Environmental Risks Assessment Of Total Mercury Accumulation At Sentani Lake Papua, Indonesia

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Abstract: This study aimed to investigate the environmental risks of Total Mercury (THg) accumulation in Sentani Lake, Papua, Indonesia. THg was assessed around the Sentani Lake. Sampling survey for Water column, sediment, bivalve, pelagic and benthic fishes samples were collected in one time collection. Furthermore and potential environmental and health risks were determined using hazard quotient (HQ) and target hazard quotient (THQ) equation. Results revealed the concentration of Hg in water column, sediment, bivalve, pelagic and benthic fishes were ranged from 0.002 to 0.076 µg L^{-1}, 115 to 184 µg kg^{-1} dw, 0.024 to 1.678 µg kg^{-1} ww, 0.017 to 1.265 µg kg^{-1}ww and 0.139 to 1.845 µg kg^{-1}ww, respectively. The elevated values of Mercury concentrations were observed in sediment and benthic fish (at the maximum concentrations of 1.678 µg kg^{-1}dw and 1.845 µg kg^{-1}ww), respectively. The magnitude of environmental Hazard Quotient for water and sediment showed that 0.001 – 0.038 and 0.66 – 1.06, respectively. In addition, the high values of target hazard quotient (THQ) for water column, bivalve, pelagic and benthic fishes were in the range of 0.004 to 0.252, 0.005 to 0.190 and 0.024 to 0.277, respectively. All those levels have not exceeded the limit standard or < 1 for potential health risks which mean safe for consumption. This study suggested that the magnitude of Hg concentration in Sentani Lake is generated primarily by the atmospheric fall down from some industries and amalgamation process where some people burn the gold ore. This area is surrounded by major number of Hg from industry and home scale industry directly emitted their waste to Sentani Lake. In case of health risks assessment by comparing with PTWIs and THQ, (based on the bivalve and fish consumption, for 70 years life span of 60 kg body weight), it indicated that people who consumed bivalve and fish from Sentani Lake were not at risk, however, they might be posed a risk due to the long and continual period of consumption.

Keywords; Environmental risk, total mercury accumulation, aquatic system,

I. INTRODUCTION

Mercury (Hg) emission to the environment may occur during the industrial wastes disposal, home industrial and mostly due to the atmospheric fall down from some kilometers away of the point source such as industry use Hg, artisanal gold mining in a variety of ways from the panning and amalgamation as well as open burn. In the incomplete process, Hg is used to amalgamate gold, which facilitates the separation of gold from the unwanted materials. Hence, some Hg escapes to the atmosphere and some releases directly into water bodies as elemental Hg droplets or as coatings of Hg adsorbed onto sediment (Telmer et al., 2006). Mercury emitted in term of gasses will vapour from soil and water then enters the air in atmosphere, where it potentially might be transported and redistributed over the all Earth's surface (Risher et al., 2003). Prior to the advent of mercury pollution, atmospheric deposition contributed an insignificant fraction of the mercury accumulated in lake sediments relative to the supply from the catchment (Bindler, Renberg, Bra¨nnvall, Emteryd, & El-Daoushy, 2001). Mercury can enter the air as a vapour, then drop both as dry and wet deposition, it settle to the bottom at sediment, be absorbed by phytoplankton, or be ingested by zooplankton and microorganisms, or small fishes and bigger fishes as a high stage of food chain as transformation process life. It is noted that as a result of anthropogenic sources, mercury deposition to aquatic systems has increased over the past century (Mason et al., 1994; Fitzgerald et al., 1998).

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2. MATERIALS AND METHODS

2.1 Research Area

This study was conducted in Sentani Lake which is located in the middle of Abepura city, Papua Province, Indonesia. Sentani Lake is a necessary, daily water sources, drinking water and main food protein source such as shellfish, bivalve, shrimps, various fishes and other aquatic seafood which are become daily consumption by local people along Sentani Lake and some areas around the the city of Abepura and Jayapura. Some fishes even are sold out to the next district around Sentani district itself. Here some interesting daily activities seen at the two mouths of the sentani Lake like weekend fishing and boating among dwelling along the Sentani Lake. The Climatology data of the area shows North wind was blowing in from January to March along with the coming dry season, and West Wind during the two months ie from April to May. (Indonesia National Meteorological Department, 2013). The topography of the regency consists of plains at the coastal area and a little bit hilly several hundred meters ahead from the shorelines toward the mine sites.

2.2 Study Design

2.2.1 Water sample collection and its procedures

Within one period of sampling, surface waters sample were collected at the mid depth at total of twelve stations from upstream to downstream of the Sentani Lake. For mercury analysis, we collected four kinds of samples of from the study aquatic tract such as; water column, sediment for the depth of 10 cm, bivalve and fishes which were consisted of two species of the pelagic and benthic fish that were commonly consumed by community. Water samples were collected at twelve varied stations at a depth of 30 cm below the water surface in high density glass bottles. Then, sediment at the top 10cm of the bottom samples were

collected at the same stations where water sample collected using the Eickman bottom sampler device (Santos, 2013). Those samples were kept in polypropylene containers (20g) for Hg analysis and in glass bottles (at least 150 g) for texture analysis. Then, The quotient analysis method served as screen level estimation of risk. The aquatic environmental and health risk assessment equations model was used to analyze exposure and ecological effects and to estimate community level risk and target hazard quotient was used to estimate the health risks (Chen, 2005). All samples were analyzed both in the field study and those that transported to the certified laboratory in Laboratory Kesehatan (LabKes) Makassar, South Sulawesi Province. Some field parameters such as Power Hydrogen (pH) using a potable pH meter, temperature and conductivity were measured using a Salinity-Conductivity-Temperature (S-C-T) meter and Dissolved Oxygen (DO) was measured using a Dissolved Oxygen meter. Suspended Particulate Matter (SPM) was measured using aliquot water sample which collected on pre-weighed TTFE membranes (polytetralfluoroethylene membrane) as the nucleopore membrane of 0.4µm pore size (Gambrell,1991). Dissolved Organic Carbon (DOC) was measured using a total organic carbon analyzer (APHA et al, 1998). Duplicate set of water samples were collected using high density bottle in the river and using non metallic convertible water sampler (Kremerer water sampler) in the sea. The laboratory analysis was done immediately following collection, then all water samples were determined against the national and international standard.

2.2.2 Bivalve and fish sample collection and its procedures

Bivalves were collected at the Sentani lake aquatic track stations where water and sediment samples collected. Approximately 20-25 bivalves with the size in the range of 5-7 cm in length for Anadara trapecia were collected. The tissues were immediately cut off and placed into polyethylene sample bags and kept in an ice box with the temperature of 4ºC before being transported to laboratory and put into a freezer (-20°C). Soft tissue of bivalve were removed and cut in section of small pieces at the end the homogenized representing samples were frozen prior being analyzed. Biota (pelagic and benthic) fishes were collected with hook-and-line to complement dock sampling efforts. At the same stations of each of these two species of fish chosen were collected (a total of 24 fish). They were placed in labeled polypropylene Falcon tubes, stored in ice and immediately transported to the laboratory. To assess the risk of population exposure, the whole fishes were used but by taking into account the conditions of consumption. Since these two species are widely distributed and consumed. At each site, three random subsamples of water column, sediment and aquatic biota were collected to ensure the feasible sample representativeness on the site. All samples were kept cool on the study field. During their transportation to the laboratory, precautions (cold storage on ice, complete filling containers, use of plastic materials for storage, avoidance of undue agitation) were taken to minimize any kind of samples disturbances (**Bennett**, 2007; Wang, Chen, & Xia, 2010).

2.3 Laboratory quality control

Samples were analyzed at Chemical Laboratory at the certified Laboratory of Health in Makassar, Indonesia. The analytical method precision and its accuracy were evaluated by comparing the concentration value of expected THg in certified reference material (SRM 1646a estuarine sediment) from the U.S. Department of Commerce, National Institute of Standard and Technology (NIST) Gaithersburg, MD 20899 with the measured values. All Hg analyses of parameters were done by three replicates. Then, method detection limit (MDL) with seven reagents blank was calculated used as a tool for verification as well.

2.4 Environmental Risks

The potential environmental risks were determined by applying the quantitative screening the environmental Hazard Quotient (HQ) approach. Here the estimates of ecotoxicity (Dose) to exposure respond is compared. The concentration at the background of the study area which is about 15 km upstream and downstream of the area concern is determined. The ratio of the exposure estimated to the effect concentration considered to represent a safe environmental concentration or screening benchmark as shown in the following formulation.

$$HQ = EEC/\text{Screening Benchmark} \quad (1)$$

EEC = Estimated (maximum) environmental contaminant concentration at the site; how much contaminant in sediment or water (e.g.mg contaminant/kg sediment). Screening Benchmark = Generally a No-Adverse Effects level concentration (NOAEL); if the contaminant concentration is below this level, the contaminant is not likely to cause adverse effect. However, If the HQ value is > 1 then it indicates the state of risks to the environment (Environmental Canada 1995; USEPA, 1997; Rayment, 2000).14-16 If: HQ < 0.1; hazard exists; HQ 0.1-1.0: hazard is low; HQ 1.1-10: hazard is moderate; HQ > 10: hazard is high (Lemly, 1996)

2.5 Provisional tolerable weekly intake (PTWI)

Health concerns of water and biota consumers in study area need to be assessed in relation to relevant guidelines. The provisional tolerable weekly intake (PTWI) guideline was recommended by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) showed appropriate safe exposure levels, which were used to estimate the amount of contaminants ingested over a lifetime without appreciable risks [13]. In case of Hg, the provisional tolerable weekly intake (PTWI) guidelines recommended for Hg in a level of 0.004 µg/kg bw (FAO, 1996) is used in this analyses, the weekly intake of Hg depends on the Hg concentration in food and the daily food consumption. In addition, the human body weight can influence the tolerance of pollutants. Estimated weekly intake was calculated by using the formulation below:

$$EWI = (CHg \times ConsR) / BW \quad (2)$$

Where; EWI is estimated weekly intakes ; CHg is Hg concentration in water, bivalve and fish; ConsR is the weekly consumption of water, bivalve and fish from Sentani

lake, Indonesia (water 14 L per week, then Bivalve 68.6 g/week and fish about 252 g/week, and BW is the human body weight (base on 60 kg adult).

2.6 Target hazard quotient (THQ)

The methodology for estimation of target hazard quotient (THQ) although does not provide a quantitative estimate on the probability of an exposed population experiencing a reverse health effect, but it offers an indication of the risk level due to pollutant exposure. This method was available in US EPA Region III Risk based concentration table (US EPA, 2000) and it is described by the following equation:

$$\mathbf{THQ} = \frac{EF \times ED \times FIR \times C}{RFD \times BW \times TA} \times 10^{-3} \qquad (3)$$

Where EF is exposure frequency (365 days/year); ED is the exposure duration (70 years), equivalent to the average lifetime; FIR is the food ingestion rate (fish: 36 g/person/day; bivalve: 9.80 g/person/day (FAO, 2005); C is the metal concentration in seafood (µg g^{-1}); RFD is the oral reference dose (Hg = 0.004 µg g^{-1}/day) (US EPA, 1997, 2000); WAB is the average body weight (60 kg), and TA is the averaging exposure time for non-carcinogens (365 days/ year x ED).

3. RESULTS AND DISCUSSION

3.1. Mercury Concentration in Water

The various magnitude level of mercury concentration in theses twelve stations are mostly affected by the purpose of the area use with its distance pollutant point sources. The maximum mean Mercury level concentration in bivalve, pelagic and benthic fishes were attained values at (1.678, 1.265 and 1.845) µg kg^{-1} ww, respectively. This may be attributed to the huge amounts of sewage, Hg fall down from the atmospheric fall down and home industrial wastewater discharged into the Sentani Lake (Abdel-Moati & El-Sammak, 1997). The water THg concentration were moderately low, from upstream to downstream in Sentani Lake, it were ranged from (0.002 to 0.013 µg L^{-1}). All stations were still lower than the maximum contaminant level considered by the U.S. Environmental Protection Agency which is permitted standard (2.0 µg L^{-1}). The highest value was at St.5 and St.10 due to the closer distance to the mining activity and amalgam processing centre. Comparing the measured Hg accumulation in the area near gold mine activity in Asturias, Spain. THg concentrations found in this local stream vary from < 0.5 to 90.8 g L^{-1}. Which have exceeded the given standard (Loredo et al., 2006).

3.2. Mercury Concentration in Bivalve and Fishes

Bivalve and both pelagic and benthic fish were available at all studied stations. The present results show that Hg concentrations in bivalve, pelagic and benthic fishes organs are closely associated with Hg content of water in Sentani Lake. This obviously may be generated to the abundance of Hg into water by the similar pattern. In addition, THg accumulation in biota have a linier association with the THg concentration in water and have a significant correlation with the THg accumulation in sediment. This finding is similar with the result from gold mining operation in Phanom Pha, Thailand where elevated THg in water (0.4 to 4 µg L-1), sediment (96 to 402 µg kg-1) and bivalve (15 to 584 µg kg-1) near the mining operation were higher than those station outside the mine operation, (Pataranawat at el, 2000).26 In this study area in fact, all biota species' were consumed by local people in Luwuk and surround area as a main protein source. The sources of THg include indirect deposition from watershed runoff, direct atmospheric deposition, point sources, and internal recycling mechanisms such as sediment resuspension (WHO, 2004). However, it is different with the study in Pocona, Philippine, concentrations of Mercury measured in all reaches of the Pocona river were found to exceed sediment quality criteria (174 mg/kg) and predicted toxic effects values (110 mg/kg). The highest Hg concentrations in the river were located adjacent to Hg point sources, paint and pigment formulating plants, and other potential Hg sources of industrial areas. These results indicate that Hg contamination of superficial sediments in Pocona river may pose a risk to aquatic biota (Bonnevie, Gunster, & Wenning, 1992).

Table 1. Concentration of Hg and its accumulation in water column, sediment, bivalve and fishes from Sentani Lake, Papua

Stations	Location	Total Mercury (THg)				
		Water column (µg L^{-1})	Sediment (15 cm depth) (µg kg^{-1}dw)	Biota (µg kg^{-1}ww)		
				Bivalve (Anadara Trapecia	Pelagic fish (pilchard)	Benthic Fish (Plectropomu s Leopardus)
St 1	Upstream, about 10 km from S	0.002	115	0.024	0.031	0.350
St 2	Upstream, 8 km from S7	0.012	157	0.050	0.051	0.555
St 3	Upstream, 6 km from S7	0.018	163	0.061	0.067	0.433
St 4	Close to river mouth in the west	0.024	172	0.067	0.017	0.956
St 5	At river mouth in the West	0.036	125	1.128	0.095	0.895
St 6	At the community Housing	0.018	136	1.262	1.265	1.074
St 7	At river mouth in the North	0.016	147	1.255	1.157	1.098
St 8	Close to river mouth in North	0.033	165	1.243	1.051	1.290
St 9	Downstream, 6 km from S6	0.013	135	1.523	1.056	1.845
St 10	Downstream , 8 km from S6	0.076	125	1.260	0.978	1.177
St 11	Downstream , 10 km from S6	0.066	184	1.678	0.165	0.163
St 12	Downstream, 15 km from S6	0.013	135	0.542	0.828	0.139

3.3 THg in Sediment

It was found that THg concentrations vary from 115 (µg kg^{-1} dw) at the upstream and to 184 (µg kg^{-1} dw) at downstream at the depth of 15 cm. The higher elevated THg in downstream potentially generated by the closer distance from the industrial process activity where some waste directly leaching to lake, amalgam open burn, compare to the other stations. As a result, Hg emitted to the atmosphere and fall down as the dry deposit on the ground, river and to the lake. The cycle process may lead to the accumulation of mercury in the sediment. THg in sediment has been indicated to change in response to changes to external Hg loading. Dated depth profiles of THg in sediment cores clearly show changes in Hg accumulation rates over time that correlate well with documented Hg utilization and environmental releases (Wang and Driscoll, 1995; Engstrom and Swain, 1997; Harris at el, 2007).23-25 Since Indonesia has no any established sediment quality guidelines at this temporary the Canadian guidelines was used as interim measure to assess whether or not the THg heavy metal in sediment.

3.4 Environmental Risks Evaluation

The potential environmental risks evaluation calculated by using hazard quotient (HQ) equation. The objective of this formulation earmarked for the estimation of environmental risks to the potential receptors that were performed in aquatic system for Hg of surface water, both for river and sea and sediment 15 cm in depth. For surface water in this study, the Screening Benchmark base on the Indonesia National Standard setting that the mercury contamination in surface water was not exceed 2.00 µg L-1. In addition the maximum allowable sediment is not exceed of (174 µg kg^{-1}dw) according to the standard of environmental Canadian. More detail of evaluation of Hq for environmental Hg concentration value is presented table 2 below.

Table 2. Concentration of Hg and its accumulation in water column, sediment, bivalve and fishes from Sentani Lake, Papua

Stations	Location	HQ	
		Water column	Sediment (15 cm depth)
St 1	Upstream, about 10 km from S7	0.001	0.66
St 2	Upstream, 8 km from S7	0.006	0.90
St 3	Upstream, 6 km from S7	0.009	0.94
St 4	Close to river mouth in the west	0.012	0.99
St 5	At river mouth in the West	0.018	0.72
St 6	At the community Housing	0.009	0.78
St 7	At the river mouth in the North	0.008	0.84
St 8	Close to river mouth in North	0.017	0.95
St 9	Downstream, 6 km from S7	0.007	0.78
St 10	Downstream , 8 km from S7	0.038	0.72
St 11	Downstream , 10 km from S7	0.033	1.06
St 12	Downstream, 15 km from S7	0.007	0.78

Results in table 2 implied that the magnitude of high three values of environmental risks for water column and sediment were 0.38, 0.33, 0.19 and 1.06, 0.95, 0.84, respectively. Those values state no risks both in water and in sediment except one station for sediment with 1,06 in St.11. This fact indicated that the concentration of Hg in this area initiate by the tailing form the amalgam centre were still less both in background and in contaminated sites. The accumulation in the sediment tended to be correlated to the THg concentration in water. However, the exception was made for the downstream and sentani lake mouth where the accumulation of Hg started to increase toward to the river. Results show that the potential environmental risks evaluation in water column and sediment of contaminated sites were slightly higher in average compare to those in the references sites.

3.5 Target Hazard Quotient (THQ)

Results on the table 3 showed that the magnitude of THQ values for Bivalve anadara Trapecia, pelagic fish and benthic fish were 0.252 for anadara trapezia, 0.174 for pelagic fish (Pilchard) and 0.277 for benthic fish (Leopardus), respectively.

Table 3. Target hazard quotient (THQ) for Hg through consumption of bivalve and fish from Sentani Lake, Papua.

Stations	Location	THQ		
		Bivalve (Anadara Trapecia)	Pelagic fish (pilchard)	Benthic Fish (Plectropomus Leopardus)
St 1	Upstream, about 10 km from S7	0.004	0.005	0.053
St 2	Upstream, 8 km from S7	0.008	0.008	0.083
St 3	Upstream, 6 km from S7	0.009	0.010	0.065
St 4	Close to river mouth in the west	0.010	0.003	0.143
St 5	At river mouth in the West	0.169	0.014	0.134
St 6	At the community Housing	0.189	0.190	0.161
St 7	At the river mouth in the North	0.188	0.174	0.165
St 8	Close to river mouth in North	0.186	0.158	0.194
St 9	Downstream, 6 km from S7	0.228	0.158	0.277
St 10	Downstream , 8 km from S7	0.189	0.147	0.177
St 11	Downstream , 10 km from S7	0.252	0.025	0.024
St 12	Downstream, 15 km from S7	0.081	0.124	0.021

Although these values are still less than 1, the potential risks might be posed due to the continual bioaccumulation and long term exposure. Similar experience was found in municipality of Montecristo, Colombia, which the THg mean level for all fish samples (0.407 ± 0.360 µg g^{-1} ww) did not exceed the recommended limit ingestion level. However, the risk assessment based on the hazard index was suggested that a fish intake of 0.12 kg per day could increase the potential health effects related to Hg exposure in the local human population. (Marrugo-Negrete, Benitez, & Olivero-Verbel, 2008). In addition, limited consumption of shellfish and fish from mercury contaminated water was recommended because most current health risks associated with seafood safety originate in the environment. It is in fact that absolute residue limits are difficult to derive because of the lack of direct evidence for critical levels in seafood. Since the HQ of majority of tuna fish exceeded current human health criteria for Hg, the contaminated fish consumption from the Sulawesi Sea which was the main route of human exposure appears to be risky for human health. (Ullrich, Ilyushchenko, Tanton, & Uskov, 2007) The magnitudes of detectable Hg concentrations were used to estimate potential total lifetime cancer and non-cancer risks based on scenario of an exposure of one meal per week.

4. CONCLUSION

The existing of Total mercury in Sentani Lake habitat is the major environmental concern due to Hg accumulation in all environmental compartments. This case potentially generate impact to environment and community surrounding the Sentani Lake, even the community in the city. The magnitude of THg concentrations were found in living organisms in Lake aquatic ecosystems such as water, sediment, bivalve and fish near and adjacent to the Sentani lake. Also, all of those aquatic contaminated food sources that are major and consumed by local people are also sold out to other cities and area particularly in Abepura and Jayapura. Furthermore, although the concentration is low, THg concentrations might be found in the long distance vicinity areas which the accumulation of Hg in aquatic biota can be merely generated from the Hg dry deposit as a result of atmospheric fall out that occur continuously.

CONFLICT OF INTEREST STATEMENT

Authors declare there are no conflicts of interest.

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