[Makara Journal of Science](https://scholarhub.ui.ac.id/science)

[Volume 25](https://scholarhub.ui.ac.id/science/vol25) [Issue 4](https://scholarhub.ui.ac.id/science/vol25/iss4) December

[Article 3](https://scholarhub.ui.ac.id/science/vol25/iss4/3)

12-30-2021

Bioaccumulation of Trace Metals for Daily Fish Consumption from Selected Former Tin Mining Ponds in Peninsular Malaysia: A Review on Safety of Fish Consumption

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Recommended Citation

Rosli, Mohd Hafiz; Md Joni, Amirul Azuan; and Malik, Nur Khaliesah Abdul (2021) "Bioaccumulation of Trace Metals for Daily Fish Consumption from Selected Former Tin Mining Ponds in Peninsular Malaysia: A Review on Safety of Fish Consumption," Makara Journal of Science: Vol. 25 : Iss. 4 , Article 3. DOI: 10.7454/mss.v25i4.1282

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Bioaccumulation of Trace Metals for Daily Fish Consumption from Selected Former Tin Mining Ponds in Peninsular Malaysia: A Review on Safety of Fish Consumption

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Received September 20, 2021 | Accepted November 9, 2021

Abstract

Tin mining was the predominate mining industry in Malaysia, especially during the mid-19th century. Most of the former mining areas have been reused for aquaculture purposes. Fish produced from former mining areas have been misunderstood as a high health risk if consumed due to their likelihood of containing high heavy metal content in their tissue. Therefore, we review national information on trace metals levels in the fish tissue of the selected former mining ponds in Peninsular Malaysia, together with background content of trace metals in their surroundings and food security and human health implications. Seven trace metals (Cu, Zn, As, Cd, Pb, Cr, and Ni) were observed in the water column and sediment. Various reports showed that background trace metal concentrations in the former mining area are mostly lower than several established limits (which were used as the main reference), except for the areas that were being treated for secondary use. Trace metal contents in the fish tissue also showed the same tendency of high contents due to improper or unfinished treatment of former mining ponds. In the context of food safety, various local reports have shown that the consumption of aquaculture products from former mining areas over a lifetime is unlikely causing any hazardous impact and hence may be regarded to be safe. However, these findings are restricted due to the limited studies in Peninsular Malaysia. Therefore, extensive study regarding this particular issue is highly recommended.

Keywords: fish consumption, food security, former mining site, tin mining, trace metals

Introduction

Mining is a significant economic field for most countries in Europe and Asia (International Council on Mining and Metals 2012). The mining sector progress in developing countries contributed in meeting the increasing demand, specifically for certain minerals of interest because of the expansion of world economy in recent decades [1]. Although such circumstances have led to a significant economic growth, they also have caused numerous environmental and social conflicts [2–4]. In Malaysia, tin mining predominated especially during the mid-19th century [5]. Two major aspects supported the speedy development of this field in that period, namely, the discovery of rich tin fields in Perak and Selangor and substantial for tin due to the expansion of tin canning [5, 6].

In consequence to these mining activities, the concentrations of trace metals in numerous ecosystems may reach unprecedented levels [7–9]. The increased utilization of metals in industries and mining works have led to severe environmental pollution, mainly come from effluents and runoff [10–12]. Under particular environmental circumstances, excess trace metals can be accumulated to toxic concentrations [13–16], resulting in negative effects on the surrounding ecology [17–19] and posing harm to human health [20–22].

Aquatic systems are susceptible and sensitive to trace metal pollutants, and the gradual raise in their content in aquatic environments, particularly caused by the anthropogenic sources, became an issue of primary concern [23–26]. This phenomenon is caused by the persistence of trace metals; which generally cannot be eradicated by any natural and chemical treatment, unlike almost all other organic pollutants. Moreover, the organic matters rottenness in aquatic systems, together with the detritus generated from natural weathering processes and effusive mining works, serves a fertile resource of nutrients in the bottom sediments and overlying water body [27].

Microorganisms, microflora, and algae may accumulate trace metals from various supply sources into their living cells [28]. Thus, in a food chain, little fishes are enriched with the accumulated substances, whereas the predatory ones show higher levels compared to their prey. Thus, after consuming these predatory fishes, humans inevitably suffer from the results of the enrichment that occurs at each trophic level [27].

Fish plays a significant part in the balanced diet of humans because of the existence of several macro and micronutrients [29–32]. In general, fish consumption has numerous benefits in regards to the wellbeing and may lower the cardiovascular risk and neurological diseases [33, 34]. However, given the excessive anthropogenic activities, the accumulation of trace metals in fish tissues has globally increased and caused severe risks for human health [35-38]. Fish from former mining areas have always been misunderstood as a high health risk if consumed due to their likelihood of containing high content of heavy metals in their tissue.

Therefore, this review aims to compile and analyze national information on trace metals levels in fish tissue, together with background concentration of these metals in former mining areas and their food security and human health implications.

Materials and Methods

We have used these following search engines to collect information: Google Search, Science Direct, Scopus, SpringerLink, Research Gate Online, Web of Science. We also performed an extensive review using these following key words in our search: i."Fish Consumption + Former Tin Mining Area", ii. "Former Tin Mining Area + Trace Metals/Heavy Metals", iii. "Fish Consumption + Food Security +Former Tin Mining Area". This review paper is based on 105 journal papers and technical reports, which includes articles from 1998 to 2021. The chronological range was decided within the suggested timeframe due to the difficulty to find online material published before 1998.

Results and Discussion

To understand the risks of consuming fish from former mining areas, studies should acknowledge the whole scenario of potential pollutant flow from the point source, especially from the surrounding area [39]. In this case, trace metals are the main health risk with a former mining pond [40]. Therefore, this section reviews and discusses the content of trace metal in the aquatic surrounding, which covers the background content of trace metals in sediment and water as reported locally.

Trace metal concentration in sediment. Trace metals could be accumulated by aquatic organisms, namely fish, through food, water, and sediment [41]. In an aquatic ecology, these metals are likely to exhibit toxicity toward benthic organisms and eventually affect human health as an ultimate consumer through the food chain, especially concerning the former mining ponds. In Malaysia, Ahmad and Sarah have reported that the total trace metals (Fe, Cu, Zn, and Ni) in sediment from the tin ex-mining catchment areas of Sg. Lembing were greater compared to the concentrations in natural earth crust [41, 42]. Given these findings, sediments in this ex-mine catchment are considered polluted with increased content of metals, especially Zn and Ni, showing a higher concentration than the established limit but still lower than the toxic effect threshold (TET) for sediment (Table 1). However, not all stations in the ex-mine catchment exhibited a similar trend, with several of these metals (Ni and Zn) showing lower concentrations than the TET [43]. Based on their report, the concentration level of trace metal was highly influenced by the distance from the old mining area. The results have shown that the largest content level of trace metal was mainly discovered in the further stations, as a result from the high accumulation of sediment in the downstream area. However, other toxic metal concentrations (such as those of Sn, As, Cr, Cd, and others) were observed in this study.

Ashraf *et al*. reported that the pollution index for trace metals (As, Cr, Cu, Zn, Pb, and Sn) in Bestari Jaya former tin mining pond was mostly within a moderate level (with

the values of 0.01–0.03, 40.21–49.23, 8.90–22.00, and 44.97–77.95 mg/kg), except that for Pb (15.55–18.73 mg/kg) and Sn (1.65–3.12 mg/kg), which was considered to be within a high range for their type [7]. However, the report highlighted that the high Sn concentration was due to the mobile nature of Sn because the site is a former tin mining area, whereas the high Pb level was due to the proximity of the station to a heavily traveled highway, which might influence the particular metal content in the station. They also concluded in their report that the total trace metal content within the study area was still low compared with that in other polluted places. This finding indicates that the point source location also contributes a significant part in influencing the level of trace metals in former mining areas [44].

Kamari *et al*. used different approaches to assess trace metals in the sediment from Bidor, a former mining pond in Perak, by utilizing risk index, with higher trace metal amount for his study, covering 12 different trace metals [45]. This type of index can be applied to evaluate the level of pollution and ecological effects of multiple trace metal pollution in sediments [18]. Their findings suggested that all trace metals in the sediment from the research site posed a low level of ecological risk, with the ranked order of metal concentrations as follows: Sn $(336–727 \text{ mg/kg})$ > Fe $(277–657 \text{ mg/kg})$ > Al $(311–327$ mg/kg) > Mn (157–196 mg/kg) > Zn (33–51 mg/kg) > Pb $(11.3-12.2 \text{ mg/kg})$ > Ni $(1.2-2.6 \text{ mg/kg})$ > Cu $(2.3-3.8 \text{ g/kg})$ mg/kg)> As (0.45–0.88 mg/kg) > Cr (0.42–0.63 mg/kg) $>$ Cd (0.062–0.150 mg/kg) $>$ Hg (0.024–0.061 mg/kg). Seasonal variations of metals were also observed in their study and showed that the metal concentration was higher during the dry season because the slower water flow during this season causes the increased deposition of finer sediment grains, which potentially contain high trace metal contents [46]. Thus, not only the spatial but also the temporal variation of the environment contributes to the metals variation in the sediment of former mining areas.

Another report on trace metal concentration in Danau Kota Lake, Kuala Lumpur [47] showed a considerably lower overall trace metal concentration in sediments compared with other former mining areas within, with the order of $Mn > As > Cu > Ni > Pb$. In general, all the trace metals, except for Mn, were reported to be substantially lower than the established limit (Table 1). To date, trace metals, such as Mn, Sn, Al, and Fe, lack a fix concentration limit in their sediment for global reference because they are naturally abundant based on natural locality and environment [48,49]. However, in this case, Mn concentration (1.18 mg/kg) was lesser than that of the study by Louhi *et al.*, with an average of $3.6 \pm$ 1.2 mg/kg, within a less polluted area [50].

Trace metal concentration in water. Former tin mining field acts not only as natural water catchment areas to control floods but can also potentially be promoted as recreational areas for the locals [51]. Several former tin mining areas were reclaimed as natural habitats for aquaculture activities, where a large number of freshwater fishes are bred [52]. Therefore, in addition to trace metal content in sediment, assessing these metal concentrations in the water column is crucial because aside from bioaccumulation, metals in the water column can be absorbed directly inside fish tissue, especially when water passes through their gills during the breathing process [53, 54]. High metal content in the water column of former tin mining ponds are a concern for most ecologists [55–59].

The concentrations of the overall target of trace metals in water (Mn, As, Cu, Ni, and Pb) in the former tin mining pond at Danau Kota Lake, Kuala Lumpur were below the safe guideline values provided by the National Lake Water Quality Criteria and Standards for Malaysia and World Health Organization (WHO), with the concentration trend of Mn $(1.18 \text{ µg/l}) > As (0.71 \text{ µg/l}) >$ Cu $(0.14 \text{ µg/l}) > \text{Ni } (0.04 \text{ µg/l}) > \text{Pb } (0.01 \text{ µg/l})$ [47]. In accordance with the directive of Interim National Quality Water Standard Malaysia, lake water is classified as class III, indicating that fishing activities are permitted. Thus, in a study, the lake site was secure for public use perhaps because of the lack of any direct contact activities in recent years. However, further treatment is recommended for the lake if it is to be used as a source of water [60].

Hashim *et al*. assessed selected trace metal concentrations, namely, those of Pb, Cu, As, and Zn, in the five former mining lakes in Perak area: Gunung Lang, Taman Indah, Kg. Temiang, Lahat, and Kg. Engku Husin [61]. The outcomes showed that Cu, As, and Zn were lower than the limit set by Department of Environment (DOE) and Ministry of Health (MOH), except for Pb concentration. For example, Kg. Engku Husin had the highest Pb concentration among the four study sites, perhaps due to several active developments in the area that the author failed to disclose. Notably, this past study [61] recorded decreases in trace metal concentrations between November 2015 and December 2015, except for Pb concentration. Meanwhile, the detection of these three trace metals such as As, Cu and Zn were expected because the lakes were former mining ponds. However, the areas were recommended as suitable and safe for water recreational activities because most of them are classified under class II as the values were considered low than the standards set from DOE and MOH.

Through the new method of sequential extraction leaching procedure, a former mining pond in Bestari Jaya Catchment was reported to have high Pb and Sn concentrations in the water column at almost all the study sites [7], with the total concentrations ranging between 28.98 ± 0.19 and 945.23 ± 8.96 µg/l. High Sn and Pb levels were mainly observed in the former tin mining

catchment bound with organics because the study sites are near the river and end up at the bottom sink within the catchment. However, these high trace metal concentrations were only present in 2 of 10 other study sites. The content of other metals, such as Zn, Cu, and As, were lower than the safe limit, with the total concentration range of 52.05 ± 1.53 , 20.13 ± 0.69 , and 1.05 ± 0.03 µg/l, respectively.

However, other places had been recorded to have high trace metal content in the water column, namely that reported in an investigation [45] of a former mining pond in Bidor, Perak. Among the trace metals tested, Sn exhibited the highest concentration (145–258 mg/l) and Cd the lowest (0.4–0.7 mg/l). Important seasonal variations were noted in the other metals tested (As, Cr, Fe, Ni, Pb, and Zn) in water samples. In addition, these levels exceed the limit value in the NWQS (Table 1). The author also highlighted that several of these metals increased by more than 100-fold during the dry season. This high concentration of trace metal availability may result from constant exposure from several point sources [62,63]; however, the author did not discuss this in detail in the report. Finally, the author explained the possible peak concentration during dry season, that is, the water volume in the pond was significantly low in the dry season.

Brief on trace metal concentration in sediment and water from former mining areas. Mining activity has often been associated with evident environmental degradation, mainly in terms of water and sediment quality [57]. Based on several reports, most trace metal levels in former mining areas which had been utilized as secondary use were remaining within the lower range from the established limit. Several reports have recorded trace metals levels exceeding these safe limits, such as those reported by Kamari *et al*. [45]. This finding was limited to areas receiving improper treatment or being treated for secondary use. Low trace metal concentration in Danau Lake, as reported by Hamid *et al.* [47], is a good example of a properly treated former mining area.

In terms of the correlation of trace metal with the sediment, water, and fishes in an area, theoretically, a high trace metal level in an aquatic environment implies the high metal content in the tissue of living organisms that inhabit the environment [66]. This condition, however, depends on uncontrolled environmental variables, such as water current, temperature, pH, electro conductivity, and others [66, 67]. The difference species of the organism also will influence the correlation of trace metal with the sediment and water as they have various regulations types within their body. For example, Merciai *et al*. stated that the content of several trace metals in fish tissue correlated well with water and sediment, whereas other fish species lacked any pattern of correlation [64]. Bervoets, L. and Blust did not find

any relationship pattern in their study [65]. Notably, we failed to review the correlation of trace metals in the sediment, water, and fish tissue in the former tin mining pond because most of the reports lacked this particular information in their study.

Trace metal content in fish and health risk assessment. According to Ashraf *et al*., the assessment of trace metal concentration in fish tissue could be significant based on two major factors: first, from the public's viewpoint, concern has been raised against the requirement to measure these metals accumulation, especially those which cause a health hazard toward humans [27]. Second, from the aquatic environment viewpoint, the major issue is to avoid or minimize biological deterioration and the source, which jeopardizes ecological balance. Determining trace metal accumulation in fish tissue, especially from former mining ponds, is a major concern for researchers around the world [68–70, 20] Hence, this section reviews and discusses the concentrations of trace metals in fish tissue and the health risk assessment of fish consumption from former tin mining ponds, as reported locally.

Low *et al*. performed a research on health risk assessment and trace metal accumulation in the tissue of tilapia fish from a former tin mining pond in Jelebu, Negeri Sembilan [48]. In their report, the metal accumulation pattern in tilapia revealed a resembling tendency to a certain extent, regardless of the sampling site location: Fe $> Zn > As > Se > Mn \approx Cu > Cd$. These patterns may have been due to the equilibrium or physiological regulation process of tilapia itself [71]. Another possible reason that might have contributed to these circumstances was the natural background level of trace metals within the proximity of the area [72–74]. Toxic metals, such as As, Cd, and Hg, are non-essential and could be toxic at trace concentrations, whereas other metals, such as Fe, Cu, Zn, Se, and Mn, are substantial to biological systems [75, 76]. However, these essential materials can also be toxic in excess concentrations. Table 2 shows a comparison with established limits and other reports studied in this paper.

To date, maximum limits are recommended for the consumption of trace metals, such as V, Mn, Fe, Co, and Se. This finding may be due to the possibility that th $+e$ ingestion of these metal species via fish (which is tilapia in this study) can be assumed non-significant scenarios because every one of them provides less than 1% hazard index (HI), which was set by the US Environmental Protection Agency (USEPA). However, the concentration was lower than the limit set by USEPA for risk-based concentration [77]. Other metals were significantly lower than the maximum limits permitted by the Malaysian Food Act 1983 and Food Regulations 1985 [78]. However, the author noted a particular metal, namely, As, that showed a higher value than the limit set by the legal Malaysian allowance. As explained in the author's report, the As species discovered in tilapia differed substantially because its concentration was typically measured based on the oxidation state and/or organic substituents from the innocuous arsenobetaine to toxic inorganic arsenic species [79, 48]. Given this condition, the USEPA advocates inorganic As uptake for the assessment of human health risk. Therefore, the mean inorganic As level should be lower (by approximately 10% than the reported concentration) than the limit set by USEPA [77]. Based on the reasonable maximum exposure analysis, the author also suggested that the consumption of tilapia over a lifetime from any production site is hardly causing any harmful impacts on Malaysians.

Another [27] research on trace metal contents in 15 fish species from a former tin mining area of Bestari Jaya, Kuala Selangor showed that the average Sn concentration was the highest (56.34–153.45 mg/kg) compared with other metal species, with the order of $Sn > Pb > As > Zn$ > Cu, in fish muscle tissue. This previous research particularly focused on metals in fish muscle given that people generally only consume the muscles and not the other parts of the fish [80–83]. With the presence of several fish species, the author highlighted that the metal concentrations varied, with several fishes reported to have extremely high concentrations, whereas other samples showed relatively low concentrations. A high Sn concentration was expected because the breeding area was a former tin mining area, showing almost the same trend as the other former tin mining studies [84, 85].

The author also highlighted the probability that the source may be derived from various origins, such as nearby oil palm and rubber plantation catchments. However, the reported concentration in the research was twice smaller compared to the allowed level set by the US Food and Drug Administration [86] (Table 2). In general, the overall trace metal content in all the fish tissues from the study site were lower than the established limits (Table 2). Their study conducted health risk assessments according to the integration data from trace metal analysis and the presumed assumption rate based on USEPA guidelines. Specifically, HI was utilized in their study to determine the probability of adverse health effects toward humans [87], and the author concluded that no instantaneous health risk due to the fish species consumption was observed in the former tin mining area. However,

constant monitoring regarding runoff discharge was highly recommended in their study.

Kamari *et al*. focused on trace metal concentration in the tissue of fish (*Anabas testudineus*; climbing perch) in former mining areas between seasons [45]. Their findings showed that several metal species, such as Pb, Mn, and Sn, did not exhibit notable distinctions in their levels between seasons. However, mostly, fish muscle accumulated greater trace metal contents in the dry season, with levels of up to 2.78 times higher compared to that in the wet season. This finding may be due to the high filtration and metabolism of aquatic organisms during the high-temperature period [88-92]. Achary *et al.* also have found that the zooplankton was discovered to be greater in the summer season [96]. Another possible reason is the change in plankton composition due to differences in environmental conditions [93-95]. Different compositions of plankton may cause changes in their ability and capacity to accumulate trace metals [96,97]. Fang *et al.* have found that the plankton had bioaccumulated more trace metals compared to the greater trophic organisms [97].

Kamari *et al*. measured the health risk assessment of fish consumption through the determination of estimated daily intake (EDI) of trace metal [45]. According to the WHO, dietary exposure approaches of observing consumption of pollutants in fish, such as the EDI method, are a reliable tool for health risk assessment [98- 100]. The authors highlighted that the EDI levels of As and Cr concentrations exceed the permissible levels in the wet and dry seasons. This finding has been explained extensively in the previous paragraph and therefore will be not explained in this section. Furthermore, all the trace metal contents in all the fish tissues in the research site were lower than the established limit (Table 2). Thus, the consumption of fish in their study sites over a lifetime is unlikely to have any harmful effects.

In general, although several limited studies showed concern regarding health risks because of the consumption of fish from tin mining ponds, specifically in Malaysia, based on reports, several types of health risk assessment of fish tissue have been implemented to ensure safety of the consumption of these fishes. The overall trace metal levels in all the fish tissues were lower than the established limit and could be regarded acceptable for human consumption.

	Place Reported	Cu	\mathbf{Zn}	As	C d	Pb	Cr	Ni	Sources
Sediment (mg/kg)	Sg.Lembing, Pahang	110.23	408.42					44.00	$[41]$
	Bestari Jaya, Selangor	8.90-22.00	44.97-77.95	$0.01 - 0.03$	\blacksquare	15.55-18.73	40.21-49.23	\blacksquare	$[27]$
	Bidor, Perak	$2.3 - 3.8$	$33 - 51$	$0.45 - 0.88$	$0.062 - 0.150$	$11.3 - 12.2$	$0.42 - 0.63$	$1.2 - 2.6$	$[45]$
	Danau Kota Lake, Kuala Lumpur	0.14		0.71	\overline{a}	0.01	$\overline{}$	0.04	$[47]$
	SQG								
	ANZECC ISQG-Low	65.00	200.00	20.00	1.5	50.00	80.00	21.00	$[102]$
	Hong Kong ISQG-Low	65.00	200.00	$8.2\,$	1.5	75.00	80.00	40.00	$[103]$
	TET	86.00	540.00	17.00	3.00	170.00	100.00	61.00	$[43]$
	Place Reported	Cu	Zn	As	Cd	Pb	Cr	Ni	Sources
Water (mg/l)	Danau Kota Lake, Kuala Lumpur	< 0.01		< 0.01		< 0.01		< 0.01	$[47]$
	Perak	$0.00 - 0.01$	$0.02 - 0.03$	$0.01 - 0.05$	\blacksquare	$0.42 - 1.30$	\blacksquare	$\overline{}$	$[51]$
	Bestari Jaya, Selangor	0.020	0.052	< 0.01	$\overline{}$	0.029	\sim	\blacksquare	$[27]$
	Bidor, Perak	$43 - 75*$	$87 - 141*$	$4.30 -$ $5.70*$	$0.40 - 0.70*$	52-98*	$2.20 - 3.60*$	$2.10 - 6.30*$	$[45]$
	WQG								
	NWQS	0.05	5.00	0.05	0.01	0.05	0.05	0.05	$[104]$

Table 1. Comparison between Locally Reported Trace Metals in Sediment and Water of Former Mining Areaa with Established Permissible Limit

SQG: Sediment Quality Guideline; **ISQG**: Interim freshwater Sediment Quality Guideline; **ANZECC**: Australian and New Zealand Environment and Conservation Council; **TET**: Toxic Effect Threshold; **WQG**: Water Quality Guideline; **NWQS**: National Water Quality Guideline. *Extreme Value

Conclusion and Perspective

Given the information gathered, the trace metal contents in the sediment, water, and fish tissues in former mining areas are mostly lower than the established limit except for those areas that are being treated for secondary use. Trace metal concentration in the fish tissue also showed the same trend, that is, a high metal concentration in fish tissue implies the improper or unfinished treatment of a former mining site. Meanwhile, in the context of food safety, local reports have shown that the consumption of aquaculture products from former mining areas over a lifetime is unlikely causing any hazardous impact and hence was regarded to be safe. However, these findings are restricted based on the limited number of studies in Malaysia. Therefore, extensive studies regarding this particular issue are highly recommended.

Acknowledgements

The authors would like to record appreciation to Universiti Putra Malaysia for providing financial support throughout this project.

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