



Assessment of Ecological and Target Hazard Risks of Mercury Contaminated Water Along Makassar Coastal Areas, Indonesia

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Abstract

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INTRODUCTION: The coastal area of Makassar city is a vulnerable area to various pollutants, most of which are generated by anthropogenic activities, such as home industry products, gold shop waste products, and household waste, starting from the coastal area in Barombong to the coastal villages in the Ujung Tanah subdistrict, with the highest level of mercury (Hg) concentration in the coastal area. Another source of potential Hg pollution in this area can be due to community activities at the fish landing base (PPI, Paotere).

AIMS: The aims of this research were to analyze the Hg content in surface water collected from coastal areas of Makassar City and to analyze the target hazard risk effect due to the excessive exposure of water from the recognized as ecologically contaminated area.

MATERIALS AND METHODS: This research applied an observational analytic design of cross-sectional studies. Direct measurement and laboratory sample analyses were conducted as well as a questionnaire was applied to obtain relevant, precise, and accurate data. Respondents' characteristics related to education, habit, lifestyle, occupational smoking habits, and dietary were requested by the administered household interview questionnaire. Assay of the laboratory to analyze the magnitude of Hg in surface water was conducted. Respondents with volunteered based selected using stratified random sampling methods.

RESULTS: Results of the analysis of Hg distribution in surface water used kriging interpolation method showed that the highest concentration of Hg was in stations (22 and 23) around Area of Anjungan Losari Beach and Area of Anjungan Mandar Losari with 1.0013 and 1.0012 mg/l, respectively. While the lowest were in station 2 with 0.0011 and in station 26 Pelindo Jetti area with 0.0013 mg/l, respectively. Ecological Risk assessment indicated that the areas with the highest risk of ecology were such as Near Dermaga Kayu Bangkoa with 9.8, the Tourism area of Karaeng Batjo Tomb 9.7, and Tanjung Bayang beach with risk ecology 9.1 units, respectively. Furthermore, the magnitude of Target Hazard Quotient (THQ) values were 0.0093 in station 1 in Tourism area of Karaeng Batjo Tomb and 0.0089 units in station 24 of Port of Sukarno Hatta, respectively.

CONCLUSIONS: To conclude, Hg levels of contamination in water are still be tolerated in the current; however, it accumulate that potentially harmful in the near future. Ecological risk value was more than one that indicates at risks, although the THQ values are still below than the one that indicates safe.

Introduction

Research related to the contamination and bioaccumulation of mercury (Hg) in coastal areas has been widely observed in many big cities in the world. The research observed various information both in developed and developing countries, such as in Indonesia, particularly in the city of Makassar. Clearly, it is found that data related to the impact of Hg contaminated is scarcely available to those people who are working and living surrounding the area of the city coastal. Hg deposit exposure from water and soil among fisheries and communities both male and female children are actually occurred daily and potentially lead to some symptoms and illness as result of the increase of Hg disposal waste generated from gold shop activities and small-scale home industries.

The exposure of Hg during a long period can initiate these substances accumulate and lead a

bio-magnification in the body may generate the hazard impact of health problems. Diseases may occur such as skin disease, headache, kidney failure, and blood pressure increase that is correlated with cardiovascular diseases occurrence as the results of Hg exposure frequently [1], [2], [3].

The Hg exposure routes through inhalation of metal fumes, dust, and steam most frequently occur little by little and tend to accumulate chronically among fishers and gold workers due to their worker activity daily. Due to the routine exposure, not all Hg pollutants in the body are removed out of the body. Instead, it may accumulate in the hair, urine, bones, and body tissue that may generate illness. Study of Hg hazards shows the facts among workers, gold, jewelry caused Hg, and other chemical exposure in jewelry making. Approximately, 20 workers who did as goldsmiths during 10 years with a period of exposure to chemical fumes >5 h/day were analyzed for their lung spirometry function parameters in forced vital capacity

was $63.95 \pm 9.77\%$ in cases and 76.95 ± 8.1 control suffered from lung disturbances [4], [5].

This observational analytic aimed to explore the relationships between Hg in water and the ecological risks and potential target hazard quotient (THQ) occurrence of coastal areas in the city of Makassar, South Sulawesi.

Materials and Methods

Quality control of laboratory test

Samples were analyzed at the certified chemical laboratory of Health Department in Makassar City, Indonesia. The analytical method of its precision and accuracy were evaluated by comparing the measured total Hg (THg) concentration value with the value in certified reference material (SRM 1646a estuarine) from the United States Department of Commerce, National Institute of Standard and Technology Gaithersburg, MD 20899. All analyses of THg parameters were conducted for three replicates. In addition, the method detection limit with seven reagent blanks was calculated and used as a tool for verification.

Sampling of Hg on surface water

Hg water samples were collected at the mid depth in a total of thirty point samples from upstream to downstream of the coastal area from Barombong till Paotere area of the city of Makassar. To analysis Hg, we collected water from 30 points varying stations at a depth of 30 cm below the water surface in high density glass bottles (Figure 1).

In addition, the quotient analysis method served as the screen level risk estimation. The aquatic ecological and health risk assessment equation were used to analyze exposure and ecological effects and to estimate community level risk, then the THQ was used to estimate the health risks [6], [7]. Samples were analyzed either in the site and transported to the certified Health laboratory of (LabKes) Makassar, Province of South Sulawesi.

Respondent interview

Respondents were selected on the voluntary base took part in the study after signing consent form, and age of about 25 years old and older in the urban coastal areas of Makassar city, South Sulawesi, Indonesia. Interview was conducted to record respondents' characteristics such as age in a year, level of education, family income, sex, holiday or on living in the area, and living and working period was obtained from every respondent by directly writing the answer and oral household interviews.

Assessment of ecological risks

The potential environmental risks were calculated by assessing the quantitative screening of the environmental Hazard Quotient (HQ) approach. It is the estimation of eco toxicity (Dose) compared to exposure respond. The magnitude of the study area background which is approximately of 6 km upstream and 5 km downstream of the area of concern. The exposure ratio was estimated from the effect concentration considered that represents a safe screening benchmark environmental concentration as described in the following formulation.

HQ = EEC/Screening Benchmark
 (1) EEC = Estimated (maximum) concentration of environmental contaminants at the site; how much contaminant in water (e.g., ml contaminant/L of water). Screening Benchmark = Generally a No-Adverse Effects level concentration; if the contaminant concentration is below this level, the contaminant is not likely to lead an adverse effects. However, if the HQ value is >1 , then it is regarded as state of risk to the environment. Where; 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 [8], [9].

THQ

Calculations of noncancer risk were conducted using the THQ, because it is the ratio of the estimated dose of a Hg contaminant to the dose level below any appreciable risk. When THQ is less than one, it is assumed to be safe for the risk of non-carcinogenic effects, however, once higher than one, it is regarded as risk. The formulation is given:

$$THQ = \frac{EF \times ED \times FIR \times C}{RFD \times BW \times AT} \times 10^{-3} \quad (1)$$

Where, EF is exposure frequency (365 days/year); ED is the exposure duration (70 years); FIR is food ingestion rate; C is Hg concentration; RFD is oral reference dose ($5.0 \times 10^{-4} \mu\text{g g}^{-1}/\text{day}$) BW is body weight (70 kg) and AT is averaging exposure time for non-carcinogens [8], [10].

Statistical analysis

Stata software statistical calculation was applied to explain and to analyze the tests of inferential. As the results, percentages, means, and standard errors were prediction in illustrating the characteristics of respondents and all variables correlation ships. All data then were calculated and analyzed using of in proportion of two Chi-Squares (χ^2). However, if there is one value is <5 ($>20\%$), then we applied the Fisher exact test accordingly.



Figure 1: Map of sampling location of water sampling in the coastal area of the city of Makassar, 2021

Results

Total Hg in surface water

The accumulation Hg in the water has a fluctuating trend that surely may give an impact on the environment and health status of communities who are living in the surround of coastal areas. The level of magnitude Hg in water from areas from Barombong district to Paotere District is presented in Table 1, also on the figure of the sampling map. Water characteristics and quality parameters, such as pH, temperature, and turbidity, were directly measured *in-situ* at each site using a YSI Sonde (YSI Inc., Yellow Springs, OH, USA), Hanna pH Tester (Hanna Instruments, Woonsocket, RI, USA), Turbidity was measured using a Hach 2100Q Portable Turbidimeter (Hach Company, Loveland, CO, USA).

Temperature

Temperature in water plays a role in regulating processes that occur in water, chemical, physical, and biological processes. When there is an increase in the temperature of water, the dissolved oxygen

will decrease, which at the same time the oxygen consumption by the aquatic habitat in the water will increase. Temperature can also affect the corrosivity of metals in water [10].

Based on the results of temperature measurements at each sampling point, the temperature value shows a range from 25 to 29°C. Compared with the standard temperature value of the quality standard based on PP No. 28 of 2001, the quality criteria for Class II are the range (24–30°C), then the temperature at all sample points of seawater on the coast of Makassar City meets the requirements.

Power hydrogen (pH)

The results of pH measurements showed that at each sampling location the pH values ranged from 5.08 to 7.06. A pH level <4.8 and >9.2 can be considered polluted [11]. Seawater has a very large buffering capacity to prevent changes in pH. A slight change in pH from the natural pH will indicate a disturbed buffer system. This can cause changes and imbalances in CO₂ levels that can endanger the life of marine biota. The pH of surface sea water in

Indonesia generally varies from location to location between 6.0 ± 8.5 . Changes in pH can have an adverse effect on the life of marine biota, either directly or indirectly [12].

Turbidity nephelometric turbidity units (NTU)

Measurement of internal turbidity (NTU) at each sampling location shows that the turbidity value ranges from 5.05 to 24.21 (NTU). The value of seawater turbidity in the waters of Tumbak-Bentenan ranged from 0.21 to 6.31 NTU with an average value of 1.07 ± 1.85 NTU. The variation coefficient value $>100\%$ indicates the turbidity value between stations is very varied. From the results of turbidity measurements, it can be seen that two stations have very high values >5 NTU, namely, station 13 and station 15, different from other stations <1 NTU [13].

Hg concentration

The highest value from the analysis of the concentration of Hg at the research location was at points 22 and 23, namely, at Makassar Container Terminal 1 and 2 with values of 1.0013 and 1.0012 mg/L, respectively.

Ecological risk and target hazard risks

Ecological risk and THQ calculation result from 30 stations is shown in the following Table 2.

Table 2: Ecological Risk and Target Hazard Risks of Hg in water from coastal areas Makassar, South Sulawesi 2021

No of Station	Description of Site	Eco Risks Water	THQ Water
1	Tourism area of Karaeng Batjo Tomb	9.7	0.0093
2	Area of Ujung Kasi	1.1	0.0012
3	Barombong beach	7.2	0.0033
4	Western cape coast of Barombong	7.6	0.0041
5	Beach of Tanjung Layar Putih	8.3	0.0077
6	Beach of Parahyangan	7.7	0.0052
7	Tanjung Bayang beach	9.1	0.0087
8	Anging Mammiri beach	8.3	0.0072
9	Area of Tanjung Bira	5.4	0.0031
10	Akarena beach	6.6	0.0070
11	Gowa development tourism PT	8.2	0.0042
12	Area of Tanjung Bunga	5.1	0.0042
13	Area of TSM side	7.2	0.0049
14	Ciputra University of Makassar	2.4	0.0012
15	Twin tower CPI	3.2	0.0023
16	Area of Anjungan Losari Beach	4.3	0.0034
17	Area of Anjungan Mandar Losari	3.3	0.0022
18	In front of Bulepe Café	4.6	0.0036
19	Near Dermaga Kayu Bangkoa	9.8	0.0082
20	Near of Makassar port police station	7.7	0.0044
21	Area of Waves Café	4.5	0.0032
22	Makassar container terminal	1.01	0.0011
23	Makassar container terminal II	1.02	0.0012
24	Port of Sukarno Hatta	9.1	0.0089
25	Near the packing plant PT. Tonasa	3.3	0.0022
26	Area of Pelindo Jetty	3.4	0.0045
27	Depot PT Port of Pertamina	6.5	0.0072
28	Near jetty II TBBM Makassar	5.5	0.0052
29	Area of TPI Lelong Paotere	8.7	0.0086
30	Port of Paotere	1.8	0.0020

THQ: Target hazard quotient.

Ecological and THQ

Ecological Risk assessment indicated that the areas with highest risks of ecology were such as Near Dermaga Kayu Bangkoa with 9.8, the Tourism area of Karaeng Batjo Tomb 9.7, and Tanjung Bayang beach with risk ecology 9.1 units less, respectively. Furthermore, the magnitude of the highest magnitude of THQ values were 0.0093 in station 1 in Tourism area of Karaeng Batjo Tomb

Table 1: Characteristics and results of analysis of Hg concentrations in water coastal areas of the city of Makassar, South Sulawesi, 2021

No of sample point	Description of sample sites	Hg Characteristics and Concentration					Coordinates	
		Temp. (Degree Celsius)	pH (pH)	Turbidity (NTU)	Hg in Water (mg/l)	Mean and Standard (0.001mg/l)	Longitude, Latitude	
1	Kampoeng wisata/makam karaeng btj	25	7.02	24.21	0.0098	0.0097	119°23'02.1"E 5°13'16.6"S	
2	Ujung kasi	26	6.08	21.02	0.0012	0.0011	119°23'08.8"E 5°12'47.7"S	
3	Pantai barombong	26	6.09	16.21	0.0071	0.0072	119°22'52.6"E 5°12'12.0"S	
4	Western cape coast barombong (muara sungai jeneberang)	28	7.01	11.11	0.0078	0.0076	119°22'47.6"E 5°11'42.9"S	
5	Pantai tanjung layar putih	26	7.01	6.05	0.0081	0.0083	119°23'01.3"E 5°11'24.7"S	
6	Pantai parahyangan	26	7.03	8.02	0.0076	0.0077	119°23'09.8"E 5°11'06.8"S	
7	Tanjung bayang beach	27	7.02	9.11	0.0090	0.0091	119°23'12.8"E 5°10'59.9"S	
8	Anging mammiri beach	25	6.02	10.01	0.0085	0.0083	119°23'14.4"E 5°10'49.7"S	
9	Tanjung biru	25	6.03	10.21	0.0055	0.0054	119°23'15.3"E 5°10'34.1"S	
10	Akarena beach	26	6.04	12.01	0.0065	0.0066	119°23'14.6"E 5°10'18.7"S	
11	Gowa development tourism PT	26	7.03	11.12	0.0081	0.0082	119°23'15.4"E 5°10'01.1"S	
12	Tanjung bunga	27	7.02	15.00	0.0052	0.0051	119°23'16.9"E 5°09'36.6"S	
13	Belakang TSM	28	6.03	12.02	0.0071	0.0072	119°23'31.0"E 5°09'24.1"S	
14	Universitas ciputra makassar/sunset quay makassar	27	6.02	8.05	0.0025	0.0024	119°23'37.4"E 5°08'54.5"S	
15	Twin tower CPI	28	6.01	7.05	0.0032	0.0032	119°24'03.0"E 5°08'36.2"S	
16	Anjungan pantai losari	27	6.05	7.20	0.0045	0.0043	119°24'29.3"E 5°08'45.7"S	
17	Anjungan mandar losari	27	5.09	6.14	0.0035	0.0033	119°24'24.3"E 5°08'29.0"S	
18	Bulepe café	26	5.08	6.04	0.0048	0.0046	119°24'21.7"E 5°08'22.1"S	
19	Dermaga kayu bangkoa	29	6.02	5.05	0.0095	0.0098	119°24'17.4"E 5°08'14.9"S	
20	Makassar port police station	29	6.01	5.50	0.0078	0.0077	119°24'16.4"E 5°08'06.8"S	
21	Masuk lewat waves café	28	7.05	9.14	0.0045	0.0045	119°24'13.2"E 5°08'02.1"S	
22	Makassar container terminal	26	7.05	11.02	1.0013	1.0011	119°24'14.9"E 5°07'47.4"S	
23	Makassar container terminal II	26	7.02	9.03	1.0012	1.0012	119°24'24.5"E 5°07'36.0"S	
24	Pelabuhan soetta	28	7.05	8.55	0.0091	0.0091	119°24'25.9"E 5°07'25.7"S	
25	Dekat packing plant PT tonasa	28	6.01	6.17	0.0032	0.0033	119°24'29.1"E 5°07'19.9"S	
26	Pelindo jetty	28	6.03	4.16	0.0031	0.0034	119°24'32.2"E 5°06'57.9"S	
27	Depot Pt pertamina pelabuhan	29	7.01	5.01	0.0066	0.0065	119°24'35.9"E 5°06'47.9"S	
28	Dekat jetty II TBBM Makassar	27	7.06	8.48	0.0056	0.0055	119°24'46.7"E 5°06'43.9"S	
29	TPI Lelong paotere/bmkg paotere	28	7.04	6.23	0.0089	0.0087	119°25'12.0"E 5°06'36.7"S	
30	Pelabuhan paotere	26	7.00	8.23	1.0019	1.0018	119°25'16.5"E 5°06'34.6"S	

NTU: Nephelometric turbidity unit.

and 0.0089 units in station 24 of Port of Sukarno Hatta, respectively.

Discussion

Hg concentrations in water coastal areas of the city of Makassar

Results of the analysis of Hg distribution in surface water used kriging interpolation method shows that the highest concentration of was in stations (22 and 23) around Area of Anjungan Losari Beach and Area of Anjungan Mandar Losari with 1.0013 and 1.0012 mg/l, respectively. While the lowest were in station 2 with 0.0011 and in station 26 Pelindo Jetti area with 0.0013 mg/l, respectively.

Hg waste from gold traders in the market causes high levels of pollution at Losari Beach in Makassar, South Sulawesi. The use of Hg in gold, especially when it cooked to facilitate the formation process. Usually, the Hg water is directly discharged through culverts and flows to Losari beach so that Hg levels mix with sea water, as a result the magnitude of Hg in surface water increases. Increasingly of the gold shops are currently located in line and next to Losari Beach.

Aquatic habitat on Losari beach was feared to be in danger of being exhausted or no longer fit for consumption due to Hg contamination. "There are many fishermen who live on the island around Makassar, eating fish catches from Losari beach. Now, they are experiencing skin problems such as itching, ulcers, and other unseen symptomatic diseases. Pollution on Losari beach has entered an alarming stage, especially for residents who live around the coast and consume caught fish," he said. The pollution radius of Losari beach is estimated to be 10 miles from the shoreline. Usually, Makassar residents eat fish with a radius of more than 10 miles from the shoreline. Fishermen who catch fish are also more often around the Makassar Strait.

Ecological risk assessment

The exposure of environmental to Hg contaminant in the water was estimated from effect concentrations considered that represents a safe screening benchmark environmental concentration. As shown on the table that the areas with the highest risk of ecology were such as Near Dermaga Kayu Bangkoa with 9.8, the Tourism area of Karaeng Batjo Tomb 9.7, and Tanjung Bayang beach with risk ecology 9.1 units less, respectively. The analysis of the spatial distribution of metals in shells with Pb method is seen that the distribution pattern of the highest Pb was station 4 Mariso district around the coastal waters Tallo and the lowest district was Tamalate District. Based on

the analysis of Pb, the spatial distribution of surface soil shows that high around station 4 in the district Tallo followed by Tamalatea district of the coastal and the lowest in Ujung Tanah district.

The Hg contamination of water faces severe diseases of human exposure in the coming years. Fishes, aquatic animals are also affected due the increasing amount of Hg in water and the peoples who caught the fishes; they are also affected by the Hg contamination. Human healthiness is straight forwardly affected by the utilization of polluted water, fishes, vegetables, plants, etc., which are the most important sources of foodstuffs for humans. Signs and symptoms related to acute toxicity of Hg are tremors, insomnia, muscle atrophy, muscle twitching, weakness, etc. It shows carcinogenic effects in humans and it also causes various types of diseases to humans [14].

Similarly, a previous study in Pangkep Regency near Tonasa industry revealed that Hg magnitude variation levels among different community's sites are highlights important to describe the ecological role condition and to the communities behavior and health who are exposed to Hg contaminants. The ecological risk point of view of Hg values in the area are >1 [15].

THQ

THQ assessment results found values of 0.0093 in station 1 in Tourism area of Karaeng Batjo Tomb and 0.0089 units in station 24 of Port of Sukarno Hatta, respectively. Hg is a globally known pollutant. However, of particular concern is MeHg, which can be converted into inorganic forms of Hg in aquatic ecosystems. Hg can enter freshwater ecosystems through atmospheric deposition or industrial wastewaters. Authman *et al.* [16] emphasized that the principal sources of Hg and mercurial organic compounds in the environment are fungicides and organic fungicides, respectively. It is noteworthy that Hg is associated with a wide spectrum of adverse health effects including damage to the central nervous system (neurotoxicity) and the kidney [17].

The main concern about the toxicity of Hg in the general population exposed to low levels of Hg in their diet relates to the potential neurotoxicity of organic forms of Hg in both children and adults. From the previous research, it is found that Hg and cyanide concentrations are still below the quality standard in accordance with Government Regulation No. 82 of 2001, where laboratory results are smaller than 0.0005 mg/L in Kayeli river water samples [18].

It is also similar with the finding from Rantetampang and Mallongi, showing that the water THg from upstream to downstream concentration were moderately low in Sentani Lake, while it were ranged from 0.002 to 0.013 $\mu\text{g L}^{-1}$. All stations were still lower

than the maximum contaminant level considered by the U.S. Environmental Protection Agency, which is a permitted standard ($2.0 \mu\text{g L}^{-1}$). The highest value was due to the closer distance to the mining activity and amalgam processing center, where it is ecological risk as well as the Target Hazard risk assessment value indicated still moderate and low risk [7], [19], [20].

Conclusions

The magnitude of Hg contamination in surface water is still be tolerated in the current situation, however accumulate Hg potentially harmful in the near future. Ecological risk value was more than one that indicates at risks, although the THQ values are still below than the one that indicates safe.

Acknowledgments

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Author Contributions

Anwar Mallongi: Conceptualization, data curation, Writing-original draft, and review – edit. Muh. Fajaruddin Natsir: data calculation and curation, referencing. Ratna Dwi Puji Astuti: Location Observing, data collection and edit draft. Annisa Utami Rauf: Location Observing, data collection and edit draft.

Informed Consent

Informed consent was obtained from each participant included in the study.

Data Availability

The data used to support the findings of this study are available from the corresponding author on request.

Ethical Approval

This study was approved by the Ethical Committee of Medical Faculty of Hasanuddin University.

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