Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2013.871338

52 (2014) 2340–2343
February

Taylor & Francis
Taylor & Francis Group

Becoming of pharmaceutical rejections in urban wastewater

Fateh Naitali, Hafida Ghoualem*

Laboratoire d'Electro-Chimie, Corrosion, Métallurgie et Chimie Minérale, Faculté de Chimie, USTHB, El-Alia Bab-Ezzouar, Alger, Algeria
Email: haghoualem@yahoo.fr

Received 3 January 2013; Accepted 22 November 2013

ABSTRACT

Many scientific studies reveal the presence of pharmaceutical compounds in aquatic environments. Pharmaceutical industry and sewage are the main sources of these compounds in the receiving environments. In this study, we investigate the fate of a simultaneous release of three drugs (antifungal, antibiotic, and antihistamine) in municipal wastewaters. To achieve this objective, a set of physical and chemical analysis is carried out. The obtained results show that releasing individual and concurrent drugs have a direct impact on the quantity and the quality of different constituent materials of waste water.

Keywords: Wastewater; Active sludge; Biological treatment; Drugs; Simultaneous releases

1. Introduction

Nowadays the wastewater treatment is becoming increasingly important worldwide, and many techniques have been developed to treat wastewater, such as physical–chemical treatment, system lagoon, activated sludge process, and anaerobic digestion [1–5]. The treatment plants use the activated sludge process to treat wastewater. However, the drugs present in the wastewater are not degraded in sewage treatment plants. They are present in the aquatic environment and they are quantified [6–9]. The occurrence, fate, and impact of drugs in aquatic environments are a recent area of concern and are taking a significant scale by scientists worldwide in the fields of environment and public health. The improvement of knowledge of the environmental impact of drugs first assumes knowledge of their fate in the wastewater. The purpose of this work is to study the fate of the selected drugs in urban wastewater through the analysis of the following physicochemical parameters: turbidity, chemical oxygen demand, electrical conductivity, NO_2^-, and SO_4^{2-} ions.

2. Materials and methods

Urban wastewater samples were collected from a purification station situated in Boumerdes (Algeria) for one to two times per week from April 2011 until April 2012. Sample collections were done at 11:00 am. The water temperature and pH were measured *in situ*. The other pollution parameters have been determined in the laboratory. The analysis of the samples concerned the physico-chemical parameters as; pH, temperature, turbidity, conductivity (σ), chemical oxygen demand (COD), NO_2^- and SO_4^{2-} ions. These

*Corresponding author.

parameters were analyzed according to the Standards Methods [10,11].

For this purpose several series of tests were performed. Method used for COD measurement was the K_2CrO_4 boiling method, and the test is performed in a temperature range of 140–150˚C. COD parameter has been analyzed by a COD-meter of mark Spectroquant TR 320 MERCK. Methods used for NO_2−N, Zambelli method and for SO_4^{2-}−S, solution of polyvinyl-pyrolidine 25% method [12]. The chemical parameters have been determined by UV–Vis spectrophotometer of JASCO V. The electrical conductivity was measured using a conductimeter of Basic 30. The turbidity is determined using a turbidimeter of Aqua Lytic. The water temperature during the tests is about 20˚C.

A pilot plant was used to treat this wastewater. The experimental device used for the biological treatment is composed of a cylindrical cone with a maximum volume of 5 L. For this purpose, all manipulations were carried out using one liter of activated sludge and two liters of wastewater. The aeration is carried out using a porous disc disposed in the bottom of a cone. The oxygen is provided by an air pump. The mixture of water and drugs was stirred for 5 min.

The experiments were conducted with three types of drugs, an antibiotic, an antifungal, and an antihistamine, following an experience factorial plan of (2^3). The equation is expressed as following:

$$Y = A0 + A1\,X\,1 + A2\,X2 + A3\,X3 + A12\,X1\,X2 + A13\,X1\,X3 + A23\,X2\,X3 + A123\,X1\,X2\,X3 \quad (1)$$

where Ai: represents the model coefficients and Xi the model factors (drug concentration); A0: average effect of drugs; A1: effect of drug 1; A2: effect of drug 2; A3: effect of drug 3; A12: effect of the interaction between drug 1 and 2; A13: effect of the interaction between drug 1 and 3; A23: effect of the interaction between drug 2 and 3; A123: effect of the interaction between drug1, 2, and 3; X1: Drug concentration 1; X2: Drug concentration 2; X3: Drug concentration 3.

The medicines and their field of study are shown in Table 1.

Experiments were performed according to the following experience matrix (Table 2):

Table 1
Studied factors and their study field

	Level (low) (mg/L)	Level (high) (mg/L)
Antibiotic (*Doxycycline*)	1	50
Antifungal (*Ketoconazole*)	2	20
Antihistamine (*Loratadine*)	1	5

Table 2
Experience matrix

No.	Antibiotic (*Doxycycline*)	Antifungal (*Ketoconazole*)	Antihistamine (*Loratadine*)
1	−1	−1	−1
2	+1	−1	−1
3	−1	+1	−1
4	+1	+1	−1
5	−1	−1	+1
6	+1	−1	+1
7	−1	+1	+1
8	+1	+1	+1

3. Results and discussion

The obtained results of the pollution parameters in the wastewater are represented in Table 3. As shown, the pollution parameters widely exceed the standards discharges [13,14]. In the presence of drugs, the pollution parameters concentration varied. In this study, the effect of each drug and drug interactions on the pollution parameters was examined. These parameters variations were analyzed by the method of the design of experiments. The obtained results are shown in Table 4.

The obtained results show that the studied drugs affect the pollution parameters in wastewater. These drugs interact with the various compounds in water that influence the biological treatment process of the effluent. Because of the drugs interaction between themselves, the drug stability is variable in wastewater. The drug combinations, whether of the same family or different families, can have different modes of action on certain parameters and similar to others in the wastewater [15].

We notice that the antibiotic reduces SO_4^{2-}, NO_2^- ions, and σ. However, the antifungal decreases only the σ parameter. The antihistamine reduces turbidity, NO_2^- ions, and σ. The interaction between the antibiotic and the antifungal reduced NO_2^- and σ. The interaction with the antibiotic and the antihistamine reduces turbidity, NO_2^-, and σ. The interaction between the antifungal and the antihistamine reduced turbidity, SO_4^{2-}, and COD. The addition of drugs has increased the organic substances and therefore COD increases. Variations of the SO_4^{2-} and NO_3^- are explained by the disruption of nitrification and sulfonation.

Table 3
Variation of the pollution parameters in wastewater

No.	Conductivity (μS/cm)	Turbidity (NTU)	COD (mg/L)	SO_4^{2-} (mg/L)	NO_2^- (mg/L)
1	1,266	130	341	287.743	0.1102
2	1,426	98	141	124.84	0.3908
3	1,321	84	400	209.629	0.2652
4	1,040	327	877	222.644	0.2256
5	1,252	91	274	259.557	0.2738
6	1,098	203	1,132	279.575	0
7	1,222	120	425	284.625	0.2954
8	1,127	173	1,081	378.537	0

Table 4
Results of the design of experiments

	Conductivity (μS/cm)	Turbidity (NTU)	COD (mg/L)	SO_4^{2-} (mg/L)	NO_2^- (mg/L)
A0	1219.00	153.25	583.88	255.89	0.20
A1	−46.25	47.00	223.88	−4.49	−0.04
A2	−41.50	22.75	111.88	17.97	0.00
A3	−44.25	−6.50	144.13	44.68	−0.05
A12	−47.75	27.00	59.38	31.23	−0.04
A13	−16.00	−5.75	154.63	32.98	−0.10
A23	41.25	−23.00	−86.88	13.04	0.00
A123	62.50	−41.75	−109.88	−12.75	0.04

Reactors were used batch for the purification of effluent that contains synthetic substrates and a mixture of disparity made up of 10 xenobiotic compound concentrations in the range of mg/L as well as heavy metals [16]. They observed a significant consumption of the ammonium inhibition.

The conductivity decrease is explained by the tendency of drug molecules to form complexes with the mineral matter present in raw water, this is a phenomenon of adsorption and/or electrostatic interaction causing a concentrations decrease in the liquid phase. The increase of the conductivity translated a toxic effect of the doses used on existing microbial pollution in the water used and includes biological aggregates and multivalent cations [17].

4. Conclusion

In this study, we successfully tested a biological treatment of wastewater from a purification station. The obtained results show that the studied drugs have a positive or a negative impact on the pollution parameters. The results explain the persistence of these drugs in wastewater, and their interaction with different constituent materials of these waters that influence the biological wastewater treatment process. For the used drugs, different trends in reduction of the turbidity, the NO_2^-, and SO_4^{2-} ions and the σ were observed. The interactions between the different drugs and the parameters reduction by these medicines are variable. Mixtures of drugs, either from the same therapeutic family or other families, can have different action modes on certain parameters and similar on others in the sewage system.

References

[1] D. Bolzenallo, F. Fatone, S. Fabio, Mesopholic, thrermophilic and temperature phased anaerobic digestion of waste active, Chem. Eng. Trans. 17 (2009) 385–390.

[2] H. Ghoualem, Assessment of a wastewater and treatment of an urban effluent, Chem. Eng. Trans. 17 (2009) 391–396.

[3] L. Loperana, M.D. Ferrari, V. Saravia, D. Muro, C. Lima, L. Ferrando, A. Fernandez, C. Lareo, Performance of commercial inoculums for the aerobic biodegradation of a high fat content dairy wastewater, Biores. Technol. 98 (2007) 1045–1051.

[4] H. Ghoualem, F. Tedjani, A. Khouider, Assessing the feasibility organic matter and nutrients removal from wastewater food industry, Proceedings of the 12th International Conference on Environmental Science and Technology, B-346–350, Greece, 2011.

[5] Metcalf and Eddy, Wastewater Engineering: Treatment, Disposal, Reuse, 4th ed., Tata McGraw-Hill, New Delhi, 2003.

[6] D. Calamari, E. Zuccato, S. Castiglioni, R. Baganti, R. Fanelli, Strategic survey of therapeutic drugs in the rivers Po and Lambro in Northern Italy, Environ. Sci. Technol. 37 (2003) 1241–1248.

[7] S. Managaki, A. Murata, H. Takada, B.C. Tuyen, N.H. Chiem, Distribution of macrolides, sulfonamides and trimethoprim in tropical waters: Ubiquitous occurrence of veterinary antibiotics in the Mekong delta, Environ. Sci. Technol. 41 (2007) 8004–8010.

[8] J. Siemens, G. Huschek, C. Siebe, M. Kaupenjohann, Concentrations and mobility of human pahrmaceuticals in the world's largest wastewater irrigation system, Mexico City, Mezquital Valley, Water Res. 42 (2008) 2124–2134.

[9] H. Ghoualem, F. Naitali, Study of mechanical behavior of active sludge after treatment of pharmaceutical effluent, Global Conference on Global Warming, Turkey, 2012.

[10] French Association for Standardization. Collection of French Standards. Water testing methods. Association Française de Normalisation (AFNOR), Recueil des normes françaises. Eaux, méthodes d'éssais, 1979.

[11] French Association for Standardization. The water quality, Volume 1: Sampling and quality control, Association Française de Normalisation (AFNOR), La qualité de l'eau. Tome 1: echantillonnage et contrôle qualité, Paris, 1979.

[12] J. Rodier, L'analyse de l'eau eaux naturelles, eaux résiduaires, eau de mer (The analysis of water, natural waters, sewage water, sea water), 8ème ed., Dunod, Paris, 1996.
[13] J.C. Boeglin, Traitements biologiques des eaux résiduaires (Biological treatment of wastewater), Edition technique de l'ingénieur. J 3 942, 1998.
[14] M.N. Pons, Analyse du cycle de vie. Épuration des eaux usées urbaines (Analysis of the life cycle. Urban wastewater), Edition technique de l'ingénieur, 1998.
[15] P. Verlicchi, M. Al Aukidy, E. Zambello, Occurrence of pharmaceuticals compounds in urban wastewater: Removal mass load and environment risk after a secondary treatment – A review, Sci. Total Environ. 429 (2012) 123–155.
[16] D. Dionisi, C. Levantesi, M. Majone, L. Bornoroni, M. De Senctis, Effect of micropolluants (organic xenobiotics and heavy metals on the activated sludge process, Ind. Eng. Chem. Res. 46 (2007) 6762–6769.
[17] J. Wingender, T.R. Neu, H.C. Flemming (Eds.), Microbial Extracellular Polymeric Substances, Springer, Heidelberg, 1999.