining activities should be sufficiently safe to consume. The reservoirs or ponds of coal mining open pits should, therefore, be used as fish habitats and fish culture areas, provided the water is treated with liming sedimentation combined with dredging regularly.

Ethical statement

The authors state that the research was conducted according to ethical standards.

Funding body

We thank KPC Enterprise for partly funding the research and permitting access to its restricted area. We also thank Berau Coal Enterprise and Mahakam Sumber Jaya Enterprise for facilitating two abandoned coal mining open pits to study. We also thank the Research and Developmental Bureau of the East Kalimantan Province Government for facilitating funding for study in Berau Regency and Kutai Kartanegara Regency, and the Faculty of Fisheries and Marine Science of Mulawarman University, who supported us with financial management.

Conflicts of interest

The authors declare no conflict of interest.

Acknowledgments

We thank the people living on the Sangatta River bank for examining our boats and fishing gear and the employees of the coal mining enterprise for accompanying us during the study of the enterprise sedimentation ponds. We also thank our field technicians for performing the field work.

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