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Advantages of liming combined to Sedimentation-Dredging in reducing heavy metals and metalloid in water from a coal mining area

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ABSTRACT

A study on the advantages of liming combined to sedimentation-dredging to reduce heavy metals and metalloid in coal mining water was performed in tailing wastewater sedimentation ponds, mixed water sedimentation ponds, and Sangatta River where water from the coal mining area flows in. The concentrations of heavy metals and metalloid showed that the results were not significantly different (P > 0.05) between water from ponds or reservoirs of coal mining and water in the Sangatta River. The liming combined to sedimentation and the followed regular dredging of the sediment was useful in reducing the metals and metalloid in water from the coal mining area before being released into the public water.

Introduction

As a source of energy, coal has many advantages and disadvantages; the advantages include its cheap cost and the fact that it is easy to mine and simple to convert into energy, but the disadvantages are its impact on the environment (Theclaycenter, 2021). In Indonesia, coal is a source of national income due to coal export. In 2020, the total coal production in Indonesia was 561 million tons, but only 132 million tons were used for domestic energy (Indonesian Ministry of Energy and Mineral Resources, 2021). Some parts of Indonesia depend strongly on coal mining activity, as it improves the economy, nevertheless, it also increases social and environmental problems (Fitriyanti, 2016).

Coal mining activities produce huge amounts of water that may contain trace metals, including heavy metals and metalloid, which could be toxic to the public (Greenpeace, 2005). However, Dang et al. (2002), de Place & Kershner (2013), and Liu, Bai, & Liu (2019) mentioned that coal mining without burning coal could not introduce concentrations of heavy metals and metalloid into the aquatic environment. Some studies have shown changed and unchanged water quality in the presence of coal mining activity as noted in Table 1. Unfortunately, no studies collected the water samples directly from coal mining sedimentation pond, they have just collected them from the surrounding waters.

As coal mining drains its water into public waters, this has become the focus of many studies to assess the effect of coal mining activities on the quality of public water. Liming to raise water pH to fulfill the government regulation, and dredging the full sediment in sedimentation pond are common practices in coal mining industries in Indonesia. However, the effect of liming, the following sedimentation, and the regular dredging of the sediment, even as high as 25% from the pond bottom to decrease heavy metals in the released water, were not addressed previously in the literature. And this is the main aim of the present study.

Methods

This study was performed in a total of 8 years, with the sampling time divided into 4 periods of replications for the active reservoir of pond 19B; a primary tailing pond or coal slurry collection pond, pond 19A of secondary sedimentation tailing pond, pond KJ a sedimentation pond for all water except tailing water, and in Sangatta River of the East Kalimantan Province. Water pH in coal mining area reservoirs except Sangatta River is controlled by the mining company, which uses volumes of calcium oxide, approximately 250 kg per year, according to the pH of the water. Liming was done if the pond was full of water, and was let to settle for some days according to the weather. If the water produced has a pH over 7 and is clear, then it would be released into the river or

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Table 1
Water quality affected by coal mining in some countries.

Country	Water Quality Affected by Coal Mining	Authors
China	The presence of anions of HCO^- $SO_4^=$, and Cl^-, also cations of Ca^{2+}, Na^+, and Mg^{2+} were the major constituents.	Zhou et al. (2020)
USA	Lowest pH, $SO_4^=$ became the major anion, with the presence of higher $Fe^{2+,3+}$ and Al^{3+}cations.	Corbett (1977)
Australia	High in Ni^{2+} and Zn^{2+}, also the water became saline.	Belmer & Wright, (2019)
Indonesia	No changes in water quality; water has shown to be suitable as raw tap water.	Said & Yudo (2021)
Indonesia	Lowest polluted level in the parameters of pH, water temperature, BOD, COD, Fe, Mn, and TSS.	Khanifa, Sari, & Bargawa (2019).

*BOD: Biological Oxygen Demand; COD: Chemical Oxygen Demand; TSS: Total Suspended Solids.

sea, and afterwards the pond would be routinely dredged if the sediment is as high as 25% from the pond bottom even if the pond is full of water. All sampling locations are in the East Kutai Regency of the East Kalimantan Province of Indonesia (Fig. 1). The sampling point in the pond was at the water gate of the drain, and was performed in surface water (at least 3 days after liming was applied) using a borosilicate bottle sampler, and the pH was adjusted to 2 by adding nitric acid. Similarly, for Sangatta River water, sampling was done in surface water at about 30 cm depth.

The sampling points on the global positioning system were confirmed using GARMIN® GPS 12 as noted below:

Pond 19B: Latitude 00° 32′ 16.3″, longitude 117° 38′ 17.5″, is a primary water collector pond for tailing water.

Pond 19A: Latitude 00° 32′ 06.0″, longitude 117° 38′ 20.2″, is a pond for collecting water from the primary tailing water pond 19B.

Pond KJ: Latitude 00° 32′ 56.4″, longitude 117° 31′ 23.4″, is an active pond for collecting runoff from the land surface, rainwater, water from surrounding aquatic environments, and acid mine drainage.

Sangatta River: Latitude from 00° 27′ to 00° 35′, and longitude from 117° 17′ to 117° 37′; along this 92 km long river, which measures 30 m at its greatest width and almost 10 m at its greatest depth, three main human activities, which produce wastewater into the river, are present: the small town of Sangatta City, huge palm plantations, and some other coal mining enterprises.

The lowest detection limit for different heavy metals was as follows; iron (Fe), selenium (Se), cadmium (Cd), and metalloid of arsenic (As) with 0.001 mg/L; copper (Cu) and manganese (Mn) with 0.02 mg/L; lead (Pb) and antimony (Sb) with 0.01 mg/L; zinc (Zn) with 0.05 mg/L; and mercury (Hg) with 0.0005 mg/L. The analysis was performed by a certified commercial laboratory. The detectable samples containing heavy metal $\times$ (n) within a total number of samples for heavy metal $\times$ (t) were expressed by ATSDR (2003) equation with the Percentage Detectable (PD) = n / t $\times$ 100%, n or t = 1, 2, 3....

Results and discussion

Table 2 shows that heavy metal and metalloid concentrations fluctuated according to the sampling period from not detected (nd) to higher concentration. Water quality in coal mining reservoirs and Sangatta River in this study showed relatively similar results of Said & Yudo (2021) that showed water in coal mining area has no change with surrounding water detected. On the other hand, The present results was different to Belmer & Wright (2019), Khanifa, Sari, & Bargawa (2019), Corbett (1977) that showed change in water quality including some heavy metals, However, this differs from the findings of Zhou, Li, Zhang, Gao, Ullah, Peng, He, & Yu (2021) which showed changes in the presence of non-heavy metallic Ca2+, Na+, and Mg2+ as the major constituents. Mercury, selenium, and antimony were below the detection limit of the measuring instrument, therefore were noted as 'nd' in all samples. Iron was detected in 100% of all samples and years, and manganese was the second most detectable metal, with concentrations ranging from not detected to as high as 87.5%. Iron and manganese are two heavy metals that were distributed abundantly in the water of organic-rich water (Hamilton-Taylor & Davyson, 1995), due to the huge areas of vegetation surrounding the ponds and Sangatta River. Fluctuations in the concentrations of metals and metalloid in the water were not specific. The study by Sukhodolska (2017) showed that the concentrations of heavy metals fluctuated in different months of the year for Zn, Mn, Fe, Pb, Cd, Co, and Ni. Moreover, Anifowose & Oyebode (2019) recorded different concentrations for Pb, Cu, Mn, Ni, Zn, Cd, Fe, Cr, and Co due to human activities along the Osun River in Nigeria and this was also in accordance with the results of Sabbir et al. (2018) for As, Cd, Cr, Pb, and Hg. The study by Mohammed, Ibfahim, Hamid, & Abdulkareem (2016) showed that the heavy metals Pb, Cr, Cd, Ni, Co, Cu, Zn, Fe, and Mn in water not only fluctuated but were affected by coal mining and were present at significantly high concentrations. This was somewhat different from the results of the present study, as much of the data fell under the "not detected (nd)" moniker because very low concentrations were detected by the measuring instruments. These low concentrations of heavy metals in the water are seemingly common because there has been no prominent source in the coal mining area that leaches heavy metals. This was also noted previously by de Place & Kershner (2013). In addition, the results of a study by Hernandi, Rositah, Zarta, Ruslim, Kustiawan, & Aipassa (2019) on the water quality of 7 rivers flowing in the surrounding area of 7 coal mining companies of East Kalimantan Province of Indonesia showed no significant or not detected concentrations of the heavy metals Co, Cr, Cu, and Pb. High concentrations of heavy metals in water would otherwise be caused by differences in the mineral composition of the coal and the surrounding soil in the area as noted by Schweinfurth (2016). Furthermore, the low concentrations of metals and metalloid seemingly may be caused by the relatively small potential for the leaching of heavy metals from coal into the environment as reported by Cao, Li, & Zhang (2018).

In the Sangatta River area, the soil is acidic (pH 4.10 to 6.46) (Mashud & Manaroinsong, 2014), and the rainwater could be washing soil that contains pyrite and its oxidised forms including sulphuric acid suggesting that the parameters of pH and water quality may be affected by the increasing dissolved minerals (including metals and metalloid), and thus liming treatment may be required. Liming in freshwater, as noted by Mant, Jones, Reynolds, Ormerod & Pullin (2013), has a beneficial effect on organisms by increasing their abundance. Somridhivej & Boyd (2016) added that liming can maintain the pH, increase conductivity, cause a high concentration of total alkalinity, and increase calcium hardness. However, as mentioned by Jeng (1992) and Pohl (2020), liming should also affect the solubility of some heavy metals in stagnant water. The study by Jamali, Kazi, & Niaz (2008) found that As, Cr, Ni, Pb, and Zn showed decreased availability to a level of 10–40% as a result of liming, but there was increased mobility for Cd and Cu to 10% and 24%, respectively. Immobility is defined as combining heavy metals or metalloid with liming anions to form insoluble particles that can precipitate into the sediment. If the sediment is then dredged and removed into other areas, the pond should show reduced levels of heavy metal and metalloid contaminants.

In order to maintain the depth of the water column in ponds of the coal mining area, it must be controlled by dredging, which

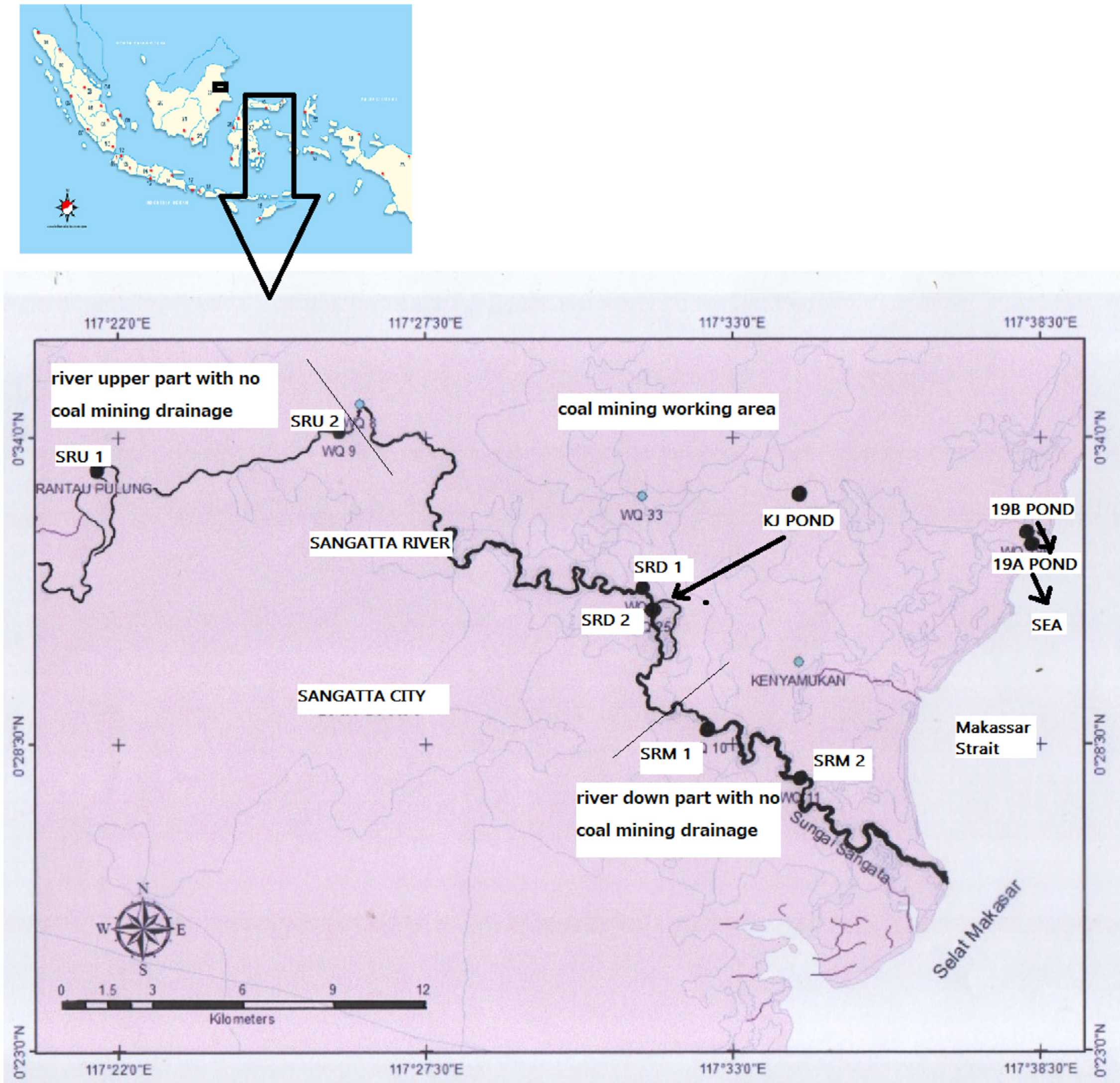

Fig. 1. Map of Indonesia (source: www.indonesia-tourism.com); the blue box shows the small black area of the present study (expanded below). The black dots are the sampling points in the present study. SRU 1 and SRU 2 are the upper part of the river with no coal mining drainage into the river, SRD 1 and SRD 2 are the river parts affected by coal mining drainage, and lastly, SRM 1 and SRM 2 are the down parts of the river with no coal mining drainage but affected by the water river current from the coal mining zone. The water of KJ Pond drained into Sangatta River (black arrow), where water from the tailing pond of 19B at first drained into 19A pond, after being treated again with sedimentation, then it is released into the sea of Makassar Strait (black arrow). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

causes high fluctuations in the parameters of transparency, turbidity, and total suspended solids. Similarly, Zhang, Zhou, Xu, Lin, Cheng & Wu (2010) found that dredging can cause a decreasing trend of phosphorus, organic matter, total suspended solids, Chlorophyll *a*, and transparency of the Secchi disc, but increased conductivity, total dissolved solids, and nitrates. However, these studies on the dredging effect did not include heavy metal and metalloid concentrations before and after dredging.

The heavy metal and metalloid concentrations in both ponds of tailing water showed no significant difference ($P > 0.05$) when using the Stratified Wilcoxon Test (Nasoetion & Barizi, 1986) for each heavy metal and metalloid in the statistical stratum. Even among the detected Fe, Cd, Pb, Mn, and As in these ponds, each individual metal and metalloid was tested by the Wilcoxon Rank Sum Test, with no single heavy metal or metalloid showing to be significantly different in both ponds ($P > 0.05$). This result has shown that even in tailing water, the liming, sedimentation and dredging ought to influence the heavy metals concentration in waters.

The percentage of detected concentrations of Cu, Pb, and Zn in all samples was higher for pond KJ than for samples from Sangatta River, but the opposite was true for Mn. The detected concentration was high in the water of pond KJ, but statistical analysis using the Stratified Wilcoxon Test and the Wilcoxon Rank Sum Test showed no significantly different results ($P > 0.05$) between the concentration of total and individual heavy metals and metalloid in water drained from the pond KJ mixed with water from Sangatta River at the SRD sampling point. This statistical result gave basic support to suggest that liming following sedimentation and regular sediment dredging was useful to reduce the presence of heavy metals and metalloid in water collected from coal mining activities to the same level as the water of Sangatta River.

Goldman & Horne (1983) and Beaumont (1975) stated that human activities including coal mining activities are an anthropogenic factor that should increase ions in the catchment areas

Table 2
The lowest, highest, and detectable percentage of heavy metals and metalloid in tailing water sedimentation ponds of 19B and 19A, the mixed water sedimentation pond of KJ pond, and Sangatta River at SRU, SRD, and SRM.

Parameter	Unit	Sampling Area					
		19B, n = 4*	19A, n = 4*	KJ, n = 4*	SRU, n = 8**	SRD, n = 8**	SRM, n = 8**
Iron (Fe)	mg/L	0.02–0.11 (100%)***	0.03–0.14 (100%)	0.04–1.78 (100%)	0.32–2.39 (100%)	0.25–2.55 (100%)	0.10–0.78 (100%)
Cadmium (Cd)	mg/L	nd −0.23 (25%)	nd-0.047 (25%)	nd	nd	nd	0.01–0.039 (50%)
Copper (Cu)	mg/L	nd	nd	nd-0.058 (25%)	nd-0.031 (75%)	nd-0.027 (12.5%)	nd-0.05 (25%)
Lead (Pb)	mg/L	nd-0.231 (25%)	nd-0.940 (50%)	nd-0.920 (25%)	nd	nd-0.01 (12.5%)	nd-0.440 (25%)
Zinc (Zn)	mg/L	nd	nd	nd-0.052 (50%)	nd-0.043 (75%)	nd-0.05 (12.5%)	nd-0.061 (25%)
Manganese (Mn)	mg/L	nd-0.032 (50%)	nd-0.021 (25%)	nd-3.050 (50%)	nd-0.141 (75%)	nd-0.169 (87.5%)	nd-0.23 (87.5%)
Arsenic (As)	mg/L	nd-0.004 (25%)	nd-0.001 (25%)	nd	nd	nd	nd
Mercury (Hg)	mg/L	nd	nd-	nd	nd	nd	nd
Selenium (Se)	mg/L	nd	nd	nd	nd	nd	nd
Antimony (Sb)	mg/L	nd	nd	nd	nd-	nd	nd

Notes:
*Kaltim (2011) stated that the government of East Kalimantan Province regulated water from both coal mining tailing wastewater and coal drainage, where the concentrations of Fe and Mn were not to exceed 7 mg/L and 4 mg/L, respectively. **The President of the Republic of Indonesia (2001) regulated that metalloid and heavy metal concentrations for raw tap water to be below: 0.3 mg/L for Fe; 0.01 mg/L Cd; 0.02 mg/L Cu, 0.03 mg/L Pb; 0.05 mg/L Zn; 1.0 mg/L Mn; 0.05 mg/L As; 0.001 mg/L Hg; and 0.01 mg/L Se; there was no regulation for Sb.
***The percentage value in parentheses is the level of heavy metals detected in all samples. nd: not detected or below the minimum level of detectable concentration by the appropriate instrument.

of a river. However, after comparing the entire concentration of heavy metals and metalloid in the upper part of Sangatta River (the SRU sampling point) with concentrations in the region close to the mouth of the river (SRM sampling point), there was no statistically significant difference between them ($P > 0.05$). It can be concluded that there is no difference between the concentrations of heavy metals and metalloid in the upper part and lowest part of the Sangatta River.

Conclusion

This present study concluded that treating the water from coal mining with liming following sedimentation and regular sediment dredging should result in a good impact on the water released into the natural river of the area. The first treatment with liming should precipitate the heavy metals and metalloid into the sediment, then the second treatment with regular sediment dredging, even if the pond is not full of water or only full as high as 25% of the pond capacity, should reduce the release of the heavy metals and metalloid from sediment into the water. Finally, we strongly recommend the liming combined to sedimentation and regular sediment dredging as standard procedure for treating the water produced from coal mining activities.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Ethical Clearance

Not Applicable.

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