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REVIEW

Removal of Endocrine Disrupting Compounds in Drinking Water

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The article reviews the information regarding endocrine disrupting compounds, including the definition of endocrine disrupting compounds, the present pollution situation in China and the treatments. Various natural chemicals and some contaminants of industrial source were classified as endocrine disrupting compounds. For the endocrine disrupting compounds could adversely affect reproductive and developmental health, it was necessary and meaningful to discuss the applicable removal methods fand processes. In this review, a number of treatment processes were discussed in terms of their potential on removal of endocrine disrupting compounds. Finally, the future prospects together with the challenges for the monitoring and removing the endocrine disrupting compounds from water were summarized.

Keywords: Endocrine disrupting compounds, Endocrine disrupter, Water pollution, Water treatment, Removal.

INTRODUCTION

Various adverse health effects of endocrine disrupting compounds (EDCs) had been reported in recent years. Endocrine disrupting compounds also called environmental hormones, were exogenous endocrine disrupting compounds discharged into the environment by industrial production processes and could interfere with the endocrine systems of humans or animals abnormal and generate carcinogenic, teratogenic and mutagenic substances. They were widely distributed in the biosphere, by precipitation, surface runoff, the use of pesticide and water pollution, through the enrichment of drinking water and food chain leading serious harm to humans and organisms. Such compounds indirectly cause humans toxic and abnormal, but induce the estrogen-like effects on the organism directly. Even if the content is very low, endocrine disrupting compounds could threaten serious harm to human health and survival. In recent years, various kinds of endocrine disrupting compounds were also detected in the rivers and lakes in China with the rapid development of economy.

Human exposure to very low dose during critical periods, for example at the cellular differentiation period, could alter the development course of these tissues and this may result in permanent character changes in the mature living beings[1]. Recent toxicity studies indicated that bisphenol A exposure in adults may be associated with reduced ovarian response and *in vitro* fertilization success, reduced fertilization success and embryo quality, implantation failure, miscarriage, premature delivery, reduced male sexual function, reduced sperm quality, altered sex hormone concentrations, polycystic ovary syndrome, altered thyroid hormone concentrations, blunted immune function, cardiovascular disease (*i.e.* heart disease, hypertension and cholesterol levels), altered liver function, obesity, albuminuria, oxidative stress and inflammation and altered epigenetic markers and gene expression[2]. Waring and Harris[3] concluded that endocrine disrupting compounds not only affected reproductive function, but also affected a range of tissues which were steroid sensitive such as the central nervous system and thyroid. Results from the latest studies suggest that endocrine disrupting compounds may also affect the immune system, glucose homeostasis and could act as epigenetic modulators resulting in transgenerational effects. However, many types of endocrine disrupting compounds were detected in a wide range of natural and engineered environments across the world at present, including surface water, ground water supplies, wastewater effluents, sea water and sediment.

Therefore, a comprehensive understanding of the status of endocrine disrupting compounds pollution in China and the removal method of endocrine disrupting compounds was meaningful and necessary. Control and removal of endocrine disrupting compounds from water has a positive and important influence on human health. In particular, the latest results and trends were assessed regarding the adverse effects of endocrine disrupting compounds, the status of endocrine disrupting compounds pollution in China and removal processes. In summary, future research aspects on endocrine disrupting compounds were suggested.

Definition of endocrine disrupting compounds: Almost all the harmful effects of endocrine disrupting compounds were based on their impact on the reproductive system previously and these compounds were synthetic estrogen-like compounds with estrogenic effects. Therefore the concept of "environmental estrogen" was proposed. Later, some compounds could interact with hormone receptors and affect the activity of the endocrine system, which were also called as "endocrine disrupting compounds". With the passage of time, the understanding on endocrine disruptors was further enhanced. UK Environment agency deemed that environmental endocrine disruptors were natural or synthetic substances interfered with the function of the endocrine system and produced abnormal effects. Environment agency of Japan stressed that the endocrine disrupting compounds were the compounds which were absorbed into the organism, interfere with endocrine system and generate abnormal effect. The endocrine disruptor screening and testing committee of the U.S. Environmental Protection Agency (EPA) put forward the definition of endocrine disrupting compounds, which are known as a class of chemicals having xenobiotic and exogenous origins while mimicking or inhibiting the natural action of the endocrine system in animals and human, such as synthesis, secretion, transport and binding. They maintained the homeostasis, reproduction, metabolism, development and/or behaviour of living species[4]. Endocrine disrupting compounds were widely existed in various environmental media and had harmful influences on endocrine system and reproductive system, through various means, such as intake, the accumulation of biological and human endocrine system and so on. While the Organization of Economic and Cooperative Development (OECD) defined EDCa as "an exogenous substance or mixture that alters the function(s) of the endocrine systems and consequently causes adverse health effects in an intact organism, or its progeny or (sub) populations"[5]. The British Royal Society published the (Endocrine disrupting chemicals (EDCs)) in the 2000 and considered the endocrine disrupting compounds as the chemicals interfering normal function of human and animal endocrine systems. Dai[6] believed that endocrine disrupting compounds were the compounds that interfered with endocrine system and led to physical abnormalities effects on human or wildlife in the environment.

The compounds which could influence endocrine system generally had four major kinds as following: natural hormones (such as estradiol, estrone, testosterone, *etc*.), fungal and plant estrogen, synthetic estrogen, environmental chemical pollutants. Except natural estrogen, the other three were classified as endocrine disrupting compounds. About 70 species of chemicals had been recognized as endocrine disrupting compounds currently. Although not all chemicals had endocrine disrupting activity, there was an urgent need to screen and confirm the chemical with the activity of endocrine disruptors for about 10 million kinds of various chemicals in the world.

With the progress of the development of analytical instruments and analysis technology, more and more synthetic compounds were identified as endocrine disrupting compounds. Examples of various types of endocrine disrupting compounds classified are summarized in Table-1, including alkylphenol-ethoxylated, nonionic surfactant and its degradation products, food additives, flavoring agents, antioxidants, flame retardants, plasticizers agents, industrial solvents, disinfectants, fecal sterols, polycyclic aromatic hydrocarbons, pesticides and herbicides, *etc*. with increasingly stringent environmental standards, more and more compounds would likely be included in the endocrine disrupting compounds.

Pollution situation of endocrine disrupting compounds in China: The main way of endocrine disrupting compounds intake into human body was the direct ingestion of food and water. So it was seriously significant to control the quality and conditions of raw water, water from water supply plant

TABLE-1
LIST OF SOME CHEMICALS CLASSIFIED AS EDCS IN THE ENVIRONEMNT [5,6]

Classification		Name of the substance
Pesticide	Herbicide	Alachlor, aminotriazole, atrazine, metribuzin, nitrofen, trifluralin, 2,4-D, 2,4,5-T, molinate, *etc.*
	Fungicide	Benomyl, carbendazim, hexachlorobenzene, ethylene thiourea, pyrimidine methanol, ethylidene double dithiocarbamate zinc, dimethyl dithiocarbamate zinc, *etc.*
	Insecticide	Lindane(β-666), chlordane, endosulfan, triazine, carbaryl, dicofol, dieldrin, DDT and its metabolite, heptachlor, H-epoxide, methomyl, methoxychlor, dechlorane, parathion, oxygen chlordane, trifluralin, synthetic pyrethroids, *etc.*
	Nematicide	Temix, carbofuran, dibromochloropropane (DBCP), *etc.*
Industrial compounds	Heavy metal	Lead, cadmium, mercury, *etc.*
	Raw materials for resin	Alkylphenol, nonyl phenol, octyl phenol, styrene, bisphenol-A, polyvinyl chloride, dibutyl phthalate, adipate, double (2-ethyl hexyl) phthalate, *etc.*
	Medicine	Tamoxifen, diethylstilbestrol, bromonaphthol, nonyl phenyl ether-9-nonyl phenol, diethylstilbestrol, 17'-ethinylestradiol, *etc.*
	Dielectric oil	Polychlorinated biphenyl, aroclor-1254, pyranol, 2,2',4,4'-tetrabrominated diphenyl ether, 2,5-dichloro-4, hydroxybiphenyl, *etc.*
	Surfactant	Nonyl phenol, tergitol NP40, tergitol, *etc.*
	Others	Dioxin [TCDD(Tetrachlorodibenzo-*p*-dioxin)], pentachlorophenol, tributyltin, dichlorophenyl, polybrominated biphenyls, phthalate, Spray shanmugaratnam sarin, Two hexyl phthalates, Hydroxyl fluoride butyl nitrate amide, daidzen and genistein, matairesol, enterodiol, enterolactone, *etc.*
	Phytoestrogen	Coumestrol, formononetin, *etc.*

and water in the pipe network. Meanwhile it was also important to take appropriate corresponding measures to ensure the quality of drinking water[7,8]. Urban drinking water was vulnerable to high intensity of human activity and pollution caused by endocrine disrupting compounds. Research had shown that even in the "acceptable" concentrations, the endocrine disrupting compounds could still have harmful and noxious impact on the ecosystem[9].

Over 100 endocrine disrupting compounds were reported at least once in waters and sediments at ng/L and ng/g level, respectively. The results of the screening level risk assessment (SLERA) also revealed that the hotspots for endocrine disrupting compounds pollution at present were the Haihe river watershed, the Pearl River watershed and the Yangtze river estuary, especially those rivers affected by the mega cities of Beijing, Tian-jin, Guangzhou and Shanghai[10]. The surface water pollution by endocrine disrupting compounds may attribute to the huge volume of wastewater discharge and limited wastewater treatment rate currently in China. For the protection of surface water, there were particularly urgent needs to improve the wastewater treatment rate to reduce the direct discharge. In some cases, endocrine disrupting compounds could be detected in source water of drinking water treatment plants or even tap water. It was significantly urgent to protect the water sources from endocrine disrupting compounds contamination[11]. Meanwhile endocrine disrupting compounds were detected in sediments of Shaying river, the largest tributary of the Huaihe river in eastern China, most of them with concentrations higher than the sediment quality guidelines, thus indicating potential biological risks to aquatic organisms[12]. Table-2 shows the endocrine disrupting compounds pollution status of the typical drinking water sources in China.

Applicable treatment methods to remove endocrine disrupting compounds from water

Conventional treatment process: The conventional processes for current drinking water treatment in China were as follows: flocculation-coagulation, sedimentation, filtration, disinfection[13,14]. Testing researches and production practices showed that conventional treatment processes for removal of organic matter was only 20-30 %[15,16]. Researches showed that the removal efficiency of long-chain alkyl phenol ethylene oxide ether (NPEO) by conventional treatment process was more than 90 %, but the removal efficiency of nonyl phenol (NP) was only about 60 % and achieved primarily by flocculation-coagulation process and oxidation process[17,18].

Zhang *et al.*[19] studied the enhanced coagulation method to remove phthalate esters (PAEs).The results show the dimethyl phthalate (DMP) removal rate is 99.87 % when initial dimethyl phthalate concentration is 0.50 mg/L with 50 mg/L PDMDAAC and 2.5 mg/L CPAM at pH 6. Therefore, the removal of phthalate esters by coagulation is effective and the application of coagulation by cationic polyacrylamide in water supply will enhance the removal of phthalate esters and also reduce the water turbidity. Meanwhile a method to remove dioctyl phthalate (DOP) form water was realized through a flocculation process with a novel polyacrylamide flocculant P(AM/AA/AMPS). The performance of flocculant was evaluated by their ability to remove dioctyl phthalate from water and a maximum dioctyl phthalate removal efficiency of 82.1 % was obtained under optimized conditions[20].

The use of Cl_2/ClO_2 is still the most widespread conventional treatment for disinfecting drinking waters. ClO_2 can be used to treat wastewater effluents to oxidize various endocrine disrupting compounds[21]. Macrolide and sulfonamide antibiotics as well as estrogens and phenazones were readily oxidized by ClO_2. Ozone seemed to be considerably more efficient for pharmaceutical control than ClO_2 because it exhibited higher rate constants and reacted with a larger number of pharmaceuticals. However, ClO_2 appeared slightly more powerful than chlorine for the oxidation of pharmaceuticals[22].

In short, conventional treatment systems are unable to completely remove a large amount of the organic micropollutants present in urban wastewaters. More effective and specific treatments are required to reduce the environmental and potential impact of effluents and comply with increasingly strict legislation[23].

Adsorption technology: Adsorption, using granular activated carbon (GAC), generally removes most organic contaminants, including endocrine disrupting compounds, such as DDT, endosulfan, (polychlorinated biphenyl) PCBs, dioxin, NPEOs and other alkylphenols in some related literature. Great attention has been focused on the selective adsorption of individual endocrine disrupting compounds using various adsorbents. The adsorption performance of the endocrine disrupting compounds by GAC was influenced by natural organic matter in water (NOM), pH and temperature. As the raw water pH was around neutral, it was meaningful to study the competitive adsorption between natural organic matter (NOM) and endocrine disrupting compounds. Lerman, *et al.*[24] pointed out that when single-walled carbon nanotubes (SWCNTs) were used as sorbent, carbamazepine was the primary adsorbate

TABLE-2
EDCS POLLUTION STATUS OF THE TYPICAL DRINKING WATER SOURCES IN CHINA

Contaminated area	EDCs pollution status
Surface water of Beijing[13]	20 Kinds of EDCs were detectable. benzene hexachloride and its isomers (exceeding China Standard), DDT and its metabolites (≥ 33·42 μg/L). Phthalate esters (detection rate ≥ 80 %)
Yangtze river in Nanjing and its downstream river[14]	PCBs in the sediment of nanjing (0.14-4 ng/g), its downstream river (10-26 ng/g)
Jialing river, Yangtze river in Chongqing[13]	Nonylphenol in April (Maximum 1.12 μg/L), Alkylphenol polyethylene oxide ether (Maximum 3.5-100 μg/L)
Huangpu river[15]	DDTs (0.1-0.12 μg/L)
Songhua river[16]	Steroid estrogens (4-44 ng/L); Four of phthalate acid esters (PAEs), bisphenol A (BPA) and 3 alkylphenols (APs) (2-163 760 ng/L)
Haihe river[17]	Polychlorinated biphenyl (0.06-3.11 μg/L), organo-chlorine pesticide (0.05-1.07 μg/L)

and bisphenol A and phenanthrene were used as competitors. Strong competition with bisphenol A and no effect of phenanthrene on adsorption of carbamazepine was obtained. They also suggested that the chemical nature of DOM could significantly affect the sorptive behaviour of polar organic pollutants with carbon nanotubes when all are introduced to the aquatic system.

Although activated carbon had a preferable removal ability of endocrine disrupting compounds, pre-dosing ozone activated would oxidize endocrine disrupting compounds into small organic molecules and enhance removal efficiency obtained by GAC adsorption. Micro-organisms in the GAC were also more conducive to degrade the organic matter absorbed by activated carbon adsorption and further prolong the service life of activated carbon greatly. So the ozone-biological activated carbon process was feasible for the removal of endocrine disrupting compounds. Addition of 5 mg/L of powder activated carbon with a 4 h contact time removed 50-98 % of endocrine disrupting compounds compounds. Separate chlorine or ozone experiments decreased the endocrine disrupting compounds initial concentration by < 10 to > 90 %. Conventional treatment (coagulation plus chlorination) in the water supply plant had low removal efficiency of many endocrine disrupting compounds, while addition of powder activated carbon and ozone could substantially improve the removal efficiency[25]. Biochar is the by-product of the pyrolytic processing of biomass to obtain biofuel such as controlled thermal process and gasification and has a potential as a promising adsorbent for the elimination of micro-pollutants. Jung *et al.*[26] had reported that chemically activated biochar produced under oxygenated (O-biochar) and oxygen-free (N-biochar) conditions were employed to investigate the adsorption of endocrine disrupting compounds *e.g.*, bisphenol A (BPA), atrazine (ATR), 17 α-ethinylestradiol (EE2) and pharmaceutical active compounds (PhACs); sulfamethoxazole (SMX), carbamazepine (CBM), diclofenac (DCF), ibuprofen (IBP), suggesting that O-biochar and N-biochar were the promising sorbents for agricultural and environmental applications. Compared with the conventional adsorbents, metal-organic frameworks presented fascinating merits due to their diverse compositions and structures, such as greater porosity, higher surface area and pore volume, tunable pore size and modifiable surface. Metal-organic framework MIL-53(Al){Al(OH)-[O_2C-C_6H_4-CO_2]} and MIL-53(Al) - F127{Al(OH)[O_2C-C\H_4-CO_2]} were synthesized and used as sorbents to remove bisphenol A (BPA) from aqueous system. The results demonstrated that the equilibrium sorption amounts of BPA on MIL-53(Al) and MIL-53(Al)-F127 reached 329.2 ± 16.5 and 472.7 ± 23.6 mg/g, respectively, far more than that of commercial activated carbons (ranging from 129.6 to 263.1 mg/g)[27].

Advanced oxidation processes (AOPs): There were a group of chemical-oxidative processes called advanced oxidation processes, characterized by the generation of hydroxyl radicals. One of the most commonly used method for treatment of toxic and hazardous substances in the environment was advanced oxidation process, especially photocatalytic degradation. Considerable research interest has recently been shown in the application of O_3, UV/H_2O_2 and UV/ TiO_2 systems for the removal of endocrine disrupting compounds from water. The hydroxyl radical is highly reactive and non-selective against refractory organic pollutants with subsequent conversion of the pollutants to CO_2, H_2O and mineral acids. In the last couple of years, advanced oxidation processes have been extensively exploited and reported in the literature for the degradation of different wastewater containing contaminants such as pesticides, pharmaceuticals, endocrine disruptors and dye molecules[28]. Table-3 lists some key studies, with a summary of the experimental conditions and results.

Biological treatment process: Now many researchers dedicated to study biological treatment process for the removal of endocrine disrupting compounds, numerous studies had demonstrated that the majority of endocrine disrupting compounds was biodegradable. Conventional biological processes, such as activated sludge, biofiltration and soil aquifer treatment, had shown some degree of endocrine disrupting compounds removals, which were mostly derived from biodegradable and/or other compounds readily attached to particles[4]. Some research studies showed the metabolic characteristics of nitrifiers to achieve the removal of endocrine disrupting compounds[36]. Jin *et al.*[37] detected the concentrations of several estrogens and their estro-genicities in the influent and effluent of the municipal sewage treatment plant (STP) with conventional primary and secondary treatments located in Wuhan, China. The results demonstrated that estrogenic compounds could not be entirely removed by the existing sewage treatment process, which should be further improved to protect aquatic ecosystems and human health. Zhou *et al.*[38] investigated the change of concentrations of eight endocrine disrupting compounds in wastewater along the treatment processes of three STPs in Beijing. The STPs could not remove alkylphenols effectively from the aqueous phase with less than 40 % reduction. Bisphenol A decreased over 90 % and steroid estrogens achieved considerable reductions from 64.8 % of 17β-estradiol to 94.9 % of estriol. Generally, biological treatment was more effective in removing alkylphenols, bisphenol A and natural estrogens from the aqueous phase than primary treatment. However, the synthetic estrogen, 17α-ethinylestradiol, was mostly removed by the primary treatment with about 63.5 % reduction. The compound might have a bearing with the waste effluents of dairy farms around urban area of Beijing. However, these studies only focused on the wastewater side, while the activated sludge side was largely overlooked. The reason probably lied in that the serious matrix interference made it difficult to accurately quantify the amount of endocrine disrupting compounds present in the activated sludge. Auriol *et al.*[39] pointed out that though the biodegradation by and/or the adsorption to activated sludge were generally presumed to be the main mechanism accounting for the removal of endocrine disrupting compounds in the STPs, more information about endocrine disrupting compounds in the activated sludge was required. Therefore, the concentrations of endocrine disrupting compounds in both wastewater and activated sludge should be simultaneously determined to comprehensively understand their fate and behaviour in the STPs. Nie *et al.*[40] investigated the fate and seasonal variation of several typical endocrine-disrupting compounds (EDCs), including estrone (E1), 17β-estradiol (E2), 17α-ethinylestradiol (EE2), estriol (E3), bisphenol A (BPA) and 4-nonylphenol (NP)

TABLE-3
DEGRADATION OF VARIOUS EDCs BY AOPs

Compound	Treatment	Operating conditions	Results and comments	References
Antibiotic (metronidazol)	Fenton	C_0 = 6 µmol /L	Fenton was less efficient than photo-Fenton	Shemer *et al.*[29]
Benzafibrate (lipid regulator)	Ozonization	$C(O_3)$ = 0.73 mmol /L; C_0 = 0.5 mmol /L; pH 6 to 8	The complete Bezafibrate abatement is achieved.	Dantas *et al.*[30]
Bisphenol A	UV UV/H_2O_2	Low pressure lamp (15 W, 254 nm); C_0 = 60 µmol /L; $C_{H_2O_2}$ = 0 to 50 mg/L; pH 5.3-4.3	UV was not enough to degrade bisphenol A. Experiments with UV/H_2O_2 give a better removal of estrogenic activity	Chen *et al.*[31]
Seven EDCs [E1, E2, E3, EE2, octylphenol (OP), NP, and BPA]	TiO_2/ UV	$C(TiO_2)$ = 0.8 g/L, UV(0.7 W, 365 nm.) EDCs = 100 ng/L	The estrogenic activity was almost totally removed by TiO_2 photocatalysis in 50 min	Zhang *et al.*[32]
32 Selected micropollutants (pharmaceuticals, corrosion inhibitors and biocides/pesticides)	UV254/ H_2O_2/ Fe^{2+}	50 mg/L of H_2O_2, 5 mg/L of Fe^{2+}	Global percentages of micropollutants removal achieved were 98 and a 97 %, respectively, after 0.5 h of treatments	Cruz *et al.*[33]
40 Selected pharmaceuticals	UV/H_2O_2	C_0 = 0.79-4.91 µg/L UV (200-300 nm)	Most of the pharmaceu-ticals are well removed when applying both UV and H_2O_2	Wols *et al.*[34]
Phenol and chlorophenol	UV/H_2O_2/TiO_2	pH 4, 0.5 g/L TiO_2, 90 min irradiation time, 75 ml/L H_2O_2. 50-200 mg/L phenol and *p*-chlorophenol	74.6 and 79.8 % phenol and chlorophenol were destroyed. UV/H_2O_2/TiO_2 combined system was more efficient than UV/H_2O_2 and UV/TiO_2	Dixit *et al.*[35]

in a municipal sewage treatment plant (STP) employing an anaerobic/anoxic/oxic (A/A/O) process. Results indicated that in summer, the examined STP could effectively remove 75.4 % of E1 and more than 90 % of all other studied endocrine disrupting compounds from the wastewater. The biodegradation in the A/A/O process was the primary removal mechanism.

Conclusions and future prospects: Endocrine disrupting compounds were of a general concern and were significant research subject. The epidemiological data gave evidence of a possible relationship between chemical exposure and harmful observed effects of endocrine disruption in the living beings. The rapid development of sensitive analytical techniques in recent years was enabling their analysis at trace levels and investigation of their fate and transformation pathways, which were still largely unknown. Information about the toxic effects of these compounds and their degradation byproducts on living organisms was also very limited. So a lot of comprehensive work needed to be carried out. A comprehensive understanding of registration usage of all kinds of endocrine disrupting compounds and the establishment of an overall database of endocrine disrupting compounds were urgently necessary. For endocrine disrupting compounds pollution in the water environment at present, it was necessary to develop some more efficient, economical, safe and practical removal techniques and facilities. The removal efficiency of endocrine disrupting compounds in various water treatment processes should be well studied. Effective measures should be taken to control the pollution of endocrine disrupting compounds. In addition, the establishment of large-scale monitoring networks was designed to enable a better understanding of fate and transport mechanisms in the environment, including soil, water and air, was necessary.

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REFERENCES

1. M. Auriol, Y. Filali-Meknassi, R.D. Tyagi, C.D. Adams and R.Y. Surampalli, *Process Biochem*., **41**, 525 (2006).
2. J.R. Rochester, *Reprod. Toxicol*., **42**, 132 (2013).
3. R.H. Waring and R.M. Harris, *Maturitas*, **68**, 111 (2011).
4. H.S. Chang, K.H. Choo, B. Lee and S.J. Choi, *J. Hazard. Mater*., **172**, 1 (2009).
5. S. Esplugas, D.M. Bila, L.G.T. Krause and M. Dezotti, *J. Hazard. Mater.*, **149**, 631 (2007).
6. S.G. Dai, Advanced in Environmental Chemistry, Chemical Industry Press, Beijing, p13/478-13/495 (2005).
7. J. Debska, A. Kot-Wasik and J. Namiesnik, *Crit. Rev. Anal. Chem*., **34**, 51 (2004).
8. A. Nikolaou, S. Meric and D. Fatta, *Anal. Bioanal. Chem*., **387**, 1225 (2007).
9. M.J. Benotti, R.A. Trenholm, B.J. Vanderford, J.C. Holady, B.D. Stanford and S.A. Snyder, *Environ. Sci. Technol*., **43**, 597 (2009).
10. Q. Bu, B. Wang, J. Huang, S. Deng and G. Yu, *J. Hazard. Mater*., **262**, 189 (2013).
11. J.L. Liu and M.H. Wong, *Environ. Int*., **59**, 208 (2013).
12. Y.Z. Zhang, C.Y. Tang, X.F. Song, Y. Dun, W. Meng and Y. Zhang, *Environ. Earth Sci*., **70**, 2237 (2013).
13. L. Lang and G. Zhang, *Environ. Eng*., **26**, 60 (2008).
14. D. Fang, Z. Mei and X. Lou, *Environ. Monit. China*, **17**, 2 (2001).
15. C. Chen, Z. Chen and C. Bi, *J. Agro-Environ. Sci*., **2**, 705 (2008).
16. X. Shao, J. Ma and G. Wen, *Environ. Sci*., **10**, 2723 (2008).
17. T. Wang, Z. Zhang and J. Huang, *Environ. Sci*., **4**, 730 (2007).
18. A. Soares, B. Guieysse, B. Jefferson, E. Cartmell and J.N. Lester, *Environ. Int*., **34**, 1033 (2008).
19. P. Zhang, H.L. Zheng, X.L. Deng, S.J. Jiang, J.J. Wang, L. Liu and B. Yang, *J. Civil Architect. Environ. Eng*., **4**, 157 (2011).

20. H. Zheng, J. Ma, C. Zhu, Z. Zhang, L. Liu, Y. Sun and X. Tang, *Separ. Purif. Tech*., **123**, 35 (2014).
21. G. Hey, R. Grabic, A. Ledin, J. la Cour Jansen and H.R. Andersen, *Chem. Eng. J*., **185**, 236 (2012).
22. M.M. Huber, S. Korhonen, T.A. Ternes and U.V. Von Gunten, *Water Res*., **39**, 3607 (2005).
23. J. Rivera-Utrilla, M. Sánchez-Polo, M.Á. Ferro-García, G. Prados-Joya and R. Ocampo-Pérez, *Chemosphere*, **93**, 1268 (2013).
24. I. Lerman, Y. Chen, B. Xing and B. Chefetz, *Environ. Pollut*., **182**, 169 (2013).
25. P. Westerhoff, Y. Yoon, S. Snyder and E. Wert, *Environ. Sci. Technol*., **39**, 6649 (2005).
26. C. Jung, J. Park, K.H. Lim, S. Park, J. Heo, N. Her, J. Oh, S. Yun and Y. Yoon, *J. Hazard. Mater*., **263**, 702 (2013).
27. M. Zhou, Y. Wu, J. Qiao, J. Zhang, A. McDonald, G. Li and F. Li, *J. Colloid Interf. Sci*., **405**, 157 (2013).
28. J.O. Tijani, O.O. Fatoba and L.F. Petrik, *Water Air Soil Pollut*., **224**, 1770 (2013).
29. H. Shemer, Y.K. Kunukcu and K.G. Linden, *Chemosphere*, **63**, 269 (2006).
30. R.F. Dantas, M. Canterino, R. Marotta, C. Sans, S. Esplugas and R. Andreozzi, *Water Res*., **41**, 2525 (2007).
31. P. Chen, K.G. Linden, D.E. Hinton, S. Kashiwada, E.J. Rosenfeldt and S.W. Kullman, *Chemosphere*, **65**, 1094 (2006).
32. W. Zhang, Y. Li, Y. Su, K. Mao and Q. Wang, *J. Hazard. Mater.*, **215-216**, 252 (2012).
33. N. De la Cruz, J. Giménez, S. Esplugas, D. Grandjean, L.F. de Alencastro and C. Pulgarín, *Water Res*., **46**, 1947 (2012).
34. B.A. Wols, C.H.M. Hofman-Caris, D.J.H. Harmsen and E.F. Beerendonk, *Water Res*., **47**, 5876 (2013).
35. A. Dixit, A.K. Mungray and M. Chakraborty, *Int. J. Chem. Eng. Appl*., **1**, 247 (2010).
36. I. Forrez, M. Carballa, H. Noppe, H. De Brabander, N. Boon and W. Verstraete, *Water Res*., **43**, 77 (2009).
37. S. Jin, F. Yang, T. Liao, Y. Hui and Y. Xu, *Environ. Toxicol. Chem*., **27**, 146 (2008).
38. H.D. Zhou, X. Huang, X.L. Wang, X.H. Zhi, C.D. Yang, X.H. Wen, Q.H. Wang, H. Tsuno and H. Tanaka, *Environ. Monit. Assess*., **161**, 107 (2010).
39. M. Auriol, Y. Filali-Meknassi, R.D. Tyagi, C.D. Adams and R.Y. Surampalli, *Process Biochem*., **41**, 525 (2006).
40. Y. Nie, Z. Qiang, H. Zhang and W. Ben, *Sep. Purif. Technol*., **84**, 9 (2012).