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The Effect of Adsorbent Composition: Quartz Sand/Andisol Soil/Zeolite/Activated Carbon Against Mn, Fe, BOD, and COD in Citarum River Eater Cleaning Progress

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ABSTRACT

The results of the water quality monitoring by Perum Jasa Tirta II mentioned that water from the Cisanti outlet (upstream of the Citarum River) contained heavy metals that exceed the quality standard threshold. This study aimed to determine the effectiveness of heavy metal absorption with various absorbents, such as quartz sand, andisol soil, zeolite, and activated carbon in the Citarum River, West Java. The method that used in this work was refining silica and synthesized various combinations of quartz sand, andisol soil, zeolite, and activated carbon. The results showed that the composition of quartz sand/andisol soil/zeolite/activated carbon by 20/20/20/40 gave the best performance in the absorption. The studies of Fe, Mn, BOD, and COD revealed that the adsorption-treated water had met the parameters of PERMENKES RI number 32 of 2017. The variations of adsorbent composition did not affect the absorption of Mn and Fe metals content from water taken from the location before the dam and Citarum River 2, as indicated by statistical analysis. Although, for other locations, the variations of adsorbent composition impacted the reduction of Mn, Fe, BOD, and COD contents.

Keywords: Adsorption, activated carbon, andisol soil, composite, quartz Sand, and zeolite

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INTRODUCTION

The Citarum River is the longest and largest river in West Java. The upstream of the Citarum River starts from Mount Wayang, Bandung Regency, and ends at the mouth of the Java Sea, located at Muara Gembong, Bekasi Regency. The Citarum River plays a vital role as raw water for PDAM drinking water, electricity supply resource in Java-Bali, and providing irrigation water for rice fields in West Java. Pratiwi (2018) stated that the Citarum watershed is dominated by the manufacturing sector, such as chemicals, textiles, leather, paper, pharmaceuticals, metals, food and beverage products, also others [1]. There are so many industries in the Citarum River watershed that caused water pollution and are identified containing dangerous heavy metals [2].

Previous work was conducted to determine the value of indicators such as E-coli, BOD, COD, and the turbidity level downstream of the Citarum River. Preliminary results indicated that the clean water threshold had been exceeded with E-coli levels reaching 1600/100 ml, BOD 10 mg/liter, COD 19 mg/liter, and turbidity with a score of 320 NTU (12.8 times normal) [3].

One of the most important bacteriological pollution indicators is E-Coli that has reached up to 1600 amount/100 milliliters from the standard 0 ppm. Therefore, Citarum river water is not suitable as clean water based on the Minister of Health Regulation RI number 32 of 2017 [3].

The results of heavy metal content parameters monitoring in river water samples exhibited that the iron (Fe) content is relatively high and above of clean water threshold. Therefore, the residents cannot consume the water, either for river water and well water [4]. Some efforts are needed to overcome these problems. Previous research of natural technology to reduce water contamination has been carried out, which has been done by Pranoto et al. (2018) using clay and active andisol soil to adsorb Cu (II) metal then processed into crafts [5]. Pranoto et al. (2018) has also researched the composition of fiber/clay/andisol soil and andisol soil/zeolite composites for Cr (VI) metal adsorption [6]. The result certainly brings hope that natural sources can save costs and become efficient purification materials to reduce water contamination in the Citarum River, which is expected to be alternative raw water for water drink production.

In this study, the andisol soil was added into activated carbon, quartz sand, and active zeolite, thus becoming a various combination of andisol soil, activated carbon, quartz sand, and active zeolite in the ceramic membrane form, then used as an adsorbent for water purification in the Citarum river, West Java according to the standard quality of Indonesian National Standard (SNI).

EXPERIMENTAL

Filtration Performance Optimization of the Quartz Sand/ Andisol Soil/ Zeolite/Activated Carbon Combination

Silica sand refining process

The silica sand refining process procedure was modified from Ramadhan (2014) [7]. Quartz sand was sieved to obtain a homogeneous powder with a size of 100 mesh. A total of 50 g of quartz sand was placed in beaker glass and poured by 3 N HCl until HCl submerged the silica sand. The mixture is then heated until the silica sand dissolves, marked by a color change into a slightly greenish-brown. After that, the pH was neutralized with distilled water until pH 7. The wet solid was dried in an oven at 105 ^{0}C for 5 minutes.

Synthesis of various combinations of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon

In this research, the andisol soil adsorbent was used in tubular form, with activated carbon, allophane, quartz sand, and zeolite were added to it. Allophane used came from the soil on the slopes of Mount Lawu.

The research instrument design was as follows.

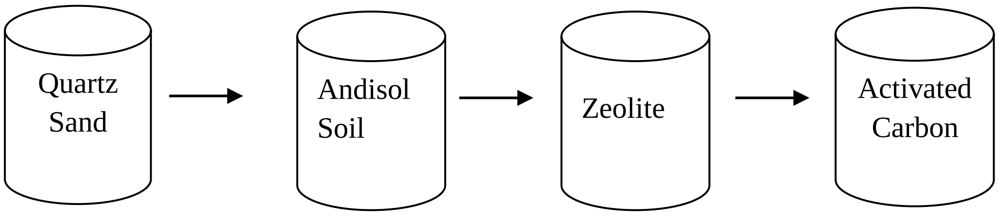

Fig. 1. The research instrument design.

Figure 1: The research instrument design of water treatment was including four processes, that was filtration by quartz sand, adsorption by allophane, zeolite, and the finishing process by activated carbon. The research variables were the best composition of each material on wastewater treatment effectiveness. The analysis was carried out in the laboratory by heavy metal Mn analysis, which caused Citarum River pollution.

RESULTS AND DISCUSSION

The effect of various adsorbent composition of the Quartz Sand/Andisol Soil/Zeolite/Activated Carbon on the reduction of Mn metal content at various sample points

The sample points that were used include before the dam, Walahar dam, Citarum 1, and Citarum 2 rivers, to determine the effect of various adsorbent composition and various research locations on the Mn metal content as shown in Figure 2 and Table 1.

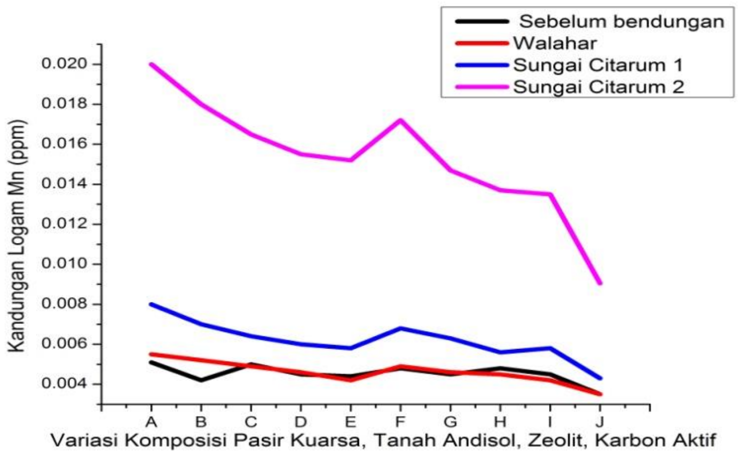

Fig. 2. The composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon A = No treatment, B = 100/0/0/0, C = 0/100/0/0, D = 0/0/100/0, E =0/0/0/100, F = 25/25/25/25, G = 40/20/20/20, H = 20/40/20/20 I = 20/20/40/20 and J = 20/20/20/40 on the Mn metal content reduction.

As seen in Figure 2 and Table 1, at the location of before the dam, the water sample was treated by nine variations and obtained that the optimum composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon was 20/20/20/40. This composition has appeared to be dominated by activated carbon due to its high surface area. The identification of the carbon surface area was analyzed using the BET method. The result revealed that activated carbon has a specific surface area of 821 m^2/g, this surface area was more significant than the surface area of quartz sand activated by 1M HCl of 222.068 m^2/g [8], the surface area of zeolite was 73.31 m^2/g [9] and andisol soil was 257.84 m^2/g [10]. The higher of the material surface area obtained, the more adsorbate content might be attached to the carbon surface [11].

Table 1. The effect of various adsorbent composition at various research locations on the reduction of Mn Metal Content.

Quartz Sand/ Andisol Soil/ Zeolite/Activated Carbon	Mn metal final concentration before dam (ppm)	Mn metal final concentration in Walahar dam (ppm)	Mn metal final concentration in Citarum River 1 (ppm)	Mn metal final concentration in Citarum River 2 (ppm)
No treatment	0.0051	0.0055	0.0080	0.0200
100/0/0/0	0.0042	0.0052	0.0070	0.0180

0/100/0/0	0.0050	0.0049	0.0064	0.0165
0/0/100/0	0.0045	0.0046	0.0060	0.0155
0 /0/0/100	0.0044	0.0042	0.0058	0.0152
25/25/25/25	0.0048	0.0049	0.0068	0.0172
40/20/20/20	0.0045	0.0046	0.0063	0.0147
20 /40/20/20	0.0048	0.0045	0.0056	0.0137
20/20/40/20	0.0045	0.0042	0.0058	0.0135
20 /20/20/40	0.0035	0.0035	0.0043	0.0090

The sample, obtained at the location before the dam, was treated by nine variations. The optimum composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon was obtained by 20/20/20/40, with the final concentration of Mn after adsorbent application of 0.0035 ppm. Furthermore, the other locations such as the Walahar Dam, Citarum 1 River, and Citarum 2 River also have the best Mn reduction after treatment by the composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon of 20/20/20/40, with final Mn metal concentrations respectively of 0.0035 ppm, 0.0043 ppm, and 0.00905 ppm. The locations with the lowest metal content of Mn are located in 2 locations, they were before the dam and after the Walahar dam, each of 0.0035 ppm. After adsorbent application, the samples of four locations have accomplished to the quality standard threshold according to PERMENKES RI number 32 of 2017. Whereas the other research, Noor (2018) reported that the elimination of Mn metal content using 25% activated carbon, 35% silica sand, 20% manganese, and 20% zeolite only obtained an outlet concentration 0.1 ppm [12].

Table 2. Statistical analysis of the effect of various adsorbent composition on the Mn metal content reduction at the four sampling locations.

Location	R Calculated	R Table	Calculated Significance	Significance 5 %	Note
Before Dam	0.518	0.632	0.125	0.05	There was no effect of the adsorbent composition on reducing the metal content of Mn
Walahar Dam	0.852	0.632	0.002	0.05	There was an effect of the adsorbent composition on reducing the metal content of Mn
Citarum River 1	0.821	0.632	0.004	0.05	There was an effect of the adsorbent composition on reducing the metal content of Mn
Citarum River 2	0.891	0.632	0.001	0.05	There was an effect of the adsorbent composition on reducing the metal content of Mn

The results of statistical analysis explained that if R Calculated > R Table or Calculated Significance < Table Significance, then the alternative hypothesis is accepted, which means to an effect of the adsorbent composition on the Mn metal content reduction. Although, if the R Calculated < R Table or Calculated Significance > Table Significance, then the null hypothesis is accepted, which means that there is no effect of the adsorbent composition on the Mn metal content reduction. The results showed that the treatment with any adsorbent composition did not affect the reduction of manganese quality, as seen in Table 2, with the statistical value from each location. The statistical results of the three locations, Walahar dam, Citarum 1 River, and Citarum 2 River, exhibited a significant effect between the adsorbent composition and the quality reduction of Mn metal, further proven by the statistical data in Table 2.

The effect of various adsorbent composition of the Quartz Sand/Andisol Soil/Zeolite/Activated Carbon on the reduction of Fe metal content at various sample points

The sample points that were used include before the dam, Walahar dam, Citarum 1, and Citarum 2 rivers, to determine the effect of various adsorbent composition and various research locations on the Fe metal content, as shown in Figure 3 and Table 3.

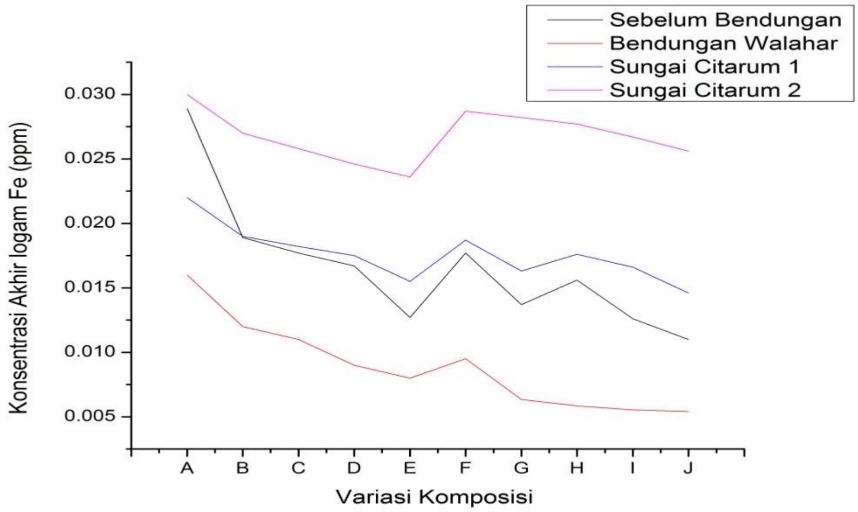

Fig. 3. The composition of Quartz Sand/Zeolite/Andisol Soil/Activated Carbon A = No treatment, B = 100/0/0/0, C = 0/100/0/0, D = 0/0/100/0, E =0/0/0/100, F = 25/25/25/25, G = 40/20/20/20, H = 20/40/20/20 I = 20/20/40/20 and J = 20/20/20/40 on the Fe metal content reduction.

The results, as seen in Figure 3, explained that the composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon by 20/20/20/40 was the best composition for water samples adsorption at four locations. The location variation in Figure 3 showed that the Walahar dam had the lowest iron content than the other four locations. A more detailed explanation can be seen in Table 3 as follows.

Table 3. The effect of various adsorbent composition at various research locations on the reduction of Fe Metal Content.

Quartz Sand/ Andisol Soil/Zeolite/Activated Carbon	Fe metal final concentration before dam (ppm)	Fe metal final concentration in Walahar dam (ppm)	Fe metal final concentration in Citarum River 1 (ppm)	Fe metal final concentration in Citarum River 2 (ppm)
No treatment	0.0289	0.0160	0.0220	0.0300
100/0/0/0	0.0189	0.0120	0.0190	0.0270
0/100/0/0	0.0177	0.0110	0.0182	0.0258
0/0/100/0	0.0167	0.0090	0.0175	0.0246
0 /0/0/100	0.0127	0.0080	0.0155	0.0236
25/25/25/25	0.0177	0.0095	0.0187	0.0287
40/20/20/20	0.0137	0.0063	0.0163	0.0282
20 /40/20/20	0.0156	0.0058	0.0176	0.0277
20/20/40/20	0.0126	0.0055	0.0166	0.0267
20 /20/20/40	0.0110	0.0054	0.0146	0.0256

The Fe can be produced from anthropogenic processes. According to Yunginger et al. (2018), anthropogenic processes are produced by human activities, for example, pollution caused by fuel, industrial waste, domestic waste, livestock waste, also agricultural and plantation waste [13]. The results in Figure 3 and Table 3 displayed the content of Fe metal in four locations, among others before the dam, Walahar Dam, Citarum 1 River, and Citarum 2 River by 0.0289 mg/L; 0.0160 mg/L; 0.0220 mg/L and 0.0300 mg/L. This Fe metal content was lower than Kirana report (2019) in the Citarum River, which is mentioned at 4.41 mg/L [4]. The four samples taken at the location of before the Walahar Dam, Walahar Dam, Citarum 1 River, and Citarum 2 River also meet the quality standards of PERMENKES RI number 32 of 2017. The metal content that has met this quality standard was initiated by the Presidential Regulation number 15 of 2018 concerning acceleration of pollution control and damage of the Citarum watershed by involving all stakeholders through discussion stages at the central, provincial, district/city government levels, private sector, academics, and environmental activists. This pollution control program is focused on solving problems in the aspects of handling critical lands; handling of the industrial, fishery, and livestock waste; handling of domestic liquid waste and solid waste; spatial/river use control; law enforcement; education and public relations; as well as monitoring water quality. Therefore, in order to support this government action, water contained metal treatment was carried out. The results of Citarum River water treatment are shown in Figure 3 and Table 4. Among nine variants of the adsorbent composition, the composition of Quartz

Sand/Andisol Soil/Zeolite/Activated Carbon by 20/20/20/40 presented the best performance at four locations. The best Fe metal content reduction was found at the location of the Walahar dam by Fe final concentration of 0.0054 ppm. In addition, the Fe metal content was reduced into 0.011 ppm, 0.0146 ppm, and 0.0256 ppm after being treated, found at other locations such as before the Walahar dam, Citarum 1 River, and Citarum 2 River.

Table 4. Statistical analysis of the effect of various adsorbent composition on the Fe metal content reduction at the four sampling locations.

Location	R Calculated	R Table	Calculated Significance	Significance 5 %	Note
Before Dam	0.796	0.632	0.006	0.05	There was an effect of the adsorbent composition on reducing the metal content of Fe
Walahar Dam	0.929	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the metal content of Fe
Citarum River 1	0.765	0.632	0.010	0.05	There was an effect of the adsorbent composition on reducing the metal content of Fe
Citarum River 2	0.153	0.632	0.672	0.05	There was no effect of the adsorbent composition on reducing the metal content of Fe

The results of statistical analysis showed that there was no effect of the adsorbent composition on the reduction of Fe metal content at the Citarum 2 River location. This result was due to the R Calculated < R Table or Calculated Significance > Table Significance, so the Nul hypothesis was accepted. Whereas the statistical analysis results of the three other locations, such as a location before the dam, Walahar dam, and Citarum River 1, indicating an effect of variations in the composition of the adsorbent with a reduction of Fe metal content. This result was due to the R Calculated > R Table or Calculated Significance < Table Significance, then the alternative hypothesis is accepted.

The effect of various adsorbent composition of the Quartz Sand/Andisol Soil/Zeolite/Activated Carbon on the reduction of BOD content at various sample points

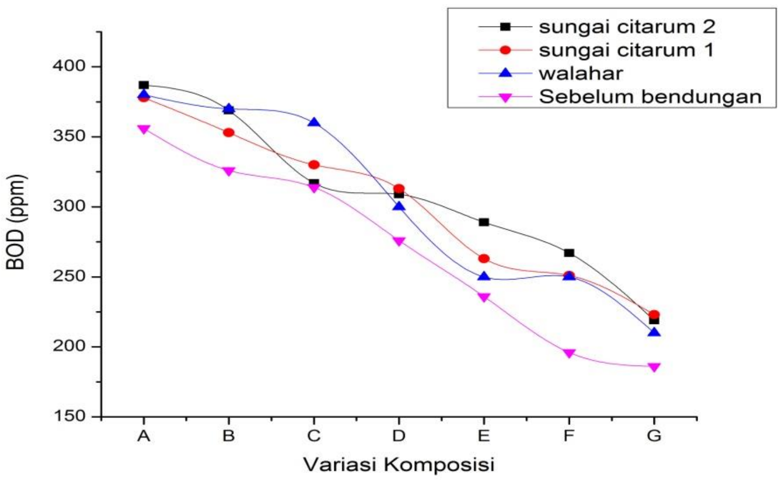

Fig. 4. The composition of Quartz Sand/Zeolite/Andisol Soil/Activated Carbon A = No treatment, B = 100/0/0/0, C = 0/100/0/0, D = 0/0/100/0, E =0/0/0/100, F = 25/25/25/25, G = 40/20/20/20, H = 20/40/20/20 I = 20/20/40/20 and J = 20/20/20/40 on the BOD content reduction.

The results of Figure 4 showed the optimum composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon was 20/20/20/40, and the sample point that had the best BOD content was at the location before the dam. The complete results are described in Table 5 as follows.

Table 5. The effect of various adsorbent composition at various research locations on the reduction of BOD Content

Quartz Sand/Zeolite/Andisol Soil/Activated Carbon	BOD final concentration before dam (ppm)	BOD final concentration in Walahar dam (ppm)	BOD final concentration in Citarum River 1 (ppm)	BOD final concentration in Citarum River 2 (ppm)
No treatment	3.56	3.80	3.78	3.87
100 : 0 : 0 : 0	3.26	3.70	3.53	3.69
0 : 100 : 0 : 0	3.14	3.60	3.30	3.17
0 : 0 : 100 : 0	2.76	3.00	3.13	3.09
0 : 0 : 0 : 100	2.36	2.50	2.63	2.89
25 : 25 : 25 : 25	1.96	2.50	2.51	2.67
40 : 20 : 20 : 20	1.86	2.10	2.23	2.19
20 : 40 : 20 : 20	1.63	1.99	2.13	1.87
20 : 20 : 40 : 20	1.59	1.88	2.10	1.76
20 : 20 : 20 : 40	1.22	1.75	1.87	1.70

The BOD provided an overview of how much oxygen is used by microbial activity during a specified period [14]. The higher the BOD value, so the greater of water pollution level by organic matter. A low BOD content indicated that the river is free from organic matter contamination [15] A high level of BOD in water is undesirable because it reduced DO [16]. The results of this study, as seen in Figure 4 and Table 5 showed that the BOD content of the four locations, before the dam, Walahar dam, Citarum 1 river, and Citarum 2 river before treated were 3.56 ppm; 3.80 ppm; 3.78 ppm and 3.87 ppm. This result was not much different from Arief et al. (2012) mentioned that the BOD concentration in the upstream Citarum River ranges from 1.6 mg/L - 20.5 mg/L [17]. After being treated using the best composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon 20/20/20/40, the BOD values were 1.86 ppm, 2.10 ppm, 2.23 ppm, and 2.19 ppm, respectively. In addition, the BOD content in four locations, before the dam, Walahar dam, Citarum 1 river, and Citarum 2 river, has appeared to meet the quality standard threshold set by the Minister of Health Regulation Number 32 of 2017 for a BOD content by 50 ppm.

Table 6. Statistical analysis of the effect of various adsorbent composition on the BOD content reduction at the four sampling locations.

Location	R Calculated	R Table	Calculated Significance	Significance 5 %	Note
Before Dam	0.988	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of BOD
Walahar Dam	0.973	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of BOD
Citarum River 1	0.983	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of BOD
Citarum River 2	0.989	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of BOD

The statistical analysis results of four locations, before the dam, Walahar dam, Citarum 1 River, and Citarum 2 River, indicated an effect of adsorbent composition on reducing BOD content. This result was according to R Calculated > R Table or Calculate Significance < Table Significance, then the alternative hypothesis is accepted.

The effect of various adsorbent composition of the Quartz Sand/Andisol Soil/Zeolite/Activated Carbon on the reduction of COD content at various sample points

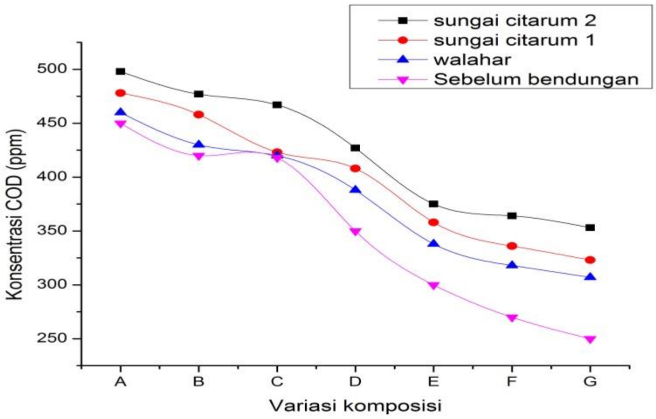

Fig. 5. The composition of Quartz Sand/Zeolite/Andisol Soil/Activated Carbon A = No treatment, B = 100/0/0/0, C = 0/100/0/0, D = 0/0/100/0, E =0/0/0/100, F = 25/25/25/25, G = 40/20/20/20, H = 20/40/20/20 I = 20/20/40/20 and J = 20/20/20/40 on the COD content reduction.

The results of Figure 5 presented that the optimum composition occurred at the composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon by 20/20/20/40. The location that had the lowest COD levels was the location before the dam. The complete results can be summarized in table 7 as follows.

Table 7. The effect of various adsorbent composition at various research locations on the reduction of COD Content

Quartz Sand/Zeolite/Andisol Soil/Activated Carbon	COD final concentration before dam (ppm)	COD final concentration in Walahar dam (ppm)	COD final concentration in Citarum River 1 (ppm)	COD final concentration in Citarum River 2 (ppm)
No treatment	4.50	4.60	4.78	4.98
100/0/0/0	4.20	4.30	4.58	4.77
0/100/0/0	4.18	4.20	4.23	4.67
0/0/100/0	3.50	3.88	4.08	4.27
0 /0/0/100	3.00	3.38	3.58	3.75
25/25/25/25	2.70	3.18	3.36	3.64
40/20/20/20	2.50	3.07	3.23	3.53
20 /40/20/20	2.20	2.87	2.98	3.21
20/20/40/20	2.10	2.77	2.77	3.11
20 /20/20/40	1.97	2.65	2.67	2.75

Minimum COD levels in sections of the Citarum River reached 14 mg/L and a maximum of 120.76 mg/L at dry season, while the minimum level was 12 mg/L and a maximum of 290 mg/L in the rainy season. Whereas, in reservoirs, the minimum concentration of COD in the dry season was 14.67 mg/L and a maximum of 43.56 mg/L, while in the rainy season was a minimum of 15 mg/L and a maximum of 43.0 mg/L. The results in Figure 6 and Table 8 showed that COD content in 4 locations, before the dam, Walahar dam, Citarum 1 River, and Citarum 2 River before treatment were 4.50 ppm; 4.60 ppm; 4.78 ppm, and 4.98 ppm. This result was smaller than its predecessor of Citarum River due to the existence of Presidential regulation number 15 of 2018 concerning the acceleration of pollution control and damage to the Citarum watershed, and then the Citarum River kept clean. This government program is also supported by COD waste treatment. The treatment using the best composition of Quartz Sand/Andisol Soil/Zeolite/Activated Carbon 40/20/20/20, exhibited a reduction of COD level into 1.97 ppm, 2.65 ppm, 2.67 ppm, and 2.75 ppm, respectively. Therefore, the COD content in the 4 locations had met the quality standard threshold set by the Minister of health regulation RI number 32 of 2017 for COD content by 90 ppm.

Table 8. The effect of various adsorbent composition on the reduction of COD Content in Citarum 1 River

Location	R Calculated	R Table	Calculated Significance	Significance 5 %	Note
Before Dam	0.979	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of COD
Walahar Dam	0.981	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of COD
Citarum River 1	0.991	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of COD
Citarum River 2	0.989	0.632	0.000	0.05	There was an effect of the adsorbent composition on reducing the content of COD

The statistical analysis results of the four locations, before the dam, Walahar dam, Citarum 1 River, and Citarum 2 River, indicated an effect of adsorbent composition on reducing the COD content. This result was due to R Calculated > R Table or Calculated Significance < Table Significance, then the alternative hypothesis is accepted.

CONCLUSIONS

The experiment results of Fe metal, Mn metal, BOD, and COD content appeared to meet the parameters of PERMENKES RI number 32 of 2017. Quartz Sand/Andisol Soil/Zeolite/Activated Carbon with 20/20/20/40 ratio was the best composition for Mn metal. The statistical analysis results showed that adsorbent composition had not affected the reduction of Mn and Fe metals at locations before the dam and Citarum 2 River. For other locations apart from before dam and Citarum 2 River on Mn and Fe metal, The statistical analysis results showed that adsorbent composition had affected the reduction of Mn, Fe, BOD, and COD content.

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