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Determination of The Performance of Sand, Zeolite and Vermiculite Mixture in Removal of Phosphorus From Wastewater

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ABSTRACT

Domestic and industrial wastewater discharged directly into the receiving environment without treatment causes serious deterioration in aquatic ecosystems due to their high phosphorus content, which threatens life. Phosphorus accelerates the eutrophication process, especially in stagnant water bodies such as lakes and dams, causing algae formation, decreased oxygen levels and disruption of ecosystem balance. For this reason, wastewater should never be discharged into the receiving environment without ensuring phosphorus removal within the limit values specified in the Water Pollution and Control Regulation of the Republic of Türkiye. This effective, phosphorus removal method is one of the natural filters found in abundance in Türkiye, can choose adsorptive phosphorus removal. This is aimed at natural filter materials sand, zeolite and vermiculite are sieved from each of them and materials remaining between sieves with a diameter of 1 and 2 cm are formed (50 cm3) homogeneous mixtures with a total volume of 150 cm³ and tested using chromatography columns with an inner diameter of 3 cm from a micro distance representing wetlands in the laboratory environment. In the experiments, synthetic wastewater solutions containing phosphorus at concentrations of 25, 50 and 100 ppm were transferred to the columns with a peristaltic pump at flow rates of 1 and 2 ml/min and samples were taken at 3rd, 6th, 12th, 24th and 48th hours and filtered, and pH, electrical conductivity (EC) and total phosphorus analyses were performed. According to the results obtained, the highest phosphorus removal efficiency was obtained as 99% in 3 hours at 2 ml/min flow rate and 50 ppm concentration, while the lowest efficiency was observed as 3% in 48 hours at 100 ppm and 2 ml/min conditions. It has been determined that the materials used in mixture form have higher performance than their pure forms.

Kum, Zeolit ve Vermikülit Karişiminin Atik Sulardan Fosfor Giderim Performansinin Belirlenmesi

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ABSTRACT

Arıtılmadan doğrudan alıcı ortama deşarj edilen evsel ve endüstriyel atık sular, yüksek fosfor içerikleri nedeniyle sucul ekosistemlerde ciddi bozulmalara neden olmakta ve bu durum canlı yaşamını oldukça tehdit etmektedir. Fosfor, özellikle göller ve barajlar gibi durgun su kütlelerinde ötrofikasyon sürecini hızlandırarak alg oluşumuna, oksijen seviyelerinin düşmesine ve ekosistem dengesinin bozulmasına neden olmaktadır. Bu nedenle atık sular, Türkiye Cumhuriyeti Çevre, Şehircilik ve İklim Değişikliği Bakanlığı'nın Su Kirliliği ve Kontrolü Yönetmeliği'nde belirtilen sınır değerler dahilinde fosfor giderimi sağlanmadan asla alıcı ortama deşarj edilmemelidir. Bu çalışmada, fosfor giderim yöntemlerinden biri olan ve Türkiye'de bolca bulunan doğal filtre materyalleri kullanılarak gerçekleştirilen adsorptif fosfor giderimi incelenmiştir. Bu amaçla, doğal filtre materyallerinden olan kum, zeolit ve vermikülitin her birinden elenerek 1 ve 2 cm gözenek çapına sahip elekler arasında kalan materyallerden 50 cm³ alınarak toplam 150 cm³ hacme sahip homojen karışımlar oluşturulmuş ve laboratuvar ortamında yapay sulak alanları temsilen mikro ölçekli iç çapı 3 cm olan kromatografi kolonları kullanılarak test edilmiştir. Deneylerde, 25, 50 ve 100 ppm konsantrasyonlarında fosfor içeren sentetik atık su çözeltileri peristaltik pompa ile 1 ve 2 ml/dak akış hızlarında kolonlara aktarılmış ve 3, 6, 12, 24 ve 48. saatlerde örnekler alınarak filtre edilmiş olup pH, elektriksel iletkenlik (EC) ve toplam fosfor analizleri yapılmıştır. Elde edilen sonuçlara göre, en yüksek fosfor giderim verimi 3 saatte 2 ml/dak akış hızı ve 50 ppm konsantrasyonda %99 olarak elde edilirken, en düşük verim 48 saatte 100 ppm ve 2 ml/dak koşullarında %3 olarak gözlenmiştir. Karışım halinde kullanılan malzemelerin saf hallerine göre daha yüksek performans gösterdiği belirlenmiştir.

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INTRODUCTION

It is known that water is essential for all living things. Water is the most critical factor in the survival of all plants and animals, including humans, in our ecosystem (Armağan and Işık, 2022). Approximately 97% of the world's total 1.4 billion km^3 of water is saltwater, and the remaining 3% is freshwater. Since most of the fresh water in our world is only found in the poles and underground, the low amount of water that living things can use can be clearly expressed in numbers. It is a known fact that usable water is limited to the increasing world population. With the growing demand for industrial, agricultural, and domestic water, competition arises between sectors in the use of water. Today, limited water resources must be utilized effectively in all sectors in a manner that is compatible with the environment (Çakmak et al., 2003). Although seas and has many dams and rivers, it creates the image that it is rich in terms of water resources. However, Türkiye is among the countries suffering from a water shortage according to international criteria. The scarcity of usable water resources also brings about a food problem. Therefore, it is necessary to utilize existing water resources efficiently and to prioritize the reuse of treated wastewater. Various technologies are used in different parts of the world for wastewater treatment. These technologies consist of physical, chemical or biological processes and are mainly preferred depending on treatment costs. Constructed wetlands are preferred especially in rural areas because of the low investment costs. Constructed wetlands are highly effective in removing nutrients, particularly due to the filter materials and plants they contain (EPA, 1993).

Constructed wetlands are a natural wastewater treatment system that has been rapidly used in recent years as an alternative to conventional treatment systems for domestic and industrial wastewater treatment and can be easily applied in small to medium-sized areas with low energy requirements (Anonymous, 1993; Anonymous, 1999; Tunçsiper and Akça, 2006; Çiftçi et al., 2007). Nitrate, phosphorus and heavy metals can mix into groundwater or groundwater through leaks from the bottoms of constructed wetland ponds, waste storage areas and resting and processing ponds built for manure management in animal shelters.

Nitrate is a factor that can cause pollution in groundwater at levels that are hazardous to the environment and human health. Pollution that occurs with water leaking from storage and treatment areas is defined as point source pollution. Spalding and Exner (1993) have demonstrated that nitrate is the most common chemical pollutant in groundwater. Water contaminated with nitrate through groundwater flows can reach surface waters and disrupt the nitrogen: phosphorus balance in these environments, causing eutrophication and algae formation (Bolger and Stevens, 1999). The most significant source of nitrate that threatens groundwater is agricultural waste.

Phosphorus, which poses another critical threat, is found in inorganic and organic forms as phosphate compounds. It is mainly in the form of orthophosphate that can be used by algae and macrophytes. Organic phosphorus is generally formed as a result of biological processes and is found in raw wastewater, food residues, and human feces. Inorganic phosphorus is usually formed as a result of the use of detergents for cleaning purposes. Phosphorus fertilizers also contain significant amounts of inorganic phosphorus. While more than one process is carried out during the treatment of phosphorus from wastewater, removal is provided until a specific capacity is reached. No additional removal occurs after this level (EPA, 1993). Phosphorus removal requires advanced treatment technologies. Various methods such as activated carbon, reverse osmosis, and electrodialysis are used for phosphorus removal from wastewater (Yeoman et al., 1988; Taş et al., 2007). In recent years, research has focused on phosphorus removal by using solid retention materials. Cui et al. (2008) in their study on phosphorus adsorption characteristics and physico-chemical properties of nine different filter materials peat, topsoil, gravel, medium-sized sand, blast furnace slag, coal slag, artificial blast furnace slag, artificial coal slag and medium-sized artificial sand materials were tested. Peat material was determined to be the best phosphorus absorbent material. Onar and Öztürk (1993) in their study on phosphorus adsorption by pumice grains obtained a phosphorus removal efficiency of over 80% and stated that pumice is a promising scavenger in phosphorus removal. Njau et al. (2003) in their study on the use of pumis as a filter material in constructed wetlands used for the treatment of domestic wastewater stated that pumis provided a pretty high level of phosphorus removal. Uzun et al. (2021) investigated the effectiveness of sand, pumis and zeolite in their study aiming at phosphorus removal from wastewater and found that pumis material was a better adsorbent than other materials.

Zeolites, which have a wide range of applications and properties among adsorbents, are increasingly being utilized in pollution control due to their ion exchange and adsorption properties. For this purpose, zeolites are used in the retention of radioactive wastes in water, retention of metal ions and nitrogen compounds in wastewater, adsorption of flue gases, cleaning of oil spills, landfilling and oxygen production. Many studies have been conducted on pollution control with zeolites. For example, they have been used for the treatment of metal pollutants such as zinc (Calvo et al., 2009), lead and copper (Wang et al., 2008; Stylianou et al., 2007; Sprynskyy, 2008), iron and chromium (Inglezakis et al., 2002) from wastewater. Clinoptilolite-type zeolite was used for the removal of NH4+ ions from landfill leachate (Doğan et al., 2018). Apart from pollution control, zeolite has many areas of use. If we need to collect them under main headings, we can classify them as energy, agriculture and animal husbandry, mining and metallurgy and other areas of use. In the energy sector, zeolite is used in coal gasification, cleaning nitrogen oxides and hydrocarbons, purification of natural gas, removal of carbon dioxide, as heat exchangers in solar energy production and as catalysts in the production of petroleum products. Zeolites can be utilized as heat exchangers in solar energy systems by leveraging their water-absorbing and water-giving properties (DPT, 2001). Natural zeolites are widely used as fertilizer carriers in fertilization and soil preparation, as well as in agricultural applications. Zeolites are used in mining, exploration of mineral deposits and retention of some heavy metals in metallurgy. Other areas of use of zeolite, apart from the areas mentioned, are the paper sector, construction sector, health sector and detergent sector.

In this study, an attempt was made to determine the phosphorus removal efficiency of homogeneous mixtures of sand, zeolite, and vermiculite materials from wastewater.

MATERIAL AND METHOD

The sand, zeolite, and vermiculite used in the study were supplied by a commercial business. The materials were sieved through sieves with 1-2 mm pore diameter, and the remaining part was used in the study. The experiments were carried out with three replications using synthetic wastewater with different concentrations (25, 50 and 100 ppm).

In this study, columns with an inner diameter of 3 cm and a length of 60 cm made of glass were used. The columns were filled with 150 cm3 filter material representing constructed wetlands at a microscale. The system was operated by continuously feeding from the top at a flow rate of 5 ml/min using a peristaltic pump with a digital display. After the system was operated, samples were taken from the outlet sections of the columns at 3, 6, 12, 24, and 48 hours in order to determine the capacities of the materials depending on time (Fig. 1). The samples obtained were subjected to pH, EC, and total phosphorus analyses. In the study, pH and EC analyses were determined using a Hanna brand device. Total phosphorus contents were determined at a wavelength of 882 nm in a spectrophotometer according to the ammonium molybdate blue color method (Kuo and Sparks, 1996).

Figure 1
The glass column test setup where the study was conducted

RESULTS

In the study carried out, solutions containing phosphorus at three different concentrations (25, 50, and 100 ppm) were applied to the filter materials in the columns at 1 and 2 ml/min flow rates for 48 hours. During this process, samples were taken at different times to determine the time-dependent capacities of the materials. pH, EC, and total phosphorus were determined in the obtained samples.

Effect of Different Concentration Ratios on pH at Different Flow Rates

As a result of the experiment, the effect of different phosphorus concentrations and mixing ratios on pH is shown in Table 1 and change in pH values with hydraulic retention times is presented in Fig. 2. The pH value of the prepared solutions was determined as 8.34. When Table 1 is examined, it is observed that the pH values in the effluent analyses of the synthetic wastewater included in the columns increased after contact with the material and decreased during the continuation of the study. As a result of the experiment, different concentrations had a significant effect on the pH change, depending on the hydraulic retention times and mixing ratios. According to the Water Pollution and Control Regulation of the Ministry of Environment and Urbanization of the Republic of Türkiye, the pH range required for discharging wastewater into the receiving environment is 6.0-9.0 (WPCG, 2004).

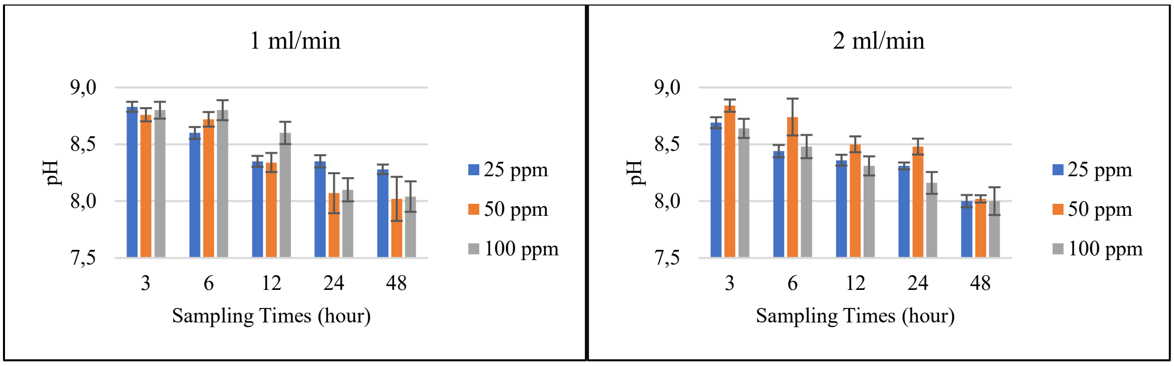
Table 1
Changes in pH values during the treatment process

Flow Rate (ml/min)	Conc. (ppm)	Sampling Times (hour)				
		3	6	12	24	48
1	25	8.83	8.60	8.35	8.35	8.28
	50	8.76	8.72	8.34	8.07	8.02
	100	8.80	8.80	8.60	8.10	8.04
2	25	8.69	8.44	8.36	8.31	8.00
	50	8.84	8.74	8.50	8.48	8.02
	100	8.64	8.48	8.31	8.16	8.00

The pH values were determined as the lowest 8.76 and 8.64, and the highest 8.83 and 8.84 at the end of the 3-hour period at 1 ml/min and 2 ml/min flow rates, respectively; the lowest 8.60 and 8.44, and the highest 8.80 and 8.74 at the end of the 6-hour ; the lowest 8.34 and 8.31, and the highest 8.60 and 8.50 at the end of the 12-hour; the lowest 8.07 and 8.16, and the highest 8.35 and 8.48 at the end of the 24-hour ; the lowest 8.02 and 8.00, and the highest 8.28 and 8.00 at the end of the 48-hour.

Figure 2

Change in pH values of mixture



Effect of Different Concentrations at Different Flow Rates on EC

As a result of the experiment, the effect of different phosphorus concentrations and mixing ratios on EC is shown in Table 2 and change in EC values with hydraulic retention times is presented in Fig. 3. The EC value of the prepared solutions was determined as 43.78 µS/cm. As a result of the experiment, different concentrations had a significant effect on the EC change, depending on the hydraulic retention times and mixing ratios. According to the Water Pollution and Control Regulation of the Ministry of Environment and Urbanization of the Republic of Türkiye, the EC amount required for wastewater to be discharged into the receiving environment should be below 2000 µS/cm (WPCG, 2004).

Table 2

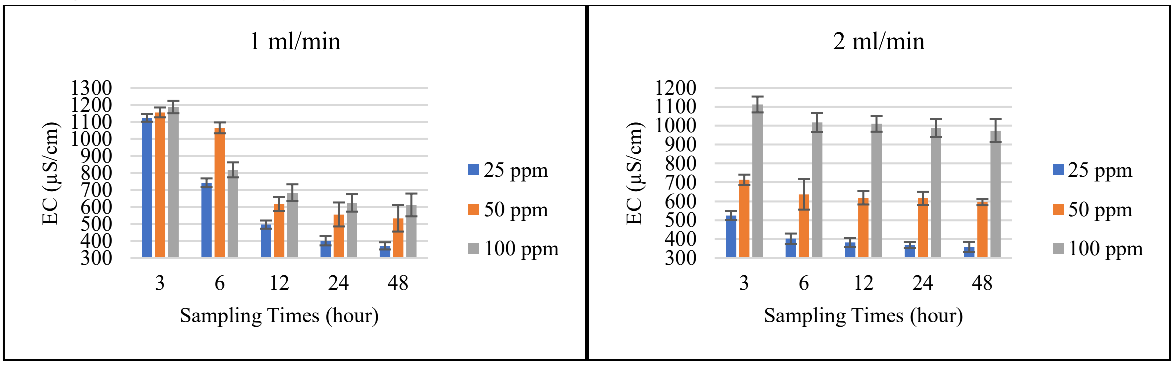
Changes in EC (µS/cm) values during the treatment process

Flow Rate (ml/min)	Conc. (ppm)	Sampling Times (hour) 3	6	12	24	48
	25	1123.00	741.67	496.60	401.23	371.53
1	50	1155.33	1064.43	617.07	556.07	533.37
	100	1187.00	817.83	683.90	623.70	611.93
	25	524.60	402.60	382.70	369.50	359.37
2	50	713.97	637.17	618.23	615.60	595.07
	100	1111.67	1016.27	1009.93	987.07	973.20

EC values (µS/cm) were determined as the lowest 1123 and 524.6, and the highest 1187 and 1111.67 after 3 hours at 1 ml/min and 2 ml/min flow rates, respectively; as the lowest 741.67 and 402.60, and the highest 1064.43 and 1016.27 after 6 hours; as the lowest 496.6 and 382.7, and the highest 683.9 and 1009.93 after 12 hours; as the lowest 401.23 and 369.5, and the highest 623.7 and 987.07 after 24 hours; as the lowest 371.53 and 359.37, and the highest 611.93 and 973.2 after 48 hours.

Figure 3

Change in EC values of mixture



Effect of Different Concentrations at Different Flow Rates on TP

The prepared phosphorus solutions of 25, 50, and 100 ppm were transferred to the columns containing the filter material and the results obtained as a result of the analyses are given in Table 3 and change in TP values with hydraulic retention times is presented in Fig. 4. As a result of the experiment, it was determined that different mixing ratios had a significant effect on the TP change depending on the hydraulic retention times of different concentrations. According to the Water Pollution and Control Regulation of the Ministry of Environment and Urbanization of the Republic of Türkiye, the TP limit required for discharging wastewater into the receiving environment is 10 ppm (WPCG, 2004).

Table 3

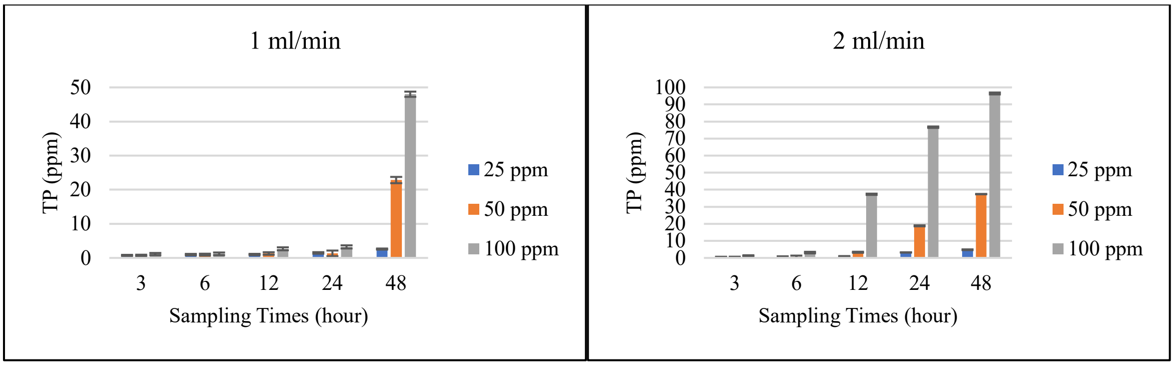
Changes in TP (ppm) values during the treatment process

Flow Rate (ml/min)	Conc. (ppm)	Sampling Times (hour) 3	6	12	24	48
	25	0.73	0.97	1.00	1.47	2.60
1	50	0.70	0.97	1.30	1.33	22.83
	100	1.13	1.20	2.67	3.23	48.00
	25	0.50	0.73	0.90	3.20	4.80
2	50	0.50	0.60	3.33	18.83	37.42
	100	1.20	3.13	37.33	76.67	96.45

TP values (ppm) were determined as the lowest 0.7 and 0.5, and highest 1.13 and 1.2 at the end of the 3-hour period at 1 ml/min and 2 ml/min flow rates, respectively; as the lowest 0.97 and 0.6, and highest 1.2 and 3.13 at the end of the 6-hour period; as the lowest 1.0 and 0.9, and highest 2.67 and 37.33 at the end of the 12-hour period; as the lowest 1.33 and 3.2, and highest 3.23 and 76.67 at the end of the 24-hour period; as the lowest 2.6 and 4.8, and highest 48 and 96.45 at the end of the 48-hour period.

Figure 4

Change in TP values of mixture



Removal Rates of Different Concentrations at Different Flow Rates

As a result of total phosphorus analyses, the removal performance of the material used in the study was determined by applying solutions with different concentrations at different flow rates to the filter material (Table 4).

Table 4
Changes in TP (%) values during the treatment process

Flow Rate (ml/min)	Conc. (ppm)	Sampling Times (hour) 3	6	12	24	48
	25	97.07	96.13	96.00	94.13	89.60
1	50	98.60	98.07	97.40	97.33	54.33
	100	98.87	98.80	97.33	96.77	52.00
	25	98.00	97.07	96.40	87.20	80.80
2	50	99.00	98.80	93.33	62.33	25.16
	100	98.80	96.87	62.67	23.33	3.55

TP removal (%) of the filter material was determined as the lowest 97.07 and 98.0, and highest 98.87 and 99.0 at the flow rate of 1 ml/min and 2 ml/min, respectively, for 3 hours; as the lowest 96.13 and 96.87, and highest 98.80 and 98.80 at the end of 6 hours; as the lowest 96.0 and 62.67, and highest 97.4 and 96.4 at the end of 12 hours; as the lowest 94.13 and 23.33, and highest 97.33 and 87.2 at the end of 24 hours; as the lowest 52.00 and 3.55, and highest 89.6 and 80.80 at the end of 48 hours.

CONCLUSION AND RECOMMENDATIONS

In this study, which was carried out in the form of column experiments, homogeneous mixtures of sand, zeolite and vermiculite were applied to constructed wastewater containing phosphorus at three different concentrations (25, 50 and 100 ppm) for 48 hours and sampling was carried out at five different times (3rd, 6th, 12th, 24th and 48th hours).

According to the data obtained as a result of the study, it was concluded that after the treatment process, pH and EC values were at the desired levels and there was no obstacle in the discharge to the receiving environments.

As a result of the phosphorus analyses, it was determined that there was no obstacle to the discharge of solutions containing phosphorus at a concentration of 25 ppm after the application of 150 cm^3 of filter material for 48 hours. However, it was also found that solutions with concentrations of 50 and 100 ppm should not be discharged.

As a result of the study, it was revealed that the choice of filter materials to be used in constructed wetlands is crucial. In the planned constructed wetlands, the homogeneous mixing of sand, zeolite, and vermiculite for phosphorus from wastewater will provide more effective results. It has been determined that the expected performance will increase in the treatment of wastewater with low flow rates and low phosphorus content in constructed wetlands in order to improve the effectiveness of the materials. In addition, it has been determined that wastewater with low pH values is easier to treat. Therefore, it will be more effective to subject wastewater with high pH values to adsorption processes after reducing their pH values. It has been suggested that focusing on increasing the exchange capacity of the materials in studies aimed at enhancing their capacities would be more effective.

Yazar Katkıları/Author Contributions

Tasarım/Design: Yazar 1 (%50) – Yazar 2 (%50)

Veri Toplama /Data Collection: Yazar 1 (%30) – Yazar 2 (%70)

Analiz ve yorum/Analysis or Interpretation: Yazar 1 (%70) – Yazar 2 (%30)

Literatür tarama/Literature Search: Yazar 1 (%60) – Yazar 2 (%40)

Yazma/Writing: Yazar 1 (%20) – Yazar 2 (%80)

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