Wastewater Treatment Using Horizontal Subsurface Flow Constructed Wetland

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Abstract: The last few decades witnessed sharp focus on environment pollution and its impact on life in nature. Wetlands can be used for biological treatment of wastewater. **Problem statement**: Scarcity of water is considered as a global problem and Iran is one the countries which is facing water shortage problem. Pollution of water bodies restrict the availability of water for various uses. Treatment of waste water before disposal contributes to water conservation efforts. Constructed wetlands are techniques aim to polish water quality and reduce the harmful effect of effluent. **Approach**: In this study, four horizontal subsurface flow wetlands (HSSF) were constructed at the Research Station of Tehran University, located in Karaj, Iran. The study was carried out from April to September, 2007. Gravel and *zeoilte* were used in this study as substrate. Gravel-beds with and without plants (called GP and G) and gravel-beds mixed with (10%) *zeolite*, with and without plants (called ZP and Z) were examined to investigate the feasibility of treating synthetic wastewater which was specifically produced and modified to imitate agricultural wastewater. **Results:** The results of this study indicated that the system had acceptable pollutant removal efficiency and that both plants were found to be tolerant under the tested conditions. The wetland system could achieve the NO_3-N removal of (79%) in ZP, (86%) in Z, (82%) in GP and finally (87.94%) in G. As for the P removal, the efficiencies of 93, 89, 81 and 76% were respectively achieved for ZP, GP, Z and G. The outflow concentrations of Pb and Cd were found to be under the detection limit; however, as for Zn, the removal efficiencies of 99.9, 99.76, 99.71 and 99.52% were concluded for ZP, Z, GP and G respectively. **Conclusions/Recommendations**: It can be concluded that constructed wetlands are efficient in removing Zn, Pb and Cd from agricultural wastewater. Plants types such as Phragmites Australis and *Juncus Inflexus* can contribute in treating wastewater, while *Zeolite* and gravel materials provide a suitable plant growth medium to replace conventional sand and gravel substrates. So it is highly recommended to use Constructed wetland for treating wastewater before disposal.

Key words: Constructed Wetland, nitrate-nitrogen, phosphorus, wastewater, zeolite, Zn

INTRODUCTION

Many countries around the world are experiencing water stress and scarcity during a large part of the year and they are exploiting reserves which are being not sufficiently replaced[1]. Iran is also going to experience water scarcity by 2025, based on the availability of less than 1000 m^3 of renewable water per person per year[2]. In addition to the natural scarcity of freshwater, the quality of the available freshwater is also getting worse; this is due to pollution which is a result of water shortage. Agricultural and residential wastewaters contain high levels of nutrients and if not treated, they can contaminate surface water and groundwater systems. The treatment of wastewater using Constructed Wetland (CW) is one of the treatment systems which are used in many parts of the world.

This system seems to have the potential to be one of the solutions in discharging the huge amount of wastes and getting access to safer drinking water. Constructed wetlands (CWs) are treatment systems that have been designed to accomplish natural processes containing wetland substrate, vegetation and the associated microbial assemblages to help in treating wastewaters and take advantage of the processes that occur in natural wetlands within a more controlled environment. Nitrogen (N) and Phosphorus (P) are the nutrients of concern for removal in wetland systems. The removal mechanisms for N include uptake by plants and microorganisms, ammonification, nitrification, denitrification, ammonia volatilization and cation exchange for ammonium[3]. The removal mechanisms for P include chemical adsorption and precipitation in substrate and biological transformations[4] and also plant uptake in a lower percentages[5].

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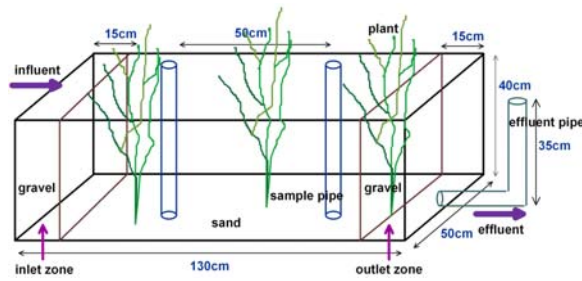
Fig. 1: Schematic 3D view of wetland cell

Industrialization in developing countries with an increasing demand for heavy metals results in a high emission of these pollutants into the biosphere. Heavy metal pollution in water bodies is a serious environmental problem, threatening the aquatic ecosystems and human health. Heavy metals are not degraded through biological processes. Some of the important heavy metals of concern in wastewaters are Ni, Mn, Pb, Cr, Cd, Zn, Cu, Fe, Hg and some toxic elements like As, B, Na. Among these we can mention functions of some them in CW. The main objectives of this study were to determine the efficiency of the horizontal subsurface flow constructed wetland (HSSF) system in treating wastewater under the climatic condition in Iran and to determine the effect of gravel and *zeolite* as the media of treatment in constructed wetlands.

Materials and methods: In this study, four HSSF CWs systems made of polyethylene, each with a surface area of 0.65 m^2 $^($1.3×0.5 m) and 0.4 m depth Fig 1 was conducted at Research Station of Tehran University, located in Karaj, Iran. A slope of (1%) was required in placing the cells to maintain the hydraulic gradient. The inlet zone consisted of 4 inlet points (one for each cell) attached to a water control tank with a float installed after the storage tank and a container as a water source. The control structure was designed as the inlet water could enter with a constant head to maintain the water level in the system, as well as to avoid shock loading from the storage tank due to water level fluctuation in the tank. The outlet zone was made of perforated PVC pipes attached at the bottom of each cell. The edge of these pipes was connected to adjustable flexible hoses which served as risers for maintaining water level in the bed. Two perforated tubes covered with *geotextile* were inserted into the middle part of the substrate 40 cm apart from inlet and outlet to allow sampling. The gravel with the size of 10-15 mm was put into the inlet and outlet zones in each cell in order to produce a uniform distributed flow. Then the remainder area of

two cells was filled with fine gravel and the other two with mixure of fine gravel and *zeolite* (10:1 weight ratio). An equal mixture of two plant species, *Phragmites australis* and *Juncus inflexus* for vegetation with an initial density of 30 plants m^2 was selected for two cells (one with *zeolite* and one with gravel only). According to the types of substratum media and conditions with or without vegetation, four treatments were thus installed: gravel-beds with (10%) *zeolite* (ZP) and without vegetation (Z) and gravel-beds (GP) and without vegetation (G). The influent for all four treatment systems was artificial wastewater prepared prior to each feeding by mixing urea $(NH_2)_2CO$ and Ammonia phosphate $(NH_4)_3PO_4$ as chemical fertilizers used in agriculture. Such synthetic wastewater contained approximately 80-100 mg L^1 of NO_3 and 10 mg L^1 of TP, 1, 2 and 10 mg L^1 of Cd, Pb and Zn respectively. The required chemicals were mixed in 1000 L polyethylene container, using tap water. A valve was used to regulate flow. The synthetic wastewater was remained 20 hours constant in order to eliminate the residual chlorine.

A first order plug flow model based on nitrate removal was used and it is described by the following equation [6]:

$$As = \frac{Q*(\ln C_o - \ln C_e)}{K_t * d * n} \qquad (1)$$

The value of K t is give by:

$$K_t = 1.15^{(T-20)} \qquad (2)$$

Where:
A = Area (m^2)
Q =Average flow (m^3 day^1)
C_o = Influent NO_3 (mg L^1)
C_e =Effluent NO_3 (mg L^1)
K_t =Temperature-dependent rate constant
d = Depth of gravel bed
n =Porosity

According to the average temperature of the site and knowing n and d we can decide the area.

For t = 25, K_t = 1.15$^{[25-20]}$ = 2.011, A = 0.65 (m^2), C_0 = 100 (mg L^1), C_e = 5(mg L^1), d = 0.4 (m) and n = 35%, the value of Q is computed using Eq. 1 and it is found to be 0.060 m^3 day^1.

The amount of evapotranspiration is calculated and found to be 7 mm day^1 for each cell, therefore value of Q for the four cells is found to be equal to Q = 0.078 m^3 day^1. HRT and HLR can be calculated using the following equations:

$$HRT = \frac{A \times d \times n}{Q} \qquad (3)$$

$$HLR = \frac{Q}{A} \qquad (4)$$

Where:
HRT = Hydraulic retention time
HLR = Hydraulic loading rate

The HRT and HLR can be calculated and their values were found to be 1.2 days and 0.12 m day[1], respectively.

The systems were installed in April 2007. After two month form the planting date, the wastewater was operated into the system and water samples were then collected for 3 months. Water samples were taken from inflow, outflow and the sampling pipes every two week in order to measure TP, NO_3-N, Zn, Pb and Cd. The analytical methods were referenced by [7].

Statistical test: Statistical analyses were performed using SPSS 15 for Windows. Normality of the distribution of the metal concentration in the influent and effluent was tested by means of the Kolmogorov-Smirnov test of normality (α = 0.05). All concentrations in the water were normally distributed. Significant differences between inflow and outflow water with regard to concentrations, was determined by the paired-samples t-test. Also the Duncan's Multiple Range test was run to determine the significant differences between 4 treatments. The same test was run to determine significant differences between location 1, 2 and 3 (effluent) in each cell. Pearson correlation coefficients (α = 0.05) were determined

between mean concentrations of wastewater of the different sampling time.

RESULTS

All macrophytes were found tolerant under the tested conditions. It was not possible to continuously monitor the variable flow rate in the outlet of the cells; therefore the removal efficiency was determined from concentrations and not from loadings. Both influent and effluent concentrations and removal statistics of TP, NO_3-N and Zn are shown in Table 1. Although it is seen that the mean outflow concentration values are not statistically different among the 4 cells, but concentration reduction was statistically significant in all treatments.

As for nitrate removal, the mean outflow concentration of all the four cells shows that the difference is not statistically significant, as proven by the statistical tests. The unplanted cell with gravel (G) had a removal efficiency of (86%). The unplanted cell with gravel (G) had a removal efficiency of (86%). Similarly, the cell with *zeolite* (10%) and gravel, without vegetation (Z) had a removal efficiency of (88.5%), while the other two planted cells ZP and GP had removal efficiencies of (81%) and (77.6%), respectively (Figure 2). A comparison of the inflow and outflow concentrations of NO_3-N during the study period is shown in Figure 3. It indicates that the planted and unplanted cells show a significant reduction in NO_3-N concentration. Nevertheless, it is obvious that the planted cells (ZP and GP) had lower removal efficiency as compared to the unplanted ones (Z and G). In the case of phosphorus removal, the planted system with (10%) *zeolite* + gravel as a substrate (ZP) had a removal efficiency of (92.7%).

Table 1: Total concentrations in the inflow and outflow of systems together with standard deviation (SD), range and removal efficiency

	Variable		NO_3-N	P	Zn
Influent Concentration (mg l[1])	I	Mean±SD	79.3±32.4	10.5±1.04	806±2.7
		Range	110-20	12.0-9.0	12.0-5.0
	A	Mean±SD	17.71±9.34a	0.76±0.58c	0.011±0
		Rnge	30-2	1.7-0	0.047-0
		Removal (%)	79.19	93.12	99.9
		Mean±SD	9.3±4.8a	1.95±0.7ab	0.019±0.018a
	B	Range	19.5-4.5	3-0.83	0.055-0
Effluent Concentration (mg l[1])		Removal (%)	86.58	81.76	99.76
		Mean±SD	15.14±8.27a	1.14±0.63bc	0.022±0.019a
	C	Range	28-1	2-0.33	0.057-0
		Removal (%)	82.39	89.47	99.71
		Mean±SD	11.0±2.6a	2.5±1.1ab	0.037±0.019a
	D	Range	13-6	4.2-1.33	0.062-0
		Removal (%)	83.38	76.65	99.52

Note: Means with the same letter(s) in each column are not significantly different, I = Inflow

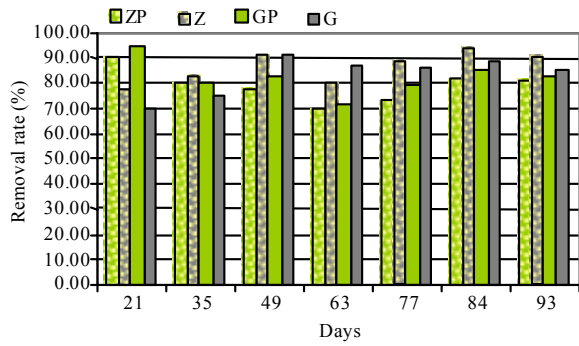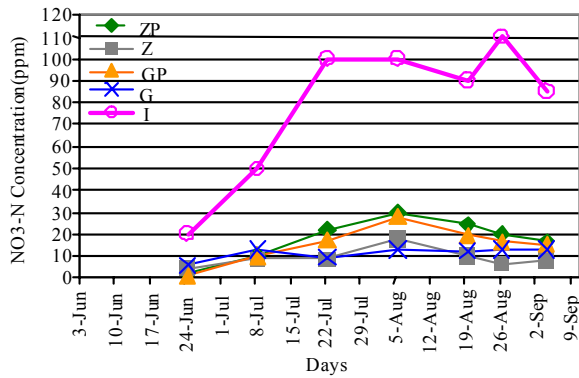Fig. 2: NO$_3$-N removal efficiency during the study period

Fig3: Inflow and outflow concentration of NO$_3$-N during the operation period

Fig 4: P removal efficiency during the study period

Fig 5: Inflow and outflow concentration of P during the operation period

The cell with the same substrate without vegetation (Z) had a removal efficiency of (81.4%). And the other two systems with gravel media (GP and G) had removal efficiency of (89%) and (76%) which refers to a planted and unplanted system respectively. The removal efficiency of P in each sampling during the study period is shown in Fig 4.

The same result got from mean values can be obtained from Figure 4. The experimental results of P inflow and outflow concentration levels are shown in Fig 5. The outflow concentrations had a significant reduction in comparison with the inflow concentration levels. A much better removal efficiency of planted cells (almost 12%) is shows in Figure 5. The concentrations of P in the influent were very low with average value of 1.5 mg L^{-1}. The same result got from mean values can be obtained from Figure 4. The experimental results of P inflow and outflow concentration levels are shown in Fig 5. The outflow concentrations had a significant reduction in comparison with the inflow concentration levels A much better removal efficiency of planted cells (almost 12%) is shows in Fig 5.
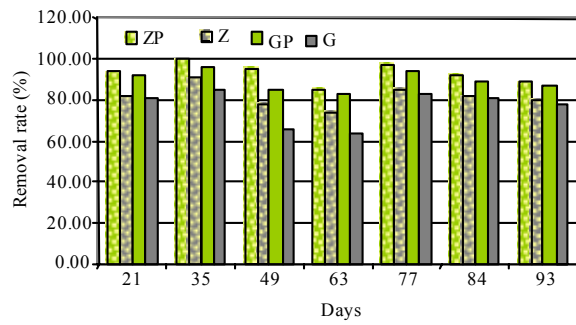
. The concentrations of P in the influent were very low with average value of 1.5 mg L^{-1}. It was also found that P had a regular behavior during the study period which is shown by the relatively low standard deviation of the efficiency values Table 1.

The removal efficiency and the concentrations of Zn in the influent and effluent are shown in Table 1. Based on the finding, the removal rates of heavy metals were found to be almost (100%). The concentrations of Cd and Pb in the effluent were lower than the detection limit, indicating that the systems were highly efficient in removing heavy metals from polluted water. Hence, the following section will put forward a discussion of the effect of Zn.The planted cell with *zeolite* and gravel (ZP) had the highest removal efficiency of (99.89%), whereas, the cell with the same substrate without vegetation (Z) had the second removal efficiency of (99.76%). The other two units with gravel, with and without vegetation (GP and G) had removal efficiencies of (99.70%) and (99.52%), respectively Figure 6. As for Zn, a decrease in the total concentration between the influent and effluent is observed during monitoring, as shown in Figure 7.

Table 2 shows the values of pollutant concentrations along the four units for the entire operation period.

Table 2: Variation of concentration along the cell's length

|		ZP			Z		
		ZP1	ZP2	ZP3	Z1	Z2	Z3
NO$_3$	Mean±SD	14.5±2.5	13.3±1.2	17.71±9.34	10.7±2.6	9.3±4.8	
	Range	25-5.5	20-10	30-2	15-5	25.5-5.5	19.5-4.5
	Removal (%)	81.71	83.15	77.66	86.40	78.38	88.47
P	Mean±SD	2.1±0.36	1.37±0.26	0.76±0.58	2.7±0.27	2.13±0.26	1.95±0.7
	Range	4-1.1	2.5-0.66	1.7-0	3.5-1.93	3.2-1	3-0.83
	Removal (%)	79.92	86.95	92.78	74.49	79.71	81.39
Zn	Mean±SD	0.28±0.01	0.055±0.014	0.011±0.017	0.1±0.15	0.017±0.045	0.019±0.018
	Range	0.65-0.015	0.111-0	0.047-0	0.77-0.014	0.14-0	0.055-0
	Removal (%)	97.11	99.34	99.90	98.53	99.47	99.76
		GP			G		
		GP1	GP2	GP3	G1	G2	G3
NO$_3$	Mean±SD	12.1±3.2	10.5±2.2	15.14±8.27	16.7±1.5	12.3±1.5	11.0±2.6
	Range	22-0.5	18.5-2	28-1	21.5-9.5	15.5-5	13-6
	Removal (%)	84.68	84.76	80.90	78.92	84.50	83.38
Zn	Mean±SD	2.66±0.47	1.84±0.38	1.14±0.63	3.27±0.39	2.7±0.4	2.5±1.1
	Range	0.723-0.016	0.277-0	0.057-0	1.2-0.035	0.575-0	0.062-0
	Removal (%)	96.62	99.30	99.71	96.55	98.73	99.52
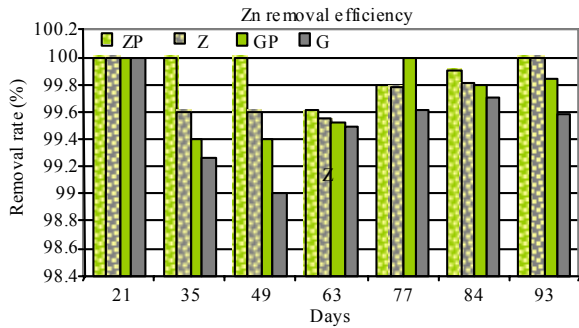
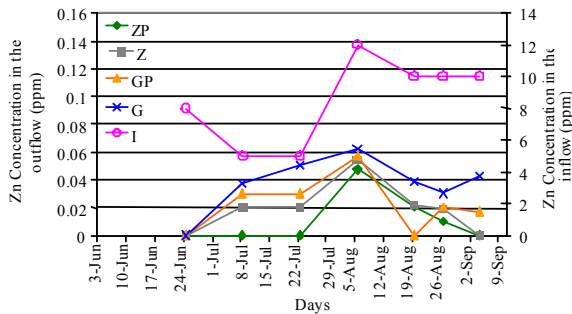Figure 6: Zn removal efficiency

Fig 7: Inflow and Outflow Concentrations of Zn during the Operation Period

It is observed that most of the pollutants are mainly removed in the first one-third of the constructed wetland length. In all four cells more than (82%) of NO$_3$-N removal occurred in the first and third parts. The concentration of NO$_3$-N in the four cells which is below 20 mg L^1 between the inlet A decrease of (0.3%) and (0.06%) in average was observed in the second. It can be seen that the NO$_3$-N

reduction has a regular function only in cell G which shows a gradual removal. Other cells did not show regular reduction along the system which may be occurred due to experimental error. Also it should be noted that the behavior of nitrate is difficult to explain because of different functions of N especially in the presence of the plant, so it is not possible to define a particular function for cell ZP and GP. More than (74%) of P removal occurred in the first one-third part of all cells. An average of (6%) and 4% reduction was observed in the other two sections. In all cells a gradual reduction is seen which shows an ordinary function for P.

In planted cells (ZP and GP) a sharper decline is observed due to plant uptake. *Zeolite* presence in cell ZP and Z is the reason for less concentration shown in Fig8 in all locations.

For Zn in all four cells more than (97%) of Zn removal occurred in the first one- third part of cells. An average reduction of (2%) and (0.5 %) was observed in the second and third parts along the cells. and location 1. In the rest part of the cells there is not a significant change in concentrationsone-third part of the cells.

DISCUSSION

Less treatment efficiencies were observed in the systems with media only (Z and G) indicating that wetland plants play an important role in pollution removal. Although the outflow concentrations from Z and G cells are higher than planted cells, significant amounts of contaminants were removed in those systems in the absence of wetland plant. This suggests that the microbial and physical/chemical processes may also result in the removal of pollutants because the media create suitable environment for microbial

growth. Biodegradation, sedimentation and sorption were the major pollutant removal mechanisms in the systems with media only. The important mechanisms for the removal of N are microbial activity (ammonification, nitrification and denitrification), plant uptake, sedimentation and ammonia volatilization[8-10]. Nitrification/denitrification and ammonification occur simultaneously in most CWs, but the extent of individual processes differs among the systems. The higher nitrate concentrations in vegetated cells are thus likely due to the root zone effect (RZE) which let more oxygen to be transferred into the soil in order to precede the activities of nitrifying bacteria and nitrification [11,12]. In other words, some vegetated beds of CWs provide high rate of nitrification, thus creating additional NO_3-N, which does not undergo denitrification due to unfavourable oxygen conditions for this process; so, nitrification produces more nitrate than what has been removed by denitrification. It is also interesting to note that the harvested mass from the ZP unit was about 1.3 times that of the GP unit, which may explain the increased nitrate amount in this unit, due to higher RZE which let more oxygen transferred into the soil to precede the activities of nitrifying bacteria and nitrification. In gravel and *zeolite* bed cells, as compared to gravel bed cells, the higher removal efficiency could be due to the cation exchange capacity of *zeolite* which affects ammonia in the system by adsorption process, in which the ammonia is exchanged by Na^+[13, 14]. In this way, *zeolite* can therefore reduce the amount of NH_4 which can affect the amount of NO_3 produced in the nitrification process. Some researches show a removal of (82%) to (99%) for NO_3-N[15], (89%) for nitrate[16] and (70.73%) for NO_3-N[17]. Constructed wetlands with subsurface flow have the major potential for phosphorus removal as adsorption and precipitation of P is effective in such systems where wastewater gets in contact with filtration substrate via these mechanisms. So, HSSF systems have higher potential for P reduction because the substrate is constantly flooded and there is not much fluctuation in redox potential in the bed [3].

CONCLUSION

The contamination of nutrients and heavy metals in water environment is a serious problem which threatens not only the aquatic ecosystems, but also human health. The present study provides evidences which prove that these treatment systems can be used effectively for decontamination of water with nutrient and Zn, Pb and Cd pollution. The emergent wetland plants (*Phragmites Australis* and *Juncus Inflexus*) have been shown to survive and perform well in treating synthetic

wastewater, while *Zeolite* and gravel materials provide a suitable plant growth medium in constructed wetlands as an alternative to conventional sand and gravel substrates. The wetland shows strong potential for the reduction of TP since TP reductions resemble efficiencies found in other similar studies. Constructed wetland cells can be expected to remove P and the removal efficiencies range from as high as (96.12%) to as low as (76.65%) while higher removal was observed in vegetated cell with a substrate of *zeolite* and gravel. The characteristics of the media type selected in this system (*zeolite*), containing higher amounts of Ca, Al and Fe oxides, was inferred to be a factor causing such high removal of P by adsorption. Therefore, it can be used effectively as a media, alone or in combination with other materials in CWs. The plant uptake was also concluded to be a factor in the P removal due to the higher removal efficiency of the planted cells. For the removal of NO_3-N, the gravel-bed wetland system without vegetation was found to be the optimal one in this study. The RZE of plants was identified to be the factor causing the increase of NO_3-N tested in the effluent of the vegetated systems. Meanwhile, the removal of NO_3-N was also more significant in unplanted *zeolite* and gravel-bed wetland systems than in the unplanted gravel-bed systems through cation exchange capacity of *zeolite,* which affected ammonia by the adsorption process. In this process, the ammonia is exchanged by Na^+ and the reduction in ammonia concentration would reduce the NO_3-N amount produced by nitrification. As for Zn removal, the planted cell with *zeolite* and gravel had the highest removal efficiency, while the system with the same substrate without vegetation had the 2nd highest removal efficiency and the other two units with gravel, with and without vegetation had lower removal efficiencies. It can be concluded from this research that adsorption in the constructed wetland was the principal process for the removal of Zn from the wastewater, while the plant tissues took up only small amounts of metals. It is also demonstrated that most of the removal occurred in the first one third part of the cells near the inlet by chemical mechanisms. The differences in the overall efficiency are therefore related to the biochemical processes operating along the length of the bed.

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