Heavy metals concentration in sediment of Makassar Strait

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Abstract: The concentration of heavy metals (HMs) camium (Cd), cuprum (Cu), chromium (Cr), nickel (Ni), lead (Pb), and zinc (Zn) in sediments of Makassar Strait was investigated. We collected sediment samples from 28 research stations. Atomic Absorption Spectrophotometer (AAS) was used to measure HMs in all samples. This research aims to know the levels of HMs’ contamination and pollution in sediments. The analysis revealed that the concentration of all HMs is still low and inlined with the sediment standard guidelines, exception of Cu and Ni. In regards to the value of L_geo and PLI, sediment in the Makassar Strait is still normal for marine life, and sediment is included in unpolluted categories (PLI<1). The sources of HMs in this strait come from human activities in the land of Kalimantan and Sulawesi Island.

1. Introduction
Makassar Strait is located between the eastern part of Kalimantan Island and the west part of Sulawesi Island. In the coastal areas of the two islands, there are various kinds of activities potentially polluting the marine environment of the region. East Kalimantan, is a very complex estuarine area and is a confluence of several large rivers such as the Mahakam river, Bungus river, Sangata river, Barumuda river, Bungalu river, Karangan river, and Manubar river. Each river has different characters. In addition, East Kalimantan is known as one of the largest foreign exchange earners in Indonesia which has abundant natural resource potential, particularly in the mining and timber sectors. Industry and settlements are growing rapidly along the coast of Sangkulirang-Samarinda. These industries are LNG, coal, organic chemical fertilizers, shipbuilding, iron and steel construction, heavy equipment repair maintenance, tire retreading industry, and wood industry along the Mahakam River. Likewise with the western part of Sulawesi Island, various activities dump waste into the sea waters, thus, it affects the condition of the Makassar Strait waters. Studies on HMs’ pollution have escalated in recent decades on a relatively large scale, especially in coastal areas [1]. This is because information on pollution is still very limited in tropical and subtropical areas such as Indonesia, where a large number of heavy metal pollutants come from nature and from human activities, such as ports, industries, agriculture, and housing around the estuarine which causes the surrounding environment to be polluted.

Sediment serves as a heavy metal adsorber which is important in the hydrological cycle. HMs emissions generally come from human activities that accumulate in river or sea sediments, these heavy metals are adsorbed by fine materials like clays. Therefore, sediments can detach HMs to the water layer through anthropogenic or natural processes, and can also have harmful effects on the ecosystem.
Benthic organisms absorb metals directly or indirectly from sediments, and in turn, leveling up the probability for these metals to enter the food chain [1, 2].

The assessment of HMs contamination in sediments in the estuarine region involves a complex data, which requires combination of physico-chemical and biological mechanisms. Unfortunately, in some areas, the imperative biological data are often unavailable. The estimation of environmental geochemical contamination status, commonly utilizing the Sediment Quality Guidelines (SQGs) or quantitative indices such as geoaccumulation index (I_geo), contamination factor (CF), which refers to reference values such as normal values in nature (background values), concentration mean of shale, continental crust, etc [3, 4, 2].

The purpose of the study is to determine the level of contamination and to assess the contamination of HMs Cadmium (Cd), Cuprum (Cu), Chromium (Cr), Nickel (Ni), Lead (Pb), and Zinc (Zn) in sediments in the Makassar Strait by considering the Sediment Quality Guidelines, and computing Contamination Factors (CF), Pollution Load Index (PLI), Geoaccumulation Index (I_geo) as well.

2. Materials and Method
2.1. Research sites
The research was conducted in the sea waters of the Makassar Strait from September to October 2004 using the RV Baruna Jaya VIII. The position of the station was determined purposively following the research objectives (Figure 1).

2.2. Sampling and Analysis
We used a gravity core to collect the sediment from bottom of the sea then we kept them into a cleaned polyethylene box. We dried the sample using an oven at 105 °C for 24 hours. To get a complete cleaning of the sample, we rewashed the dried sampled with distilled water. The samples wer re-dried using the same heating protocol as previous step till they obtained a constant weight. We ground the samples to gain a fine particle. Those fine samples, 5 grams for each sample, were then underwent a digestion protocol using HNO₃/HCl. First step of digestion is to let the mixture in room temperature for at least 4 hours. The second step was heating the mixture at 90 °C for 8 hours [5]. The quantification of Cadmium (Cd), Cuprum (Cu), Chromium (Cr), Nickel (Ni), Lead (Pb), and Zinc (Zn) of the pre-treated/digested samples was carried out using AAS Varian Techtron model IBQ AA-1475. The quality of data was ensured by analyzing a reference material PACS-2. The recovery varied from 95% to 100% and the deviation of reference material’s measurement is <5%. The concentration of HMs is expressed in part per million, ppm (dry weight).
2.3. Determination of Sediment Pollution Level

The pollution of HMs in sediments was assessed applying several indices for sediment standards such as PLI and $I_{geo}$ [6-10] as the following formula:

$$I_{geo} = \log_2 \left[ \frac{C_x}{1.5B_n} \right]$$  \hspace{1cm} (1)

$C_x$ = concentration of HM $x$ in the example, $B_n$ = background concentration of HM $x$ in nature $1.5$ = constanta

$$CF = \frac{C_x}{C_{background}(B_n)}$$  \hspace{1cm} (2)

$$PLI = [CF1 \times CF2 \times CF3 \times ... CFn]^{1/2}$$  \hspace{1cm} (3)
The calculation of aforementioned indices produced values and the interpretation of the value was presented in Table 1.

### Table 1. The interpretation of indices used in the study

| Criteria | Geoaccumulation Index, \( I_{geo} \) | Pollution Load Index, PLI | Contamination Factor, CF |
|----------|------------------------------------|--------------------------|------------------------|
| Unpolluted | \( I_{geo} < 0 \) | \(<1\), unpolluted | \( CF < 1 \) |
| Unpolluted to moderately polluted | \( 0 < I_{geo} < 1 \) | 1-2 | 1<\( CF < 3 \) |
| Moderately polluted | \( 1 < I_{geo} < 2 \) | 2-4 | 3<\( CF < 6 \) |
| Moderately to strongly polluted | \( 2 < I_{geo} < 3 \) | 4-6 | \( CF < 6 \) |
| Strongly polluted | \( 3 < I_{geo} < 4 \) | 6-8 | Very high contamination |
| Strongly to very strongly polluted | \( 4 < I_{geo} < 5 \) | 8-10 | |
| Very strongly polluted | \( I_{geo} > 5 \) | | |

3. Results and Discussion

3.1. Results

The measurements results of HMs levels in sediments at the Makassar Strait (Table 2 and Figure 2).

### Table 2. HMs content in the sediment of Makassar Strait

| St | Pb ppm | Cd ppm | Cu ppm | Cr ppm | Zn ppm | Ni ppm |
|----|--------|--------|--------|--------|--------|--------|
| 1  | 5.32   | 0.15   | 10.95  | 12.70  | 36.24  | 12.80  |
| 2  | 10.74  | 0.26   | 21.21  | 28.79  | 74.68  | 35.69  |
| 3  | 18.16  | 0.26   | 28.15  | 41.02  | 81.05  | 32.92  |
| 4  | 12.77  | 0.13   | 11.90  | 23.53  | 47.55  | 20.41  |
| 5  | 18.20  | 0.10   | 27.03  | 27.35  | 78.33  | 28.95  |
| 6  | 16.83  | 0.27   | 28.82  | 29.94  | 92.39  | 33.46  |
| 7  | **19.11** | **0.29** | **33.03** | **33.81** | **98.52** | **40.11** |
| 8  | 10.53  | **0.35** | 26.17  | 28.08  | 99.84  | 36.07  |
| 9  | 6.55   | 0.14   | 11.13  | 21.71  | 47.80  | 21.32  |
| 10 | 9.68   | 0.06   | 13.93  | 20.53  | 73.93  | 29.01  |
| 11 | 8.94   | 0.16   | 10.71  | 16.80  | 56.54  | 18.24  |
| 12 | 0      | 0      | 0      | 0      | 0      | 0      |
| 13 | 0      | 0      | 0      | 0      | 0      | 0      |
| 14 | 12.40  | 0.10   | 25.14  | 30.38  | 82.90  | 34.56  |
| 15 | 7.40   | 0.08   | 16.76  | 26.27  | 53.45  | 23.66  |
| 16 | 11.46  | 0.22   | 26.31  | 24.40  | 98.70  | 41.92  |
| 17 | 9.20   | 0.08   | 13.80  | 23.25  | 66.75  | 22.82  |
| 18 | 5.79   | 0.19   | 8.17   | 15.95  | 40.17  | 13.50  |
| 19 | 11.78  | 0.18   | 27.10  | 25.68  | 101.85 | **43.29** |
| 20 | 14.50  | 0.29   | 28.53  | 28.60  | 89.59  | 32.84  |
|    | Pb  |  |  | Cu  |  | Cr  |  |  |  |
|----|-----|---|---|-----|---|-----|---|---|---|
|    | 21  | 15.40 | 0.10 | 17.03 | 43.04 | 77.25 | 29.52 |    |    |
|    | 22  | 13.78 | 0.19 | 31.82 | 33.01 | 103.03 | 39.33 |    |    |
|    | 23  | 9.43  | 0.19 | 11.48 | 17.93 | 52.44 | 18.66 |    |    |
|    | 24  | 10.63 | 0.17 | 10.14 | 22.41 | 62.13 | 21.07 |    |    |
|    | 25  | 6.62  | 0.07 | 7.80  | 12.93 | 53.44 | 15.00 |    |    |
|    | 26  | 6.80  | 0.02 | 9.35  | 24.76 | 53.25 | 24.80 |    |    |
|    | 27  | 8.84  | 0.02 | 10.16 | 22.82 | 62.40 | 24.55 |    |    |
|    | 28  | 9.78  | 0.04 | 13.46 | 24.35 | 78.29 | 27.46 |    |    |
| Min| 5.32 | 0.02 | 7.8  | 12.7  | 36.24 | 12.8  |    |    |
| Max| 19.11| 0.35 | 33.03| 43.04 | 103.03| 43.29 |    |    |
| SD | 3.972 | 0.091 | 8.460| 7.381 | 20.435| 9.015 |    |    |
| Average| 11.178 | 0.160 | 18.464| 25.386 | 70.635 | 27.896 |    |    |

ND (No Data)
Figure 2. Graphic of HMs concentration of Pb, Cd, Cu, Cr, Zn, and Ni (ppm)

From the table above, it can be seen that the levels of Cadmium (Cd), Cuprum (Cu), Chromium (Cr), Nickel (Ni), Lead (Pb), and Zinc (Zn) were diverged at each station. On average, the highest heavy metal content is Zn> Ni> Cr> Cu> Pb> Cd.

3.2. Discussion

3.2.1. Distribution of HMs

The distribution of Pb metal at each station is uneven (Table 2). Overall, Pb levels ranged from 5.32-19.11 ppm with an average of 11.178 ppm. The highest and lowest Pb content was found at station 7 and station 1, respectively. This data shows some stations accumulate greater lead compared to others. The enrichment of metal lead in sediments can come from anthropogenic sources or geological weathering.

The average Pb levels from these observations were relatively high. Edward (2010; 2011) reported that Pb levels in sediments in Elat waters (Southeast Maluku) were relatively unpolluted and still natural, ranging from <0.001–0.274 ppm with a mean of 0.042 ppm [11, 12]. Other locations also showed unpolluted water by Pb such as in Ngilngo (Tual, Southeast Maluku) 3.187-3.921 ppm, and in Ohoimas (Southeast Maluku) 2.781-4.033. This Pb level is still lower than the normal Pb level found in nature, which is 12.5 ppm [13]. The Canadian Council of Ministers of the Environment (CCME, 2002) assigns 35 ppm as the threshold value of lead in sediments for the protection of biota [14]. Meanwhile, the Sediment Quality Guidelines (SQG) assigns Pb content of <40 ppm as the uncontaminated sediment [13, 15]. KMNLH (2010) determined the Pb Threshold Value in sediments for biota to be 36.8 ppm [16]. Based on the CCME, SQG, and KMNLH criteria above the Pb level are still safe for marine biota that lives and forages in sediments.

Cd levels in the sediment ranged from 0.02-0.035 ppm with an average of 0.158 ppm. Cd levels are also not evenly distributed at each station. One station showed the highest levels of Cd (station 6) and two stations (26 and 27) exhibited the lowest concentration. Station 6 was suspected to receive more Cd input than others. Cd levels in these three locations were relatively low. Edward (2010; 2011) found Cd levels in Elat waters (Southeast Maluku) which were relatively unpolluted and still natural ranged <0.001-0.0172 ppm (mean= 0.009 ppm), in Ohoimas (Southeast Maluku) 0.216-0.295 ppm.
The British Columbia Ministry of Water, Land and Air Protection (BCMWLAP) (2006b) sets 16 ppm as that the lowest value of Ni in sediment causing negative effects [20]. The normal average level of Ni in nature is 75 ppm. The SQG determine the uncontaminated sediments by Ni content is <20 ppm, and 20-50 ppm as moderately polluted sediments [13]. Based on the BCMWLAP, Ni content is dangerous for marine biota.

3.2.2. Contamination Factors (CF) and Pollution Load Index (PLI)
The calculation of the CF and PLI is presented in Table 3. CF Pb value ranges from 0.266-0.955 with a mean of 0.558. The highest CF Pb value was found at Station 7, namely 1.528, and the lowest was at Station 1, namely 0.425. This data shows that station 7 has a higher level of contamination than other stations. Station 1, 2, 8, 10-19, 23-27 showed CF value ranges from 0.425-0.992. This value is <1, which means that the stations have a low contamination level, while stations 3-7, 20-22, the CF value ranges from 1.021-1.528, this value is fell between 1 and 3 (1 < CF <3) and included in the moderately contaminated category. On average, the level of Pb contamination was <1.

The value of the CF Cd varied from 0.1 to 1.75 (average 0.790). This value <1 determines low contaminated category (low contamination). Station 8 was the highest concentration of Cd contamination and stations 26 and 27 were the lowest.

| St | CF Pb | CF Cd | CF Cu | CF Cr | CF Zn | CF Ni | PLI |
|----|-------|-------|-------|-------|-------|-------|-----|
| 1  | 0.266 | 0.5   | 0.156 | 0.141 | 0.381 | 0.170 | 0.239|
| 2  | 0.537 | 0.866 | 0.303 | 0.319 | 0.786 | 0.475 | 0.505|
| 3  | 0.908 | 0.866 | 0.402 | 0.455 | 0.853 | 0.438 | 0.614|
| 4  | 0.638 | 0.433 | 0.170 | 0.261 | 0.500 | 0.272 | 0.344|
| 5  | 0.91  | 0.333 | 0.386 | 0.303 | 0.824 | 0.386 | 0.473|
| 6  | 0.841 | 0.9   | 0.411 | 0.332 | 0.972 | 0.446 | 0.595|
| 7  | 0.955 | 0.966 | 0.471 | 0.375 | 1.037 | 0.535 | 0.669|
| 8  | 0.526 | 1.166 | 0.373 | 0.312 | 1.050 | 0.480 | 0.574|
| 9  | 0.327 | 0.466 | 0.159 | 0.241 | 0.503 | 0.284 | 0.306|
| 10 | 0.484 | 0.200 | 0.199 | 0.228 | 0.778 | 0.386 | 0.331|
| 11 | 0.447 | 0.533 | 0.153 | 0.186 | 0.595 | 0.243 | 0.315|
| 12 | ND   | ND    | ND    | ND    | ND    | ND    | ND  |
| 13 | ND   | ND    | ND    | ND    | ND    | ND    | ND  |
| 14 | 0.62  | 0.333 | 0.359 | 0.337 | 0.872 | 0.460 | 0.464|
| 15 | 0.37  | 0.266 | 0.239 | 0.291 | 0.562 | 0.315 | 0.326|
| 16 | 0.573 | 0.733 | 0.375 | 0.271 | 1.038 | 0.558 | 0.539|
| 17 | 0.46  | 0.266 | 0.197 | 0.258 | 0.702 | 0.304 | 0.331|
| 18 | 0.289 | 0.633 | 0.116 | 0.177 | 0.422 | 0.18  | 0.256|
| 19 | 0.589 | 0.600 | 0.387 | 0.285 | 1.072 | 0.577 | 0.537|
| 20 | 0.725 | 0.966 | 0.407 | 0.317 | 0.943 | 0.437 | 0.577|
| 21 | 0.77  | 0.333 | 0.243 | 0.478 | 0.813 | 0.393 | 0.460|
| 22 | 0.689 | 0.633 | 0.454 | 0.366 | 1.084 | 0.524 | 0.587|
| 23 | 0.471 | 0.633 | 0.164 | 0.199 | 0.552 | 0.248 | 0.331|
| 24 | 0.531 | 0.566 | 0.144 | 0.249 | 0.654 | 0.280 | 0.354|
| 25 | 0.331 | 0.233 | 0.111 | 0.143 | 0.562 | 0.200 | 0.227|
| 26 | 0.34  | 0.066 | 0.133 | 0.275 | 0.560 | 0.330 | 0.230|
| 27 | 0.442 | 0.066 | 0.145 | 0.253 | 0.656 | 0.327 | 0.247|
| 28 | 0.489 | 0.133 | 0.192 | 0.270 | 0.824 | 0.366 | 0.317|
| Min| 0.266 | 0.066 | 0.111 | 0.141 | 0.381 | 0.170 | 0.227|
| Max| 0.955 | 1.166 | 0.471 | 0.478 | 1.084 | 0.577 | 0.669|
| Ave| 0.558 | 0.526 | 0.263 | 0.281 | 0.753 | 0.369 | 0.413|
| SD | 0.198 | 0.302 | 0.120 | 0.081 | 0.215 | 0.118 | 0.139|

ND (No Data)

The Cu contamination factor value ranged from 0.111 to 0.60 with a mean of 0.263 (<1), which means it was categorized as low contamination. The highest level of contamination was found at station 7 and the lowest at station 5. The value of the Cr contamination factor ranged from 0.127 to 0.430 with a mean of 0.253 (<1) which means it is a low level of contamination.

The highest level of Cr contamination was found at station 21 and the lowest at station 1.
The value of the Zn contamination level ranged from 0.517 to 1.471 with a mean of 1.022 (<1), which means it is also in the low contamination category. The highest level of Zn contamination was found at station 19 and the lowest at station 1.

The value of the Ni contamination level ranged from 0.17 to 0.577 with a mean of 0.369 (<1), which means it is also in the low contamination category. The highest level of Ni contamination was found at station 19 and the lowest at station 1.

Based on the data above (Table 2) the mean concentration of contamination are as follows: Zn>Pb>Cd>Ni>Cr>Cu. This shows that Pb has a higher level of contamination than other metals. The overall PLI value varied from 0.227-0.669 (mean 0.413), this value is <1 meaning unpolluted condition in the sediment in these waters, except for station 20. The PLI is 1.1534 (>1) implying a lightly polluted category.

3.2.3 Geoaccumulation Index (Igeo)

The results for the geo accumulation index’s calculation are presented in Table 4. The Igeo value of Pb spread from -2.495 to -0.65 (mean of -1.512). The Igeo value of Pb is in the negative range and the mean <0, reflecting unpolluted condition by Pb. The Igeo values of cadmium is ranged from -4.491 to -3.362 with a mean of -1.828. This value means the sediment is unpolluted by Cd (Igeo value < 0). Igeo values of Cu spread between -3.75 and -1.668 (mean= -2.66). The sediment is categorized uncontaminated with Cu (value <0).

Tabel 4. Geoakumulation Index (Igeo) in Sedimen in Makassar Strait

| St | Pb   | Cd   | Cu   | Cr   | Zn   | Ni   |
|----|------|------|------|------|------|------|
| 1  | -2.495 | -1.584 | -3.261 | -3.410 | -1.975 | -3.135 |
| 2  | -1.481 | -0.791 | -2.307 | -2.229 | -0.932 | -1.656 |
| 3  | -0.724 | -0.791 | -1.899 | -1.718 | -0.814 | -1.772 |
| 4  | -1.232 | -1.791 | -3.141 | -2.520 | -1.583 | -2.462 |
| 5  | -0.721 | -2.169 | -1.957 | -2.303 | -0.863 | -1.958 |
| 6  | -0.833 | -0.736 | -1.865 | -2.172 | -0.625 | -1.749 |
| 7  | -0.650 | -0.633 | -1.668 | -1.997 | -0.532 | -1.487 |
| 8  | -1.510 | -0.362 | -2.004 | -2.265 | -0.513 | -1.641 |
| 9  | -2.195 | -1.684 | -3.237 | -2.636 | -1.575 | -2.399 |
| 10 | -1.631 | -2.906 | -2.914 | -2.717 | -0.946 | -1.955 |
| 11 | -1.746 | -1.491 | -3.293 | -3.006 | -1.333 | -2.624 |
| 12 | ND   | ND   | ND   | ND   | ND   | ND   |
| 13 | ND   | ND   | ND   | ND   | ND   | ND   |
| 14 | -1.274 | -2.169 | -2.062 | -2.151 | -0.781 | -1.702 |
| 15 | -2.019 | -2.491 | -2.647 | -2.361 | -1.414 | -2.249 |
| 16 | -1.388 | -1.032 | -1.996 | -2.468 | -0.529 | -1.424 |
| 17 | -1.705 | -2.491 | -2.927 | -2.537 | -1.094 | -2.301 |
| 18 | -2.373 | -1.243 | -3.683 | -3.081 | -1.826 | -3.058 |
| 19 | -1.348 | -1.321 | -1.954 | -2.394 | -0.484 | -1.377 |
| 20 | -1.048 | -0.633 | -1.879 | -2.238 | -0.669 | -1.776 |
| 21 | -0.962 | -2.169 | -2.624 | -1.649 | -0.883 | -1.930 |
| 22 | -1.122 | -1.243 | -1.722 | -2.031 | -0.467 | -1.516 |
| 23 | -1.669 | -1.243 | -3.193 | -2.912 | -1.442 | -2.591 |
| 24 | -1.496 | -1.404 | -3.372 | -2.590 | -1.197 | -2.416 |
| 25 | -2.180 | -2.684 | -3.750 | -3.384 | -1.414 | -2.906 |
| 26 | -2.141 | -4.491 | -3.489 | -2.446 | -1.420 | -2.181 |
| 27 | -1.762 | -4.491 | -3.369 | -2.564 | -1.191 | -2.196 |
| 28 | -1.617 | -3.491 | -2.963 | -2.470 | -0.864 | -2.034 |
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Table 6. Correlation among HMs in Sediment of Makasar Strait (n=28)

|       | Pb  | Cd      | Cu      | Cr       | Zn       | Ni       |
|-------|-----|---------|---------|----------|----------|----------|
| Pb    |     |         |         |          |          |          |
| Cd    | 0.579** | 1       |         |          |          |          |
| Cu    | 0.853** | 0.707** |         | 1        |          |          |
| Cr    | 0.868** | 0.496** | 0.782** | 1        |          |          |
| Zn    | 0.816** | 0.617** | 0.903** | 0.811**  | 1        |          |
| Ni    | 0.785** | 0.590** | 0.905** | 0.826**  | 0.967**  | 1        |

** significance at 0.01 level (2-tailed).

4. Conclusion
The measurement of HMs indicated different accumulation in each station. The concentration of each HMs also differs for Zn> Ni> Cr> Cu> Pb> Cd. Among those HMs, cuprum and nickel concentration were above the guideline for marine biota, meanwhile, cadmium, chromium, zinc, zinc and lead were below the guideline. Considering the Pollution Load Index (PLI) value, the sediment in the Makassar Strait has not been polluted by HMs cadmium, cuprum, chromium, lead, nickel and zinc metals, and based on the criteria set by the Sediment Quality Guideline (SQG), Cu and Ni levels in the sediment are relatively high and already above the criteria set by several SQGs.

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