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Research Article

Spatial Distribution and Contamination Assessment of Lead (Pb) in the Seawater and Surface Sediments of the Coastal Area of Prigi Bay, Trenggalek, East Java

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Abstract

This study attempts to measure the spatial distribution of Pb in the seawater and surface sediments and to analyze its potential harmful effect in the surface sediments of the coastal areas of Prigi Bay, Trenggalek. Prigi Bay has been considered as one of the harbors with substantial activities from fisheries, tourism, and domestic events in Indonesia. These activities may discharge heavy metals, e.g., lead (Pb) into the environment and bring hazard to animal and human life. Data for Pb concentrations were collected from two stations near Prigi Harbor (Stations 1 and 2), one station in the Prigi Beach (Station 3) and two stations near the river mouths (Stations 4 and 5). Water quality parameters (temperature, salinity, pH, and DO) were also measured. Stations 1 and 4 displayed a higher concentration of Pb in the seawater than in the surface sediments. On the other hand, Stations 3 and 5 showed a higher concentration of Pb in the surface sediments than in the seawater. Pb concentration in the seawater was 0.22-0.60 mg/kg, and in the surface, sediments were in the range of 0.40-0.57 mg/kg. According to the analysis of contamination factor (CF), the study sites have a low degree of contamination from heavy metal Pb (CF < 1). The result was also supported by the potential toxicity of Pb to the benthic environment around Prigi Bay that was still considered low (E < 40). In spite of the low degree of contamination and low potential toxicity from Pb, concern regarding the heavy metal pollution in the study areas is still needed due to substantial activities that can contribute to the accumulation of heavy metal in a long time.

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1. Introduction

Lead (Pb) is one of the heavy metals, amongst many others that can be a threat to the marine environment because of their persistence, difficulty to degrade, and potential toxicity. It originates from both natural and anthropogenic sources. Once heavy metals enter the aquatic environment, it mixes with the water and is deposited in the sediments. Heavy metal in the sediment varies less in space and time; thus its distribution in the sediment provides useful data for monitoring (Hwang *et al*., 2016). Moreover, the study of heavy metals in water and deposits could be used to assess the impact of anthropogenic and industrial activities to the aquatic environment (Li *et al*., 2017; Ochieng *et al*., 2009; Syakti *et al*., 2015; Vu *et al*., 2017; Yona *et al*., 2016).

Pb is categorized as non-essential metal, which means its biological function has not been known and toxic even in low concentration (Duffus, 2002). Pb enters the aquatic environment mostly from anthropogenic activities such as traffic exhaust, paints, battery industry, and also oil pollution from ship activities (Esmaeilzadeh *et al*., 2016; Saher and Siddiqui, 2016). Environmental conditions such as salinity, pH, and redox potential control its solubility in the water and also the release from sediment to the seawater (Li *et al*., 2017; Ochieng *et al*., 2009). Moreover, the presence of the other metals in the environment could also influence Pb concentration. Song *et al*. (2017) discovered that the correlation between Mn and Pb as Mn might be a significant control on the abundance of Pb. The study also revealed that the relationship between Pb and As showed both metals had a similar source or enrichment mechanisms.

Contamination factor (CF) can be used to assess the degree of anthropogenic contamination of single metal in the marine environment (Ali *et al*., 2016). It measures the ratio of metal concentration in the sediment of the study area to the background value. CF is classified into four level of contamination: low degree ($CF < 1$), moderate level ($1 < CF < 3$), considerable ($3 \leq CF < 6$) and high degree ($CF \geq 6$). To evaluate its potential risk to the ecology (E) is measured by multiplying CF to the toxicity response coefficient (Hakanson, 1980). It is classified into five ecological risk potential: low ($E < 40$), moderate ($40 \leq E < 80$), higher ($80 \leq E < 160$), high ($160 \leq E < 320$) and serious ($E > 320$). There have been many studies of the used of CF and E in assessing the contamination of Pb in the environment (Ali *et al*., 2016; Bastami *et al*., 2014; Lin *et al*., 2013; Nethaji *et al*., 2017; Song *et al*., 2017; Sundararajan *et al*., 2017). In those studies, Pb has been found in moderate to high values of contamination and mostly due to the anthropogenic and industrial activities.

Bioavailability of heavy metal is known as the transfer of metals in the environment to the organisms. It depends on the physical, chemical, and biological factors (Riba *et al*., 2003). If accumulated metals in the organisms exceeds a certain threshold, it could lead to toxicity (Appenroth, 2010). Heavy metals have been reported to affect cellular organelles and interact with cell components such as DNA causing DNA damage (Tchounwou *et al*., 2012).

Prigi Bay is busy with the fisheries, domestic, and tourism activities. Prigi harbor, located in the Prigi Bay is one of the busiest ports in Indonesia connected to the Indian Ocean. It is home to many fishing vessels with high fisheries activities and industries (Farikin *et al*., 2015; Nikmah *et al*., 2018). According to Farikin *et al*. (2015), the number of fishing vessels in the harbor everchanging, depending on the class of tonnage. Prigi Bay is also one of the tourist destinations because of its calm and pretty beach. It attracts many visitors all year round. These busy activities in Prigi Bay may contribute to environmental pollution, including metal pollution. Therefore, this study was conducted to assess the distribution and ecological risk potential of metal, especially Pb in the water and surface sediments in the Prigi Bay.

2. Materials and Methods

2.1 *Study area and samples collection*

Data were collected on May 2016 from five sampling sites of the coastal areas of Prigi Bay (Figure 1): east side of Prigi Harbor (Station 1), west side of Prigi Harbor (Station 2), Prigi Beach (Station 3) and two stations near the river mouths (Stations 4 and 5). Surface seawater samples for Pb analysis were collected by immersing 500 ml polyethylene bottle, acidifying it with 10 % HNO3, and was placed in an icebox for further study in the laboratory. Sediment samples were obtained from the top 5 cm of the surface using Ekman grab. The sediment collected was the portion that did not come into contact with the sample walls to avoid metal contamination and placed in polyethylene bags, preserved in icebox before laboratory analysis. Seawater was used to wash the grab between sampling sites. All sampling materials were pre-washed with nitric acid and distilled water before sampling. Temperature, salinity, pH, and DO were measured using thermometer, refractometer, pH meter, and DO meter (Hanna Instrument Ltd), respectively. For each station, seawater and surface sediment samples for Pb analysis and the water quality parameters were taken in duplicate.

2.2 *Sample analysis*

In the laboratory, seawater samples were filtered through 0.45 mm Whatman glass filter paper and acidified to reduce the pH < 2 with concentrated HNO_3. About 50 ml of well-mixed, acidified samples were added with another 5 ml concentrated HNO_3 and boiled at 130°C on the hot plate. The process was repeated until the solution becomes light color or clear (Ali *et al*., 2016).

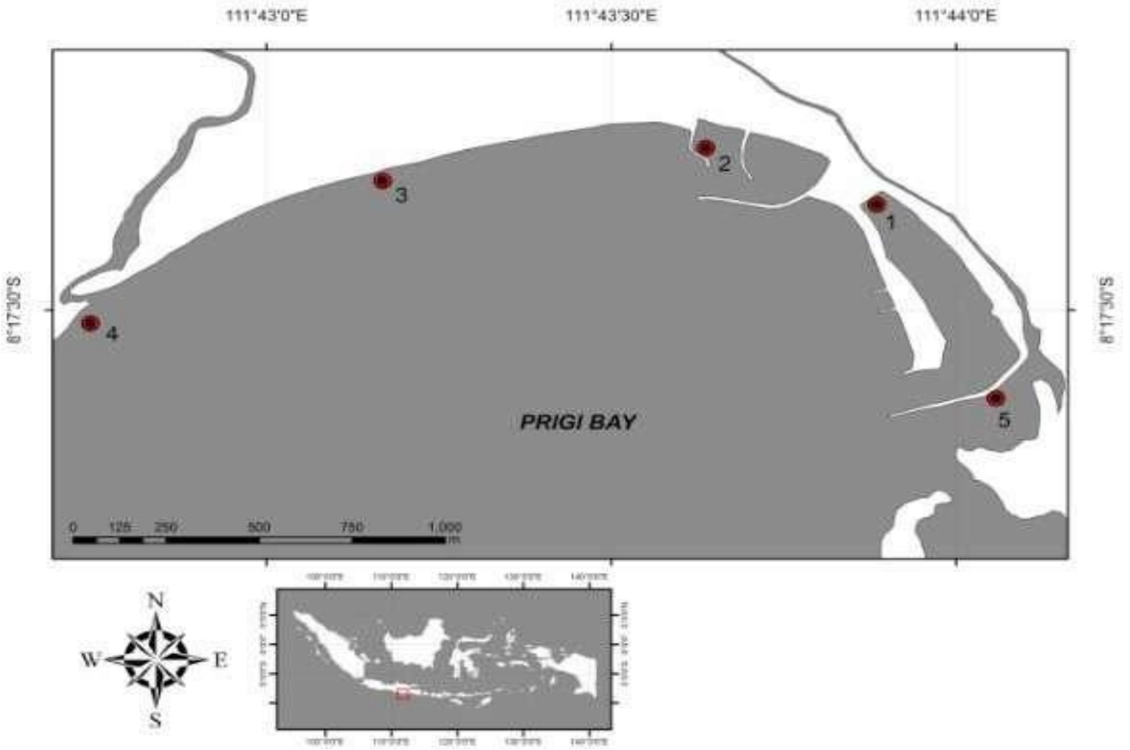

Figure 1. The location of sampling sites at Prigi Harbor, Trenggalek. Numbers show the sampling stations: 1 and 2 are the east and west side of the Prigi Harbor stations, 3. Prigi Beach station, 4 and 5 are near river mouth stations

For sediment samples, after oven drying, the sediment samples were ground using hand mortar and screened with 0.5 mm sieve to remove large particles. About 1 g of sediment sample was digested using a mixed solution (1:3 HNO_3:HCl) for three h, cooled, filtered through Whatman – 42, diluted and adjusted to volume of 50 ml using distilled water (Bastami *et al*., 2014). Seawater and sediment samples were then measured by a Shimadzu Flame Atomic Absorption Spectrophotometer (Model AA-6800). Blank and standard samples were used to monitor the performance of the instrument by developing calibration curves.

2.3 Lead contamination indexes and statistical analysis

Sediment has been studied as the representative of the source of metal contamination to the aquatic environment (Guimarães *et al*., 2011). Contamination factor (CF) was calculated using background value from the abundance of elements in crustal rocks for Pb that is 13 mg kg^{-1} (Greenwood and Earnshaw, 1997), while the toxic biological response (Tr) of Pb to calculate the potential risk index for the ecology (E) was retrieved from Hakanson (1980) with the value of five. SPSS (Version 16.0) was used to analyze the data statistically. One-way ANOVA was used to compare Pb concentrations among the sampling stations. Pearson`s correlation coefficients were used to identify correlations among water parameters and Pb concentrations. Independence student`s t-test was performed to compare the level of Pb between seawater and sediments. Normality data was checked using the Shapiro-Wilk test, and the data were normally distributed ($p = 0.55$ and $p = 0.92$ for Pb concentration in the seawater and sediments, respectively).

3. Results and Discussion

3.1 Water Quality Parameters

Water quality parameters are presented in Table 1, and it shows among the study sites the values were almost in a similar range for all the physicochemical parameters. Lower salinity was found at Stations 4 and 5,

Table 1. Water quality parameters (average ± SD) in the Prigi Harbor, Trenggalek

Station	Temperature (°C)	Salinity	pH	DO (mg L-1)
1	32.74 ± 0.16	31.17 ± 0.31	6.06 ± 0.21	6.15 ± 0.07
2	33.07 ± 0.31	31.20 ± 0.05	6.20 ± 0.14	6.50 ± 0.14
3	30.55 ± 0.77	31.90 ± 0.05	7.90 ± 0.28	8.10 ± 0.28
4	31.62 ± 0.73	26.47 ± 1.40	6.40 ± 0.42	7.22 ± 0.02
5	29.3 ± 0.28	26.38 ± 2.70	8.40 ± 0.28	8.50 ± 0.14

Table 2. Pearson`s correlation of Pb in the seawater, sediments and water parameters in the Prigi Harbor, Trenggalek (n=10)

	Pb in seawater	Pb in sediment	Temperature	Salinity	pH	DO
Pb in seawater	1					
Pb in sediment	-0.34	1				
Temperature	0.88*	-0.26	1			
Salinity	0.41	-0.11	0.48	1		
pH	-0.90*	0.21	-0.95*	-0.25	1	
DO	-0.94*	0.15	-0.91*	-0.46	0.92*	1

Table 3. Comparison of Pb concentration in water and sediments with local and international studies in the world

Location	Pb concentration		References
	Seawater	Sediments	
Prigi Harbor, Trenggalek (Indonesia)	0.22-0.60 mg/kg	0.17-0.57 mg/kg	This study
Lekok coastal areas, Pasuruan (Indonesia)	0.20-0.28 ppm	0.25-0.42 ppm	(Yona *et al*., 2016)
Perairan Segara Anakan, Cilacap (Indonesia)	0.12-0.24 mg/L	3.96-21.99 mg/kg	(Hidayati *et al*., 2016)
Houjing River (Taiwan)	0.569 mg/L	15.6-258 mg/kg	(Vu *et al*., 2017)
Yalujiang estuary (China)	0.40-1.80 μg/L	12.3-29.9 mg/kg	(Li *et al*., 2017)
Northern Liadong Bay (China)	3.98 μg/L	18.77 mg/kg	(Zhang *et al*., 2017)
Karnaphuli River (Bangladesh)	5.29-27.45 mg/kg	21.98-73.42 mg/kg	(Ali *et al*., 2016)
Indian Ocean coast (Kenya)	7.0-68 μg/L	4.09-27.29 μg/g	(Ochieng *et al*., 2009)

Table 4. Contamination factor (CF) and ecological risk factor (E) of Pb (average ± SD) in the sediments of Prigi Harbor, Trenggalek

Station	Ecological risk assessment			
	CF	Grade of contamination	E	Grade of risk
1	0.04 ± 0.01	Low	0.19 ± 0.06	Low
2	0.04 ± 0.02	Low	0.19 ± 0.10	Low
3	0.04 ± 0.00	Low	0.19 ± 0.01	Low
4	0.03 ± 0.00	Low	0.16 ± 0.01	Low
5	0.04 ± 0.00	Low	0.22 ± 0.02	Low

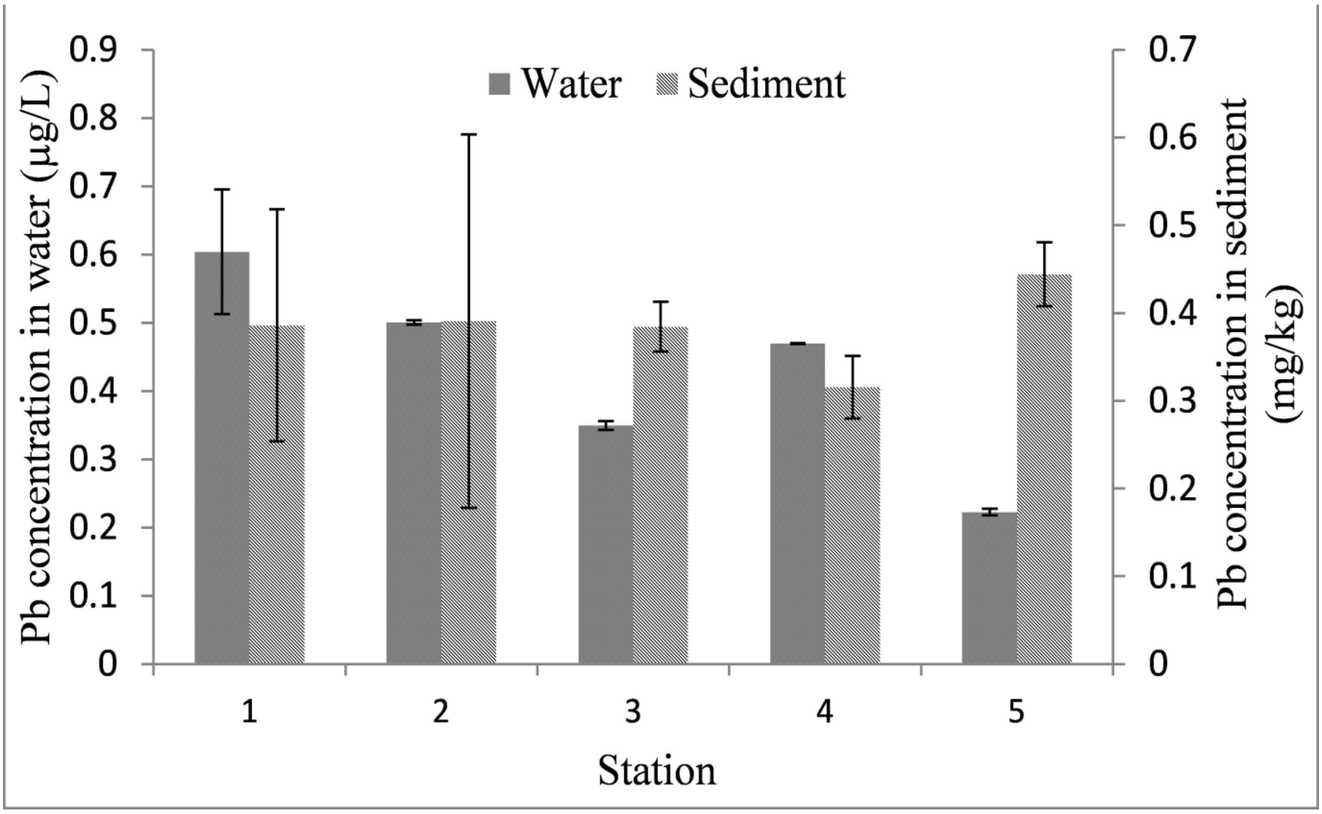

Figure 2. Pb concentration in seawater and sediments (mean ± SD) in the Prigi Harbor, Trenggalek

and it is the result of the intrusion of freshwater from the adjacent rivers. On the other hand, lower DO value at Stations 1 and 2 compared to the other stations could be due to higher temperatures during the sampling period. The solubility of dissolved oxygen in the water decrease with increasing the temperature (Ali *et al.*, 2016), and according to Pearson`s correlation test (Table 2), there was a significantly negative correlation between temperature and DO (r = -0.91, p = 0.01). pH showed significant difference among the sampling stations from the acid condition at Stations 1, 2, and 4 to alkaline condition at Stations 3 and 5. Lower pH values were the result of higher fisheries and domestic activities in those stations. Similar to DO, there was significantly negative correlation between temperature and pH (r = -0.95, p = 0.01). The high temperature would increase metabolic activities of the organisms that contributed to the increase of CO_2 resulted in lowering pH value. Overall, physicochemical parameters of the study sites are within the permissible limit of regulations regarding water quality for harbor and marine organisms set by the Indonesian Ministry of Environment, 2004.

3.2 Pb Concentration in Seawater and Sediments

Pb concentrations in the seawater were found in the range of 0.22-0.60 mg/L, and the level in the surface sediments was in the range of 0.17-0.57 mg/kg (Figure 2). Compared to the other studies (Table 3), the result of this study was in the similar range with the one in the Lekok coastal areas, Pasuruan (Yona *et al.*, 2016) for both Pb concentration in the seawater and surface sediments, but higher compared to the study in the Indian Ocean coast (Ochieng *et al.*, 2009). Pb concentration in the surface sediments of this study was observed very much lower compared to the study conducted in the Segara Anakan waters (Hidayati *et al.*, 2016), Houjing River (Vu *et al.*, 2017), northern Liadong Bay (Zhang *et al.*, 2017) and Yalujiang estuary (Li *et al.*, 2017). Moreover, the result of this study found Pb concentration was very much lower in both seawater and surface sediments compared to the study in the Karnaphuli River (Ali *et al.*, 2016).

Pb concentrations in the seawater and surface sediments were found to be similar. The results were supported with Student`s Independent t-test that there is no significant difference between Pb concentration in the water and the surface sediment (p = 0.291). One-way Anova test revealed that Pb concentrations in both seawater and surface sediments ($p < 0.01$) were significantly different among the stations. According to Pearson`s correlation (Table 2), there is a negative but not significant correlation between Pb concentration in the water and the surface sediments (r = - 0.34). Although statistically, the relationship between Pb in the seawater and the surface sediments was weak, the interaction of metal concentration might still occur because there is a continuous inflow of metals from seawater to the sediments (Wojtkowska *et al*., 2016).

Among the water quality parameters, only temperature that was found to have significant positive correlation with Pb concentration in the seawater ($r = 0.88$, $p < 0.01$) ,explaining the lowest concentration of Pb at Station 5 (Figure 2) that was related with the lowest temperature (Table 1) among the sampling stations. The temperature might influence heavy metal concentration related to the precipitation process that possibly contributes to the input of heavy metal to the environment (Zhang *et al*., 2017). Moreover, high temperature has been observed to cause the higher metal release from the sediment to the overlying water (Li *et al*., 2013). Unlike temperature, pH and DO displayed strong negative correlation with Pb concentration in the seawater ($r = -0.90$ and $r = -0.94$ for pH and DO, respectively, $p < 0.01$). The higher concentrations of Pb in the seawater occurred in Stations 1 and 2 when these stations experienced lower pH and DO values. Yona *et al*. (2018), in their study of heavy metal distributions on the southern coast of Pacitan, found the distribution of metals is related to the distribution of pH and DO. Moreover, Atkinson *et al*. (2007), in their experimental study of pH and DO on metal release from marine sediments, stated that during low pH experiments, Pb concentration was higher in the water due to the release of metal from the sediment to the overlying water. Similar to that, low DO and disturbed sediments will release Pb in the sediments to the water. It is related to the presence of organic matter in the aerobic condition that can binding heavy metals (Akan *et al*., 2013; Li *et al*., 2013).

Among the sampling sites, Pb concentrations at Stations 1 and 4 were found slightly higher in the seawater compared to the concentration in the surface sediments. On the other hand, at Stations 3 and 5, Pb concentrations were notably higher in the surface sediments rather than in the seawater. Unlike the other stations, Pb concentrations in the seawater and surface sediments of Station 2 were almost in a similar value. Heavy metal concentration in the seawater highly related to the terrestrial input such as riverine, domestic, and industrial inputs (Zhang *et al*., 2017). Stations 4 and 5 were the sampling sites that were closest to the river mouth, however Station 5 contained the lowest concentration of Pb in the seawater and the highest concentration in the sediment. Data sampling at Station 5 was conducted during low tide with low water flow causing lower level in the seawater and high accumulation of the metal in sediment (Ali *et al*., 2016). Station 1 was observed to have the highest Pb concentration in seawater, and this might be the result of high amount of activities in the harbor such as boat painting, gasoline emitted from the boat, and also the extensive discharge of untreated effluents from the fish market.

3.3 Ecological Risk Assessment for Pb in the Sediments

The calculation of the contamination factor revealed that all sampling stations of the study area have a low degree of contamination from heavy metal Pb (Table 4). The higher value of background level compared to the actual Pb concentration in the samples resulted in the low contamination of Pb in the study area. Similar results with low concentration of the measured Pb compared to the background value were also observed in the study along southeast coast of Caspian Sea (Bastami *et al*., 2014), Danjiang Harbor, Tianjin (Guo *et al*., 2010), Pulau Morotai waters, Maluku Utara (Edward, 2015) and some parts of the observed areas in the Segara Anakan waters, Cilacap (Hidayati *et al*., 2016). On the other hand, moderate degree contamination of Pb ($1 < CF < 3$) was observed in another study such as the study along the coastal sediment of Pakistan (Saher and Siddiqui, 2016), Karnaphuli River, Bangladesh (Ali *et al*., 2016) and Segara Anakan waters, Cilacap (Hidayati *et al*., 2016). These studies found Pb concentration in the sediment in the range of 32-35 µg/g, 56-67 mg/kg, and 19-21 mg/kg, respectively, which is much higher compared to the concentration found in this study which is 0.17-0.57 mg/kg.

The ecological risk factor of Pb in the study area exhibited a low grade of risk to the ecosystem (Table 4). The results were similar to the effects of most studies about ecological risk factors for Pb (Bastami *et al*., 2014; Guo *et al*., 2010; Saher and Siddiqui, 2016). Although present in the same category with those studies, Pb concentrations in this study were found very much lower compared to the one found along southeast coast of the Caspian Sea (Bastami *et al*., 2014) and the coastal sediment of Pakistan (Saher and Siddiqui, 2016) with the concentration of 9-16 ppm in winter and 4.20-9.10 ppm in summer and 32-35 µg/g, respectively.

4. Conclusion

In this study, we evaluated Pb distribution in the seawater and surface sediments around the coastal areas of Prigi Bay, Trenggalek, and the results revealed that there was no significant difference of Pb concentration between seawater and surface sediments. Some study sites showed higher level in the seawater, and some others showed higher level in the sediments, but but the difference was not significant. Water quality parameters such as temperature, pH, and DO displayed a significant correlation with Pb concentration in the seawater only. Pb concentrations found in this study were considered low, and it resulted in a low grade of contamination and low grade of risk according to the Contamination Factor (CF < 1) and Ecological Risk Index (E < 40). Although the results showed a low degree of contamination and low ecological risk, Pb pollution in the study area could not be ignored due to substantial activities that can contribute to the accumulation of the metal in the sediments. Monitoring is needed to observe the increasing concentration of Pb in the coastal areas of Prigi Bay, Trenggalek. Moreover, to assess in more detail the impact of Pb to the ecosystem, an ecological risk assessment should involve the interaction of Pb with the organisms, not only from the surface sediment samples.

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Author`s Contributions

DY designed the sampling process and drafted the manuscript; DV designed the sampling process and collected the data; RDK and SHJS analized the data and drafted the manuscript. All authors finalized the manuscript.

Conflict of Interest

The authors declare that they have no competing interests.

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