

SPATIAL DISTRIBUTION OF MERCURY CONCENTRATION IN TRADITIONAL GOLD MINING OF BULADU

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ABSTRACT

This research is aimed at mapping and examining downstream and upstream mercury concentration spatially in water and sediment in the traditional gold mining of Buladu. 94 Samples consisting water and sediment were taken through random sampling at Pasolo River, Sulawesi Sea and tailings with three replications. The samples of mercury in water and sediment were analyzed in the Integrated Research and Testing Laboratory of UGM by using mercury analyzer and at the *Quality Control* and Testing Laboratory of Fisheries Products of Gorontalo Province by using non-flame Atomic Absorption Spectroscopy (AAS). Data were analyzed using a map and then interpreted.

In the period of Sampling I, The result of the analysis that mercury concentration in water and sediment at the upstream was 0.0006 mg/l and 10.8731 mg/kg. The location of ASH₂ and SSH₂ had very extreme values because they were the initial location of mine processing, namely 0.00103 mg/l and 55.0680 mg/kg. In Sulawesi Sea, the average concentrations in water and sediment were 0.0008 mg/l and 17.55 mg/kg. In the period of Sampling II, namely ASH₂, and SSH₂, the concentrations were 0.0055 mg/l and 136.70 mg/kg and in the Celebes Sea the average concentrations were 0.37141 mg/l and 0.9477 mg/kg. In the period of sampling III, the concentrations of mercury in ASH₂ and SSH₂ were <0.00006 mg/l and 244.16 mg/kg. In the Sulawesi Sea, the average concentration was 0.00084 mg/l and 0.6408 mg/kg. In tailings of the Sampling I, II and III, the concentrations in both water and sediment were very high, with the average values of 0.00528 and 22.500, 0.3074 and 50.048 and 0.00147 mg/l and 162.42 mg/kg. An appropriate management model is required to overcome mercury pollution in the Traditional Gold Mining of Buladu.

Keywords: *Spatial ; Mercury ; Gold mining.*

A. INTRODUCTION

To date, people still need the support of mining resources and mining commodities in order to maintain and improve their welfare. Significantly, the existence of mining is a strategic sector in developing mankind. Mineral resources are a unit of the geological structure as a part of the ecosystem. The mining resources can be divided into metal and non-metal as well as into quality and quantity, the existence of the mining sector is still strategic for Indonesia and the backbone of local revenue for mining resource-rich areas.

In general, gold mining in Gorontalo Province is one of the potential natural resources, which offers a better prospect of increasing economic levels and social welfare. Such economic improvement includes income, employment, and opportunities for new activities, in addition to agricultural and plantation sectors.

Gold mining uses mercury amalgam to separate gold from raw materials. This process will produce wastewater containing mercury, which will pollute the waters and endanger lives in the waters (Sutomo, 1998).

Waters of rivers, lakes, and seas are contaminated by mercury from wastewater of industries that use mercury. Particles that contain mercury salts (Hg⁰) and organic mercury enter the water and settle with sediment, and undergo oxidation-reduction. In the sediment, there are anaerobic bacteria which are able to restructure methane and produce methylcobalamin through the process of decomposition in water. Methylcobalamin is a compound that can transform inorganic mercury into methylmercury (CH₃Hg) in neutral or alkaline water. Methylmercury accumulates through the food chain ranging from plankton, zooplankton, small fish and big fish. Eventually, the fish is contaminated with methyl mercury, which is harmful to human health (Sutomo, 1998 in Mahmud, 2012). Research

conducted by Mahmud (2012) shows that pollution load of mercury that enters the river and settles in the bottom sediments at low and medium discharge criteria will be 1.4 - 2.1 times of its initial state. This occurs because of the accumulation of heavy metal in the water. At high discharge, the mercury concentration is affected by dilution in the river.

The research was conducted in the gold mining of Buladu because this gold mining dump wastes into rivers and then into the sea. This is dangerous because the sea is the place for aquatic animal life, particularly fishes, which are consumed by many people who live along the waters of the Sulawesi Sea. Results of a research conducted by Withiatna (2005) indicate that high levels of mercury in the tailings are generally caused by imperfect amalgamation. The research conducted in the Cineam Region indicates that one-cycle amalgamation produces mercury by 9%. The results of the research conducted by Withiatna (2005) indicate that the concentration of mercury in the tailings in the Cineam Region by 201-595 ppm tends to be the same as the concentration of mercury in the flow of the Tulabolo River (Mahmud, 2012). A research conducted by Mahmud et al., (2014) in a traditional mining site of Buladu indicates that the concentration of mercury in the Celebes Sea is 0008 mg/l. A research conducted by Kitong, et al., (2012) indicates that the concentrations of mercury in Ranoyapo River range from 0.05 - 1.3 mg/kg. Gold Processing using amalgamation has contaminated river sediments. Sediments contaminated with mercury have a potential negative environmental impact and are harmful to the people who live around the river. River sediments contaminated by mercury in gold mining areas around, the people are directly related to the gold processing of by means of metal amalgamation, along with mercury metal wasted as a smooth mixture of tailing materials (Kitong, et al., 2012).

This research is aimed at mapping and assessing the concentration of upstream and downstream mercury in the water and sediment which was conducted spatially in the traditional gold mining of Buladu.

B. LITERATURE STUDY

Public mining with no license or it is usually called as traditional mining is a public activity in mining aspect in a particular area without government's license. Usually the miners use

simple equipments. Some of the examples are entrenchment C mining activity and other general mining activities like gold mining activity and entrenchment of valuable items (Balihristi, 2008).

Inorganic mercury can transform to organic mercury with help of microbes activity both in aerobe and in anaerobe condition; in the low concentration of inorganic mercury there will form di-methyl mercury, and in high concentration of inorganic mercury there will form mono-methylmercury. In natural watershed area, the concentration of mono-methyl mercury and di-methyl mercury are influenced by microbes, organic carbon, concentration of inorganic mercury, pH, and temperature. Both forms of methyl mercury compound can be broken down by bacteria living in the sediment. Methyl mercury can get bioaccumulation and biomagnifications in watershed biota both directly and indirectly through food web (Efendi, 2003; Palar, 1994; Darmono, 1995).

C. METHODOLOGY

The research was conducted in the traditional mining site of Buladu in Sumalata Subdistrict. Samples of water and sediment were taken in Pasolo River, Sulawesi Sea and tailings. The samples from the river were taken with 3 repetitions. Sampling I and III were taken from 7 points in the river, 3 points in the sea and 5 points in the tailings. Sampling II was taken from 8 points in the river, 6 points in tailings and 3 points in the sea. The number of samples in total was 94 samples. The samples of mercury in the water and sediment were analyzed using a mercury analyzer at the Integrated Research and Testing Laboratory of UGM by using mercury analyzer and at the *Quality Control* and Testing Laboratory of Fisheries Products of Gorontalo province by using non-flame Atomic Absorption Spectroscopy (AAS). Data were analyzed using a map and then interpreted.

D. RESULTS AND DISCUSSION

a. Results of Spatial Analysis on Mercury Concentration in Water and Sediment of River and Sea

Sampling I

The results of the analysis indicated that the concentration of mercury in the water at the upstream of Pasolo River, i.e. ASH1 point, was

0.0006 mg/l and at the downstream was 0.00024 mg/l. At the ASH1 location, the concentration of mercury was low because it was a control point. The concentration of mercury existed at the control point of ASH 1 because there is a traditional gold mining of Padengo at the upstream of Pasolo River. Sulawesi Sea as the waste receiver had the concentration of mercury in water by an average of 0008 mg/l. A Map of the results of the spatial analysis of mercury concentrations in water and sediment is shown in Figure 1.

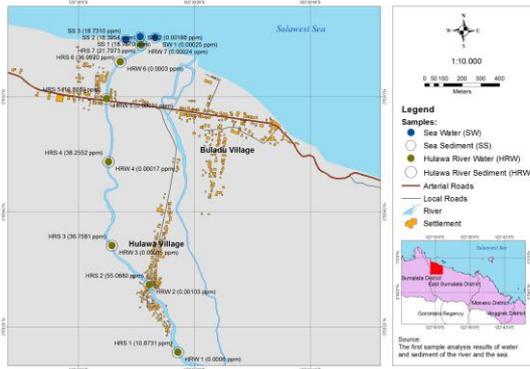


Figure 1.Map of Results of Spatial Analysis on Mercury Concentration in Water and Sediments in Pasolo River and Sulawesi Sea

(Source :Primary Data, Mahmud, et al., 2016)

The location of ASH3 had a very high concentration of mercury by 0.00035 mg/l. The reason was that the ASH 3 location was the processing location which was only within 5 m to the river. The concentrations mercury tended to be similar along the river due to processing activities very close to Pasolo River. Pasolo River flows into the Sulawesi Sea, where the concentrations of mercury ranged from 0.00025 to 0.00027 mg/l. This result was relatively high because the distance between the river as waste disposal and the sea were quite close, approximately 1 Km.

The map shows that the concentration of mercury in the upstream of Pasolo River (SSH1) was 10.8731 mg/kg. The location of SSH1 was the control location before the gold mining of Buladu. The concentration of mercury existed in this location because in the upstream there was another gold mining, namely Padengo gold mining. The location of SSH 2 is the beginning of the traditional gold mining of Buladu with a very high concentration of 55.0680 mg/kg. In the downstream of the river the concentration was 21 mg/kg. At the location of the SSH1, the concentration was the control point. In general, the closer to the location of the waste source, the higher the concentration of

mercury in the river sediment. The pattern of mercury concentrations in the river sediments indicated that the closer to the downstream was, the lower the concentration would be. Sulawesi Sea as the waste receiver indicated that it had mercury concentrations in sediment ranged between 8.7870 mg/kg and 25.4630 mg/kg with an average result of 17.548 mg/l. Accordingly, Sulawesi Sea has received waste by an average of 17.548, which would endanger aquatic animals that live in the sea. Therefore, the sediment in the Celebes Sea was above the European Safety Standard which could not exceed 2 mg/kg. The concentration of mercury in the sediment of Pasolo River was very high when compared to the concentration of mercury in Ranoyapo River ranging between 0.05 and 1.3 mg/kg. The high concentration of mercury in Pasolo River was due to gold mining of Buladu because the processing location was very close to the river. Moreover, there was the amalgam processing of which the tailings were directly discharged into the river. This is a cause of high levels of mercury from mining gold in the river Buladu compared with Ranoyapo. The results of research conducted by Mahmud (2012) indicated that the probability of significance was 0.05 so that it can be concluded that the concentration of mercury in sediments was affected by distance. Based on the mathematical model, it was found that the greater the distance reviewed, the lower the concentration of mercury in sediments at various water discharge criteria. Many factors can affect the concentration of mercury in the bottom sediments. The shape, size and weight of soil particles determine the amount of sediment transport. A research conducted by Ikhsan (2007) shows that the smaller the diameter of the sediment is, the larger the amount of bedload can be transported. The greater the discharge is flowing, the larger the amount of bedload can be transported. The same thing also occurred with the concentration of mercury in Manyuke River. At the period of Sampling I, the concentration of mercury in Pasolo River was already very high compared with the concentration of mercury in Manyuke River, Landak Regency. The research conducted by Subandri (2008) indicates that the average levels of mercury in the water were 0.5334 ppb and sediment was 3.1417 ppb.

Sampling II

The analysis results on Sampling II at the upstream of Pasolo River, ASH1 point, the concentration was 0.9852 mg/l and at the downstream, ASH8, and the concentration was 0.00565 mg/l. The concentration of mercury fluctuated from the upstream to the downstream due to some processing activities which were very close to the river. The concentration at the location of ASH3 was very high by 2.4284 mg/l because the ASH3 point was very close to the processing activities, which was within 5 m to the river. Sulawesi Sea as the waste receiver from Pasolo River was identified to have mercury concentrations ranging between 0.00063 and 1.1122 mg/l. Such results were very high and would endanger the lives of aquatic creatures. The map the results of the spatial analysis of mercury concentrations in water and sediment in Sampling II is shown in Figure 2.

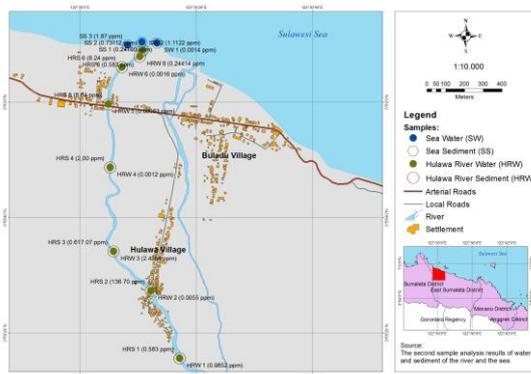


Figure 2. Map of Spatial Analysis on Mercury Concentration in Water and Sediment of Pasolo River and Sulawesi Sea in Sampling II
 (Source :Primary Data, Mahmud, et al., 2016)

The map shows that the concentration of mercury in the upstream was 0582 mg/l. This location is the control location. At the location of SSH2, the concentration was very high by 136.70 mg/kg. This location was very close to the location of the activities and the tailings were directly discharged into the river. This was one of the factors that made the concentration high at this location. At the downstream, the concentration was 0.24414 mg/kg. The closer to the downstream was, the lower the concentration of mercury would be. Sulawesi Sea as the waste receiver from Pasolo River had the mercury concentration ranging between 0.24190 mg/kg and 1.87 mg/kg. The average results of the concentration in Sulawesi Sea

were 0.94 mg/kg. This would endanger aquatic animals that live in the waters of Sulawesi.

Metal speciation in freshwater and seawater mainly differ in terms of ionic strength, lower surface absorption in sea water, the concentration of trace metals, the concentration of main cations and anions and usually a higher concentration of ligand organic in the system of freshwater (Connell and Miller, 2006).

The behavior of metals in natural waters is strongly influenced by the interaction between the phases of liquid and solid, especially water and sediment. Decomposed ions and metal compounds disappear rapidly from solution when in contact with the surface of the particulate material through several types of surface bonding phenomena. Additions and remobilization of trace metals in a sedimentary environment depend on factors such as chemical structure (e.g. amount of decomposed iron and carbonates), salinity, pH, the value of the redox and hydrodynamic state (Stumm and Morgan, 1970; Hart and Davies, 1977; Fostner, 1979b in Connell and Miller, 2006).

Sampling III

The average concentration of mercury in sampling III was very small by <0.00006 mg/l. In Sulawesi Sea, the mercury concentrations in water ranged between 0.00078 mg/kg and 0.00141 mg/kg with an average concentration of 0.00084 mg/kg.

The map shows that the concentration of mercury in sediments in the downstream of Pasolo River was 0.55255 mg/kg. The highest mercury concentrations were in the SSH2 by 244.16mg/kg. The concentration in the SSH2 was extreme because this location was always active and very close to the river. At the downstream of the river, the concentration was 0.12340 mg/kg. In Sulawesi Sea as the waste receiver, the mercury concentrations in sediments ranged between 0.27745 mg/kg and 1 mg/kg with an average concentration of 0.6408 mg/kg. The concentration of mercury in gold mining traditional of Buladu was higher than that of Acre State by 0.042 ug/g in Amazon, Brazil (Brabo, et al., 2003) and tended to be the same as that in the zone north to Sado Estuary, Portugal (Lilibo, et al. , 2011).

The results of the spatial analysis of mercury concentrations in water and sediment of Sampling III are shown in Figure 3.

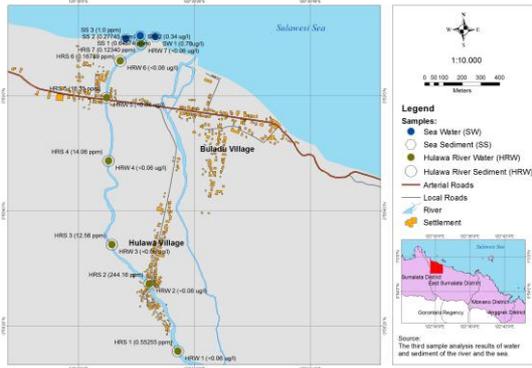


Figure 3. Map of Spatial Analysis on Mercury Concentration in Water and Sediments in Pasolo River and Sulawesi Sea in Sampling III
 (Source :Primary Data, Mahmud, et al., 2016)

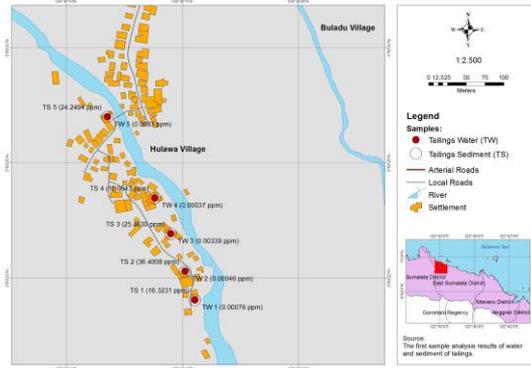


Figure 4. Map of Results of Spatial Analysis on Mercury Concentration in Water and Sediments in Tailings of Sampling I
 (Source :Primary Data, Mahmud, et al., 2016)

3.2. Results of Spatial Analysis on Mercury concentrations in Water and Sediments of Tailings

Sampling I

The map shows that the mercury concentrations in the water of tailings were between 0.00076 and 0.00339 mg/l. The concentration of mercury in the water of the tailings tended to be similar. The highest concentrations of mercury at the locations of AT2 and AT3 were 0.00046 and 0.00339 mg/l. The reason is that the average processing activities in the location were only once a week on Saturdays, except the location of AT3. Particular locations such as AT2 and AT3 worked continuously depending on the mining results obtained. The concentration of mercury in sediments reflect the characteristics of the contaminants that were passed by that: (1) deposit and introduction of mercury is the mobilization of the result of burning process and evaporation of amalgamation (2) inputs of particulate mercury will be decomposed coming from the washing process of contaminants mineral in the processing in the tailings (3) the remains of mercury from the breakdown of absorption at the surface (Appleton et al, 2001 in Mahmud, 2012). The results of the spatial analysis of mercury concentrations in water and sediments in the tailings in Sampling I are shown in Figure 4.

The map shows that the concentration of mercury in sediments of the tailings ranged between 16.3231 and 36.4008 mg/kg. This result was already above the European Safety Standard that should not exceed 2 mg/kg. The highest concentration of mercury occurred in the location of ST2 by 36.4008 mg/kg and ST3 by 25.4630 mg/kg. This occurred because the location was the most active processing activities.

Sampling II

The map shows that the highest mercury concentrations were in the location AT 2 and AT 3 by 0.8748 mg/l and 0.9423 mg/l. The concentrations of mercury were very high and fluctuated. The high concentration of mercury in the tailings was because the bonding process of gold and rock used mercury to form the amalgams. In the process of forming amalgams, the machine was rotated for 3.5 hours using water. In the washing process, the remains of mercury would enter the tailings and increase the load on the waste. Furthermore, most of the wastewater would flow and enter the rivers and end at the sea and some would remain at the processing facility. This resulted in very high concentrations of mercury in the tailings. The map of the results of spatial analysis in mercury concentrations in water and sediments in the tailings in Sampling II is shown in Figure 5.

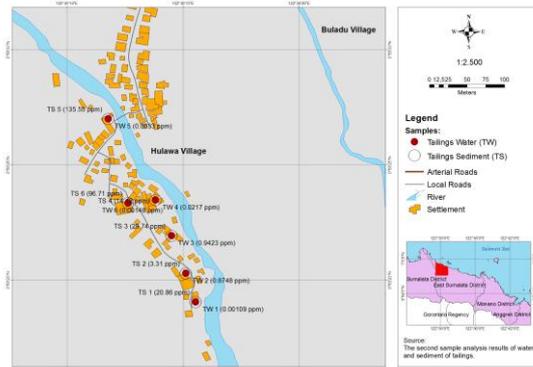


Figure 5. Map of Mercury Concentration Spatial Analysis Results in Waste Water and Sediments in Tailing Sampling II (Data Primer, Mahmud, et al 2016)

The map shows that the results of the analysis of mercury in sediments conducted on the Sampling II indicated the highest mercury was in the ST5 by 135.55 mg/kg and the lowest was in the ST2 point by 3.31 mg/kg. The concentrations of mercury in sediments fluctuate and many factors influence, namely waste load produced, processing activities, pH, temperature and particle size. In the aquatic environment, metals are in the forms of free ions, pairs of organic ions and complex ions. Metal solubility in water is controlled by pH of the water. The increase in pH lowers the solubility of metals in water. The increase in pH changes the stability of the form of the carbonate into hydroxide, which forms a bond with particles on the body of water, so that it will settle and form mud (Palar, 2004). The solubility of trace metals in natural water in principle is governed by (1) pH, (2) types and concentrations of ligand and chelating substances and (3) the state of oxidation of the mineral component and the redox environment of the system (Leckie and James, 1974 in Connell and Miller (2006).

Sampling III

The map shows that the highest concentrations of mercury were at the location of AT4 by 0.07003 mg/l. The lowest concentration at the location of AT5 was 0.00036 mg/l. The gold processing usually uses amalgamation technique, by mixing ore and mercury to form amalgams (Au-Hg alloy) using water media. The results of the spatial analysis of mercury concentrations in water and sediments in the tailings in Sampling III are shown in Figure 6.

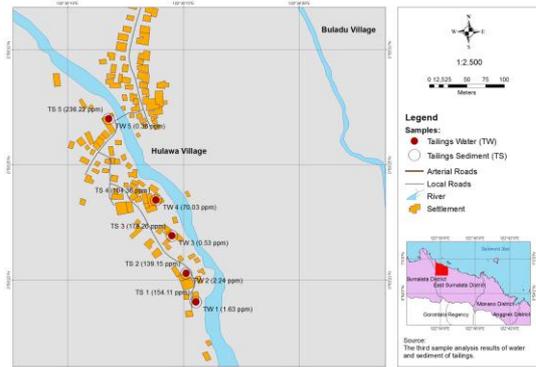


Figure 6. Map of Results of Spatial Analysis on Mercury Concentration in Water and Sediment of Tailings in Sampling III (Primary Data, Mahmud, et al., 2016)

The map shows that the results of the analysis of the concentrations of mercury in sediment ranged between 104.36 mg/kg and 236.22 mg/kg. The highest concentration of mercury in sediments was 236.22 mg/kg in the location of ST 5. On average, the results of mercury concentration in Sampling III had very extreme values. This would be dangerous because these particles would be carried away by the water and enter into rivers and the sea as the waste receiver. These particles could get into the roots of the plant and would harm plants that live in the area. Accordingly, a good gold management model is recommended to be implemented in order to reduce mercury in the tailings so that it does not pollute soil, water, and vegetation in the vicinity.

E. CONCLUSION

In the period of Sampling I, The result of the analysis that mercury concentration in water and sediment at the upstream was 0.0006 mg/l and 10.8731mg/kg. The location of ASH₂ and SSH₂ had very extreme values because they were the initial location of mine processing, namely 0.00103 mg/l and 55.0680 mg/kg .In Sulawesi Sea, the average concentrations in water and sediment were 0.0008 mg/l and 17.55 mg/kg. In the period of Sampling II, namely ASH₂, and SSH₂, the concentrations were 0.0055 mg/l and 136.70 mg/kg and in the Celebes Sea the average concentrations were 0.37141 mg/l and 0.9477 mg/kg. In the period of sampling III, the concentrations of mercury in ASH₂ and SSH₂ were <0.00006 mg/l and 244.16 mg/kg. In the Sulawesi Sea, the average concentration was 0.00084 mg/l and 0.6408 mg/kg. In tailings of the Sampling I, II and III, the concentrations in both water and sediment were

very high, with the average values of 0.00528 and 22.500, 0.3074 and 50.048 and 0.00147 mg/l and 162.42 mg/kg. An appropriate management model is required to overcome mercury pollution in the Traditional Gold Mining of Buladu.

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REFERENCES

- Appleton, J.D., Williams, T.M., Orbea, H, and Carrasso, M. (2001). Fluvial Contamination Associated With Artisanal Gold Mining in the Ponce Enriquez, Portovelo-Zaruma and Nambija Areas, Equador. *Journal of Water, Air, and Soil Pollution* 131: 19 – 39.
- BALIHRISTI, (2008). Laporan Akhir Kegiatan Pengawasan Pelaksanaan PETI. Provinsi Gorontalo.
- Brabo, E.S, Angelica, R.S, Silva, A.P., Faial K.R.F., Mascarenhas, A.F.S., Santos, E.C.O., Jesus, M and Loureiro, E.C.B. (2003). Assessment of Mercury Levels in Soils, Waters, Bottom Sediments and Fishes of Acre State in Brazilian Amazon. *Water, Journal Air and Soil Pollution* 147: 61 – 77, 20
- Connell, D.W., dan Miller, G.J. (1995). *Kimia dan Ekotoksikologi Pencemaran*. Universitas Indonesia Press, Jakarta.
- Darmono, (1995). *Logam Dalam Sistem Biologi Makhluk Hidup*. Universitas Indonesia, Jakarta.
- Effendi, Hefni. (2003). *Telaah Kualitas Air*. Kanisius, Yogyakarta.
- Ikhsan, C. (2007). Pengaruh Variasi Debit Air Terhadap Laju Bed Load Pada Saluran Terbuka dengan Pola Aliran Steady Flow. *Jurnal Penelitian Media Teknik Sipil*. UNS Hal 63-68.
- Lilebo, A.I., Coelho, P.J., Pato, P., Valega, M, Nargalho, R., Reis M., Raposo, J., Pareira, E, Duarte, A.C. and Pardal, M.A., (2010). Assessment of Mercury in Water, Sediments, and Biota of a Southern European Estuary (Sado Estuary, Portugal), *Journal of Water Air Soil Pollut* (2011) 214 : 667 – 680.
- Mahmud, M. (2012). Model Sebaran Spasial Temporal Konsentrasi Merkuri Akibat Penambangan Emas Tradisional Sebagai Dasar Monitoring dan Evaluasi Pencemaran di Ekosistem Sungai Tulabolo Provinsi Gorontalo. *Dissertation*. Study Program.
- Mahmud, M, Lihawa, F, Patuti I, (2014). *Kajian pencemaran Merkuri terhadap Lingkungan di Kabupaten Gorontalo Utara*. Laporan Penelitian PNBPU Universitas Negeri Gorontalo.
- Mahmud, M., Desei, F., Banteng B.C., Saleh, Y, (2016). Model Pengelolaan Penambangan Emas Tradisional Buladu Kabupaten Gorontalo Utara. Laporan Penelitian PUPU. Tahun I. Universitas Negeri Gorontalo. Gorontalo.
- Palar, H. (2004). *Pencemaran dan Toksikologi Logam berat*. Rineka Cipta, Jakarta.
- Kitong M. T, Abidjulu, J., Koleangan H.S.J. (2012). Analisis Merkuri (Hg) dan Arsen (As) di Sedimen Sungai Ranoyapo Kecamatan Amurang Sulawesi Utara. *Jurnal MIPA Unstrat Online* 1 (1) 16-19.
- Subandri, (2008). Kajian Beban Pencemaran Merkuri (Hg) terhadap Air Sungai Manyuke dan Gangguan Kesehatan pada Penambang sebagai akibat penambangan emas tanpa izin (PETI) di Kecamatan Manyuke Kabupaten Landak Kalimantan Barat. *Thesis*. Program Pasca Sarjana Magister Kesehatan Lingkungan Universitas Diponegoro, Semarang.
- Sutomo, S. (1998). Merkuri dan Bahayanya. *Jurnal Kedokteran dan Farmasi. Medika*, No12 14 Desember 1988, pp.: 1126-1129.
- Widhiyatna, D. (2005). *Pendataan Penyebaran Merkuri Akibat Pertambangan Emas di Daerah Tasikmalaya, Propinsi Jawa Barat*. Kolokium Hasil Lapangan-DIM 2005.