

Comparing the performance of wastewater treatment using activated sludge and aerated lagoons processes in the removal efficiency of estradiol hormones

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Abstract

Introduction: Due to the risks of hormones on human health and aquatic life, the aims of this study were to determine the efficiency of estrogen removal in the different stages of Shiraz wastewater treatment plant using activated sludge process and Marvdasht wastewater treatment plant using aerated lagoons process and also comparing the performance of both systems in removing the hormone of estrogen.

Methods and Materials: In this study, the instantaneous sampling was used to take a sample from the wastewater treatment plants. The sampling was done for 3 times to determine the removal efficiency of estrogen hormone E2. The samples were transported to the laboratory within the maximum period of 3 hours. The Enhanced Chemiluminescence (ECL) was used to measure the hormone. The device (Elecsy 2010) was made by co-production tech companies (Roche) Germany and (Hitachi) Japan.

Results: The results showed that the amount of estrogen in the influent wastewater treatment plants is approximately 40 pgmL⁻¹. The percent of the removal of the hormone in Marvdasht wastewater treatment plant using lagoon aeration process was 52.5%. The hormone removal efficiency in Shiraz wastewater treatment plant using activated sludge was more than 82%. Primary sedimentation removes only 3 to 4 percent of the estradiol hormone.

Conclusions: Although the primary wastewater treatment plant was able to remove a trace amount of estrogen, biological treatments plant has an important role in removing the hormone. The capability of activated sludge process in removing estrogen is higher than aerated lagoon.

Keywords: estradiol E2, wastewater, activated sludge, aerated lagoons, Shiraz, Marvdasht.

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Introduction

Nowadays a wide variety of chemical pollutants in huge amounts enter into water resources causing high expenses for their treatments. The evidences show that many of these pollutants are endocrine disruptor and have an adverse effect in humans, domestic and wild animals.

Many studies demonstrate the existence of the wide range of endocrine disruptor compounds (EDCs) such as estrogen which are most likely found in the aquatic organisms (1). Estrogens are basically a group of female sex hormones. A wide range of synthetic compounds have been identified that have the same estrogenic activity. Natural estrogens are named steroidal hormones, while the synthetic estrogens are called non-steroidal hormones. Estrogen is an organic compound with four cyclo-alkanes (Figure 1). Estradiol is the predominant steroid which is produced during pregnancy. Estrogenic compounds are a group of estrogenic medicine (such as retina estradiol) using as a birth control pill or contraceptive pill for hormone therapy. Most of the estrogen medicines were excreted from the body. Up to 90 percent of estrogens find their way into sewer system.

Estrogen medicines are sometimes used as estradiol patch (Estraderm). The researches have shown that the mean daily excreted estrogen via urine in a healthy woman is about 17.6 micrograms. The free estrogens were never detected in urine samples. The exerted estrogen is mostly conjugated estrogens. A laboratory biodegradation test confirmed that conjugated estrogens are readily deconjugated in raw domestic wastewater, presumably due to the large amounts of the β -glucuronidase enzyme produced by fecal bacteria (*Escherichia coli*). Deconjugation is continued in the wastewater collection network as well (2).

The concerns are increased about the direct effects of estrogen pollution on animals and

humans. Many evidences showed that fish inhabiting in polluted water that receive either raw or treated municipal wastewater treatment plants (MWWTPs) are exposed to the estrogen effects. Due to the estrogenic effect, feminization occurred in male fish and the fish reproductivity was decreased to 50 percent. Some studies have shown the extinction of carp and trout population after exposure to a synthetic estrogen (3, 4). The analysis of liver samples in fish exposed to chemicals in 113 ambient water samples in California's Central Valley and Northern area indicated that six samples (5% of total body weight of fish) have the EDCs. This amount represents the cumulative effect of the hormone in fish, while the concentration of estrogenic endocrine disrupting chemicals in ambient water samples was low (5). Taking estrogen medicine without prescription plays an important role in the etiology of breast, uterus and testis cancer. Estrogens could also result in early puberty (6).

Jafari et.al (2009) showed that the estrogen compounds in wastewater were ten times more than drinking water (7). There are several investigations that have been done in order to evaluate the efficiency of wastewater treatment plants in removing estrogenic hormones. Andersen et al. (2003) showed the efficiency of activated sludge with nitrification-denitrification in removing estrogen from MWWTP (8). Nygym et al. (2004) showed the feasibility of removing both estrogen hormone and estradiol using eight different commercial type of the nano-filtration and low pressure reverse osmosis membranes (9). In addition, Hansen (2010) showed that ozonation of the effluents from MWWTPs was successful in removing estrogens and xeno-estrogens (10).

According to the risk of hormone pollution for aquatic and human health, the main aim of this research is to (i) determine estradiol

E2 (as the strongest and the most common steroid hormone) in the MWWTP including screening, grit removal chamber with skimmer, primary sedimentation tank and biological treatment in Shiraz MWWTP using activated sludge process and Marvdasht MWWTP using aerated lagoons and (ii) to compare the performance of these two processes for the efficient removal of estrogen.

Methods and Materials

The characteristic of municipal wastewater treatment plants

1-Shiraz municipal wastewater treatment plant using activated sludge process:

Shiraz municipal wastewater treatment plant is located in the southeastern region of the city. The first module was operated for the population of 409000. The second module was designed for the total populations of 548000 which will be used in the near future. The wastewater treatment plant is using primary and secondary treatment and the order of the sampling places are as follow:

1 - Screening, 2 - Grit removal chamber with skimmer, 3- Primary sedimentation tank, 4 - Aeration tank, 5 - Secondary sedimentation tank, 6 - chlorination unit (Figure 2).

2-Marvdasht municipal wastewater treatment plant using aerated lagoon process:

Marvdasht municipal wastewater treatment plant is using primary and secondary treatment and the order of the sampling places are as follow:

1 - Screening (bar rack), 2 - Grit removal chamber with the skimmer, 3- Two aerated lagoon with the capacity of 38000 m³, 4 - Clarifier with the capacity of 12000 m³, 5 - Chlorination unit (Figure 3).

Sampling from wastewater treatment plants

In this study, the grab sampling was used to take a sample from the wastewater treatment plants. The sampling was done under standard condition in a glass container with the capacity of one liter. The samples were transported to the laboratory within the maximum period of 3 hour to analyze the hormone of estrogen E2. All the sampling was performed in three replications. Each sample only shows the characteristics of the wastewater at the time of sampling.

To determine the removal efficiency of the hormone estrogen E2 at different units' processes of Shiraz MWWTP using activated sludge process (Case Study Shiraz MWWTP No. 1) and Marvdasht MWWTP using aerated lagoon process (Case Study Marvdasht MWWTP), the samples were taken at the different stages of MWWTP as follows:

1- Influent wastewater before screening, 2- After screening, 3- After primary sedimentation tank, 4- The effluent from secondary sedimentation tank, 5- After chlorination unit

These order of sampling described above showed the role of primary wastewater treatment including screening, grit removal chamber with skimmer and primary sedimentation tank in the removal of the hormone. In addition, the estrogen removal efficiency of secondary wastewater treatment plant including activated sludge reactor, secondary sedimentation tank and chlorination unit was examined in this study. The following sampling points were selected at Marvdasht MWWTP to determine the hormone removal efficiency at different processes used in the treatment plant.

1- Influent wastewater before screening, 2- After screening, 3- After grit removal chamber with skimmer, 4- After the first aerated lagoon, 5- After the second aerated lagoon, 6- After the clarification tank, 7- After chlorination unit

Analytical method

The Enhanced Chemiluminescence (ECL) was used to measure the hormone. The device (Elecsy 2010) made by co-production tech companies (Roche) Germany and (Hitachi) Japan. The accuracy of the instrument is 5 pgr/ml (5ngr/l).

The following formula was used to determine the efficiency of each unit.

$$E = (S_0 - S_1 / S_0) \times 100$$

S_0 and S_1 are the concentration of the estrogen E2 before and after each unit process, respectively.

Results

Table 1 shows the results of sampling at Shiraz MWWTP using activated sludge. The unit of concentration is based on picogram (pictograms = 10^{-12} gram) per milliliters or nanogram per liters. According to the

results, Shiraz MWWTP decreased the estrogen concentration from 31.53 pg/ml to 5 pg/ml. The Screening increased the concentration of hormone comparing to the influent raw wastewater (approximately 9.04 pg/ml). Results also demonstrated that the primary sedimentation tank didn't have a considerable effect for the estrogen removal. The concentration of hormone in the effluent from secondary sedimentation was 5.81 pg/ml. Therefore, the role of activated sludge process was significant in removing estrogen.

Generally, the concentration of hormone was higher in Marvdasht wastewater influent (47.45 pg/ml) compared to Shiraz wastewater influent. Both treatment plants had the capability to decrease the hormone concentration to 22.53 pg/ml.

Table 1: The Mean and maximum concentration of estradiol E2 hormones at different sampling place in Shiraz municipal wastewater treatment plant using activated sludge process

Sampling place	Wastewater influent	After screening	After primary sedimentation tank	After secondary sedimentation tank	Wastewater effluent
Mean (pgmL ⁻¹)	31.53	40.54	35.09	5.81	5
Maximum (pgmL ⁻¹)	31.98	41.3	35.59	6.2	5
Standard deviation (pgmL ⁻¹)	0.193733	0.442633	0.605033	0.303333	0

Table 2: The Mean and maximum concentration of estradiol E2 hormones at different sampling place in Marvdasht municipal wastewater treatment plant using aerated lagoon process

Sampling place	Wastewater influent	After screening	After grit removal chamber with skimmer	After primary aerated lagoon	After secondary aerated lagoon	After clarification tank	After chlorination unit
Mean (pgmL ⁻¹)	47.45	30.87	35.07	18.44	21.98	18.52	22.53
Maximum (pgmL ⁻¹)	51.1	32.88	36.51	20.10	22.38	19.20	23.33
Standard deviation (pgmL ⁻¹)	3.035	1.891	1.5780	1.893	0.863	0.844	0.650

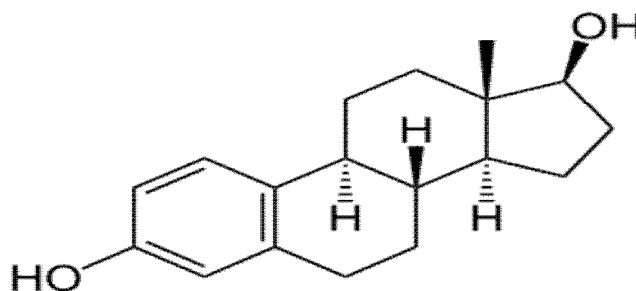


Figure 1: The structural formula for estrogen hormone

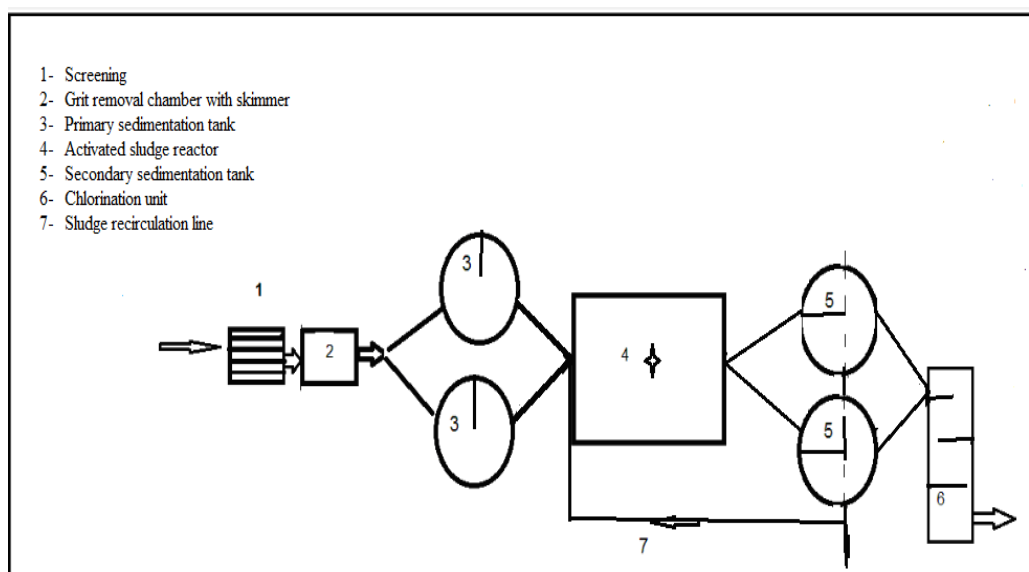


Figure 2: The flow diagram of Shiraz municipal wastewater treatment plant using activated sludge process

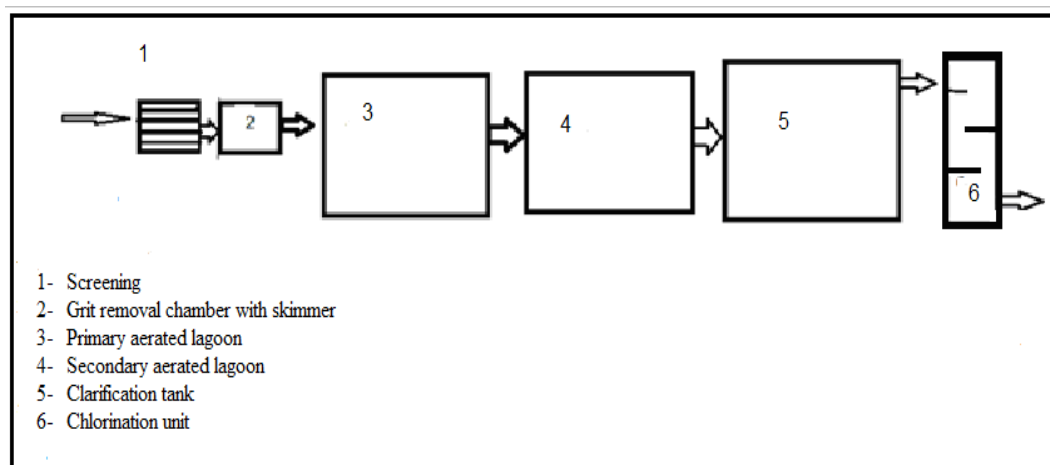


Figure 3: The flow diagram of Marvdasht municipal wastewater treatment plant using aerated lagoon process

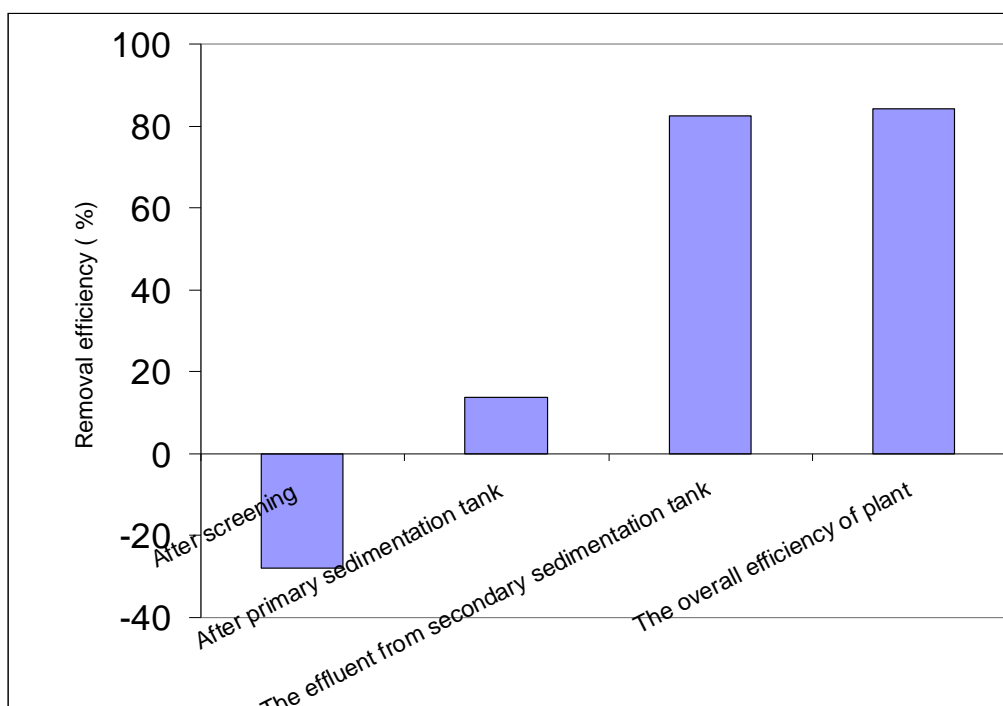


Figure 4: The removal efficiency of estradiol E2 in different sampling place of Shiraz municipal wastewater treatment plant using activated sludge process

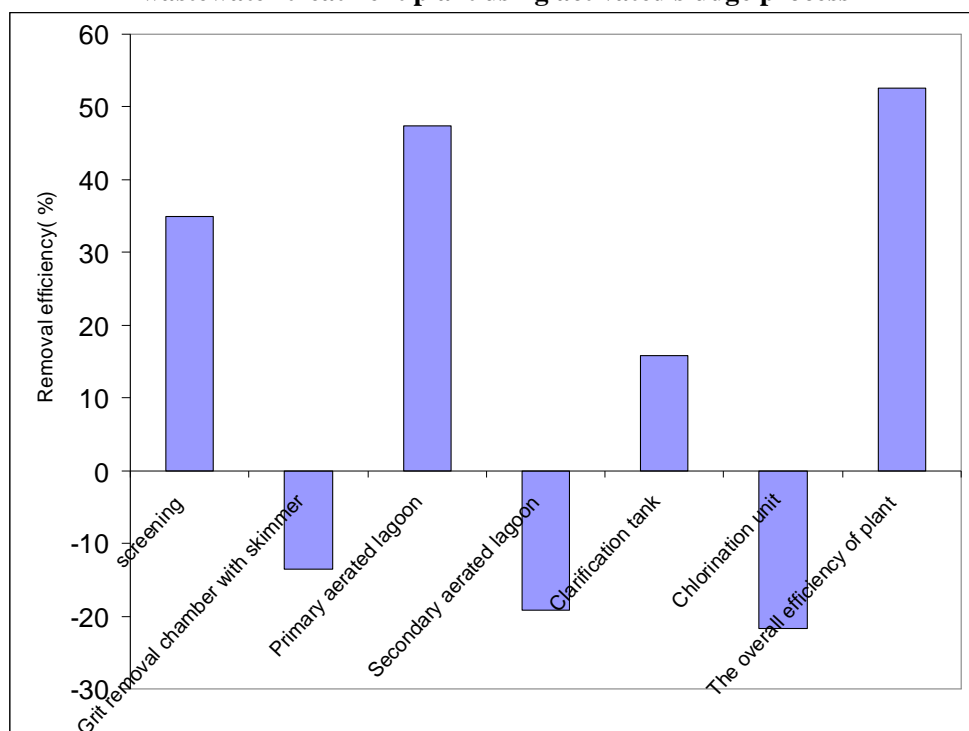


Figure 5: The removal efficiency of estradiol E2 in different sampling place of Marvdasht municipal wastewater treatment plant using aerated lagoon process

Conclusions

With regard to cycloid structure of estrogen which contains four phenolic rings connected to each other by hydrogen bonding (Figure 1), the hormone compound could not be degraded as fast as a linear organic compound such as glucose. Therefore, the special enzyme activity is required for the biodegradation of the estrogen. Due to the high molecular weight and low water solubility of the estrogen, the hormones initially attach to the bacteria mass and then the bacterial enzymes begin to decompose the hormone in the settled sludge. Since the concentration of estrogen in municipal wastewater is low, the rate of contact between the bacterial mass and hormone is low and the oxidation of hormone is slow. Therefore, high volume of bacterial enzyme is required for decomposition of the estrogenic compound. It is highly expected that trace organic material such as estrogen have higher chance of removal in activated sludge process rather than in the aerated lagoon process. Activated sludge process with high concentration of mixed liquor volatile suspended solids (MLVSS) between 2000 and 5000 mg/l and high amount of external enzymes is a suitable media to degrade estrogen in the settled sludge zone.

The efficiency of Shiraz MWWTP using activated sludge process for the removal of the hormone of estrogen E2 is at least 84%, while the removal efficiency of estrogen hormone E2 in Marvdasht MWWTP using aerated lagoon system is approximately 52.5%. Zoellick et al showed that the hormone removal efficiency of MWWTP using activated sludge was 80-93% (11). The reason for the superiority of the activated sludge process to the aerated lagoon process could be related to the difference between bacterial activities that exist in these two processes. Since activated sludge process has higher MLVSS

concentration, the rate of degradation is expected to be higher in this process. The bacterial external enzymes not only hydrolyze the organic materials but also they coagulate the suspended solid to facilitate the settling process as well. In activated sludge systems with higher biomass concentration MLVSS, the efficiency of treatment will be increased. Aerated lagoon process has used the same technology as the activated sludge process. Due to the lack of sludge recirculation in the aerated lagoon process, the concentration of MLVSS in the reactor was lower. The population of active bacteria will also be decreased because of the long retention time in the aerated lagoon. The surface mechanical aeration is used in the aerated lagoon. The essential mechanism for removing the organic material in the aerated lagoon is oxidation rather than biodegradation. The oxidation in the aerated lagoon process is more likely related to the contact of bacteria with the organic material. According to Figure 4 and 5, the variation of hormone removal efficiencies was observed at different sampling place in MWWTP. For example, an increase in the hormone concentration was observed after screening. In general, the accumulation of various compounds behind the screening bar (such as birth control pills or Estraderm) may cause an increase in the estrogen levels. Furthermore, the conversion of combined estrogen into free estrogen by the bacterial enzyme could cause an increase in the concentration of hormone in some parts of plants. Many studies demonstrated that the excreted estrogen combined with the other compounds in the wastewater and then the existing bacterial inside the sewage collection network convert the combined estrogen into the free estrogen (12-14). On the other hand, the holding capacity of wastewater and also the density of the material behind the bar screen can be effective on the performance of screening.

Conclusion

In conclusion, conventional primary wastewater treatment is not efficient in removing the estradiol hormone. Although the chemical formula of estradiol hormone is close to cholesterol, the removal efficiency of estrogen was not appropriate at the grease removal chamber and the aerated lagoon system. Due to the high molecular weight of hormone and its neutral buoyancy, the hormone of the estrogen cannot be removed in the grease removal chamber. Therefore, it was concluded that the efficiency of primary wastewater treatment in removing estrogen was negligible. Biological wastewater treatment had an essential role in the removal of estrogen from municipal wastewater. Moreover, the activated sludge process is more efficient than the aerated lagoon in removing the hormone.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

All authors have made extensive contribution into the review and finalization of this manuscript. All authors read and approved the final manuscript.

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References

- 1-ZuehlkeS, Duennbier U, Heberer T, Fritz B. Analysis of endocrine disrupting steroids: investigation of their release into the environment and their behavior during bank filtration. *Ground Water Monit R* 2004;24(2):78-85.
- 2-Rachel L, Gomes M, Mark D, Scrimshaw D, Lester NJ. Fate of conjugated natural and synthetic steroid estrogen in crude sewage and activated sludge batch studies. *Environ Sci Technol* 2009;43(10):3612–3618.
- 3-Lazorchak JMME, Smith DL, Lattier BE, Wiechman A, Braam GP, Toth RC, et al. National

screening survey of edcs in municipal wastewater treatment facilities. (United States Environmental Protection Agency, Cincinnati) 2004 EPA Pub No 600/R-04/171.

4-Kidd KA, Blanchfield PJ, Mills KH, Palace VP, Evans RE, Lazorchak JM, et al. Collapse of a fish population after exposure to a synthetic estrogen. *Proc Natl Acad Sci USA* 2007;104(21):8897–901.

5-de Vlaming V, Biales A, Riordan D, Markiewicz R, Holmes P, Otis C, et al. Screening California Surface Waters for Estrogenic Endocrine. 2006 Disrupting Chemicals (EEDC) with a Juvenile Rainbow Trout Liver Vitellogen in mRNA Procedure, SWAMP. Available From: [URL://http://www.swrcb.ca.gov/rwqcb5/water-issues/water-quality-studies/swamp-eedc-rpt-2006-4](http://www.swrcb.ca.gov/rwqcb5/water-issues/water-quality-studies/swamp-eedc-rpt-2006-4). Accessed month day, year.

6-Sharma BK, Ray A. Breast and prostate cancer. *Indian J Clin Biochem* 2000;15(Suppl 1):110-6.

7-Jafari AJ, Pourkabireh Abasabad R, Salehzadeh A. Endocrine disrupting contaminants in water resources and sewage in Hamadan city of Iran. *Iran J Environ Health Sci Eng* 2009;6(2):89-96.

8-Andersen H, Siegrist H, Halling-Sørensen B, Ternes TA. Fate of estrogens in a municipal sewage treatment plant. *Environ Sci Technol* 2003;37(18):4021-6.

9-Nghiem LD, Manis A, Soldenhoff K, Schäfer AI. Estrogenic hormone removal from wastewater using NF/RO membranes. *J Membrane Sci* 2004;242(1-2):37-45.

10-Hansen KM, Andersen HR, Ledin A. Ozonation of estrogenic chemicals in biologically treated sewage. *Water Sci Technol* 2010;62(3):649-57.

ZuehlkeS, Duennbier U, Heberer T. Determination of estrogenic steroids in surface water and wastewater by liquid chromatography-electrospray tandem mass spectrometry. *J Separation Sci* 2005;28(1):52-8.

11-ZuehlkeS, Duennbier U, Heberer T. Determination of estrogenic steroids in surface water and wastewater by liquid chromatography-electrospray tandem mass spectrometry. *J Sep Sci* 2005;28(1):52-8.

12-D'Ascenzo G, Di Corcia A, Gentili A, Mancini R, Mastropasqua R, Nazzari M, et al. Fate of natural estrogen conjugates in municipal sewage transport and treatment facilities. *Sci Total Environ* 2003;302(1-3):199-209.

13-Racz L, Goel RK. Fate and removal of estrogens in municipal wastewater. *J Environ Monit* 2010;12(1):58-70.

14-Xu N, Xu YF, Xu S, Li J, Tao HC. Removal of estrogens in municipal wastewater treatment plants: a Chinese perspective. *Environ Pollut* 2012;165:215-24.