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Designing activated carbon and zeolite amended biosand filters: optimization using response surface methodology

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ABSTRACT

Biosand filters can be amended with activated carbon and zeolite in order to improve their efficiency at removing micropollutants. The tested pollutants here include ammonia (5 mg/L), lead (210 µg/L), phosphate (12 mg/L), COD (400 mg/L), and iron (0.4 mg/L). The response surface methodology (RSM) is used to evaluate the influence of the adsorbents' column heights (H) and lead ([Pb]) on system efficiency. The results show that (i) increased H values enhanced filter efficiency; (ii) increased [Pb] values only influenced the lead concentration of the effluent; (iii) there was a higher removal of ammonium, COD, and phosphate; and (iv) the removal of iron was below the allowable level. The RSM results suggest that heights of 33 cm and 26 cm could be considered optimal for zeolite and activated carbon, respectively. The optimized filter quantitatively removed biological pollutants and was capable of removing ammonium, lead, COD, and phosphate pollutants at upto 98, 98, 97, and 87%, respectively. Amending biosand filters with efficient adsorbents enables them to be used to remove micropollutants.

Keywords: Activated carbon; Biosand filter; Box-Behnken; Response surface methodology; Runoff; Zeolite

1. Introduction

Access to ample, clean, high-quality water is a matter of great importance for sustainable development [1,2]. Half a billion people live in countries that will likely face water scarcity, and due to the increasing population, this number is expected to reach three billion by 2025 [3]. Nearly a billion people around the world do not have access to potable water, with most of these people living in rural areas of developing countries [4,5]. The World Health Organization (WHO) encourages the development of household treatment technologies [6,7]. Surface water is generally used for drinking and agricultural use, such as irrigation [8,9]. An increase in impervious surfaces in an area (such as roads and rooftops) prevents rain from soaking into the soil, so runoff is created that collects pollutants [10]. The impact of storm water runoff in rural areas is particularly substantial, with the most rural streams yielding high total organic carbon concentrations [11]. TKN, TP and heavy metals are the most commonly found contaminants in surface waters, and many of them are harmful. High concentrations of COD and ammonium have serious effects on people's wellbeing [12], while metals such as lead, cadmium, mercury, iron, and arsenic are harmful even in very low concentrations and have undesirable long-term consequences [13]. Neurological disorders; cancer; respiratory and cardiovascular disorders; damage to the liver, kidneys and brain; hormonal imbalance; abortion; arthritis; osteoporosis; and in extreme cases, death are among the effects that heavy metals have on the body [14]. It is therefore important to develop cheap, efficient, filtering systems to remove pollutants from runoff water.

Biosand filters are a cost-effective technology built from commonly available materials, so they are a suitable alternative to expensive water-treatment systems [15,16]. A biosand filter is a modified version of a slow sand filter on a smaller scale, and it is designed for frequent use [17]. In this filter, four actions occur, mechanical trapping, predation,

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adsorption, and natural death. Microorganisms living in the media bed of the biosand filter significantly reduce biological pollutants [18–20], and biosand filters can also reduce turbidity [21,22]. Biodegradation, as well as nitrification and denitrification processes, occur in biosand filters [19,23]. In addition to the microbial pollutants, chemical pollutants are a major obstacle to healthy water. The presence of heavy metals in water resources can occur both naturally and through contamination [24], causing considerable concern [25,26]. Metals such as lead, cadmium, mercury, silver, aluminum, barium, and arsenic are detrimental to one's health at any concentration, and they have adverse long-term effects on the body [12]. Unfortunately, biosand filters cannot remove some chemical pollutants and heavy metals, so they need to be modified and improved.

An efficient way to remove chemical contaminants is to use adsorbents. Many adsorbents have been found to be cost effective, and some of them can be made from available natural materials [27,28]. As a result, combining a biosand filter with these adsorbents is not just cost effective but also leads to improved results. In 2016, Ghebremichael et al. used pumice to enhance the hydraulic performance of biosand filters. Biosand filter also led to reduced water turbidity (<0.23 NTU) [29].

Using metallic iron, such as scrap iron or iron nails can also improve biosand filter performance [30–32]. The process of aqueous iron corrosion in a packed bed has been proven to be efficient for the removal of unspecific aqueous contaminants [33]. In 2012, Noubactep enhanced the performance of household Fe0/sand filters by using bimetallics and MnO_2 [34]. Changing sand filters into GAC-sand dual media filters (GSF) leads to a more efficient removal of organic matter and ammonium [35].

In this study, zeolite and activated carbon adsorbents are examined. Many researchers have conducted work on activated carbon and shown its great potential to remove organic pollutants [36,37] and heavy metals such as lead [38,39]. Zeolites, meanwhile, are naturally occurring hydrated aluminosilicate minerals. Zeolite's structure comprises a 3D framework of SiO_4^+ and AlO_4^+ tetrahedral [40]. Many studies have been conducted to assess zeolite's ability to remove various pollutants, such as ammonium [41,42], lead [43], and phosphate [44]. Also, in 2001, Mwabi et al. used sand and zeolite as a base layer in biosand filters. The removal efficiency of fluoride, calcium, iron, magnesium, nitrate, phosphate, and arsenic was 99, 90, 64, 57, 18, 39, and 68%, respectively [45].

Response surface methodology is a set of statistical and mathematical techniques that employ experimental data sets for modeling and optimizing both responses and variables [46]. Traditional optimization techniques consider only one factor at a time, with other factors being constant, which makes these techniques expensive and time-consuming due to the high number of experiments required. In contrast, with fewer experiments, the response surface methodology can examine and optimize the interactive effect between variables [47]. In 2015, Tundia et al. studied the influence of three parameters (pause time, charge volume, and influent turbidity) on the performance of biosand filters using response surface methodology [48].

The aim of this study is to develop an optimal and efficient system as a supplemental treatment step for water purification with biosand filters. For this reason, columns of zeolites and activated carbon are placed after the biosand filter. In order to investigate their effects and how to optimize them, the height of the zeolite and activated carbon adsorbent columns, as well as the influent lead level, were considered to be variable. (The height of the activated carbon and zeolite adsorption columns are considered to be variable so as to measure their effects beyond using the minimum amount of materials and occupying the least space.) To study the effect of parameters involved in the removal of contaminants, as well as optimizing the heights for the adsorption columns, the test was designed using the response surface method.

2. Materials and method

2.1. Materials and pilot construction

In order to build a pilot, Plexiglas columns were used with a height of 1 m and a diameter of 30 cm. In the columns, a 5 cm layer of gravel was placed at the bottom to allow drainage, with layers of sand placed above it to support the filtration sand and prevent it from escaping through the drainage layer and out through the outlet tube (Fig. 1). The main layer was then added. The main layer of the biosand filter was a 99% pure silica sand with a uniformity coefficient of 1.7 and an effective size of 0.2 mM. (The maximum sand grain size was about 0.4 mM.) The main layer of the adsorbent column contained activated carbon and zeolite with a grain size of 1–3 mM. The granular activated carbon (GAC) used in this study was prepared from hard-skinned fruits like walnuts. Natural zeolite of the clinoptilolite type with a purity of 99% was obtained from a mine located in Semnan, Iran. (It should be noted that to reduce errors in the experiments, three duplicate columns were constructed).

Since the biological layer needed to form in the biosand filter, the biosand filter was given some time to do this, and this took almost 30 d. (For a better, quicker forming of the biological layer, microorganisms were added to the water synthesis.) This layer could be effective in removing contaminants. After one month of operation time, the tests started in April. The initial flow rate was measured at around 0.5 L/min, and the pilot produced around 13 L of water each time. It should be noted that the pause time (the resting time) for the biosand was set at 6 h.

2.2. Water synthesis

In this study, runoff purification was examined. To this end, pollutants from Tehran's runoff were measured to obtain the input data shown in Table 1.

Based on Table 1, the sample used for this study was simulated. The pollutants and their values, along with alternative chemical compositions and their values, are shown in Table 2. As can be seen, the influent lead was considered variable because of its potential hazard and its changing value in the runoff.

The differences between the amounts of pollutants in Tables 1 and 2 can be explained as follows:

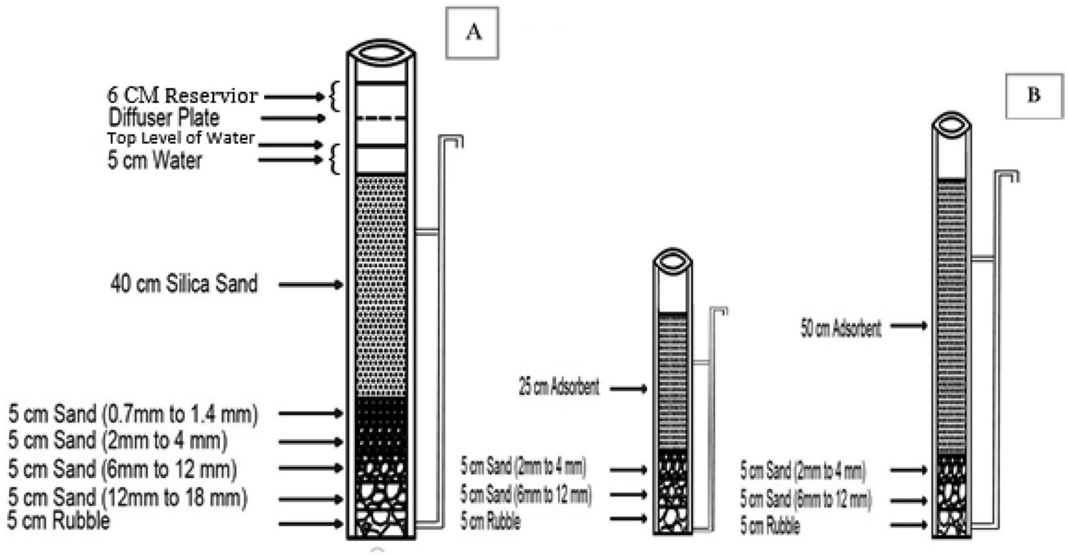

Fig. 1. (A) The biosand filter pilot and (B) The adsorbents column pilot.

Table 1
Pollutants in Tehran's runoff water

Sampling Location	T_p (mg/L)	PO_4^-(mg/L)	NH_4^+(mg/L)	NO_3^- (mg/L)	COD (mg/L)	Fe (µg/L)	Zn (µg/L)	Pb (µg/L)	Cu (µg/L)
Razan Area	4.8	3.5	4	4.3	396	220	–	25	–
Rusty Galvanized Roof	0.49	0.1	4.5	0.6	–	110	850	5	5
Not Rusty Galvanized Roof	0.8	0.11	3.8	0.46	–	250	440	5	5
KNTU University Asphalt Surface	1.17	0.1	0.88	1.48	–	–	–	6	6
Valiasr Street Asphalt Surface	1.6	0.6	1.18	2.6	–	180	780	74	14
Khashayar Area	4.7	1.4	4.15	1.64	277	300	40	5	20
Niyavaran Street	4.98	0.49	2.2	1.26	250	350	–	44	–

- A number of pollutants were not synthesized because their levels were lower than the allowed limits.
- The values of some pollutants were considered only slightly higher than the original safe values.
- An amount of 12 mg/L of phosphate was considered roughly equivalent to 4 mg/L of total phosphorus [49].

To synthesize the water, tap water from the laboratory was used to prepare the test sample as follows: First, the alternative compounds (see Table 2) were added to 0.5 L of water and mixed well using a magnetic stirrer to obtain a homogeneous combination. Next, the solution was diluted to the desired volume, which was generally 50 L. Since the tap water had very little bacterial content, microorganisms were introduced by adding sewage (0.1%v/v), which resulted in influent total coliform and *E. coli* concentrations of 2.0 × 104 ± 220 and 1,820 ± 258 MPN/100 mL, respectively, in the solution [48].

Table 2
Quantities for the pollutants and their alternative chemical compositions

Pollutant	Pollutant concentration	Alternative chemical composition	Alternative chemical composition concentration
Phosphate	12 mg/L	($NaH_2PO_4{\cdot}2H_2O$)	70.19 mg/L
Ammonium	5 mg/L	($(NH_4)2SO_4$)	34.18 mg/L
Iron	4.0 mg/L	($FeCl_3{\cdot}6H_2O$)	94.1 mg/L
	10 µg/L		013.0 mg/L
Lead	110 µg/L	($Pb(NO_3)_2$)	14.0 mg/L
	210 µg/L		270 mg/L
COD	400 mg/L	(CH_3COONa)	400 mg/L

2.3. Methodology

To perform the experiments, the samples containing the pollutants were first made as explained in the previous section and then introduced to the pilot. The final concentrations of pollutants were then measured, with each test run being repeated twice. A third test was also conducted in case of error or substantial discrepancies. Also, during each run, five samples were taken at various time intervals to ensure the values of the samples were sufficiently close to each other (e.g., the standard deviation of the samples for run 1 was 0.13). The concentrations of some pollutants—including ammonium, iron, phosphate, and COD—were measured using spectrophotometry, while the concentration of lead was measured by ICP. The sample was first entered into the biosand filter, with the filtration time and pause time being 1 h and 6 h respectively, and then into the adsorbent column. Since an activated carbon adsorbent is more expensive than a zeolite adsorbent [50], the activated carbon was placed after the zeolite adsorbent so less contamination would reach it and extend the time before it became saturated. In order to examine the effective factors on pollutant removal and system optimization, the height of the adsorbent column was considered variable. Similarly, the lead concentration was also considered variable due to its harmful effects and in accordance with the discussion in the previous section. By using the Design-Expert® software, the response surface methodology was used based on the Box-Behnken method to evaluate the effects of the independent variables on the efficiency of the system and determine the optimum configuration. The Box-Behnken design is an experimental design for response surface methodology (RSM), and it requires only three levels to run an experiment. This special three-level design does not contain any points at the vertices of the experiment region [51]. For this reason, the column heights for the zeolite and activated carbon adsorbents, along with the initial lead level, was changed at three levels, as shown in Table 3. The selection of the upper and lower levels for each variable was performed in such a way that, in addition to enabling the investigation of a wide range of parameters and variables, laboratory equipment could provide values close to the previous research. Afterwards, 17 runs were designed randomly by BBD using different operating conditions (Table 4). As a result, the efficiency of the system at removing ammonia, lead, phosphate, COD, and iron could be examined, and the results are shown in Table 4.

Table 3
Experimental ranges and levels for the independent variables

Variables	Levels		
	1–	0	1+
Influent Lead (µg/L)	10	110	210
Zeolite Height (cm)	0	25	50
Activated Carbon Height (cm)	0	25	50

3. Results

3.1. Model development

The Design-Expert® software was used for regression and graphical analyses of the obtained data. The results from the 17 experiments (runs) are given in Table 4. Based on the results, the most appropriate model was fitted, namely the one that had the highest correlation between observed and predicted values, as well as the one with the highest R^2 and lowest P-values. The best-fitting models are quadratic for each of the four responses (effluent ammonium, lead, phosphate and COD), and these are formulated in Eqs. (1)–(4):

$$\text{Sqt}\left(\text{Effluent}NH_4^+\right) = -0/0014C_0 - 0/06H_Z - 0/015H_G - 0/00017H_ZH_G + 0/000006C_0^{\ 2} + 0/0005H_Z^{\ 2} + 0/00006H_G^{\ 2} + 1/74 \quad (1)$$

$$Ln(EffluentPb) = 0/031C_0 - 0/073H_Z - 0/019H_G - 0/00006C_0^{\ 2} + 0/0009H_Z^{\ 2} - 0/46 \quad (2)$$

$$\left(\text{EffluentPO}_4^-\right) = -0/2H_Z - 0/037H_G + 0/0019H_Z^{\ 2} + 8/3 \quad (3)$$

$$\left(\text{Effluent COD}\right)^{1/5} = -5/13H_Z - 207/42H_G + 0/06H_ZH_G + 2/67H_G^{\ 2} + 3806 \quad (4)$$

Table 4
The box-behnken experimental design with experimental responses for ammonia, lead, phosphate, COD, and iron

Run	Influent Lead (µg/L)	Zeolite Height (cm)	Activated Carbon Height (cm)	Effluent Ammonium (mg/L)	Effluent lead (µg/L)	Effluent Phosphate (mg/L)	Effluent COD (mg/L)	Effluent Iron (mg/L)
1	110	25	25	0.1	4.0	3.5	38	<0.1
2	10	25	0	0.4	3.0	3.8	238	<0.1
3	110	50	0	0.06	7.2	3.1	233	<0.1
4	110	25	25	0.15	6.1	3.1	36	<0.1
5	210	25	50	0.08	4.5	3	18	<0.1
6	110	0	50	1.2	3	5.8	24.5	<0.1
7	110	25	25	0.2	1.8	3.2	37	<0.1
8	10	0	25	1.9	0.7	7.1	44	<0.1
9	10	50	25	0.05	0.09	1.87	20	<0.1
10	210	25	0	0.5	7.5	3.98	239	<0.1
11	210	50	25	0.06	3.9	1.87	19	<0.1
12	110	0	0	2.8	14	9.21	243	<0.1
13	210	0	25	1.9	8	7.4	44	<0.1
14	110	50	50	0.01	1.4	0.8	12	<0.1
15	110	25	25	0.11	1.6	3.5	36	<0.1
16	10	25	0	0.8	0.1	2.9	15	<0.1
17	110	50	0	0.12	2.4	4	32	<0.1

where C_0 is the influent lead (µg/L), H_z is the zeolite column height (in cm), and H_G is the activated carbon column height (in cm). The values of R^2_{adj} and R^2 were closer to 100% than with the other models, so the quadratic model was chosen.

According to Table 5, based on an analysis of the variance (ANOVA—the assumptions were checked, and the residuals were normally distributed and the variances were equal), all of the four models showed statistically significant *F*-values over the *F*-critical value at a 95% confidence level, and the *P*-values were lower than 0.05 at a 95% confidence level. The correlation coefficient of the model (R^2) for responses in ammonium, lead, phosphate and COD levels was 99.5, 89.6, 96.3 and 99, respectively. The R^2_{adj} values of the models are also close to the R^2 values, indicating a high correlation between the models' results and the observations. This means that the regression model provides an excellent explanation of the relationship between the independent variables and the responses. The non-significant value for the lack of fit (more than 0.05) showed that the quadratic model was valid for the present study, and the model fit the data very well [52,53].

3.2. Analysis of the results

In these tests, contaminants were first subjected to the biosand filter where the four actions (mechanical trapping, predation, adsorption, and natural death) occurred, reducing some of the contaminants. Using fine silica sand increased the surface adsorption. The remaining contaminants then made it through to the adsorbent columns, where they were (hopefully) adsorbed. Because of the inherent characteristics of the materials, the adsorption abilities of the activated carbon and zeolite are different. The ability of each adsorbent and the effect of column height will also be discussed. Fig. 2 shows the effect of column heights for the activated carbon and zeolite in the form of a 2D plot. As demonstrated in Fig. 2, increasing the height for zeolite leads to further ammonium removal. This is more distinctive at a lower height for activated carbon. Generally, both adsorbents are effective at ammonium removal, and increasing the height enhances the rate of removal. However, zeolite appears to be stronger than activated carbon at this, and zeolite exchangeable ions could lead to a better removal. In addition, the red points show that if adsorbents had not been used, the biosand filter alone could not have reduced pollutants to acceptable levels. The slope from the adsorbent graph is extremely steep at heights from 0 to 25 cm, but it then decreases for heights of 25–50 cm, and it is almost non-existent at greater heights. Thus, it can be concluded that the impact of greater height diminishes as it approaches 50 cm, and heights above 50 cm are not required. It should be noted that because there was a set of pollutants rather than a single pollutant, the removal rate was lower.

Fig. 3 shows the interactive effect of column heights for the activated carbon and zeolite on lead removal in 2D plots (considering the initial lead level to be 110 µg/L). As it stands, increasing the height of zeolite leads to further

Table 5
Analysis of variance for the four proposed models

	Responses	Model	Residuals	Lack of fit	Pure error	Total
Sum of Square	R1	3.74	0.016	0.004	0.011	3.75
	R2	30.25	3.50	1.55	1.95	33.75
	R3	73.10	2.77	2.28	0.49	75.88
	R4	37,890,000	5106	3479	1626	37,900,000
DF	R1	7	9	5	4	16
	R2	5	11	7	4	16
	R3	3	13	9	4	16
	R4	4	12	8	4	16
Mean Square	R1	0.53	0.0017	0.0008	0.0027	–
	R2	6.05	0.32	0.22	0.49	–
	R3	24.37	0.21	0.25	0.12	–
	R4	9,473,000	425.53	434.93	406.75	–
F–Value	R1	308.40	–	0.32	–	–
	R2	19.02	–	0.45	–	–
	R3	114.16	–	2.06	–	–
	R4	22262	–	1.07	–	–
P–Value	R1	<0.0001	–	0.87	–	–
	R2	<0.0001	–	0.82	–	–
	R3	<0.0001	–	0.25	–	–
	R4	<0.0001	–	0.5	–	–
R^2	R1	99.5		–	–	–
	R2	89.6	–	–	–	–
	R3	96.3	–	–	–	–
	R4	99	–	–	–	–
R^2_{adj}	R1	99.2	–	–	–	–
	R2	84.9	–	–	–	–
	R3	95.5	–	–	–	–
	R4	99	–	–	–	–

lead removal, but this is more evident at a lower height for activated carbon. Generally speaking, both adsorbents are effective at lead removal, and height does enhance their removal rates. However, zeolite is again better than activated carbon at lead removal, because it reaches the same efficiency as activated carbon at lower heights, and zeolite exchangeable ions could lead to better removal. The red points also again show that if adsorbents had not been used, the biosand filter alone would not have reduced lead pollutants to an acceptable level. We also again see how the slope from adsorbents graph is extremely steep at heights from 0 to 25 cm before easing off for heights from 25 to 50 cm and becoming almost level at greater heights (35–50 cm). Heights above 50 cm had no major influence on lead removal. It should also be noted that a low height for the zeolite along the biosand filter can reduce lead pollutants to an acceptable level, and this shows the positive impact of the biosand filter in removing lead (i.e., the adsorption process occurs). It should be noted that because there was a set of pollutants rather than a single pollutant, the removal rate was lower.

Fig. 4 shows the interactive effect of column heights for activated carbon and zeolite on phosphate removal in the form of a 2D plot. As can be seen, increasing the height of zeolite leads to better phosphate removal, which is also evident at a greater height for activated carbon. Both adsorbents seem effective at phosphate removal, and increasing height generally increases the removal rate. However, zeolite is again stronger at phosphate removal, with it demonstrating a better efficiency at lower heights, and zeolite exchangeable ions could lead to better removal. The slope from the graph in the direction (axis) of both

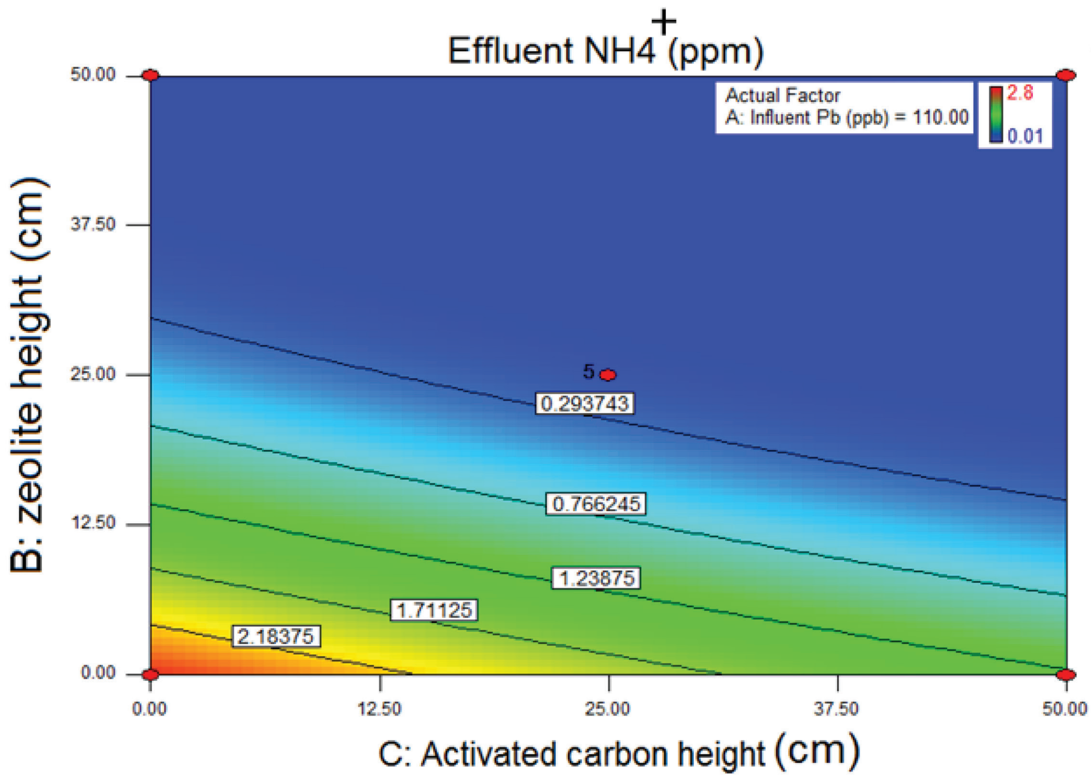

Fig. 2. Contour plot for the effect of column heights for activated carbon and zeolite on NH_4^+ removal.

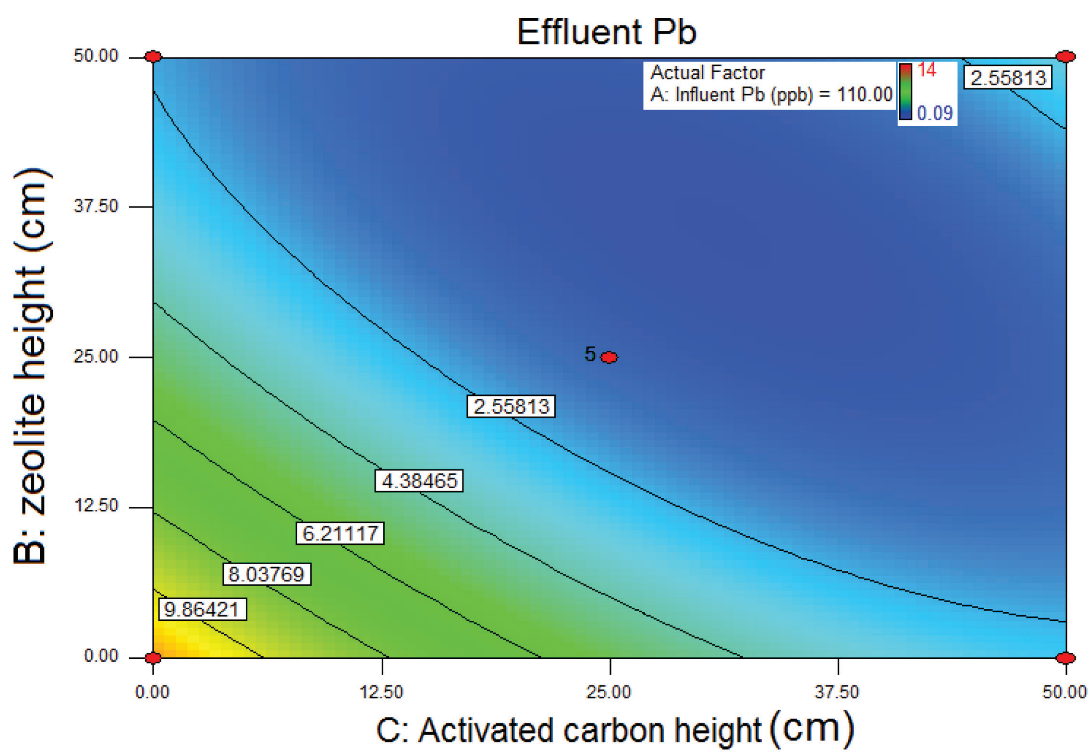

Fig. 3. Contour plot for the effect of the column heights for activated carbon and zeolite on lead removal.

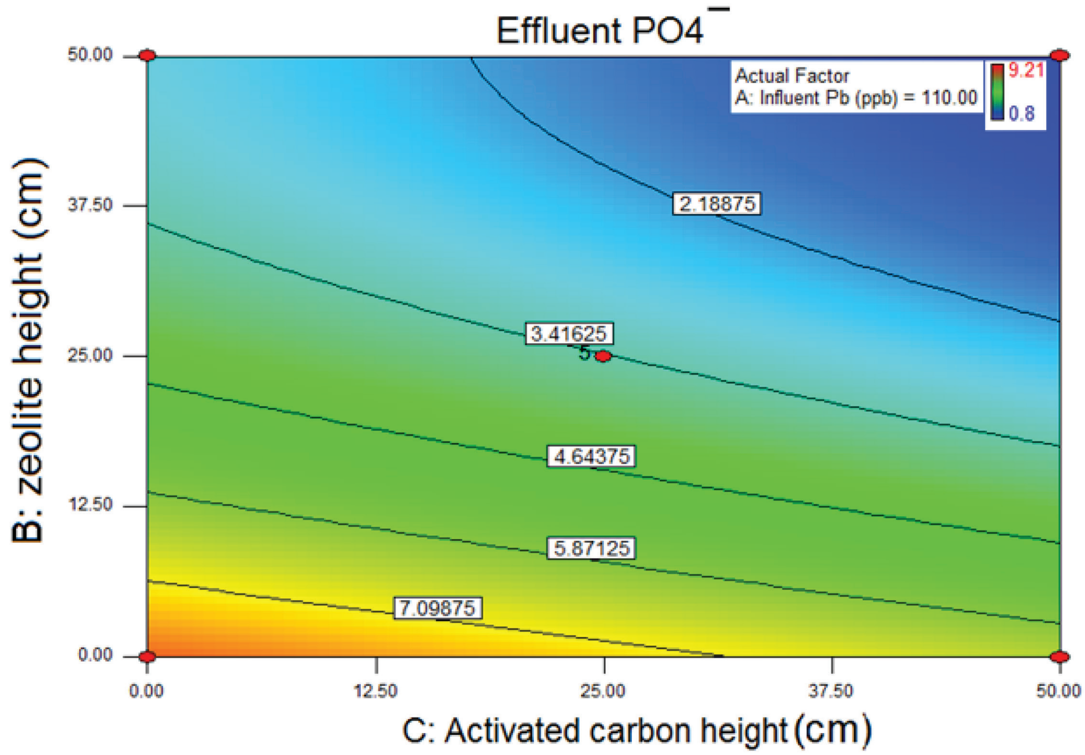

Fig. 4. Contour plot for the effect of the column heights of activated carbon and zeolite on PO_4^+ removal.

adsorbents is almost constant and does not approach zero, indicating that further increasing the adsorbents' heights can also have a positive effect on phosphate removal. Again, because a set of pollutants were used rather than a single pollutant, the removal rate was lower.

Fig. 5 shows the interactive effect of column heights for activated carbon and zeolite on COD removal in the form of a 2D plot. For a better view of the diagram, the COD concentration to the power of 1.5 was used rather than its actual concentration. As it stands, the column height for zeolite does not have a significant impact on COD removal, as the slope in the graph along the zeolite height axis (B) is almost nonexistent, with COD concentration only slightly reducing at greater heights. However, the graph along the activated carbon height axis (C) is extremely steep, which shows the marked effect of activated carbon on COD removal. In addition, the slope of this graph is very steep for heights from 0 to 25 cm, but it levels off at heights from 25 to 50 cm. It can therefore be concluded that the slope will be almost zero, or possibly even negative, at greater heights, indicating that heights above 50 cm will not have any major influence on COD removal. It should be mentioned that the amount of COD was also decreased by the biosand filter.

Graphs 6 and 7 are intended to establish the impact of lead on the experimental results. As shown in Fig. 6, the impact of lead concentration on the removal of ammonium pollutants is negligible, since the slope of the graph along the A axis is almost nonexistent, with it increasing only slightly at high lead concentrations. This slight increase can be attributed to the creation of competitive conditions between pollutants for adsorbance by the zeolite [54]. It should be noted that influent lead also has almost zero effect on the graphs for phosphate and COD.

Fig. 7 shows the interactive effect of influent lead and column heights for the adsorbents on lead removal in the form of a 3D plot. Increasing the influent lead clearly causes the effluent lead to increase, as can be seen in Fig. 7. In addition, an increase in the adsorbents' heights results in better lead removal, with it being more evident at higher concentrations because the slope of the graph is steeper. It can be inferred from Fig. 7 that 15 cm of zeolite combined with 25 cm of activated carbon is necessary at higher lead concentrations. More specifically, increasing the height for activated carbon leads to better lead removal, and this effect is more visible at higher lead concentrations. It can be concluded from Fig. 7 that 25 cm of zeolite performs adequately, even at high lead concentrations.

According to the results and diagrams, one could conclude that a portion of the contaminants is first biodegraded by the biological layer that has formed on the surface of the biosand filter. This is then absorbed by the sand grains, and the contaminants are reduced, as can be seen in Figs. 2–7. The water then flows through the adsorbent columns, where physical and chemical adsorption reduces the contaminants to an acceptable level. It is clear that the combination of the biosand filter with the activated carbon and zeolite adsorbents leads to improved final results compared to the biosand filter alone [55,56]. It also performs better at removing COD and phosphate when compared to the hybrid system described in the study of Zhang et al. [57]. However, the addition of iron scraps could help remove

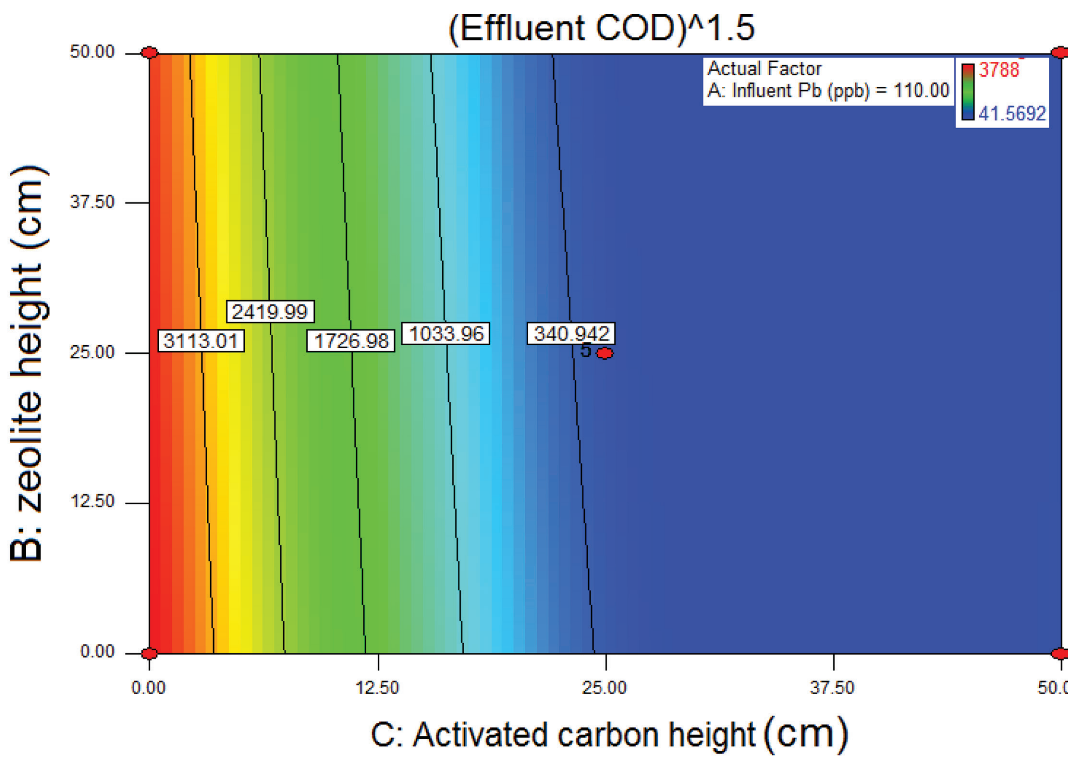

Fig. 5. Contour plot for the effect of the column heights of activated carbon and zeolite on COD removal.

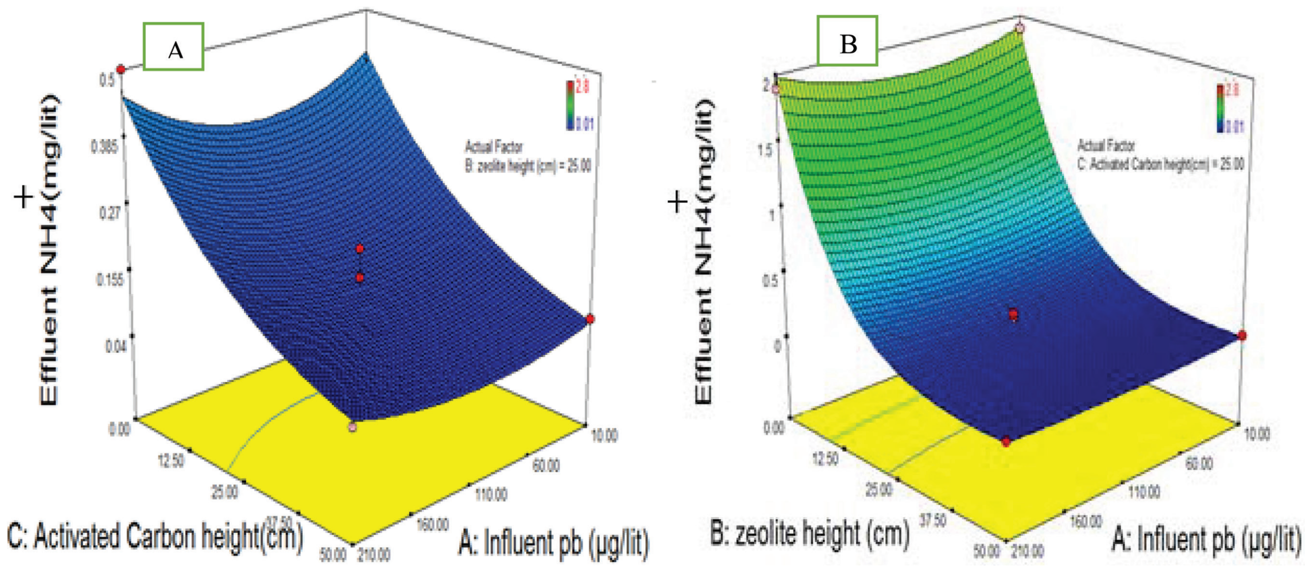

Fig. 6. (A) A 3D plot showing the effect of influent lead concentration and activated carbon column height on ammonium removal; and (B) A 3D plot showing the effect of influent lead concentration and zeolite column height on ammonium removal.

other contaminants, particularly those contaminants that are not addressed in this study (e.g., arsenic) [57].

3.3. Optimization

As shown in the previous section, the pollutants were decreased to acceptable levels by the proposed system. In order to achieve greater efficiency in the removal of pollutants, thus enabling a more cost-effective and efficient system, software was used to try to optimize the system. In order for this to happen, the optimal range for each parameter and variable and their degrees of importance were determined. The initial lead concentration needed to be in standard range due to uncertainty over the level of influent

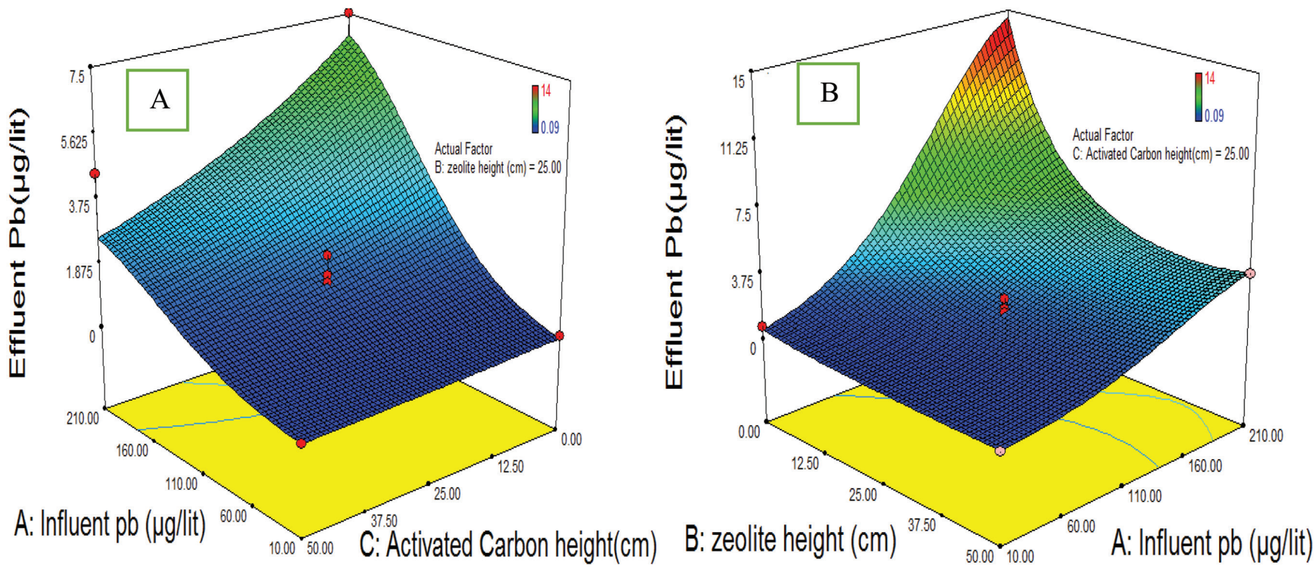

Fig. 7. (A) 3D Plot for the effect of influent lead and column height for activated carbon on lead removal, and (B) 3D Plot for the effect of influent lead and column height for zeolite on lead removal.

lead. Since the system is intended for household treatment, it is better for it to occupy a minimal space, so the height of the adsorbent columns was considered for minimization. In addition, activated carbon adsorbent is expensive [50], so this is taken more into account than zeolite, so the system will be more cost-effective. Effluent parameters should also be minimal, or at least below the allowed limits, so as to achieve maximum efficiency. Based on the considered circumstances, the best solution was generated by the software. The results showed that 33 cm of zeolite together with 26 cm of activated carbon has a utility of approximately 74.4%, which is suitable, as well as high utility and parameters for ammonium, lead, phosphate and COD at 0.06 mg/L, 3 µg/L, 2.5 mg/L and 12 mg/L, respectively. It should be noted that influent lead had the highest value.

Since the tests were carried out in fixed laboratory conditions, a change in other parameters, such as temperature or pH, could affect the performance of the system. Runoffs generally have a neutral pH, but their temperature could change with the ambient temperature (e.g., because of the season and/or region). This could affect the biological activities and the adsorption process, so it is recommendable to install the system in a closed environment (e.g., inside a house) where temperatures will be relatively normal (e.g., around 25°C).

Sources of natural zeolite are absent in many countries, and it may be near impossible to obtain activated carbon, so optimized systems require continued investigation. To this end, the optimal level of activated carbon and zeolite adsorbents is defined as zero. As can be seen in Table 6, in the absence of zeolite, pollutants are reduced to an acceptable level, although the ammonium and lead pollutants have reached the allowed limit. If activated carbon is not used, COD pollutants are only reduced by about 50%. This reduction is also due to the presence of the biosand filter, but COD levels are reduced better with activated carbon. It thus follows that regions with low COD levels have no specific need to use activated carbon, while in regions where zeolite is inaccessible, using activated carbon alone can reduce pollution to acceptable levels. Finally, in regions with high COD levels but where activated carbon is not accessible, zeolite adsorbent is suitable, but it will require another complementary adsorbent to remove COD.

To confirm the results, the optimized system was tested in real situations (with rainwater runoff). The amounts of ammonium, lead, phosphate, COD, and iron pollutants were measured before and after entering the system, as shown in Table 7. As it stands, pollutant removal performs well, with the final concentration being slightly higher than the predicted value, but this is negligible. This slight difference could be due to the presence of other pollutants–such as copper, zinc, et cetera–causing competition for adsorption and making the removal of the contaminants more challenging. In addition, the turbidity and TSS were extremely high in the real sample but not in the synthesized sample. To reduce this difference, the actual sample could be first kept at sedimentation conditions to decrease the

Table 6
The optimal system in different situations

	Zeolite height	GAC height	$C_e(NH_4^+)$	$C_{e(pb)}$	$C_e(PO_4^-)$	C_e(COD)
No Limits	33	26	0.06	3	2.5	12
Lack of Zeolite	0	48	1.2	6	6.5	12
Lack of GAC	43	0	0.11	5	3	234

Table 7
System performance compared with actual conditions

Pollutant	Influent	Effluent	Predicted effluent	Removal percentage	Predicted removal percentage
Ammonium (mg/L)	2.5	0.08	0.06	96.8	98.8
Lead (µg/L)	50	5	0.38	90	99.2
Phosphate (mg/L)	10	3	2.5	70	78
COD (mg/L)	600	41	12	93	97
Iron (mg/L)	1.01	0.25	–	75.2	–

TSS and turbidity before entering it into the system. All the above factors could explain the differences between the predicted final concentration and the measured final concentration. It should be noted that if other contaminants in the runoff were added to the synthesized sample, and if other parameter such as temperature and the number of pollutants reaching the adsorbent were considered, it could further improve the model.

4. Conclusion

In general, based on the experiments and the analysis of the results, it could be said that increasing the column height for the adsorbents enables better pollutant removal. The column height for the zeolite adsorbent had the greatest impact on the removal of ammonium and phosphate. The column height for activated carbon had the greatest impact on COD removal, while zeolite showed an insignificant ability to remove this pollutant. Lead pollutants were effectively removed by both adsorbents, with the two adsorbents having a high and almost identical level of efficiency, although zeolite was observed to be slightly more effective than activated carbon at this. Increasing the influent lead caused the effluent lead to increase, but it had a negligible effect on other pollutants.

In order to achieve an efficient and affordable system, the system was optimized through software. Column heights of 33 cm and 26 cm were considered to be optimal for zeolite and activated carbon, respectively. Therefore, to supply the water for a village or in other critical conditions, two consecutive columns could be used. This first column should comprise 55 cm of sand and 5 cm of water, while the second column should contain 26 cm of activated carbon and 33 cm of zeolite (from bottom to top). At the bottom of each column, two layers of sand should be considered for separation and drainage. The second column should be placed lower than the first column, so water can easily flow into the second column.

5. Recommendations for future studies

Suggestions for further research include:

- Identify cost-effective adsorbents with a good ability to remove COD, so they can be used instead of activated carbon.
- Investigate the lifetime of the proposed optimized system, so adsorbents can be replaced or restored at the right time.
- Investigate the use of the proposed optimal system as a pretreatment before reverse osmosis system, nanofiltration, and so on.
- Optimize other system parameters, such as sand height, flow rate, and the initial concentration of other pollutants (as we did for lead in this study).

Symbols

C_0 — influent lead (µg/L);
H_Z — column height for zeolite (cm);
H_G — the column height for activated carbon (cm).

References

[1] T. Srebotnjak, G. Carr, A. de Sherbinin, C. Rickwood, A global water quality Index and hot-deck imputation of missing data. Ecol. Indic., 17 (2012) 108–119.
[2] J. Waage, C. Yap, S. Bell, Caren Levy, G. Mace, T. Pegram, E. Unterhalter et al., Governing the UN Sustainable Development Goals: interactions, infrastructures, and institutions, Lancet Global Health, 3(5) (2015) 251–252.
[3] M. Falkenmark, Shift in thinking to address the 21st century hunger gap, In Integrated Assessment of Water Resources and Global Change, Springer Netherlands, (2006).
[4] World Health Organization, Progress on sanitation and drinking water: 2015 update and MDG assessment, World Health Organization, (2015).
[5] World Health Organization, UN-water global analysis and assessment of sanitation and drinking-water (GLAAS) 2014 report: investing in water and sanitation: increasing access, reducing inequalities, World Health Organization, (2014).
[6] WHO/UNICEF Joint Water Supply, and Sanitation Monitoring Programme, Progress on drinking water and sanitation: 2014 update, World Health Organization, (2014).
[7] M.D. Sobsey, S. Water, World Health Organization, Managing water in the home: accelerated health gains from improved water supply, World Health Organization, (2002).
[8] J.D. Brookes, C.C. Carey, D.P. Hamilton, L. Ho, L. van der Linden, R. Renner, A. Rigosi, Emerging challenges for the drinking water industry, Environ. Sci. Technol., 48(4) (2014) 2099–2101.
[9] B. Rickert, I. Chorus, O. Schmoll, Protecting surface water for health. Identifying, assessing and managing drinking-water quality risks in surface-water catchments, Protecting surface water for health, Identifying, assessing and managing drinking-water quality risks in surface-water catchments, (2016).
[10] C.H Hsieh, P.D Allen, Evaluation and optimization of bioretention media for treatment of urban storm water runoff, J. Environ. Eng., 131(11) (2005) 1521–1531.

[11] M.A. Mallin, V.L. Johnson, S.H. Ensign, Comparative impacts of stormwater runoff on water quality of an urban, a suburban, and a rural stream, Environ. Monit. Assess., 159(1) (2009) 475–491.
[12] J.S. Choe, K.W. Bang, J.H. Lee, Characterization of surface runoff in urban areas, Water Sci. Technol., 45(9) (2002) 249–254.
[13] N. Kalantari, Z. Sajadi, M. Makvandi, M. Keshavarzi, Chemical properties of soil and groundwater of the Assaluyeh alluvial plain with emphasis on heavy metals contamination, J. Geotech. Geol., 7 (2012) 333–342.
[14] P. Wongsasuluk, S. Chotpantarat, W. Siriwong, M. Robson, Heavy metal contamination and human health risk assessment in drinking water from shallow groundwater wells in an agricultural area in Ubon Ratchathani province, Thailand, Environ. Geochem. Health., 36(1) (2014) 169–182.
[15] A.J. Sisson, P.J. Wampler, R.R. Rediske, J.N. McNair, D.J. Frobish, Long-term field performance of biosand filters in the Artibonite Valley, Haiti, Amer. J. Tropic. Med. Hygiene, 88(5) (2013) 862–867.
[16] W.F. Duke, R.N. Nordin, D. Baker, A. Mazumder, The use and performance of BioSand filters in the Artibonite Valley of Haiti: a field study of 107 households, Rural Remote Health, 6(3) (2006) 570.
[17] M. Kubare, J. Haarhoff, Rational design of domestic biosand filters, J. Water Supply: Res. Technol.-Aqua, 59(1) (2010) 1–15.
[18] P. Earwaker, Cranfield University, Evaluation of Household BioSand Filters in Ethiopia, Silsoe Institute of Water and Environment, (2006).
[19] C.E. Stauber, M.A. Elliott, F. Koksal, G.M. Ortiz, F. A. DiGiano, and M.D. Sobsey, Characterisation of the biosand filter for E. coli reductions from household drinking water under controlled laboratory and field use conditions, Water Sci. Technol. 54(3) (2006) 1–7.
[20] C.E Stauber, The microbiological and health impact of the biosand filter in the Dominican Republic: A randomized controlled trial in Bonao, PhD diss., University of North Carolina at Chapel Hill, (2007).
[21] S.S.K. Tiwari, W.P. Schmidt, J. Darby, Z.G. Kariuki, M.W. Jenkins, Intermittent slow sand filtration for preventing diarrhoea among children in Kenyan households using unimproved water sources: randomized controlled trial, Tropic. Med. Int. Health, 14(11) (2009) 1374–1382.
[22] T.L. Lee, Biosand household water filter project in Nepal, PhD diss., Massachusetts Institute of Techno., (2001).
[23] M.A. Elliott, C.E. Stauber, F. Koksal, F.A. DiGiano, M.D. Sobsey, Reductions of E. coli, echovirus type 12 and bacteriophages in an intermittently operated household-scale slow sand filter, Water Res. 42(10) (2008) 2662–2670.
[24] R.A. Wuana, F.E. Okieimen, Heavy metals in contaminated soils: a review of sources, chemistry, risks and best available strategies for remediation, ISRN Ecology, (2011).
[25] M. Rabani, A. Ashtiani, A. Sharif, The measurement of heavy metal (Ni. Pb. Hg) pollution in sediments of the Persian Gulf/ operational area Assaluyeh, J.Exp. Pro., (2008) 53–58.
[26] A. Shahriari, Measurement of Heavy Metals (Cd, Cr, Pb, Ni) in the edible tissue of Persian Gulf's fish (Frenzy and Red snapper), JGUMS, 7(2) (2005) 65–70.
[27] K.Y. Hor, J.M.C. Chee, M.N. Chong, B. Jin, C. Saint, P.E. Poh, R. Aryal, Evaluation of physicochemical methods in enhancing the adsorption performance of natural zeolite as low-cost adsorbent of methylene blue dye from wastewater, J. Cleaner Prod., 118 (2016) 197–209.
[28] V.K. Gupta, A. Rastogi, A. Nayak, Adsorption studies on the removal of hexavalent chromium from aqueous solution using a low cost fertilizer industry waste material, J. Colloid Interface Sci., 342(1) (2010) 135–141.
[29] K. Ghebremichael, S. Ergas, N. Alcantar, Enhancement of the performance of a biosand filter using pumice media and natural coagulant dosing, In World Environmental and Water Resources Congress, (2016) 256–264.
[30] C. Noubactep, E. Temgoua, M.A. Rahman, Designing iron-amended biosand filters for decentralized safe drinking water provision, Clean-Soil Air Water, 40(8) (2012) 798–807.
[31] T.K. Ngai, R.R. Shrestha, B. Dangol, M. Maharjan, S.E. Murcott, Design for sustainable development-Household drinking water filter for arsenic and pathogen treatment in Nepal, J. Environ. Sci. Health Part A, 42(12) (2007) 1879–1888.
[32] H. Chiew, M.L. Sampson, S. Huch, S. Ken, B.C. Bostick, Effect of groundwater iron and phosphate on the efficacy of arsenic removal by iron-amended biosand filters, Environ. Sci. Technol., 43(16) (2009) 6295–6300.
[33] C. Noubactep, Metallic iron for safe drinking water worldwide, Chem. Eng. J., 165(2) (2010) 740–749.
[34] C. Noubactep, S. Caré, K. Btatkeu, B. Donald, C.P. Nanseu-Njiki, Enhancing the sustainability of household Fe0/sand filters by using bimetallics and MnO_2, Clean-Soil Air Water, 40(1) (2012) 100–109.
[35] J. Kim, B. Kang, DBPs removal in GAC filter-adsorber, Water Res. 42(1) (2008) 145–152.
[36] D.P. Sounthararajah, P. Loganathan, J. Kandasamy, S. Vigneswaran, Column studies on the removal of dissolved organic carbon, turbidity and heavy metals from stormwater using granular activated carbon, Desal. Water Treat., 57(11) (2016) 5045–5055.
[37] L. Paredes, E. Fernandez-Fontaina, J.M. Lema, F. Omil, M. Carballa, Understanding the fate of organic micropollutants in sand and granular activated carbon biofiltration systems, Sci. Total Environ., 551 (2016) 640–648.
[38] J.N. Sahu, J. Acharya, B.K. Sahoo, B.C. Meikap, Optimization of lead (II) sorption potential using developed activated carbon from tamarind wood with chemical activation by zinc chloride, Desal. Water Treat., 57(5) (2016) 2006–2017.
[39] K. Singh, M. Gautam, B. Chandra, A. Kumar, Removal of Pb (II) from its aqueous solution by activated carbon derived from Balam Khira (Kigelia Africana), Desal. Water Treat., 57(51) (2016) 24487–24497.
[40] D.W. Breck, Zeolite Molecular Sieves, Krieger, (1984).
[41] S.W. An, Y.C. Jeong, H.H. Cho, J.W. Park, Adsorption of NH^{4+}–N and E. coli onto Mg^{2+}-modified zeolites, Environ. Earth Sci., 75(5) (2016) 1–11.
[42] F. Aydın Temel, A. Kuleyin, Ammonium removal from landfill leachate using natural zeolite: kinetic, equilibrium, and thermodynamic studies, Desal. Water Treat., 57(50) (2016) 23873–23892.
[43] I. Nuiģ M. Trgo, and N.V. Medvidoviģ The application of the packed bed reactor theory to Pb and Zn uptake from the binary solution onto the fixed bed of natural zeolite, Chem. Eng. J., 295 (2016), 347–357.
[44] M.K. Zabochnicka-ćwiđek ģ Removal of nitrogen and phosphorous compounds by zeolites and algae, Environmental Engineering IV, CRC Press Ta Francis Group, (2013) 123–128.
[45] J.K. Mwabi, F.E. Adeyemo, T.O. Mahlangu, B.B. Mamba, B.M. Brouckaert, C.D. Swartz, G. Offringa, L. Mpenyana-Monyatsi, M.N.B. Momba, Household water treatment systems: a solution to the production of safe drinking water by the low-income communities of Southern Africa, Phys. Chem. Earth, Parts A/B/C, 36(14) (2011) 1120–1128.
[46] J.N. Sahu, J. Acharya, B.C. Meikap, Response surface modeling and optimization of chromium (VI) removal from aqueous solution using Tamarind wood activated carbon in batch process, J. Hazard. Mater., 172(2) (2009) 818–825.
[47] A.I. Khuri, Response surface methodology and related topics, World Scientific, (2006).
[48] K.R. Tundia, M.M. Ahammed, D. George, The effect of operating parameters on the performance of a biosand filter: a statistical experiment design approach, Water Sci. Technol.: Water Supply, 16(3) (2016) 775–782.
[49] J.F. Kopp, G.D. McKee, Methods for chemical analysis of water and wastes, National Technical Information Service. Springfield, Va., Report No. PB 297686, 32319, (1979) 490.
[50] S. Zhu, D. Yin, N. Gao, S. Zhou, Z. Wang, Z. Zhang, Adsorption of two microcystins onto activated carbon: equilibrium, kinetic, and influential factors, Desal. Water Treat., 57(50) (2016) 23666–23674.

[51] U. Groemping, Cran task view: Design of experiments (doe) & analysis of experimental data, (2016).
[52] D.R. Hamsaveni, S. G. Prapulla, S. Divakar, Response surface methodological approach for the synthesis of isobutyl isobutyrate, Process Biochem., 36(11) (2001) 1103–1109.
[53] A.T. Nair, A.R. Makwana, M.M. Ahammed, The use of response surface methodology for modelling and analysis of water and wastewater treatment processes: a review, Water Sci. Technol., 69(3) (2014) 464–478.
[54] M. Ansari, A. Raisi, A. Aroujalian, B. Dabir, M. Irani, Synthesis of nano-NaX zeolite by microwave heating method for removal of lead, copper, and cobalt ions from aqueous solution, J. Environ. Eng., 141(5) (2014) 04014088.
[55] A. Mangoua-Allali, L.C. Lacina Coulibaly, J.M.P. Ouattara, G. Gourene, Implementation of biosand filters in rural area for drinking water production, Afr. J. Food Sci., 6(24) (2012) 574–582.
[56] R.R. Shrestha, Eco home for sustainable water management: A case study in Kathmandu, Nepal, In International Conference on Sustainable Sanitation: Food and Water Security for Latin America, (2007).
[57] Y. Zhang, Y. Cheng, C. Yang, W. Luo, G. Zeng, L. Lu., Performance of system consisting of vertical flow trickling filter and horizontal flow multi-soil-layering reactor for treatment of rural wastewater, Bioresour., Technol., 193 (2015) 424–432.