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The Effect of Antibiotics on Urban Wastewater Cod Assessment and Their Control by Magnetically Activated Carbon

Mahsa Molasalehi[1]*

[1]Graduated with a master's degree in water and wastewater environmental engineering from West Tehran University, Tehran, Iran
*Corresponding author

Abstract

In this study, the effect of ten pharmaceutical micro pollutants from the categories of antibiotics drugs on the activated sludge process was studied. In order to express the extent of this undesirable effect, two indices, inhibition percentage and toxicity index, were calculated. The COD trend during the sludge process was considered as an evaluation factor.

In reactors containing 2 mg/L of each drug, it was observed that the COD removal rate was reduced by 49% compared to the control reactor. This impairment in COD removal varied depending on the type of drug.

Before evaluating the efficiency of this pretreatment method, the separation of activated carbon by two methods of coagulation, flocculation and magnetization was also evaluated. To this end, by adding using magnetized activated carbon for pretreatment up to a concentration of 5000 mg/L, the inhibition COD removal rate for wastewater contaminated with 20 mg of the drug combination reaches 16%. Also, the optimal contact time for effective removal of drugs by magnetic activated carbon is 90 minutes. The results showed that the adverse effect of pharmaceutical pollutants in wastewater reactors containing 2 mg/L of all drugs when passing through the pretreatment stage COD is reduced by 71%.

Keywords: COD –wastewater –Medicine-activated carbon–Activated sludge -Toxicity

1. Introduction

Medicines are a very important and integral part of modern life and are used to treat human and animal diseases. The presence of these substances in wastewater causes them to enter wastewater treatment plants, and given that in many pharmaceutical wastewater treatment sites, due to the absence or inefficiency of the drug removal system, high concentrations of them may enter the wastewater leading to urban treatment plants and, as a result, return to the water cycle. Among pharmaceutical compounds, antibiotics have a special place due to their high medical and veterinary uses. Some drugs have a long life in nature and can accumulate and combine with each other and reach a biologically active level, which has a direct impact on organisms. If the wastewater contains micro pollutants and toxic substances, it affects the quality of the wastewater. Therefore, it is important to examine the quality indicators of wastewater. One of the most important quality indicators of wastewater is COD (Chemical oxygen demand), therefore investigated in this study, the effect of drugs on the COD removal rate.

Therefore, 10 pharmaceutical micro pollutants from the antibiotics category, including (Ceftriaxone, Cefxime, Penicillin, Cephalexin, Ampicillin, Amoxicillin, Flocking, Gentamicin, Tetracycline and Trimethoprim) they were used separately and then in combination. In order to determine the effect of drugs on the activated sludge process, two parameters, inhibition rate and toxicity index, were introduced, and in order to reduce the adverse effects of pollutants, the adsorption process by activated carbon in the form of coagulation, flocculation, and magnetization was used as a pretreatment method. Finally, the undesirable effect of pharmaceutical pollutants in wastewater reactors was investigated in combination with all drugs when they pass through the pretreatment stage on COD removal rate.

2. Materials and methods

The drugs used in this study were used without purification and as received from the manufacturer .Taken Also, activated carbon, ferric chloride, nitric acid, ferric chloride II and III used in the synthesis of magnetic activated carbon, sulfuric acid used to stabilize wastewater samples obtained from the activated sludge process from Merck were purchased During the experiment of double distilled water to prepare the solution has been used.

Table 1. Drugs used in the tests

Manufacturer	Purity percentage(Weight)	Material
Pharma Chemistry	99<	Tetracycline
Tehran Medicine	99<	Ofloxacin
Lutman	99<	Cefixime
Pharma Chemistry	99<	Ampicillin
Pharma Chemistry	99<	Trimethoprim
Tehran Medicine	99<	Amoxicillin
Elixir	99<	Gentamicin
Pharma Chemistry	99<	Cephalexin
Lutman	99<	Penicillin
Tehran Medicine	99<	Ceftriaxone

2.1. Preparation of solutions

For preparing drug stock solutions(1000 mg/L) The required amounts of each drug were weighed and transferred to a 100 ml volumetric Balloon .liters were transferred and made up to volume with distilled water, acid or base. In order to prepare the solution High stock of Cefixime, Trimethoprim, ceftriaxone and penicillin, which has low solubility in water, Ethanol and acetone were used. Also, any amount of ethanol poured into the reactors The same amount was poured into the control reactor .The solvent was removed from the test. An ultrasonic bath was also used to completely dissolve the drug .Other solutions The required dilution of this solution The concentrations used in this study were obtained from the method of Ali Qardashi et al. (Alighardashi et al 2009)

2.2. Tools and devices used

In this study, a Haila air pump made in China was used for the pilot aeration process .Activated sludge has been used. Spectrophotometer model DR 5000 HACH Made in the USA, it was used to measure ammonium concentration. The device DO The meter, made in the United States, is used to measure the amount of dissolved oxygen during the activated sludge process. The device pH Hannah model made in the USA for measurement pH Sample Wastewater meters and a digital scale from Japan were used to weigh the samples. An ultrasonic bath was used to prepare the solutions. In order to separate the solid phase of the products from the liquid phase, filter paper was used.mm25 Whatman companies have been used.

2.3. Collecting wastewater samples

Sample Wastewater samples were obtained daily from the South Tehran Treatment Plant from the inlet to the activated sludge process.

Table 2. Collecting wastewater samples

COD	mg L-1 155
$N\text{-}NH_4$	mg L-1 24
TN	mg L-137

TP	mg L-1 4/8
pH	7/4
TSS[1]	mg L-1 110
Temperatures	°C 25

2.4. Synthesis of magnetic activated carbon

2.4.1. Synthesis of activated carbon washed with nitric acid

To perform this part, the method developed.(Jafri Kang et al. 2016) First 40 grams Slowly add the activated carbon to 200 ml .Add 1 liter of nitric acid (65%)and at a temperature of 80 degrees Celsius Grad is subjected to magnetic rotation conditions for 3 hours Let's give.

After the synthesis process is complete a vacuum filter was used to separate the activated carbon washed with nitric acid and wash it. For this purpose, a filter paper was placed at the bottom of the Buchner funnel and after turning on the vacuum pump, the mixture obtained from the synthesis was poured onto the filter and its nitric acid was separated from the activated carbon. Finally, the nitric activated carbon remaining on the filter was neutralized with soda. After that, it was washed several times with distilled water to prepare it for drying. Finally, the activated carbon washed with nitric acid was placed in the oven. Let it dry at 50 degrees for 24 hours.

2.4.2. Iron oxide synthesis

To make iron oxide from Fe^{+3} and Fe^{+2} Use for this purpose, 2.92 grams of ferric chloride2and 1.05 grams of ferric chloride3 Pour into distilled water and place under a nitrogen flow at 80°C in the absence of oxygen. Then 80 ml .Add 1 liter of ammonia (25%) dropwise over 30 minutes .While the mixture is under magnetic rotation, the iron oxide obtained from this step is finally separated by a magnetic magnet. and by 100 ml .Liters of hydrogen peroxide for washing let's give.

2.4.3. Synthesis of magnetic activated carbon

To perform this part, the method performed.(Baghdadi et al 2016) In the presence of nitrogen gas, the activated carbon washed with nitric acid obtained from the first step and the magnetic nanoparticles obtained from the previous step were mixed in a ratio of 1 to 4 in 500 ml of distilled water mixed Let's do it ,pH The mixture was brought to 4 and subjected to magnetic rotation in the presence of nitrogen gas for 1 hour at room temperature .The fabricated nanoparticles can be easily separated using a magnetic magnet and incubated at 50°C for 12 hours .Grad is placed in the oven .Then the product is exposed to a temperature of 110 degrees Celsius for 4 hours In order to remove iron ions adsorbed on the surface of magnetic activated carbon, the final product is washed with 0.2 M hydrochloric acid solutions, then washed with distilled water and dried at room temperature.

2.5. How to perform the test

2.5.1. Investigating the separate effect of drugs on the activated sludge process

In the first stage, we add one liter of activated sludge and two injections to each of the test reactors. Then, except for the control reactor, we add 20 mg L-1 of the desired drug to each of the reactors and aerate them with an air pump. The aeration process is carried out in such a way that the DO meter always shows the dissolved substances between 3 mg L-1 and 5 mg L-1 After the desired time, we take samples from each pilot and filter them with filter paper. To prevent the activity of microorganisms in the samples and stabilize them, we acidify the environment with sulfuric acid. Finally, COD content of the samples is measured.

[1]- Total suspended solids

2.5.2. Investigating the combined effect of drugs on the activated sludge process

In this stage, in order to investigate the combined effect of drugs on the activated sludge process, exactly the same as the previous stage, where the effect of drugs was investigated separately, with the same working conditions, with the only difference being that To the Your From Reactor Witness to Reactor Sample 12 mg- L Of all 10 Add medicine and finally the COD of the samples is measured.

2.5.3. Method review Activated carbon separation after the pretreatment stage

At this stage, in order to compare the efficiency of two methods of using coagulation and flocculation and magnetization of activated carbon for separation, Activated carbon synthesis at the end of the pretreatment stage, a test stage was designed and carried out.

For this purpose, 5 g/L of activated carbon was added to one of the two 2-liter volumes contaminated with 2 mg/L of drugs and 5 g/L of magnetic activated carbon was added to the other volume, and the volume was placed under rotation at 150 rpm for 90 minutes.

After this time, to separate Activated carbon separation was carried out using a magnet in the volume where magnetic activated carbon was used, and coagulation and flocculation processes were used in the volume where activated carbon was present to separate the activated carbon. The coagulation and flocculation processes were carried out using a Jar test device and 20 mg/l of ferric chloride was used as a coagulant. In this process, the mixture was placed in a jar machine for three minutes at a speed of 180 rpm for coagulation and then for 15 minutes at a speed of 20 rpm for flocculation. Finally, the solution after passing through this pretreatment stage, the wastewater entered the activated sludge process. In order to properly compare the results of two other reactors, including one reactor containing wastewater taken from the treatment House and another with sewage contaminated by the merger mg /l 2 of the drugs and without a pretreatment step were evaluated under conditions similar to the main reactors. For this purpose, the content COD of the samples are measured.

2.5.6. Investigating the effect of magnetic activated carbon in the pretreatment stage on the activated sludge process

To 2 liter wastewater samples containing concentrations mg L-12 of the 10 types of drugs are specified, desired amounts of magnetic activated carbon are added the samples were then mixed well for 90 minutes using a stirrer After this period, the magnetic activated carbon sample by iron the magnetic magnet completely separates the sewage. Example The results obtained from this step were introduced into a wastewater sample containing different concentrations of the 10 specified drugs (described in the previous two steps).Finally, the COD content of the sample It is measured.

2.5.7. Investigating the effect of time in the pretreatment stage of magnetic activated carbon

To 2 liter wastewater samples containing concentrations mg /l 2 of the 10 types of drugs are specified, adding 5 gr/l of magnetic activated carbon. Then the sample in time Different mix together. After that, the sample is magnetically activated carbon are completely separated from the wastewater by a magnetic magnet. Example the results obtained from this stage are also included in the development process as in the previous stage .No Wastewater is a mixture of pharmaceuticals. Finally, the COD content of the sample are measured be.

2.5.8. Measurement of COD parameter in wastewater samples

The prepared samples were analyzed for COD. For COD, according to the standard method, 2 ml of the sample was added to the COD measurement kit and after thorough mixing, heated to about 150°C for two hours. In the cold room, their COD levels were read and recorded by the DR5000 device.

2.5.9. Parameter introduction Evaluation of inhibition rate and toxicity index

In order to investigate the effect of drugs on the activated sludge process, two parameters, inhibition rate and its corresponding toxicity index, were used. These parameters, which are obtained from comparing the drug-contaminated reactors and the control reactor, were introduced. The next step is to obtain a measure of the toxicity of the drugs introduced into the process. To perform these calculations, use the method (Louver et al. 2010)Used.

2.5.10. Deterrence rate

To compare the rate of COD reduction in drug-contaminated reactors and the control reactor, a parameter called the inhibition rate per hour was used. The eighth and twenty-fourth sampling points are defined as follows.

Eq.1. Inhibition percentage

Inhibition percentage=

$$\frac{\text{Control reactor reduction rate} - \text{Drug contaminated reactor reduction rate}}{\text{Control reactor reduction rate}} \times 100$$

In this equation, the control reactor reduction rate is the amount of COD in the control reactor at hour zero minus the eighth or twenty-fourth hour, and the drug-contaminated reactor rate is the amount of COD in the drug-contaminated reactor at hour zero minus the eighth or twenty-fourth hour..

2.5.11. Toxicity index

Considering the first-order equation, the reaction rate for COD removal can be the following equation can be reached:

$Eq.2.$ rate for COD removal $$Ln\,\frac{COD_t}{COD_0} = -Kt$$

In this equation, the value COD_t
is COD In time, the value at the beginning of the process andNH_{4t}KThe coefficient of the equation is by considering the above equation for each reactor at any time from the start of the process in the reactor, the equation coefficient for each reactor will be obtained. If the coefficient obtained for the control reactor is K0 Named Let's put it, The toxicity index can be introduced as the following equation.

$$\frac{K}{K_0} = \text{Toxicity index} \qquad Eq.3.\,Toxicity\ index$$

By introducing this indicator, the toxicity level in each reactor can be determined at any hour from the start of the process. This allows the toxicity of drug-contaminated reactors can be easily compared, so the smaller the numbers obtained, the better. The toxicity of the drug to the activated sludge process is higher if it is wet. A distinctive feature of this index is that in determining it, all the numbers read from the sample are taken per hour the test results have the same role, while according to the inhibition coefficient formula, in the inhibition coefficient obtained for the eighth hour, the result obtained from the COD reading at the eighth hour plays a decisive role. And more than any other time, this issue is happening. Can cause errors in the results.

3. Results and discussion

Initially, wastewater COD parameters were studied to separately determine the effect of drugs on activated sludge.

3.1. Investigating the effect of drugs on reduction COD process in samples

In order to investigate the effect of drugs on COD reduction, the first 20 mg /L of each drug is added to the treatment samples separately, while the control sample is monitored without addition at the same time and under the same conditions. The test reactors were monitored for 24 hours. Sampling from the reactors to investigate COD levels was done at zero, one, two, three, four, five, six, seven, eight, twenty-three and twenty-four hours.

Results presented in Tables 3 to 6 has shown The COD content of drug-contaminated samples at the beginning of the experiments (time zero) was higher than that of the control reactor. This increase in COD is caused by the addition of pharmaceuticals to wastewater samples. Efficiency in all reactors is very low in the first hour due to the shock caused by changing conditions and entering the reactor environment. On the other hand, in all cases, the control reactor performs better in COD removal than the drug-contaminated reactor. To demonstrate the better performance of the control reactor compared to the drug-contaminated reactors, the percentage inhibition rate defined in Equation 1, which is a measure of the interference in COD removal in wastewater by drugs during the activated sludge process, is used. As is clear from the results shown in Table 4, the highest and lowest percentages of inhibition at the eighth hour are for cephalexin with 51% and tetracycline with 15%, respectively. At the twenty-fourth hour, the highest and lowest percentages of inhibition are observed for penicillin with 36% and tetracycline with 12%. Table 4 also shows the classification of drugs based on percentage of inhibition.

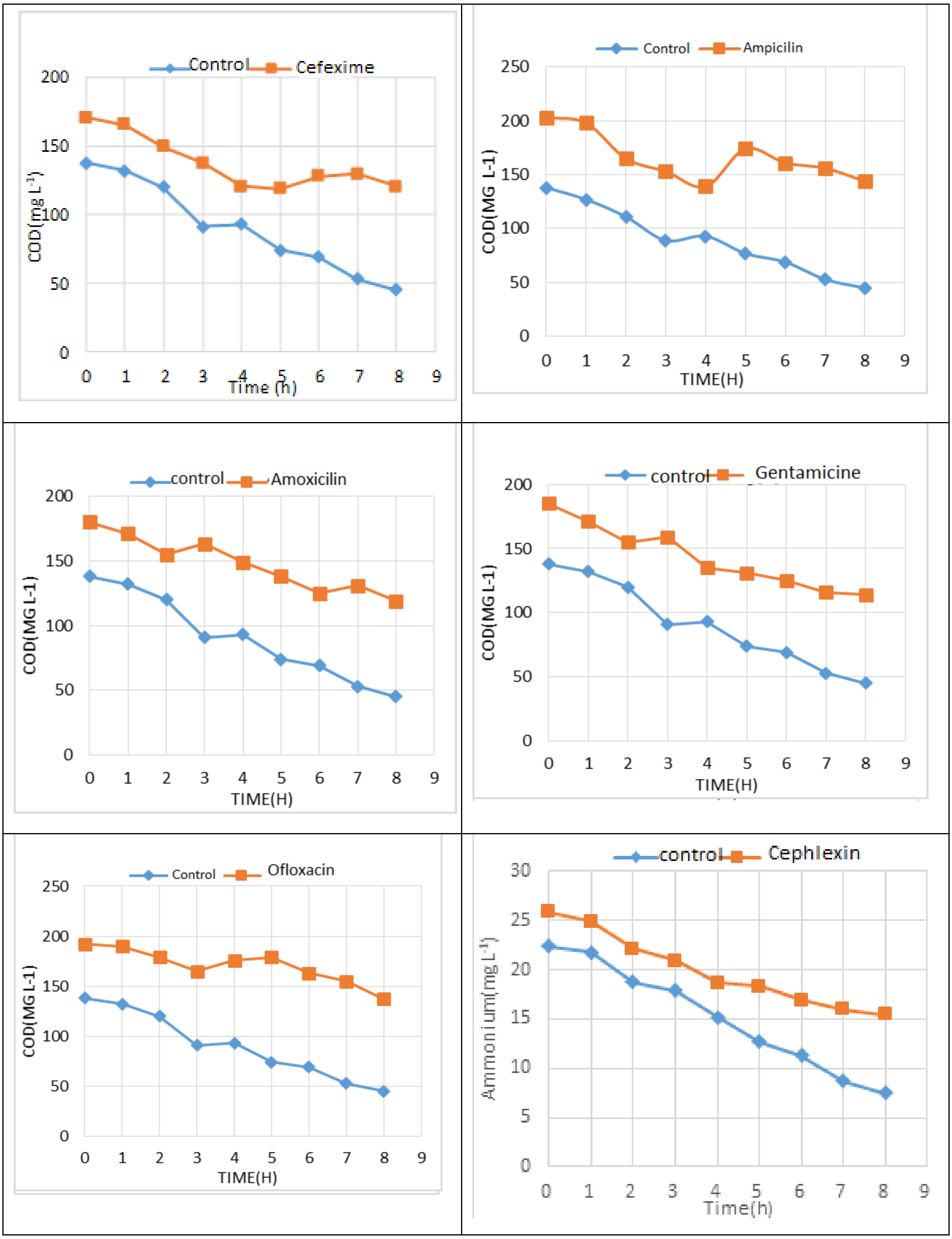
The reduction in the ability of the activated sludge system to remove COD is due to the fact that pharmaceutical micro pollutants introduced into the environment reduce the growth of active microorganisms in the process. According to the results obtained from Table 4, the inhibition rate for most drug-contaminated reactors and the control reactor at 24 hours is lower than at 8 hours. This could be due to the reduction of contaminants, which are the food sources of microorganisms, and the increased proliferation of microorganisms compatible with drug contamination over time.(Pasquini et al. 2013)

According to Figure 1, it can be seen that in the second to fifth hours of sampling, there is an increase in the COD level of the drug-contaminated reactors. This phenomenon may be due to the death of active microorganisms in the sludge and its conversion to COD in the test environment. (Louvet et al. 2010)

Table 3. COD levels in drug-contaminated and control samples at different sampling times in mg/l.

) hour time / reactor	**0**	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**23**	**24**
Control(the witness)	138	127	111	89	93	77	69	53	45	20	16
Amoxicillin	179	174	151	142	125	128	114	120	111	96	98
Cefixime	171	166	150	138	121	119	128	130	121	83	77
Tetracycline	181	179	158	141	136	128	133	120	115	103	104
Ampicillin	174	171	173	158	142	124	119	136	128	90	94
Ceftriaxone	203	198	165	153	139	174	161	156	144	123	129
Cephalexin	180	171	155	163	149	138	125	131	119	99	98
Ofloxacin	192	190	172	165	176	179	163	155	137	104	109
Penicillin	185	171	155	159	135	131	125	116	114	108	100
Gentamicin	162	151	144	128	109	103	94	91	83	51	49
Trimethoprim	186	182	171	151	149	162	155	135	123	110	102

Figure 1. COD content in drug-contaminated reactors compared to the control reactor in mg/l at different hours



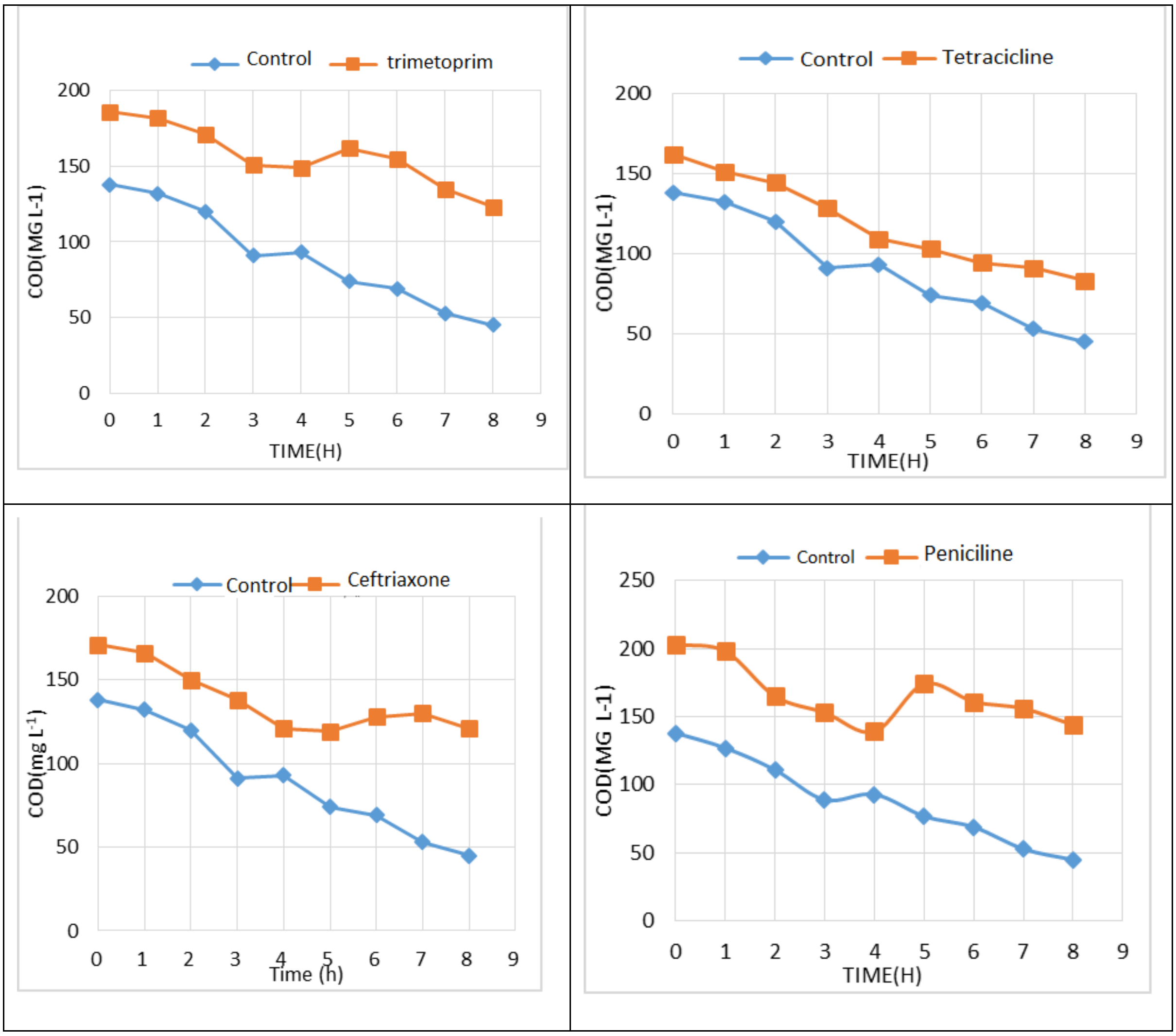


Table 4. COD removal rate of reactors and inhibition percentage in drug-contaminated wastewater samples and control samples at the eighth and twenty-fourth hours of sampling.

Sample name	Reactor of COD removal rate in mg/l per 24 hours	Percentage of inhibition at 24 hours	Reactor of COD removal rate in mg L-1 at 8th hour	Percentage of inhibition at 8 hours
Control(the witness)	115	-	93	-
Ceftriaxone	81	30	68	27
Cefixime	94	19	50	47
Sample name	Reactor of COD removal rate in mg/l per 24 hours	Percentage of inhibition at 24 hours	Reactor of COD removal rate in mg L-1 at 8th hour	Percentage of inhibition at 8 hours
Penicillin	77	36	66	30
Tetracycline	109	12	79	15
Cephalexin	80	29	46	51
Amoxicillin	88	22	70	25
Ampicillin	80	29	59	35
Ofloxacin	83	28	55	41

Gentamicin	85	26	71	23
Trimethoprim	84	27	63	32

Table 5. Classification of drugs according to their percentage inhibition of COD removal in eight hours

1٠% - 2٠%	2٠% - 3٠%	3٠% - 4٠%	40% - 5٠%	5٠% - ۶٠%
Tetracycline	Ceftriaxone	Penicillin	Cefixime	Cephalexin
-	Gentamicin	Amoxicillin	Ofloxacin	-
-	Ampicillin	Trimethoprim	-	-

Table 6. Toxicity index in COD removal for various drugs

reactor	K_0	K	Toxicity index
Control(the witness)	0/1422	-	-
Ceftriaxone	-	0/0615	0/432
Cefixime	-	0/043	0/302
Penicillin	-	0/057	0/400
Cephalexin	-	0/048	0/337
Ampicillin	-	0/033	0/232
Amoxicillin	-	0/0509	0/357
Ofloxacin	-	0/034	0/239
Gentamicin	-	0/0621	0/436
Trimethoprim	-	0/044	0/309

According to the results presented in Table 6 and Figure 1, the toxicity index can be introduced for each of the reactors. The advantage of this parameter over the inhibition percentage is that the reactor performance in the hours before the eighth hour also affects it. As can be deduced from Table 4, the drugs gentamicin and ampicillin with toxicity indices of 0.436 and 0.232 have the lowest and highest toxicity for the activated sludge process, respectively.

As is clear, the results obtained for the toxicity index are in predictable agreement with the results obtained for the inhibition percentage.

3.2. Investigating the combined effect of drugs on the rate of COD removal rate in samples

At this stage, in order to investigate the adverse effects of drugs on the activated sludge process, the concentration2 mg per liter of each drug in different reactors per sample Sewage disposals were added, so the total concentration of drugs in each reactor is equal to 20 mg per liter. The control sample was the same as the previous step without adding any Medicinal species, too the reactors were tested for 24 hours and under similar conditions. Sampling from the reactor To check the amount of COD per hour The experiments were carried out at times zero, one, two, three, four, five, six, seven, eight, twenty-three and twenty-four. Again, to compare the rate of COD reduction in reactors contaminated with the combination of drugs and the control, the inhibition rate parameter per hour was used .The eighth and twenty-fourth samples Vector was used.

The results showed that as the amount of drugs used in wastewater increases, Find the inhibition rate in COD removal at an increasing rate .It increases .Find it too Such a reduced toxicity index indicates an increase in the toxicity of drugs in the activated sludge process .The fact that the presence of pollutants Pharmaceuticals cause the death of microorganisms in activated sludge and as a result, serious disturbances in its efficiency Can it be confirmed It will be done.

Table 7. COD levels in combined drug-contaminated wastewater samples and control samples at different sampling times in milligrams per liter

) hour time / reactor	control	1mg L^{-1}	2mg L^{-1}	4mg L^{-1}
0	24/2	26/5	27/7	30/4
1	23/7	26/2	27/1	29/8
2	22/5	24/7	24/7	28/5
3	18/6	21/1	24/0	26/1
4	17/8	20/3	25/5	31
5	12/1	19/2	23/8	29/6
6	11/5	15/0	19/7	26/2
7	9/7	14/5	17/7	26/1
8	8/4	12/9	17/9	24/6
23	3/5	9/1	13/4	21/7
24	3/3	9/0	12/9	21/1

Figure 2. Logarithmic graph of reaction rates in COD removal for drug combinations

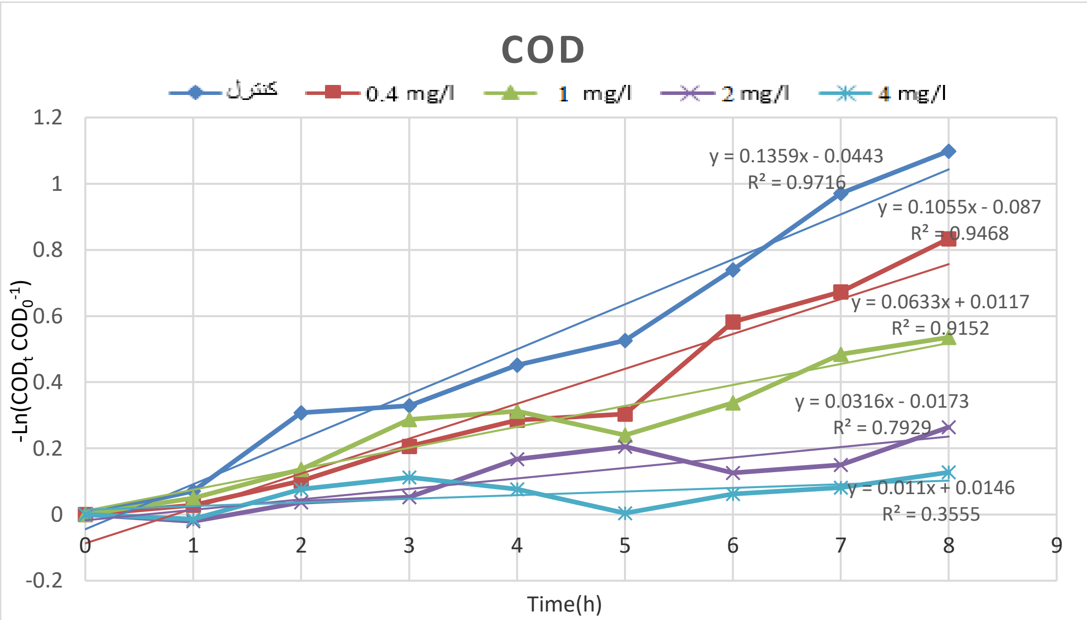


Table 8. Toxicity index in COD removal for combined drugs

	control	1 mg L^{-1}	2 mg L^{-1}	4 mg L^{-1}
K_0	0/1359			
K		0/0633	0/0316	0/011
Toxicity index		0/46	0/23	0/08

Considering this That the inhibition rate for 20 mg per liter of drug combination from the inhibition rate for a concentration of 20 mg per liter of most drugs taken individually, over 8 and 24 hours It is wet, may It can be said that this phenomenon occurs due to the intensifying effects of drugs. These effects are amplified .Can be disrupted by Molecule action The drugs interact with each other or with metabolites due to their different half-lives. Produced by it this issue highlights the importance of the presence of micro pollutants. Shows pharmaceutical applications in wastewater treatment processes Although the concentration of these substances in urban wastewater is low, due to their wide and increasing diversity and the possibility of their synergistic effects on each other, they can create new problems for the efficiency of the activated sludge process and, consequently, for treatment plants and the environment. (Wilson et al. 2003; Kim and Aga 2007)

3.3. Choosing the right activated carbon separation method for the pretreatment stage

From the results obtained in the previous sections, it is clearly evident that the presence of micro pollutants Pharmaceuticals in wastewater will cause disturbances in the activated sludge process. In addition, the failure to reliably remove these micro pollutants by wastewater treatment systems, resulting in their presence in surface waters, can have irreparable environmental consequences. Therefore, the use of pretreatment systems seems essential to prevent or reduce these destructive effects. (Bolong et al. 2009; Huerta-Fontela et al. 2011)

Considering the aforementioned information about the properties of activated carbon and its high ability to remove a wide range of pollutants Pharmaceuticals, its use to remove micro pollutants Pharmaceuticals in the pre-treatment stage can be a good option. On the other hand, to prevent any interference between the activated carbon and the activated sludge system and to allow a correct comparison between the performance of the sludge with and without the pretreatment stage, it is necessary that the activated carbon is removed from the environment after the pretreatment stage. To achieve this, two methods can be used: coagulation, flocculation, and magnetization of activated carbon.

The results in Tables 9 and 10 showed that using the method Pretreatments provide a good improvement in the inhibition rate and consequently increase the performance of activated sludge. It is also observed by comparing the two separation methods of coagulation and flocculation and magnetization of activated carbon. A reactor that uses the method of magnetizing carbon and separating it with iron The reactor used in the pretreatment showed better performance than the reactor in which activated carbon was used to separate the coagulation and flocculation process. Among the reasons for proper performance Wet separation by magnetic mixing ratio 4C: N: P this is when coagulation and flocculation are used in pretreatment. The proper ratio for aerobic pond operation is between 100:5:1 and 100:10:1(Thompson et al. 2006) . Also so, considering this although the coagulation and flocculation process performs well in removing phosphorus, there is a possibility that this ratio will be disturbed and as a result, there will be disturbances in the activated sludge process (Amuda and Amoo 2007). On the other hand, observations indicate that May the coagulation and flocculation process 20 g/l Chloroferric as a coagulant will not be able to completely remove activated carbon from the wastewater environment. This phenomenon has caused the activated carbon to enter the activated sludge process reactors, which as a result, in addition to some Materials Non-biodegradable added to the environment, not a suitable comparison with the control reactor was made. In addition, due to the high volume of sludge produced in the coagulation and flocculation process as a pretreatment method, it itself causes secondary problems for the removal and separation of the produced sludge. According to the results obtained, the magnetic activated carbon method was used in the subsequent experiments.

Table 9. COD changes in activated carbon reactors separated by coagulation and flocculation and magnetic methods, contaminated with drugs without pretreatment and control, over a period of four hours in mg/l.

) hour time / reactor	**control**	**With medicine and without pretreatment**	**Contains the drug and after pre-treatment and separation by coagulation and flocculation**	**Contains the drug and after pre-treatment and separation by magnetization of activated carbon**
0 hours	150	196	92	139
4 hours	91	160	49	87

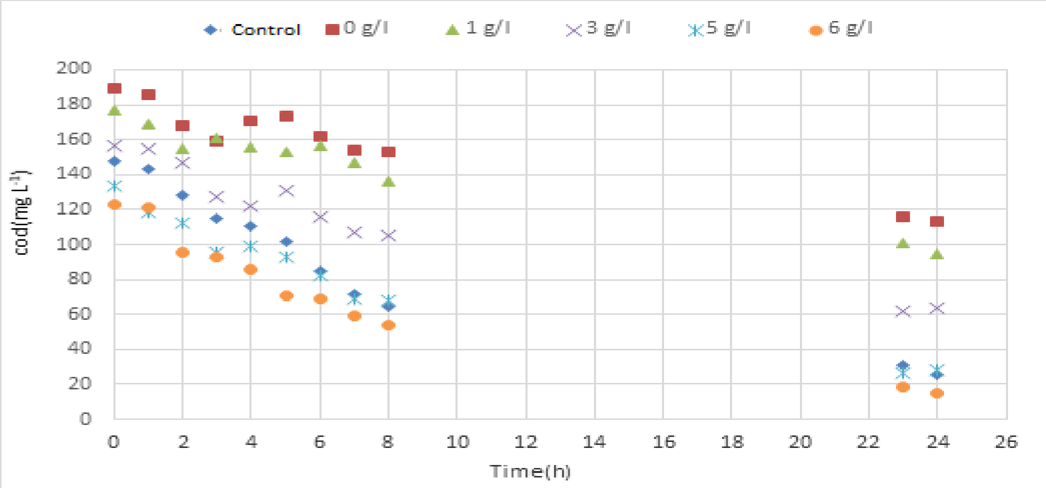
Table 10. Reduction rate and inhibition percentage for COD in activated carbon reactors separated by coagulation and flocculation methods and magnetic methods, contaminated with pharmaceuticals without pretreatment and control.

	Control	Contains medicine and no pretreatment	containing the drug and after pre-treatment and separation by coagulation and flocculation	containing the drug and after pretreatment and separation by magnetization of activated carbon
COD reduction rate in the reactor in the fourth hour in mg/l	**59**	**36**	**36**	**52**
Percentage of inhibition at the fourth hour	**-**	**39**	**39**	**12**

3.4. Using magnetic activated carbon pretreatment stage

At this stage, in order to investigate the effect of magnetic activated carbon, the concentration0, 1, 3, 5 and 6grams per liter of it to 2 liters of contaminated wastewater solution with 20 mg per liter of drug combination 2 mg per liter of each drug) was added. Then, after 90 minutes, the magnetic activated carbon was completely separated from the medium by a magnet. Finally, the solution Wastewater obtained in the reactor Separate reactors were subjected to the activated sludge process similar to the previous stages. In order to accurately compare with the previous stages, a reactor containing wastewater taken from the treatment House (control) in conditions similar to the reactor The originals were used.

Figure 3. COD recovery in pretreatment reactors with different activated carbon and reactor controllers



However, the effect of activated carbon in the pretreatment stage is low in reducing the initial amount of COD .The predominant mechanism of adsorption of substances on activated carbon is physical .Given that activated carbon does not have a surface charge due to its synthesis method, which is often carried out at high temperatures, its tendency to chemically adsorb charged species is very low (Park and Kim 2005).

The same As can be seen from the results and their comparison, with a drug-contaminated reactor without pretreatment (g L -1It is clear that the use of magnetic activated carbon has clearly increased the efficiency of the activated sludge process. In other words, the efficiency of the reactor with a pretreatment step using 6 grams of magnetic activated carbon compared to the drug-contaminated reactor without pretreatment (g L -10), increased by 70% in eight hours. Also, for COD an increase in efficiency of 56% was observed in eight hours.

Table 11. Toxicity index in COD removal for pretreatment reactors with different amounts of activated carbon and control reactor

	control	0 g /l	1g /l	3 g /l	5g /l	6 g/l
K_0	0/104	-	-	-	-	-
K	-	0/023	0/025	0/052	0/094	0/106
Toxicity index	-	0/22	0/24	0/5	0/91	1/01

The results showed that increasing the amount of activated carbon from 1 to 6 grams in the pretreatment stage had a positive effect on higher micro pollutant removal.in reactors as well as reducing the toxicity of drugs (increasing the toxicity index) in the COD removal process relative to the control reactor.

To achieve maximum efficiency of the activated sludge system, the effect of different concentrations of magnetic activated carbon on the efficiency of the pretreatment system was tested. The results showed that the inhibition rate decreased with increasing magnetic activated carbon concentration up to a concentration of 5000 mg per liter Significant increase By increasing the concentration of magnetic activated carbon to 6000 mg/min, per liter does not show much increase in system efficiency .Therefore, in order to save on the consumption of magnetic activated carbon, the amount of 5000 mg per liter of it as the optimal amount of absorbent to continue the experiment were considered.

3.5. Investigating the effect of magnetic activated carbon contact time in the pretreatment stage

In the last stage of this study, in order to investigate the effect of magnetic activated carbon contact time on the efficiency of the pretreatment stage, the concentration5Grams per liter of magnetic activated carbon to 2 liters of wastewater solution contaminated with 20 mg/liter of drug combination Then magnetic activated carbon was added at30, 90 and 120 minutes in contact with the sample After this time ,Magnetic activated carbon by iron The magnet was completely separated from the medium. At the end of the sample pretreatment stage Pre-treated wastewater in the reactor Separate reactors were introduced into the activated sludge process as in the previous stages. In order to accurately compare with the previous stages, two control reactors, one of which Contains wastewater taken from treatment House and another contaminated with 20 mg of drug combination (drug-contaminated control), in reactor-like conditions The main ones were used.

Table 12. COD levels in combined drug-contaminated wastewater samples after passing through the pretreatment stage and control samples at different sampling times in milligrams per liter

) hour time / reactor	control	Drug contaminated control	30 min (5 g/l)	90min (5 g/l)	120min (5 g/l)
0	145	183	135	125	117
1	141	173	129	113	106
2	128	157	109	94	99
3	108	192	98	92	84
4	102	178	94	91	76
5	86	173	99	75	80
6	79	146	91	71	54
7	61	138	75	58	53
8	56	140	73	53	42
23	27	118	44	21	14
24	21	111	40	22	16

Table 13. Toxicity index for COD removal in pretreated reactors with different pretreatment times and control reactors

	control	Drug contaminated control	30min 5gr/l	90min 5 gr/l	120min 5gr/l
K_0	0/125	-	-	-	-
K	-	0/033	0/074	0/103	0/124
Toxicity index	-	0/246	0/592	0/8۴	0/99

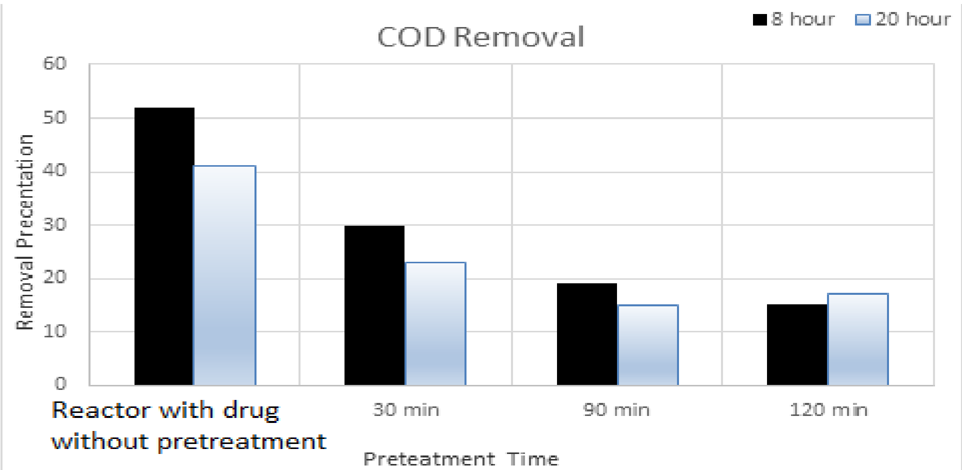


Figure 4. Column chart of the percentage inhibition in COD removal in pretreated reactors with different pretreatment times at eight and twenty-four hours.

According to the results obtained and their comparison identified with the drug-contaminated control reactor By increasing the contact time of magnetic activated carbon with the sample wastewater, the inhibition rate for COD removal is significantly reduced, it shows .But in time Higher doses did not significantly change the inhibition rate after 90 minutes .For example, at eight hours after the start of the activated sludge process, the COD inhibition rate at 90 minutes compared to 120 minutes only decreased by 4 and 1 percent, respectively .This also applies to the toxicity index Slow. In other words, it can be said that 90 minutes is enough time for the efficiency of magnetic activated carbon in the pre-treatment stage to reach its maximum value.

4. Conclusion and suggestions

The results of this research are listed separately below:

1) Presence of micro pollutants Pharmaceuticals in wastewater have a negative effect on the efficiency of the activated sludge process as a common they have the most advanced wastewater treatment method.
2) The effectiveness of the different drugs used in this study varies .Be.
3) The highest inhibition in COD removal was related to cephalexin with an inhibition rate of 51% and the lowest effect was related to tetracycline with 15%.
4) The highest and lowest toxicity according to the defined index in COD removal are related to ampicillin with a toxicity index of 0.232 and gentamicin with an index of 0.436, respectively.
5)
6) After the test It was found that the inhibition rate for 20 mg per liter of drug combination from the inhibition rate for a concentration of 20 mg per liter of most drugs are given individually, at the time Various sampling methods The reason for this phenomenon It is can cause the effects of medications to intensify and interfere with Its action with one It may be another.
7) Due to disturbances in the efficiency of the activated sludge system due to the presence of medicinal wastewater, using a pretreatment system to remove micro pollutants Medicinal properties were investigated. Considering the properties of the idea Magnetic activated carbon in micro pollutant removal as well as its ease of separation from wastewater, this material was tested as a suitable option. Using this system increased the efficiency of the system in removing COD by up to 71%.Be.
8) Using magnetic activated carbon as a pretreatment agent reduces the COD content of the sample .Sewage treatment plants this reduction amount varies depending on the amount of magnetic activated carbon used.
9) By increasing the amount of magnetic activated carbon used to a concentration of 5000 mg per liter of May The efficiency of the activated sludge process was increased. So The inhibition rate in COD removal for wastewater contaminated with 20 mg of drug combination (2 mg) of each of the 10 drugs) only accounts for 14 percent It arrives.
10) Sufficient contact time between samples Drug-contaminated wastewater and magnetic activated carbon increase the efficiency of the pretreatment system .The minimum time required for effective drug elimination is by magnetic activated carbon is 90 minutes. The results of this experiment showed that time Times above 90 minutes will not have much effect on the performance of the pretreatment system.

Suggestions:

Given the high per capita drug consumption in Iran compared to global standards and its very destructive effects, on human health and the environment. Initiate comprehensive research to determine the exact concentration of these substances in wastewater .and water Surface treatments seem necessary .Due to the lack of complete knowledge of all the destructive effects of this micro pollutant The impact on the environment requires serious attention to reforming the pattern of drug consumption in the country. Also, by strictly implementing regulations and guidelines Relevant regulations prevent any pharmaceutical and hospital wastewater from entering the wastewater Urban and water Finally, necessary measures to improve the performance of the system Urban wastewater treatment plants should be considered.

-Research work proposals

The following suggestions are made for future research work:

-A detailed examination of break again works Action Effects of drugs on each other and metabolites the results of that on system efficiency Wastewater treatment plants.

- System usage review other pretreatment methods such as zonation to improve the efficiency of the activated sludge process.

-Due to the low efficiency of the discontinuous system, research is needed to improve the efficiency of the pretreatment process and reduce the amount of activated carbon consumed, using the method other pretreatment systems including the Column or ultrafiltration.

-Due to the relatively high price of synthetic activated carbon, the use of the adsorbent Natural carbon-based materials that are very low in price More and more compatibility Environmentally friendly materials are being investigated and researched as an alternative to activated carbon.

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Conflict of interest:

The author has no financial conflicts to declare related to this article.

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