



## A study of treated municipal waste leachate and Zeolite effects on soils

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**ABSTRACT-** As the world's population has grown and become more urban and affluent, waste production has raised drastically. Wastewater reuse has been identified as a way to alleviate water scarcity and improve crop productivity and environmental sustainability. To address the issue, a soil column experiment was carried out in a  $3 \times 3$  factorial randomized block design including three treatments of adsorbents (non-pretreated leachate (L1), rice husk filtered leachate (L2), activated carbon filtered leachate (L3)) and three levels of zeolite (0, 5 and 10% by soil weight). A decrease in drainage water volume through the experiment period was observed. Application of zeolite at 5% level could improve soil removal efficiency and had a positive impact on the quality of the wastewater, as indicated by changes in EC,  $\text{Na}^+$ ,  $\text{Ca}^{2+}+\text{Mg}^{2+}$ , Cl<sup>-</sup> (decreased by 22%, 15%, 24%, 15% respectively) and total P (increased by 12%) for treatment of leachate. However, adding 10% zeolite did not make a significant difference ( $p < 0.05$ ). Adsorbents used in the experiment had a significant effect ( $p < 0.05$ ) on the parameters such as  $\text{N-NH}_4^+$ , SAR, total P and  $\text{Na}^+$  content. Changes in most parameters for the L<sub>3</sub> treatment were statistically significant ( $p < 0.05$ ) compared to other leachates (less  $\text{N-NH}_4^+$  (40%), total P (33%) and more  $\text{Ca}^{2+}+\text{Mg}^{2+}$  (14.3%),  $\text{Na}^+$  (14%)) indicating an increase in adsorbent efficiency due to rice husk activation. Therefore, it can be concluded that application of zeolite can improve soil removal efficiency for treatment of leachate, but the rates of application can be case sensitive depending on the soil and the type of zeolite.

### INTRODUCTION

Leachate is high strength wastewater due to high content of organic and inorganic compounds. Ammonium, suspended solids, and soluble metals are examples of inorganic compounds and ammonia and acetic acid are volatile inorganics. Discharging leachate without efficient treatment may result in severe pollution of water resources and serious environmental problems for receiving water bodies (Chernicharo, 2006; Marks et al., 1994). Many biological, chemical and physical technologies have been used for the removal of pollutant materials from leachate. Thus, the land treatment is considered one of the necessities of the environment. The most important consideration in application of methods is the cost of operation for the treatment. Several studies have used landfill drainage water in water management (Akkaya et al., 2010; Li et al., 2013). Adsorption is one of the most effective ways for leachate. Adsorbent is assumed as "low cost" if it requires little processing, is abundant in nature or is a by-product or drainage material from another industry. Of course, improved sorption capacity may compensate the cost of additional processing. Lee et al. (2005) have demonstrated the feasibility of using rice husk as a source of siliceous material. Rice husk with original composition silica and carbon, the granular structure

and low solubility in water, has chemical stability and high mechanical resistance, which its raw form is suitable for the removal of organic and inorganic contaminants (Apichat and Iekachai, 2010). Activated carbon is the most popular adsorbent that is used for a wide range of pollutants in the drainage water (Hale et al., 2013). Due to its high adsorption ability and biological and chemical characteristics, this material is a natural alternative as a filtering material. Zeolites are naturally occurring silicate minerals, which can also be produced synthetically. Clinoptilolite is the most abundant of more than 40 zeolite species (Ming and Dixon, 1987). Their adsorption properties result from ion-exchange capabilities. Sodium, calcium, potassium and other positively charged exchangeable ions occupy the channels within the structure. It holds several effective sites in its structure (Tabatabaei and Liaghat., 2004). Significant rainfall takes place in the southern strip of the Caspian Sea (Golian et al., 2010), where a number of municipal landfills are located. Drainage water from these landfill areas is part of the water basins, which supply the underground water resources. So, the leachate from them will eventually enter the water system. The pretreatment of leachates is important due to the porous and permeable texture of this area. To examine this, the present study intends (i) to investigate the effect of different adsorbents including rice husk and

activated carbon from rice husk as biological filters for leachate pretreatment and compare it with a situation lacking these materials based on changes in drainage water and (ii) to assess the feasibility of using Zeolite for land treatment of soil from landfill leachate.

**MATERIALS AND METHODS**

A soil column experiment was set up using a clay loam texture (sand: 32%, silt: 32% and clay: 36%) (Buyoucos, 1962) collected from a landfill area at 30 kilometers southwest of Babol in, Mazandaran province

in I.R. of Iran. The columns were made of P.V.C. 16cm (diameter) by 50cm (height) and were packed with soil mixed with zeolite in three levels to the original bulk density up to a height of 40 cm. Because collecting landfill leachate and drainage water from the area was not possible, leachate was obtained from the drainage of a compost factory at Kahrizak, Tehran (Mavaddati et al., 2010). Zeolite source used in the experiment was Clinoptilolite obtained from Afrand Tosca mine located at Semnan, Iran which was sieved to 2-4 mm in size (Table 1).

**Table 1.** Chemical analysis of the zeolite used in this experiment

SiO <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	K <sub>2</sub> O	MnO	TiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>	Na <sub>2</sub> O	Loss of Ignition(LOD)
66.5%	1.3%	3.11%	0.72%	11.81%	3.12%	0.04%	0.21%	0.01%	2.01%	12.05%

In this study, activated carbon was prepared from rice husk (Mohan et al., 2008). The experiment was set as a 3 × 3 factorial arrangement in a randomized complete block design with four replications in greenhouse conditions. The first factor was leachate pretreatment, using L1: primary leachate (no pretreatment), L2: leachate pass through rice husk, L3: leachate pass through activated carbon from rice husk and the second factor was the use of zeolite in three levels Z0; Z5; Z10: (zero, 5 and 10 wt%, respectively. After each irrigation, columns were set for a 6-day rest (MDE, 2003). For twelve weeks, they were irrigated with leachate. The amount of leachate was 750 mm (equal by 5 cm) added in three stages and time interval of 4 hours. The

drainage water of the soil columns were collected after 24 hours and collected samples were analyzed at time intervals of 1(W1), 3(W3), 5(W5), 8(W8) and 12(W12) weeks. Analysis performed on the water samples and soils are described in Table 2. The experimental set up and all administrative procedures were performed in the open air. Parameters measured in the leachate included COD, N-NH<sub>4</sub><sup>+</sup>(APHA, 1998), N-org, total phosphorus (T.P), pH, EC, Na<sup>+</sup>, Ca + Mg, Cl<sup>-</sup> and OC (Sparks et al., 1982). Statistical analysis was done using SAS 9.2, and Duncan is multiple range tests (5% level) were used for comparing means. Graphs were drawn using Excel 2010.

**Table 2.** Some properties of soil and leachate before the experiment

Parameter	pH	EC (ds/m)	Na <sup>+</sup> (mg/kg)	Ca <sup>2+</sup> +Mg <sup>2+</sup> (me/L)	SAR (meq/L)0.5	Ca <sup>+</sup> (meq/L)	COD (mg/L)	N-org (mg/L)	N-NH <sub>4</sub> <sup>+</sup> (mg/L)	N (%)	P (mg/kg) (mg/L)
Soil	7.8	0.74	48.7	4.47	1.45	2.0	-	-	-	0.04	15.27
Leachate	4.4	19.4	2998.4	122.3	16.67	34.47	60300	491.6	458.71	-	131.51

**RESULTS AND DISCUSSION**

A decrease in drainage water volume through the experiment can be attributed to the changes in the temperature and the increased evaporation since the columns were set in open air from mid-March to mid-June (Fig. 1). At the same time, the increased soil temperature resulted in higher water consumption to obtain the desired water content in the columns as the experiment progressed. A reduction in soil hydraulic conductivity can also be expected due to blocking of pore spaces, reducing the volume of drainage water. The maximum amount of loss was related to Z<sub>5</sub> treatment,

which represents the high capacity in outflow of water from columns.

Table 3 shows the changes in EC and SAR of leachates for different pretreatments and at different zeolite levels over the time of experiment comparing the values with primary leachate. A significant increase in Ca+Mg concentration and EC of leachates and decrease in Na concentration and SAR can be an indication of positive effects of pretreatments. A one-unit decrease in pH was observed over the experimental period (twelve weeks) (Table 5).

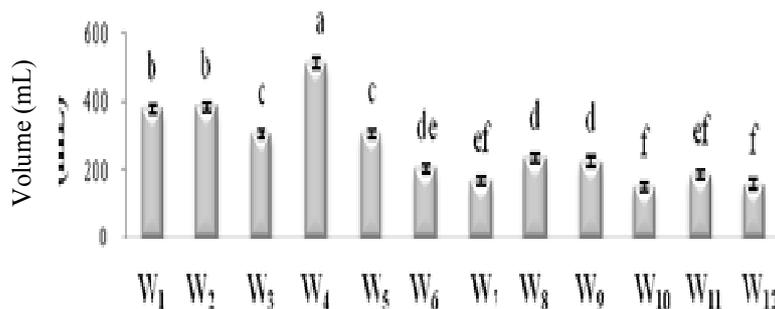


Fig. 1. Average volume of drainage water of the columns during 12-weeks (W) of the experiment

Table 3. Interaction effects of leachate (L<sub>1</sub>, L<sub>2</sub> and L<sub>3</sub>) and zeolite (Z<sub>0</sub> (only soil), Z<sub>5</sub> and Z<sub>10</sub>) on some chemical properties of drainage water columns compared to Pri.Leachate\* (without soil and only pretreatment)

		EC (dS/m)	Na <sup>+</sup> (mg/L)	Ca <sup>2+</sup> +Mg <sup>2+</sup> (meq/L)	SAR (meq/l) <sup>0.5</sup>
<b>Z<sub>0</sub></b>	L <sub>1</sub>	21.12a	1844.6d	248.47b	7.13d
	L <sub>2</sub>	27.97ab	4760.8a	374.47a	12.55ab
	L <sub>3</sub>	17.67c	1835.2d	224.67bc	8.42cd
<b>Z<sub>5</sub></b>	L <sub>1</sub>	17.45c	2320cd	224.2b	10.17bc
	L <sub>2</sub>	18.25c	2433.8cd	213.33c	10.92a-c
	L <sub>3</sub>	16.95c	2433.8c	209.07c	11.28ab
<b>Z<sub>10</sub></b>	L <sub>1</sub>	30.62a	4008.4b	245.37b	12.35ab
	L <sub>2</sub>	20.02bc	2841.5c	199.8c	12.77ab
	L <sub>3</sub>	26.99ab	4521.2ab	399.33a	13.37a
<b>Pri. leachate</b>	L <sub>1</sub>	19.41	2998.23	122.29	19.53
	L <sub>2</sub>	19.73	2771	146.36	16.8
	L <sub>3</sub>	20.77	2560.92	107.05	15.7

Table 4. Some chemical properties of drainage water after treatment in three levels of zeolite (Z<sub>0</sub>, Z<sub>5</sub> and Z<sub>10</sub>) and leachate (L<sub>1</sub>, L<sub>2</sub> and L<sub>3</sub>) during 12-weeks of the experiment

	pH	EC (dS/m)	Na <sup>+</sup> (mg/L)	Ca <sup>2+</sup> +Mg <sup>2+</sup> (meq/L)	SAR (meq/L) <sup>0.5</sup>	Cl <sup>-</sup> (meq/L)	COD (mgO <sub>2</sub> /L)	N-NH <sub>4</sub> <sup>+</sup> (mg/L)	N-Org (mg/L)	T.P (mg/L)	Vw (mL)
<b>Zeolite</b>											
<b>Z<sub>0</sub></b>	7.2a	22.25ab	2813.6b	282.5a	9.36b	222.1ab	26725a	3.78b	19.83a	3.47c	254b
<b>Z<sub>5</sub></b>	7.3a	17.5b	2383c	215.5b	10.79b	189b	19699a	4.66a	15.61a	3.96b	328a
<b>Z<sub>10</sub></b>	7.4a	25.88a	3685.7a	281.5a	12.8a	252.8a	28385a	3.78b	17.41a	4.58a	221b
<b>Leachate</b>											
<b>L<sub>1</sub></b>	7.3a	23.06a	2595.5c	239.34b	9.88b	218.8a	26152a	4.79a	13.53a	4.51a	267a
<b>L<sub>2</sub></b>	7.3a	22.08a	3332.6a	262.5a	12.08a	211.3a	27346a	4.59a	21.1a	4.48a	275a
<b>L<sub>3</sub></b>	7.3a	20.54a	3029.1b	277.7a	11.02ab	235.7a	21311a	2.85b	17.97a	3.03b	261a

Table 5. Some chemical properties of drainage water during 12-week treatment of the experiment

	pH	EC (dS/m)	SAR (meq/L)	Cl <sup>-</sup> (meq/L)	COD (mgO <sub>2</sub> /L)	N-NH <sub>4</sub> <sup>+</sup> (mg/L)	N-Org (mg/L)	T.P (mg/L)
<b>W<sub>1</sub></b>	8a	1.58d	1.26d	71e	89c	1.72e	3.9c	0.25d
<b>W<sub>3</sub></b>	7.3b	13.63c	6.6c	119.1d	22222b	2.42d	9.07bc	3.76c
<b>W<sub>5</sub></b>	7.4b	25.62d	6.51c	172.2c	25630b	3.21c	15.72b	3.38c
<b>W<sub>8</sub></b>	6.8d	29.32d	11.5b	291.5b	37926a	4.53b	12.23bc	4.5b
<b>W<sub>12</sub></b>	7c	39.33a	29.11a	452.6a	38815a	8.49a	50.9a	8.14a

\*Primary leachate (It was only transmitted pretreatment)

The decrease was perhaps due to organic and inorganic acids present in the leachate and increase in microbial activity of the soil or related to addition of some nutrient elements during the experimental period (Saber, 1986) (Table 4). The 5% zeolite treatment had the lowest hydraulic conductivity value which was 21% less than that of  $Z_0$  treatment. This treatment had the most volume of drainage water, which implies a decrease in EC through dilution of soluble salts (Table 4). EC mean of drainage water of the columns during 12 weeks was more than the initial value of EC in leachates (Table 5), indicating that water flow from the column increased EC in the drainage water. This result is contrary to Li (2013) who reported a decrease in EC of drainage water over a similar period from suitable flocculation of soil particles and the increased percentage of organic materials although soil texture in this study was different from that of the above experiment. The value of EC increased from the fifth week onwards in the drainage water and was higher than the EC of the input leachate. The reasons were (i) completion of soil adsorption ability and saturation of soil particle surfaces by presence of salts in the leachate and (ii) the heat of weather and evaporation reducing the volume of drainage water and dry weather or wind which increased evaporation and thus thickened the leachate solution (iii) blocking pores of soil due to accumulation of suspended solids of leachate decreasing hydraulic conductivity. In the 5% zeolite treatment, the

electrical conductivity of drainage water using three types of leachate did not show any statistically significant difference.

**Chemical oxygen demand:** COD is considered as the equivalent oxygen for containing organic material susceptible to oxidation by a strong oxidized agent (APHA, 1998). In this experiment, COD in the leachate changed from 60300 mg/L in  $L_1$  to approximately 45700 mg/L in  $L_3$  and 54667 mg/L in  $L_2$ . Activated carbon and rice husk removed COD by 25% and 10%, respectively. Mohan et al. (2008) reported that activated carbon from rice husk removed COD by 45-73%. Halim et al., (2011) reported 24-36 percent of COD removal by frequent application of activated carbon compared to 13-27% for rice husk. They stated that cation exchange capacity is an important factor in adsorption variations. Zeolite 5% had the least COD (Table 4). Over time, wastewater COD increased. This increase was more intense from the third week through the experiment. Apparently, the removal efficiency was not suitable up to this time. Thus, for satisfactory results, pretreatment operation on rice husk was required (due to the high initial COD) (Table 4). Interaction of zeolite and leachate was significant. With  $L_2$ , zeolite application caused to decrease COD. Adding zeolite preserved organic material of soil and prevented its removal. (Organic carbon of zeolite 5% = %1.46 and zeolite 10% = %1.97) (Fig. 2).

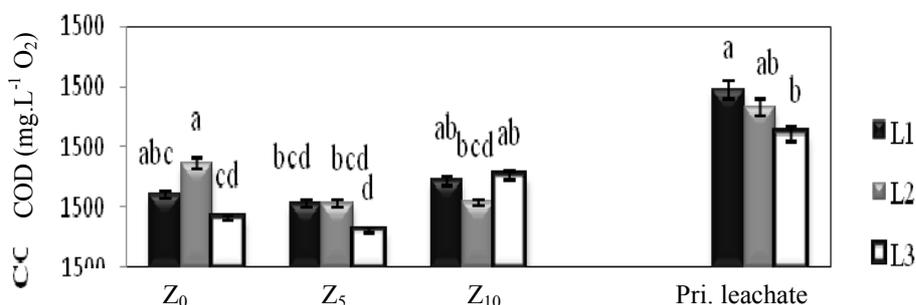


Fig. 2. Interaction effects of leachate and amounts of zeolite on COD of drainage water of the columns

**N-NH<sub>4</sub><sup>+</sup> and N-Org:** Zeolite 10% and  $L_3$  had the least amount of N-NH<sub>4</sub><sup>+</sup> (4% and 40% decrease, respectively) (Table 4). According to  $Z_{10}$  treatment, N-NH<sub>4</sub><sup>+</sup> concentration decreased while Na<sup>+</sup>, Ca<sup>2+</sup>+Mg<sup>2</sup> and EC increased. Ammonium ions were removed from aqueous solutions by zeolites via exchange with cations or by adsorption in pores of alumino silicate systems (Wang et al., 2006). Our results for changes in drainage water and comparison with the leachate content showed a 98% reduction of N-NH<sub>4</sub><sup>+</sup> through land treatment during this experiment, and in the last week, its amount reached 8.49 mg/L (Table 5). Leachate and zeolite interaction was statistically significant ( $P < 0.05$ ) (Fig. 3).

It was also observed that a significantly better performance for N-ammonia removal was obtained with the finer size particles than with the coarser particles. This is due to the slow diffusion of the ammonium ion into the coarser particles (Lazarova and Bahari., 2005). The removal efficiency of N-Org was 95-97% in this experiment. N-Org of initial leachate from 491.6 mg/L reached 487.1 mg/L in  $L_2$  and 372.2 mg/L in  $L_3$ . In the activated carbon, it was observed to decrease 24%. Although initial N-Org in  $L_3$  was less than the content after the experiment, it was not different in types of leachates (Fig. 3). Time changes of N-Org in drainage water had an ascending trend (Table 4).

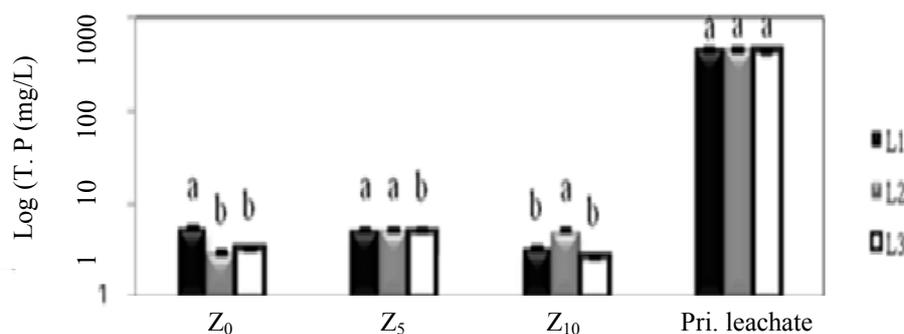


Fig. 3. Interaction effects of leachate and amounts of zeolite on N-NH<sub>4</sub><sup>+</sup> of drainage water of the columns

**Phosphorus:** Phosphorus content of initial leachate from 131.51 mg/L reached 120.92 mg/L in L<sub>2</sub> and 116.27 mg/L in L<sub>3</sub>. In the activated carbon and rice husk, it was observed to decrease 8% and 11%, respectively. L<sub>3</sub> had the least (13% decrease) and Z<sub>10</sub> had the highest increase of total P (Table 5). The interaction of zeolite and leachate was also significant (p<0.05). The application of L<sub>3</sub> leachate caused the 5% zeolite to have more P in its drainage water. It is

anticipated that for activated carbon treatment, the presence of carbonyl and hydroxyl groups on the carbon favored the adsorption (Kotdawala et al., 2008). While this was not observed in the other treatments (Fig. 4). P removal efficiency was very good (i.e., 97%). This is mostly controlled by physical (sedimentation) and chemical (adsorption) and biological processes of soil (Yalcuk and Ugurlu., 2009).

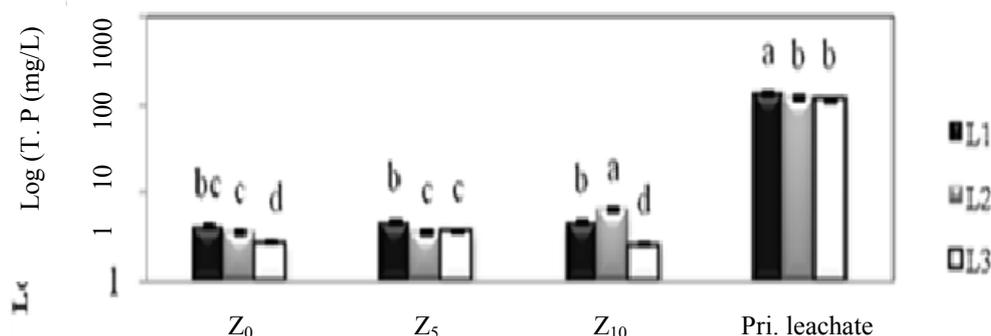


Fig. 4. Interaction effects of leachate and amounts of zeolite on Total phosphorus of drainage water of the columns

**Soluble Cations and Anions:** Table 4 shows the changes in the concentration of sodium, calcium and magnesium as well as chloride in the drainage water over the course of the experiment. The sodium of drainage water of L<sub>2</sub> had a 28% increase and 5% zeolite treatment had the least amount of sodium (15% decrease) compared to non-treated leachate. Acidic pH of leachate and replacement of ions in the exchangeable sites resulted in increasing sodium in drainage water and soil solution, which was proportionate to the zeolite content (Table 4). Ca<sup>2+</sup>+Mg<sup>2+</sup> concentrations of L<sub>1</sub> and 5%zeolite decreased 24% compared to non-treated leachate. This could be related to amorphous surface of rice husk that adsorb and release more Ca<sup>2+</sup>+Mg<sup>2+</sup> (Kamath and Proctor, 1998). The interaction of leachate and zeolite was statistically significant (p<0.05). The overall pattern of its changes was similar to those of EC and Na<sup>+</sup>. The 5%zeolite treatment had the least

concentration of chloride in the drainage water (Table 4). Apparently, at first, chloride ions were added to the columns by leachate, and then removed from the columns through the experiment with irrigation water. In the first week, they were 35 and 71 meq/L, respectively. Since the volume of drainage water collected were half of the leachate volume, it seemed that all chloride added to the soil was removed and the increase in concentration was due to a decrease in the volume. Over time, soluble chloride concentration increased (Table 5). Other researchers (Lazarova and Bahari, 2005) also reported increasing Cl<sup>-</sup> concentration through irrigation by wastewater. In general, the trend of changes of chloride was similar to that of EC in different treatments and during time. It was shown that the presence of Cl<sup>-</sup> ion played a role in the electrical conductivity of drainage water. Finally, the sodium adsorption ratio of treatments was calculated and

compared which indicates that zeolite 0, 5% and L<sub>1</sub> had the least SAR (Table 4). The results showed that rice husk and activated carbon could remove ions from drainage water and release them through exchange and adsorption mechanisms (Kadirvelu et al., 2001). This was observed in the present experiment about zeolite treatment. The SAR increased for all three types of leachate with increasing zeolite content (Table 3). Increase in SAR has been reported by other researchers following the use of wastewater and associated to sodium solubility in soil (Jahantigh, 2008).

## CONCLUSIONS

This experiment simulated the application of rice husk and activated carbon along with zeolite in land treatment of municipal waste leachate by comparing drainage water from soil. Our results showed that COD was the most sensitive (450 times increase) and chloride concentration was the most resistant parameter (6 times decrease) in drainage water with changes over time

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- (Table 5). The maximum amount of drainage water loss was related to 5% zeolite treatment and the amendment improved the structure and hydraulic conductivity of soil (Table 4). The increase in zeolite content increased adsorption of Na<sup>+</sup>, disturbed soil structure and reduced drainage water. Generally, 5% zeolite effects were greater on parameters of (EC, Na<sup>+</sup>, Ca<sup>2+</sup>+Mg<sup>2+</sup>, N-NH<sub>4</sub><sup>+</sup>, total P, Cl<sup>-</sup> and Vw). Adsorbents (L<sub>2</sub> and L<sub>3</sub>) were more efficient in changes of Na<sup>+</sup>, Ca<sup>2+</sup>+Mg<sup>2+</sup>, SAR, N-NH<sub>4</sub><sup>+</sup> and Total P (Table 5). Pretreatment of leachate with rice husk and activated carbon had only a significant effect on N-Org, Total P and COD. According to our results; applying activated carbon as a pretreatment and 5% zeolite in land treatment of municipal waste leachate can improve the quality of drainage water, effectively change chemical properties and increase its potential reuse. The experiment also showed that using adsorbents such as rice husk is more effective in the early stages of application.
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## مطالعه اثرات استفاده از شیرابه تیمار شده پسماند زباله شهری و زئولیت بر خاکها

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#### واژه های کلیدی:

زه آب

شیرابه

تصفیه زمینی

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جاذب

**چکیده** - همانطور که جمعیت جهان در حال رشد بوده و شهر نشینی و رفاه افزایش یافته، تولید زباله نیز به شدت در حال افزایش می باشد. استفاده مجدد از فاضلاب به عنوان یک راهکار برای کاهش کمبود آب، بهبود تولید محصول و پایداری محیط زیست شناخته شده است. به جهت بررسی این مسئله، آزمایش ستون خاک براساس طرح بلوک تصادفی فاکتوریل ۳×۳ شامل سه تیمار جاذب (شیرابه بدون پیش تصفیه (L<sub>1</sub>)، شیرابه عبور نموده از پوسته برنج (L<sub>2</sub>)، شیرابه عبور نموده از کربن فعال (L<sub>3</sub>)) و سه سطح زئولیت (۰، ۵ و ۱۰٪ وزنی خاک) انجام پذیرفت. حجم آب زهکشی شده در طول آزمایش کاهش یافت. کاربرد سطح ۵٪ زئولیت کارایی برداشت خاک را می تواند بهبود بخشد و اثر مثبتی بر کیفیت زه آب دارد. همچنان که به صورت تغییرات در EC، Na<sup>+</sup>، Ca<sup>2+</sup>+Mg<sup>2+</sup>Cl<sup>-</sup> (تا ۲۲٪، ۱۵٪، ۲۴٪، ۱۵٪ کاهش یافته) و فسفر کل (تا ۱۲٪ افزایش یافته) در تیمار شیرابه نشان داده شده است. اما افزودن ۱۰٪ زئولیت تفاوت معنی داری نداشت (p < ۰/۰۵). جاذب های مورد استفاده در این آزمایش اثر معنی داری بر پارامترهایی مانند SAR، N-NH<sub>4</sub><sup>+</sup>، فسفر کل و مقدار سدیم داشتند. تغییرات در اغلب پارامترها در تیمار L<sub>3</sub> از نظر آماری به طور معنی دار (p < ۰/۰۵) در مقایسه با دیگر شیرابه ها ((N-NH<sub>4</sub><sup>+</sup> ۴۰٪) و فسفر کل (۳۳٪) بیشتر و Ca<sup>2+</sup>+Mg<sup>2+</sup> (۱۴٪) و سدیم (۱۴٪) کمتر) متفاوت بود، که نشان دهنده افزایش کارایی جاذب ناشی از فعال سازی پوسته برنج می باشد. کاربرد زئولیت می تواند کارایی برداشت خاک را برای تیمار شیرابه بهبود بخشد، اما سطوح کاربرد بسته به خاک و نوع زئولیت متفاوت خواهد