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RESEARCHARTICLE

IMPACT OF RESIDUES OF DRUGS (PARACETAMOL, CODEINE, AMBROXOL) IN THE TREATMENT OF WASTEWATER ON THE METAZOAN SCRUBBERS

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ABSTRACT

In the activated sludge wastewater treatment process of many metazoan scrubbers are associated to organic colloids and organ metallic present in wastewater. Some agencies may use the carbon and the nitrogen component drugs to feed or use it as a source of energy. The study of the biological degradation of micro pollutants (secondary pollution), including the pharmaceutical compound reveals a great importance for the process of degradation and environmental. In this work we are interested in the study of the effect of three drugs (NSAIDs, hormone and a corticosteroid) rejected at the same time, in wastewater on the most abundant metatozoans according to the method of the design of experiments (2^3 factorial) and study some parameters directly linked pollution has this kind of micro-organisms. The results obtained show that: studied medicines interfere with the process of nitrification and they have an adverse effect, in certain doses, on the metazoan present.

INTRODUCTION

Out of the body, pharmaceutical substances can also undergo structural transformation by biotic or abiotic processes. Biotic transformations are performed by the bacterial and fungal cultures present in the environment and in the sewage waste water (Groning *et al*., 2007). Biological treatment of wastewater for the elimination of the organic matter and nutrients (nitrogen, phosphorus) by processes which include, intensively, the principle of assimilative (Abolghasem Alighardashi, 2007). Two main families of microorganisms exist in the sludge: the protozoan and the metazoan. They play a very important role in the STEP and contribute to the balance of the ecosystem. The main roles of the metazoan are: Reduce turbidity by predation of free bacteria and feeding of certain pathogenic bacteria. The most frequently detected pharmaceutical compounds in the sewage are anti-inflammatory, antibiotics, anti-epileptics, anticancer, contrast media and regulators of lipid (Ternes and Joss, 2006). These compounds may have effects on the biomass of the sewage as they are, by definition, intended to have an impact on the organism. Microscopic observation of sludge is a technique that is, so far, not enough used in routine sewage treatment operators and local specialists who advise (Duchene and Coutteux, 1993). This justifies our interest to study the impact of simultaneous of three drugs (Paracetamol, Codeine, Ambroxol) on metazoans in biological sewage process according to the method of design of experiments (factorial 2^3) that while analyzing the following pollution parameters in this work: TSS and index of mud (SVI), BOD_5, COD, report of biodegradability, NO_2^- and interfacial surface before and after treatment.

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MATERIALS AND METHODS

Wastewater samples were collected from a station of purification situated in Boumerdes (located 45 km east of Algiers). A biological treatment was used to treat this wastewater. The experimental device used for the biological treatment is composed of a cylinder-conical reactor whose maximum volume is of 5 liters. For this purpose all manipulations were carried out using one liter of activated sludge and two liters of wastewater. Oxygen is provided by a compressor. The treatment was performed over a period of 5 hours. The physical and chemical parameters were analyzed every hour.COD, BOD_5 and NO_2pollution parameters were analyzed by COD meter, BOD_5 meter and visible

spectrophotometer (Rodier, 1996). The biodegradability ratio (k) was calculated using the following relationship:

$$K = \frac{DCO}{DBO}$$

The surface tension (γ) was measured by the stalagmometry method. The TSS sludge is analyzed by drying at 105 ° C for 24 h. Mud (SVI) index (Forster C. F. 1996) is the volume occupied by one gram of mud after 30 minutes of settling in a test tube of oneliter. Mud index is obtained by calculating the ratio of the volume of sludge decanted and the TSS concentration of samples introduced into the tube. Microscopic results have been obtained using an optical microscope. The analysis is based on the following microorganisms 'metazoan': Digonontasand filaments. The water temperature during the tests is in the vicinity of 20±2 ° C. The experiments were conducted with three drugs .The drugs are the ambroxol, the codeine and the paracetamol.The experiments were performed according to factorial experimental design (2^3).
The equation is expressed as following:

Y = A0 + A1 X 1 + A2 X 2 + A3 X 3 + A12 X 1 X 2 + A13 1 X 3 + A23 X 2 X 3 + A123 X 1 X 2 X 3.

Where Ai, are the coefficients of the model and Xi, are the factors of the model (drug concentration).
Experiments were performed according to the following matrix of experience (Table 1)

Table 1. Experience matrix

	Codeine (A) (mg/L)	Paracetamol (B) (mg/L)	Ambroxol (C) (mg/L)
1	1	10	10
2	40	10	10
3	1	100	10
4	40	100	10
5	1	10	100
6	40	10	100
7	1	100	100
8	40	100	100

Studied drug and their field of study are reported in Table 2

RESULTS AND DISCUSSION

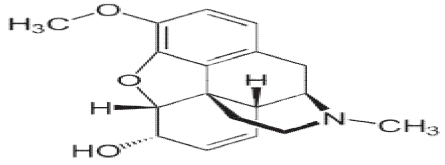
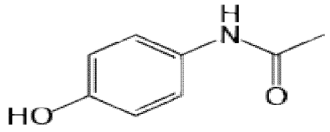
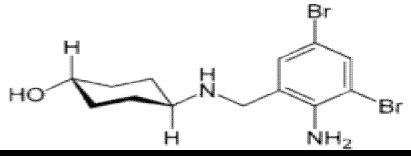
The results of the parameters of the pollution obtained in the wastewater before (a) and after (b) addition of drugs are shown in figures (1 and 2). We notice that after the addition of drugs the concentration, COD decreases for all tests and BOD_5 decreasing so except for the tests 6 and 7, the report of biodegradability increases only for the first two tests, interfacial tension increases except for testing 5 and 6, the nitrites increase only for test 4 and 6.The report of biodegradability increases for the first two tests and decreases for the others.

Before treatment: k < 2 for tests 1 and 5 (biodegradable effluent), 2 < k < 5 for other tests (not readily biodegradable effluent). After purification k < 2 for tests 1, 3, 4, 5, 6, 7 and 8 (effluent biodegradable), k > 5 test 2 (non-biodegradable effluent).

The interfacial tension values for all the tests is below the value of pure water (72 mN/m), none exceeds 60,11mN/m after the rejection of the drug. This is the fact that drug use act as detergents, or surfactants and damage the bacterial cytoplasm membrane (Meyer, 2004).The nitrite concentration increases only for the4 and 6 tests. It does not exceed the barrier of 0.7 ppm. This explains the harmful effect of drugs studied in nitrifying microorganisms. (Huang *et al*., 1995) have noted that in the presence of an assault, localized bacteria in the interface between the liquid phase and the aggregate firstlylose their activities.

The TSS concentration decreases for the1st and 3rd and increases for other tests. The content does not exceed 25 g/L.SVI increases only for the 3rd test. It does not exceed the value of 110 mL/g (easily sediment mud). The increase of TSS and the decrease of SVI showed that organic matter increases in the sludge which is in agreement with the results of (Sponza, 2002) 'Sponza observes the SVI index improves with the increase in protein content, which is an organic'. Thus, levels of protein and DNA of a flock are respectively positive and negative on the decrease of SVI (Bura, 1998) and (Sponza, 2003).

Table 2. Studied factors and their field of study

Drugs	Chemical formula		Lowlevel(mg/L)	High level(mg/L)
Codéine (A)		Opioidanalgesic	1	40
Paracetamol (B)		NSAIDS	10	100
Ambroxol (C)		Corticosteroid	10	100

Results of design of experiments

To quantify the effect of each drug and the drug interactions on the parameters of pollution examined in this study, changes

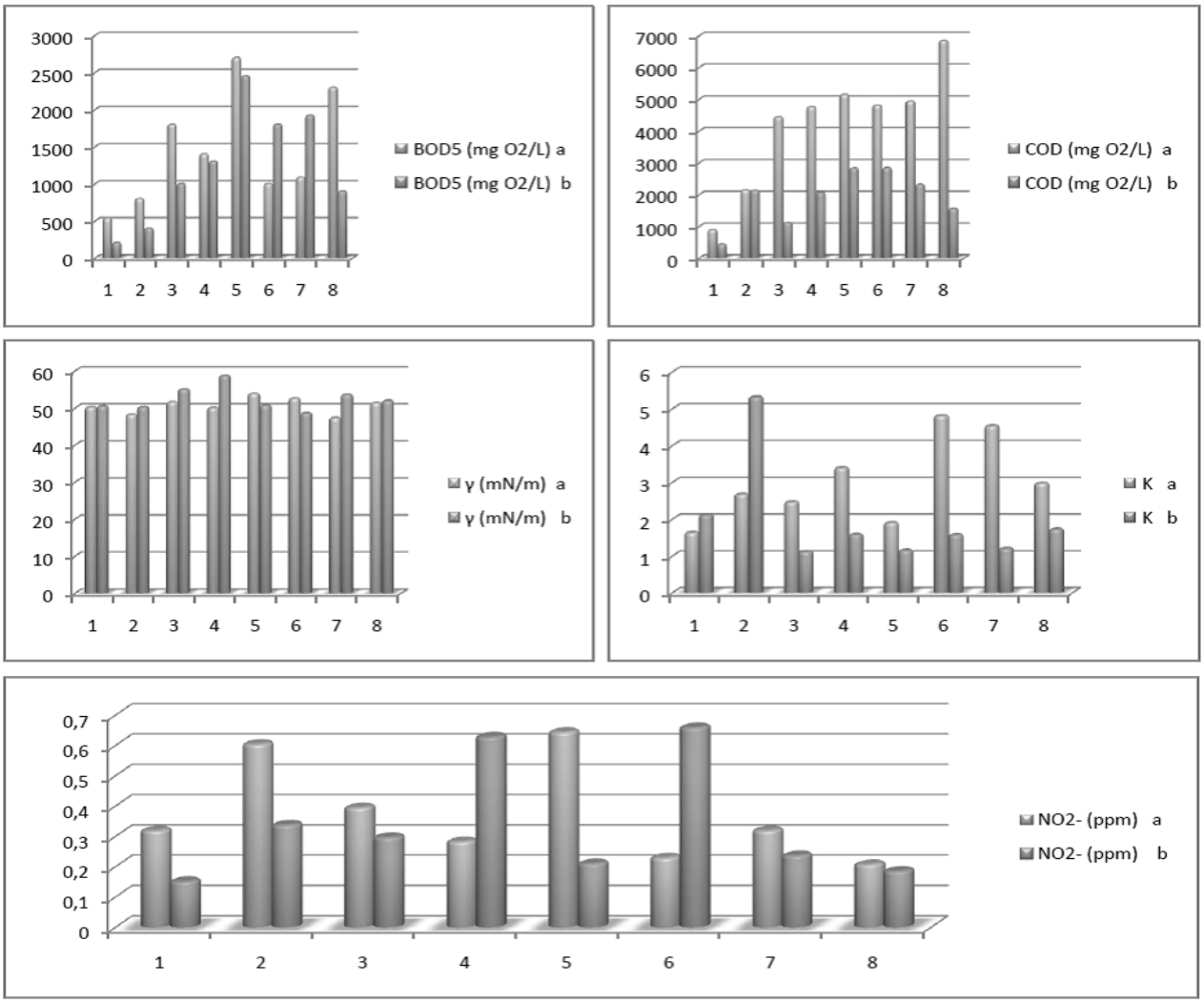


Fig.1. Parameters of pollution (a) before and after (b) treatment in wastewater

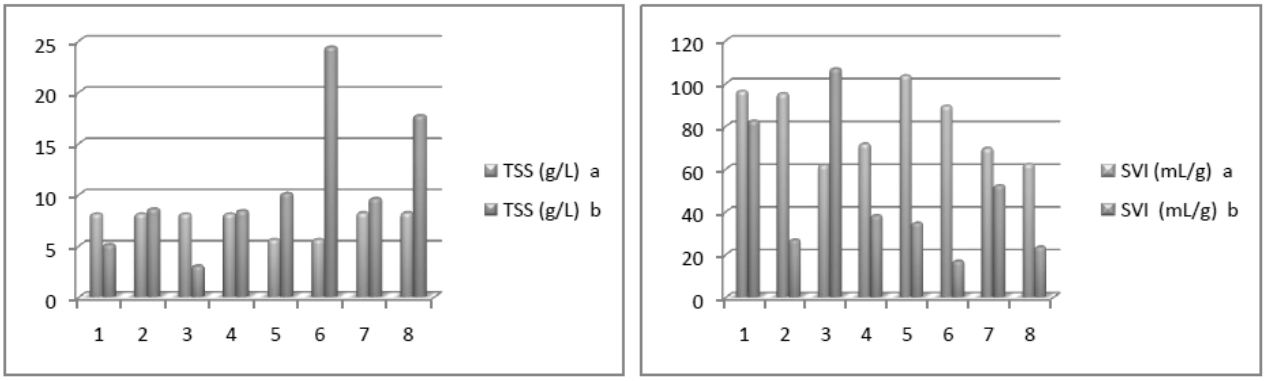


Fig2: TSS and SVI before (a) and after (b) purification in thesludge

Table 3. Results of design of experiments

	γ (%)	DCO (%)	DBO_5 (%)	K (%)	NO_2^- (%)
A0	3,67	-49,88	-9,59	-25,32	10,50
A1	0,06	5,9725	0,08	9,34	53,14
A2	5,87	-15,62	0,59	-30,57	4,67
A3	-3,44	-4,26	31,17	-30,23	10,35
A12	-0,39	-7,46	-25,09	-1,13	-12,60
A13	-3,49	-10,98	-12,10	-8,41	14,29
A23	1,06	4,53	-14,38	28,39	-43,77
A123	-2,15	0,38	-31,55	15,91	-47,23

in these parameters were analyzed by the method of design of experiments. Results obtained are reported in Table 4.

We notice that the studied drugs affect the parameters of pollution in waste water such as the COD, the BOD_5, the surface tension, the k, and the NO_2^-.The analgesic increases all parameters studied.

The NSAID decreases the parameters COD and k. It increases the parameter γ, BOD_5, and NO_2^- The corticosteroid reduces the parameters COD, γ, and k however it increases the BOD_5 and NO_2^- parameters. The interaction between the analgesic and NSAID decreases all parameters. The interaction between the analgesic and the corticosteroid reduces all parameters except nitrites. The interaction between the NSAIDs and the corticosteroid reduces the BOD_5 and NO_2^- parameters. It increases the parameters COD, γ and k.

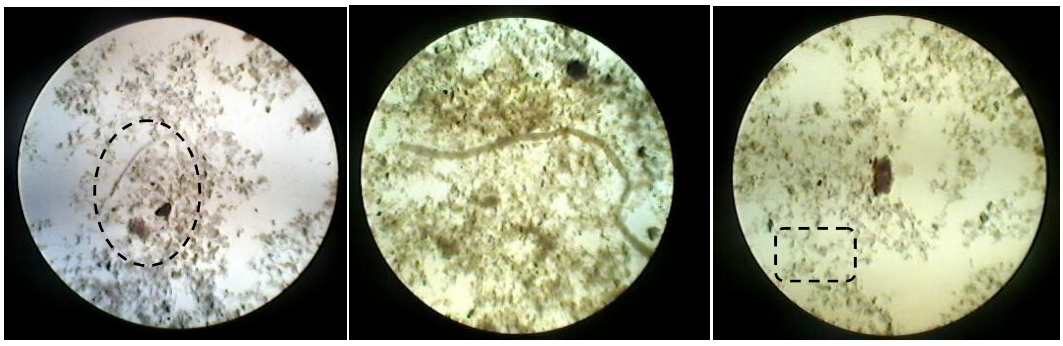
The interaction between the three drugs has the opposite effect of the NSAIDs. The analgesic and the corticosteroid, the interaction between the analgesic and NSAID, the analgesic and the corticosteroid, the NSAIDs and the corticosteroid have the lowest effect on γ. The NSAIDs has the lowest impact on BOD_5.The interaction between the three drugs has the lowest impact on COD.

Results of microscopic analysis

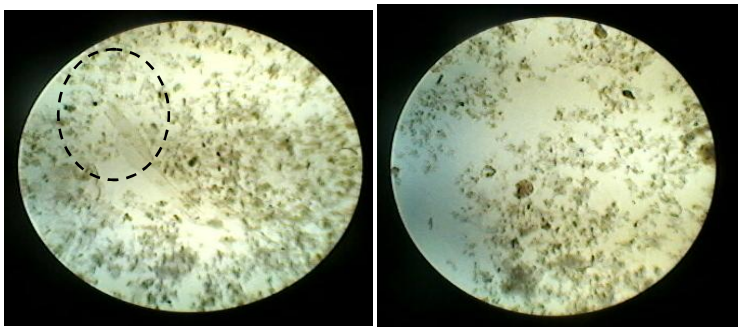
Microscopic tests were conducted on the activated sludge before and after the treatment of wastewater. The bacterial filaments increase in number for all experiments. Pebbles of trickling are noticed. A viscous layer of bacteria and metazoan covers the surface of these stones. The microorganisms studied are observed related at mud and in the water between the muds that is due to the phenomenon of Chemo taxis (Prescott *et al*., 2003). Microscopic observation results are reported in Table 4

Table 4. Results of microscopic observations

	1	2	3	4	5	6	7	8
Filaments	Increases in number	Increases in number	Increases in size and number	Increases in number	Increased number and decreased size	Increased number and decrease in size	Increases in number and in size	Increased number and decrease in size
Digononta	No change	Increased number and decreased size	Decreases in size	Decreases in size	Manyspeciesstill (dead).	Absence	Spherical in shape (a mixture of small and large size).	Spherical in shape (a mixture of small and large size).



Digononta (n°7)Filaments (n°3) Digononta (n°8)



Digononta (n°1) Filaments (n°2)

Figure4: Microscopic aspects of the metazoan studied.

Microscopic observation results are covtable:

We notice that the spherical shape of Digononta is due to the increase in osmotic pressure (Figure 4). The results of the test indicate that drugs do not affect on the same type of metazoans in the same way. The change of form of Digonontais explained by: this species have the power to transform into oval or spherical unit equipped with extraordinarily high resistance when the middle runs nutrient or external physicochemical conditions change. The death of microorganisms is due to the presence of corticosteroid that promoting the phenomenon of antisepsis, the NSAIDs and analgesic that amending the osmotic pressure by changing the surface tension. Despite the addition of drugs, some species undergo no change for some tests, this can be explained as follows: certain organic materials can, for example, protect bacteria against an agent because wastewater containing the agent can easily reach the bacteria; either the agent is absorbed by the material surrounding it (Meyer, 2004) and (Higarashi, 2002) considers that the degradation of 7 to 8 mg of organic material by microorganisms is enough to consume the oxygen content in one liter of water.

Conclusions

During this study, we have observed that the three drugs alter the physicochemical properties of wastewater and have an influence on metazoans. Treatment of wastewater containing pharmaceutical residues can lead effluents containing of byproduct more dangerous than the initial product. The inhibition of nitrification is caused by the degradation products which themselves affect the physicochemical parameters such as COD, BOD_5, surface tension, TSS and SVI mud. The supply of organic matter (drugs) rich in carbon seems to play a dual role: it can act as an accelerator, but also as a retardant of biological activity. Results of the BOD_5 and COD variation is due to the biological activity of drugs used and has the existence of sorption phenomenon (studies on pharmaceutical substances themselves are essentially confined to studies phase dissolved so that very little data of presence in the solid phases are identified. The use of the method of design of experiments showed that the effect of drugs refers according to its concentration when other drugs coexist in the middle, hence, we must not neglect interactions between drugs in aquatic environments (Naitali, 2012) and (Ghoualem, 2012).

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