

Application of conventional and O₃-BAC processes to treat organic matter and antibiotic pollutants in a lake in East China

X. B. Liao, X. J. Zhang, J. Wang, X. B. Li, C. K. Wang and C. Chen

ABSTRACT

The occurrence and treatment of antibiotics in a micro-polluted lake which serves as a drinking water source in East China was surveyed. A pilot plant with conventional and O₃-BAC (biological activated carbon) processes was set up to investigate its effectiveness in dealing with the contaminants. Solid phase extraction (SPE) coupled with electro-spray ionization-tandem mass spectrometry (LC-ESI-MS-MS) was applied to detect various antibiotics simultaneously. Three groups of antibiotics, i.e. sulfonamides, fluoroquinolones and tetracyclines, were detected in the source water. The gross concentrations of them in the lake are up to 471, 23.4 and 1,039 ng/L, respectively. The conventional and O₃-BAC processes could remove 78.9, 62.4 and 70.2% of them, respectively. Among the antibiotics, tetracyclines could be effectively removed by ozonation, while fluoroquinolones could be removed by the coagulation–sedimentation process. BAC could not degrade fluoroquinolones but enabled the reduction of the other two antibiotics. In addition, O₃-BAC was an effective technology for the removal of bulk organic matter. The concentrations of chemical oxygen demand (COD_{Mn}), UV₂₅₄ and dissolved organic carbon (DOC) in the effluent of the up-flow BAC process were 2.31 mg/L, 0.034 cm⁻¹ and 1.76 mg/L, respectively, with the corresponding removal rates of 45.1, 67.3 and 65.1%, respectively. In all, the combined conventional and O₃-BAC process was the best available technology to remove organic matter as well as antibiotics.

Key words | antibiotics, biological activated carbon, drinking water, organic matter, ozone

X. B. Liao
X. J. Zhang
J. Wang
X. B. Li
C. K. Wang
C. Chen (corresponding author)
School of Environment,
Tsinghua University,
Beijing, 100084,
China
E-mail: chen_water@tsinghua.edu.cn

INTRODUCTION

Antibiotics are extensively used in animal farming and aquaculture for preventing or treating infections, as well as for promoting growth (Holmstrom *et al.* 2003). In recent years, antibiotics have been detected in the drinking water treatment plants of Canada, the USA, China and other countries (Xu *et al.* 2007b; Araceli *et al.* 2010). The occurrence of antibiotics in the aquatic environment is of increasing concern. The harm caused by antibiotics can be attributed to the following aspects: possible adverse effects on aquatic ecology, potential for increased antibiotic resistance among microbial populations continuously exposed to low levels of antibiotics, and the negative influence on human health (Gilliver *et al.* 1999). Their presence in drinking water probably has an unknown health effect on humans

due to the chronic low-level exposure to these substances over a lifetime. Therefore, antibiotics should be removed during the treatment of drinking water (Pomati *et al.* 2006).

The physical-chemical properties of antibiotics affect their removal. For instance, conventional processes are unable to eliminate soluble and hydrophilic compounds such as sulfonamides (Nakada *et al.* 2007); however hydrophobic compounds, such as carbamazepine, could be effectively removed by a GAC (granular activated carbon) filter (Stackelberg *et al.* 2007). In addition, Chu & Fernandes (1989) indicated that quinolones had a small or linear substituent at the C-7 position and exhibited lower potency than analogues substituted with heterocyclic rings such as ciprofloxacin, thus they may be more difficult to be absorbed

than ciprofloxacin. Therefore, appropriate processes should be chosen to address antibiotics with different properties.

Some researchers investigated the removal of antibiotics in the drinking water treatment process. The conventional practices are not effective in removing antibiotics. Paul *et al.* (2007) found that GAC filtration accounted for 53% of the removal of antibiotics from the aqueous phase; disinfection accounted for 32%, and clarification accounted for 15%. However, GAC removal capacities have been found to be higher for the lower molecular weight components (Karanfil *et al.* 1996). These findings have important practical implications for the design and operation of GAC adsorbers and pretreatment processes. Some of the hydrophilic antibiotics may be difficult to be absorbed or restrict the adsorption capacities of carbon (Choi *et al.* 2008).

Ozonation is proposed as a process for the abatement of antibiotics (Andreozzi *et al.* 2005); but this process leads to the formation of oxidation intermediates that are mostly unknown at this point. In addition, unwanted oxidation byproducts, such as formaldehyde or bromate, form in the oxidation process. A BAC (biological activated carbon) unit following the ozone unit can remove the oxidation byproducts formed (Mara *et al.* 1999). Maybe the extension of conventional treatment with the ozone-biological activated carbon (O₃-BAC) technology can solve this problem.

Organic substances should be removed for they are the precursors of disinfection byproducts, which are either carcinogenic or potentially carcinogenic (Rehan & Manuel 2004). O₃-BAC technology was also proved to be an effective method to reduce organic substances (Karnik *et al.* 2005). As has been reported in our previous paper (Liao *et al.* 2012), BAC filtration in the down-flow mode is a common practice for the treatment of drinking water. However, in China, there have been several full-scale applications of BAC filtration in the up-flow mode within several cities (Zha *et al.* 2010). BAC

filtration in the up-flow mode is believed to avoid the excess growth of biomass and reduce hydraulic head loss. Moreover, due to the microdilancy of the BAC reactors, it could remove more organic matter than the down-flow system. The application of BAC filtration in the up-flow mode is now under consideration in many other regions in order to meet increasingly stringent drinking water quality criteria.

An investigation was conducted to help one city in East China determine the water treatment process for their new water source. A lake in this city, contaminated by human activity, has been used as a water source due to the increasing water supply demand and water resource shortage. The water quality concern about this lake arises due to the inner contamination from aquaculture and outer contamination from industry outflow, sewage and agriculture. Antibiotics and other organic substances are the most important contaminants in this source water. As far as the authors know, the ability of up-flow O₃-BAC technology to deal with antibiotics has rarely been reported. Hence, the aim of this paper was to: (1) develop a sensitive method for the determination of antibiotics species in the drinking water resource; (2) study the removal of antibiotics by O₃-BAC technology; (3) evaluate the ability of up-flow O₃-BAC technology to deal with water sources containing high levels of organic substances.

MATERIALS AND METHODS

Experimental system

A pilot plant with a flux of 1 m³/h was set up for this study, as shown in Figure 1. Water samples were collected from raw water and the effluent of each unit process, including pre- O₃, sedimentation, post- O₃ I, post- O₃ II, BAC and post sand filter.

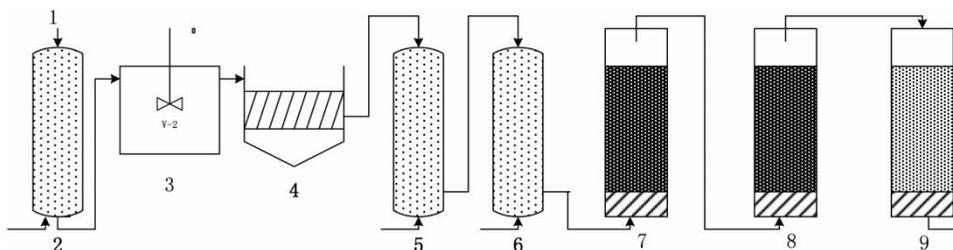


Figure 1 | Diagram of O₃-BAC technology. 1. Raw water; 2. Pre-O₃; 3. Coagulation; 4. Sedimentation; 5. Post O₃ I; 6. Post-O₃ II; 7. BAC I; 8. BAC II; 9. Sand filtration unit.

Dosages of pre- O₃ and post- O₃ used in this study were 0.5 and 1 mg/L, respectively, and 4 mg/L of aluminum coagulant (as Al₂O₃) was added. This pilot plant was operated continuously for about 1 year. The parameters of each unit process are shown in Table 1.

Sampling for antibiotics analysis

All the samples were collected and stored in pre-cleaned amber glass bottles, then filtered through a 0.45 µm membrane filter (Millipore, USA). Solid-phase extraction (SPE) with Oasis HLB cartridges (Waters, USA) was preconditioned with 6 ml of methanol and 6 ml of deionized water. Thereafter, the samples were passed through the SPE columns at a flow rate of approximately 5 ml/min. The HLB column was then dried under nitrogen gas for 1 h and then eluted with 15 ml of methanol and concentrated to 1 ml. The concentration solution was ready for the liquid chromatography-mass spectrometry (LC-MS) test.

Detection of antibiotics

An HP 1100 LC/MS instrument (Agilent Technologies, Palo Alto, CA, USA) was used for antibiotic detection in this investigation with a controlled gradient system. A chromatograph

column (2.1 × 50 mm, C18, Waters, USA) was operated at 35 °C. Optimum separation was achieved using gradient elution. The mobile phase consisted of A: acetonitrile; and B: water with 0.2% (v/v) formic acid. The injection volume was 20 µl, and the flow-rate was 0.4 ml/min. All the compounds were eluted out of the column within 20 min.

Other parameters

Turbidity was determined by a turbidimeter (2100P, HACH, USA). Ammonia was analyzed using the colorimetric phenate method, nitrate was measured by the UV spectrophotometric screening method, and nitrite was measured by the colorimetric method (APHA, AWWA & WEF 1995). The potassium permanganate method was used to determine chemical oxygen demand (COD_{Mn}) (Standards for Drinking Water Quality GB 5749/50-2006). UV₂₅₄ was measured with a UV-VIS spectrophotometer (Model T6, Puxi Corp, China). The dissolved organic carbon (DOC) was measured by the combustion method with a TOC analyzer (5000A, Shimadzu, Japan). Samples for both UV₂₅₄ and DOC were pre-filtered with a 0.45 µm membrane before measuring. A pH meter (PHB-4, LeiCi Corp., China) was used to measure the pH and temperature in the water.

Table 1 | Operating parameters of the pilot plant

Process	Parameters	Designed values
Pre-ozonation	Contact time	7.5 min
	Ozone dose	0.5–1 mg/L
Coagulation	Mixing system	Three stages
	Hydraulic retention time	18.66 min
	Coagulant dose	4 mg/L (calculated as Al ₂ O ₃)
Sedimentation	Hydraulic retention time	Inclined tube 7.97 min
Post-ozonation I	Contact time	7.5 min
	Ozone dose	0.5–1.5 mg/L
Post-ozonation II	Contact time	7.5 min
	Ozone dose	0.5–1.5 mg/L
GAC filter	Size	Φ400 mm × 2,210 mm × 2 column
	Media depth	1,000 mm × 2 column
	Filtration velocity	8 m/h
	Empty bed contact time	15 min
Sand filter	Media depth	1,150 mm
	Grain size	0.8 mm
	Filtration velocity	8 m/h

A fluorescence spectrophotometer (F-7000, Hitachi, Japan) was used to obtain the three-dimensional fluorescence excitation-emission matrix (EEM) spectra. A xenon lamp was the excitation source and the bandwidths on excitation and emission modes were both set at 5 nm. The wavelength coverage of the excitation spectrum was 200 to 400 nm, and that of the emission spectrum was 250 to 550 nm. Ultrapure water was used as a blank adjustment to remove the Rayman effect in the sample. The photomultiplier tube (PMT) voltage was set at 600 V, and the scanning rate was 1,200 nm/min. EEM data were quantified following the fluorescence regional integration (FRI) technique developed by *Chen et al. (2003)*.

RESULTS AND DISCUSSION

Raw water quality

A comprehensive sampling campaign was conducted from June 1 to June 16, 2011 when the pilot test plant

had been operated continuously for over 2 months. The basic water quality profile of raw water is given in *Table 2*.

From *Table 2* it can be seen that this lake presented a profile typical of a water source contaminated by human activity. This water source had low turbidity, which may be detrimental for coagulation, but fairly high concentrations of organic matter, which might originate from phytoplankton secretion and was of great concern for drinking water safety.

Three-dimensional fluorescence EEM spectroscopy provides a valuable insight into the characteristics of natural organic matter (NOM) in this lake, as illustrated in *Figure 2*.

Regions I, II, III, IV and V correspond to aromatic protein I, aromatic protein II (biochemical oxygen demand, BOD₅), fulvic acid, microorganism metabolites and humic acid, respectively. Since fluorescence intensity is proportional to concentration as well as quantum yield of the fluorophores, differences in the relative fluorescent intensities of the components may be a reflection of differences in concentrations and/or quantum efficiencies of the components. The three-dimensional fluorescence integrated intensity ($\Phi_{i,n}$) was used to calculate the content of pollutants (*Chen et al. 2003*).

Table 2 | Characteristics of source water

	Turbidity (NTU)	Ammonia (mg/L)	DOC (mg/L)	UV ₂₅₄ (cm ⁻¹)	COD _{Mn} (mg/L)	Temperature (°C)
Maximum	1.28	0.25	5.05	0.137	5.68	26.6
Minimum	1.17	0.23	4.68	0.120	4.08	24.2
Average	1.23	0.24	4.81	0.132	4.96	25.2

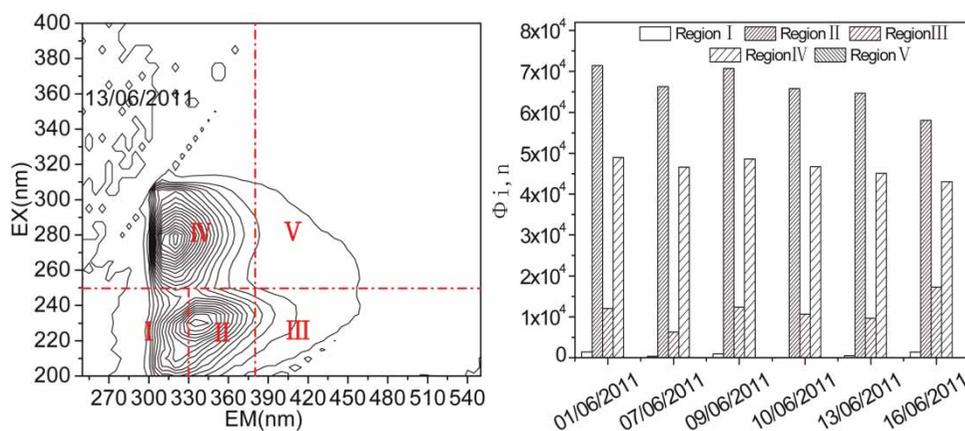


Figure 2 | EEM spectra and cumulative fluorescence response in the raw water samples.

Results in Figure 2 indicate that the dominant fluorophores were aromatic protein and soluble microbial products. According to their fluorescence intensity, the sequence of organic matter content is: aromatic protein II (BOD₅) > microorganism metabolites > fulvic acid > aromatic protein I > humic acid, which confirmed that the lake has been polluted by anthropogenic pollutants (Holbrook *et al.* 2005). The discharge of domestic wastewater from the vicinity and aquaculture waste in the lake is the cause.

Antibiotics in the water source

The detection of antibiotics by SPE-LC-electro-spray ionization (ESI)-MS was conducted to screen the antibiotics in the source water. The results are shown in Figure 3.

Various kinds of antibiotics were detected in this lake, including ciprofloxacin (CIP), sulfathiazole (ST), oxytetracycline (OTC), fluoxetine, diltiazem, ofloxacin (OFL), oxacillin, V penicillin, sulfamethazine (SM1), sulfamethizole (SM) and sulfamethoxazole (SMZ). The concentrations of fluoxetine, diltiazem and V penicillin were too low to quantify them.

In this study the main antibiotics were divided into three groups: sulfonamides, fluoroquinolones and tetracycline to make the interpretation more condensed.

Sulfonamides are frequently detected in the aquatic environment (Brown *et al.* 2006), they are broad spectrum antibiotics used against both Gram-positive and

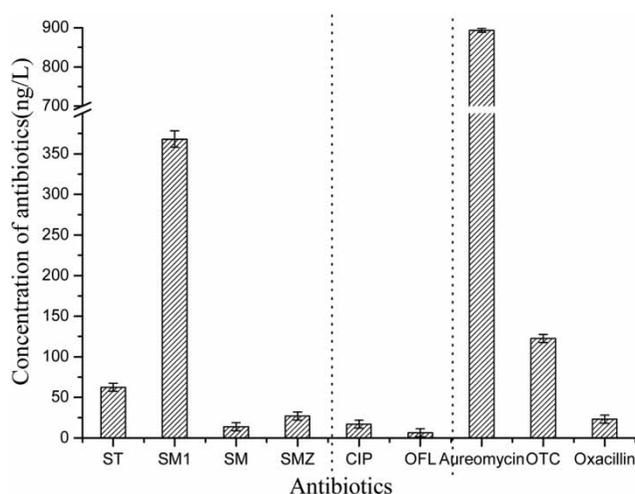


Figure 3 | Concentration of antibiotics in the water source.

Gram-negative aerobic bacteria. They are also effective against protozoa, and their combination with trimethoprim is largely used to treat respiratory diseases like pneumonia (Henney 1986). As shown in Figure 3, the detected sulfonamides in this study included SM1, ST, SM and SMZ. Their concentrations were 368.2, 62.5, 13.8 and 26.9 ng/L, respectively. Two fluoroquinolones CIP and OFL, were detected at concentrations of 16.9 and 6.5 ng/L, respectively. Fluoroquinolones are potent antibiotics with low minimal inhibitory concentrations against a wide range of bacterial species (Wiedemann & Grimm 1996). The tetracycline group contained aureomycin, OTC and oxacillin, the concentrations of which were 893.6, 122.6 and 23.2 ng/L, respectively. Aureomycin was the dominant antibiotic in raw water, followed by OTC. Tetracyclines are used extensively worldwide in human therapy and the farming industry and have been shown to disrupt microbial soil respiration, Fe(III) reduction, nitrification, and phosphatase activities (Gao *et al.* 2012). The concentrations of sulfonamides, fluoroquinolones and tetracyclines in raw water were 471.4, 23.4 and 1039.2 ng/L, respectively.

Water quality after treatment by conventional and O₃-GAC processes

Removal of bulk organic matter

COD_{Mn} is easy to test as an index for bulk organic matter concentration and is regulated in the Chinese *Standards for Drinking Water Quality* (GB 5749/50-2006). As shown in Figure 4 the COD_{Mn} concentration of raw water was 4.96 mg/L, and the mean overall removal rate of COD_{Mn} was just 14.1% by conventional water treatment (coagulation-sedimentation), while after the subsequent O₃-BAC unit the removal rate was 27.2%. Hence, O₃-BAC was necessary to remove more organic matter to meet the standard of 3 mg/L. Although ozonation contributed little to direct COD_{Mn} reduction, it helped enhance the biological performance of BAC and extended the life of the carbon media.

UV₂₅₄ is a surrogate index of the aromatic materials in water. The results in Figure 5 show that the value of raw water was only 0.104 cm⁻¹, and ozonation processes just result in a small reduction of UV₂₅₄. This may be due to

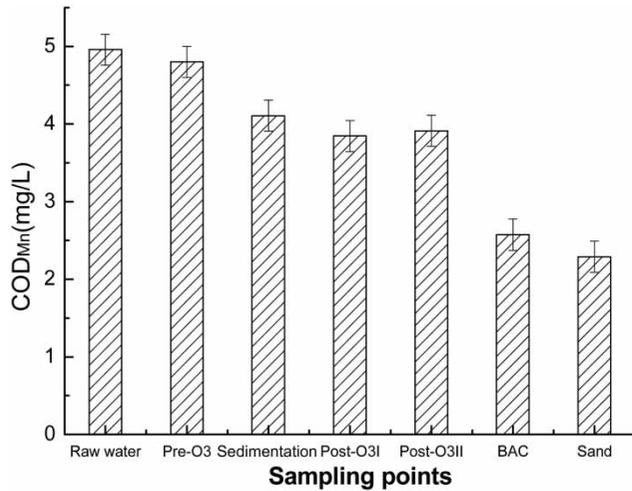


Figure 4 | COD_{Mn} concentration after each process.

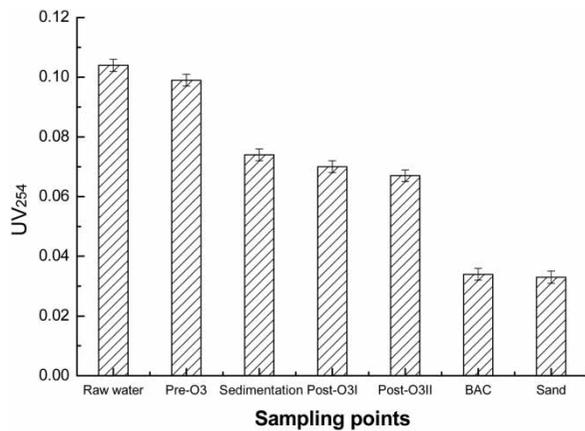


Figure 5 | UV₂₅₄ value after each process.

the water quality (high percentage of micro-molecular material) and the low ozone dosage. While UV₂₅₄ absorption was lowered by about 24% after the coagulation–sedimentation units and by more than 31.7% after BAC, the result is consistent with that of Xu (Xu *et al.* 2007a, b).

The DOC concentration in the effluent of each unit process followed the same trend as the COD_{Mn} and UV₂₅₄, as shown in Figure 6. The DOC of the raw water was 4.81 mg/L, and ozonation did not lead to any significant removal but a slight increase of DOC. The result is consistent with the findings of Xu (Xu *et al.* 2007a, b). Ozonation is known to decrease the molecular weight of the organic substances by breaking the macro-molecular materials into

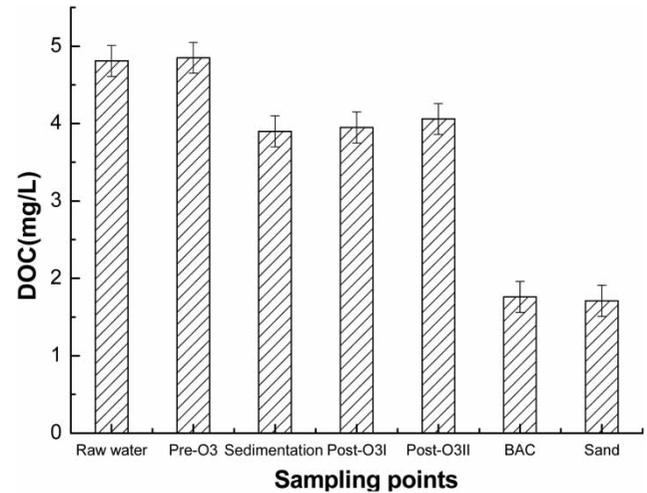


Figure 6 | DOC concentration of effluent from each process.

small pieces. DOC decreased from 4.85 to 3.9 mg/L in the conventional process and 47.8% of DOC removal was attributed to the O₃-BAC unit.

Removal of antibiotics

The change in antibiotics concentration also showed the necessity for advanced treatment since these emerging contaminants were removed only slightly by the conventional process but significantly by the O₃-BAC process. As can be seen in Figure 7, the conventional coagulation–sedimentation process only removed 0.25, 15.7 and –5% of

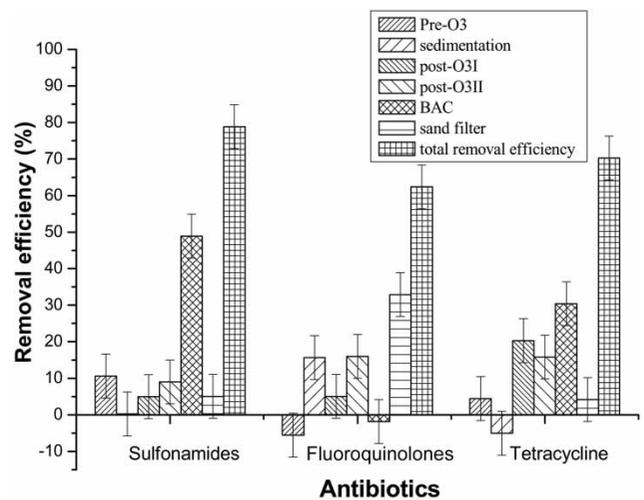


Figure 7 | Removal efficiency of three groups of antibiotics during O₃-BAC process.

sulfonamides, fluoroquinolones and tetracycline. However, the total removal efficiencies of sulfonamides, fluoroquinolones and tetracycline by the O₃-BAC process were 78.9, 62.4 and 70.2%, respectively.

Sulfonamides were recalcitrant in the conventional process and much more easily removed by the O₃-BAC process. The coagulation–sedimentation unit had little effect on their removal, and the sand filtration process also only removed 5.1% of them. An average removal efficiency of 10.6% was achieved pre-O₃ and that of two-stage post-O₃ was about 14% in total. The majority of sulfonamides removal occurred in the BAC filter with a significant reduction of nearly 50%.

Fluoroquinolones were more easily reduced by conventional treatment (15.7% for sedimentation and 32.9% for the sand filter) compared to sulfonamides. Similar to sulfonamides, fluoroquinolones could also be slightly removed by ozonation processes. However, the BAC unit was quite ineffective in fluoroquinolones removal, which indicated that fluoroquinolones are difficult to be adsorbed or biodegraded. Moreover, these new generation antibiotics are so powerful that they could probably inhibit the growth of bacteria in the filter.

The tetracyclines tested could be removed by adsorption processes (removal 30.4%), ozonation (40.6%) and conventional technology to a lesser extent (4.2% for sand filter). Tetracyclines have been shown to form relatively stable complexes with particulates and metal cations (Alexy *et al.* 2004), so they could not easily be removed by the coagulation–sedimentation unit. The appearance of a negative value of the removal efficiency may be attributed to a measuring error.

In brief, the conventional coagulation–sedimentation technology was not effective for antibiotics removal. Although there was a removal efficiency of 15.7% for fluoroquinolones, there was nearly no removal of sulfonamides and tetracycline. This is consistent with the result of Watkinson *et al.* (2007), who reported the limited removal of sulfonamides by conventional technology. The oxidation process only removed 14 and 24.6% of sulfonamides and fluoroquinolones, but it achieved a high removal (40.6%) of tetracycline. BAC was very effective for both sulfonamides and tetracycline removal, but quite ineffective in fluoroquinolones removal.

CONCLUSION

High concentrations of antibiotics were detected in one impacted source water. The gross concentrations of sulfonamides, fluoroquinolones and tetracycline in raw water were 471.4, 23.4 and 1,039.2 ng/L, respectively. The combined conventional and advanced processes were proven to be the feasible treatment solution for this water source. The conventional process was effective in removing fluoroquinolones, ozonation was effective for tetracycline, and the BAC process was very powerful in removing sulfonamides. The O₃-BAC process was also very efficient in removing COD_{Mn}, UV₂₅₄ and DOC, with efficiencies up to 45.1, 67.3 and 65.1%, respectively. The total removal efficiencies of sulfonamides, fluoroquinolones and tetracycline by the O₃-BAC process were 78.9, 62.4 and 70.2%, respectively.

REFERENCES

- Alexy, R., Kumpel, T. & Kunnerer, K. 2004 Assessment of degradation of 18 antibiotics in the closed bottle test. *Chemosphere* **57**, 505–512.
- Andreozzi, R., Canterino, M., Marotta, R. & Paxeus, N. 2005 Antibiotic removal from wastewaters: the ozonation of amoxicillin. *Journal of Hazardous Materials* **122**, 243–250.
- APHA, AWWA & WEF 1995 *Standard Methods for the Examination of Water and Wastewater*, 19th edition. American Public Health Association, Washington, DC, USA.
- Araceli, G. A., Romain, B., Vincent, S., Barbeau, B., Prévost, M. & Sauvé, S. 2010 Oxidation kinetics of cyclophosphamide and methotrexate by ozone in drinking water. *Chemosphere* **79**, 1056–1063.
- Brown, K. D., Kulis, J., Thomson, B., Chapman, T. H. & Mawhinney, D. B. 2006 Occurrence of antibiotics in hospital, residential, and dairy effluent, municipal wastewater, and the Rio Grande in New Mexico. *Science of the Total Environment* **366**, 772–783.
- Chen, W., Westerhoff, P., Leenheer, J. A. & Booksh, K. 2003 Fluorescence excitation-emission matrix regional integration to quantify spectra for dissolved organic matter. *Environmental Science Technology* **37**, 5701–5710.
- Choi, K. J., Kim, S. G. & Kim, S. H. 2008 Removal of antibiotics by coagulation and granular activated carbon filtration. *Journal of Hazardous Materials* **151**, 38–43.
- Chu, D. T. & Fernandes, P. B. 1989 Structure-activity relationships of the fluoroquinolones. *Antimicrobial Agents and Chemotherapy* **33**, 131–135.

- Gao, Y., Li, Y., Zhang, L., Huang, H., Hua, J. J., Shah, S. M. & Su, X. G. 2012 Adsorption and removal of tetracycline antibiotics from aqueous solution by graphene oxide. *Journal of Colloid and Interface Science* **368**, 540–546.
- Gilliver, M. A., Bennett, M., Begon, M., Hazel, S. M. & Hart, C. A. 1999 Antibiotic resistance found in wild rodents. *Nature* **401**, 233–234.
- Henney, C. R. (ed.) 1986 *A Handbook of Drugs*. 2nd edn, Churchill Livingstone, Edinburgh, UK.
- Holbrook, R. D., Breidenich, J. & DeRose, P. C. 2005 Impact of reclaimed water on select organic matter properties of a receiving stream - fluorescence and perylene sorption behavior. *Environmental Science Technology* **39** (17), 6453–6460.
- Holmstrom, K., Graslund, S., Wahlstrom, A., Pongshompoo, S., Bengtsson, B. E. & Kautsky, N. 2003 Antibiotic use in shrimp farming and implications for environmental impacts and human health. *Journal of Food Science and Technology* **38**, 255–266.
- Karanfil, T. J., Kilduff, J. E., Schlautman, M. A. & Weber, W. J. 1996 Adsorption of organic macromolecules by granular activated carbon. 1. Influence of molecular properties under anoxic solution conditions. *Environmental Science and Technology* **30**, 2187–2194.
- Karnik, B. S., Davies, S. H., Baumann, M. J. & Masten, S. J. 2005 The effects of combined ozonation and filtration on disinfection by-product formation. *Water Research* **39**, 2839–2850.
- Liao, X. B., Chen, C., Chang, C. H., Wang, Z., Zhang, X. J. & Xie, S. G. 2012 Heterogeneity of microbial community structures inside the up-flow Biological Activated Carbon (BAC) filters for the treatment of drinking water. *Biotechnology and Bioengineering* **117**, 881–886.
- Mara, A., Takako, A. W., Takayuki, M. K., Wataru, N. M., Akihisa, T. & Yasumoto, M. 1999 Bromate removal during transition from new granular activated carbon (GAC) to biological activated carbon (BAC). *Water Research* **33** (12), 2797–2804.
- Nakada, N., Shinohara, H., Murata, A., Kiri, K., Managaki, S., Sato, N. & Takada, H. 2007 Removal of selected pharmaceuticals and personal care products (PPCPs) and endocrine-disrupting chemicals (EDCs) during sand filtration and ozonation at a municipal sewage treatment plant. *Water Research* **41**, 4373–4382.
- Paul, E., Jacob, G., Furlong, E. T., Meyer, M. T., Zaugg, S. D. & Lippincott, L. R. 2007 Efficiency of conventional drinking-water-treatment processes in removal of pharmaceuticals and other organic compounds. *Science of the Total Environment* **377**, 255–272.
- Pomati, F., Castiglioni, S., Zuccato, E., Fanelli, R., Vigetti, D., Rossetti, C. & Calamari, D. 2006 Effects of a complex mixture of therapeutic drugs at environmental levels on human embryonic cells. *Environmental Science and Technology* **40**, 2442–2447.
- Rehan, S. & Manuel, J. R. 2004 Disinfection by-products (DBPs) in drinking water and predictive models for their occurrence: a review. *Science of Total Environment* **321**, 21–46.
- Stackelberg, P. E., Gibb, J., Furlong, E. T., Meyer, M. T., Zaugg, S. D. & Lippincott, R. L. 2007 Efficiency of conventional drinking water-treatment processes in removal of pharmaceuticals and other organic compounds. *Science of Total Environment* **377**, 255–272.
- Standards for Drinking Water Quality* GB 5749/50 2006, China.
- Watkinson, A. J., Murby, E. J. & Costanzo, S. D. 2007 Removal of antibiotics in conventional and advanced wastewater treatment: implications for environmental discharge and wastewater recycling. *Water Research* **41** (18), 4164–4176.
- Wiedemann, B. & Grimm, H. 1996 Susceptibility to antibiotics: species incidence and trends. In: *Antibiotics in Laboratory Medicine* (V. Lorian, ed.). Williams and Wilkins, Baltimore, MD, pp. 900–1168.
- Xu, B., Gao, N. Y., Sun, X. F., Xia, S. J., Simonnot, M. O., Causserand, C., Rui, M. & Wu, H. F. 2007a Characteristics of organic material in Huangpu River and treatability with the O₃-BAC process. *Separation Purification Technology* **57**, 348–355.
- Xu, W. H., Zhang, G., Zou, S. C., Li, X. D. & Liu, Y. C. 2007b Determination of selected antibiotics in the Victoria Harbour and the Pearl River, South China using high-performance liquid chromatography-electrospray ionization tandem mass spectrometry. *Environmental Pollution* **145**, 672–679.
- Zha, R. G., Xu, B., Zhu, H. T. & Zhang, P. 2010 Application of upflow biological activated carbon filter in water treatment plant. *Water & WasteWater Engineering* **36** (6), 14–17.

First received 30 November 2012; accepted in revised form 11 April 2013. Available online 13 September 2013