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# BIOLOGICAL TREATMENT OF A WASTEWATER CONTAINING PHARMACEUTICAL CONTAMINANTS

**Lotfy, H. R. and Eiseb, N. B.**

*Department of Chemistry and Biochemistry, Science Faculty,*
*University of Namibia, Windhoek, Namibia.*

## ABSTRACT

*In this article the biodegradation studies of some of the frequently used pharmaceutical drugs, aspirin, ibuprofen, triphasil and amoxicillin are presented. It is found that aspirin is readily biodegradable at all tested concentrations. Ibuprofen is found biodegradable up to 35 mg/l, and at a concentration of 40 mg/l, it showed some toxicity to microorganisms. The existence of ibuprofen as a pure substrate caused a temporary inhibition to the bacterial activities for a period of 5 days after which the bacterial activities increased. In case of effluents containing aspirin, triphasil and amoxicillin biodegradation seems to be a reasonable way of treatment.*

***Keywords :*** *Biodegradability, lag time, toxicity, plateau.*

## INTRODUCTION

The continued growth in human population has created a corresponding increase in the demand for the Earth's limited supply of freshwater. Thus, protecting the integrity of our water resources is one of the most essential environmental issues of the 21st century. Recent decades have brought increasing concerns for potential adverse human and ecological health effects resulting from the production, use, and disposal of numerous chemicals that offer improvements in industry, agriculture, medical treatment, and even common household conveniences.

Many pharmaceutically active compounds, residues and their metabolites could resist wastewater treatment plant discharges and they could be transported into the surface waters. The widespread of antibiotics has led to the development of antibiotic-resistant bacteria, capable of penetrating and surviving different treatment processes. The occurrence of pharmaceutically active compounds in wastewater has been reported in several areas of the world (Jawad et al, 2007, Frenandez et al, 2010). The biological treatment of wastewater contaminated with some pharmaceutically active compounds has also been reported ( Adriano et al, 2005, Jesus et al, 2005 and Jermy et al, 2008).

Household chemicals, pharmaceuticals, and other consumables as well as biogenic hormones are released directly to the environment after passing through wastewater treatment processes (via wastewater treatment plants, or domestic septic systems), which often are not designed to remove them from the effluent. Veterinary pharmaceuticals used in animal feeding operations may be released to the environment with animal wastes through overflow or leakage from storage structures or

land application. As a result, there are a wide variety of transport pathways for many different chemicals to enter and persist in environmental waters.

Little is known about the extent of environmental occurrence, transport, and ultimate fate of many synthetic organic chemicals after their intended use, particularly hormonally active chemicals, personal care products, and pharmaceuticals that are designed to stimulate a physiological response in humans, plants, and animals. These organic waste chemicals are potentially associated with human, industrial, and agricultural wastewaters and include antibiotics, other prescription drugs, nonprescription drugs, steroids, reproductive hormones, personal care products, products of oil use and combustion, and other extensively used chemicals.

The fate of chemicals and their partitioning over the environments should include evaluation of their biodegradability determination as pure chemicals in a synthetic wastewater.

Microbial degradation of wastewater containing organic waste chemicals has been documented by several authors (Rozich et al, 1985, Kumaran, 1986, Lallai and Mura, 1989, Rashed and Lotfy, 1993, Lotfy et al, 1997, Lotfy and Rashed, 2002, Goivanjn et. al, 2003 ).

Aspirin ($C_9H_8O_4$), is a salicylate drug, often used as an analgesic to relieve minor aches and pains, as an antipyretic to reduce fever, and as an anti-inflammatory medication. Aspirin is also used in prevention of heart attacks and strokes. It is used in veterinary uses in treating animals to relief pain. Today, aspirin is one of the most widely used medications in the world, with an estimated 40,000 tons of it being consumed each year and that's why it is very important to study its influence on wastewater and the environment.

Ibuprofen ($C_{13}H_{18}O_2$), is a non-steroidal anti-inflammatory drug used for relief of symptoms of arthritis, primary dysmenorrhea, fever, and as an analgesic, especially where there is an inflammatory component. Ibuprofen is sometimes used for the treatment of acne, because of its anti-inflammatory properties. The drug may also be used to treat low blood pressure when standing up. Ibuprofen works by blocking the production of substances in the body that cause pain and inflammation.

Triphasil ($C_{20}H_{24}O_2$), ethinyl estradiol and levonorgestrel (generic name) contains a combination of female hormones that prevent ovulation. This medication also causes changes in the cervical mucus and uterine lining, making it harder for sperm to reach the uterus and harder for a fertilized egg to attach to the uterus.

Amoxicillin ($C_{16}H_{19}N_3O_5S$) is used to treat bacterial infections caused by susceptible microorganisms. It acts by inhibiting the synthesis of bacterial cell wall. Amoxicillin is regarded as having a fairly broad spectrum against many bacteria thus it is used both on organisms known to be sensitive to it plus it is a good selection when the sensitivity of bacteria is unknown. It is especially helpful in anaerobic infections. Amoxicillin is used in infected bite wounds, upper respiratory infections, infected teeth and bladder infections.

In this article we are presenting the biodegradation studies of some of the frequently used pharmaceutical drugs, aspirin, ibuprofen, triphasil and amoxicillin.

Synthetic wastewater composition

**Experimental**

Reagents and chemicals

Aspirin, Ibuprofen, Triphasil and Amoxicillin are commercially available, solvents and other chemicals are all reagent-grade.

**Table 1.** Composition of the Synthetic Wastewater (Patting et al, 1980).

| Constituent | Stock concentration g/l | Quantity used ml/5 liter | Final concentration mg/l |
|---|---|---|---|
| Bacto-Peptone | 32.25 | 27.3 | 352.73 |
| $MgSO_4.7H_2O$ | 10.00 | 25.0 | 50.00 |
| $MnSO_4. H_2O$ | 1.00 | 25.0 | 5.00 |
| $FeCl_3. 6H_2O$ | 0.175 | 25.0 | 1.00 |
| $CaCl_2$ | 100.00 | 25.0 | 3.75 |
| $KH_2PO_4$ | 52.25 | 33.4 | 349.4 |
| $K_2HPO_4$ | 107.00 | 33.4 | 715.56 |

In our experiments using unadapted inoculums from domestic sewage (as a seed) the degradation of aspirin, ibuprofen, triphasil and amoxicillin at different concentrations in synthetic wastewater, incubated for 15-31days under aerobic condition was studied. The BOD of the seed (municipal wastewater) alone was measured, under the same experimental conditions in each run, for seed corrected data calculations. Experiments were done at 25$^{o}$C with magnetic stirring and in the dark. The synthetic wastewater used was not inoculated.

The aspirin run was carried out by dissolving aspirin in the synthetic wastewater in the following concentrations 5 mg/l, 10 mg/l, 15 mg/l, 20 mg/l, 25 mg/l, 30 mg/l, 35 mg/l and 40 mg/l respectively in addition to the blank. 4

The ibuprofen run was carried out by dissolving ibuprofen in the synthetic wastewater in the following concentrations 5mg/l, 10 mg/l, 15 mg/l, 20 mg/l, 25 mg/l, 30 mg/l, 35 mg/land 40 mg/l respectively in addition to the blank. The triphasil run was carried out

by dissolving triphasil in the synthetic wastewater in the following concentrations 1 mg/l, 2 mg/l, 3 mg/l, 4 mg/l, 6 mg/l, 8 mg/l, 10 mg/land 12 mg/l respectively in addition to the blank. The amoxicillin run was carried out by dissolving amoxicillin in the synthetic wastewater in the following concentrations 5mg/l, 10 mg/l, 15 mg/l, 20 mg/l, 25 mg/l, 30 mg/l, 35 mg/land 40 mg/l respectively in addition to the blank.

Seed corrected data is obtained by subtracting the blank values from all other corresponding values for different concentrations.

## RESULTS

BOD readings for different experiments are tabulated and presented graphically as shown below. The results of biodegradation of aspirin are shown in Fig. 1-3.

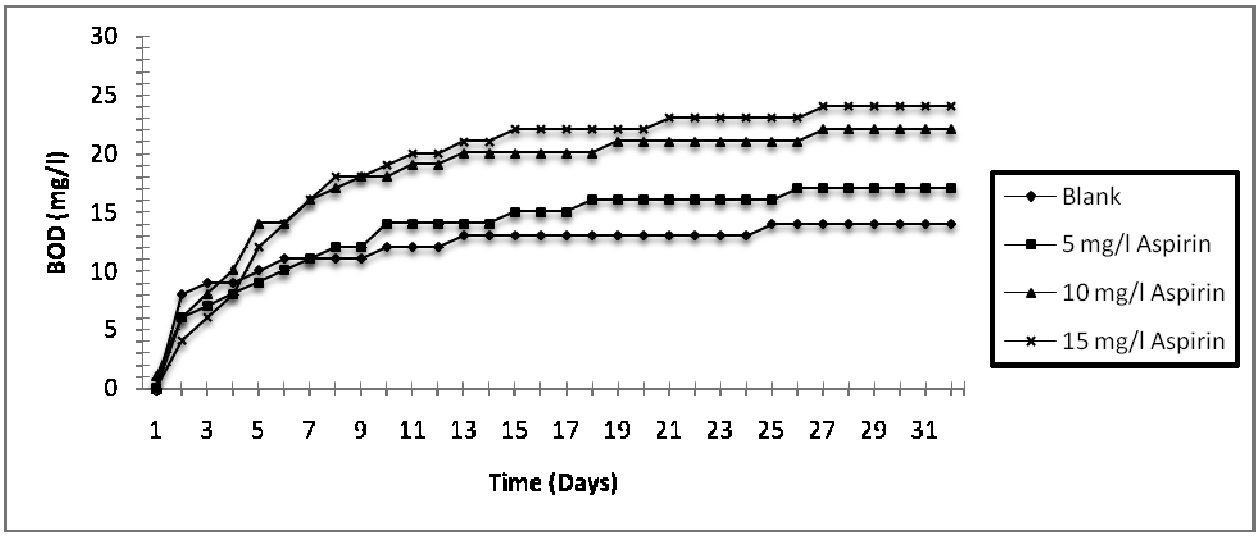


**Figure 1.** Biodegradation of a wastewater containing 5, 10 and 15 mg/l aspirin.

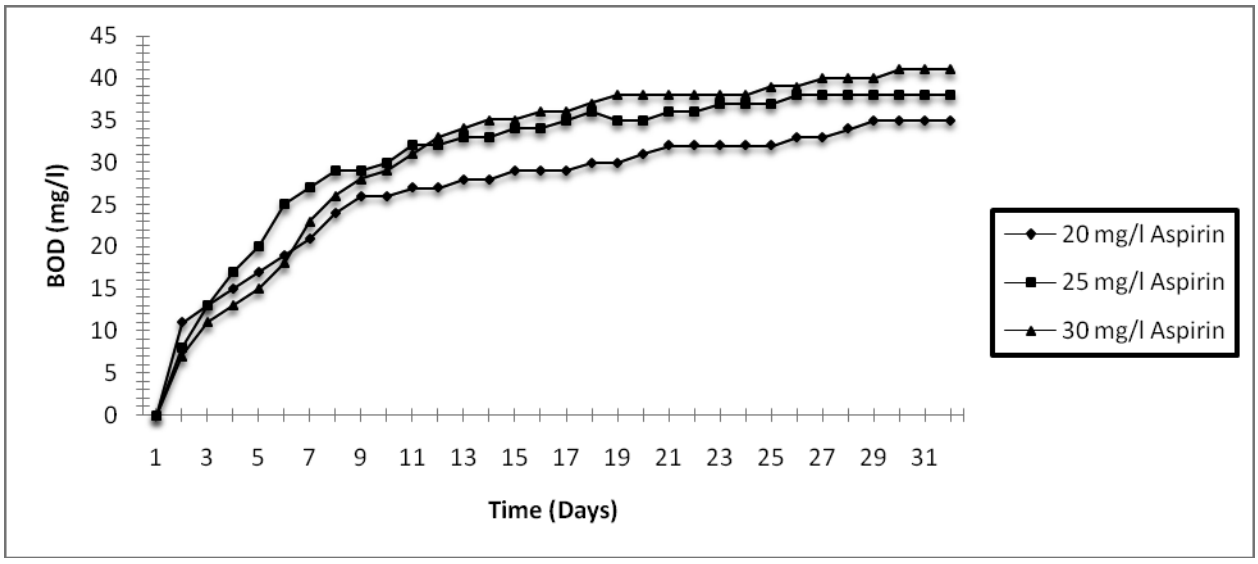


**Figure 2.** Biodegradation of a wastewater containing 20, 25 and 30 mg/l aspirin.

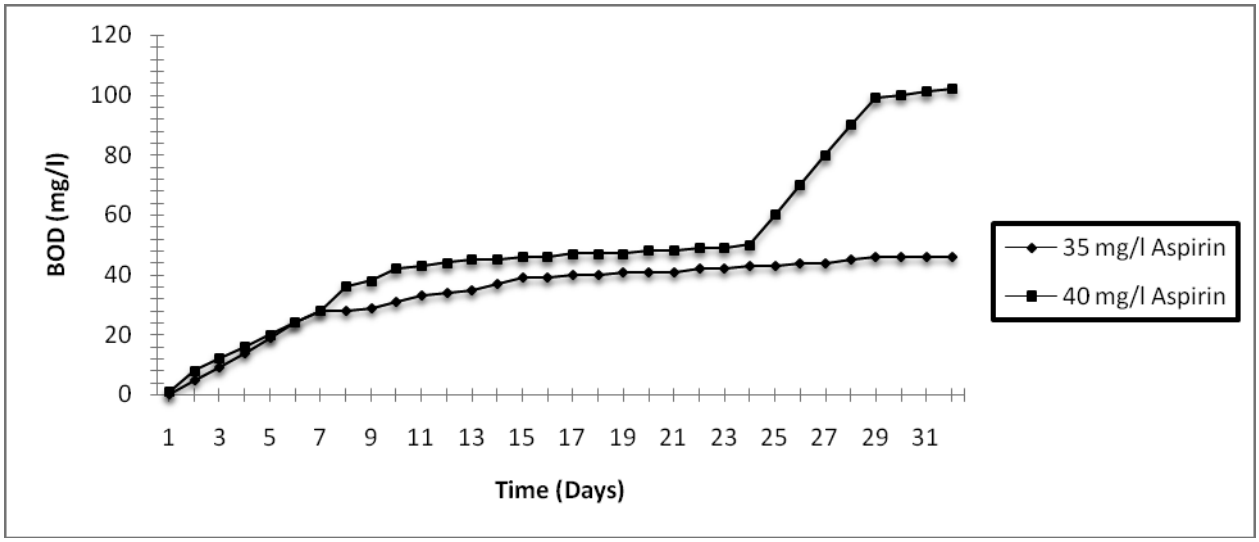


**Figure 3.** Biodegradation of a wastewater containing 35 and 40 mg/l aspirin.

The results of biodegradation of ibuprofen are shown in Fig. 4-6.

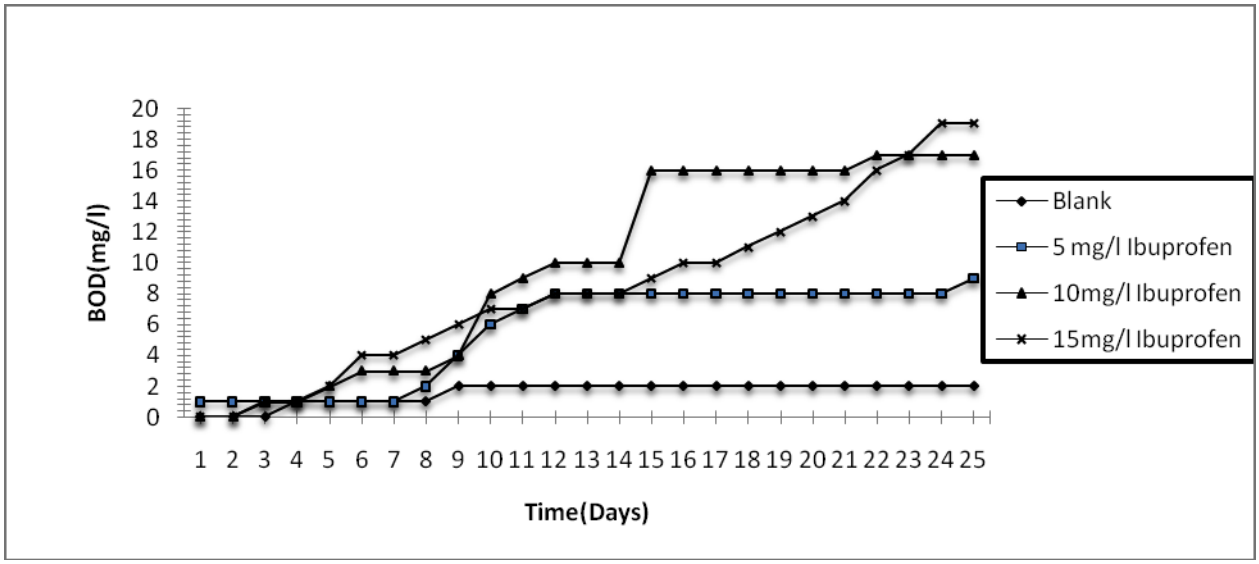


**Figure 4.** Biodegradation of a wastewater containing 5, 10 and 15 mg/l ibuprofen.

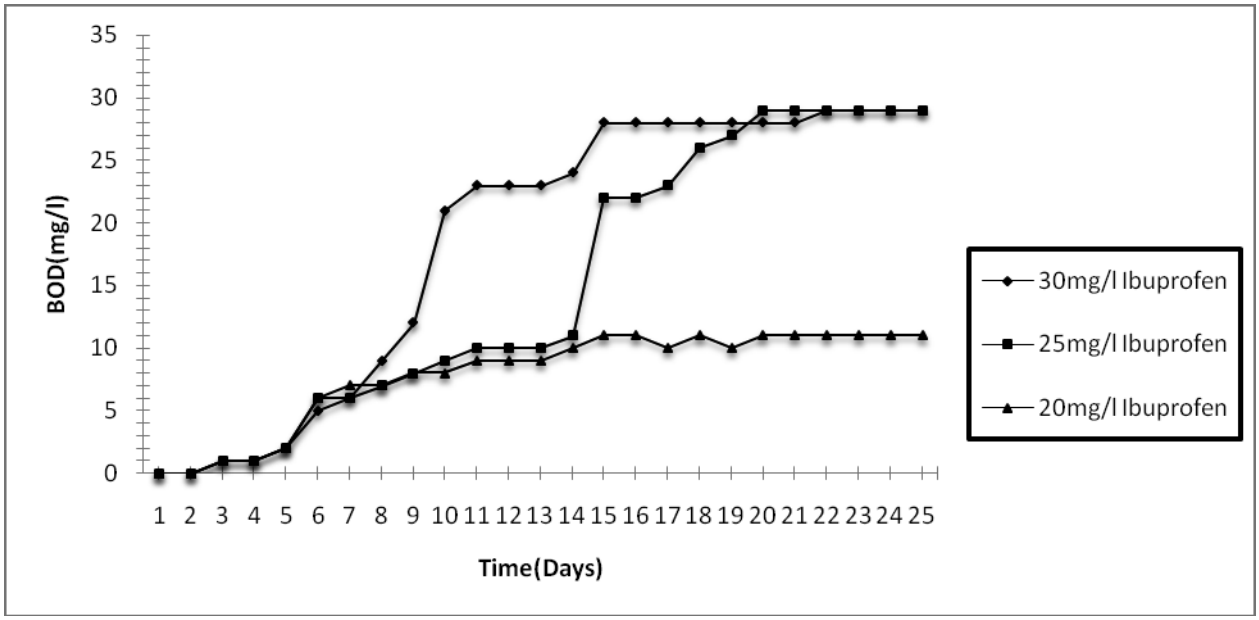


**Figure 5.** Biodegradation of a wastewater containing 35 and 40 mg/l aspirin.

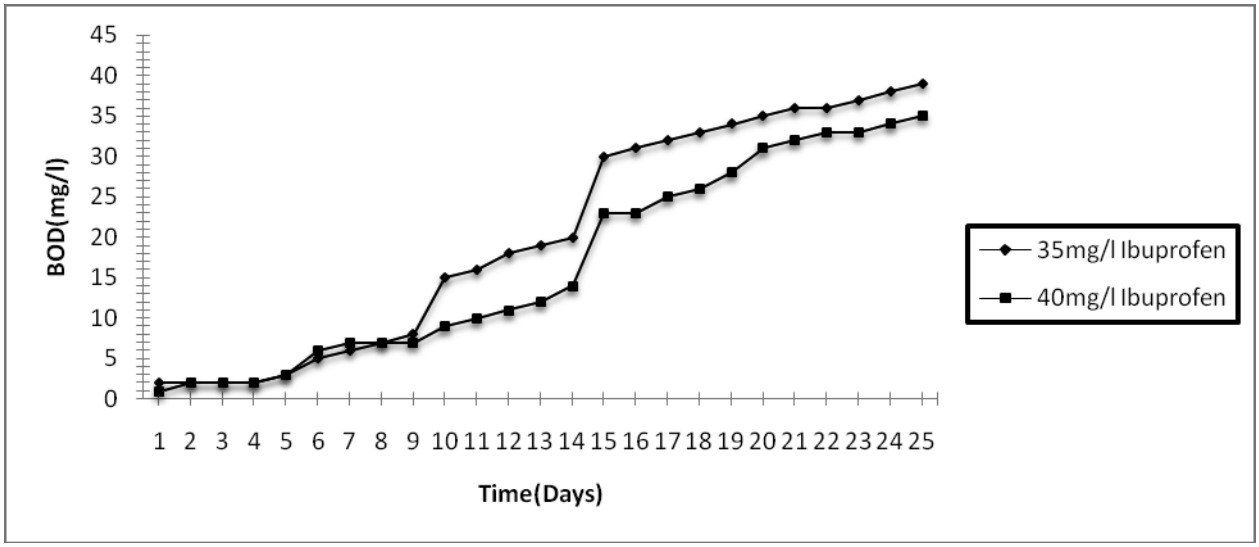


**Figure 6.** Biodegradation of a wastewater containing 35 and 40 mg/l ibuprofen.

The results of biodegradation of triphasil are shown in Fig. 7-9.

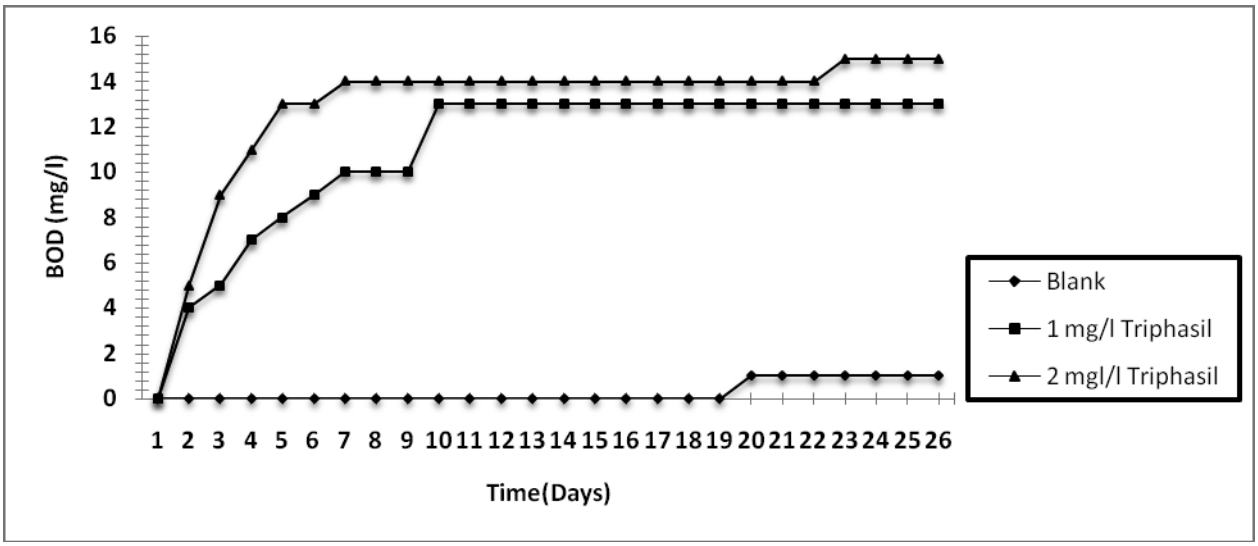


**Figure 7.** Biodegradation of a wastewater containing 1 and 2 mg/l triphasil.

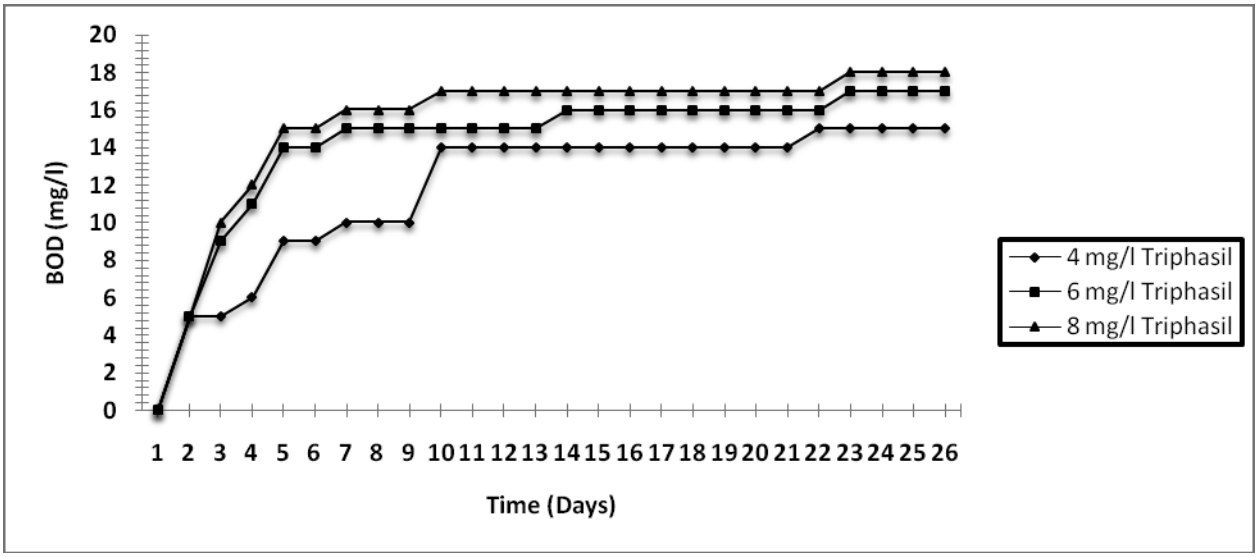


**Figure 8.** Biodegradation of wastewater 4, 6 and 8 mg/l triphasil.

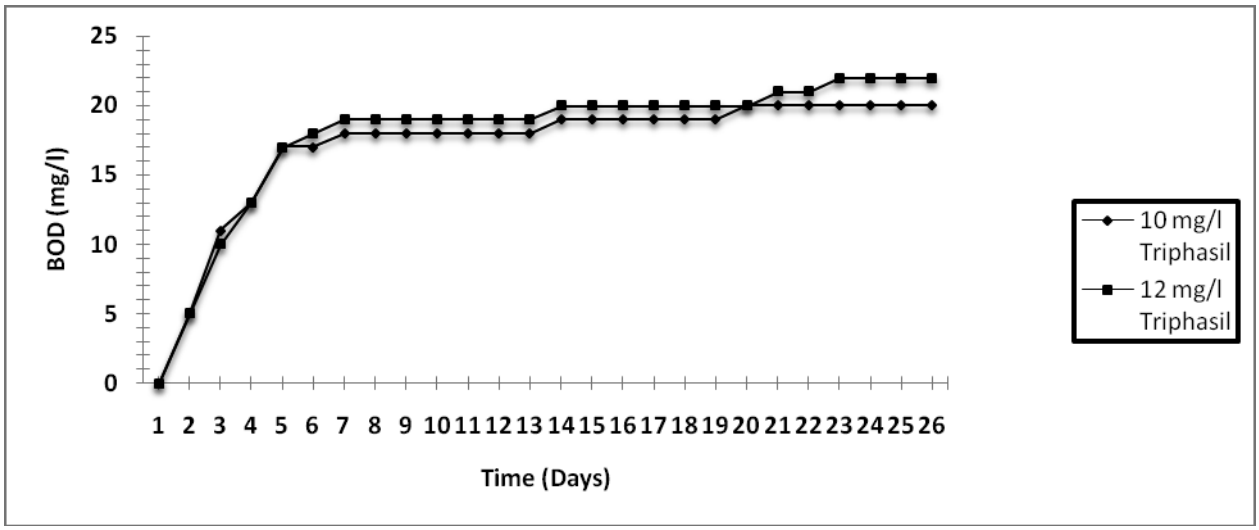


**Figure 9.** Biodegradation of wastewater containing 10 and 12 mg/l triphasil.

The results of biodegradation of amoxicillin are shown in Fig. 10-12.

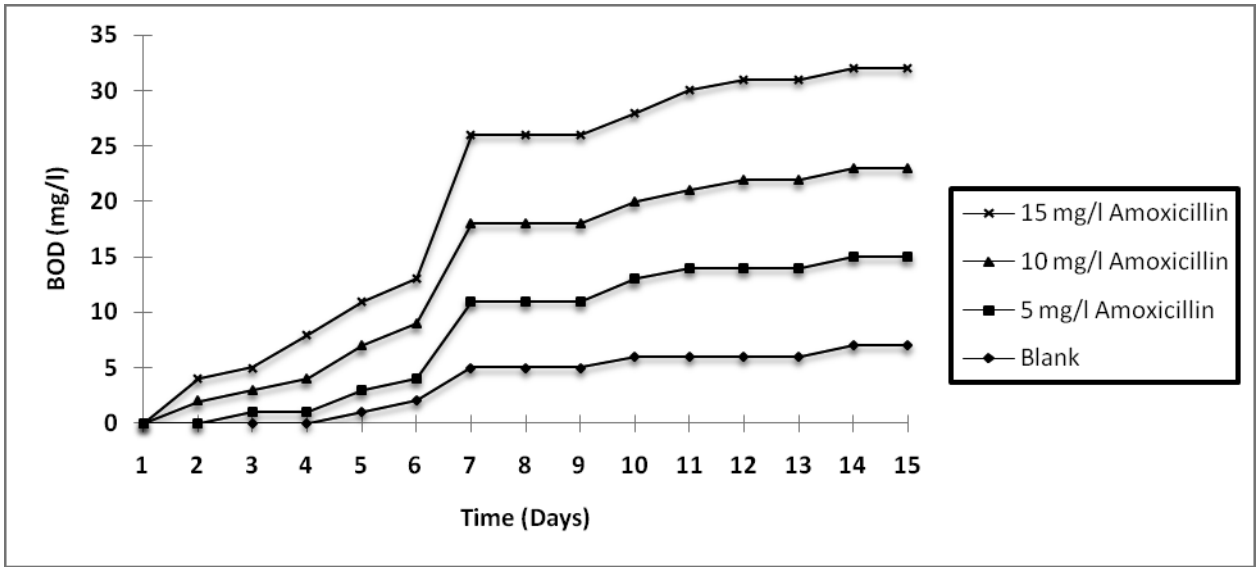


**Figure 10.** Biodegradation of wastewater containing 5, 10 and 15 mg/l amoxicillin.

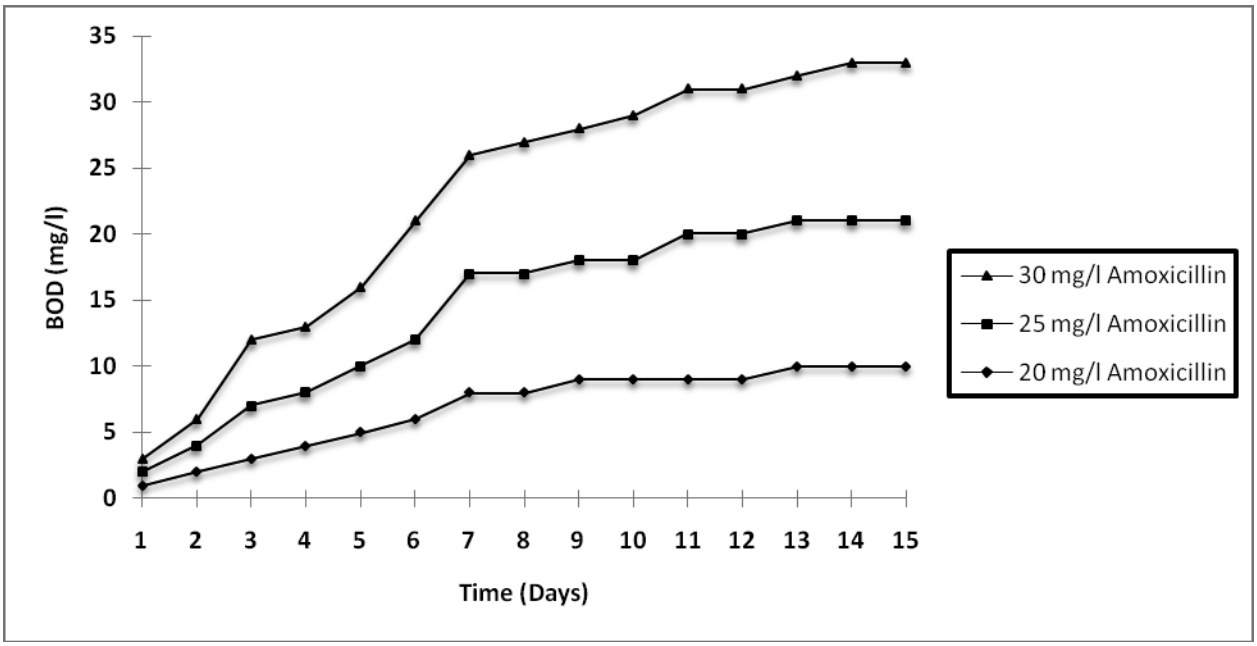


**Figure 11.** Biodegradation of a wastewater containing 20, 25 and 30 mg/l amoxicillin.

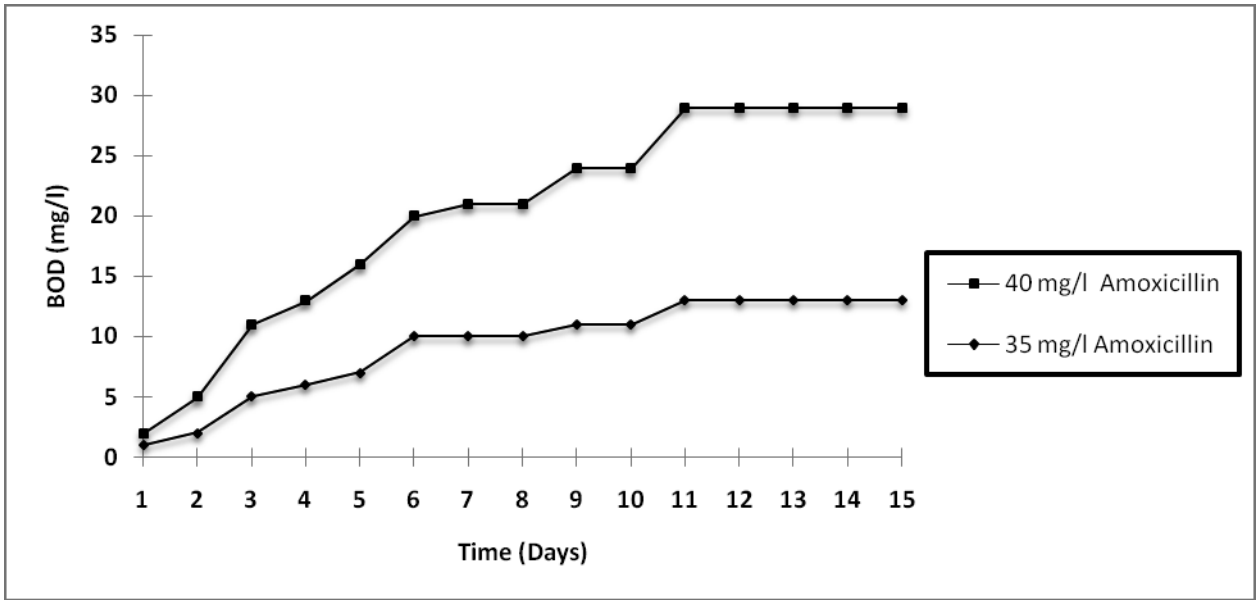


**Figure 12.** Biodegradation of a wastewater containing 35 and 40 mg/l amoxicillin.

Seed corrected data were obtained in all cases and the results are shown in Fig. 13-16.

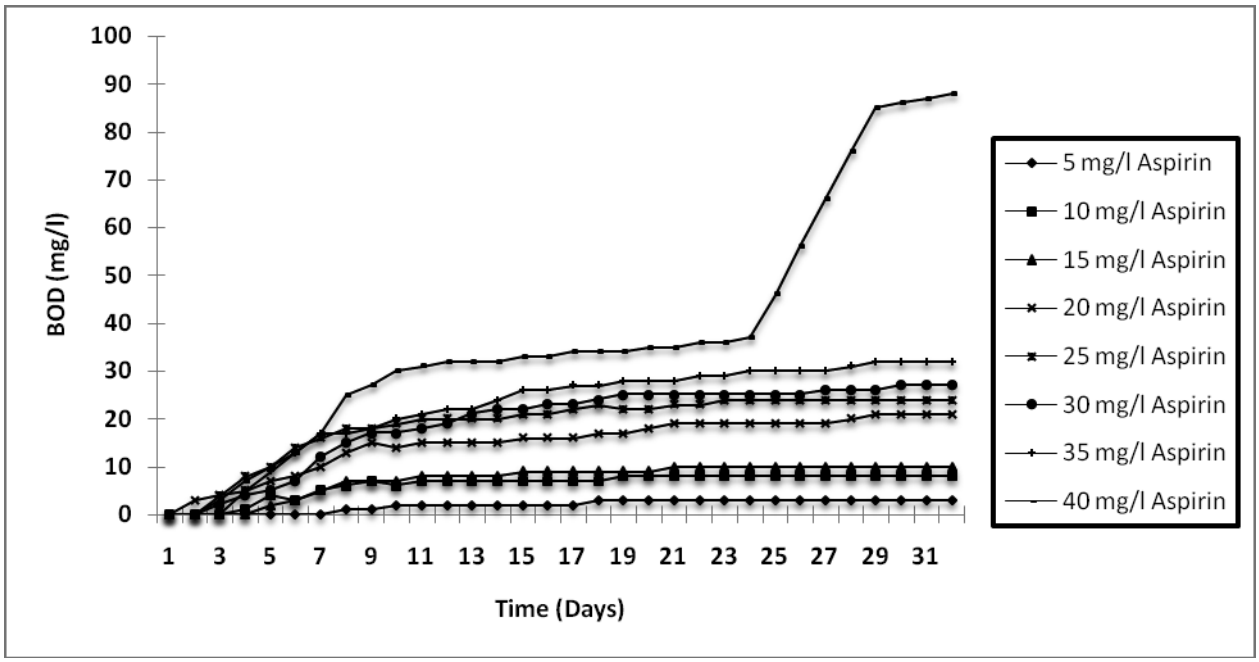


**Figure 13.** Seed corrected data for the different aspirin concentrations.

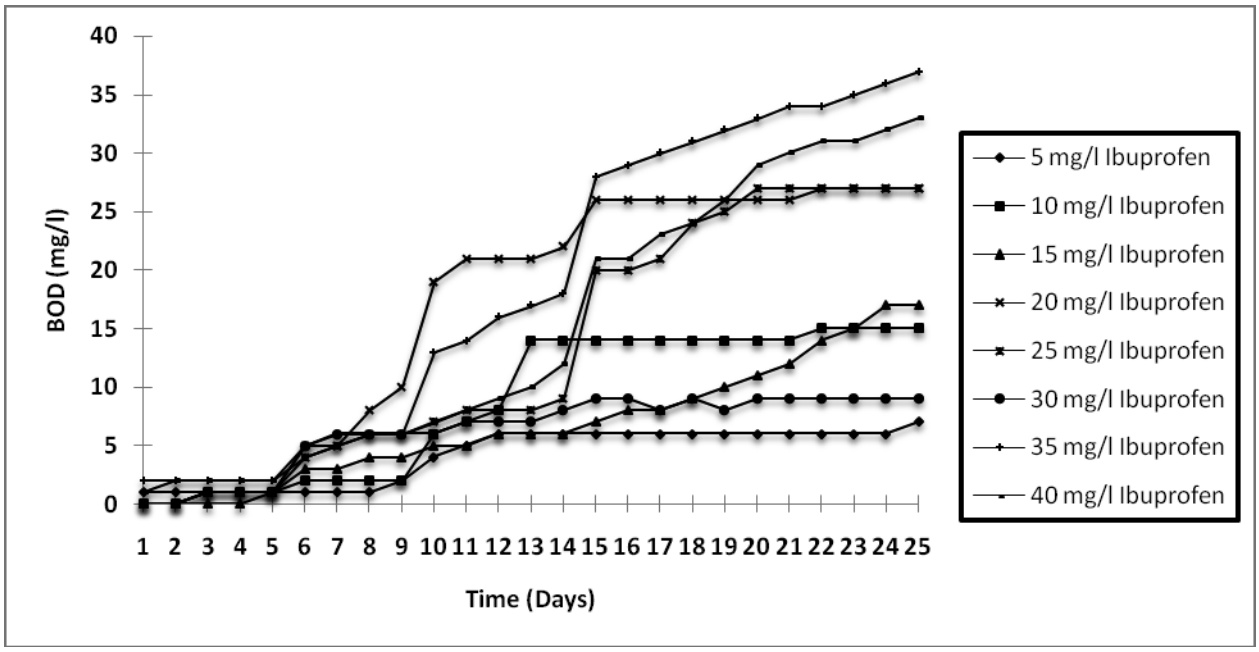


**Figure 14.** Seed corrected data for the different ibuprofen concentrations.

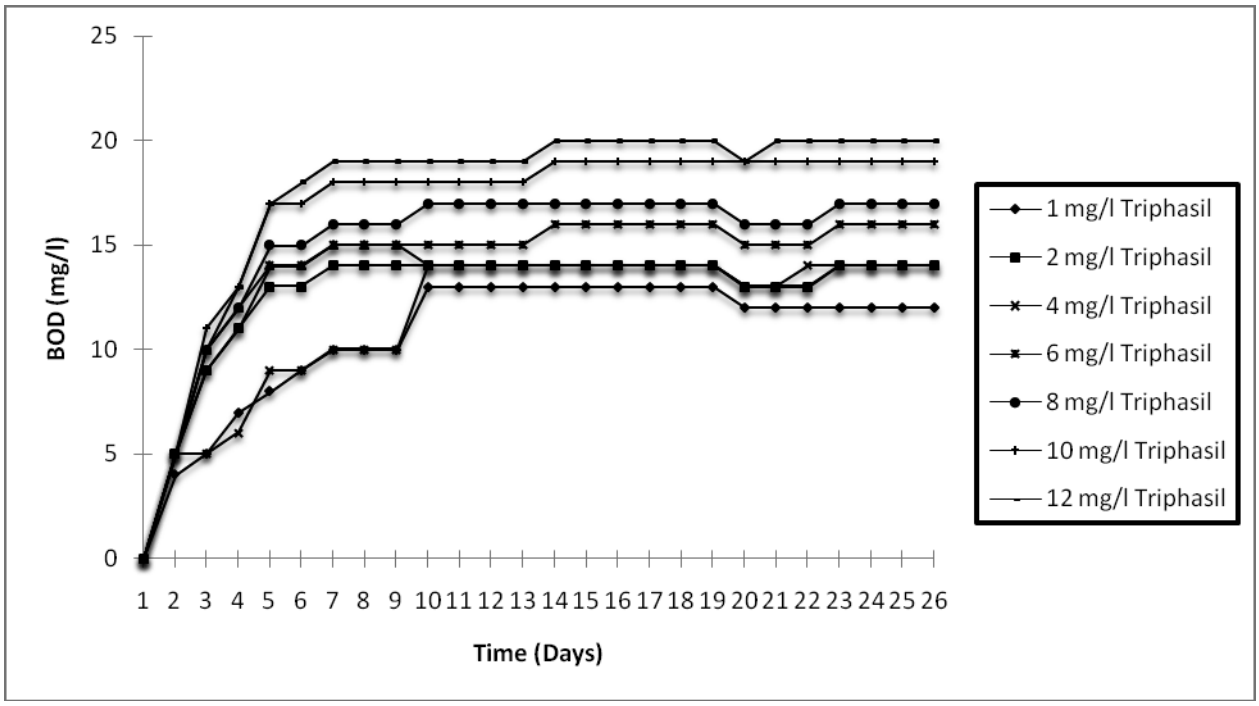


**Figure 15.** Seed corrected data for the different triphasil concentrations.

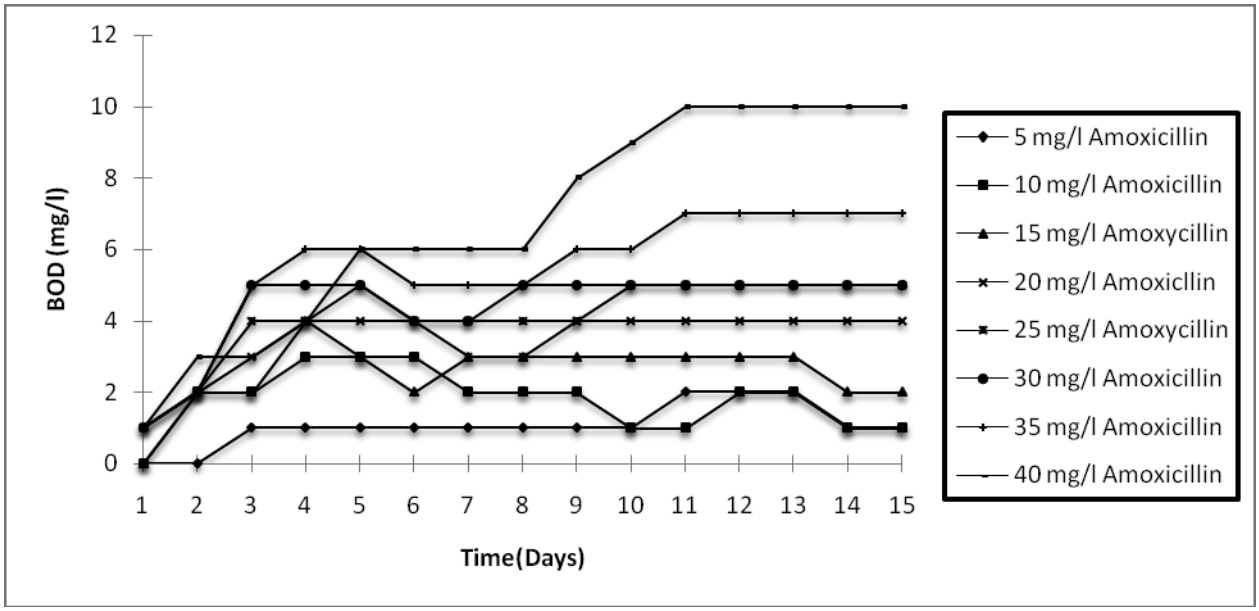


**Figure 16.** Seed corrected data for the different amoxicillin concentrations.

## DISCUSSION

In a set of experiments using unadapted inoculums from domestic sewage (as a seed) the biodegradations of aspirin at concentrations of 5-40 mg/liter (Figs. 1-3), ibuprofen at concentrations of 5-40 mg/liter (Figs. 4-6), triphasil at concentrations of 1-12 mg/liter (Figs. 7-9) and amoxicillin at concentrations of 5-40 mg/liter (Figs. 10-12) in synthetic wastewater, incubated for appropriate period of time were studied. It is shown that the plateau values for the different concentrations of aspirin increased as the concentrations of the substrate increased (Fig. 13), indicating the ability of the microorganisms to biodegrade aspirin at all tested concentrations.

The existence of ibuprofen as a pure substrate caused a temporary inhibition to the bacterial activities for a period of 5 days after which the activities started to increase. The plateau values for the different concentrations of ibuprofen increased as the concentrations of the substrate increased except for concentration 40 mg/l, where the plateau values are less than the corresponding values for the concentration 35 mg/l (Fig. 14), indicating appearance of some toxicity at the concentration 40 mg/l.

In case of effluents containing aspirin, triphasil and amoxicillin biodegradation seems to be a reasonable way of treatment. It appears that 10 days is a convenient time to reach satisfactory biodegradation in case of wastewater containing aspirin and triphasil (Fig. 13 and 15 respectively). In case of amoxicillin 4 days seems to be enough time for biodegradation (Fig. 16) and about 14 days in case of ibuprofen (Fig. 14).

## CONCLUSION

In case of effluents containing aspirin, triphasil and amoxicillin biodegradation seems to be a reasonable way of treatment. It appears that 10 days period is a convenient time to reach satisfactory biodegradation in case of wastewater containing aspirin and triphasil. In case of amoxicillin 4 days period seems to be enough time for biodegradation and about 14 days in case of ibuprofen.

The existence of ibuprofen as a pure substrate caused a temporary inhibition to the bacterial activities for a period of 5 days after which the activities increased. The plateau values for ibuprofen show that biodegradation is possible for concentrations equal to or less than 35 mg/l, after which the toxicity of ibuprofen may upset the biological treatment process.

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# الملخص العربى

# المعالجة البيولوچية لمياه صرف تحتوى على بقايا بعض الأدوية

هشام ربيع لطفى ونيفـل أويســب

قسم الكيمياء - كلية العلوم - جامعة ناميبيا - ويندهوك - ناميبيا

تناولت هذه الدراسة التحلل البيولوچى لبعض الأدوية الشائعة الاستخدام والتى يتوقع وجودها فى مياه الصرف الصحى، الأدوية التى تم دراسة تحللها البيولوچى فى هذا البحث هى الأسبرين والبروفين والترای فاسيل والأموكسيسيلين، وقد وجد فى هذه الدراسة أن الأسبرين يسهل تحلله البيولوچى عند التركيزات المستخدمة فى هذه الدراسة، وقد وجد أن البروفين يمكن تحلله بيولوچياً حتى تركيز ٣٥ ملجم / لتر بالرغم من حدوث سمية مؤقتة استمرت لمدة خمسة أيام عند كل التركيزات المستخدمة، أما تركيز ٤٠ ملجم / لتر فقد تسبب فى بعض السمية الدائمة للكائنات الدقيقة وتبين ذلك فى ضعف النشاط البكتيرى حتى نهاية التجربة، أثبتت الدراسة أيضاً أن الصرف الصحى المحتوى على تركيزات من أدوية الترای فاسيل والأموكسيسيلين يمكن تحللها بيولوچياً عند التركيزات المستخدمة فى الدراسة.

JOESE 5

# BIOLOGICAL TREATMENT OF A WASTEWATER CONTAINING PHARMACEUTICAL CONTAMINANTS

**Lotfy, H. R. and Eiseb, N. B.**

*Department of Chemistry and Biochemistry, Science Faculty,*
*University of Namibia, Windhoek, Namibia.*

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