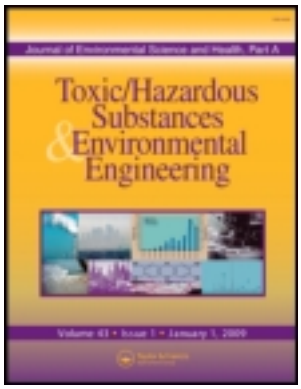

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Efficiency of membrane technology, activated charcoal, and a micelle-clay complex for removal of the acidic pharmaceutical mefenamic acid

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The efficiency of sequential advanced membrane technology wastewater treatment plant towards removal of a widely used non-steroid anti-inflammatory drug (NSAID) mefenamic acid was investigated. The sequential system included activated sludge, ultrafiltration by hollow fibre membranes with 100 kDa cutoff, and spiral wound membranes with 20 kDa cutoff, activated carbon and a reverse osmosis (RO) unit. The performance of the integrated plant showed complete removal of mefenamic acid from spiked wastewater samples. The activated carbon column was the most effective component in removing mefenamic acid with a removal efficiency of 97.2%. Stability study of mefenamic acid in pure water and Al-Quds activated sludge revealed that the anti-inflammatory drug was resistant to degradation in both environments. Batch adsorption of mefenamic acid by activated charcoal and a composite micelle (otadecyltrimethylammonium (ODTMA)–clay (montmorillonite) was determined at 25.0°C. Langmuir isotherm was found to fit the data with Q_{max} of 90.9 mg g^{-1} and 100.0 mg g^{-1} for activated carbon and micelle-clay complex, respectively. Filtration experiment by micelle-clay columns mixed with sand in the mg L^{-1} range revealed complete removal of the drug with much larger capacity than activated carbon column. The combined results demonstrated that an integration of a micelle-clay column in the plant system has a good potential to improve the removal efficiency of the plant towards NSAID drugs such as mefenamic acid.

Keywords: Anti-inflammatory drugs, wastewater, membrane technology, activated carbon, micelle-clay complex, adsorption isotherms, mefenamic acid.

Introduction

The consumption of water throughout the world is increasing, and the demand on water resources for household, commercial, industrial, and agricultural purposes is rising. This soaring demand is due to a rapidly expanding population, industrial expansion, and the need to expand irrigated agriculture. It is worth noting that there is a decrease in fresh water resources and low water availability.[1] Thus the treatment and reuse of wastewater for agriculture irrigation becomes a necessity to overcome this shortage.[2]

The occurrence of drugs in the environment is a subject of concern. The contamination is due to the consumption and the excretion of large quantities of pharmaceuticals via urine and feces in wastewaters. In fact, many administered pharmaceuticals are excreted without any chemical change or are released in conjugated or more polar transformed forms. The unused portion of pharmaceuticals can be also a source of sewage contaminations. In fact, a survey conducted by Kuspis and Krenzelok in the United States[3] indicated that more than 35% of people flushed medications down the toilet or sink. However, the percentage of unused medicines sold is still unknown.[4]

Even though residues of pharmaceuticals and their metabolites are usually detectable in the environment at trace levels, the low concentration level (ng/L-μg/L) can induce toxic effects, as in the cases of antibiotics and steroids that cause resistance in natural bacterial populations, or endocrine disruption effects.[5] Another main concern is the chronic and/or synergistic effects of "cocktails" of pharmaceuticals that humans release to water, which

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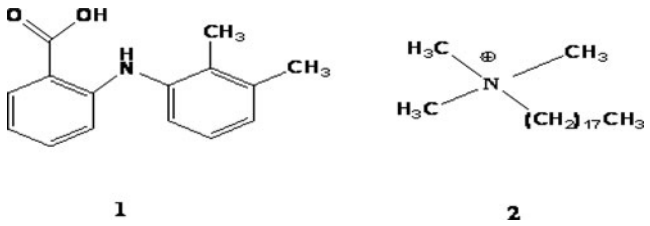
may enter into drinking water and be consumed by the population.[6–8]

Pharmaceuticals, especially medicines are designed to interact with receptors in human and animal bodies, but in aquatic environment the organisms having the same or similar enzymes or receptors as humans could experience similar pharmacodynamic effects, resulting in the destruction of organisms that are vital to environment.[6]

There are several methods for removal of pharmaceuticals from wastewater, among them:

(a) biodegradation process by which biological degradation can take place in wastewater by means of aerobic/anaerobic microbial degradation of the drug substance leading to reduction of the parental pharmaceuticals and/or their metabolites during wastewater treatment. The microbes include bacteria, yeasts, fungi, protozoa, and unicellular plants and rotifers; some of these organisms have the ability to degrade some of most hazardous and recalcitrant chemicals;
(b) deconjugation process by which pharmaceuticals are often metabolized in the liver, consequently sulfate and gluconoride conjugates of the parental drug are excreted. These excreted compounds will further deconjugate in domestic wastewater and within sewage treatment plants for organic compounds such as steroid hormones. These compounds were detected and attributed to the presence of large amounts of β-glucoronidase enzyme.
Gluconorides and sulfates of drugs will be degraded by the same process. This effect increases the excreted contribution of the active drugs to sewage and effluents;
(c) partitioning between the aqueous and organic biomass phase is considered the key component in determining the ultimate concentration of organic pollutants. Compounds with high log P (lipophylic molecules) values are known to sorb to sludge, whereas substances with lower values are more likely to remain in the aquatic phase, depending on the individual compound, and other substances sorbing to solids;
(d) removal during sludge treatment by which drugs may also be degraded by a biotic process (hydrolysis) during sewage treatment process. Many pharmaceuticals are not thermally stable, and so they might be expected to break down during chemical and biological processes at higher temperatures; and
(e) photodegradation by which several pharmaceuticals might be degraded due to the action of sunlight. Some pharmaceuticals such as diclofenac, which is analgesic/anti-inflammatory drug have been shown to degrade in aquatic environment due to ultraviolet (UV) light.[9–12]

Due to an incomplete elimination in wastewater treatment plants using the conventional treatment method, residues of pharmaceuticals are found in both wastewater and surface water.[13] To improve this situation an application of advanced treatment techniques, such as membrane filtration technology is required. A significant number of studies reported a satisfactory efficiency of nanofiltration, reverse osmosis,[14] advanced oxidation processes,[15] and activated carbon adsorption in removing pharmaceuticals from wastewater.[14,16].



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Scheme 1. Chemical structures for Mefenamic acid (**1**), and ODTMA (**2**).

Mefenamic acid [(2,3-dimethyl diphenyl) amino-2-carboxylic acid] (structure 1 in Scheme 1) belongs to the acidic, nonsteroidal, and anti-inflammatory drugs (NSAIDs). It is used for the relief of short-term moderate pain lasting less than 1 week, such as muscular aches and pains, menstrual cramps, headaches, and dental pain. It is typically prescribed for oral administration.[17,18]

Anti-inflammatory drugs such as ibuprofen, naproxen, mefenamic acid and ketoprofen were frequently detected in the discharges of wastewater treatment plants (WWTPs) and river water in Japan. The average concentrations range for those NSAIDs, ibuprofen, naproxen, mefenamic acid and ketoprofen, was 60–1580 ng/L in the effluent.[19] High removal efficiencies of the drugs were observed in the WWTP that has longer hydraulic retention time.[19]

In Sweden, removal of five NSAIDs, ibuprofen, ketoprofen, naproxen, diclofenac and mefenamic acid was assessed by batch experiments, with suspended biofilm carriers and activated sludge from several full-scale wastewater treatment plants. Biofilm carriers from full-scale nitrifying wastewater treatment plants, demonstrated considerably higher removal rates per unit biomass (i.e., suspended solids for the sludge and attached solids for the carriers) of diclofenac, ketoprofen and mefenamic acid compared to the sludge.[20]

The goal of this study was to explore the efficiency of advanced treatment technologies which consist of integration of activated sludge process with ultra-filtration membranes, hollow fiber and spiral wound membranes, activated carbon adsorbent, micelle-clay filters, and reverse osmosis in removing mefenamic acid.

The integrated components assembled in the wastewater treatment plant at Al-Quds University along with a micelle-clay complex octadecyltrimethylammonium (ODTMA, structure 2 in Scheme 1) and activated charcoal were tested for the removal of mefenamic acid from wastewater.

The micelle-clay composite used in this study was positively charged, had large surface area and included large hydrophobic domains. It was shown by X-ray diffraction, electron microscopy and adsorption experiments that the material characteristics of the micelle-clay complex are

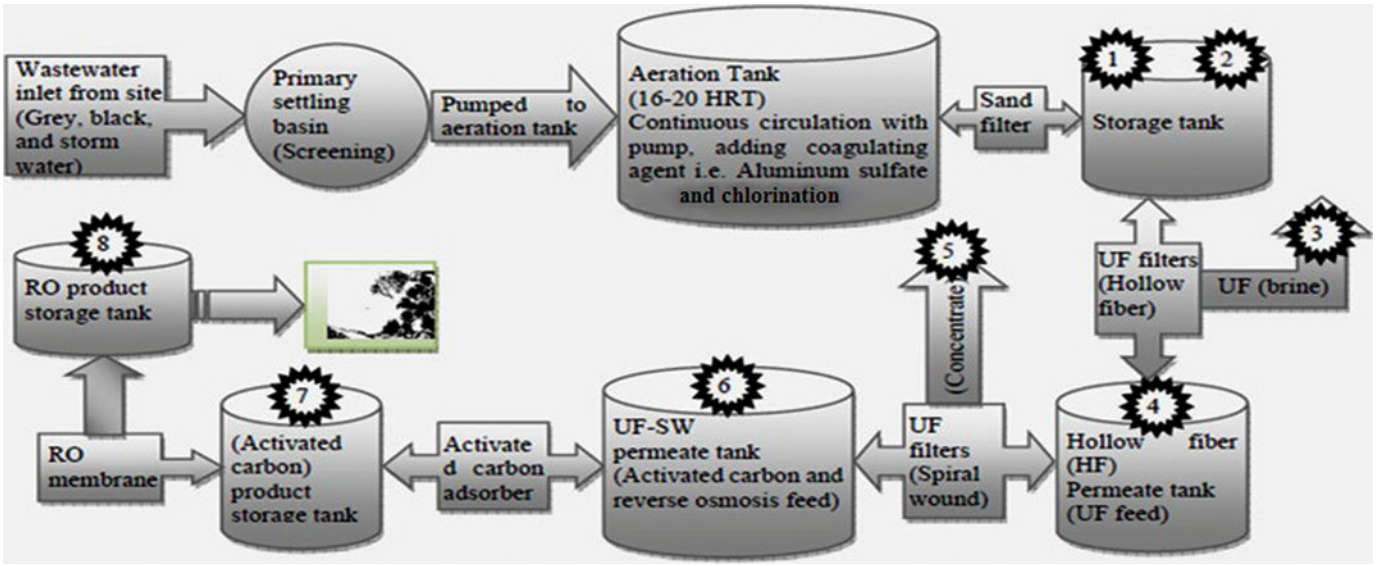

Fig. 1. Flow diagram of the waste-water treatment plant (WWTP) at Al-Quds University (color figure available online).

different from those of an organo-clay complex, which is formed by adsorption of the same organic cation ODTMA (Octadecyltrimethylammonium) as monomers.[21] Micelle-clay composites have already been proven useful in the removal of about 20 neutral and anionic pollutants.[22–24]

Experimental

Instrumentation

HPLC analyses were performed with High Performance Liquid Chromatography (HPLC-PDA) system, which consists of an alliance 2695 HPLC (Waters, Milford, MA, USA), and a Waters Micromass® Masslynx™ detector with Photo-diode array (Waters 2996). Data acquisition and control were carried out using Empower™ software (Waters). Analytes were separated on a 4.6 mm × 150 mm C18 XBridge® column (5 µm particle size) used in conjunction with a 4.6 mm × 20 µm XBridge™ C18 guard column. To construct the calibration curve for mefenamic acid, several concentrations of the drug (0.8, 2.0, 4.0, 10.0, 20.0, 80, 100, 200 and 400.0 ppm) were prepared. Then, 20 µL of each solution was injected into the HPLC and the peak for mefenamic acid was recorded using the following HPLC conditions: C18 column, wavelength = 350 nm, flow rate = 1.0 mL/min. Peak area vs. concentration of mefenamic acid (mg/L) was then plotted, and R^2 of the plot was recorded. Microfilter was used with 0.45 µm (Acrodisc® GHP, Waters); pH meter model HM-30G: TOA electronics™ was used in this study to measure the pH value for the samples. The C18 (1 g) cartridges (6cc single use) for general laboratory use were purchased from the Waters company.

The wastewater treatment plant (WWTP) at Al-Quds University consists of a primary treatment (two-stage primary settling basin), secondary (activated sludge with a hydraulic retention time of 16–20 h, coagulation and chlorination) treatment (Fig. 1). Then the secondary effluent is introduced to a sand filter before entering the ultrafiltration (UF) membrane (hollow fiber and spiral wound). After the ultrafiltration process, the effluent is subjected to activated carbon absorbers followed by a reverse osmosis (advanced treatment). Then a blend of all effluents is used for irrigation. The ultrafiltration process is made of two small scale membrane treatment plants with a capacity of 12 m^3/day. The first UF unit is equipped with two pressure vessels (AST Technologies, model number 8000 WW 1000^{-2}M) that house the hollow fiber membranes with 100 kD cutoff (AST Technologies, Model No. 8000- WWOUT-IN-8080, Sigma-Aldrich, Munich, Germany). The two units are designed to deliver 1.5 m^3/h. The second unit is equipped with 2 × 4 inch pressure vessels with pressure resistance up to 150 psi.

Each vessel holds two separation membranes (spiral wound with 20 kD cutoffs, which are equivalent to 0.01 micron separation rate). The designed permeate capacity of the system is 0.5–0.8 m^3/h. This membrane can remove bacteria, suspended solids, turbidity agents, oil and emulsions. The reverse osmosis (RO) system consists of 1 × 4 inch pressure vessel made from composite material with pressure resistance up to 400 psi. The vessel holds two 4-inch special thin film separation membranes made of polyamide with pH range 1–11 (BW30-4040 by Dow Film Tec., Edina, MN, USA). Membrane anti-scalants made of phosphate in water with active ingredient of *o*-phosphonic acid disodium salt (Product NCS-106-FG, Dow Film Tec.) are continuously dosed to the RO feed at concentration of 4 ppm in order to prevent deposition of divalent ions. The system is designed to remove major ions and heavy metals. The designed RO permeate capacity of the system is 0.45–0.5 m^3/h.

Chemicals and reagents

Pure standard of mefenamic acid (>99%) was obtained from Beir-Zeit pharmaceutical company (Ramallah, Palestine). Acetonitrile and methanol HPLC grade, *o*-phosphoric acid, magnesium sulfate, charcoal activated granules with particle size (≤700.0 μ), and octadecyltrimethyl ammonium (ODTMA) complex were purchased from Sigma Chemical Company (Munich, Germany).

Methods

Preparation of micelle-clay complex

The ODMTA micelle-clay complex was prepared by mixing a clay-mineral (montmorillonite) with cationic surfactant octadecyltrimethylammonium (for the chemical structure of ODTMA, see Scheme 1) with a critical micelle concentration (CMC) value of 0.3 mM as described previously.[22] Briefly, 12 mM of ODTMA was stirred with 10 g L^{-1} clay for 72 h. Suspensions were centrifuged for 20 min at 15,000 *g*, supernatants were discarded, and the complex was lyophilized. The obtained complex by virtue of its positive charge with hydrophobic region is capable of efficiently binding negatively charged organic molecules.[21–26]

Efficiency of micelle (ODTMA)-clay complex and charcoal for removal of mefenamic acid

Here the ODTMA-clay complex was tested for a removal of mefenamic acid from water using the following procedure: 200 mg L^{-1} of mefenamic acid was prepared in buffer at pH = 8.0. The pH was adjusted by sodium hydroxide (1M); 100 mL from this solution were transferred to a 200-mL Erlenmeyer flask (Darmstadt, Germany), 0.5 g of charcoal, or micelle (ODTMA)-clay complex was then added to the flask. Then the flask was shaken for 180 min. Samples were taken at different time intervals: 0, 5, 10, 20, 30, 40, 50, 60, 80, 100, 120, 150 and 180 min. Then, each sample was centrifuged for 5 min at 250 rpm, passed through 0.45-μM filter and analyzed by HPLC.

Adsorption isotherms

Equilibrium relationships between adsorbents (micelle-clay complex and activated charcoal) and adsorbate (mefenamic acid) was accomplished by studying the percentage adsorbate removal whose concentrations ranged from 50 to 1000 mg L^{-1} in a volume of 100 mL. The pH of the initial solutions was adjusted to 8.0. The sorbent dose was 0.5 g of either charcoal or ODTMA-clay complex.

Analysis of adsorption isotherms

Adsorption isotherms were analyzed by the linear form of the Langmuir isotherm, Eq. 1, as in:[26]

$$C_e/Q_e = 1/(k\,Q_{max}) + Ce/Q_{max} \quad (1)$$

in which C_e (mg/L) is equilibrium concentration of mefenamic acid, Q_e (mg/g) is equilibrium mass of adsorbed mefenamic acid per gram of complex or charcoal, k (L/mg) is Langmuir binding constant, and Q_{max} (mg/g) is maximum mass of drug removed per gram of complex, or charcoal.

Column experiments. Column filter experiments were performed with 100/1 (w/w) mixtures of quartz sand and ODTMA-clay complex (20 cm layer) in a column of 25 cm length and 5 cm diameter, which included either 6.5 g of micelle-clay complex, or activated carbon. The bottom of the column was covered by 3 cm layer of quartz sand. Quartz sand was thoroughly washed by distilled water and dried at 105°C for 24 h. A solution of mefenamic acid at a concentration of 9.42 ppm was passed through the above columns. The flow rate was 52 mL min^{-1}. Fractions were collected for assay of mefenamic acid.

Stability study of mefenamic acid

Stability study in distilled water. Stability study of mefenamic acid was performed with a 50 mg L^{-1} solution, prepared by dissolving mefenamic acid in distilled water adjusted to pH 8.0 using 1M sodium hydroxide. Samples at specific time intervals: 0, 1, 2, 4, 5, 10, 15, 20, 25 and 30 days were taken, and analyzed by HPLC.

Stability study in the presence of Al-Quds sludge. Stability study for mefenamic acid in the presence of Al-Quds sludge was performed by adding 1000 mL of suspended sludge (25% concentration) to 0.5 g of sorbent under continuous aeration to maintain bacterial growth in the sludge environment. Reproducibility studies were performed in triplicate and the average values were recorded together with standard deviations.

Physical, chemical and biological parameters of Al-Quds sludge

The physical, chemical and biological qualities of Al-Quds University sludge used in the stability studies are presented in Table 1. Careful inspection of Table 1 reveals that the sludge is rich in N and P with high values of BOD, COD, TDS, settable solids and bacterial count. These values are expected since the analyzed samples were taken from the aeration tank in the activated sludge unit within the wastewater treatment plant.

Data obtained by physiological and biochemical characterization of bacterial community in Al-Quds activated sludge allowed the identification of the following bacterial species: *Escherichia coli, Enterobacter sakazakii, Citrobacter freundii, Pseudomonas aeruginosa, Klebsiella pneumonia, Enterobacter cloacae*, *Enterobacter amnigenus, Enterobacter aerogenes, Salmonella spp., and Serratia liquefaciens.*

Table 1. Physical, chemical and biological parameters for wastewater sample (Al-Quds University WWTPs).

Parameter	*Results*	*Units*	*Parameter*	*Results*	*Units*
pH	7.32	—	TSS	3700 ± 100	mg/L
Conductivity	2000	μSm/cm	BOD	900 ± 150	mg/L
Temperature	15.5	°C	COD	1900 ± 300	mg/L
Turbidity	5000 ± 200	NTU	NH4-N	59.5	mg/L
DO	0.4	mg/L	PO4-P	14.3	mg/L
TS	4200 ± 200	mg/L	FC (E.coli)	290,000 ± 150,000	cfu/100 mL
TDS	620 ± 50	mg/L	TC	6,500,000 ± 3,000,000	cfu/100 mL
Settable solids	240	mL/L	TAC	2,600,000 ± 1,300,000	cfu/100 mL

DO: Dissolved Oxygen, TAC: Total Aerobic Count.

Efficiency of the wastewater treatment plant (WWTP) of Al-Quds University for removal of mefenamic acid

The efficiency of different units of the advanced integrated wastewater treatment plant of Al-Quds University for the removal of mefenamic acid from wastewater was studied by spiking the secondary effluent with 40 mg L^{-1} of drug in a storage tank (25 g mefenamic acid in 625 L secondary treated wastewater). Samples were taken from the following points of the WWTP: (1) storage tank (before running wastewater treatment plant) (2), (3), and (4) feed-, brine- and product-points of the HF-UF membrane, respectively (5) and (6) concentrate-, and permeate-UF point of the HF-SW membrane, (7) activated carbon point, and (8) reverse osmosis point (Fig. 1). These samples were treated as follows: 10 mL of sample was loaded into a C18 cartridge, and allowed to pass through a cartridge by effect of gravity. Mefenamic acid adsorbed on the C18 cartridge was then eluted using 10 mL of acetonitrile; 20 μL of the eluted solution was injected into the HPLC and chromatograms were taken.

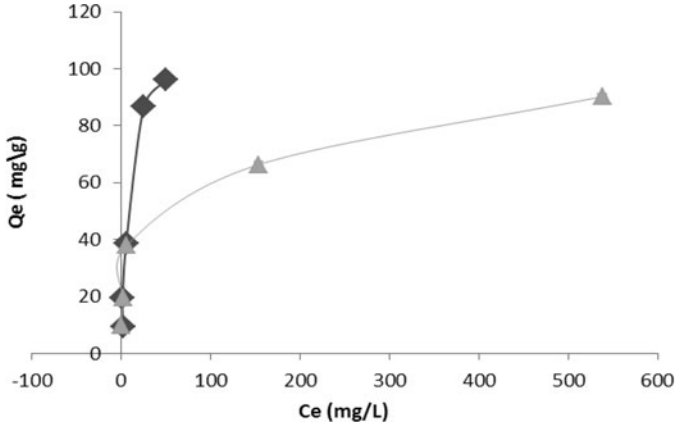

Fig. 2. Adsorption isotherms for the removal of mefenamic acid by micelle-clay complex [■] and by activated charcoal (▲) (pH 8.0, Temp. 25°C).

Results and discussions

Stability study of mefenamic acid

The results showed that mefenamic acid was stable in both distilled water and Al-Quds University activated sludge during a period of more than one month.

Adsorption isotherms

The adsorption of mefenamic acid at several initial concentrations (50, 100, 200, 500, and 1000 mg/L) on a micelle-clay complex and activated charcoal was investigated (Fig. 2). The data fitted the Langmuir isotherm (Eq. 1) with R^2 greater than 0.99 for both cases (Fig. 3). The Langmuir constants (k and $Q_{max)}$ were calculated and their values are presented in Table 2. Inspection of Table 2 revealed that the micelle-clay complex has larger k and Q_{max} values than activated carbon, thus rendering the former as a better adsorbent for the removal of mefenamic acid.

Kinetics of adsorption of mefenamic acid on the micelle-clay complex and activated charcoal

Mefenamic acid removal by a micelle-clay complex and activated charcoal was studied at pH 8.0. Samples were taken at different time intervals (0–180 min). The results revealed that for a micelle-clay complex only 5 min sufficed for complete removal of mefenamic acid (97% removal), whereas for activated charcoal only 28% was removed during the

Table 2. Langmuir adsorption parameters (k and Q_{max}) and the correlation coefficient (R^2) values obtained from the adsorption of mefenamic acid on both adsorbents, a micelle-clay complex and activated charcoal.

	Langmuir		
Adsorbents	*K (L mg)*	*Q_{max} (mg g)*	*R^2*
Micelle-clay complex	0.105 ± 0.02	100.0 ± 5	0.990
Charcoal	0.065 ± 0.01	90.9 ± 3	0.999

Table 3. Removal of Mefenamic acid by filtration of its solution (9.42 mgL^{-1}) through a laboratory filter, which included either a micelle-clay complex, or activated carbon mixed with excess sand at 1:100 (w/w).[a]

Volume filtered (L)	*Type column*	*Emerging concentration (mg L)*		*% Removed exp. (avg)*
		Column 1	*Column 2*	
0.78	Micelle clay complex	0	0	100 ± 0
	Activated carbon	3.10	3.90	63 ± 5
1.56	Micelle clay complex	0	0	100 ± 0
	Activated carbon	4.21	4.91	51.6 ± 5
2.34	Micelle clay complex	0	0	100 ± 0

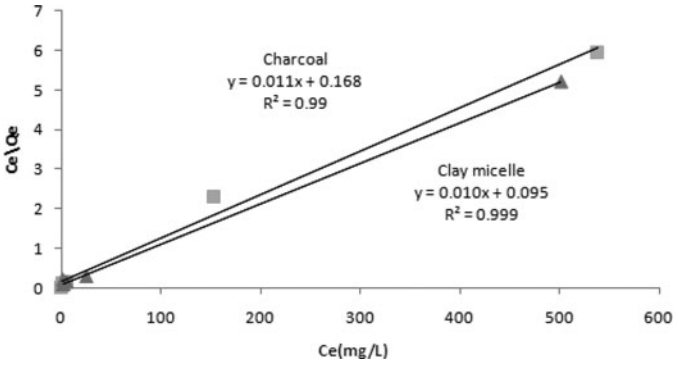

Fig. 3. Langmuir isotherms for the removal of mefenamic acid by micelle-clay complex (▲) and by activated charcoal [■] (pH 8.0, Temp. 25°C).

same period. These results indicate that the adsorption of mefenamic acid on a micelle-clay complex is much faster than that on activated charcoal. This difference between the two adsorbents can be attributed to electrostatic interactions between the micelle-clay complex and the hydrophilic moiety of mefenamic acid, which are absent in activated carbon. However, the result that the activated carbon is characterized by slow kinetics of adsorption is also observed in the case of neutral pollutants, e.g., bromacil.[27] The relatively slow kinetics of adsorption is more striking in filtration, as demonstrated next.

Filtration results

Mefenemic acid solutions in the mg L^{-1} range were passed through filters which included the micelle-clay complex or activated carbon mixed with excess sand at 1:100 ratios (w/w). The results are presented in Table 3. The results indicate a significant advantage of the micelle-clay filter in removing mefenamic acid compared to that removed by activated charcoal. This was not surprising, since the results for the adsorption isotherm and in particular the kinetics have clearly shown that the micelle-clay-complex was more efficient than activated carbon in removing mefenamic acid from water.

Efficiency of the wastewater treatment plant (WWTP) at Al-Quds University for mefenamic acid removal

The efficiency of WWTP at Al-Quds University for a removal of mefenamic acid was studied. Samples were taken from the same locations as described previously. The results of the samples taken from the hollow fiber points (UF-HF) indicated that mefenamic acid was 74.0% removed at this stage, whereas 94.3% of mefenamic acid was removed after passing the spiral wound (SW) stage (Tables 4 and 5). The sample taken after passing the point of activated carbon column showed that mefenamic acid was almost completely removed (98.8%). In addition, samples taken after passing the RO unit demonstrated that complete removal of mefenamic acid was achieved (99.5%).

However, it should be realized that the initial concentrations of drug encountered by treatment elements at the

Table 4. Removal of mefenamic acid through a hollow fiber (UF-HF), spiral wound (UF-SW), activated carbon and reverse osmosis from wastewater treatment plant at Al-Quds University.

No.	*Sample description*		*Conc. of mefenamic Acid (mg L) First trial*	*Conc. of mefenamic acid (mg L) Second trial*	*Conc. of mefenamic acid (mg L) Third trial*
1	Blank (before addition of Mefenamic acid)		0	0	0
2	The initial concentration of Mefenamic acid in storage tank (after addition of Mefenamic acid)		42.00	40.0	39.5
3	HF-UF	Feed point	18.80	37.90	38.3
		Brine point	42.00	38.00	36.0
		Product point	1.10	11.30	18.4
4	HF-SW	Concentrated UF point	1.10	11.20	16.0
		Permeated UF point	0.15	1.90	4.7
5	Activated carbon point		0.12	0.70	0.6
6	Reverse osmosis (RO)	Brine RO point	0.45	0.72	0.6
		Permeated RO point	0.07	0	0.5

Table 5. The cumulative percentage removal of mefenamic acid by the different purification elements in Table 4.

Trial No.	*Hollow fiber (HF)*	*Spiral wound (SW)*	*Activated carbon*	*Reverse osmosis (R.O)*
1	97.8%	99.6%	99.7%	99.8%
2	71.8%	95.2%	98.2%	100.0%
3	53.4%	88.1%	98.5%	98.7%
Average	74.3%	94.3%	98.8%	99.5%
SD	18.2%	4.7%	0.8%	0.7%

Al-Quds WWTP diminish along their sequence. One deduction from Table 5 is that the combination of sequence of purification elements enables to achieve almost complete removal of mefenamic acid from its initial value of 40 mg L^{-1}. On the other hand the actual percentages of removal relative to the initial solutions at some of the purification elements are smaller. Thus the AC removes only an average of 75.8% relative to the initial concentration of mefenamic acid that reaches it, which is in general accord with Table 3. The actual average percentage of removal of the drug by the RO relative to the concentration of mefenamic acid that reaches it is only 62%.

Conclusions

The kinetic study revealed that the NSAID mefenamic acid was stable in distilled water and in Al-Quds sludge for more than 30 days. Hence, there is a pressing need to find an appropriate method for the removal of this anti-inflammatory drug from wastewater.

Adsorption isotherms and kinetics demonstrated that the micelle (ODTMA)–clay complex has much higher efficiency in the removal of mefenamic acid from water than activated carbon. Filtration results showed the superiority of micelle-clay complex over activated charcoal in the removal of mefenamic acid from the environment. The current combination of purification elements enables the removal of mefenamic acid from wastewater almost completely, but the combined results suggest that incorporation of micelle-clay filters in wastewater treatment plants is promising in removing mefenamic acid efficiently from the environment.

Acknowledgments

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