The distribution of pyrite (FeS_2) in brackish water ponds and its mitigation efforts in the Mahakam Delta, Indonesia

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Abstract. This study aims to analyze the distribution of pyrite in water and soil of brackish water ponds in the Mahakam delta and its mitigation efforts. Water and soil sampling were carried out on several brackish water ponds that have different salinity, i.e. salinity of 0–10 ppt, 11–20 ppt, and 21–30 ppt. Each type of salinity was represented by 10 plots of brackish water ponds. The measured parameters were the content of pyrite in water and soil, salinity, and sulfate (SO_4) in the embankment and the bottom of brackish water ponds. The specific focus in this study was the mitigation efforts by observing the absorption of iron (Fe) content by clams. It was carried out by using the Randomized Block Design (RBD) applied in the laboratory and the field. The analysis for the distribution of pyrite by digitizing, georeferencing, and editing was completed using QGIS v. 3.4 software. The employed data analysis was descriptive statistics that were presented in a box plot. The Anova and Manova methods were used to determine the results of the absorption test of iron (Fe) content at the level of confidence of $p<0.05$. The results showed that the distribution of pyrite in brackish water ponds is influenced by watersheds and distance from the sea. Furthermore, pyrite is indicated to have a strong relationship with sulfate (SO_4). Mangrove clams (*Polymesoda erosa*) can absorb iron (Fe) content in the soil of brackish water ponds and have potential as biomitigation. The class size of 61-80.99 mm has a significant effect ($p<0.05$) on the absorption of iron (Fe) content.

Key Words: brackish water pond, Mahakam delta, mitigation, pyrite, *Polymesoda erosa*.

Introduction. According to Zwieten et al (2006), Sidik (2010) and Bosma et al (2012), there is a massive change in mangrove ecosystems mainly due to the conversion of mangrove forests into brackish water ponds. Brackish water ponds in the Mahakam delta generally apply the traditional brackish water ponds system by implementing the polyculture system (Susilo et al 2019). The productivity of brackish water ponds in the Mahakam delta is below the standard of the productivity of traditional brackish water pond (nationally), which is 600-1000 kg/ha, while in 2008 it was only 14.92 kg/ha (Hartono & Almadi 2008). One of the causes of the low productivity of brackish water ponds in the Mahakam delta is high and acidic pyrite content, that kills the biotas breeding in the brackish water ponds. Pyrite content (FeS_2) in brackish water ponds in the Mahakam delta has a high tendency. This is indicated by a study conducted by the Maritime and Fisheries Office of Kutai Kartanegara (Ind. Dinas Kelautan dan Perikanan (DKP) Kutai Kartanegara) in 2014 in which pyrite content in the brackish water ponds of the Mahakam delta ranged from 1,125-1,834 ppm or 1.125–1.834 mg/kg. Furthermore, generally, pyrite content is higher in brackish water ponds compared to coastal waters. Almadi (2014) stated that pyrite in the soil of brackish water ponds is 6.15%±0.36, meaning that it is in the low content category. According to Noor (2004), the average pyrite concentration in the soil of brackish water ponds, during a study that he conducted, was also in the low content category, because the average of pyrite concentration was >5.6%.

The high pyrite content in the brackish water ponds of the Mahakam delta is determined by the type of soil in the ponds, which is classified as Acid Sulfate Soils. The oxidized Acid Sulfate Soils will cause the substrate of the brackish water pond to become toxic. Besides having high pyrite (FeS_2) content, brackish water ponds in the Mahakam delta are generally built in locations that have different salinity concentrations.

High pyrite content in the brackish water ponds of the Mahakam delta is one of the causes of the low productivity of brackish water ponds in the region. The main cultivated species in the region are tiger prawns (*Penaeus monodon*), spotted prawns (*Metapenaeus affinis*), and milkfish (*Chanos chanos*). With high pyrite content in the soil at the bottom of the brackish water pond, it makes the cultivated species susceptible to disease. Meanwhile, pyrite content also becomes toxic for cultivated biota life.

Pyrite content is an important factor influencing the productivity in the Mahakam delta. To overcome the problem of high pyrite concentration in the Mahakam delta, there is a need for mitigation efforts to be carried out, i.e. by using mangrove clams (*Polymesoda erosa*). Clams can absorb heavy metals such as Fe (iron). The reduction in Fe content is expected to reduce the pyrite formation process naturally. Clams are the best bioaccumulators for metals, compared to other organisms (Darmono 1995).

Research related to shellfish or aquatic biota as a biofilter to reduce heavy metals has been carried out by Tiwari et al (2006) and Achmadi et al (2019). It is expected that the reduction in pyrite can increase the productivity of brackish water ponds. This study aims to analyze the distribution of pyrite in the water and soil of brackish water ponds in the Mahakam delta and its mitigation efforts.

Material and Method

Description of the research location. This study was conducted in the local farmers brackish water ponds in the Mahakam delta area, Kutai Kartanegara, East Kalimantan (Figure 1). Sampling was carried out three times with intervals once every 4 months for 1 year (March 2017 until March 2018). The brackish water ponds were selected based on differences in salinity, i.e. salinity of 0–10o/oo, 11–20o/oo, and 21–30o/oo. According to Sidik et al (2014), brackish water ponds with a size of 5-10 ha have better productivity compared to other brackish water ponds with different sizes. The number of brackish water ponds available in the area of the research was ±30 plots. Therefore, samples were colected for 30 plots. They were then divided based on the level of salinity. Each level of salinity was represented by 10 plots. The measured parameters were the content of pyrite in water and soil of brackish water ponds. Water sampling was conducted on the surface and bottom of the pond, while soil sampling was done from the embankments and the bottom of the brackish water ponds. In addition, researchers also carried out salinity measurements in each brackish water pond.

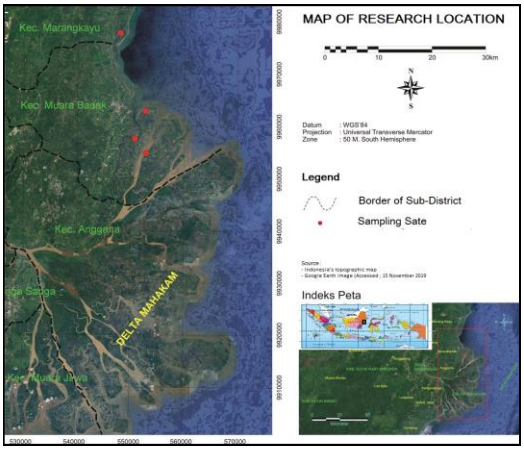

Figure 1. Location of the research in the Mahakam Delta, East Kalimantan (the red dots show the brackish water ponds sampling locations).

The specific focus of this study was to evaluate how efficient are clams for the mitigation effort (biomitigation) of pyrite, by observing the absorption rate of Fe by clams. It was carried out by using the Randomized Block Design (RBD) (Table 1), applied in the laboratory and the field. After we tested the absorption rate in the laboratory, we further tested in the field the absorption rate of clams. We tested for 1 month in the laboratory and in the field. 30 samples from the substrate of the brackish water ponds were taken and then brought to the laboratory. The soil was put into an aquarium with a size of 40 cm x 40 cm and a height of 10 cm from the bottom of the aquarium. We then added 5 cm of water from the brackish water ponds. After that, clams were added to the aquarium. Before the clams were placed into the trial sites in the laboratory and in the field, researchers carried out an initial measurement of the Fe content in the clams. Furthermore, at the end of the trial, researchers conducted a measurement again to see the accumulation of Fe in the clam body. In the applied randomized block design, clams were classified into 3 classes based on their size, namely 40–60.99 mm class (A1), 61–80.99 mm class (A2), and 81–100 mm class (A3). The results of the final measurement in each class were subtracted by the results of the initial measurement in each class by using the following formula:

$$Cf=Ct-Co$$

where Cf is the concentration of Fe (mg), Ct is the concentration of Fe (mg) in clams at the end of the trial and Co is the concentration of Fe (mg) in clams at the beginning of the trial.

Table 1

The randomized block design applied in the laboratory and in the field

Treatment : B Clams	*Class Size : A*		
	A1 (KU 1) 40 – 60.99 mm	A2 (KU 2) 61 – 80.99 mm	A3 (KU 3) 81 – 100 mm
(B) Soils of the Pond	A1B1	A2B1	A3B1

Data analysis. The analysis for the distribution of pyrite by digitizing, georeferencing, and editing was completed by using QGIS v. 3.4 software. The employed data analysis was descriptive statistics that were presented in a box plot. The Anova and Manova tests were used to determine the trial results for clams being used for pyrite mitigation. If the results of Anova and Manova indicate real differences, it will be continued with the Tukey's test at a confidence level of $p<0.05$ using the SPSS program.

Results and Discussion

Distribution of pyrite. Brackish water ponds in the Mahakam delta are built on sediment areas. Therefore, the shape and size of the ponds are not the same as each other. The area of plots in the Mahakam delta ranges from 2-100 ha. Different salinity also affects the productivity and management methods of ponds in this region.

Studies related to brackish water ponds in the Mahakam delta have been conducted by several researchers. For example, Almadi et al (2008) examined the productivity of silvofishery in brackish water ponds using polyculture systems in the Mahakam delta. Rizal (2012) studied the cultivation of *Polymesoda erosa* (Solander 1786) in brackish water ponds with mangroves and without mangroves in the Mahakam delta. Almadi et al (2013) examined the presence of oxygen in silvofishery brackish water ponds in the Mahakam delta. Asikin (2013) conducted a study on the pattern and strategy for post-harvest management of giant tiger prawns (*Penaeus monodon*) in brackish water ponds in the Mahakam delta. Nursigit (2013) conducted a study on the analysis of the compatibility of forestry cultivation area zoning in the Mahakam delta, East Kalimantan. Bosma et al (2010) studied mangrove ecosystems and their impact on sustainable livelihood.

The distribution of pyrite in the water and soil of the brackish water ponds had different distribution patterns (Figure 2). The results of this study indicated that pyrite in the soil at the bottom of the brackish water pond was higher than the soil in the embankment of the brackish water pond. It is possible because the pyrite in the soil at the bottom of the brackish water pond did not undergo oxidation. The distribution of pyrite in the water of the brackish water ponds depended on the condition of the distribution of salinity in the ponds.

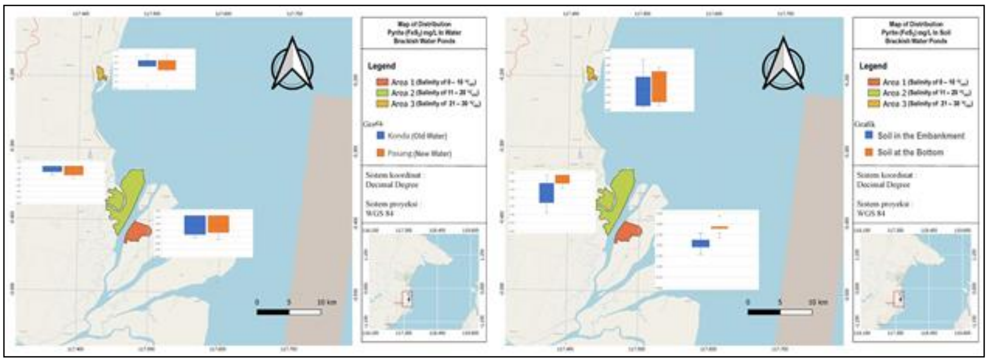

Figure 2. Distribution of pyrite in water and soil of brackish water ponds.

Studies on salinity problems in acid sulfate soils in coastal areas was carried out by Fanning (1993). Hadikusumah and Sumanjuntak (2011) examined the box model of freshwater, salinity, and soil nutrients in the Mahakam delta in which they specifically studied the transport mechanism of freshwater mass, salinity, and soil nutrients in the riverbank of the Mahakam River that interacts directly with seawater. The vertical distribution of salinity in the Mahakam delta was found to be highly stratified. Phamvan et al (2012) observed the modeling of the salinity distribution and water movement time in the Mahakam delta. The results of the simulation indicated that salinity at the forefront of the Mahakam delta directly adjacent to the Makassar Strait is 35 PSU (Practical Salinity Unit). Furthermore, salinity in the middle of the delta was less than 20 PSU. Meanwhile, 15 km upstream, it significantly decreased to 5 PSU due to the flow of the Mahakam River. Hidayanto et al (2004) and Atjo (2005) conducted similar studies in which the results indicated that aquaculture management had a direct effect on the performance of cultivation and the quality of the physical-chemical environment. In their studies, they also found out that the use of iron-rich soils has the potentials to control hydrogen sulfide concentrations in aquaculture systems. Under anaerobic conditions, sulfide removal was conducted by applying redox reactions and then creating iron sulfide (Fe_2S) (Lahav et al 2003).

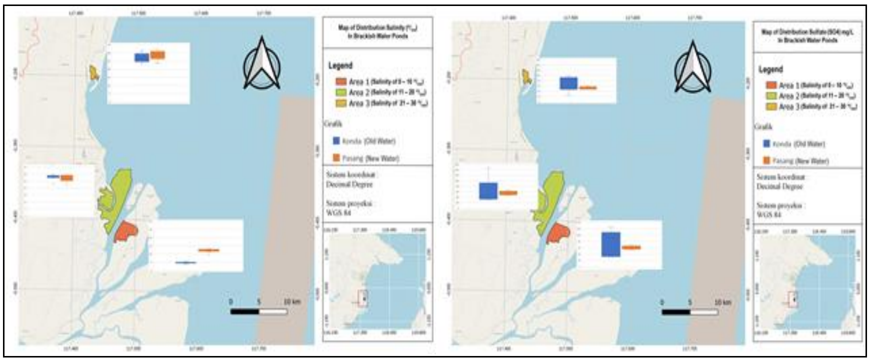

Figure 3. The relationship between salinity and SO_4 in the Mahakam delta.

Figure 3 shows that there is a strong relationship between the distribution patterns of salinity and SO_4. This is caused by the previous types of mangroves that were present in the brackish water ponds. One type is the mangrove palm (*Nypa fruticans*) which generally grows in low salinity. This type of mangrove is easier to deforest than true mangrove species belonging to the genera of *Rhizophora*, *Avicennia*, *Brugeria* etc. This also indicates the ability of farmers in deforesting for making brackish water ponds in the Mahakam delta. In addition, to make a brackish water pond in a true mangrove species area, it requires a substantial cost in terms of renting equipment. Based on a study conducted by Golez and Kyumah (1997), it indicated that pyrite oxidation causes a reduction in some nutrients such as calcium (Ca), magnesium (Mg), zinc (Zn), and copper (Cu).

Table 2

Correlation between water and soil parameters

	Pyrite (P)	*Pyrite (D)*	*SO_4 (P)*	*SO_4 (D)*	*Ca (P)*	*Ca (D)*	*Pyrite (water) Old Water*	*Pyrite (water) New Water*	*Salinity (water) Old Water*	*Salinity (water) New Water*	*SO_4 (water) Old Water*	*SO_4 (water) New Water*	*Ca (water) Old Water*	*Ca (water) New Water*
Pyrite (P)	1													
Pyrite (D)	0.360	1.000												
SO_4 (P)	***1.000***	0.360	1.000											
SO_4 (D)	***0.540***	*0.475*	***0.540***	1.000										
Ca (P)	-0.115	-0.047	-0.115	-0.567	1.000									
Ca (D)	-0.301	-0.417	-0.301	-0.332	-0.018	1.000								
Pyrite (water) Old water	0.167	0.373	0.167	0.253	-0.094	0.100	1.000							
Pyrite (water) New Water	0.138	0.305	0.138	0.240	-0.065	0.109	*0.757*	1.000						
Salinity (water) Old water	-0.202	-0.154	-0.202	-0.196	0.276	0.062	0.008	-0.036	1.000					
Salinity (water) New Water	0.025	0.151	0.025	0.157	-0.073	-0.029	0.207	0.208	-0.244	1.000				
SO_4 (water) Old water	-0.034	0.277	-0.034	0.023	-0.161	-0.120	0.186	0.058	-0.229	0.300	1.000			
SO_4 (water) New Water	0.285	0.102	0.285	0.255	-0.272	-0.022	0.301	0.347	-0.544	0.389	0.068	1.000		
Ca (water) Old water	-0.178	-0.126	-0.178	-0.231	0.339	0.094	-0.040	-0.074	***0.972***	-0.248	-0.212	-0.582	1.000	
Ca (water) New Water	0.301	-0.363	0.301	0.061	0.037	-0.031	0.000	-0.036	0.081	-0.035	-0.193	0.026	0.133	1.000

The results of water and soil measurements at P (embankment), D (pond bottom), Old Water and New Water are found in the correlation results (Table 2), which indicated that there was a strong relationship (r=1000) between pyrite and SO_4 on the embankment of the brackish water ponds. Furthermore, it also was found that there was a strong relationship between pyrite on the embankment of the ponds and pyrite on the bottom of the pond that was covered by water (r=0.540). As mentioned by Tood (1980), the salinity of the water that is stored for a long time in a brackish water pond provides a strong relationship with the presence of Ca in the water. According to Priatmadi and Haris (2008), the process of the pyrite formation is found in tidal swamp soils as in brackish water ponds. From a study conducted by Ritvo et al (2004), soaked microcosm can be used in controlling sulfate soils. A study conducted by Gosavi et al (2004) indicated that biomonitoring of algae can be applied to find out metal contamination in acid sulfate ponds. Furthermore, in a study conducted by Avnimelech and Ritvo (2003), it found out that there is a correlation between redox components such as sulfides, ammonium concentrations, or soil oxygen requirements. Lifting sediments at the bottom of the brackish water ponds can provide changes to aquaculture activities (Yuvanatemiya & Boyd 2006). Brackish water ponds that use acid sulfate ponds are recommended to not be drained so that pyrite is not formed. Acidification is influenced by the solubility of aluminum (Al), iron (Fe), potassium (K), and sulfur (S) (Golez 1995).

Mitigation of pyrite compounds. In this study, efforts to mitigate biological pyrite compounds were conducted by using mangrove clams (*Polymesoda erosa*). In this case, the absorption of Fe content (an agent for the formation of pyrite compounds) is applied as biological mitigation.

According to Tood (1980), the acidic state is related to the number of Fe content so that the reduction of Fe content through the absorption by clams as biological agents can decrease the acidity of brackish water ponds. Clams as filter feeders and deposit feeders are highly effective in reducing the number of pyrite compounds by absorbing Fe content which is the basic element for the formation of these compounds. Some of the factors that influence the rate of absorption of Fe content in sediments and water are salt content, the presence of other chemical compounds, temperature, pH, size of the organism, and starvation conditions of the organism. The tolerance rate of the clams to Fe content does not depend on the rate of absorption into their body. Physiological conditions are also very influential because optimal physiological conditions can cause an increase in the absorption rate. Oxidative soils of brackish water ponds will increase the overhaul and mineralization of organic matter, whereas reductive soils of brackish water ponds will inhibit the overhaul and mineralization of organic matter.

The increase in Fe concentration is influenced by soil organic matter content, soil nature, Fe content, pH, and soil temperature. The element Fe_{2+} has the potential to form pyrite compounds so that the potential redox of the soil of brackish water ponds can decrease dramatically. The alternative method to reduce Fe content by using clams is significant as an effort to reduce the accumulation of pyrite compounds.

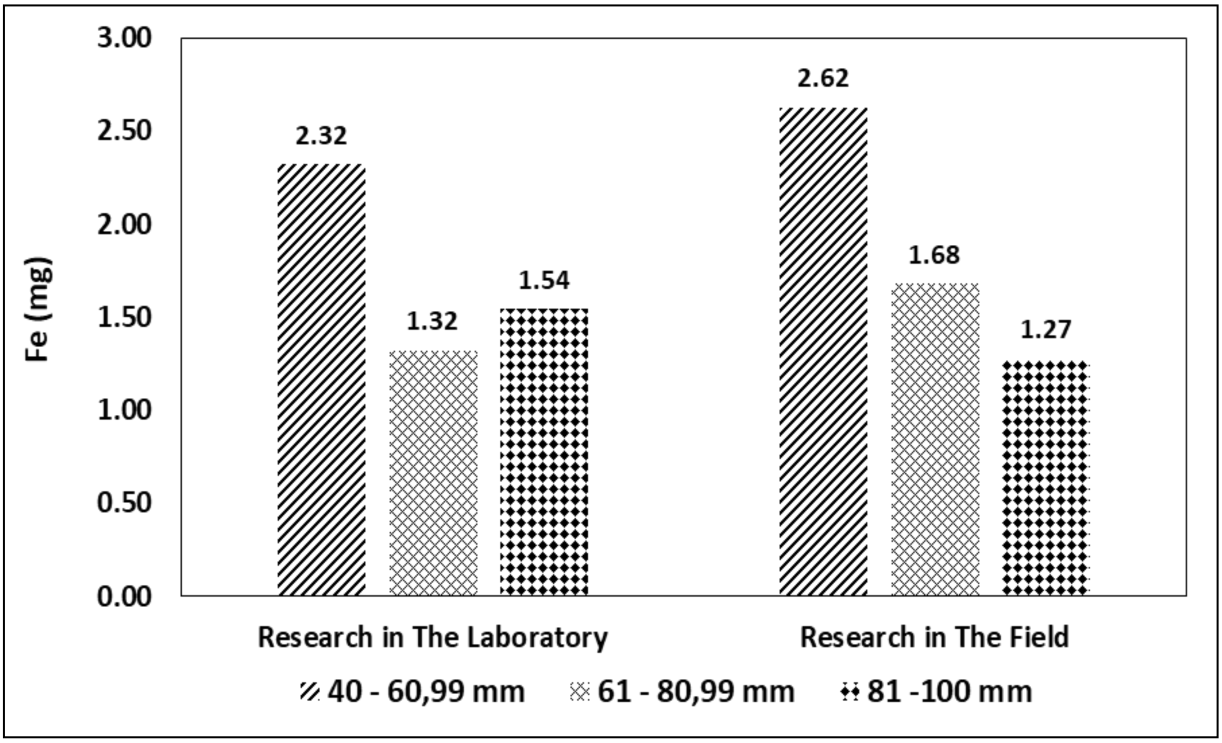

Figure 4. Absorption rate of Fe content by mangrove clams (*P. erosa*).

Based on the results of trials of mangrove clam (*P. erosa*) in the laboratory and the field, it indicated that the metabolic pattern and specific habitat of these clams can cause a difference in the level of absorption of Fe content quantitatively. The level of sensitivity of clams to Fe content can affect the proportion of absorption. The results of the analysis indicated that mangrove clams (*P. erosa*) can absorb Fe content (Figure 4). The absorption of Fe content can reduce the occurrence of the process of compounding FeS_2. The size of mangrove clams (*P. erosa*) with 40-60.99 mm had a higher average absorption of Fe content compared to other classes of clam sizes. The class size of 61-80.99 mm had a significant effect ($p<0.05$) on the absorption of Fe content. This indicated that mangrove clams can be used as biomitigators to overcome the high pyrite compound in the soil of brackish water ponds, the results tested for 1 month mud clam reduce the Fe content in a study conducted by Setyono (2002), it found out that clams can absorb Fe content which is an element for pyrite formation. The pyrite compound (FeS_2) is significantly different in each condition of brackish water ponds.

Conclusions. The distribution of pyrite in brackish water ponds is influenced by watersheds and distance from the sea. Furthermore, pyrite is indicated to have a strong relationship with SO_4. Mangrove clams (*Polymesoda erosa*) can absorb Fe content in the soil of brackish water ponds and have the potential as biomitigators. The class size of 61-80.99 mm has a significant effect ($p<0.05$) on the absorption of Fe content. Mangrove clams (*Polymesoda erosa*) can be used to mitigate the Fe content in brackish water ponds.

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