

CONTRIBUTION OF FISH CONSUMPTION TO CADMIUM AND LEAD INTAKES IN COASTAL COMMUNITIES OF WEST KALIMANTAN, INDONESIA

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ABSTRACT

Consumption of both marine and freshwater fishes is considered as a healthy way to obtain diet, but the ingestion of contaminated fishes remains a concern. Our study is conducted to assess cadmium (Cd) and lead (Pb) contents in various fishes consumed by coastal communities in six districts of West Kalimantan (Indonesia) and their health risks to the communities. Specifically, the objectives of this study are to determine the average weekly intakes (AWIs) of cadmium and lead based on dietary intake of fishes, then estimate health risk associated with fish consumption using the risk quotients (RQs) method. We also use food frequency questionnaires (FFQ) filled by 696 respondents to assess fish consumption habit of the communities in West Kalimantan who consumed up to 30 fish species in 2012 and 2013. Our results show that their average weekly fish consumption is 135.7 ± 66.3 g per week, which contributes to AWI values of 0.38 ± 0.55 $\mu\text{g/kg}$ body weight (bw) per week for Cd and 0.87 ± 0.65 $\mu\text{g/kg}$ bw per week for Pb. The RQ values indicate a low risk of Pb coming from fish consumption, but the RQ value for Cd is high for the children age group. In conclusion, fish consumption in the six districts of coastal communities in West Kalimantan generally does not pose any health risk from Pb. Conversely, Cd intake from fish consumption has a potential risk for children. As such, regular monitoring and additional studies are needed on potential contribution of Cd intake from other food sources.

Keywords: average weekly intake (AWI), risk quotients (RQs), heavy metals, health risk, Kalimantan

INTRODUCTION

Heavy metals, especially cadmium (Cd) and lead (Pb), have pervasive distribution and rising concentrations in aquatic ecosystems, thus posing threats to the environment and human health (Domiano *et al.*, 2011; Adel *et al.*, 2016). These heavy metals, released into aquatic systems by anthropogenic activities, are accumulating in water, suspended solids and sediments, and subsequently in aquatic biota. Therefore, communities with a dietary habit based on fish consumption may have a higher risk to be exposed to and ingest heavy metals.

Ongoing anthropogenic activities in the West Kalimantan Province of Indonesia may contribute to heavy metals intake in humans. Three main sources of anthropogenic activities in the province are large usage of pesticides in palm oil industries, a large number of boats as river transportation and illegal mining activities (Arifin *et al.*, 2014). These activities increasingly release and remobilize heavy metals into aquatic systems that finally enter the food chain. Previous studies have shown that almost all commercial fishes and shellfishes contain heavy metals, with some of them accumulate high Pb and Cd in their tissues (Setyawati and Nofritra, 2002; Purbonegoro

et al., 2014). Indeed, more than 90% of heavy metals uptake to humans is mainly through water and food intake (Hassan *et al.*, 2016).

On the other hand, traditional fisheries contribute up to 67% of the total fishery production in West Kalimantan (BPS, 2013), where the coastal communities rely on marine and freshwater fishes as a main source of protein. High in micronutrients but low in saturated fatty acids, fishes are known to be beneficial to health (Hoekstra *et al.*, 2013; Afonso *et al.*, 2013; Neff *et al.*, 2014). However, contaminated fishes bring health risks to humans (Harris *et al.*, 2009; Gao *et al.*, 2016). Previous studies have reported that exposure to Cd and Pb through fish consumption causes major diseases in humans such as renal failure, liver damage and symptoms of chronic toxicity in the kidney (Bosch *et al.*, 2016).

With the need for estimating the health risks of contaminants through dietary intake, previous literatures have employed a number of methods including the hazard quotient (HQ), target hazard quotient (THQ) and hazard index (Zhu *et al.*, 2015; Saleh and Marie, 2015; Zhu *et al.*, 2016). Additionally, the food frequency questionnaire (FFQ) approach can be performed to study fish consumption habit in coastal communities. The FFQ is simple and low cost, yet it can obtain complete information from large populations, therefore the method is commonly used in dietary intake assessments (Athanasiadou *et al.*, 2016; Saeedi *et al.*, 2016; Moghames *et al.*, 2016; Nybacka *et al.*, 2016).

Therefore, our study assesses the risk of heavy metals intake to humans via fish consumption in West Kalimantan. The objectives of our study are two-folds; firstly to assess the average weekly intakes (AWIs) of Cd and Pb based on dietary intake of fish; secondly to estimate risk levels of Cd and Pb contaminants to the coastal communities in West Kalimantan. The risk quotient (RQ) method is adopted in this study for estimating the risk of contaminants via fish consumption as has been applied in other studies (MPP-EAS, 1999; USEPA, 2000; Chien *et al.*, 2006; Sioen *et al.*, 2009).

MATERIALS AND METHODS

Determination of Metal Concentration in Fish Samples

The field sampling component was conducted between 2012 and 2013. Fish species were selected based on the dietary habits of coastal communities, with a total of 30 species of fish/shellfish consisted of 8 species of freshwater fish and 24 species of marine fish. Cd and Pb contents in fish tissues were determined following a method by the ASEAN-Canada Cooperative Programme on Marine Science II (McPherson *et al.*, 1999). Each sample of fish or shellfish was analyzed in triplicate in the following manner. Tissue sample of each specimen was heated in an oven at 105°C for approximately 12 hours, then the dried tissue sample was ground using an agate mortar. A portion of the powdered sample (about 1 g weighed with a precision of 0.1 mg) was poured into a glass beaker, added with a strong acid (HNO₃) and heated on a hot plate at 85°C for approximately 8 hours. Then H₂O₂ was added to the sample to achieve a complete digestion process.

The liquid sample was then analyzed using a Flame Atomic Absorption Spectrophotometry (Varian SpectrAA-20 Plus). The detection limits of the instrument for Cd and Pb are 2 µg/L and 10 µg/L, respectively. The reference material (SRM NIST 1566B), which has a certified Cd content of 2.48 ± 0.08 mg/kg and Pb content of 0.3808 ± 0.09 mg/kg, was used to validate the methods; where we obtained 2.51 ± 0.06 mg/kg for Cd and 0.337 ± 0.17 mg/kg for Pb (N = 3) in our study.

Dietary Survey

The fish consumption habit of the communities was investigated using food frequency questionnaires (FFQ). There were 696 respondents from six districts of the West Kalimantan Province, *i.e.*, Sambas, Kota Singkawang, Bengkayang, Kota Pontianak, Kubu Raya and Sanggau (Figure 1). The fish consumption habit information include fish species, the frequency and the quantity of fishes consumed. Respondents were also interviewed for their profile which consists of information on their age, sex, body weight and occupational groups.

Cadmium and Lead Intake

The estimated average weekly intakes (AWIs) of Cd and Pb from fish consumption are calculated based on a formula from MPP-EAS (1999) and USEPA (2000):

$$AWI = \frac{A \times B}{BW} \quad (\text{Eq.1})$$

where AWI is the average weekly intake of each contaminant ($\mu\text{g}/\text{kg}$ body weight per week), A is weekly fish consumption (g per week), B is concentration of contaminant ($\mu\text{g}/\text{g}$ ww) and BW is body weight (kg).

The health risk of respondents due to fish consumption was calculated using the risk quotient (RQ) method (MPP-EAS, 1999):

$$RQ = \frac{AWI}{TWI} \quad (\text{Eq.2})$$

where AWI is the average weekly intake ($\mu\text{g}/\text{kg}$ bw per week) and TWI is the tolerable weekly intake. The TWI for Cd is $2.5 \mu\text{g}/\text{kg}$ bw per week based on the Panel on Contaminants in the Food Chain of the European Food Safety Authority (Alexander *et al.*, 2009). For Pb, we adopt a provisional tolerable weekly intake (PTWI) of $25 \mu\text{g}/\text{kg}$ bw per week based on the FAO/WHO Joint Expert Committee on Food Additives and Contaminants, JECFA) (Arnich *et al.*, 2012).

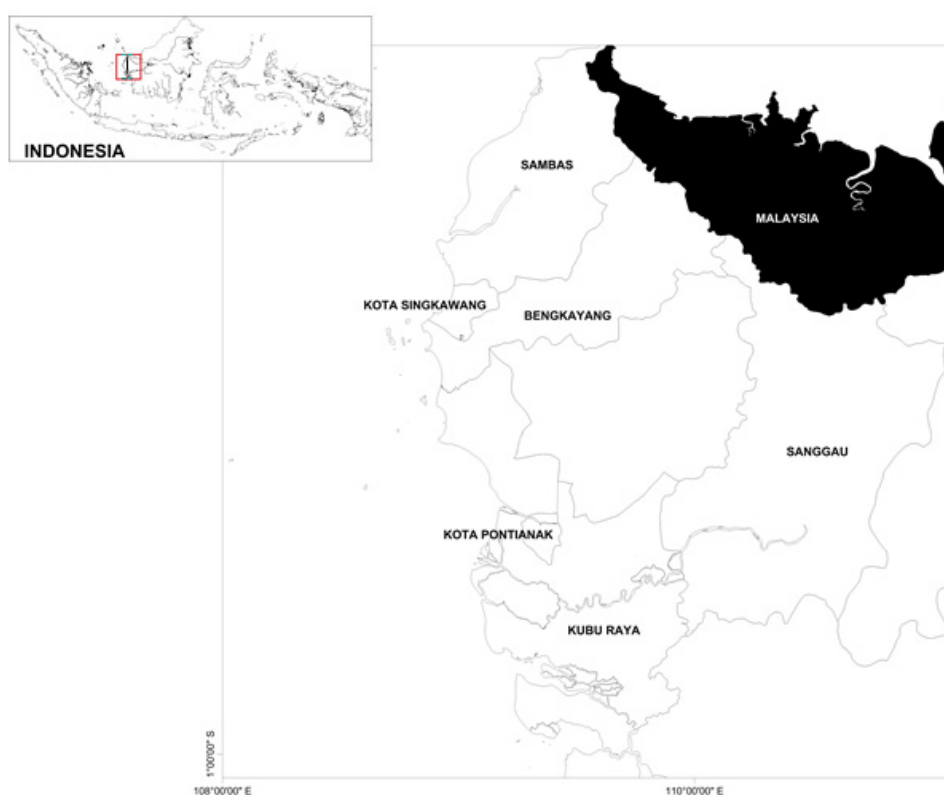


Figure 1. Study sites consisting six districts in the West Kalimantan Province.

Statistical Analysis

All statistical analyses are estimated using SPSS23 (IBM Corp.) and figures are presented using the ggplot2 function in the R environment (R v3.3.1; R Core team). The normality and homogeneity of data are determined using the Shapiro-Wilkinson and the Levene's tests. If the data are not normally distributed, a non-parametric procedure (Kruskal-Wallis H test) is applied to the data.

RESULTS

Profile of Respondents

A total of 696 respondents were interviewed from six districts of the West Kalimantan Province, with their ages ranging from 12 to 65 years old. The respondents are composed of almost equal numbers of male and female (Table 1). The average values of their body weight are 51.4 kg (range of 22.3 - 62.0 kg) for the male

group and 45.4 kg (range of 20.7 - 59.2 kg) for the female group.

Table 1. Profile of respondents of the FFQ survey in West Kalimantan, Indonesia.

	Respondents (N)	Frequency (%)
Gender:		
Male	332	47.7
Female	364	52.3
Age (year):		
Children (<14)	38	5.5
Adolescent (15-24)	200	28.7
Adult (25-44)	299	43.0
Senior (45-65)	159	22.8
Occupation:		
Student	191	27.4
Civil servant	137	19.7
Private sector	188	27.0
Fisherman	58	8.3
Housewife	122	17.5
Residence:		
Kubu Raya	153	22.0
Sanggau	44	6.3
Sambas	133	19.1
Kota Pontianak	131	18.8
Kota Singkawang	119	17.1
Bengkayang	116	16.7

Average Weekly Intake of Fish

The fish consumption of six coastal communities in West Kalimantan is highly variable, ranging from daily to once a week. In general, the frequency of fish consumption of the coastal communities in West Kalimantan is 2.1 ± 0.8 times per week, with an average weekly intake of fish of 134.6 ± 59.5 g per week. The fish consumption habit of the coastal communities is significantly different between gender groups (Kruskal-Wallis H test, $\chi^2(1) = 4.68$, p-value = 0.03), but similar between age groups (Kruskal-Wallis H test, $\chi^2(3) = 4.63$, p = 0.20).

Fish species consumed by the respondents from six districts are: island mackerel, hairtail fish, barred queenfish, Indo-Pacific king mackerel,

Indian mackerel, goldband goatfish and all marine catfishes (Table 2).

Estimation of the Average Weekly Intake of Cd and Pb

The estimated weekly intake (AWI) of Cd and Pb through fish consumption for different age and gender groups is presented in Figure 2. Fish consumption contributes to an average Cd intake of 1.01 ± 1.68 , 0.35 ± 0.28 , 0.35 ± 0.35 , and 0.31 ± 0.30 $\mu\text{g/kg bw}$ per week for the children, adolescent, adult and senior age groups, respectively. In contrast, Pb intake is more than twice higher than the Cd intake, on average 1.78 ± 1.40 , 0.88 ± 0.65 , 0.80 ± 0.45 and 0.73 ± 0.36 $\mu\text{g/kg bw}$ per week for the children, adolescent, adult and senior age groups, respectively. The weekly intakes of Cd and Pb are significantly different between age groups with children having the highest intake (Kruskal-Wallis H test, $\chi^2_{\text{Cd}}(3) = 25.06$, $\chi^2_{\text{Pb}}(3) = 40.20$, $p < 0.05$). And, male and female gender groups have similar intake rates (Kruskal-Wallis H test, $\chi^2_{\text{Cd}}(1) = 0.04$, $\chi^2_{\text{Pb}}(1) = 3.3$, $p > 0.05$) (Figure 2).

Estimation of Risk Quotient of Cd and Pb

Risk assessment due to the dietary intakes of Cd and Pb shows that RQ values, calculated based on the gender and age groups are mostly far below the maximum risk quotient value (*i.e.* RQ = 1.0) for all respondents from all the six districts (Table 3). The RQ values of Cd and Pb range from one-hundredth to half of the maximum RQ. Both RQ_{Cd} and RQ_{Pb} for male and female respondents are significantly different (Kruskal-Wallis H test, $\chi^2_{\text{Cd}}(1) = 0.04$, $\chi^2_{\text{Pb}}(1) = 0.75$, $p > 0.05$) and highly vary among respondents. Based on the age of respondents, RQ_{Cd} values range from 0.12 to 0.41 (with an average of 0.15), and RQ_{Pb} values range from 0.03 to 0.07 (average of 0.20). There is also a significant difference between age groups (Kruskal-Wallis H test, $\chi^2_{\text{Cd}}(3) = 25.06$, $\chi^2_{\text{Pb}}(3) = 91.64$, $p < 0.05$).

Table 2. Cadmium and lead concentrations (mean \pm standard deviation), N = number of fish samples.

No	English Name (Local Name)	Scientific Name	N	Cd	Pb
				(µg/g ww)	(µg/g ww)
Freshwater fFsh					
1	Striped snake head (gabus)	<i>Channa striata</i>	6	0.05 ± 0.04	0.32 ± 0.10
2	Striped snake head (toman)	<i>Channa</i> sp1.	2	0.03 ± 0.01	0.09 ± 0.12
3	Wallago (juara)	<i>Wallago attu</i>	7	0.06 ± 0.04	0.30 ± 0.13
4	Sheatfish (lais sengarat)	<i>Belodontichthys dinema</i>	6	0.03 ± 0.00	0.18 ± 0.00
5	Three spot gourami (biawan/sepat)	<i>Trichogaster trichopterus</i>	3	0.02 ± 0.00	0.26 ± 0.00
6	Common sheatfish (lais lajong)	<i>Cryptogoterus apogon</i>	7	0.03 ± 0.00	0.29 ± 0.11
7	Asian redbtail catfish (baung)	<i>Macrones nemurus</i>	10	0.04 ± 0.04	0.29 ± 0.08
8	Pangas catfish (patin laut)	<i>Pangasius pangasius</i>	1	0.01 ± 0.00	0.18 ± 0.00
Marine Fish					
9	Hairtail fish (timah-timah)	<i>Trichiurus</i> sp.	12	0.05 ± 0.04	0.33 ± 0.26
10	Marine catfish (duri kera)	<i>Arius</i> sp1.	11	0.04 ± 0.02	0.32 ± 0.04
11	Marine catfish (duri udang)	<i>Arius</i> sp2.	14	0.04 ± 0.04	0.46 ± 0.69
12	Marine catfish (belukang)	<i>Arius</i> sp3.	11	0.03 ± 0.03	0.30 ± 0.10
13	Marine catfish (duri putih)	<i>Arius</i> sp4.	20	0.04 ± 0.03	0.53 ± 0.68
14	Marine catfish (duri moncong)	<i>Arius</i> sp5.	11	0.04 ± 0.04	0.32 ± 0.30
15	Marine catfish (sembilang)	<i>Arius</i> sp6.	9	0.05 ± 0.08	0.32 ± 0.18
16	Spotted catfish (mayung jahan)	<i>Arius maculatus</i>	1	0.04 ± 0.00	0.01 ± 0.00
17	Croaker fish (gulama tora)	<i>Johnius</i> sp1	10	0.07 ± 0.03	0.26 ± 0.24
18	Croaker fish gulama papan)	<i>Johnius</i> sp2.	12	0.04 ± 0.03	0.33 ± 0.34
19	Goldband gotfish (biji nangka)	<i>Upeneus moluccensis</i>	9	0.05 ± 0.00	1.39 ± 0.00
20	Doublelined Tonguesole (sebelah merah)	<i>Paraplagusia bilineata</i>	5	0.03 ± 0.02	0.51 ± 0.35
21	Barred queenfish (talang-talang)	<i>Chorinemus tala</i>	3	0.10 ± 0.00	0.31 ± 0.00
22	Indian mackerel (kembung laki-laki)	<i>Rastrelliger kanagurta</i>	4	0.10 ± 0.00	0.54 ± 0.00
23	Indo Pasific king mackerel (tenggiri papan)	<i>Scomberomorus guttatus</i>	2	0.05 ± 0.00	1.14 ± 0.00
24	Island mackerel (kembung)	<i>Rastrelliger faughni</i>	3	0.35 ± 0.00	0.28 ± 0.00
25	Kelee shad (tamban)	<i>Hilsa kelee</i>	8	0.10 ± 0.04	0.40 ± 0.38
26	Black pomfret (bawal peda-peda)	<i>Parastromateus niger</i>	5	0.06 ± 0.00	0.15 ± 0.00
27	Cuttlefish (sotong kodok)	<i>Sepia</i> sp.	4	0.13 ± 0.00	0.27 ± 0.00
28	Mangrove cockle (kerang kepah)	<i>Anandontea alba</i>	38	0.07 ± 0.03	0.29 ± 0.13
29	Squid (cumi)	<i>Teuthida</i> sp.	2	0.11 ± 0.00	0.25 ± 0.00
30	Blood cockle (kerang darah)	<i>Anadara</i> sp.	52	0.10 ± 0.11	0.16 ± 0.24
National Standard (Indonesia)				0.10	0.30
European Union Standard				0.05	0.30

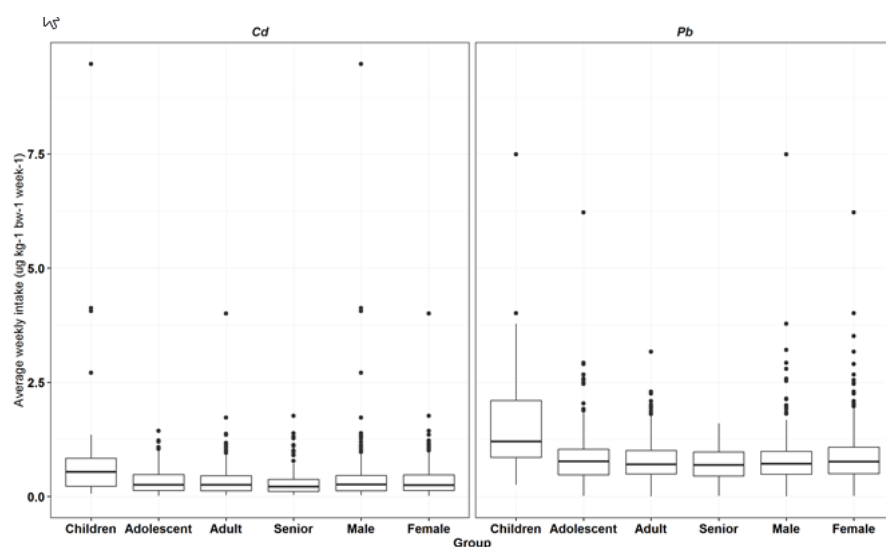
**Figure 2.** Distribution of average weekly intake of Cd and Pb ($\mu\text{g/kg bw}$ per week) among different age and gender groups. The graph is shown as median (5, 25, 75, and 95 percentile) and outliers.

Table 3. RQ values of Pb and Cd intake.

Classification		RQ value	
		Cd	Pb
Sex	Male	0.16 ± 0.26	0.03 ± 0.02
	Female	0.14 ± 0.14	0.04 ± 0.03
Age	Children (<14)	0.41 ± 0.66	0.07 ± 0.06
	Adolescent (15-24)	0.14 ± 0.11	0.04 ± 0.03
	Adult (25-44)	0.14 ± 0.14	0.03 ± 0.02
	Senior (>45)	0.12 ± 0.12	0.03 ± 0.01

Note: maximum RQ value = 1.0

DISCUSSION

Fish is a major part of human diet in many provinces in Indonesia, however, the problems of coastal pollution due to industrial and development activities resurface almost every year. Therefore, it is not surprising that numerous studies have been carried out on measuring non-essential and toxic metals such as Hg, Cd, and Pb in many parts in Indonesia (Kamaruzzaman *et al.*, 2010; Putri *et al.*, 2012; Koesmawati and Arifin, 2015). Fish consumption is one of the routes for heavy metal uptake from food sources (Waheed *et al.*, 2013). And, there is a positive correlation between the amount of fish consumed and heavy metal intake by humans (Sioen *et al.*, 2009). Therefore, it is important for the community to have reliable information related to risks and benefits of fish consumption (*e.g.* Sidhu, 2003; Neff *et al.*, 2014).

Our study shows that out of 30 species of fish consumed in West Kalimantan, 13% of fish species exceed the safety level for Cd concentration ($0.10 \mu\text{g/g ww}$) set by the Indonesian National Standard in SNI 7387 2009 (SNI, 2009). However, if the number is compared to the regulation established by the EU, there are 57% of fish species exceeding the permitted value ($0.05 \mu\text{g/g ww}$) (European Commission, 2006). In contrast to Cd, 67% of Pb concentrations in fish species are beyond the maximum value ($0.30 \mu\text{g/g ww}$) set by the SNI and EC standard (SNI, 2009; European Commission, 2006). Therefore, it is clearly urgent to inform the consumers, not only the benefits of consuming fishes, but also the risk of consuming fishes contaminated with heavy metals Cd and Pb.

In our current study, the age group, body weight group and consumption habit are major factors in differentiating the average weekly intakes (AWIs). The weekly intakes of Cd and Pb highly vary between age groups, likely reflecting different preferences and quantities in fish consumption as well as body weights. In general, the average weekly intakes of cadmium and lead for all age groups are below the established TWI of Cd ($2.5 \mu\text{g/kg bw per week}$) and PTWI of Pb ($25 \mu\text{g/kg bw per week}$); though for several respondents, their Cd intakes exceed the safety level. The weekly intakes of Cd and Pb in the children group, for example, tend to be higher compared to the rest of age groups (Figure 2). Although children have a similar amount of fish consumption with other age groups in our study, because of their smaller body weights, they have higher doses of toxicant per kilogram of body mass. In all, our study suggests that with the current habit of fish consumption and the concentration of Cd and Pb in fishes, the consumption of fishes from traditional markets in West Kalimantan does not lead to high intake of Cd and Pb for the local coastal communities.

It should be noted that the intake of heavy metals depends on several associated factors, such as ethnicity, consumption habit, gender, age and body weight (Wilhelm *et al.*, 2002; Vieira *et al.*, 2011). In our study, the children age group shows the highest intake of Cd and Pb compared to other age groups. High intakes of both metals in the children age group also could be contributed to the feeding behaviour and food preference of their parents, especially for fisherman communities whose majority are low income families. Indeed, a previous study (Afonso *et al.*, 2013) showed that consumption habits have a significant role in heavy metal intake. This condition raises a concern on potential cadmium over-exposure in children.

The estimated Cd and Pb AWIs herein are only based on fish consumption, thus do not consider other sources of cadmium and lead intake such as vegetables, meat and rice (*e.g.* Zhou *et al.*, 2016). Consequently, the total dietary intake of cadmium might be higher than the current estimation for the coastal communities of West Kalimantan, especially for the children age group. Therefore, future works need to consider possible Cd intake from other food sources.

Risk assessment due to dietary intakes of cadmium and lead shows that the RQ values are generally far below the maximum risk quotient value ($RQ = 1.0$) for all respondents, except for several respondents belonging to the children age group. Pb concentrations in fish from coastal waters of West Kalimantan do not pose any health problem. In contrast, Cd intake from fishes may risk a portion of the children age group whose RQ values could reach the safely level ($RQ_{Cd} = 1.0$). Therefore, preventive steps should be taken particularly for children as different age groups have different potential health risks. In addition, children with their lightweight body masses, limited knowledge on food preference, developing nervous systems, higher intestinal absorption and lower renal excretion; are the most vulnerable group to heavy metal exposure (Ilmiawati *et al.*, 2015; Bosch *et al.*, 2016). Therefore with long term exposure, children are more susceptible to the risk of heavy metals toxicity especially cadmium compared to adults. Furthermore, any risk assessment of heavy metals also should takes into account some potential Cd and Pb intakes coming from other food sources and a combination effect from other paths of heavy metals intake.

CONCLUSION

The average weekly fish consumption for six districts in West Kalimantan in 2012 and 2013 is 135.7 ± 66.3 g per week, which contributes to the intakes of Cd and Pb up to 0.38 ± 0.55 µg/kg bw per week for Cd and 0.87 ± 0.65 µg/kg bw per week for Pb, respectively. The Risk quotient (RQ) values of Pb are less than 1. Hence, there is no significant health risk of Pb to the coastal communities in West Kalimantan, if dietary intakes only come from fish consumption. In contrast, the risk of Cd intake is potentially high for children age group.

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