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Total mercury of marine fishes in Natuna Islands area, Indonesia: Risk assessment for human consumption

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Abstract. Bioaccumulation of mercury (Hg) in marine has been a widespread health concern. It is important to assess Hg concentration in fish and evaluate the health risks in the Natuna, a region with high fish consumption in Indonesia. Research carried out in 2018 in the Natuna Islands aimed to provide information about the mercury content in fish consumption and the approach steps to minimize the risk of bioaccumulation. The assessment was carried out on 77 commercial fish samples in the Natuna area, Indonesia. The results showed that the concentration of total Hg in *Lutjanus sebae*, *Epinephelus aerolatus* and *Gymnocranius frenatus* exceeded the permissible limit (0.5 mg/kg for fish and fishery products) according to BPOM/SNI/JECFA. The sources of mercury contamination are not elaborated in detail in this study, but a number of natural processes including vegetation surfaces, water bodies, wild fires, may be the main factor. It is a fact that mercury contamination still provides a risk for human health because of bioaccumulation. So, it is recommended to introduce Mean Weekly Intake (MWI) limit, which is a simple tool for guidance on fish consumption based on body weight and age. MWI will guide how many fishes can be consumed for an adult with 60 kg and a child with 10 kg body weight in relation to Hg concentration in the consumed fish.

1. Introduction

More than 75% of the world's marine resources are fish, which are important commodities for human consumption. As an archipelago, Natuna located in the Karimata Strait, Indonesia, has a high level of fish consumption. Fish is an important commodity because it is a source of protein and nutrients for humans, especially children [1]. However, the development and increase of human activity on land and in the open seas has produced anthropogenic contaminants such as metal and organic compounds to be aware of. Mercury is a highly toxic metal in organic and inorganic forms. Organic mercury is harmful to organisms and can be accumulated in human through seafood consumption [1].

Mercury can be found in the environment from natural and anthropogenic sources [2]. Anthropogenic sources from human activities release elemental mercury (Hg^0), reactive gaseous mercury (RGM) and particulate mercury. Elemental mercury had a long lifetime in the atmosphere, while the others are short-lived in the air and deposited near the emission source [3]. Several sources of natural processes release mercury, including vegetation surfaces [4,5], water bodies [6,7], wild fires [8–10], as well as the re-emission of deposited mercury [3]. These processes are believed to release mainly Hg^0 [3]. Methyl mercury, CH_3Hg^+ , and dimethyl mercury, $(\text{CH}_3)_2\text{Hg}$ in the water reservoirs can emerge to the environment, both as a result of chemical reactions and under the impact of biological factors like



microorganisms [11,12]. Present in organic and inorganic forms, methyl mercury is the most common form of organic mercury in the environment [13]. It is also considered the most toxic compounds in the aquatic food web [14]. Food based on marine sources other than fish and other products may contain mercury but mostly in the form of inorganic mercury. Inorganic mercury is considerably less toxic than methyl mercury. Methyl mercury is highly toxic, mainly to the nervous system and also the developing brain as the most sensitive target organ.

The Food and Agriculture Organization/World Health Organization Joint Expert Committee on Food Additives (JECFA) has defined a provisional tolerable weekly intake (PTWI) for total mercury (THg) of 1.6 $\mu\text{g/kg-body weight/week}$. The PTWI estimated the acceptable level of a substance in food, expressed on body weight (BW) basis, that can be ingested weekly for a long-time exposure without adverse effect [15]. The PTWIs for a 60 kg adult is equivalent to 0.096 $\mu\text{g/week}$ and child equivalent to 0.016 $\mu\text{g/week}$. Almost 100% Hg present in fish and seafood occurs as methyl mercury, so measurement of MeHg exposure often uses the total Hg concentration in fish as a basis [16]. The estimated intakes of mercury vary by country, depending on the amount and the type of fish consumed [17]. A population that consume large predatory fish, often have a higher concentration of methyl mercury because of bioaccumulation at the top of the food chain, will have high intakes of mercury. To estimate the risks to human health from the consumption of the fishes, assessments were made such as:

- a) Direct comparisons with maximum permissible limits (MPLs) by the SNI 7387.2009 [18], JECFA 2006 [19] and BPOM no.5 2018 [20].
- b) The weight of fishes that would need to be consumed weekly (Mean Weekly Intake/MWI) [21].

Head of National Agency of Drug and Food Control Republic of Indonesia (BPOM) and Indonesian National Standard (SNI) assigned that the maximum limit for mercury in seafood at 0.5 mg/kg. Some previous studies in Yogyakarta [22], Teluk Kao [23] and Bintan [24] showed that some fishes contained higher Hg concentration than the permissible limit. So far, the food safety standard for Hg contamination on fishery products in Indonesia is still emphasized on total Hg. This research aimed to measure Hg concentration in fish and evaluate the health risks for human based on fish consumption in Natuna.

2. Methods

2.1. Sampling and sample preparation

All fishes were collected directly from the fisherman in Natuna on April 2018 and frozen until further analysis. Samples of large fishes were filleted, but for the medium-sized fishes only the heads, skin, and tail were removed, and the rest was analyzed. The small fish was analyzed whole. The length and weight of fishes were measured before sample preparation. Fish tissues were dried in an oven (40° C, 48 h) [25], and mashed into a fine powder then stored in plastic bags. Separated tissues were dried at 105°C, 24 h for water content analysis [26].

2.2. Measurement/analysis methods

The total Hg was measured by a direct mercury analyzer (DMA NIC3000). The morphometric analysis used a digital calliper to estimate the length, and an analytical balance (Sartorius BP 210 S) was used to estimate the weight of samples. The dried sample was then ground until fine powder using mortar, and pestle. Twenty-fifty mg of fine-powder sample was weighed directly in the sample boat and analyzed in mercury analyzer. All samples were made in three replicates. To maintain the quality control of analytical methods, standard reference material (DORM 4) for mercury was analyzed periodically.

3. Result and discussion

The mercury concentrations (Table 1) varied from 0.03 mg/kg-dry weight (*Loligo* sp) to 1.75 mg/kg-dry weight (*Gymnocranius frenatus*). The order of total mercury concentration was *Gymnocranius frenatus* > *Lutjanus sebae* > *Epinephelus aerolatus* > *Pristipomoides multidens* > *Lethinus amboinensis*

> *Sepia* sp > *Selar crumenophthalmus* > *Thunnus tonggol* > *Katsuwonus pelamis* > *Loligo* sp. *Gymnocranius frenatus*, *Lutjanus sebae* and *Epinephelus aerolatus* were exceeding the BPOM/SNI/JECFA permissible limit (0.5 mg/kg) for fishery products.

Table 1. The morphometric and mercury concentration (dry weight) in fish samples from Natuna.

Scientific name	n	Local name	Weight (mg)	Length (cm)	Total moisture (%)	THg ^a (mg/kg dw)
<i>Katsuwonus pelamis</i>	6	Cakalang	1793-2399	47-52	76.62	0.23 ± 0.04
<i>Thunnus tonggol</i>	6	Tuna	1462-1671	45-50	74.68	0.27 ± 0.08
<i>Lutjanus sebae</i>	5	Kakap merah	728-1620	33.5-47	78.98	0.88±0.34
<i>Pristipomoides multidens</i>	10	Kerisi Bali	635-805	36-42	74.81	0.49±0.21
<i>Gymnocranius frenatus</i>	11	Asoh	290-534	26,5-31,5	79.80	1.75±0.53
<i>Selar crumenophthalmus</i>	10	Selar	113-203	21-24,5	78.12	0.28±0.05
<i>Lethrinus amboinensis</i>	6	Lentjam	99-142	19-22	78.83	0.34±0.05
<i>Epinephelus aerolatus</i>	10	Kerapu	314-488	28-34	78.47	0.57±0.16
<i>Sepia</i> sp.	3	Sotong	295-736	50-60	77.15	0.33±0.11
<i>Loligo</i> sp.	10	Cumi	12-25	16-26.5	77.79	0.03±0.01

^a Value is expressed as average ± standard deviation. Values in bold indicate concentration higher than 0.5 mg/kg permissible guideline (SNI 7387, JECFA and BPOM no. 5 2018).

Table 2. The main food of fishes collected in Natuna [31].

Scientific name	Local name	Main food
<i>Katsuwonus pelamis</i>	Cakalang	Fishes, crustaceans, cephalopods, and molluscs
<i>Thunnus tonggol</i>	Tuna	Fishes, cephalopods, and crustaceans, particularly stomatopod larvae and prawns
<i>Lutjanus sebae</i>	Kakap merah	Fishes, crabs, stomatopods, other benthic crustaceans, and cephalopods
<i>Pristipomoides multidens</i>	Kerisi Bali	Fishes, shrimps, crabs, lobsters, stomatopods, squids, gastropods and urochordates
<i>Gymnocranius frenatus</i>	Asoh	Small bottom-living gastropods
<i>Selar crumenophthalmus</i>	Selar	Shrimps, invertebrates, and forams when inshore, and zooplankton and fish larvae
<i>Lethrinus amboinensis</i>	Lentjam	Molluscs, crustaceans, and sea urchins
<i>Epinephelus aerolatus</i>	Kerapu	Fish and benthic invertebrates, primarily prawns and crabs
<i>Sepia</i> sp.	Sotong	Crustaceans and small fishes, crustaceans and small fishes
<i>Loligo</i> sp.	Cumi	Small, juvenile fishes, other cephalopods, crustaceans, polychaetes

Gymnocranius frenatus lives in habitat consists of sand, mud and rubble areas. Food preference of this organism is small bottom-living gastropods [27]. *Lutjanus* sp feed on fishes, crabs, stomatopods, other benthic crustaceans and cephalopods [28]. *Epinephelus aerolatus* feed on fish and benthic invertebrates, primarily prawns and crabs [29-30]. Based on the feeding behavior, mercury would accumulate higher on these three fishes compared with other fish (Table 2).

Table 3. Mean weekly intake based on Hg concentration for a 60-kg adult and a 10-kg child per week.

Scientific name (Local name)		Hg (mg/kg DW)	MWI 60 kg BW	MWI 10 kg BW
<i>Katsuwonus pelamis</i> (Cakalang)	min	0.17	2.43	0.40
	max	0.29	1.39	0.23
	mean	0.23	1.87	0.31
<i>Thunnus tonggol</i> (Tuna)	min	0.20	1.88	0.31
	max	0.46	0.82	0.27
	mean	0.27	1.48	0.25
<i>Lutjanus sebae</i> (Kakap merah)	min	0.66	0.69	0.12
	max	1.58	0.29	0.05
	mean	0.88	0.57	0.09
<i>Pristipomoides multidens</i> (Kerisi Bali)	min	0.12	3.07	0.51
	max	0.97	0.39	0.07
	mean	0.49	1.00	0.17
<i>Gymnocranius frenatus</i> (Asoh)	min	0.98	0.49	0.08
	max	2.59	0.18	0.03
	mean	1.75	0.30	0.05
<i>Selar crumenophthalmus</i> (Selar)	min	0.21	2.08	0.35
	max	0.37	1.19	0.20
	mean	0.28	1.60	0.27
<i>Lethrinus amboinensis</i> (Lentjam)	min	0.26	1.72	0.29
	max	0.40	1.14	0.19
	mean	0.34	1.37	0.23
<i>Epinephelus aerolatus</i> (Kerapu)	min	0.34	1.32	0.22
	max	0.89	0.50	0.08
	mean	0.57	0.85	0.14
<i>Sepia</i> sp (Sotong)	min	0.23	1.81	0.30
	max	0.49	0.85	0.14
	mean	0.33	1.37	0.23
<i>Loligo</i> sp (Cumi)	min	0.02	20.23	3.37
	max	0.05	8.93	1.49
	mean	0.03	15.55	2.59

It is a fact that mercury contamination still provides a risk for human health because of bioaccumulation. If safety standard for fishery product is only determined by total mercury, it is very susceptible, so it is recommended to introduce Mean Weekly Intake (MWI) limit as a simple tool for guidance on fish consumption based on body weight, age and total mercury concentration. MWI will guide how many fishes can be consumed for an adult with 60 kg and a child with 10 kg body weight in relation to Hg concentration in seafood, so there is not excessive consumption. *Katsuwonus pelamis*, for example, in which the lowest concentration of Hg was 0.17 mg/kg-DW while the highest concentration was 0.29 mg/kg-DW, can be consumed at the amount of 1.39 to 2.43 kg per week without a possible

health risk of Hg. Fishes that can be consumed per week by a 10-kg child for *Katsuwonus pelamis* is 0.23 to 0.40 kg.

The sources of mercury contamination are not elaborated detail in this study, but some natural processes including vegetation surfaces, water bodies, wild fires, may be the factor. The results of this study will provide baseline data and information about mercury concentration in fishery product in Natuna and also take off another challenge concerning methyl mercury and source of mercury contamination.

4. Conclusion

Baseline data concerning total mercury identified that mercury concentrations in *Gymnocranius frenatus*, *Lutjanus sebae* and *Epinephelus aerolatus* in Natuna have exceeded maximum permissible limit. Food standard for fishery product especially mercury contaminant only based on total mercury is very susceptible. MWI became a simple tool for guidance on fish consumption based on body weight and age due to mercury concentration. Future studies about methyl mercury level are necessary because it reflects the main threats of organic mercury.

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