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Artículos Científicos

Zeolitas nativas en el tratamiento de agua residual doméstica

Native Zeolites in Domestic Wastewater Treatment

Zeólitas nativas no tratamento de águas residuais domésticas

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Resumen

En esta investigación zeolitas de río y de cerro fueron evaluadas para ver su potencial en el tratamiento de aguas residuales. En primer lugar, se diseñó y construyó un sistema experimental de tres biorreactores de lecho fijo de flujo ascendente (BLFFA) con una altura de 1.5 metros y diámetro de cuatro pulgadas, seguidamente se evaluó el potencial de las zeolitas nativas y una testigo comercial en el tratamiento de aguas residuales. Se arrancaron los BLFFA con un diseño factorial 3 x 3, operando con zeolita de río, zeolita de cerro y zeolita comercial (control), todas con partículas de ¼ de pulgada, utilizando tres diferentes alturas de lecho (0.75, 0.90 y 1.10 m) y tres tiempos de retención hidráulica (6, 12 y 24 horas). Se evaluaron las variables de respuesta mediante un análisis de varianza multifactorial y para discriminar entre las medias se empleó el método de diferencia mínima significativa (LSD) de Fisher ($p < 0.05$, 95 % de confianza). La zeolita de cerró presentó (promedio, $N = 10$) las mejores características fisicoquímicas con densidad real y aparente de 2700 y 1470 kg/m^3 respectivamente, peso específico de 2240 kg/m^3, porosidad de 62 %, absorción de 15.20 %, solubilidad en ácido clorhídrico de 29.96 %, pH de 7.4 y conductividad eléctrica de 70 mS/cm. Al agua residual con las que se desarrollaron los tratamientos se le aplicó previo al experimento un tratamiento primario (rejilla, desarenador, desnatador y fosa séptica) y presentó valores (promedio, $N = 9$) de temperatura de 25.26 °C, pH de 8.63, sólidos disueltos totales con 950.50 mg/L, color de 1305.10 UC, turbiedad 96.37 UNT y demanda química de oxígeno de 373 mg/L. De las zeolitas evaluadas ($N = 81$) en el BLFFA, la que presentó el mejor tratamiento fue la zeolita de cerro operando con una altura de lecho de 0.9 m y con 24 horas de tiempo de retención hidráulica; obtuvo las mejores eficiencias de remoción con 71.42 % en sólidos disueltos totales, 84.34 % para turbiedad, 96.33 % para color y 97.28 % para la demanda química de oxígeno. La altura óptima en los BLFFA fue 0.9 m, factor importante de evaluar, ya que a mayor altura se aumentan los costos y las eficiencias no son relevantes para justificar la inversión y a menor altura cae el rendimiento en calidad del agua tratada. En conclusión, las zeolitas nativas cuentan con propiedades idóneas para ser utilizadas en el tratamiento de agua residual doméstica y podemos recomendar la implementación de los BLFFA en el tratamiento secundario de efluentes domésticos en sistemas descentralizados del sureste de México como una alternativa viable en el tratamiento de sus aguas residuales domésticas.

Palabras clave: biorreactor de lecho fijo de flujo ascendente, demanda química de oxígeno, eficiencia de remoción, tiempo de retención hidráulica.

Abstract

Natural zeolites have a high purification capacity in wastewater due to their porosity, specific surface, cation exchange capacity and they are a product available in nature. In Tabasco, Mexico, river and hill zeolites were evaluated to see their potential in wastewater treatment. First, an experimental system of three upflow fixed-bed bioreactors (UFBB) with a height of 1.5 meters and diameter of 4 inches was designed and built, then the potential of native zeolites and a commercial control in the field were evaluated. sewage treatment. The UFBB were started with a 3x3 factorial design, operating with river zeolite, hill zeolite and commercial zeolite (control), all with ¼ inch particles, using three different bed heights (0.75, 0.90 and 1.10 m) and three times. hydraulic retention (6, 12 and 24 hours). The response variables were evaluated by means of a multifactorial ANOVA and to discriminate between the means the Fisher's least significant difference (LSD) method ($p < 0.05$, 95 % confidence) was used. The closed zeolite presented (average, $N=10$) the best physicochemical characteristics with are real and apparent density of 2700 and 1470 kg/m^3 respectively, specific weight 2240 kg/m^3, porosity of 62 %, absorption of 15.20 %, solubility in hydrochloric acid of 29.96 %, pH of 7.4 and electrical conductivity of 70 mS/cm. The residual water with which the treatments were developed was applied prior to the experiment a primary treatment (grid, sand trap, skimmer and septic tank) and presented temperature values (average, N = 9) of 25.26 ° C, pH of 8.63, Total dissolved solids with 950.50 mg/L, color of 1305.10 UC, turbidity 96.37 NTU and chemical oxygen demand of 373 mg/L. Of the zeolites evaluated (N = 81) in the UFBB, the one that presented the best treatment was the hill zeolite operating with a bed height of 0.9 m and with 24 hours of hydraulic retention time, obtaining the best removal efficiencies with 71.42 % in total dissolved solids, 84.34 % for turbidity, 96.33 % for color and 97.28 % for the chemical oxygen demand, followed by the commercial zeolite that had very similar responses and finally the river zeolite treatment that also presents good results in comparison with other means of support. The optimal height in the UFBB was 0.9 m, an important factor to evaluate since the higher the elevation costs are increased and the efficiencies are not relevant to justify the investment and the lower the performance in quality of the treated water falls. Finally, native zeolites have ideal properties to be used in the treatment of domestic wastewater and we can recommend the implementation of UFBB in the secondary treatment of domestic effluents in decentralized systems in the southeast of Mexico as a viable alternative in the treatment of their domestic wastewater.

Keywords: upflow fixed bed bioreactor, chemical oxygen demand, removal efficiency, hydraulic retention time.

Resumo

Nesta pesquisa, zeólitas de rios e morros foram avaliados para ver seu potencial no tratamento de águas residuais. En primer lugar, se diseñó y construyó un sistema experimental de tres biorreactores de lecho fijo de flujo ascendente (BLFFA) con una altura de 1.5 metros y diámetro de cuatro pulgadas, seguidamente se evaluó el potencial de las zeolitas nativas y una testigo comercial en el tratamento de águas residuais. Os BLFFAs foram depenados em esquema fatorial 3 x 3, operando com zeólita de rio, zeólita de colina e zeólita comercial (controle), todos com partículas de ¼ de polegada, usando três alturas de leito diferentes (0,75, 0,90 e 1,10 m). vezes (6, 12 e 24 horas). As variáveis de resposta foram avaliadas por meio de uma análise de variância multifatorial e o método da diferença mínima significativa de Fisher (LSD) foi usado para discriminar as médias (p <0,05, 95% de confiança). A zeólita fechada apresentou (média, N = 10) as melhores características físico-químicas com densidade real e aparente de 2700 e 1470 kg / m3 respectivamente, peso específico de 2240 kg / m^3, porosidade de 62%, absorção de 15,20%, solubilidade em clorídrico ácido de 29,96%, pH de 7,4 e condutividade elétrica de 70 mS / cm. A água residual com a qual os tratamentos foram desenvolvidos foi submetido a um tratamento primário prévio ao experimento (grade, caixa de areia, skimmer e fossa séptica) e apresentou valores de temperatura (média, N = 9) de 25,26 ° C, pH de 8,63 , Total de sólidos dissolvidos com 950,50 mg / L, cor de 1305,10 UC, turbidez 96,37 NTU e demanda química de oxigênio de 373 mg / L. Das zeólitas avaliadas (N = 81) no BLFFA, a que apresentou melhor tratamento foi a zeólita de morro operando com altura de leito de 0,9 me com tempo de retenção hidráulica de 24 horas; obtiveram as melhores eficiências de remoção com 71,42% no total de sólidos dissolvidos, 84,34% para turbidez, 96,33% para cor e 97,28% para demanda química de oxigênio. A altura ótima no BLFFA foi de 0,9 m, fator importante a ser avaliado, visto que quanto maiores os custos de elevação aumentam e as eficiências não são relevantes para justificar o investimento, e menor o desempenho em qualidade das quedas d'água tratadas. Concluindo, zeólitas nativas possuem propriedades ideais para serem utilizadas no tratamento de águas residuais domésticas e podemos recomendar a implementação do BLFFA no tratamento secundário de efluentes domésticos em sistemas descentralizados no sudeste do México como uma alternativa viável no tratamento de suas águas residuais domésticas.

Palavras-chave: biorreator de leito fixo de fluxo ascendente, demanda química de oxigênio, eficiência de remoção, tempo de retenção hidráulica.

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Introduction

The purpose of wastewater treatment is to purify pollutants using unit operations and processes through a combination capable of sanitizing wastewater to be able to reuse it in various activities (Noyola, Morgan-Sagastume, Güereca, 2013). In the world, more than 80% of wastewater is discharged without any treatment, according to a frequently cited estimate (United Nations Educational, Scientific and Cultural Organization [Unesco], 2017). This obviously has negative repercussions on human health, economic productivity, the quality of freshwater resources and ecosystems (Unesco, 2017).

There are several levels to treat wastewater (pre-treatment, primary treatment, secondary treatment, advanced treatment and special). The most studied are secondary (biological). This type of treatment is essential in the development of wastewater treatment trains for construction, operation and maintenance times (National Water Commission [Conagua], 2016a). One of the biological technologies that offers low operating, maintenance, energy reduction and sludge production costs are packed bed reactors: they operate efficiently by the support medium used, achieve optimal adherence of microorganisms and provide maximum surface area to the flow of the liquid under treatment, which can be aerobic or anaerobic (Ganesh, Rajinikanth, Thanikal, Ramanujam and Torrijos, 2010). On the other hand, wastewater treatment with sustainable technologies is based on natural procedures that do not use chemical additives and take advantage of the resources of the areas where they are generated. In addition, because the elimination of the polluting substances in the water is achieved by taking advantage of the aquatic vegetation, the soil, local materials or microorganisms, the initial investment and the maintenance of this type of system are more economical compared to other options. (Morató, Pires y Subirana, 2009).

The efficiency of packed bed reactors with different support media and with various applications has been extensively investigated. For example, Vian, Vigueras, Velasco and Puebla (2020) evaluated an upflow anaerobic reactor packed with sludge bed in the degradation of fruit and vegetable waste and obtained a removal of volatile solids of 67% in four days of time of hydraulic retention (TRH) with a methane (CH4) productivity between 0.5 and 3.6 LCH4 / (L · d) for loads between 1 and 10 g SV / (L · d). The productivity expressed in LCH4 / (g SV consumed · d) was between 0.027 and 0.116 LCH4 / (g SV consumed · d). Previously, Vigueras, Vian, Velasco and Zafra (2016) had designed a rector of the same type that obtained a solids removal of 67% in 12 days with methane productivities of 1.45 LCH4 / (L · d), values proportional to the loads organic substances applied to the system (0.66-2.5 g SV / (L · d).

Fernández et al. (2020), on the other hand, studied a mining-metallurgical effluent that contained Cu, Fe and Pb in a bioadsorbent filter with lignocellulosic fibers (banana, coconut and orange peels). The best removal efficiency for Cu was presented with coconut shells (96.36%). In the case of Fe, the best results were obtained with half coconut shells and half orange (92.05%). And the highest removal of Pb was achieved with a combination of orange and coconut-orange peels (97.34%). In all the treatments, a pH of 7.3 was used, with a TRH of three hours and with a particle size of 0.25 mm.

Yet another example is the research carried out by Pérez et al. (2019), who evaluated the effect of the addition of zeolite in an expanded granular bed (EGSB) anaerobic reactor for the treatment of pig wastewater with high volumetric organic loads. Pérez et al. (2019) found that zeolite did not affect the hydrodynamics of the EGSB. Adjusted to the model of tanks in series, two reactors (R1 and R2) with a volume of 3.04 L, a temperature of 30 ° C, a TRH of 12 hours and a flow of 4 mL / min operated. In the first run, they evaluated the organic load shock strategy with an ascent rate of 6 m / h for 180 days: the reactor with zeolite inclusion reached an efficiency of 80% at 32 kg of chemical oxygen demand (COD) / m3d. In the second run, they evaluated a gradual increase in organic load and an increase in ascent speed up to 10 m / h for a period of 255 days: here an increase in efficiency of up to 90% was observed, the highest speed of organic degradation. Finally, these researchers assure that adding 40 g / L of zeolite to EGSB allows the system to stabilize and it becomes robust to variations in operational conditions, with favorable changes in microbial diversity.

Likewise, Guerrero, Vázquez and Rodríguez (2019) evaluated nitrogen removal in a tubular system conditioned with four layers of zeolite of different grain sizes. The zeolite of the clinoptilolite type was purchased commercially. They bought 1.70mm, 2mm, 2.63mm and 4.75mm grains. The results include that it was possible to remove between 50% and 75% of the total nitrogen and especially the ammoniacal nitrogen. The phosphate content was reduced between 50% and 95%, as well as the biochemical oxygen demand, which could be improved by up to 45%. In sum, between 2% and 8% of the dissolved solids were eliminated, as well as between 35% and 85% of the suspended solids, and it was also possible to improve the electrical conductivity up to 3% and the dissolved oxygen between 2% and 7 %.

Cárdenas and Ramos (2009) evaluated four contact media (seashell, synthetic material, vitrified material and river gravel) in an upflow anaerobic filter (FAFA). The seashell achieved the highest removals (COD of 89.7% and oxygen dioxide demand [BOD] of 87.8%) due to its physical structure, which allowed an adequate microenvironment, and its chemical composition, natural source of alkalinity and micronutrients. In Colombia, Rodríguez, Pinzón and Arámbula (2007) developed and implemented a ceramic packed bed bioreactor where they treated domestic wastewater. They were able to remove 78% of volatile suspended solids, 77% of BOD5, 57% of COD, of 79% total coliforms (TC) and 51% of P with a HRT of pulse and step of 17.75 seconds and 34.577 seconds, respectively.

In addition to all the works already cited, Castillo, Solano and Rangel (2006) studied an anaerobic biofilm on cob cobs, scourer and synthetic polyurethane as support material in a downward flow fixed bed reactor. The evaluated water was taken from the Río Frío Wastewater Treatment Plant (Girón, Santander, Colombia) and another from a lagoon for the treatment of pig excreta (Mesa de los Santos, Santander, Colombia). Castillo et al. (2006) obtained for the biofilm on cob cobs the highest percentages of COD removal (50-75%) and CH4 composition in the biogas (45-75%) when they fed it with organic loads greater than 1500 mg / L of COD with a temperature of 38 ºC, a HRT between 6 and 24 hours and a pH between 6.5-7.5. Finally, Rivas, Nevárez, Bautista, Pérez and Saucedo (2003) studied wastewater to be used in the agricultural industry with a fixed-bed biological system of ascending flow on a semi-pilot scale, with variation in the height of the bed (H) and The diameter of the support medium, keeping the water and air flow constant, observed that the highest removal efficiency of organic matter (91.9%) was obtained when the bioreactor was operated with an H of 0.65 m and a particle size of 1.87 mm.

Despite being a well-studied topic, in Tabasco, Mexico, there are no reports of the use of native zeolites in upflow fixed-bed bioreactors (BLFFA). Although native material is available in the region, its effectiveness in wastewater treatment has not been proven. For this research, an experimental BLFFA system was designed and built to treat primary effluent waters and the efficiency of native zeolites as a support medium was studied. Thus, the physicochemical characteristics of the zeolites and the removal of control parameters such as temperature, pH, total dissolved solids (TDS), color, turbidity and COD, were evaluated to demonstrate the potential of these materials and encourage their use in communities where this type of wastewater treatment process is implemented. The final scope is to demonstrate that native zeolites are feasible to implement in secondary treatment through BLFFAs in the Mexican southeast.

Materials and method

The BLFFA experimental system was built in the Academic Division of Biological Sciences (DACBiol) of the Universidad Juárez Autónoma de Tabasco (UJAT). The wastewater came from the sanitary facilities of that space in Villahermosa, Tabasco. The native zeolites were obtained in stone material banks (from hills and rivers) (17 ° 34 ′ 27.62 ″ N and 92 ° 55 ′ 44.25 ″ W, 17 ° 32 ′ 45.91 ″ N and 92 ° 55 ′ 56.68 ″ W) in Teapa, Tabasco. The zeolites were characterized in the Civil Engineering Laboratory of the Technological Institute of Villahermosa. The water quality of the experiments was characterized in the Water Technology Laboratory of DACBiol.

Characteristics of the experimental system

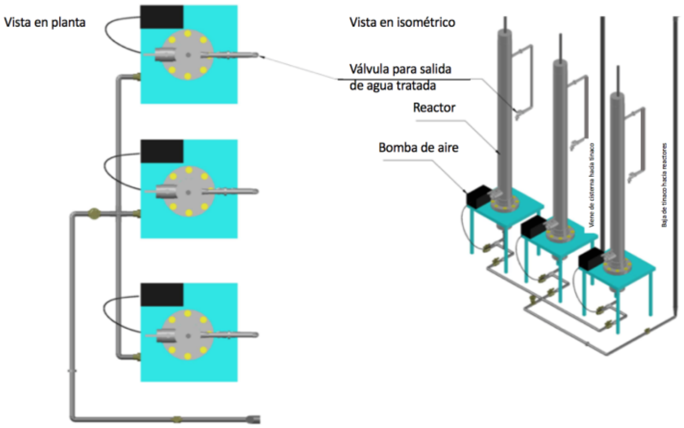
The BLFFA experimental system was installed next to the DACBiol-UJAT wastewater sump. A primary treatment was previously applied to the wastewater used in the experiments to remove sedimentable and suspended solids by means of the DACBiol treatment system, which consists of a screen, a sand trap, a skimmer and a septic tank. This water was transported through ½-inch pipes by a ¼ horsepower pump to a tank with a capacity of 2.1 m3, which, by gravity, fed the BLFFA. The characteristics of this are shown in table 1, figure 1 and figure 2.

Tabla 1. Características y dimensiones de cada biorreactor del sistema experimental

Descripción	Unidad	Valor
Base de cada reactor y altura	m	0.5 x 0.5 x 0.5
Diámetro del plénum	m	0.1016
Altura del plénum	m	0.20
Área de la base del plénum	m^2	0.0081
Volumen del plénum	m^3	0.0016
Diámetro de plato distribuidor con siete toberas y malla	m	0.1016
Altura de la columna de PVC	m	1.50
Volumen efectivo de la columna de PVC de cada reactor	m^3	0.0122

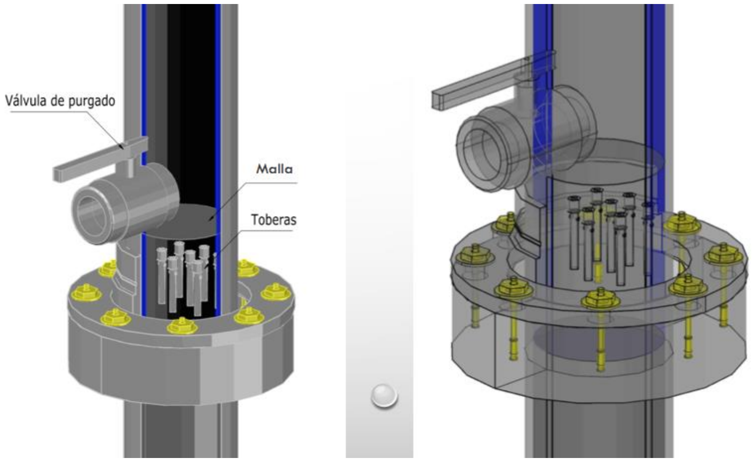
Fuente: Elaboración propia

Figura 1. Configuración del sistema experimental de BLFFA



Fuente: Elaboración propia

Figura 2. Detalle interno del reactor, toberas y malla para no permitir el paso de las partículas, sobre la malla se colocó el lecho empacado de zeolitas.



Fuente: Elaboración propia

Zeolite characteristics

To characterize the zeolites, five samples of 25 kg of river zeolite, cerro zeolite (both crushed from waste in the plant) and commercial zeolite (it was acquired in a water treatment store) were taken. 10 random samples (5 kg) of each zeolite were processed. The preparation included the transfer, reception, registration, drying, sieving, homogenization and storage for its conservation, avoiding contamination of the sample and ensuring precision and accuracy in the analysis. The samples were determined the pH, electrical conductivity (EC), apparent and real density, specific weight, porosity and solubility in hydrochloric acid, as established by Muñoz, Soler, López and Hernández (2015) and the NOM- 021-RECNAT-2000.

Monitoring of the experimental system

Sampling was carried out under NOM-AA-3-1980 after three months of stabilization (February-April 2018). In the experimental phase with the BLFFA (May-October 2018), the control parameters were measured Monday, Wednesday and Friday, taking three simple samples obtained every six hours (6:00 a.m., 12:00 p.m. and 6:00 p.m.). The parameters determined were temperature, pH, TDS, color, turbidity and COD. These were measured at the beginning and end of each experiment, taking the wastewater sample (influent) from the gully and the outlet sample (effluent) after each experiment. The methods used to measure the parameters were taken from Standard Methods for the Examination of Water and

Wastewater (2017): 2550 Temperature (2017), 2540 Total Dissolved Solids and 4500-H + pH Value, measured with the Hanna HI98129 equipment. The turbidity was determined by the EPA method 180.1, TC-300e, ISO 7027, TC-300i, using a Hanna HI 98703 equipment. The color was determined by the 2120 Color method (Standard Methods for the Examination of Water and Wastewater, 2017) , with a Lamotte team. Dissolved oxygen (DO) was determined by the 4500-H + pH Value (Standard Methods for the Examination of Water and Wastewater, 2017). The COD by the EPA 410.4 method (O'Dell, 1993). Each experiment was performed in triplicate, that is, a total of 27 experiments and 81 samples from the experiments. The experimental BLFFA system was operated at room temperature, with an air supply that allowed the reactor to maintain 4.5 ± 0.7 mg / L of DO respectively in the treatments.

Experimental design

In the experimental run, a completely randomized design was used in a factorial arrangement of 3 x 3, with three repetitions for the treatment. Three BLFFAs were used. Each run lasted two months (Rivas et al., 2003) and the factors were the three zeolite types with a particle Ø of ¼ inch of irregular shape, the height of the bed (0.75 m, 0.90 m, 1.10 m) and the TRH (6, 12, 24 hours) with flow rates of 48.6, 24.3, 12.2 L / day respectively in each reactor.

Statistical analysis and efficiency in the experimental system

All the results obtained were evaluated in the statistical program Statgraphics Centurion XVI.II. The quantitative response variables temperature, pH, TDS, turbidity, color and COD of the different types of zeolite treatments, the bed heights and the TRH were compared. To determine if there were significant statistical differences, a multifactorial analysis of variance (Anova) was performed and the method used to discriminate between the means was Fisher's least significant difference (LSD) procedure with a $p < 0.05$ and with a 95% confidence level. The performance of the bioreactor was evaluated based on the results of the concentration differences between the influent and the effluent using the average data of the pollutants, as established Torres, López, Cerino, Vázquez and Comparán (2020).

Results

Zeolite characteristics

Table 2 shows the results of the physicochemical characterization carried out on the zeolites. These present very similar real and apparent density between the three. However, it stands out that cerro zeolite has higher porosity attributes, a very important characteristic in the support of bacterial biomass; it also presented the lowest solubility in hydrochloric acid medium.

Tabla 2. Características de las zeolitas. Valores promedio (*N* = 10)

Parámetro	Zeolita de río	Zeolita de cerro	Zeolita comercial
Densidad real (kg/m^3)	2600	2700	2650
Densidad aparente (kg/m^3)	1550	1470	1600
Peso específico (kg/m^3)	2290	2420	2350
Absorción (%)	14.10	15.20	15.00
Porosidad (%)	53.00	62.00	51.00
CE (mS/cm)	65.00	70.00	67.00
pH (UpH)	7.30	7.40	7.00
Solubilidad en ácido clorhídrico (%)	59.66	29.96	51.30

Fuente: Elaboración propia

Evaluation of control parameters and basic contaminants

The residual water with which the experiment was developed presented (mean ± SD, N = 9) a temperature of 25.26 ± 1.76 ° C, pH of 8.63 ± 0.23, SDT with 950.50 ± 115.05 mg / L, color of 1305.10 ± 141.46 UC, turbidity 96.37 ± 7.79 NTU and COD 373 ± 43 mg / L. This residual water is of medium concentration, since the COD is in the medium-weak range (250-500 mg / L), according to Tchobanoglous, Burton and Stensel (2003). Table 3 shows the results of the effluents in the different treatments of the BLFFA. There it is observed that the treatment with river zeolite presented the lowest COD concentration in the H of 0.90 m and TRH of 24 hours with 23.46 mg / L; Regarding the cerro zeolite, the lowest concentration occurred in the H of 0.90 m and TRH of 24 hours with 9.86 mg / L, and with the commercial zeolite the lowest concentration was raised with the H treatment of 0.90 m and TRH of 12 hours with 11.87 mg / L.

Tabla 3. Contaminantes básicos del agua residual en el efluente de los BLFFA en cada tratamiento. Valores promedio y DE (*N* = 81)

	Parámetro	H = 0.75 m			H = 0.90 m			H = 1.10 m		
		6 h	12 h	24 h	6 h	12 h	24 h	6 h	12 h	24 h
Zeolita de río	Temp. (ºC)	28.93	29.77	28.83	28.87	29.17	28.77	29.53	30.07	29.53
	pH (UpH)	9.67	8.80	8.73	8.00	7.50	7.27	7.93	7.47	7.23
	SDT (mg/L)	993.07	902.30	889.40	830.77	817.77	805.20	827.47	820.40	805.43
	Color (UC)	824.05	777.42	728.34	489.13	235.13	234.10	478.53	237.27	236.27
	Turb. (UNT)	16.82	15.87	14.86	39.27	20.87	8.17	38.67	19.83	7.97
	DQO (mg/L)	48.31	45.58	42.70	112.81	59.95	23.46	111.08	56.98	22.89
Zeolita de cerro	Parámetro	H = 0.75 m			H = 0.90 m			H = 1.10 m		
		6 h	12 h	24 h	6 h	12 h	24 h	6 h	12 h	24 h
	Temp. (ºC)	29.83	30.00	30.40	29.17	28.77	29.17	30.07	29.53	30.07
	pH (UpH)	9.40	8.50	8.47	7.97	6.97	6.90	7.87	6.97	6.90
	SDT (mg/L)	965.80	873.53	873.00	591.13	271.67	266.63	528.00	271.13	266.70
	Color (UC)	820.75	768.39	768.42	470.80	204.43	203.43	470.80	208.97	206.97
	Turb. (UNT)	16.75	15.68	15.68	20.80	3.53	3.43	20.53	3.67	3.60
	DQO (mg/L)	48.12	45.05	45.05	59.75	10.15	9.86	58.99	10.53	10.34

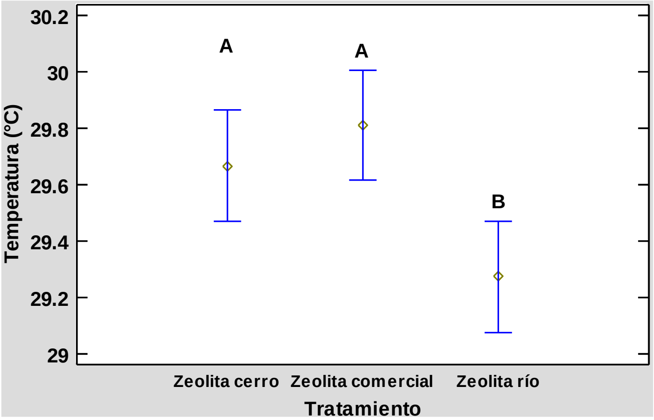
	Parámetro	H = 0.75 m			H = 0.90 m			H = 1.10 m		
		6 h	12 h	24 h	6 h	12 h	24 h	6 h	12 h	24 h
Zeolita comercial	Temp. (ºC)	30.07	30.87	31.53	28.77	29.17	28.77	29.53	30.07	29.53
	pH (UpH)	9.37	8.53	8.43	7.93	6.83	6.77	7.97	6.80	6.83
	SDT (mg/L)	965.53	874.30	873.20	731.47	698.70	424.40	732.60	681.40	412.13
	Color (UC)	820.68	768.37	768.53	476.00	206.90	205.90	475.10	206.83	205.83
	Turb. (UNT)	16.75	15.68	15.68	20.97	4.13	4.33	20.63	4.27	4.23
	DQO (mg/L)	48.12	45.05	45.06	60.23	11.87	12.45	59.28	12.26	12.16

Fuente: Elaboración propia

Temperature

The multifactorial Anova analysis ($p < 0.05$, 95% confidence level) showed that the Zeolite factor has a statistically significant effect on Temperature. The lowest temperature was shown in river zeolite (29.27 ° C), followed by hill zeolite (29.66 ° C) and finally commercial zeolite (29.81 ° C) (figure 3). The Height factor has a statistically significant effect on Temperature. The lowest temperature was shown with the H of 0.9 m (28.95 ° C), followed by the H of 1.1 m (29.77 ° C) and finally with the H of 0.75 m (30.02 ° C) (figure 4). For the TRH factor, none of the factors had a statistically significant effect on Temperature. The lowest temperature was recorded at six hours of TRH (29.62 ° C) (figure 5).

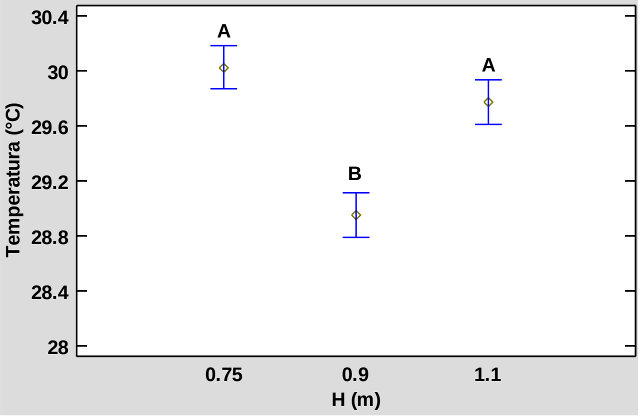
Figura 3. Valores de temperatura en cada Zeolita. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

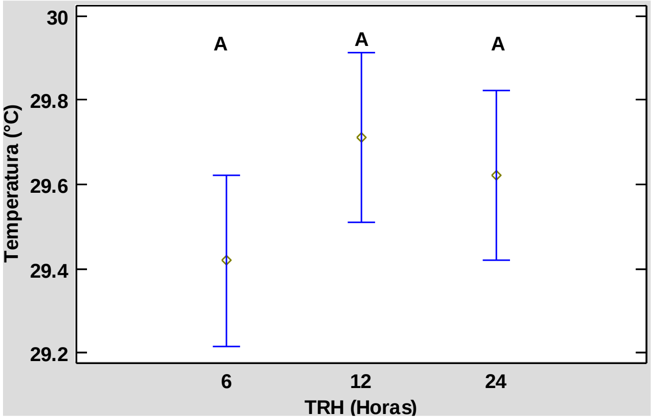
Figura 4. Valores de temperatura en cada H. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Figura 5. Valores de temperatura en cada TRH. Contraste de medias (medias ± EE)



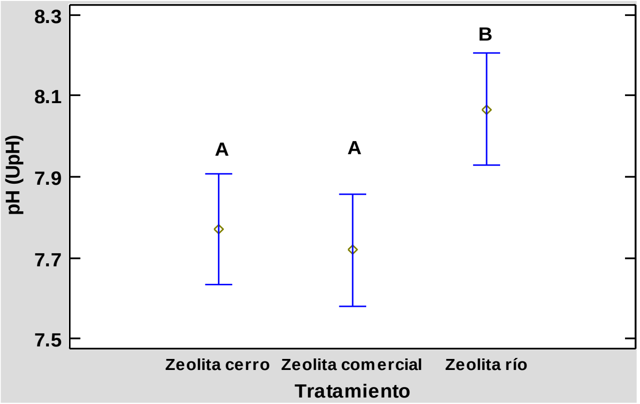
Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

pH

The multifactorial Anova analysis (p <0.05, 95% confidence level) showed that two values have a statistically significant effect on pH. River zeolite was the one that presented a pH with an alkaline tendency (8.07) and the hill zeolite and commercial zeolite treatments tended to behave more stable (7.77 and 7.71) (figure 6). The Height factor had a fairly homogeneous behavior for the pH at 0.9 m and 1.1 m (7.35 and 7.33) and the pH at 0.75 m showed an alkaline tendency (8.88) (figure 7). Finally, in the six-hour treatment an alkaline pH (8.45) was presented and for the other times the pH tends to neutrality (figure 8).

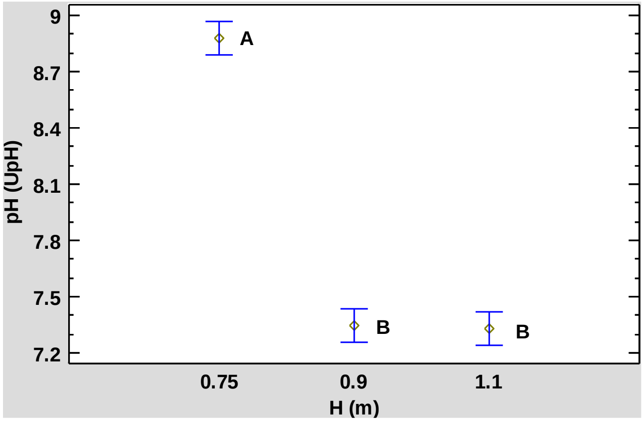
Figura 6. Valores de pH en cada zeolita. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

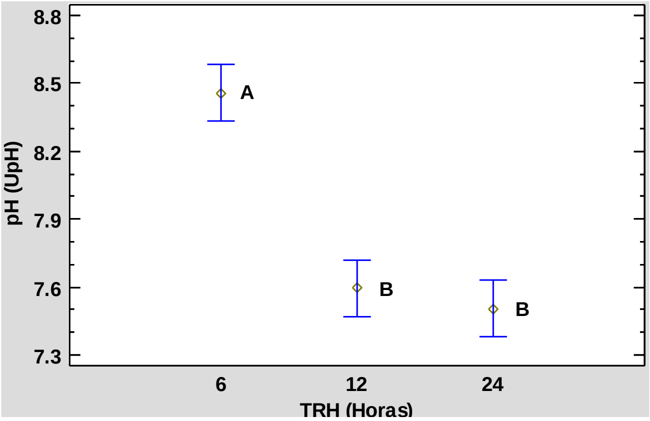
Figura 7. Valores de pH en cada H. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Figura 8. Valores de pH en cada TRH. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

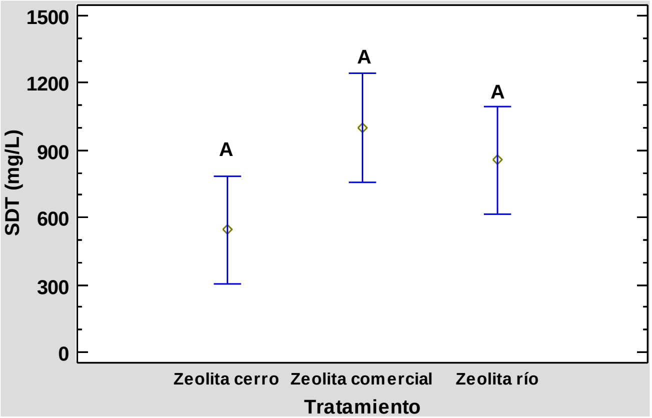
Fuente: Elaboración propia

Total dissolved solids

The multifactorial Anova analysis (p <0.05, 95% confidence level) shows that the Zeolite factor has a statistically significant effect on TDS. Here, cerro zeolite was the one that presented the lowest average (545.28 mg / L), followed by river zeolite (854.64 mg / L) and finally the commercial zeolite (1000.55 mg / L) (figure 9). The Height factor has a significant effect on the TDS. The lowest concentration was for the H of 1.1 m (593.91 mg / L), followed by the H of 0.9 m (604.19 mg / L) and finally the H of 0.75 (1202.37 mg / L) (figure 10). For the TRH factor, the lowest concentration occurred at 12 hours (690.13 mg /

L), followed by six hours (796.20 mg / L) and the highest at 24 hours (914.14 mg / L) (Figure 11).

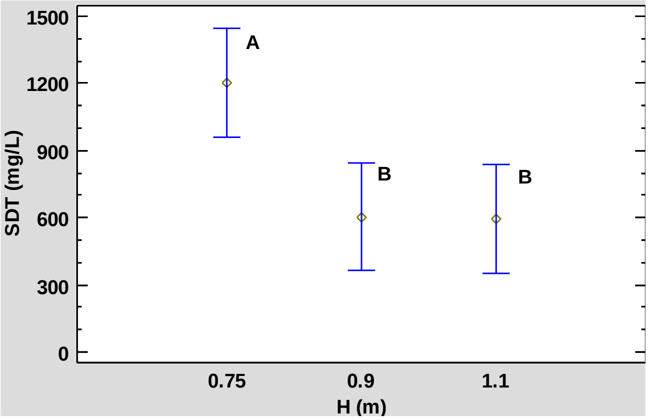
Figura 9. Valores de SDT en cada zeolita. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

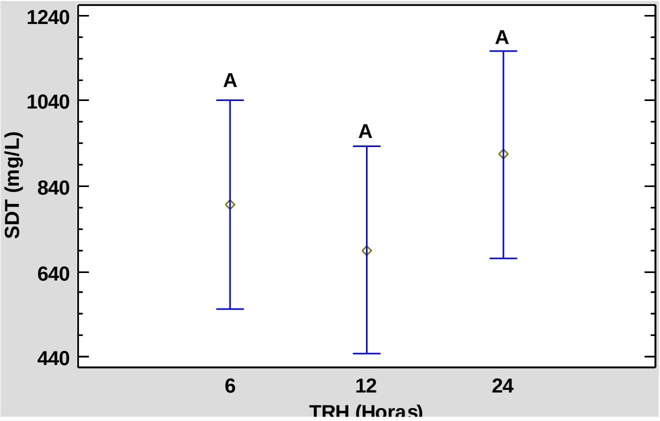
Figura 10. Valores de SDT en cada H. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Figura 11. Valores de SDT en cada TRH. Contraste de medias (medias ± EE)



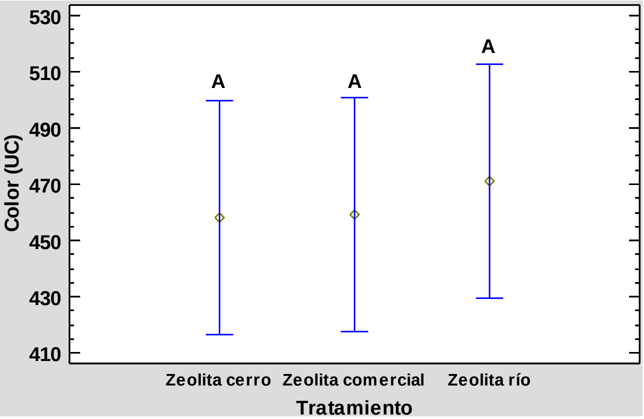
Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Color

The multifactorial Anova analysis (p <0.05, 95% confidence level) shows that the Zeolite factor does not have a statistically significant effect on color. Cerro zeolite was the one with the lowest average (458.10 UC), followed by commercial (459.35 UC) and finally river zeolite (471.14 UC) (figure 12). The Height factor has a significant effect on color. The lowest concentration was for the H of 0.9 m (302.87 UC), followed by the H of 1.1 m (302.95 UC) and finally the H of 0.75 (782.77 UC) (figure 13). For the TRH factor, the lowest concentration occurred at 24 hours (395.30 UC), followed by 12 hours (401.52 UC) and the highest at six hours (591.76 UC) (Figure 14).

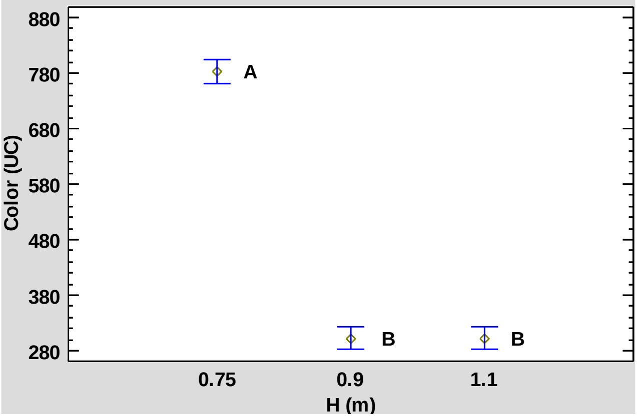
Figura 12. Valores de color en cada Zeolita. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

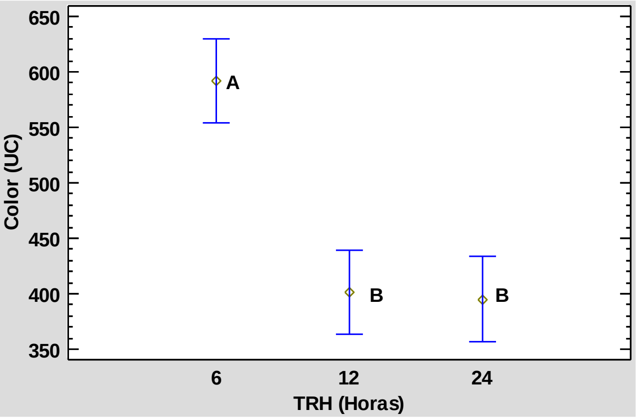
Figura 13. Valores de color en cada H. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Figura 14. Valores de color en cada TRH. Contraste de medias (medias ± EE)



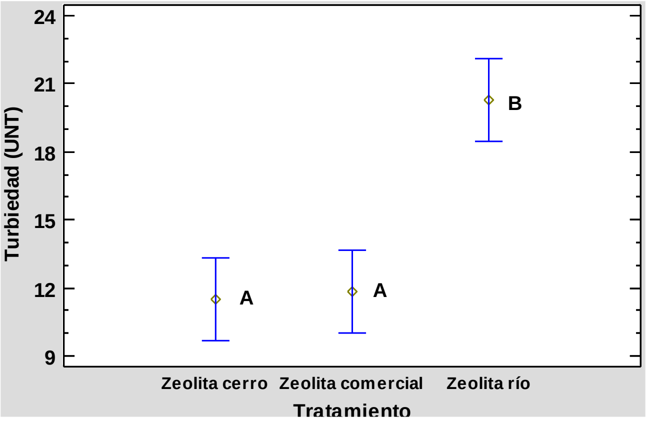
Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Turbidity

The multifactorial Anova analysis (p <0.05, 95% confidence level) shows that the Zeolite factor has a statistically significant effect on turbidity. The cerro zeolite presented the lowest average (11.52 NTU), followed by the commercial one (11.85 NTU) and finally the river zeolite (20.26 NTU) (figure 15). The Height factor has a significant effect on turbidity. The lowest concentration occurred in the H of 1.1 m (13.71 NTU), followed by the H of 0.9 m (13.94 NTU) and finally the H of 0.75 (15.98 NTU) (figure 16). For the TRH factor, the lowest concentration occurred at 24 hours (8.67 NTU), followed by 12 hours (11.51 NTU) and the highest at six hours (23.46 NTU) (Figure 17).

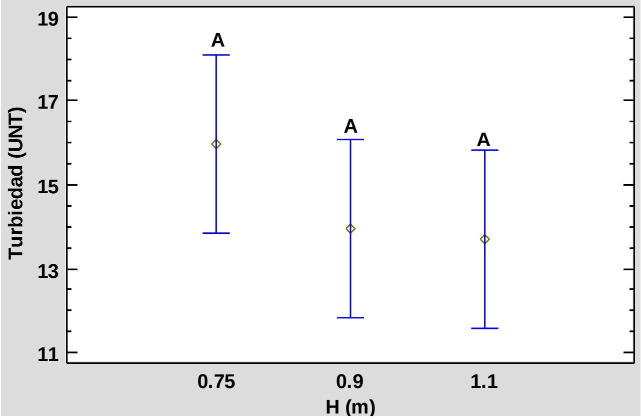
Figura 15. Valores de turbiedad en cada zeolita. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

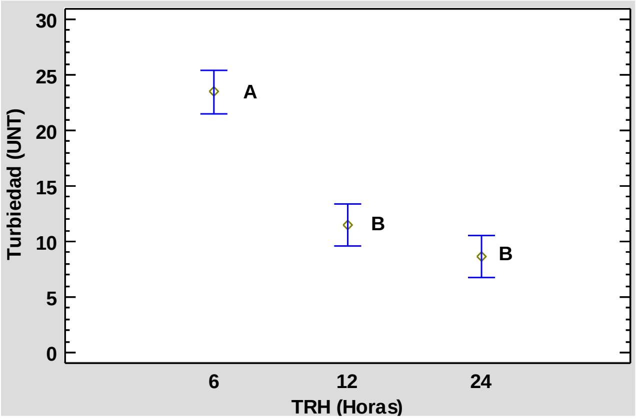
Figura 16. Valores de turbiedad en cada H. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Figura 17. Valores de turbiedad en cada TRH. Contraste de medias (medias ± EE)



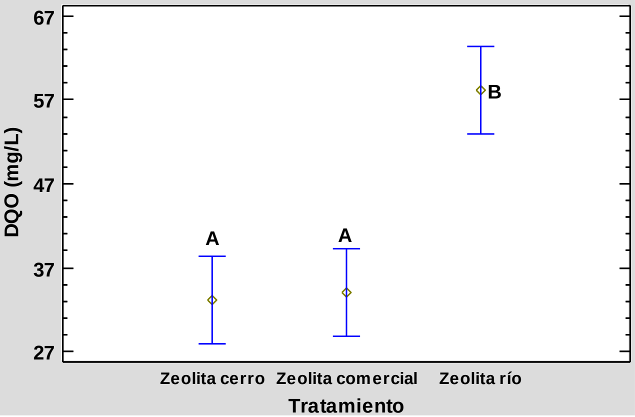
Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Chemical oxygen demand

The multifactorial Anova analysis (p <0.05, 95% confidence level) shows that the Zeolite factor has a statistically significant effect on COD. The cerro zeolite was the one that presented the lowest average (33.09 mg / L), followed by the commercial one (34.05 mg / L) and finally the river zeolite (58.20 mg / L) (Figure 18). The Height factor does not have a significant effect on COD. The lowest concentration occurred in the H of 1.1 m (39.40 mg / L), followed by the H of 0.9 m (40.07 mg / L) and finally the H of 0.75 (45.88 mg / L) (figure 19). For the TRH factor, if there is a significant effect and the lowest concentration occurred at 24 hours (24.90 mg / L), followed by 12 hours (33.05, mg / L) and the highest at 6 hours (67.40 mg / L) (Figure 20).

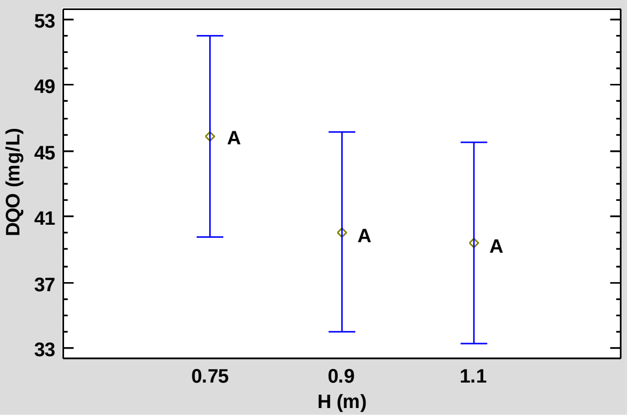
Figura 18. Valores de DQO en cada Zeolita. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

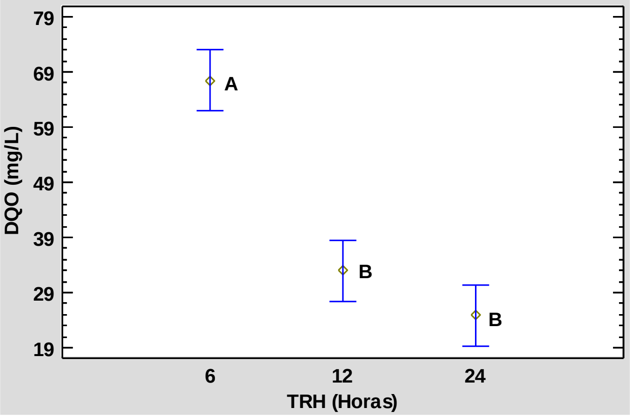
Figura 19. Valores de DQO en cada H. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Figura 20. Valores de DQO en cada TRH. Contraste de medias (medias ± EE)



Nota: Letras distintas muestran diferencias estadísticamente significativas (*N* = 18).

Fuente: Elaboración propia

Basic contaminant removal efficiency

Table 4 describes the removal efficiencies of the parameters evaluated in the treatments. In river and hill zeolite, the most optimal treatment was presented with an H of 0.9 and with 24 hours of HRT; for the commercial zeolite, with the H treatment of 0.9 m with 12 hours. This allows us to observe that the best treatment is cerro zeolite, since it reached the highest efficiency with 71.95% for SDT, 84.41% for color, 96.44% for turbidity and 97.36% for COD. It is important to clarify that the negative sign (-) in the removal efficiency indicates that the value of the parameter is higher at the output and is lower at the input, important for the case of temperature, which shows us the increase in this value in the effluent as a consequence of microbial activity in the BLFFA.

Tabla 4. Eficiencias de remoción de contaminantes básicos en los BLFFA para diferentes tratamientos (zeolita, H y TRH) (*N* = 81)

	Parámetro	H = 0.75 m			H = 0.90 m			H = 1.10 m		
		6 h	12 h	24 h	6 h	12 h	24 h	6 h	12 h	24 h
Zeolita de río	Temperatura ºC	-14.54	-17.84	-14.15	-14.28	-15.47	-13.88	-16.92	-19.03	-16.92
	pH (UpH)	-11.96	-1.92	-1.15	7.34	13.13	15.84	8.12	13.52	16.22
	SDT (ppm)	-4.48	5.07	6.43	12.60	13.96	15.29	12.94	13.69	15.26
	Color (UC)	36.86	40.43	44.19	62.52	81.98	82.06	63.33	81.82	81.90
	Turbiedad (UNT)	82.55	83.54	84.58	59.25	78.35	91.53	59.88	79.42	91.73
	DQO (mg/L)	87.05	87.78	88.55	69.76	83.93	93.71	70.22	84.72	93.86
Zeolita de cerro	Parámetro	H = 0.75 m			H = 0.90 m			H = 1.10 m		
		6 h	12 h	24 h	6 h	12 h	24 h	6 h	12 h	24 h
	Temperatura ºC	-18.11	-18.76	-20.35	-15.47	-13.88	-15.47	-19.03	-16.92	-19.03
	pH (UpH)	-8.87	1.55	1.94	7.73	19.31	20.08	8.89	19.31	20.08
	SDT (ppm)	-1.61	8.10	8.15	37.81	71.42	71.95	44.45	71.47	71.94
	Color (UC)	37.11	41.12	41.12	63.93	84.34	84.41	63.93	83.99	84.14
	Turbiedad (UNT)	82.62	83.73	83.73	78.42	96.33	96.44	78.69	96.20	96.26

	DQO (mg/L)	87.10	87.92	87.92	83.98	97.28	97.36	84.19	97.18	97.23
Zeolita comercial	Parámetro	H = 0.75 m			H = 0.90 m			H = 1.10 m		
		6 h	12 h	24 h	6 h	12 h	24 h	6 h	12 h	24 h
	Temperatura ºC	-19.03	-22.20	-24.84	-13.88	-15.47	-13.88	-16.92	-19.03	-16.92
	pH (UpH)	-8.49	1.17	2.32	8.12	20.86	21.63	7.73	21.24	20.86
	SDT (ppm)	-1.58	8.02	8.13	23.04	26.49	55.35	22.92	28.31	56.64
	Color (UC)	37.12	41.13	41.11	63.53	84.15	84.22	63.60	84.15	84.23
	Turbiedad (UNT)	82.62	83.73	83.73	78.24	95.71	95.50	78.59	95.57	95.61
	DQO (mg/L)	87.10	87.92	87.92	83.85	96.82	96.66	84.11	96.71	96.74

Fuente: Elaboración propia

Discussion

The results show cerro zeolite as the best support medium compared to river and commercial zeolite (although it is similar to commercial zeolite, which is currently used in wastewater treatment). We attribute it to the physical, chemical and mechanical characteristics: the cerro zeolite excelled in parameters such as porosity, absorption and humidity, determining factors in wastewater treatment. The optimal height (H) in the BLFFA was 0.9 m, an important factor to evaluate since the higher the height increases the costs and the efficiencies are not relevant to justify the investment and the lower the performance in quality of the treated water falls. In this sense, native zeolites have physical and chemical properties to be used in the treatment of domestic wastewater, as suggested by Florencia (2012), who evaluated a native zeolite ($Fe0.53Ca1.87Mg0.61Na0.93K0.80Si29.00Al7.25O72$) in the removal of ammoniacal nitrogen and obtained up to 78% with a TRH of 96 hours. On the other hand, Hernández et al. (2010) identified that the natural zeolites of Mexico from the Clinoptilolite and Mordenite deposits have the potential for application in water treatment, biomedicine, agriculture and air pollution, as well as the natural zeolites in this study, which proved to be efficient in the sewage treatment.

Most of the biological processes used today are stabilization ponds, activated sludge, biofilters, biodisks and anaerobic digestion. Although these offer acceptable removals of organic matter (> 80%), one of their disadvantages is TRH, which is too large. While in BLFFA the TRH is much lower and allows the bacteria to be in contact with the food present in the wastewater, which is enough for them to assimilate or stabilize the organic matter, because if the TRH is very small, not all the organic matter will be removed and the effluent will have high COD values, and if the HRT is very large, good performance is not guaranteed (Conagua, 2016b). The essential goal in biological treatments such as BLFFA is to stabilize organic matter, coagulate and remove floating colloidal solids found in water, including in some cases the removal of nutrients such as nitrogen and phosphorus (Noyola et al., 2013). Biomass converts dissolved carbonaceous organic matter in a colloidal state into different gases and cellular tissues, forming biological layers composed of cellular matter and important organic colloids in the support medium in BLFFAs (Conagua, 2016b). In this experiment, the 24-hour TRH contributed to a higher COD removal efficiency in the river and hill zeolites, which favored a greater presence of microorganisms in the support medium within the BLFFA. It has also been shown that TRH is important in fluidized bed reactors (3.6 mm Ø bead beds), since with TRH of 36 hours and with Methylobacterium sp (30-37 ° C) the degradation of toxic compounds such as formaldehyde up to 98% (Qiu et al., 2014).

Temperature plays an important role in wastewater treatment as it accelerates biochemical reactions, reduces gas solubility, and intensifies flavors and odors during treatments. During the experiments a variation of 28.95 ° C to 30.02 ° C was presented, which allowed the development and establishment of mesophilic microorganisms responsible for the degradation of organic matter (Miao, Zhang, Jia, Liao and Li, 2018; Torres et al. ., 2020), complying with NOM-001-SEMARNAT-1996 in the discharge value, which is less than 40 ° C. Regarding the pH, it presented variations in the treatments, ranging from neutral to slightly alkaline (from 7.3 to 8.9), an important condition for the establishment of microorganisms, and maintaining the range of the discharge criteria established by NOM-001-SEMARNAT-1996 , which is 6.5 to 10 units. Espinosa, Delgado and Hidalgo (2020) worked on a wastewater treatment plant with a combined process (an anoxic reactor, two aerobic full-mix reactors, and a membrane biological reactor) and with a high nitrogen content. Regarding temperature, the behaviors were 38 ° C for the anoxic, 40 ° C for the aerobes and 39 ° C for the membrane, while the pH behavior remained at eight units in all processes. When comparing these results with ours, we can observe that the temperature is lower because it is subjected to a single process such as the packed bed and because of the characteristics of the support medium that are zeolites; in the case of pH, the range is slightly alkaline as in the processes evaluated by Espinosa *et al*. (2020).

The TDS are a parameter that is not generally used to measure the performance of the rectors, however, in this case it is important to determine the adsorption capacity of dissolved ions. In this experiment, it was shown that cerro zeolite is the most efficient, removing up to 71.95%. The values that were reduced are dissolved inorganic salts (Hadad, Maine and Bonetto, 2006). With this performance it is corroborated that this stone material from the hill is a zeolite that captures dissolved ions, therefore, following Mietto, Politeo, Breschigliaro and Borin (2015), so that common support materials (biological filters or biofilters) can have similar values This removal must have prolonged HRT (> 6 days) to favor the elimination of ions or nutrients combined with the temperature effect. The SDT values in the cerro and commercial zeolite effluents of our study are within what is allowed for irrigation discharge (Secretary of the Navy, Natural Resources and Fisheries [Semarnap] -Conagua-Secretary of Urban Development and Ecology [Sedue], 1989), with 266.63 mg / L and 424.40 mg / L, respectively, since the concentration of TDS that does not present harmful effects in any crop are less than 500 mg / L and river zeolite did present values above this criterion with 805.20 mg / L.

Turbidity and color are not parameters regulated by NOM-001-SEMARNAT-1996, but they are used as quick measurement variables in the performance of wastewater treatment systems and are directly related to suspended solids (Ortiz, López, Torres and Pampillón, 2018; Torres et al., 2020). In this sense, the support medium fulfills a filtering function for these solids, retaining them by adhesion; and in the case of colloidal matter, it is stabilized by biomass (Crites and Tchobanoglous, 2000), which is why zeolites present high efficiencies in removing turbidity and color (> 91% and> 84% respectively). The hill zeolite (0.9 m of H and 24 hours of TRH) is the one that achieved 96.44% removal of turbidity and 84.41% of color.

COD quantifies the oxygen required to chemically oxidize organic (non-putrescible) and inorganic matter present in wastewater (O'Dell, 1993); It is the most important variable in biological processes to measure the performance of wastewater treatment systems (Crites and Tchobanoglous, 2000). Espinosa et al. (2020) evaluated a biological treatment system (anoxic-aerobic-membrane biological reactor) operated by a company dedicated to the production of sardine and tuna meals, whose tributaries are very high in COD (7728.9 ± 513.8 mg / L) and effluents (353.8 ± 62.1 mg / L) are above the draft standard PROY-NOM-001-SEMARNAT-2017 (Semarnat, January 5, 2018) for discharge in marine areas and estuaries (85 mg / L of COD), however the removal efficiency of this system is reported high at 95.4% for this parameter. In the treatments evaluated in this experiment, it can be observed that the cerro (97.36%) and commercial (96.66%) zeolite have a lot of similarity in the removal of COD with 24 hours of retention and 0.9 of H; the one with the lowest efficiency is river zeolite (93.71%). However, in our case, when treating domestic wastewater, the influent has an average concentration of 373 mg / L, which allows the effluents (9.8 mg / L of COD) to meet the permissible limits of PROY-NOM-001- SEMARNAT-2017 (Semarnat, January 5, 2018).

Comparing the zeolites in this study with other support media and anaerobic environments that have been used in fixed-bed bioreactors or similar, we can establish that cerro zeolite has a high potential for its use in wastewater due to its high removal efficiencies. obtained. Rivas et al. (2003) reached 91% removal with initial COD loads of 463 mg / L, very similar to those of our study (domestic water); Castillo et al. (2006), with cob waste material as a support medium, recorded removals between 50 and 70% of COD from wastewater and pig water; Rodríguez et al. (2007), using a ceramic support medium, recorded 57% removal of COD from a domestic wastewater; Cárdenas and Ramos (2009) found that seashells allow up to 89.7% removal of COD. Finally, this experimental treatment with BLFFA presents similar performances (> 93% of COD) to the aerobic processes of biomass in suspension such as the sequential discontinuous reactors reported by Torres et al. (2020), where its best treatment is given with a 20-hour HRT, which reaches 92.4% COD removal. Undoubtedly, this shows that TRH is decisive in the removal by contact time of microorganisms with wastewater.

Conclusion

Native cerro and river zeolites should be implemented in the treatment of domestic wastewater in BLFFA as they are efficient in the removal of basic pollutants and favor operating times and costs.

Experiments show that, although there is no statistically significant difference between the use of 12 or 24 hours of 20-hour HRT in the removal of color, turbidity and COD, the best removal efficiencies for these parameters are achieved in 24 hours of HRT. .

The cerro zeolite is the one that presents the best performance in the removal of basic pollutants operating with 0.9 m of H and with 24 hours of TRH.

For the use of BLFFA it is necessary that prior to the process there is a pretreatment system for the elimination of sedimentable and suspended solids.

Finally, we can recommend the implementation of the use of BLFFAs for the secondary treatment of domestic effluents in decentralized systems in the southeast of Mexico as a viable alternative in the treatment of their wastewater.

Future lines of research

To know the potential of native zeolites in packed bed reactors, the study in the reactors should be continued in a short term and to evaluate if the hydraulic retention times with the current operation are sufficient to be able to remove nutrients (nitrogen and phosphorus) and others. Xenobiotics in concentration values that allow discharge to receiving bodies in a safe way, as established by the current environmental regulatory framework. We know that it is necessary to configure a treatment train tailored to the type of water to be processed, so another line of research in the medium term would be to evaluate the total efficiency of a pilot-experimental treatment train with water of medium and strong concentration. with different configurations in the (secondary) train with units in series, aerobic, anaerobic, etc., so that a treatment train can be recommended for different efficient and economical cases.

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