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Modification of rapid sand filtration by activated carbon powder (ACP) for heavy metals (HMs) adsorption: Case study Babol River

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Abstract

Heavy metals (HMs) release in the surface water and river should be treated. Sand filtration (SF) is known as traditional in situ water treatment. However, SF technique may not be able to remove HMs from water efficiently. In this research, the performance of traditional SF on HMs removal was modified by adding activated carbon (AC). Real samples of contaminated water from Babol River was used to evaluate Pb and Cu removal by SF and modified SF with AC. The effect of operation parameter such as AC dosage, contact time, initial concentration of HMs and pH on removal efficiency were studied. Moreover, SEM, EDX and BET analyses were performed to investigate HMs removal mechanism. According to the results, adding 50 g/kg of AC to SF could increase the removal efficiencies of Pb and Cu from 46% (SF) to 78% and 68%, respectively within 30 min contact time. Initial concentrations of HMs and pH variation showed negative effect on HMs removal. The results of SEM-EDX and BET analyses revealed that AC has high surface area which can effectively adsorb HMs through physico-chemical mechanisms. This study suggest that modification of traditional sand filtration by activated carbon, as a promising approach for heavy metals removal from contaminated surface water.

Key words: Sand filtration; River water treatment; Activated carbon; Babol River



1. Introduction

Water is considered as the most essential and abundant resource for life on earth, so water pollution has turned out as a significant concern around the world [1]. In line with that, increasing heavy metals pollution represents a major worldwide issue of water contamination because of their toxicity and non-biodegradability, as well as accumulation effects on human and aquatic life [2]. Among heavy metals, Cu and Pb are the most common ones derived from both natural and anthropogenic sources [3]. Cu is necessary nutrient element for human growth and health in trace quantities; nonetheless, excessive intake of Cu may cause many health risks, including kidney or liver damage, brain or nerve inflammation, or anemia [4]. Pb can cause severe damage in nervous and genital system if it gets enrichment in the human body [5]. Therefore, exploring economical and renewable techniques removing above heavy metals from water is important for safe water production. In recent decades, many techniques have been introduced for treating contaminants in water, which include electrocoagulation, distillation[6], desalination[7], membrane filtration[8], reverse osmosis[9], electrodialysis[10], coagulation[11], filtration[12], ion exchange, precipitation, adsorption[13].

Sand filtration is one of the methods used to treat wastewater[14]. The sand filter is considered to be a simple technique for potable water treatment, low cost, efficient, and reliable [15]. Sand filters were initially designed to remove suspended solids and reduce turbidity in source water based on the physical retention and adsorption of filter material[16]. Bio-sand filters have been identified as one of the most sustainable technologies for improving household water treatment. Although, sand filtration is well known as a simple operation and low-cost method for water and wastewater treatment, the final removal capacity of heavy metals is not satisfactory [17], [18].

In order to improve the removal efficiency in sand filtration systems, many research groups suggested modification of sand filtration by coating nanomaterial and adding different adsorbents such as clay, zeolites, activated carbon and biochar [15]–[17], [19]. In this direction, Saad et al. [14] attempted to modify the sand filtration by adding activated carbon to treat the grey water and could successfully improve the removal efficiencies of TSS, BOD, COD, and PO_4^{3-} up to 94%, 96%, 93% and 57%, respectively [14]. Further investigation on sand filter revealed that activated carbon had improved the efficiency of the filters. The results showed that the percentage removals for all parameters tested were increased when coconut shell activated carbon was added as part of the media. This is because activated carbons based on coconut shells are the least dusty and are mainly micro porous which ideal for organic chemical adsorption. It also has the highest hardness compared to other types of activated carbon, which makes it the best carbon for purifying water[14]. However, there is a real need to develop alternate, cost-effective, and large-scale activated carbon for industries related to chemicals/catalysis and environmental remediation. As of now, there are



several materials such as bagasse pith, wood, peat, etc. are being used for the preparation of activated adsorbent carbon.[20]

In this study, activated carbon(AC) which was obtained from coconut shells was added to sand filtration as a sustainable adsorbent to increase HMs removal efficiency. AC is inexpensive material (almost free of charge) and available worldwide. Therefore, in this research, we evaluate the activated carbon (AC) application of as an adsorbent to modify sand filtration performance for Pb and Cu metals removal. In line with that, first, the AC characterization was conducted by using Energy dispersive X-ray (EDX), scanning electron microscopy (SEM) and Brunauer-Emmett-Teller (BET) analyses. Then, the sand filtration was modified using different dosages of activated carbon. Then, Effect of contact time, initial concentrations of heavy metals and pH on the adsorbent performance and heavy metals removal was also investigated.

2. Materials and methods

2.1. Polluted water sampling

Water samples (~1 L) were collected from 5 different points of Babol River, Mazandaran, Iran (Fig.1). The average concentrations of heavy metals in the river at different sampling points were presented in Table 1. According to the Table 1, samples from point 4 showed the highest level of heavy metals due to industrialization in the urban area and wastewater injection in to the river. In particular, the level of Pb and Cu were much higher than the accepted standard level for drinking (Pb< 10 µg/L and Cu < 1000 µg/L) and agriculture (Pb< 5m g/L and Cu <0.2mg/L) purposes (EPA).



Fig. 1. Sampling locations from Babol River, Mazandaran, Iran



Table 1. Heavy metals content of Babol River water at different sampling points

Code	Location	pH	T _w	Cu	Pb	Co	Cr	Cd	Al	Zn
			(°C)	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L	µg/L
S1	"N 13.06'1436° "E5.72'4852°	5.5	10°	2262.2	88.57	5.58	1.504	0.78	87.7	0.187
S2	"N 5.0434'2636° "E49.301'4152°	6.0	11°	2362.9	292.2	5.59	2.643	0.97	145	0.548
S3	"N 5.3455'2636° "E49.255'4152°	6.3	15°	2419.5	306.9	6.34	5.99	1.33	173	0.662
S4	"N 39.617'3636° "E30.485'3852°	6.6	16.5°	2483.5	537.5	12.65	6.11	1.49	182	1.065
S5	"N 5.147'4236° "E44.097'3852°	6.8	14°	2506.9	498.1	1.24	4.04	1.86	146	1.367
Standard (EPA)		7		1300	15	300	50	5	200	5000

2.3. Sand filtration column and experimental procedure

The activated carbon was purchased from Jacobi Activated Carbon Company with 12-16 mesh. A lab scale column with size of 18 cm high, 100 cm² area was filled with 300 g fine sand with average diameter of (200 – 500µm). A mesh screen with average pore size of 2µm and one tab was located at the bottom of the set up for sampling. To modify the sand filtration, AC with different dosage of 10, 20, 30, 40 and 50 g/kg were hand mixed with the sand filtration. Then, the columns were poured with polluted river water until it becomes saturated. The porosity of the sand filtration was measured as 34 %. For each filtration test the polluted water was remained for different time of 10 to 60 min to investigate the effect of contact time on the modified sand filtration performance. After each test 15 mL of the treated water was collated to determine the final concentration of heavy metals.

2.4. Analytical methods

The HMs concentrations in the polluted water before and after adsorption process were measured using an atomic absorption spectrometer (ICP-OES, USA). The removal efficiency (%) and adsorption capacity (mg/g) of the HMs were calculated using Eq. (1) and Eq. (2), respectively:

$$\text{Removal efficiency (R)} = [(C_0 - C_e)/C_0] \times 100 \quad (1)$$

$$\text{Adsorption capacity (q}_e) = (C_0 - C_e)V/M \quad (2)$$

Where C_0 and C_e (mg/L) are the initial and final concentrations of HMs in the water before and after treatment, respectively. M is the mass of the adsorbents (g) and V is the volume of the polluted water (L) which was equal to 0.25 L in this study.



The morphology and shape of the AC before and after treatment were observed under a scanning electron microscope (SEM) (SNE-4500 M; SEC Co., Tokyo, Japan) and light microscopic (CX31, Olympus Corporation, China). The surface area is determined with Brunauer-Emmett-Teller (BET) instrument (Micromeritics, ASAP, Model 2010) with N₂ adsorption-desorption isotherms were recorded at 77 K. All the measurements were carried out in triples and the results were reported as mean values ± Standard deviation (SD).

3. Results and discussions

3.1. Removal performance of the modified sand filtration with AC

The removal performance of lead (Pb) and copper (Cu) from contaminated river water through the sand filtration modified by AC was investigated in terms of contact time, adsorbent dosage, initial concentration of adsorbate (Pb and Cu) and pH.

3.1.1. Effect of contact time on HMs removal

Fig. 2 shows the removal efficiencies of Pb and Cu as a function of contact time under different dosages of AC adsorbents. According to the results, contact time showed a direct effect on the HMs adsorption by the AC. The HMs removal efficiency increased rapidly with an increase in contact time during the initial stages (approximately 30 min) in all the cases. It was observed that after 30 min contact time, the HMs removal efficiency became almost constant which means the surface of adsorbent was fully occupied by the HMs ions and no more room was left for further adsorption.

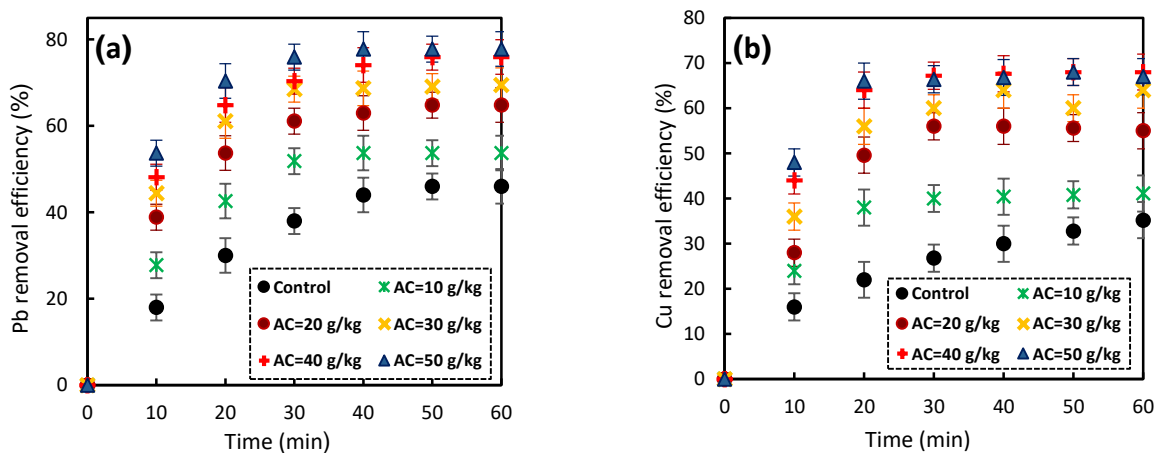


Fig. 2. Effect of contact time on the removal efficiency of (a) Pb and (b) Cu using AC



3.1.2. Effect of adsorbent dosage on HMs removal

The adsorbent dosage is an important parameter in the application of AC for the modification of sand filtration performance in large scale. Fig. 3 shows the effect of adsorbent dosage of AC on the removal efficiencies of HMs (Pb and Cu) through sand filtration process at particular contact time of 60 min. According to the results, 50 and 40 g/kg of AC could remove Pb and Cu up to 78% and 68%, respectively. However, the final concentrations of Pb in the treated water by AC (0.12 mg/L) did not meet the standard level for drinking water (the dotted line the Fig. 2a: $Pb < 0.015$ mg/L) according to EPA 2020[21]. While, in the case of Cu, the final concentrations reached to below the standard level for drinking water (the dotted line the Fig. 2b: $Cu < 1.3$ mg/L) after treatment with AC (0.8 mg/L).

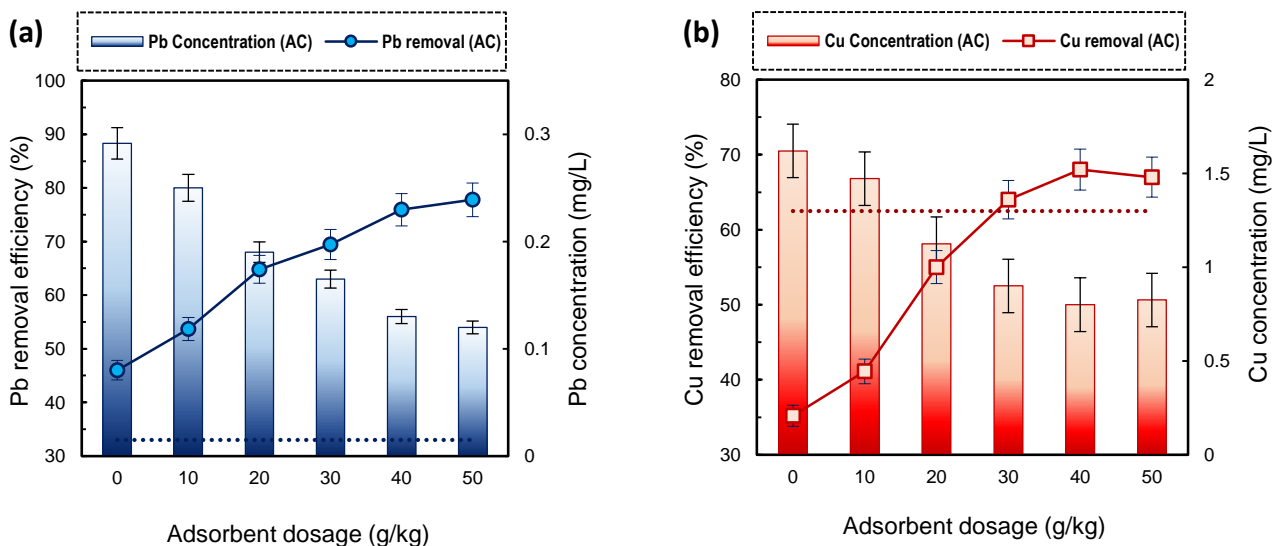


Fig. 3. Effect of AC dosage on (a) Pb and (b) Cu removal

3.1.3. Effect of initial concentration of adsorbate on HMs removal

The initial concentration of the HMs affect the adsorption performance [3]. Fig. 4 shows the effect of various initial concentrations of Cu (from 2.5 to 50 mg/L) on the final removal efficiency of the HMs by the AC. The other parameters such as adsorbents dosage and contact time were kept constant. According to the results, an increasing in the initial concentration of Cu and Pb up to 10 mg/L resulted in an increase of removal efficiency, but then the removal efficiencies were decreased by and increasing of the HMs initial concentrations.

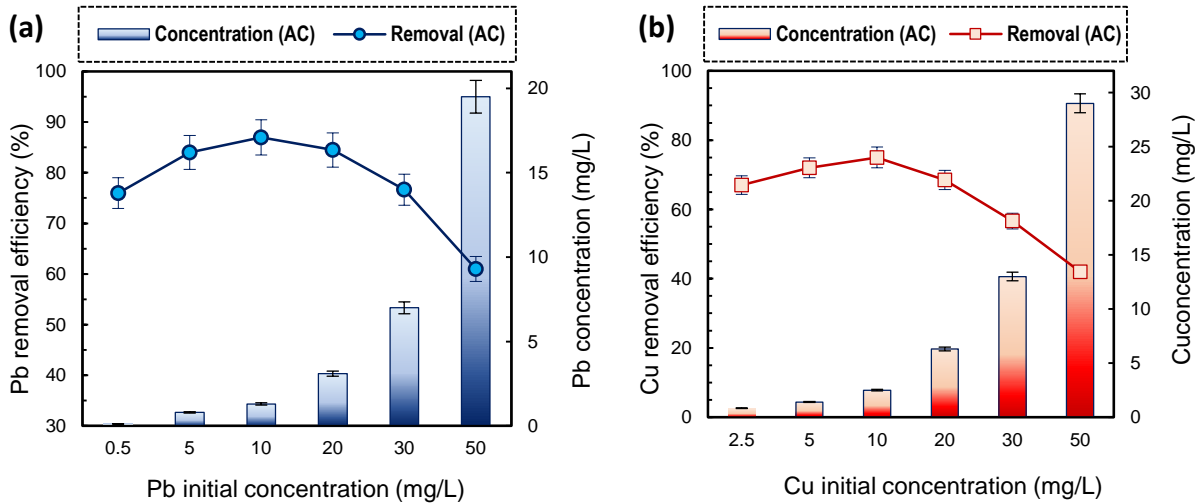


Fig. 4. Effect of AC dosage on (a) Pb and (b) Cu removals

3.1.4. Effect of pH on HMs removal

The initial pH of the water directly affects the HMs adsorption process [22]. Fig.5 shows the effect of pH variation (from 3 to 11) on the removal efficiency and final concentrations of Pb and Cu after 60 min treatment by the sand filtration process modified with AC. The adsorbent showed their best performance at pH around 7 which was the initial pH of the river water ~ 6.6 (Table 1). The maximum adoption of the HMs under natural condition ($6 < \text{pH} < 7$) using different adsorbent has been previously reported [3], [17], [22], [23]

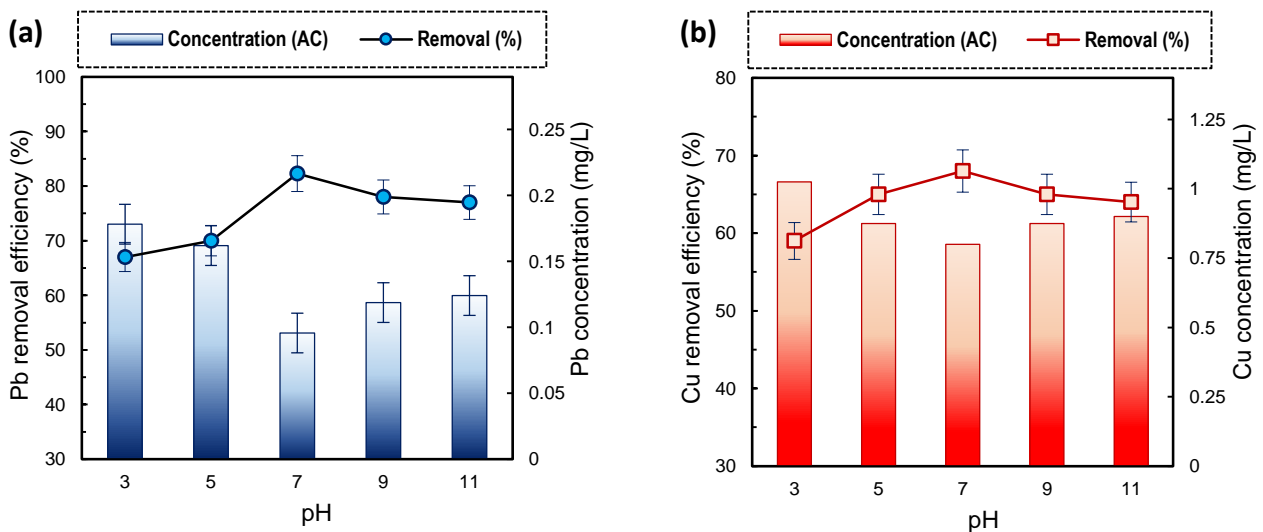


Fig. 5. Effect of pH on (a) Pb and (b) Cu removal



3.2. Adsorption mechanism

FE-SEM, EDX and BET analyses were performed to characterize the mechanism of HMs adoption by the adsorbent of AC which are used for modification of sand filtration under the optimal conditions: AC dosage = 50 g/kg, pH = 7, contact time = 30 min and initial concentrations of Pb = 0.5 mg/L and Cu = 2.5 mg/L.

3.2.1. SEM analysis of the adsorbents

FE-SEM analysis was carried out to examine the morphology, distribution size, and physical properties of adsorbent. Figure 6a shows the surface morphology of the AC adsorbent determined by SEM analysis with a magnification of 1000X. According to the SEM image, a rough structure with fine porous distributed on the surface was observed for the AC which indicates a high capability of HM adsorption by the AC. Moreover, FE-SEM analysis was conducted before and after the adsorption process to investigate the presence of Pb and Cu elements in the adsorbent and the way of distribution of these elements at the material surface. In figure 6b, yellow points represent lead metal (Pb) and red points show Copper metal (Cu). It was observed that the number of yellow points were more than red points which indicates higher adsorption of Pb than Cu by AC. The results of FE-SEM analyses proved the adoption and removal results obtained in the previous sections.

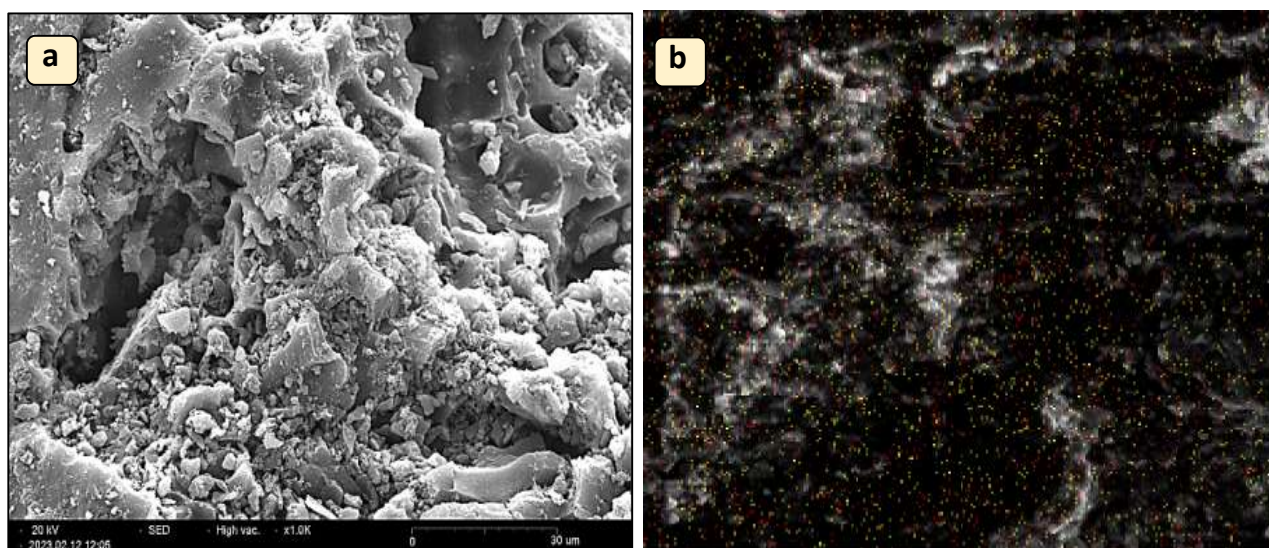


Fig. 6. (a)SEM image before and (b) EDX mapping of AC after treatment with a magnification of 1000X



3.2.2. EDX analysis of the adsorbent

Figure 7 shows the energy dispersive X-Ray (EDX) analysis of the AC adsorbent. As might be expected, the weight percentages (w %) of carbon elements in AC (C ~ 100 w %). Therefore, it has been reported in the literature that carbon composition acts as a binding agent which results in better adsorption process. The EDX result proved that carbon (C) was almost the only element of the AC adsorbent used in this study. Ultimately, the result of EDX analysis proves that the adsorbent have the potential of being used as appropriate adsorbent for heavy metals.

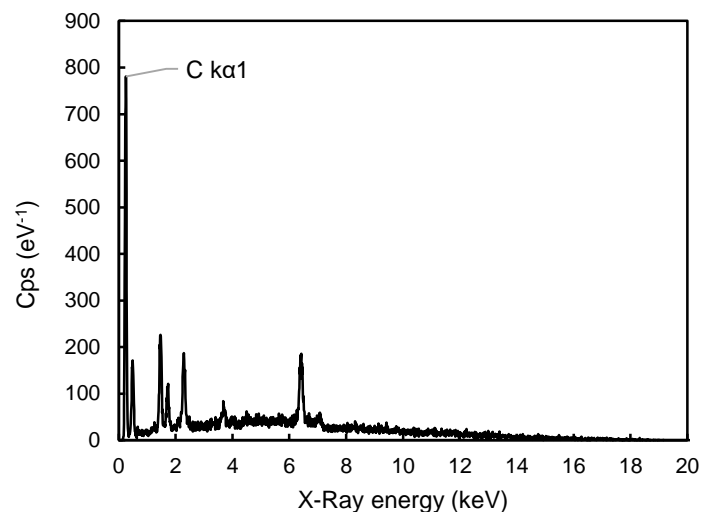


Fig.7.EDX analysis of AC

3.2.3. BET analysis of the adsorbent

BET analysis was performed by N₂ adsorption-desorption at temperature of 77 K (-196 °C) to investigate the specific surface area and porosity of the AC (Figure 8). According to the results, AC was classified as a type III isotherm based on the IUPAC classification [24] which means that the adsorbent used in this study had macro-porous structure. The specific surface area of the AC was recorded as 4.278 m²/g. In addition, the pore size distributions of the AC are shown in Fig. 8. Total pore volume of 0.012 cm³/g and average pore diameter 11.29 nm were determined for the AC. The results of the pore size distributions also confirm the macro-porous nature of the adsorbents. The BET analysis revealed that AC has high specific surface area and porosity which can significantly improve the adsorption capability.

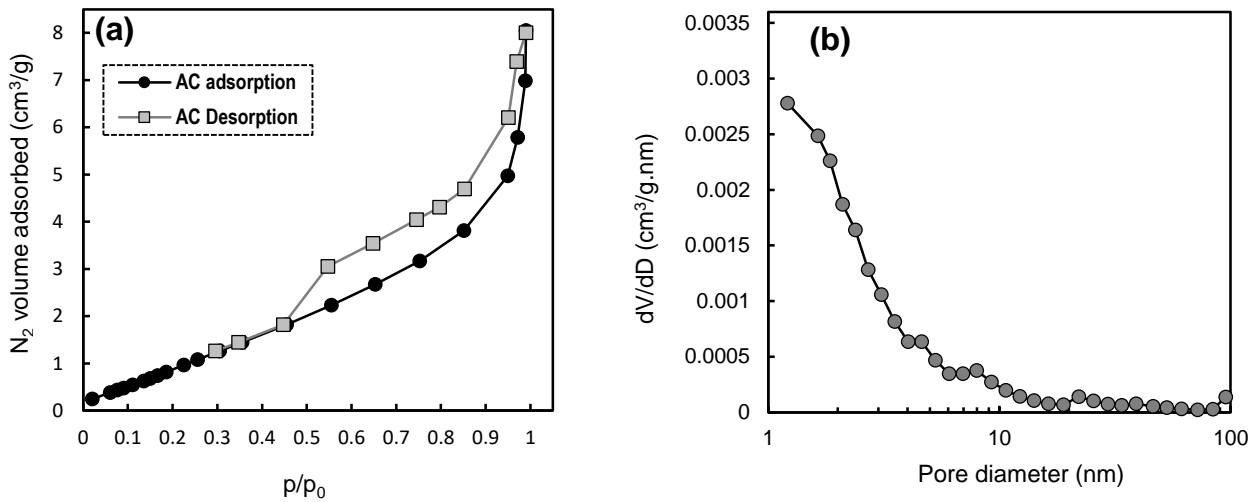


Fig. 8. BET analysis: (a) N₂ adsorption-desorption isotherms and (b) pore size distribution curve adsorbent of AC

3.3. Comparison with previous

A comparison between the AC adsorbents suggested in this study and that in the literature is presented as Table 2. According to Table 2, using sand filtration alone failed to successfully remove heavy metals from water. However, adding AC could significantly improve the SF performance on HMs removal. Adding other materials for SF were also reported for improving SF performance. For example, Kumar et al. 2022 [16] added Aminoalkyl-organo-silane to SF to improve Ar removal from wastewater.

Table 2. Comparison between the AC adsorbents suggested in this study and that in the literature

Sand modification with	pollution	Removal	Ref.
Aminoalkyl-organo-silane	Arsenic (As ⁵⁺)	86%	[17]
Biochar	Gram-negative and Gram-positive bacteria	42.9%	[16]
Zero-valent iron	Bacterial capture and inactivation	99%	[25]
Activated carbon	Organic micro pollutants	80%–99.5%	[26]
Activated carbon	Phosphate (PO ₄ ³⁻)	57.07%	[14]
Activated carbon	Lead (Pb ²⁺)	78%	This study
Activated carbon	Copper (Cu ²⁺)	68%	This study



4. Conclusion

This study investigates modification of sand filtration with activated carbon (AC) for the removal of lead Pb (II) and Copper Cu(II) from the river. The specific surface area and pore volume of the resulting AC were obtained as $4.287\text{m}^2/\text{g}$ and $0.012\text{cm}^3/\text{g}$, respectively. This research revealed that the removal of HMs was pH dependent, and the optimum pH was determined as around 7. The highest percentage removal of Pb and Cu was found to be 78% and 68% for AC (50 and 40 g/kg), respectively, at 30 min of contact time. The experimental results proved that AC could be employed successfully as a cost effective, benign, environmentally friendly adsorbent for the remediation of heavy metals in this environment.

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