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Heavy Metals Source Identification by Enrichment Factor in Bangka Island Sediments, Indonesia

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Abstract

The concentration of heavy metals in the sediments of the waters of Bangka Island, Indonesia results in a decrease in water quality. The source of heavy metals is unknown; therefore, it has become a controversial issue for the coastal communities of Bangka Island. This study aimed to determine the source of the heavy metals Zn, Cu, Pb, Cd, Sn in the sediments of the waters of Bangka Island. Sediment sampling at 12 stations was carried out in July 2019, using the APHA; 2012 method. Heavy metal concentrations were measured using the Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) technique, whereas heavy metal sources were analysed using the Enrichment Factor (EF) approach. The results showed EF > 1 at stations 1, 2, 3, 4, 5, 6, 8, 9, 10 and 11. Zn and Sn were found to be enriched in the minimum to moderate category, Pb was seen to be highly enriched, while Cu and Cd did not show any enrichment (EF < 1). The EF values of the heavy metals Zn, Cu, Pb and Cd at stations 7 and 12 showed EF < 1, indicating that the dominant metals were of natural origin. The sources of the heavy metals Zn, Cu, Pb, Cd and Sn at stations 1, 2, 3, 4, 5, 6, 8, 9, 10 and 11 were a mixture of anthropogenic and natural materials. Anthropogenic sources in the waters of the Bangka Island include: tin mining waste, fuel and paint spills, and smoke from tin mining machines (pontoons). These results inform the public and the government that community activities (anthropogenic), natural resources, and their mutual interactions constantly enrich and increase the concentrations of Zn, Cu, Pb, Cd and Sn.

Keywords: Anthropogenic sources; Bangka Island; Natural sources; Sediments

1. Introduction

The waters of Bangka Island are located in an area that has seen lead mining activities, both on land and in water, since 1998 , with a marked rise in 2001 (Nurtjahya and Agustina, 2015). This activity leads to a decrease in water quality, chiefly as a result of the increased water turbidity (Nurtjaya *et al.,* 2017).

In addition, suspended particles also contain heavy metals (Blodau, 2006; Harmesa *et al.,* 2020); hence, their concentration increases in the sediment (Yang *et al.,* 2009; Riani *et al.,* 2018, Suteja *et al.,* 2020). Heavy metals can also disperse and enter other locations due to physical processes such as water currents, waves, and tides.

Non-mining activities on the shores of Bangka Island (ice factories, shipyards, ship repair shops, SPBN) have also increased waste and heavy metal content in the waters (Arifin, 2011). Coastal communities deduce that the increased concentration of heavy metals in sediments causes a decrease in fishing resources. Prohibition of activities that can increase heavy metal concentration in the waters has become a matter of conflict and debate. The concentration of heavy metals in sediments is always higher than that in water (Riani *et al.,* 2018; Suteja *et al.,* 2020). The high current speed enhances the resuspension process (the return of metals from sediments to the water column), thereby increasing the heavy metal concentration in water and endangering organisms (Pertiwi *et al.,* 2021). It is not known if the sources of Zn, Cu, Pb, Cd, Sn in the sediments of the coastal regions of Bangka Island are natural or anthropogenic. Certainty in this regard is needed to find a solution for these problems. Research on sources of heavy metals in sediments using the Enrichment Factor (EF) approach has been carried out in the Cimadur River, Banten, Indonesia (Mulyaningsih and Suprapti, 2015), Lampung Bay, Indonesia (Permata *et al.,* 2018), Java Sea (Pugung *et al.,* 2018), Bali Strait (Yona *et al.,* 2018), Xiamen Bay, China (Zhang *et al.,* 2007), Taiwan (Lin *et al.,* 2013), South Coast of India (Salas *et al.,* 2017), and Cimanuk Waters, Indramayu, Indonesia (Harmesa *et al.,* 2020).

However, such research has never been carried out in the waters of Bangka Island. Water and sediment have similarities with regard to the content of heavy metal in them, which relates to heavy metal solubility in water (Pachana *et al.,* 2010). Heavy metals lead to various chemical and physical reactions in the water column, and subsequently affect chemical composition and particle weight (Suteja *et al.,* 2020). While particles run into processing upon ion exchange, the flocculates formed are heavier and have higher aggregate specific gravity than water. These later settle in the sediments, which causes the heavier metals to accumulate in the sediments rather than in the water column (Turekian, 2010).

Heavy metals easily bind organic matter and particles also settle at the bottom of the water; hence the concentration of heavy metals in sediments has become higher (Permata *et al.,* 2018). Several studies have revealed that heavy metal concentration in sediments is higher than that in water, i.e. Pb and Cd in Karya Island and Panggang Island (Riani *et al.,* 2017); Pb, Cd, Cu, and Zn in the Cimanuk Estuary (Harmesa *et al.,* 2020), Sn in the Mediterranean Coast of Egypt (Hamed *et al.,* 2013). Previous studies have applied the EF to determine whether the source of heavy metals in aquatic sediments is natural or anthropogenic (Barbieri, 2016). The EF confirms the input levels of heavy metals in the waters (Barbieri, 2016), as well as the status and level of environmental pollution (Feng *et al.,* 2004). The EF is used for several reasons: sediment-hosted stratiform mineralization (Perelló *et al.,* 2021; Fan *et al.,* 2021), a valuable record of human activity (Álvarez-Iglesias *et al.,* 2020), evaluation of heavy metal absorbers and carriers (Zhang *et al.,* 2017), and assessment of the ecological risk of trace metals in sediments (Hanif *et al.,* 2016). Therefore, the current study applied EF to assess the source of heavy metals in Bangka Island, focusing on a specific set of heavy metals (Zn, Cu, Pb, Cd, and Sn).

2. Materials and Methods

2.1 Sediment Sampling

Sampling was conducted at 12 stations with three replications. All of these stations represent each district on Bangka Island: Station 1 (Front of the mouth of Kelabat Bay), station 2 (Limau Beach, Kelabat Bay), station 3 and 4 (Cupat Beach, Kelabat Bay), station 5 (Semulut Beach, Kelabat Bay), station 6 (Nanas Beach, Kelabat Bay) West Bangka Regency, station 7 (Rebo Beach, Bangka Induk Regency), station 8 (Tanah Merah Beach, Central Bangka Regency), station 9 (Tukak Beach), station 10 (Anak Air Island), station 11 (Sadai Beach), and station 12 (Permis Beach) South Bangka Regency (Figure 1).
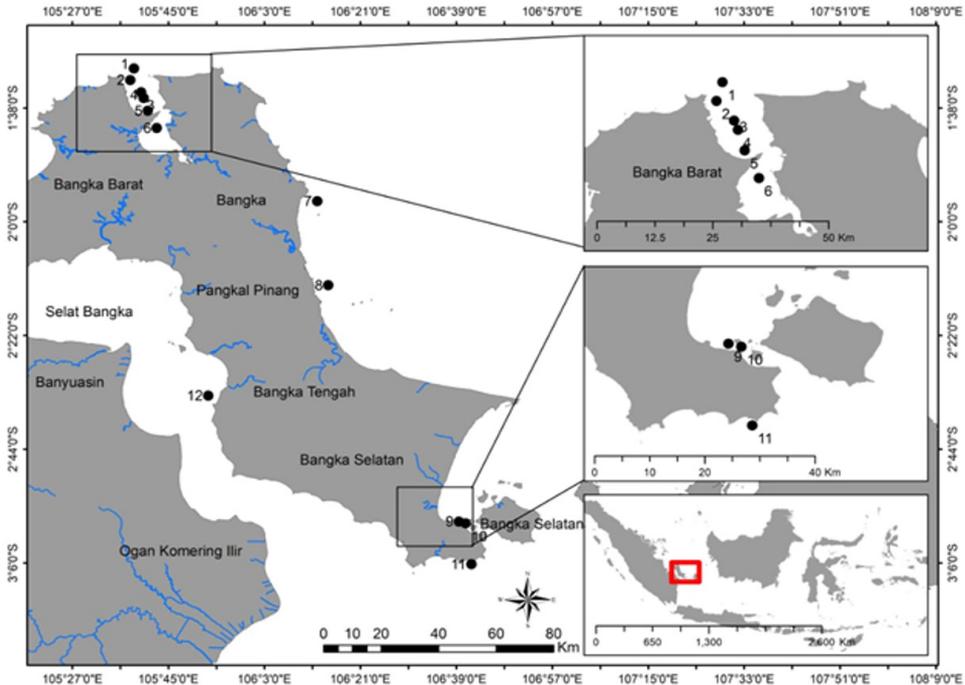
Figure 1. Map of Research Sites and Sediment Sampling in Bangka Island Waters

Station 1 is located in the outer part of Kelabat Bay. The dominant substrate with clay texture at this station is at a higher level than other stations, and the current is faster than in Kelabat Bay. Stations 2, 3, 4, 5 and 6 are located in the Kelabat Bay area, which has tin mining leading to a high turbidity level. Station 7 consists of open water with lots of granite on the shore. Station 8 has open water with local, traditional tin mining. Station 9 is located in seagrass waters close to residential areas and community activities. Station 10 is a very small island with sandbars and seagrass beds. Station 11 is a waterway with an inter-island ferry port, hence sand dredging often reduces siltation. Station 12 represents a location with several granite rocks by the beach and tin mining, far away from the mainland.

Sampling was conducted in July 2019 at 12 stations, with each station having 3 replications, thereby leading to a total of 36 sediment samples. Sediment was collected using an Eckman grab, and the sample used was from the middle portion (untouched by the Eckman

grab). The sample was put into a flip-lid plastic container to measure heavy metals and sediment texture. All samples were stored in a coolbox at 4 °C during transportation. Sediment texture analysis was performed using tiered sieving and pipetting methods.

2.2 Analysis of sediment texture

Samples of the sediment were collected in sufficient quantity and dried in an oven for 16 h at 105 - 110 °C. Samples of dry sediment were mashed with a mortar, and 25 gms of the samples were collected, placed and sieved in a sieve shaker for 20 min, using sieves with sizes of 2 mm, 0.5 mm, 0.125 mm and 0.063 mm. A sediment sample measuring 0.063 mm was put in a 1000 mL volumetric measuring cup, homogenized and subjected to pipetting process, based on the distance and time of the pipette. The percentage value of each fraction was then calculated, plotted, and named according to Wentworth's classification (Sanders *et al.,* 2010; Romano *et al.,* 2017).

2.3 Analysis of Total Concentration of Heavy Metals (Zn, Cu, Pb, Cd, Sn) in Sediment

Each station has three replications, so we had a total of 36 sediment samples. Sediment samples from each station were cleaned of litter and shell fragments. The sediment samples were dried in an oven at 60 °C for 24 h. The dry sediment samples were then pulverized and homogenized using a pestle and mortar (Suteja *et al.,* 2020); 0.5 g of the fine sediment sample was put in a vessel tube; subsequently 5 mL of nitric acid (HNO_3) and 1 mL of concentrated hydrogen peroxide (H_2O_2) were added to the sample and allowed to stand for 20 - 30 min. Furthermore, the sediment sample was destroyed using a microwave in a closed vessel at a temperature of 190 °C for 1 h. The sample was removed from the microwave, and the vessel lid was opened slowly. The sample was poured into a 50 mL volumetric flask, then dissolved with distilled water to the limit of 50 mL. If there was a precipitate, it was filtered using a 42 micron filter paper (APHA, 2002). Heavy metal analysis was carried out by measuring the heavy metal concentrations at the Regional Health Laboratory DKI Jakarta using ICP-OES: Thermo Scientific iCAP 7000 series ICP OES.

2.4 Assessment of Sediment Enrichment Factor (EF)

EF is an assessment criterion used to determine the source of heavy metals in the aquatic environment, whether anthropogenic or natural (Choi *et al.,* 2012; Barbieri, 2016). The EF value of heavy metals was calculated by normalizing the concentration of heavy metals in the sediments with their natural concentrations in the earth's crust. Aluminium (Al) was used as a normalization metal because of the abundance of water, and also as it is known to be stable and uninfluenced by chemical-biological processes (Barbieri, 2016). Moreover, the coastal sediments had predominance of alumina-silicate content (Chen *et al.,* 2007). The calculation of the EF value was based on and used the following equation (Barbieri, 2016):

$$EF = \frac{X_{(a)} \, / \, Al_{(a)}}{X_{(b)} \, / \, Al_{(b)}} \qquad (1)$$

$X_{(a)}$: concentration of metal X in sediment samples (mg/kg)

$Al_{(a)}$: concentration of metal Al in sediment samples (mg/kg)

$X_{(b)}$: concentration of metal X in background value (mg/kg)

$Al_{(b)}$: concentration of metal Al in background value (mg/ kg)

In this study, the background value refers to the average natural value, with the following EF criteria: EF less than 2 (Less enrichment, at a minimum level); EF 2 – 5 (Moderate enrichment); EF 5 – 20 (Moderately high enrichment); EF 20 – 40 (Very high enrichment); and EF over 40 (Extremely high enrichment) (Zhang *et al.,* 2007; Mmolawa *et al.,* 2011).

2.5 Data analysis

Characteristics of the aquatic environment were analysed, including heavy metals in sediment (Zn, Cu, Pb, Cd, Sn), C-Organic content, sediment texture, pH, DO, salinity, temperature, current speed, depth, and TSS. The distribution of these characteristics was analysed using Principal Component Analysis (Bengen, 2000). The relationship between sediment texture, C-Organic and heavy metals was analysed using Pearson correlation. The EF values of heavy metals were analysed descriptively. The EF values were processed using R software and presented in a graphical form.

3. Results and Discussion

3.1 The Concentration of Heavy Metals (Zn, Cu, Pb, Cd, Sn, Al) in the Sediments of Bangka Island Waters

The concentration of heavy metals (Zn, Cu, Pb, Cd, Sn, Al) at the research station showed varying values. Predominantly high concentrations of Zn and Pb were found at stations 1, 2, and 4 (Table 1). Heavy metal concentrations of Zn and Pb ranged at

29.295 ± 0.490 mg/kg and 26.665 ± 0.501 mg/kg at station 1; these ranged at 15.877 ± 0.421 mg/kg and 19.214 ± 0.551 mg/kg at station 2. The Zn and Pb metals concentration at station 4 was higher than other stations, that is, in the range of 26.179 ± 0.168 mg/kg and 34.942 ± 0.515 mg/kg, respectively. The high concentration of Pb indicates a greater Pb adsorption capacity compared to other metals.

Heavy metals are usually bound more to the mud or clay substrate. Smooth textures tend to have a large surface area and a small diameter (Mlayah *et al.,* 2009). The sediment texture causes stations 1, 2, and 4 to contain higher concentrations of heavy metals than other stations. On the other hand, at station 11, the dominant sediment texture was mud (81.7%), but it showed low heavy metal content. The association between heavy metal concentrations and sludge substrate was statistically proved. Pearson analysis showed a positive correlation between the mud substrate and heavy metals (Zn, Cu, Pb, Cd, Sn), although the correlation was quite low (r = 0.362, 0.316, 0.407, 0.370, 0.206). The dominance of different textures (sandy substrate) was found at stations 3, 5, 6, 7, 8 and 12 (Table 2). Statistically, the sandy substrate negatively correlated with the concentration of heavy metals. This resulted in the mud texture substrate having a higher metal content.

The different effects at station 11 are thought to be due to dredging. Station 11 is the Sadai Port area at the crossing point of routes between Sadai and Tanjungpandan, as well as Sadai to Manggar and Pontianak. At this port, routine dredging activities are carried out to avoid silting. This activity results in sediment extraction not being carried out on the sediment surface and causes low heavy metal content in the sample. Several studies have shown that the greater the depth, the lower the heavy metal concentrations (Zhu *et al.,* 2019).

Heavy metal concentrations (Zn, Cu, Pb, Cd, Sn) at 12 stations can be the effect of external or natural enrichment. The source of heavy metals at each station was identified using the EF (Figure 2). This study can thus provide answers to questions about heavy metal sources, which are being debated by the

coastal community of Bangka Island.

Based on the research data, it is known that the sediment texture of station 1 predominantly includes clay (61.76%), while stations 2 and 4 show the dominance of mud ranging at 80.67% and 84.63%, respectively (Table 2). The metallic bond with a fine texture causes stations 1, 2 and 4 to have higher heavy metal content than other stations. Several studies have shown that the concentration of heavy metals in sediments is closely related to sediment texture (Liang *et al.,* 2019). Heavy metals usually bind more strongly to mud or clay, because the fine texture has a large surface area, small diameter and high adsorption capacity (Warni *et al.,* 2017).

3.2 Enrichment Factor (EF)

The results showed that the EF values of Zn and Pb in the sediments at stations 1, 2, 3, 4, 5, 6, 8, 9, 10 and 11 showed EF values > 1 (Figure 2). Therefore, Zn and Pb enrichment occurs due to the external environment (anthropogenic) (Mmolawa *et al.,* 2011). Zn enrichment is in the minimal-moderate category, whereas Pb is in the moderately high category. The difference in the enrichment of Pb and Zn occurs due to the varied chemical properties of each heavy metal. These affect the binding process or incorporation of metallic compounds and their concentration in sediments (Darmansyah *et al.,* 2020). In addition to this, the enrichment of Pb and Zn also depends on the waste entering the waters due to community activities. Wastes from community activities include fuel spills, engine fumes, ballast water, ship paint waste, and the shipping industry (Agustriani *et al.,* 2019).

The high enrichment levels of Pb and Zn in the sediment occured because the oil and paint spills and ballast water scattered in the water contain Pb and Zn. These metals are insoluble and are carried in the water column, thereby experiencing dilution and settling in the sediment. This condition occurred at stations 1, 2, 3, 4, 5 and 6 at Kelabat Bay, wherein fishing boat activities largely dominated the area. In addition to fishing activities and crossing routes, there were also mass tin-mining activities using mining

Table 1. Average concentration of heavy metals Zn, Cu, Pb, Cd, Sn, Al (mg/kg) and standard deviation in sediment samples from the waters of Bangka Island

Station	Zn	Cu	Pb	Cd	Sn	Al
1	29.295 ± 0.490	6.841 ± 0.153	26.665 ± 0.501	0.029 ± 0.001	2.896 ± 0.015	31706.367 ± 1891.773
2	15.877 ± 0.421	3.935 ± 0.422	19.214 ± 0.551	0.024 ± 0.002	0.912 ± 0.207	11856.450 ± 2925.373
3	4.903 ± 0.206	0.633 ± 0.131	5.144 ± 0.194	0.012 ± 0.001	0.073 ± 0.012	5461.787 ± 1215.794
4	26.179 ± 0.168	4.485 ± 0.432	34.942 ± 0.515	0.059 ± 0.001	2.473 ± 0.008	24313.667 ± 1238.117
5	3.971 ± 0.059	2.452 ± 0.334	9.292 ± 0.217	0.022 ± 0.001	0.261 ± 0.008	3025.300 ± 1201,282
6	7.058 ± 0.049	2.337 ± 0.467	6.948 ± 0.133	0.023 ± 0.001	0.054 ± 0.039	2521.600 ± 418.463
7	2.537 ± 0.028	1.188 ± 0.107	0.000 ± 0.000	nd	0.287 ± 0.067	6129.912 ± 6077.740
8	13.195 ± 0.092	2.296 ± 0.065	10.001 ± 0.102	nd	1.440 ± 0.282	10043.238 ± 2877.280
9	8.112 ± 0.132	0.726 ± 0.187	6.859 ± 0.037	0.004 ± 0.002	0.688 ± 0.114	2760.576 ± 2474.411
10	7.245 ± 0.486	0.586 ± 0.256	6.272 ± 0.116	0.002 ± 0.001	0.292 ± 0.050	5454.149 ± 965.343
11	10.488 ± 0.299	2.509 ± 0.094	4.932 ± 0.148	0.000 ± 0.000	0.253 ± 0.054	5319.672 ± 874.802
12	3.089 ± 0.042	1.413 ± 0.038	0.000 ± 0.000	0.000 ± 0.000	0.047 ± 0.034	10620.519 ± 5091.057
background value*	70	55	12.5	0.2	2	82300

Note : *background value** (Taylor, 1964); Cd : nd (not detected)

equipment (pontoons) equipped with diesel fuel engines. Many diesel fuel spills occurred in the waters, while the steam engines emanated thick smoke. Smoke from burning fuel has been known to contain Pb (Takarina *et al.,* 2013). This combustion comes from tetraethyl-Pb and tetramethyl-Pb compounds present in the fuel (Stancheva *et al.,* 2013). Pb metal in smoke binds to particles in the atmosphere, and falls into the water when it rains. If these compounds do not undergo a dissolution process, they bond to gain weight

and sink in the sediment. This process shows that the particulate metal fraction is adsorbed by the sediment, whereas the dissolved fraction is carried by water through advection and dispersion processes (Wu and He, 2009). Moreover, the enrichment of heavy metals (Zn, Cu, Pb, Cd, Sn) in the sediments of Bangka Island waters occurred as a result of oxidation of sulfide minerals present in the tailings of tin mining, as the process is known to release heavy metals As, Cd, Cu, Pb, Al, Zn (Blodau, 2006; Mlayah *et al.,* 2009).

Table 2. Composition (%) of sediment texture (sand, mud, clay) at 12 research stations, Bangka Island waters

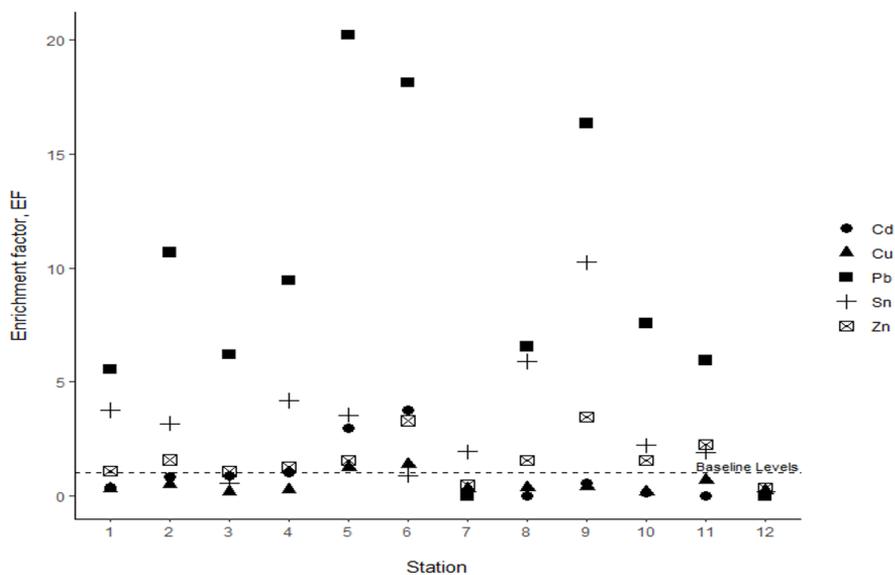| Stations	Sand	Mud	Clay
1	27.77	10.47	61.76
2	0.94	80.86	18.19
3	50.79	13.08	36.13
4	1.49	84.63	13.88
5	70.61	10.90	18.48
6	55.96	11.57	32.47
7	51.86	37.72	10.41
8	52.39	16.69	30.91
9	53.55	13.46	32.99
10	42.25	11.30	46.45
11	6.99	81.76	11.25
12	52.26	23.63	24.10

Figure 2. The Value of the Enrichment Factor (EF) of Heavy Metals Zn, Cu, Pb, Cd, Sn in Bangka Island Waters Sediments

The EF value of Sn at station 1, 2, 4 and 5 indicated a moderate enrichment, while station 7 revealed a minimum enrichment. The low Sn enrichment in sediments occurred because the main target of community tin mining was the heavy metal Sn. Sn found in the sediment was the Sn that had escaped filtering and was included in the waste. The minimum enrichment of Sn at station 7 can also be due to the impact of the location being quite far from the tin mining location. Meanwhile, station 8 was close to the mining location, so the Sn enrichment was relatively high. The different distance ranges showed that Sn enrichment was influenced not only by the current factor that drives flocculates from mining waste, but also by the spread of the metal source in increasing heavy metal enrichment. The high enrichment of Sn also occurred at station 9. Likewise, the high enrichment of Pb occurred at stations 9, 10, and 11 (Figure 2). These three stations were located in South Bangka waters, and there was a post-tin mining area on Tanjung Kerasak Beach, so there may still be heavy metal residues. The current can increase the enrichment of heavy metals. Several research results showed that post-tin mining residues still contain heavy metals, such as As, Cr, Cu,

Pb, Zn, Fe, and Sn (Blodau, 2006, Gyang et al., 2010; Ashraf et al., 2012). Stations 9 and 11 were also ports, wherein fishing boats docked and paint boats resulted in a lot of fuel and paint spills in the water and sediment. This condition increased the concentration of Pb, because apart from being used for fuel mixtures, Pb is also used as a paint mixture (Järup, 2003).

The EF values of Cu and Cd at stations 1, 2, 3, 4, 7, 8, 9, 10, 11 and 12, of Sn at station 3 and 6; and Zn and Pb at station 7 indicated EF < 1 (Figure 2), i.e., no enrichment or natural occurrence (Mmolawa et al., 2011). Natural sources of metals Zn, Cu, Pb, Cd, and Sn come from weathering (Duruibe et al., 2007) and erosion of mineral rocks around the waters, for instance at station 1, 2, 3, and 4 (Kelabat Bay). There are granite rocks on Putri Island, so that granite can be eroded due to the physical factors of seawater. The erosion can also cause an increased concentration of heavy metals. Several studies have proven that Bangka Island is composed of granite rocks that spread along the coast (Bramastya et al., 2018). Heavy metal enrichment can also occur because of the deposition and aggregation of Sn from granite due to hydrothermal fluids with water (Su et al., 2021).
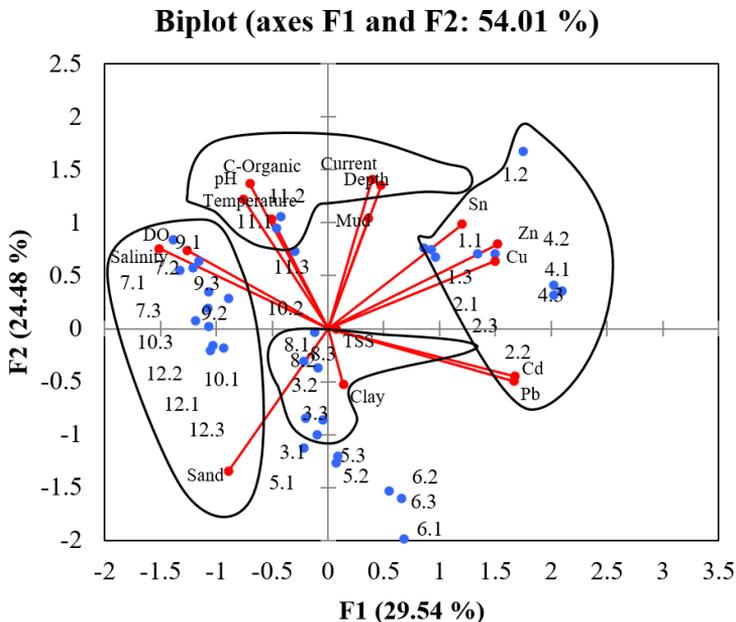

Figure 3. The results of the Principal Component Analysis (PCA) distribution of water characteristics and heavy metals based on research stations on axis 1 (F1) and axis 2 (F2)

The results of the analysis of the characteristics of the aquatic environment and heavy metals, were based on Principal Component Analysis (PCA). Stations 1, 2 and 3 are characterized by heavy metals Zn, Cu, Pb, Cd, Sn, while stations 7, 9, 10, 12 are characterized by salinity and DO values. Station 11 is characterized by high temperature, depth, current velocity, pH, and C-organic values. The characteristics of waters having large TSS are seen at stations 4 and 8 (Figure 3). This is presumably because the particles are dispersed due to the influence of spray from tin mining waste.

4. Conclusion

At stations 1, 2, 3, 4, 5, 6, 8, 9, 10 and 11, minimum to moderate enrichment was found for heavy metals Zn and Sn; while for the heavy metal Pb, moderate to very high enrichment was observed. This condition was caused more due to anthropogenic conditions, primarily because of the waste generated by human activities. Sand waste from tailings containing several Pb effluents, ship paint waste, fuel spills, and fuel fumes from tin-mining pontoon engines were known to contain heavy metal Pb from tetraethyl-Pb tetramethyl-Pb compounds. Pb in smoke binds to particles in the atmosphere, and then falls into the water when it rains. Furthermore, it binds with other compounds in the water to become flocculated, gaining weight and aggregating in the sediment. In addition to anthropogenic sources, heavy metals Zn, Cu, Cd, Sn at station 1, 2, 3, 4, 5, 6, 8, 9, 10, and 11 were also found to come from natural sources.

In contrast, there was no enrichment at station 12, and the five heavy metals present were from natural sources. This was influenced by the physical factor of seawater, which caused erosion of granite rocks on the coast of Bangka Island, thereby naturally increasing the heavy metal concentrations. The overall EF value of the heavy metals Zn, Cu, Pb, Cd, Sn at station 10 in Bangka Island waters showed that anthropogenic sources were more dominant and mixed with natural sources.

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