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Fate of Environmental Pollutants

Lokesh P. Padhye^{1*}, Ulas Tezel²

ABSTRACT: A review of the literature published in 2012 on topics relating to the fate of emerging environmental pollutants during wastewater treatment is presented. This review is divided into the following sections: biological agents, estrogens, industrial wastewater, metals, nanomaterials, persistent organic pollutants, pharmaceuticals and personal care products, and trace organic contaminants.

KEYWORDS: emerging contaminants, pharmaceuticals and personal care products, nano materials, industrial wastewater, estrogens, pathogens.

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Persistent organic pollutants

Perfluorinated Compounds (PFC). A PFC is an organofluorine compound with all hydrogens replaced by fluorine on a carbon chain but the molecule also contains at least one different atom or functional group.

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PFCs have unique properties to make materials stain, oil, and water resistant, and are widely used in diverse applications. PFCs are persistent organic pollutants, but unlike polychlorinated biphenyls (PCBs), they are not known to be degraded by any natural processes due to the strength of the carbon–fluorine bond, as a result PFCs may accumulate in the environment and pose environmental and human health risks. The most commonly used PFCs are perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA), which have been used in many kinds of products. They have been found in wastewater, surface and drinking water in both developed and developing countries around the world including North America, Europe and Asia.

In a study, occurrence of PFOS and PFOA in rivers of 15 countries around the world during 2004 to 2010 was investigated. In total, 539 samples were collected from the rivers in 41 cities. The average concentration of PFOS in each city was up to 70.1 ng/L. The average concentration of PFOA in each city was in the range 0.2-1,630 ng/L. The industrialized areas showed higher contamination in both PFOS and PFOA concentrations than non-industrialized areas suggesting that industrial activities are some of the major sources of PFCs contamination in rivers (Kunacheva et al., 2012).

In another study, the concentrations of eighteen PFCs (C5-C14 carboxylates, C4, C6-C8 and C10 sulfonates and 3 sulfonamides) were determined in wastewater and sludge samples originating from two different water resource recovery facilities (WRRFs). According to the results, perfluoropentanoic acid (PFPeA), PFOA and PFOS were dominant in wastewater and sludge samples from both WRRF. The average concentrations in the raw and treated wastewater ranged up to 75.7 ng/L (perfluorotridecanoic acid, PFTrDA) and 76.0 ng/L (PFPeA), respectively. Concentrations of most PFCs were higher in effluents than in influents, indicating their formation during wastewater treatment processes. In sewage sludge, the average concentrations ranged up to 6.7 ng/g-dry weight (PFOS). No significant seasonal variations in PFCs concentrations were observed, while higher concentrations of PFOA, PFOS and perfluorononanoic acid (PFNA) were determined in the WRRF receiving municipal and industrial wastewater (Arvaniti et al., 2012).

Chen, Zhang et al. (2012) determined the concentrations of PFOS and PFOA in influents, effluents, and sludges of twelve WRRFs in China. The highest concentrations of PFOS and PFOA in influents were found to occur in municipal and industrial WRRFs, respectively. They also indicated that PFC concentrations in the effluents are relatively higher than the influents.

Concentrations and isomer profiles for 24 per- and polyfluoroalkyl substances (PFASs) were monitored over 5 months in municipal landfill leachate in another study. Perfluoropentanoate and perfluorohexanoate were typically the dominant PFASs in leachate. Overall,

degradation of PFAS-precursors and changes in leachate pH, conductivity, and 24-h precipitation were important factors controlling PFAS occurrence in leachate (Benskin et al., 2012).

Kim, Im et al. (2012) stated that PFCs were found in higher concentrations in influent, effluent and sludge of industrial WRRFs than that of domestic WRRFs. Individual PFCs were found to have differing industrial sources, with perfluorocarboxylates used in fabric/textiles, paper-mill, and dyeing industries, and perfluoroalkylsulfonates occurring in oil/chemical and metal-plating/processing industries. Among those PFCs, 63% of PFOA was from domestic wastewater, and 75% of PFOS was from industrial wastewater. Authors concluded that main source of PFCs in natural water resources was WRRFs.

Yan et al. (2012) measured PFAs in sewage sludge samples collected from 25 WRRFs in Shanghai, China to evaluate the levels and profile of C3-C14 PFAs. The results showed a ubiquitous PFAs contamination of sewage sludge in Shanghai with the total PFAs range of 126-809 ng/g-dry weight. PFOA was the major PFA found at concentration ranged from 23.2 to 298 ng/g-dry weight, much higher than the levels in other countries.

PFCs are resistant to most conventional treatment processes. For instance, although TiO_2 -based photocatalysis is strong enough to decompose most organics, it is not effective for PFOA decomposition. It was found that indium oxide (In_2O_3) is very effective on PFOA decomposition under UV irradiation compared to TiO_2 . The major intermediates of PFOA decomposition by In_2O_3

were C2-C7 shorter-chain perfluorocarboxylic acids. PFOA decomposition in wastewater was inhibited by bicarbonate and other organic matters; however, their adverse impacts can be mostly avoided via pH adjustment and ozone addition (Li, Zhang et al., 2012).

In another study electrochemical decomposition of PFOA in aqueous solution was investigated over Ti/SnO₂-Sb, Ti/SnO₂-Sb/PbO₂, and Ti/SnO₂-Sb/MnO₂ anodes. Ti/SnO₂-Sb/PbO₂ anode's PFOA decomposition performance was better than the others tested. The results indicated that under optimal condition, the degradation rate constant and the degradation half-life were 0.064 per min and 10.8 min, respectively. The intermediate products including short-chain perfluorinated carboxylic acids and perfluorocarbons were detected. These studies suggest that electrochemical technique could be employed to degrade PFOA from contaminated wastewater as well as to reduce the toxicity of PFOA (Lin et al., 2012).

Polybrominated Diphenylethers (PBDEs).

Polybrominated diphenylethers or PBDEs, are organobromine compounds that are used as flame retardant. Like other brominated flame retardants, PBDEs have been used in a wide array of products, including building materials, electronics, furnishings, motor vehicles, airplanes, plastics, polyurethane foams and textiles. Owing to their ubiquitous distribution in the environment, toxicity to humans, and increasing accumulation in the environment, the fate of PBDEs is of serious concern for public health.

Gentes et al. (2012) detected high levels of PBDEs in ring-billed gulls breeding in a highly

industrialized section of a river. This study showed that PBDE contamination in the environment can reach to a level that it may harm human health through biomagnification.

Occurrence of alkylphenols (APs) and PBDEs in raw wastewater during dry and wet-weather periods and their removal by physico-chemical lamellar settling and biofiltration techniques were investigated in a study. Raw effluents exhibited 1.5 to 5 times higher AP and PBDE concentrations during wet periods compared with dry ones. The lamellar settler obtained high removal of APs and PBDEs under both dry and wet-weather flows. Adjusting the addition of coagulant-flocculent allows for efficient removal at high pollutant concentrations. By combining physical and biological processes, the biofiltration unit treats nutrient pollution, as well as AP and PBDE contamination efficiently (Gilbert et al., 2012).

Wei, Aziz-Schwanbeck et al. (2012) measured PBDEs in sediment cores collected from six water bodies in Arkansas, including three located close to the flame retardant manufacturing facilities in El Dorado and Magnolia, to investigate past and recent deposition histories. A total of 49 PBDEs and decabromodiphenyl ethane (DBDPE) were detected, with concentrations as high as 57,000 and 2,400 ng/g dry weight for decabromodiphenyl ether (BDE209) and DBDPE, respectively. They also found that debromination of PBDEs occurred in the sediments of a previously used wastewater sludge retention pond which indicated that biological processes play a role in the fate of PBDEs in the sediments.

Degradation of 4-monobrominated diphenyl ether (BDE-3) in anaerobic sludge and the effect of carbon source addition on removal were investigated. It was shown that BDE-3 was transformed into diphenyl ether (DE) and bromide ions. Co-metabolism with glucose facilitated BDE-3 biodegradation in terms of kinetics and efficiency (Shih et al., 2012).

Polychlorinated Compounds (PCCs). PCCs are persistent organic pollutants that have a tendency to biomagnify in the food chain (Nizzetto et al., 2012). Exposure to these compounds at high levels may pose health risks.

PCBs are a group of PCCs. PCBs were widely used as dielectric and coolant fluids, for example in transformers, capacitors, and electric motors. Due to PCBs' environmental toxicity and classification as a persistent organic pollutant, PCB production was banned by the United States Congress in 1979 and by the Stockholm Convention on Persistent Organic Pollutants in 2001. According to the U.S. Environmental Protection Agency (EPA), PCBs have been shown to cause cancer in animals, and there is also evidence that they can cause cancer in humans. The maximum allowable contaminant level in drinking water is set at zero, but due to water treatment technologies a level of 0.5 parts per billion is the defacto level. Although their production is banned in many countries, PCBs are still present in environmental media posing a risk to human health.

Wang, Hunag et al. (2012) indicated that stable colloidal suspensions of buckminsterfullerene (nC(60)) in aqueous environments can significantly affect the fate and

transport of hydrophobic organic contaminants by serving as a contaminant carrier. nC(60) alone and in the presence of natural organic matter (NOM) and surfactants greatly increases the mobility of PCBs in saturated sandy soil. This may facilitate the introduction of PCBs deposited into river and lake sediments into surrounding water environment.

Advanced oxidation processes (AOPs) are effective way of removing PCBs from water environment. In a study, Fang et al. (2012) investigated the effect of chloride ion on the degradation of 2,4,4'-CB (PCB28) and biphenyl (BP) by persulfate, based on the produced $SO_4^{\cdot-}$. The results showed that the presence of chloride ion greatly inhibited the transformation of PC828 and BP.

Polychlorinated dibenzodioxins/furans (PCDD/Fs) are another type of PCCs that are significant environmental pollutants. Members of the PCDD family bioaccumulate in the food chain because of their lipophilic properties, and may cause developmental disturbances and cancer. PCDD/Fs occur as by-products in the manufacture of some organochlorines including pesticides, in the incineration of chlorine-containing substances such as polyvinyl chloride (PVC), in the chlorine bleaching of paper, and from natural sources such as volcanoes and forest fires. They are present in wastewater; however, they preliminary accumulate in the wastewater sludge due to their hydrophobic properties.

The presence of PCDD/Fs and BFRs in sludge generated at municipal WRRFs and industrial WRRFs was investigated in a study. The concentrations of these pollutants were in the following ranges: 5.38-7,947 ng/kg

for 17 PCDD/Fs, 17.5-66,761 µg/kg for 27 PBDEs, 1.55-29,604 µg/kg for hexabromocyclododecanes (HBCDs) (alpha-, beta-, and gamma-diastereomers), and 4.01-618 µg/kg for tetrabromobisphenol A (TBBPA). Generally, the levels of each compound in the sewage-sludge samples were higher than those in the industrial-sludge samples with some exceptions. The characteristic distribution profiles of target compounds were observed for different types of sludge and different sources of wastewater. For instance, high-chlorinated PCDD/Fs were dominant in all samples except those from the textile industry (Hwang et al., 2012).

In a similar study, PCDD/Fs were measured in sewage sludge samples collected from 24 WRRFs from 18 cities of 13 provinces in China. The total concentration of PCDD/Fs ranged from 104 to 1,661 ng/kg-dry weight (Lu et al., 2012).

Besides the other complex PCCs, short chain chlorinated paraffins (SCCPs) are listed as persistent organic pollutant candidates in the Stockholm Convention and are receiving more and more attentions worldwide. In a study by Zeng, Wang et al. (2012), SCCPs were investigated in sewage sludge samples collected from 52 WRRFs in China. Concentrations of total SCCPs (in sludge) were in the range of 0.80-52.7 mg/kg dry weight (dw), with a mean value of 10.7 mg/kg dw. Most of SCCPs in the sludge samples showed a similar congener distribution patterns, and C(11) and Cl(7,8) were identified as the dominant carbon and chlorine congener groups.

Fate of 2,4,6-trichlorophenol (TCP) which is a typical metabolic uncoupler used to sludge generation

during the activated sludge process during biological treatment was investigated. The results showed that half of the TCP applied ends up in the effluent where the rest associates with the activated sludge. Limited biodegradation of TCP was observed. Dosages of more than 4 mg/L TCP (corresponding to 2.5 mg/L residual concentration) for sludge reduction in sequencing batch reactors treating municipal wastewater is not recommended due to acute toxicity (Qiao et al., 2012).

Alkylphenols (APs). The long-chain APs are used extensively as precursors to the detergents, as additives for fuels and lubricants, polymers, and as components in phenolic resins. These compounds are also used as building block chemicals that are also used in making fragrances, thermoplastic elastomers, antioxidants, oil field chemicals and fire retardant materials. APs are present in the wastewater due to their extensive domestic and industrial and release into environment through WRRFs.

Removal efficiencies of tetrabromobisphenol A (TBBPA) in a full-scale conventional activated sludge (CAS) reactor with tertiary treatment and three pilot-scale membrane bioreactors (MBRs) having different sludge retention times (SRTs) were compared in a study. All four reactors were fed the same influent. Among those treatment processes, significant TBBPA removal was observed in the membrane aerated bioreactor. Removal of TBBPA from wastewater was found to be due to a combination of adsorption and biological degradation. It is indicated that nitrification is likely a key process in the TBBPA removal (Potvin et al., 2012).

In that line, Sun et al. (2012) investigated the transformation of bisphenol A (BPA) by ammonia-oxidizing bacteria (AOB) *Nitrosomonas europaea* (*N. europaea*) ATCC 19718. They found *N. europaea* could transform BPA into nitro- and dinitro-BPA, suggesting that abiotic nitration between the biogenic nitrite and BPA played a major role in the transformation of BPA in the batch AOB system. This study indicated that nitrite concentrations, temperature, and pH values were the major factors to influence the reaction rate. Biologically facilitated transformation resulted in reduction in the estrogenicity of the treated water. They also identified nitro-BPA and dinitro-BPA in the effluent of a local WRRF indicating that nitration of BPA is also a pathway for removal of BPA in the actual treatment systems.

On the other hand, Toyama et al. (2012) showed that a novel bacterium *Novosphingobium* sp. strain TYA-1 can utilize BPA and 4-alkylphenols (4-APs) as its carbon and energy source. Biotransformation of BPA follows oxidative skeletal rearrangement involving the cytochrome p450 monooxygenase system. Strain TYA-1 was able to degrade wide variety of 4-APs except the ones with linear side alkyl chains. Degradation of 4-APs, like that of BPA, involved the cytochrome p450 monooxygenase system in strain TYA-1. It was shown that a sequencing batch bioreactor operating at 6 h of reaction time/cycle; 12 cycles in total and containing alginate-immobilized TYA-1 cells (15 mg dry cells) simultaneously removed BPA, 4-tert-BP, and 4-tert-OP from complex polluted waters.

Pharmaceuticals and Personal Care Products (PPCPs)

Hasan et al. (2012) studied adsorptive removal of naproxen and clofibric acid, two typical PPCPs, using metal-organic frameworks (MOFs) for the first time. They observed that adsorption kinetics and capacity of PPCPs generally depend on the average pore size and surface area (or pore volume), respectively, of the adsorbents. It was suggested that MOFs having high porosity and large pore size can be potential adsorbents to remove harmful PPCPs in contaminated water.

Santoke et al. (2012) studied the direct and indirect photochemical fate and treatment by advanced oxidation of three antidepressant compounds (duloxetine, venlafaxine and bupropion) in solutions of humic acid in order to elucidate their behavior in the natural environment. They showed that the hydroxyl radical was an order of magnitude more effective than the singlet oxygen in the solar-induced photochemical degradation of the antidepressants.

Phototransformation of the widely used benzodiazepine pharmaceuticals diazepam and human metabolites nordiazepam, temazepam and oxazepam under simulated sunlight in water was investigated by West and Rowland (2012). Half-lives for each of the benzodiazepine pharmaceuticals were <200 h (under all conditions) suggesting that phototransformation is an important process for such chemicals in the photic zone of receiving waters. Two of the identified photoproducts, 5-chloromethylaminobenzophenone and 2-amino-5-chlorobenzophenone, showed relatively higher persistence

than some of the benzodiazepines, suggesting that the fate and effects of photoproducts should also be incorporated into future risk assessments and environmental models of the fate of benzodiazepines.

Kuroda et al. (2012) examined the city-wide occurrence of selected PPCPs (diethyltoluamide, crotamiton, ethenzamide, propyphenazone, carbamazepine (CBZ), and caffeine) and *E. coli* in confined and unconfined aquifers in Tokyo, where unintended groundwater contamination was suspected to have taken place due to decrepit sewer networks. CBZ and crotamiton were most frequently detected PPCPs. Using CBZ as a sewage marker, Authors estimated that 0.8%-1.7% of the dry-weather flow of sewage was leaking out into the unconfined aquifers.

The competition between the autochthonous fungal/bacterial communities and *T. versicolor* was studied using denaturing gradient gel electrophoresis (DGGE) and the cloning/sequencing approach by Rodriguez, Jelic et al. (2012). Their results suggest that a fungal treatment with *T. versicolor* could be a promising process for the remediation of some pharmaceuticals in complex matrices such as biosolids.

Oxytetracycline (OTC) lab degradation test was performed in liquid medium using the ligninolytic fungus *Pleurotus ostreatus* by Migliore et al. (2012). They found that the OTC removal was mediated by fungal absorption in the mycelia where the OTC molecule underwent a degradation step.

Lee, Zhao et al. (2012) reported a triclosan-degrading wastewater bacterial isolate, *Sphingopyxis* strain

KCY1, capable of dechlorinating triclosan with a stoichiometric release of chloride. In another study, the utility of biofilm-based PPCP removal as a means to prevent environmental PPCP contamination was investigated by Onesios and Bouwer (2012). The examined PPCPs included biosol, biphenylol, p-chloro-m-cresol, p-chloro-m-xyleneol, chlorophene, sodium diclofenac, gabapentin, gemfibrozil, 5-fluorouracil, ibuprofen, ketoprofen, naproxen, triclosan, and valproic acid. This laboratory-scale experiment suggested that biofilm-based water treatment strategies, such as soil aquifer treatment and slow sand filtration, may be well-suited for the removal of many PPCPs from impacted water.

Hillebrand et al. (2012) provided a highly sensitive methodology for the estimation of the mass of untreated wastewater entering karst aquifers with rapid recharge. The approach was based on the mass flow of caffeine in spring water, the load of caffeine in untreated wastewater and the daily water consumption per person in a spring catchment area. The calculated mean contribution of untreated wastewater entering the aquifer was found to be approximately 0.4% of the total amount of wastewater within the spring catchment.

Fenet et al. (2012) determined the sorption isotherm of CBZ, an anti-seizure medication, and two of its metabolites, i.e. CBZ-10,11-epoxide (CBZ-EP) and 10,11-dihydro-10,11-dihydroxycarbamazepine (DiOH-CBZ), in two soils in laboratory assays, and investigated the presence of CBZ and its metabolites in soil and in groundwater underlying an irrigated area with treated wastewater. Their analysis revealed that CBZ and its two

metabolites were present in treated wastewater used for irrigation and in groundwater.

Kim, Kim et al. (2012) conducted a study to evaluate the degradation and mineralization of antibiotics (sulfamethoxazole (SMX) and chlortetracycline) using an electron beam, ozone and UV, and the change of toxicity. Their results showed that an electron beam was effective for the removals of both SMX and chlortetracycline in aqueous solutions, and was more efficient than ozone and UV. Authors concluded that electron beam treatment could be an effective and safe method for the removal of antibiotic compounds.

The study by De la Cruz et al. (2012) focused on the removal of 32 selected micropollutants (pharmaceuticals, corrosion inhibitors and biocides/pesticides) found in an effluent coming from a municipal WRRF based on activated sludge using AOP. The highlight of the paper was to point out the possibility of the micropollutants degradation in spite the presence of dissolved organic matter (DOM) in much higher concentrations.

Braeutigam et al. (2012) applied cavitation, the formation, growth, and subsequent collapse of gas- or vapor-filled bubbles in fluids, as an efficient and cost-effective technique for to stop the entry of CBZ into the aquatic environment. The technique of Hydrodynamic-Acoustic-Cavitation was used showing high synergistic effect. Under optimized conditions in their study, CBZ (5 µg/L) was transformed by pseudo-first order kinetics to an extent of >96% within 15 min (27% by hydrodynamic cavitation, 33% by acoustic cavitation).

Quesada-Penate et al. (2012) evaluated the catalytic wet air oxidation (CWAO) of paracetamol (PCT) on activated carbon as a water treatment technique using an autoclave reactor and as a regenerative treatment of the carbon after adsorption in a sequential fixed bed process. Respirometry tests with activated sludge revealed that in the studied conditions the use of CWAO enhanced the aerobic biodegradability of the effluent.

Ben et al. (2012) assessed the potential of ozone for the degradation of nine commonly used veterinary antibiotics (five sulfonamides, three tetracyclines, and one macrolide) in swine wastewater pretreated by sequencing batch reactor (SBR). Study revealed that in the SBR effluent, the dissolved organic materials strongly competed for oxidants against antibiotics, while the suspended solids had insignificant influence on antibiotics degradation. Authors concluded that ozonation is a feasible polishing process, subsequent to biological treatment, for removal of residual antibiotics from swine wastewater, and the primary reaction mechanism was direct oxidation via ozone rather than indirect oxidation via hydroxyl radicals.

Gomez et al. (2012) presented a collection of data of trace organic micropollutants in an urban WRRF in North East Spain using reverse osmosis (RO) membrane treatment. Their results indicated that secondary effluents from the WRRF contained most of the studied organic compounds indicating incomplete removal of organics in the conventional treatment of the plant. However, the rejection of most micropollutants was high for all three RO membrane types (low energy, high rejection, fouling resistant) tested; however, some selected micropollutants

were less efficiently removed (e.g. the small and polar and the more hydrophobic) and the molecular weight and membrane material influenced removal efficiencies.

Empirical correlations for differential UV254 absorbance and total fluorescence were developed by Gerrity et al. (2012) for the oxidation of 18 trace organic contaminants (TOrcs), including 1,4-dioxane, atenolol, atrazine, BPA, CBZ, diclofenac, gemfibrozil, ibuprofen, meprobamate, naproxen, N,N-diethyl-meta-toluamide (DEET), para-chlorobenzoic acid (pCBA), phenytoin, primidone, SMX, triclosan, trimethoprim, and tris-(2-chloroethyl)-phosphate ($R^2 = 0.50-0.83$) and the inactivation of three microbial surrogates, including *E.coli*, MS2, and *Bacillus subtilis* spores ($R^2 = 0.46-0.78$).

The fate of 14 antidepressants along with their respective N-desmethyl metabolites and the anticonvulsive drug CBZ was studied in 5 different WRRFs across Canada by Lajeunesse et al. (2012). Their results showed that venlafaxine (VEN), its metabolite O-desmethylvenlafaxine (DVEN), citalopram (CIT), and CBZ were detected at the highest concentrations in raw influent, and there was a strong evidence that primary treatment and trickling filter/solids contact has limited capacity to remove antidepressants from sewage, while activated sludge, biological aerated filter, and biological nutrient removal processes yielded moderate results (mean removal rates: 30%).

Study by De Corte et al. (2012) showed that doping of bio-Pd nanoparticles with Au(0) can be a promising approach for the reductive treatment of wastewaters containing halogenated contaminants. The

study by Li, Nanaboina et al. (2012) examined effects of Fenton oxidation on trace level PPCPs commonly occurring in wastewater. All tested PPCPs in that study, except atrazine and iopromide, were completely removed by Fenton treatment. The relative decrease of effluent organic matter absorbance was correlated with the removal of tested PPCPs.

The study by Dolar et al. (2012) dealt with application of RO and nanofiltration (NF) membranes for removing of antibiotic residues (SMX, trimethoprim, ciprofloxacin, dexamethasone and febantel) and their mixture. According to the results obtained by Authors the use of RO and the tight NF membranes were recommended to achieve a high level of retention (>95%) of all selected veterinary antibiotics.

In the study by Kohler et al. (2012) the efficiency of the pharmaceutical removal process was assessed by examining 14 micropollutants associated with hospitals. While the membrane bioreactor (MBR) treatment generally showed only a low degradation capacity for persistent pharmaceuticals, much better degradation was obtained by applying UV irradiation and H_2O_2 as AOP.

Kasprzyk-Hordern and Baker (2012) discussed the enantiomer-specific fate of chiral drugs during wastewater treatment and in receiving waters. Their results revealed the extent of stereoselectivity depended on several parameters including: type of chiral drug (high stereoselectivity was recorded for atenolol and MDMA), treatment technology used (activated sludge showed higher stereoselectivity than trickling filters), and season (higher

stereoselectivity was observed in the aqueous environment over the spring/summer time).

Keen et al. (2012) traced the transformation products of the C-14-labeled CBZ during UV/hydrogen peroxide advanced oxidation and subsequent biotransformation by mixed, undefined cultures. Their results show that the oxidation byproducts of CBZ containing a hydroxyl or carbonyl group can be fully mineralized by a mixed bacterial inoculum. A tertiary treatment approach that includes oxidation and biotransformation has the potential to synergistically mineralize persistent pharmaceutical compounds in WRRF effluents.

The study by Kosjek et al. (2012) revealed the presence of benzodiazepine derivatives in wastewater influent and effluent samples. Results of the study highlighted recalcitrant nature of benzodiazepine derivatives as it required coupled biological and photochemical treatment followed by the adsorption to activated carbon to achieve a removal efficiency of 99.99%.

Nodler et al. (2012) studied microbially mediated abiotic formation of reversible and non-reversible SMX transformation products during denitrification processes. Based on the results, retransformation of 4-nitro-SMX to SMX was postulated during denitrification.

Ottmar et al. (2012) investigated the environmental behavior of two widely prescribed cholesterol-lowering statin drugs that are expected to be present at significant concentrations in wastewater influents, namely: atorvastatin and simvastatin. Results

suggested that 85-90% of each drug is removed during conventional treatment, with sorption accounting for less than 10% of overall removal, and hence statin active ingredients were not considered to pose a significant environmental threat.

The sorption and biodegradation of three sulfonamide antibiotics, namely SMX, sulfadimethoxine (SDM), and sulfamonomethoxine (SMM), in an activated sludge system were investigated by Yang, Lin et al. (2012). Their measured kinetics implied that with typical hydraulic retention time provided by WRRF the removal of sulfonamide compounds from the wastewater during the activated sludge process would approximate 2 µg/L.

Gasser et al. (2012) studied the stereoselectivity of R,S-venlafaxine and its metabolites R,S-O-desmethylvenlafaxine, N-desmethylvenlafaxine, O,N-didesmethylvenlafaxine, N,N-didesmethylvenlafaxine and tridesmethylvenlafaxine in three processes: (i) anaerobic and aerobic laboratory scale tests; (ii) six WRRFs operating under different conditions; and (iii) a variety of wastewater treatments including conventional activated sludge, natural attenuation along a receiving river stream and storage in operational and seasonal reservoirs. From the laboratory and field studies, results suggested that the levels of the drug and its metabolites and the stereoisomeric enrichment of the metabolite and its parent drug can be used for source tracking and for discrimination between domestic and nondomestic wastewater pollution.

Kovalova et al. (2012) installed a pilot-scale MBR to study the efficiency of micropollutant elimination in raw hospital wastewater that comprises a complex matrix

with micropollutant concentrations ranging from low ng/L to low mg/L. No inhibition by antibacterial agents or disinfectants from the hospital was observed in the MBR. The hospital wastewater was found to be a dynamic system in which conjugates of pharmaceuticals deconjugate and biological transformation products were formed, which in some cases were pharmaceuticals themselves.

Bialk-Bielinska et al. (2012) investigated the hydrolytic stabilities of 12 sulfonamides. Their results showed that under typical environmental conditions (pH and temperature) sulfonamides were hydrolytically stable with a long half-life.

Bahlmann et al. (2012) monitored CBZ, caffeine and cetirizine by enzyme-linked immunosorbent assays (ELISAs) in surface and wastewaters from Berlin, Germany. Results revealed that caffeine concentrations found in surface waters were elevated by combined sewer overflows after stormwater events; while during the hay fever season, the concentrations of the antihistamine drug cetirizine increased in both surface and wastewaters. Caffeine was almost completely removed during wastewater treatment, while CBZ and cetirizine were found to be more persistent.

Sulfapyridine (SPY), sulfonamide typically used in human therapies, and veterinary sulfonamide sulfamethazine (SMZ) and their acetylated metabolites, AcSPY and AcSMZ, were studied under artificial irradiance conditions by Garcia-Galan et al. (2012). Estimated degradation rate constants (k) ranged from 0.063 per h (SPY) to 2.808 per h (AcSPY) with corresponding half-lives ($t(1/2)$) of 10.93 h and 0.25 h, respectively.

The influence of operation condition on the fate of 16 PPCPs in a single-sludge nitrifying/denitrifying pilot plant was assessed by Suarez et al. (2012). SRT was termed as an important factor for the removal of compounds that significantly sorb onto sludge while the internal recirculation ratio was termed significant for the removal of moderately biodegradable compounds. Results revealed most compounds were removed due to biotransformation, although for fragrances sorption and volatilization played a role.

Albero et al. (2012) developed a rapid method for determination of seven parabens and two chlorinated by-products in sewage sludge based on matrix solid-phase dispersion and gas chromatography-tandem mass spectrometry. In order to study the temporal variation of parabens and two chlorinated parabens during a four-year period, sludge samples were collected from 3 selected WRRFs in the study. The levels of methylparaben encountered were rather constant throughout the sampling period whereas propylparaben levels slightly increased.

The occurrence and distribution of six psychoactive compounds (primidone, phenobarbital, oxazepam, diazepam, meprobamate, and pyrithyldione) and a metabolite of primidone (phenylethylmalonamide) were investigated in WRRF effluents, surface water, groundwater of a bank filtration site, raw and final drinking water, and in groundwater affected by former sewage irrigation by Hass et al. (2012). Primidone and its metabolite phenylethylmalonamide were found to be ubiquitous in environmental water samples in Berlin. Their results indicated a high persistence of some of the

investigated compounds in the aquatic system; hence, Authors concluded that these pollutants may potentially reach drinking water resources via bank filtration if present in WRRF effluents and/or surface waters in partly closed water cycles such as Berlin's.

The study by Jia et al. (2012) developed a method for analysis of nineteen quinolone and fluoroquinolone antibiotics (FQs) in sludge samples, and investigated the occurrence and fate of the FQs in a municipal WRRF with anaerobic, anoxic, and aerobic treatment processes. Eleven compounds, including pipemidic acid, fleroxacin, ofloxacin, norfloxacin, ciprofloxacin, enrofloxacin, lomefloxacin, sparfloxacin, gatifloxacin, moxifloxacin, and sarafloxacin (only in sludge), were detected in the WRRF while the mean removal efficiencies of FQs in the WRRF were found to be 56-75%, except for new generation drugs such as moxifloxacin (40 +/- 5%) and gatifloxacin (43 +/- 13%). Authors found a significant positive correlation between removal efficiencies and K_d of FQs as the major factor in the removal of FQs in the WRRF was sorption to sludge, which was not governed by hydrophobic interactions.

Ruel et al. (2012) analyzed more than 100 substances through on-site mass balances over 19 municipal wastewater treatment lines according to their occurrence in raw wastewater, in treated wastewater and in sludge, and assessed their fate in wastewater treatment processes. Authors found that main micropollutants of concerns were cadmium, di-2-ethylhexyl phthalate (DEHP), diuron, alkylphenols, and chloroform while the emerging substances of particular concern were by-

products, organic chemicals (e.g. triclosan, benzothiazole) and pharmaceuticals (e.g. ketoprofen, diclofenac, SMX, CBZ). About 80% of the load of micropollutants was removed by conventional activated sludge plants, but about two-thirds of removed substances were mainly transferred to sludge.

The study by Bueno et al. (2012) summarized results obtained during almost two-years of a monitoring program carried out in five municipal WRRF located in the north, center, and south-east of Spain which evaluated the occurrence and persistence of a group of 100 organic compounds belonging to several chemical groups (pharmaceuticals, personal care products, pesticides and metabolites). The average removal efficiencies of the WRRFs studied varied from 20% (erythromycin) to 99% (acetaminophen). The study also identified 20 of the mostly detected and persistent compounds in wastewater effluent, of which hydrochlorothiazide, atenolol, gemfibrozil, galaxolide and three metabolites (fenofibric acid, 4-AAA and 4-FAA), presented the highest average contribution percentages, in relation to the total load of contaminants for the different WRRFs effluent studied.

In the study by Falas, Andersen et al. (2012), influent and effluent concentrations compiled from the Swedish screening programs were used to assess the occurrence and reduction of non-antibiotic pharmaceuticals for human usage. The influent concentrations were compared with the sale volumes and for many pharmaceuticals it was shown that only a small fraction of the amount sold reached WRRFs as dissolved parent compounds. Further comparison based on the biological

treatment showed lower removal rates for several pharmaceuticals in trickling filter plants compared with activated sludge plants with nitrogen removal.

The scope of the study by Gracia-Lor (2012) included understanding occurrence and fate a wide variety of pharmaceuticals belonging to different therapeutic classes from 112 samples, including influent and effluent wastewater, from different conventional WRRFs. Results indicated that 17 compounds were detected in the samples, with analgesics and anti-inflammatories, cholesterol lowering statin drugs and lipid regulators being the most frequently detected groups. 4-Aminoantipyrine, bezafibrate, diclofenac, gemfibrozil, ketoprofen, naproxen and venlafaxine were the compounds most frequently found. Removal of most of the selected compounds occurred during the treatment process of influent wastewater, although it was incomplete.

Martin et al. (2012) evaluated the occurrence of sixteen pharmaceutically active compounds in influent and effluent wastewater and in primary, secondary and digested sludge in one-year period. All the compounds found in wastewater were also found in sewage sludge, except diclofenac, while ibuprofen, salicylic acid, gemfibrozil and caffeine were the compounds found at the highest concentrations. The highest ecotoxicological risk in effluent wastewater and digested sludge was due to ibuprofen (risk quotient (RQ): 3.2 and 4.4, respectively), 17 alpha-ethinylestradiol (RQ: 12 and 22, respectively) and 17 beta-estradiol (RQ: 12 and 359, respectively) while ecotoxicological risk after wastewater discharge and sludge

disposal was limited to the presence of 17 beta-estradiol in digested-sludge amended soil (RQ: 2.7).

Gao, Ding et al. (2012) studied the occurrence and removal efficiencies of fifteen pharmaceuticals in a conventional municipal WRRF in Michigan, and found that the pharmaceuticals detected in wastewater were also found in the corresponding sludge phase. The removal efficiencies of chlortetracycline, tetracycline, sulfamerazine, acetaminophen and caffeine were >99%, while doxycycline, oxytetracycline, sulfadiazine and lincomycin exhibited relatively lower removal efficiencies (e.g., <50%). Based on the mass balance analysis, biotransformation is believed to be the predominant process responsible for the removal of pharmaceuticals (22% to 99%), whereas contribution of sorption to sludge was relatively insignificant (7%) for the investigated pharmaceuticals.

Prieto-Rodriguez et al. (2012) selected TiO₂ concentrations of only tens of mg/L to evaluate the influence of catalyst load, initial hydrogen peroxide dose and radiation intensity on the degradation rate of five emerging contaminants (100 µg/L of ofloxacin, SMX, flumequine, CBZ, and 2-hydroxy-biphenyl) spiked in a real municipal WRRF effluent. It was demonstrated that the use of hydrogen peroxide is highly recommendable for working with TiO₂ at low concentrations and high photon flux must be avoided for effective removal of these contaminants. It was also demonstrated that too low (less than 40 mg/L) TiO₂ concentration was not recommendable.

Heeb et al. (2012) investigated the origin, temporal magnitudes, and annual mass fluxes of a wide

range of pharmaceuticals, household chemicals, and pesticides in the Haihe River System (HRS), which drains 70% of the wastewater discharged by 20 million people living in Beijing. Mass-flux analyses revealed that pharmaceuticals (5930 kg/year) and most household chemicals (5660 kg/year) originated from urban wastewaters, while the corrosion inhibitor benzotriazole entered the rivers through other pathways. Per capita loads of pharmaceuticals in wastewater were lower than those in Europe, but were expected to increase in the near future, and hence, Authors warned that the loads of polar organic micropollutants transported with the water might pose a serious threat to food safety and groundwater quality as 95% of the river water is diverted to irrigate agricultural soil.

This study by Wang, He et al. (2012) investigated the reactions of trimethoprim (TMP) and two sub structure model compounds [2, 4-diamino-5-methylpyrimidine (DAMP) and 3, 4, 5-trimethoxytoluene (TMT)] with chlorine dioxide (ClO_2), one of the common water disinfection oxidants, in batch reactors. Comparison among TMP, DAMP, and TMT revealed that the reactivity of TMP to ClO_2 resided mostly in its diaminopyrimidinyl moiety at neutral to alkaline pH, but centers in its trimethoxybenzyl moiety at acidic conditions. Overall, transformation of TMP by ClO_2 was expected under typical ClO_2 disinfection conditions.

In the study by Chen, Richard et al. (2012) removal of triclosan by aqueous ozone was investigated and the degradation products formed during ozonation of an aqueous solution of triclosan were analyzed by GC-MS

and liquid chromatography tandem mass spectrometry (LC-MS/MS). The following transformation products were identified: 2,4-dichlorophenol, chlorocatechol, mono-hydroxy-triclosan and di-hydroxy-triclosan during treatment process. Cytotoxicity and genotoxicity of pure triclosan and 2,4-dichlorophenol were investigated and the results showed reduced genotoxic effects after ozonation, though the respective chlorophenol is harmful to aquatic organisms.

This pilot-scale research project by Lee, Howe et al. (2012) investigated and compared the removal of PPCPs and other micropollutants from treated wastewater by ozone/biofiltration and RO. The reduction in UV254 absorbance as a function of ozone dose correlated well with the reduction in non-biodegradable dissolved organic carbon and simultaneous production of biodegradable dissolved organic carbon (BDOC). When wider environmental impacts such as energy consumption, water recovery, and waste production were considered by Authors, they concluded that ozone/biofiltration was a more desirable process than RO for removing PPCPs and other trace organics from treated wastewater although ozone alone did not mineralize organics in treated wastewater.

The photo-Fenton degradation of PCT was evaluated by Trovo et al. (2012) using FeSO_4 and the iron complex potassium ferrioxalate (FeOx) as iron source under simulated solar light. Their results showed that the degradation was favored in the presence of FeSO_4 in relation to FeOx , and the higher concentration of hydroxylated intermediates generated in the presence of

FeSO₄ in relation to FeOx probably enhanced the reduction of Fe(III) to Fe(II) improving the degradation efficiency. The considerable decrease of the sample toxicity during photo-Fenton treatment using FeSO₄ indicated a safe application of the process for the removal of PCT.

The effect of pH on chlorination behaviors of 12 antibiotics, including beta-lactams, sulfonamides, fluoroquinolones, tetracyclines, macrolides, and others at environmentally relevant concentrations was systematically examined by Li and Zhang (2012) in the effluent matrix of activated sludge process. The elimination rates of ciprofloxacin, norfloxacin, anhydro-erythromycin, and roxithromycin increased while that of SMX decreased significantly with the increase of pH while Sulfadiazine, ofloxacin, and trimethoprim exhibited the highest reactivity with free available chlorine under the pH of 6 -7, 7, and 7.5, respectively. Anionic antibiotic species were found to be much more reactive (1-3 orders of magnitude greater) than cationic antibiotic species toward free available chlorine.

Moctezuma et al. (2012) investigated the advanced oxidation of PCT promoted by TiO₂/UV system in aqueous medium. The formation of hydroquinone, benzoquinone, p-aminophenol and p-nitrophenol in the reaction mixtures was demonstrated. Continuous monitoring by IR spectroscopy demonstrated the breaking of the aromatic amide present in PCT and subsequent formation of several aromatic intermediate compounds such as p-aminophenol and p-nitrophenol which were eventually converted into trans-unsaturated carboxylic acids.

Chong and Jin (2012) assessed a titanium dioxide (TiO₂) nanofiber based wastewater treatment process as a pre-treatment system to treat and enhance the biodegradability of a representative pharmaceutical compound, 5,000 µg/L of CBZ, in synthetic hospital wastewater. Results showed that the stand-alone TiO₂ pre-treatment system was capable of removing 78% of CBZ, 40% of COD and 23% of PO₄ concentrations from the influent wastewater within a 4 h reaction time. Eventually, it was found that the photodegradation profile for high concentration CBZ in synthetic hospital wastewater was perfectly fitted to the Langmuir-Hinshelwood kinetics model.

Hernandez-Ruiz et al. (2012) assessed the interactions between three common PPCP contaminants and two distinct DOM sources: a WRRF and the Suwannee River, GA (SROM). Aqueous DOM solutions (8.0 mg/L C, pH 7.4) were spiked with a range of concentrations of bisphenol-A, CBZ and ibuprofen to assess the DOM fluorophores quenched by PPCP interaction in excitation-emission matrices (EEM). Their results suggested that presence of DOM at environmentally-relevant concentration gave rise to PPCP interactions that could potentially affect their environmental transport, but these DOM-contaminant interactions did not suppress the accurate assessment of target analyte concentrations by aqueous injection ultra-high performance LC-MS/MS.

Jin, Shah et al. (2012) compared the rejection efficiency of the pharmaceutical compounds between commercial cellulose triacetate (CIA) based membranes and thin film composite (TFC) polyamide based

membranes. The rejection behavior was related to membrane interfacial properties, physicochemical characteristics of the pharmaceutical molecules and feed solution pH. TFC polyamide membranes exhibited excellent overall performance, with high water flux, excellent pH stability and great rejection of all pharmaceuticals investigated (>94%) in that study.

Schroder et al. (2012) evaluated the treatment of wastewater containing three nonsteroidal anti-inflammatory drugs (NSAIDs) (acetaminophen, ketoprofen and naproxen) and three antibiotics (roxithromycin, SMX and trimethoprim) performed in two MBRs with SRTs of 15 (MBR-15) and 30 (MBR-30) days over a period of four weeks. It was observed that NSAIDs were removed with higher efficiencies than the antibiotics for both MBRs, and the MBR-30 presented higher removal efficiencies for all the compounds than obtained by MBR-15. Removal rates ranged from 55% (SMX) up to 100% (acetaminophen, ketoprofen).

Ding et al. (2012) examined virgin and exhausted adsorbents via thermal and Fourier transform infrared spectroscopy (FTIR) analyses to elucidate adsorption mechanisms of pharmaceuticals. Maximum adsorption capacities for the 6 materials tested ranged from 80 to 300 mg/g, comparable to the adsorption capacities of antibiotics on various activated carbons (200-400 mg/g) reported in the literature. A large volume of pores similar in size to the adsorbate molecules with hydrophobic carbon-based origin of pore walls was indicated as an important factor promoting the separation process.

Pharmaceutical treatment using a series of wastewater treatment processes of the dissolved air flotation (DAF)–MBR–ozone oxidation was conducted in the study by Choi et al. (2012). Despite the high removal rate of the wastewater treatment system, pharmaceuticals were eliminated to be about 50–99% by the MBR system, depending on specific pharmaceuticals. Ibuprofen was well removed by MBR system up to over 95%, while removal rate of bezafibrate ranged between 50 and 90%. With over 5 mg/l of ozone oxidation, most pharmaceuticals which survived the DAF–MBR process were removed completely or resulted in very low survival rate within the range of few µg/L; however, some pharmaceuticals such as bezafibrate and naproxen tended to be resistant to ozone oxidation.

Rodriguez, Baron et al. (2012) assessed a biotechnological approach which employed the fungus *Trametes versicolor* in a sludge-bioslurry reactor in order to remove several groups of emerging pollutants. Fifteen out of 24 detected pharmaceuticals were removed at efficiencies over 50% after the treatment, including eight completely degraded. Removal ranged between 16-53% and 22-100% for the brominated flame retardants and the UV-filters, respectively.

Yang, Ying et al. (2012) investigated the removal efficiencies of 68 selected endocrine disrupting chemicals (EDCs) and PPCPs spiked in a wastewater matrix by ferrate (Fe(VI)) and further evaluated the degradation of these micropollutants present in secondary effluents of two WRRFs by applying Fe(VI) treatment technology. Fe(VI) treatment resulted in selective oxidation of electron-rich organic moieties of these target compounds, such as

phenol, olefin, amine and aniline moieties. The results from this study clearly demonstrated the effectiveness of Fe(VI) treatment as a tertiary treatment technology for a broad spectrum of micropollutants in wastewater.

Zeng, Ji et al. (2012) investigated the mechanism on aquatic photodegradation of atenolol with an emphasis on the role of DOM as well as other natural water compositions (nitrate, bicarbonate and ferric ions). Significant acceleration of photodegradation of atenolol was observed in the presence of each DOMs added in the study. Hydroxyl radical ((OH)-O-center dot) was determined as the main reactive species in this process, instead of singlet oxygen or excited triplet of DOM.

Occurrence and fate of four common synthetic musks (SMs) (galaxolide (HHCB), tonalide (AHTN), musk xylene (MX) and musk ketone (MK)) were studied by Sang et al. (2012) in different WRRFs of Shanghai, China among different seasons. Results showed that total dissolved concentrations of the four SMs were 536-3,173 ng/L in influent, 351-2,595 ng/L in effluent and 147-6,839 µg/kg dry weights in sludge. There was no significant removal of SMs in most of the sewage samples of the four WRRFs.

Liu, Nanaboina, and Korshin (2012) examined effects of ozonation on thirteen fluoroquinolone, macrolide and lincosamide antibiotics present in municipal wastewater. Concentrations of aldehydes and carboxylic acids generated via the oxidation of effluent organic matter (EfOM) were also determined in the study. The removal of antibiotics and production of aldehydes and carboxylic acids were strongly correlated with the changes in both EfOM absorbance and fluorescence.

Liu, Nanaboina, Korshin, and Jiang (2012) addressed the formation and properties of degradation products of ciprofloxacin, norfloxacin and lomefloxacin formed during ozonation of secondary wastewater effluent containing these fluoroquinolone antibiotics. The generation of the degradation products was interpreted in the context of transformations of EfOM tracked via absorbance measurements. These pathways were largely similar for all studied fluoroquinolones and involved attacks on the piperazine ring and the quinolone structure.

Berset and Ochsenbein (2012) presented a LC-MS/MS method for the simultaneous determination of frequently used artificial sweeteners (ASs) and the main metabolite of aspartame (ASP), diketopiperazine (DKP), in environmental water samples using the direct-injection (DI) technique. Acesulfame (ACE), saccharin (SAC), cyclamate (CYC) and sucralose (SUC) were found in water samples tested in the study. Highest concentrations up to 61 µg/L of ACE were found in wastewater effluents while the metabolite DKP was only detected in wastewater up to 200 ng/L and at low detection frequencies.

The stability of nine illicit drugs and metabolites in influent wastewater at relevant conditions (20 C and at pH 7.5) was evaluated by van Nuijs et al. (2012) during 26h (in 1 h intervals) to assess whether significant in-sewer processes of degradation or formation occur. The results of the study showed that amphetamine, methamphetamine, ecstasy and EDDP (metabolite of methadone) were stable in wastewater for 12 h and longer. For benzoylecgonine and methadone, a modest formation in the course of time was observed (0.2% per h and 0.4% per h, respectively),

while for cocaine (40% degradation after 12 h), ecgonine methylester (20% degradation after 12 h), and 6-monoacetylmorphine (20% degradation after 12h), a clear decrease in concentrations in time was seen.

Falas, Baillon-Dhumez et al. (2012) assessed the removal of seven active pharmaceutical substances (ibuprofen, ketoprofen, naproxen, diclofenac, clofibric acid, mefenamic acid, and gemfibrozil) by batch experiments, with suspended biofilm carriers and activated sludge from several full-scale WRRFs. A distinct difference between nitrifying activated sludge and suspended biofilm carrier removal of several pharmaceuticals was demonstrated. Biofilm carriers from full-scale nitrifying WRRFs, demonstrated considerably higher removal rates per unit biomass (i.e. suspended solids for the sludges and attached solids for the carriers) of diclofenac, ketoprofen, gemfibrozil, clofibric acid and mefenamic acid compared to the sludges.

The study by de Godos et al. (2012) determined the mechanisms of tetracycline removal during wastewater treatment in high rate algal ponds (HRAPs). The results from the batch assays suggested that the light-shading and/or pollutant-sequestering effects of the biomass limited tetracycline removal in the pond. These results demonstrate that the shallow geometry of HRAPs was advantageous to support the photodegradation of antibiotics during wastewater biological treatment but that the presence of these pollutants could hamper biomass recovery.

Luo et al. (2012) investigated the sunlight mediated photochemical fate, and treatment using advanced

oxidation and reduction (free radical) processes with respect to their effect on trimethoprim (TMP). The reaction of TMP with excited state natural organic matter is postulated as a significant degradation pathway for the loss of TMP in sunlit waters containing natural organic matter. There was no effect of pH on the direct or indirect photolysis of TMP.

Estrogens

The removal mechanisms of a natural estrogen, 17 β -estradiol (E2), and a synthetic estrogen, 17 α -ethinylestradiol (EE2) were investigated in MBRs with and without powdered activated carbon (PAC) addition (Yang, Zhou et al., 2012). The experimental results showed that the average removal rates of E2 and EE2 by the MBR without PAC addition were 89.0% and 70.9%, respectively; PAC addition in the MBR increased the removal rate of E2 and EE2 by 3.4% and 15.8%, respectively. The study also showed that the biodegradation was the dominant mechanism for the removal of E2 and EE2 in MBRs.

The study by Baynes et al. (2012) assessed both steroid removal and intersex induction in adult and early life stage fish (roach, *Rutilus rutilus*) when exposed directly to either secondary (activated sludge process (ASP)), tertiary (sand filtered (SF)), or advanced (ClO₂, granular activated charcoal (GAC)) treated effluents for six months. Their results showed that both the advanced GAC and tertiary SF treatments (but not the ClO₂ treatment) significantly removed the intersex induction associated with the ASP effluent; this was not predicted by the steroid

estrogen measurements, which were higher in the tertiary SF than either the GAC or the ClO₂.

Zheng et al. (2012) investigated the transformation kinetics and mechanisms of three natural estrogenic hormones (17 alpha-estradiol, 17 beta-estradiol, and estrone) in aqueous solutions blended with dairy lagoon water under anaerobic conditions to assess the potential risk of hormone contaminants derived from animal wastewater. Their observations suggested that these hormones are relatively stable over time and may accumulate in anaerobic or anoxic environments and anaerobic concentrated animal feeding operations (CAFO) lagoons. A racemization reaction between 17 alpha-estradiol and 17 beta-estradiol via estrone was observed in aqueous solutions in the presence of CAFO wastewater under anaerobic conditions.

The photochemical behaviors of the isoflavones, plant-derived chemicals that are potential endocrine disruptors, daidzein, formononetin, biochanin A, genistein, and equol were studied by Felcyn et al. (2012) under simulated solar light and natural sunlight. The indirect photolysis rates for these compounds scaled with NOM concentration, and NOM from microbial origin was found to be a more potent photosensitizer than NOM from terrestrial sources. Results of these studies implicated reaction with both singlet oxygen and excited state triplet NOM.

Phillips et al. (2012) collected data at a WRRF to assess the relative contribution of combined sewer overflow (CSO) bypass flows and treated wastewater effluent to the load of steroid hormones and other

wastewater micropollutants (WMPs) from a WRRF to a lake. Although CSO discharges represented 10% of the total annual water discharge (CSO plus treated plant effluent discharges) from the WRRF, CSO discharges contributed 40-90% of the annual load for hormones and WMPs with high (>90%) wastewater treatment removal efficiency. By contrast, it was noted that compounds with low removal efficiencies (<90%) have less than 10% of annual load contributed by CSO discharges.

The fate and behavior of estrone-3-sulfate (E1-3S), estradiol-3-sulfate (E2-3S), estrone-3-glucuronide (E1-3G) and estradiol-3-glucuronide (E2-3G) were studied by Kumar et al. (2012) in raw sewage, activated sludge and river water using microcosms. Field observations at three activated sludge processes suggested complete transformation of the glucuronide conjugates in the sewer largely to their parent estrogens. Field studies also indicated transformation of sulfate conjugates across the sewage treatment, although a proportion escaped into the effluent.

Kelly and Arnold (2012) explored the environmental fate of genistein and daidzein, two estrogenic compounds derived from plants, focusing on direct and indirect photolysis. Their results suggested that daidzein was transformed mainly via direct photolysis and singlet oxygenation, while genistein was transformed mainly via reaction with triplet-state natural organic matter.

The effect of inorganic ions and dissolved organic matter (DOM) on the TiO₂ photocatalytic removal of estrogenic activity from secondary effluents of municipal WRRFs was investigated by Zhang, Li, Su et al.

(2012). The presence of HPO_4^{2-} , NH_4^+ , and HCO_3^- resulted in a significantly negative impact on the photocatalytic removal of estrogenic activity from synthetic water due to their strong adsorption on the surface of TiO_2 . However, only a weak impact was noted during photocatalytic removal of estrogenic activity from secondary effluent with these ions added, since the presence of DOM in real wastewater played a more important role in inhibiting photocatalytic removal of estrogenic activity than inorganic ions.

Paterakis et al. (2012) evaluated the fate and behavior of two groups of endocrine disrupting chemicals, steroid estrogens and nonylphenol ethoxylates, during the anaerobic digestion of primary and mixed sewage sludge under mesophilic and thermophilic conditions. The biodegradation of total steroid estrogens observed was >50% during primary sludge digestion with lower removals observed for mixed sludge, which reflected bulk organic solids removal efficiencies. The removal of nonylphenol ethoxylates was greater in mixed sludge digestion (>58%) compared with primary sludge digestion and did not reflect bulk organic removal efficiencies.

Literature review by Hamid and Eskicioglu (2012) highlighted contribution of estrogenic hormones (estrone (E1), E2, estriol (E3), EE2) as the major contributor to the total estrogenicity in waterways. Their review of the fate studies revealed that activated sludge system with nutrient removal shows very high (>90%) removal of estrogenic hormones in most of the cases. Although, aerobic digestion showed better attenuation of

estrogenic compounds, anaerobic digestion increased the overall estrogenicity of biosolids.

Barber et al. (2012) presented a series of integrated chemical and biological investigations into the occurrence, fate, and effects of endocrine-disrupting chemicals in the City of Boulder Colorado's WRRF and Boulder Creek, the receiving stream. Results showed the effects of a full-scale upgrade of the WRRF from a trickling filter/solids contact process to an activated sludge process on the removal of endocrine-disrupting compounds and other contaminants (including nutrients, boron, bismuth, gadolinium, and ethylenediaminetetraacetic acid) through each major treatment. The upgrade of the WRRF resulted in improved removal efficiency for many endocrine-disrupting chemicals, particularly 17 beta-estradiol and estrone, and fish exposed to the postupgrade effluent indicated reduction in endocrine disruption relative to preupgrade conditions.

Tertiary WRRF effluent before and after ozonation was tested by Altmann et al. (2012) for aquatic ecotoxicity in a battery of standardized microbioassays with green algae, daphnids, and zebrafish eggs. In addition, unconjugated estrogen and 17 beta-hydroxyandrogen immunoreactive substances were quantified by means of enzyme immunoassays. Based on the presented screening, ozonation effectively reduced steroid hormone levels in the WRRF effluent without increasing the effluent's ecotoxicity.

Two estrogen fate and transformation models were integrated with a comprehensive activated sludge model (ASM) by Lust et al. (2012) to predict estrogen

removal based on biomass and solids production. Their estrogen fate model relating the rate of total estrogen degradation to soluble estrogen concentrations successfully predicted estrogen removals when compared with measured concentrations.

Abargues et al. (2012) compared the fate of the alkylphenols-APs (4-(tert-octyl) phenol, t-nonylphenol and 4-p-nonylphenol and the hormones (estrone, 17 beta-estradiol and 17 alpha-ethinylestradiol) in a submerged anaerobic membrane bioreactor (SAMBR) pilot plant and in a conventional activated sludge WRRF (CAS). Their results showed that the APs soluble concentrations in the SAMBR effluent were always significantly higher than the CAS ones. Moreover, the analyses of the suspended fraction revealed that the AP concentrations in the SAMBR reactor were usually higher than in the CAS reactor, indicating that under anaerobic conditions the APs were accumulated in the digested sludge.

In the work by Bellet et al. (2012), extracts of raw wastewater were tested for agonist and antagonist activities on estrogen, androgen, progesterone, mineralocorticoid and glucocorticoid receptors. Authors detected strong estrogenic activities that correlated well with the concentration of natural estrogens (estrone, estriol and 17 beta-estradiol) measured by chemical analysis. Based on the results, Authors identified pregnenolone (the precursor to all steroid hormones in humans) as a major endocrine disrupting chemical which accounted for most of the anti-mineralocorticoid activities present in raw wastewater.

The study by Reungoat et al. (2012) investigated the fate of trace organic chemicals (TrOCs) in three full-scale reclamation plants using ozonation followed by biological activated carbon (BAG) filtration to treat WRRF effluents. Estrogenicity was reduced by more than 87% indicating that the transformation products of the estrogenic chemicals lost their specific toxicity potential. Overall, the combination of ozonation and BAG filtration achieved removals of 50% for DOC and more than 90% for a wide range of TrOCs as well as a reduction of 70% of non-specific toxicity and more than 95% of estrogenicity.

Li, Nandakumar et al. (2012) identified metabolism pathways that might be involved in microbial estrogen degradation. *Stenotrophomonas maltophilia* strain ZL1 was used as a model estrogen degrading bacterium and its protein expression level during E2/E1 degradation was studied using quantitative proteomics. Based on the results, Authors hypothesized that *S. maltophilia* strain ZL1 was able to convert E1 to amino acid tyrosine through ring cleavage on a saturated ring of the E1 molecule and then utilize tyrosine in protein biosynthesis.

Zhang, Li, Wu et al. (2012) investigated the photocatalytic removal of EDCs, estrogenic activity, and *E. coli* in secondary effluents from a WRRF in a cylindrical photocatalytic reactor which was evaluated for application at advanced wastewater treatment processes. Filtration was evaluated as a pretreatment method in TiO₂ photocatalysis by comparing the reduction rate of estrogenic activity and *E. coli* in filtered samples with unfiltered wastewater samples. The inactivation rate of *E. coli* increased

significantly after filtration, whereas the estrogenic activity reduction rate only slightly increased.

Bagnall et al. (2012) assessed the influence of ammonia oxidizing bacteria and bulk organic competition during laboratory scale activated sludge treatment under short and long hydraulic retention time (HRT) and SRT conditions with bioreactors which were supplied with synthetic sewage spiked with steroid estrogens with and without ammonia as a nitrogen source. Results revealed that estrogen biodegradation was unrestricted and estrogen could be removed at higher than environmental concentrations following a pseudo-first order relationship. During this study, bulk organic loading appeared not to have any appreciable influence upon estrogen biodegradation.

The photocatalytic degradation of a mixture of phenol, Bisphenol-A (BPA), and 17 alpha-ethynylestradiol (EE2) spiked in secondary treated wastewater by means of simulated solar radiation over titania suspensions was investigated. Study results indicated that reaction rates increased linearly with catalyst concentration and photon flux, confirming the photo-induced nature of the activation of the catalytic process: likewise, a linear dependence of rate on initial concentration occurred denoting first order kinetics. Degradation in wastewater was slower than in pure water by an order of magnitude, implying the scavenging behavior of effluent's constituents against hydroxyl radicals.

The objective of the study by Cai et al. (2012) was to evaluate the ability of the constructed wetland systems (CWS) to reduce estrogenic and androgenic

hormone concentration in dairy wastewater. Their results demonstrated that CWS can be efficient system for the treatment of hormones in dairy wastewater.

Xu et al. (2012) provided a perspective on the removal of estrogens in municipal WRRFs in China. The extensive investigations on estrogen removal rates in global and Chinese WRRFs and the estrogen biodegradation kinetics results in batch laboratory experiments were reviewed in this study. The effects of estrogen initial concentration and nitrifying activated sludge were highlighted.

Other Trace Organic Contaminants

There are several other emerging organic contaminants that became a subject of research. Below we tried to summarize couple of important ones:

A study was conducted to evaluate the performance of biotrickling filters (BTFs) for treating low concentrations of *dimethyl disulfide* (DMDS), using different bacterial consortia adapted to consume reduced sulfur compounds under alkaline (pH approximate to 10) or neutral (pH approximate to 7) conditions. Batch experiments demonstrated that DMDS biotransformation favored the neutral pH and sulfate was the main product of the DMDS degradation. On the other hand, 100% removal of DMDS was achieved in BTFs in both alkaline and neutral pHs (Arellano-Garcia et al., 2012).

One study evaluated the aerobic degradation of *sulfanilic acid* (SA) by an acclimatized activated sludge. SA removal efficiency up to 97% was achieved and conversion yielded stoichiometric amounts of sulfate and

ammonium. The ammonium released from the SA oxidation was predominately converted into nitrate, revealing the presence of ammonium-oxidizing bacteria (AOB) in the mixed culture. No apparent inhibitory effect of SA (>500 mg/L) on the nitrification was noted (Chen, Cheng et al., 2012).

1-Naphthylamine wastewater causes severe environmental pollution because of its acute toxicity and carcinogenicity in humans, which makes it difficult to reuse by conventional technologies. In a study, electrochemical catalytic oxidation of 1-naphthylamine in synthetic wastewater in a 150 mL electrolytic batch reactor with Ti/Sb-SnO₂/PbO₂ as anode and steel plate as cathode, where the reaction was assisted by MnO₂ and Sn_{1-x}Sb_xO₂ composite materials as the catalyst and gamma-Al₂O₃ as the carrier (MnO₂-Sn_{1-x}Sb_xO₂/gamma-Al₂O₃) was investigated. It was found that MnO₂-Sn_{1-x}Sb_xO₂/gamma-Al₂O₃ exhibited excellent catalytic activity in the electrochemical degradation of 1-naphthylamine wastewater in 20 min at pH 7.0 (Chen, Yu et al., 2012).

Four series of *dicephalic cationic surfactants*, considered as new antielectrostatic agents have been investigated in order to establish their toxicity and biodegradability. None of the tested surfactants have shown antimicrobial activity against gram-negative bacteria and yeasts at a concentration below 1000 µg/mL, however some of them were moderately active against gram-positive bacteria (*Staphylococcus aureus*, *Bacillus subtilis*). Their toxicity to *Vibrio fischeri* depended upon the alkanoyl chain length with the EC₅₀ values in a range of 2.6-980

mg/L. All of the four tested surfactants were also biodegradable (Pietka-Ottlik et al., 2012).

Tetramethylammonium hydroxide (TMAH) is widely used in manufacturing thin-film transistor liquid crystal displays (TFT-LCD) and semiconductors. Because of its toxicity and nitrogen content, TMAH-containing wastewater has to be properly treated before discharge. Prahas et al. (2012) studied the TMAH adsorption on several commercial activated carbons. They found that activated carbon with micropores showed higher adsorption capacity for TMAH and higher pH was favorable for TMAH adsorption, while there was no TMAH adsorption at pH 4.7±0.2. In addition to high TMAH adsorption efficiency, exhausted carbon could also be easily regenerated by 0.1 N HCl, with no considerable reduction in performance even after five cycles of adsorption-desorption.

p-Nitrophenol (PNP) is widely used as a raw material in several industries, therefore it can be released to the environment, being mandatory the treatment of the PNP-contaminated industrial wastewaters. In a study, the influence of various factors on the catalytic wet air oxidation (CWAO) of a highly concentrated PNP wastewater on Pt and Ru-based catalysts was studied. The results showed that PNP elimination was higher than 90% in most cases, being the temperature the most important operating parameter upon CWAO. Additionally, CWAO increased the biodegradability by more than 50% in most of the tested conditions, being the carboxylic acid fraction the key factor to be taken into account, as the best biodegradability enhancement was observed when this

fraction was the highest. The best CWAO conditions for this pre-treatment to work were determined as 180 °C under stoichiometric oxygen pressure (i.e. 7.6 bar of oxygen partial pressure) with a Ru/TiO₂ catalyst (Martin-Hernandez et al., 2012).

Diphenylarsinic acid (DPAA) is known to be the major contaminant in soils where diphenylchloroarsine and diphenylcyanoarsine were abandoned after World Wars I and II. Contamination of water resources with DPAA is possible. In a study, biotransformation of DPAA under anaerobic soil conditions was investigated. Transformation of DPAA was notably enhanced by addition of exogenous sulfate together with acetate, cellulose or rice straw. Addition of molybdate, a specific inhibitor of sulfate reduction, resulted in the stagnation of DPAA transformation, suggesting that indigenous sulfate reducers play a role in DPAA transformation under anaerobic conditions (Guan et al., 2012).

Biotransformation of *MNX* (*hexahydro-1-nitroso-3,5-dinitro-1,3,5-triazine*) and *trinitroso* (*TNX*) derivatives which are anaerobic transformation byproducts of explosive RDX (*hexahydro-1,3,5-trinitro-1,3,5-triazine*) by an unusual cytochrome P450 XplA that mediates metabolism of RDX in *Rhodococcus rhodochrous* strain 11Y was studied. This study revealed that XplA can transform MNX in both aerobic and anaerobic conditions but TNX in only anaerobic conditions in the presence of RDX and/or MNX, demonstrating the potential for complete RDX removal in explosives-contaminated sites (Halasz et al., 2012).

Stenuit et al. (2012) studied *TNT* denitration under aerobic conditions using two phenazine secondary metabolites excreted by *P. aeruginosa*. They found that pyocyanin facilitates the TNT denitration by catalyzing superoxide-driven nucleophilic attack on the nitro- group of TNT.

Treatment of *phytosanitary effluents* which contain residues of plant protection products from a grape yard was investigated. The aerated biological storage ponds at three wineries, followed by a sand or reed-bed filter, were used for the treatment of the total annual volume of the grape yard effluents and validate the laboratory experiments. The results showed that the biological purification of pesticides by activated sludge is possible by allowing approximately 8 days for biomass adaptation. Stability of purification occurs between 20 and 30 days (Massot et al., 2012).

The electrochemical transformation of the organophosphorous insecticide *chlorpyrifos* (CPF) in wastewater was investigated by Robles-Molina et al. (2012). The oxidation of CPF was carried out in a single-compartment electrochemical flow cell working under batch operation mode, using diamond-based material as anode and stainless steel as cathode. Results showed that CPF is completely removed at the end of treatment time resulting in transient formation of six byproducts including diethyl 3,5,6-trichloropyridin-2-yl phosphate (chlorpyrifos oxon) and 3,5,6-trichloropyridin-2-ol.

Treatment of a pesticide-containing wastewater resulting from phytopharmaceutical plastic containers washing was achieved using sequential biological oxidation

process, leading to almost complete removal of the biodegradable organic carbon fraction; solar photo-Fenton process using CPCs, enhancing the bio-treated wastewater biodegradability, mainly due to pesticides degradation into low-molecular-weight carboxylate anions; and a final polishing step to remove the residual biodegradable organic carbon, using a biological oxidation process. The biological oxidation/solar photo-Fenton/biological oxidation treatment system achieved pesticide removals below the respective detection limits and 79% mineralization, leading to a COD value lower than 150 mg O₂/L (Vilar et al., 2012).

In a study by Zeng, Chin et al. (2012), the abiotic reductive transformation of seven *dinitroaniline* pesticides was investigated in a lake sediment porewaters containing naturally abundant levels of reduced sulfur species and polysulfides and dissolved organic matter (DOM). Target dinitroanilines underwent rapid degradation in PPL porewaters and were transformed into corresponding amine products. Not only sulfur species but other reactive species were responsible for reduction of dinitroanilines.

Degradation of *phenol* in a modified version of the electro-Fenton process with bipolar graphite electrodes and activated carbon was investigated. Results showed that removal efficiency of phenol was 93 at a pH of 3.0, a current density of 3.75 mA/cm², a dosage of ferrous ion of 0.10 mM, and a reaction duration of 60 min. When the reaction time was increased to 180 min, the phenol concentration in the wastewater was below the detection limit. Results indicated that both electro catalytic oxidation and radical oxidation contributed to the degradation of phenol. Modified electro-Fenton process is more efficient

in removing phenol compared to the conventional one (Yang, Liu et al., 2012).

Cyanotoxins such as saxitoxins, geosmin and 2-methylisoborneol (MIB) are present in wastewater due to enrichment of cyanobacteria as a result of high nutrient content. Wastewaters have the potential to proliferate excessive numbers of cyanobacteria due to high nutrient levels. Ho et al. (2012) showed that biological sand filters are efficient in removing geosmin and MIB but not saxitoxins. On the other hand, granular activated carbon filters efficiently removed saxitoxins for a limited time before the saturation of the adsorption sites.

Boehler et al. (2012) investigated the efficiency PAC addition on the removal of various *micropollutants* in pilot and full scale WRRFs. This paper presents the salient outcomes of pilot- and full-scale applications of PAC addition in different flow schemes for micropollutant removal in municipal WRRFs. They found that the sorption efficiency of PAC is reduced with increasing DOC, so effluent treatment should be favored. Adequate treatment of the secondary effluent requires 10 - 20 g PAC/m³ at 5 - 10 g DOC/m³ of effluent, but a PAC dosage of 30 - 40 g/m³ influent directly to the biology tank also reaches MP elimination efficiencies of more than 80% relative to the primary effluent. They concluded that the application of PAC in WRRFs seems to be an adequate and feasible technology for efficient micropollutant elimination (>80%) from wastewater comparable with post ozonation.

Zeolites are natural adsorptive media that are alternative to activated carbon in wastewater treatment. In a study, the adsorption capability and mechanism of

hexadecyltrimethylammonium modified zeolite, which was synthesized from coal fly ash, for the removal of ionizable phenolic compounds (phenol, p-chlorophenol and BPA, with different pK_a and non-ionizable organic compounds (aniline, nitrobenzene, and naphthalene, with different hydrophobicity) was examined. Modified zeolite showed great ability for the removal of organic chemicals whereas little adsorption by zeolite was observed. Uptake of ionizable compounds depended greatly on pH, increasing at alkaline pH conditions. In contrary, adsorption of non-ionizable chemicals was essentially the same at all pH levels studied. Hydrophobicity of the compounds is the driving force for the removal of these pollutants by the modified zeolite (Xie et al., 2012).

Membrane bioreactors have become very popular in wastewater treatment. Boonyaroj et al. (2012) investigated the treatment of landfill leachate from a solid waste disposal site using a two-stage (anoxic/aerobic) MBR system. The removal of *micropollutants* from landfill leachate along the treatment period of 300 days was monitored. The results indicated that two-stage MBRs could remove organic micropollutant with an efficiency ranging between 50 and 76%. The removal efficiencies varied according to the hydrophobic characteristic of compounds but they were improved during long-term MBR operation without sludge discharge.

Industrial Wastewater

Oil Refinery and Petrochemical Industry. As the oil demand increases, oil companies started to seek for new oil resources. Currently, crudes from older reservoirs

as well as oil sands are used to produce oil to buffer the current demand. Refining older crudes and oil sands introduced new challenges to oil refineries. These older oil resources contain high amounts of oxidized hydrocarbons called naphthenic acids. After desalting process most of the naphthenic acids (NAs) are introduced into wastewater. Generation of process waters contaminated by naphthenic acids is a serious environmental concern associated with refining low grade crudes and processing of the oil sands. This together with the necessity for sustainable use of water highlights the need for development of effective technologies for treatment of refinery wastewaters.

Treatment of NAs by UV/H₂O₂ oxidation was investigated in a study. Relative rate measurements using binary mixtures of model NA compounds confirmed that reactivity favored compounds with more carbons, and also favored NAs with one saturated ring, relative to the corresponding linear NA. However, for model compound with three rings, no increased reactivity was observed relative to monocyclic NA. Therefore, UV/H₂O₂ treatment favors the degradation of NAs with more carbons but less number of rings (Afzal et al., 2012).

Anderson et al. (2012) studied the ozonation of oil sands process-affected water (OSPW) which contains soluble organic compounds (OCs), including naphthenic acids (NAs) and potential toxicity of the wastewater after ozone treatment. Ozonation decreased concentrations of OCs in OSPW however toxicity of treated water to *Chironomus dilutes* and its larvae increased.

The degradation of a model naphthenic acid, cyclohexanoic acid (CHA), and real NAs from OSPW in

the presence of peroxydisulfate ($S_2O_8^{2-}$) and zerovalent iron (ZVI) was investigated. Combined ZVI/ $S_2O_8^{2-}$ treatment achieved 90% NA removal from OSPW at room temperature. Temperature and $S_2O_8^{2-}$ concentration were the main factors affecting the removal efficiency. Increasing temperature $S_2O_8^{2-}$ favored higher removal efficiencies (Drzewicz et al., 2012).

In a study, a circulating packed bed bioreactor was used to elucidate continuous biodegradation of trans-4-methylcyclohexane carboxylic acid (trans-4MCHCA), a mixture of cis- and trans-4-methyl-cyclohexane acetic acid (4-MCHAA), and mixture of these three naphthenic acids. Experimental result revealed that biodegradation of trans-4MCHCA was more favored than two other tested NAs at a residence time of 0.15 h. This suggested that both carbon number and the spatial arrangement of the alkyl side branch influenced the biodegradability of the NAs. (Huang et al., 2012).

Terephthalic acid is a petrochemical that is used principally as a precursor to the polyester PET which is utilized to make clothing and plastic bottles. PETR production creates a wastewater mainly composed of acetate, benzoate, terephthalate and p-toluate. Feng et al. (2012) studied the anaerobic treatability of purified terephthalic acid (PTA) wastewater in a novel, rapid mass-transfer fluidized bed reactor using brick particles as porous carrier materials. The reactor was very efficient, and required a low hydraulic retention time of 8 h to achieve about 70% removal both TA and COD from the high-concentration PTA wastewater. The system also has high resistance capacity to varied organic loading rates.

Treatment of PTA wastewater in two-stage upflow anaerobic sludge blanket (UASB) process was investigated. The two-phase system achieved complete removal of acetate, benzoate, terephthalate except p-toluate, while 38 and 62% of p-toluate originally present in the wastewater were consecutively degraded in the individual stages. Stoichiometric methane production was observed. Acidogenic microorganisms were identified as the key players in the biodegradation of aromatic pollutants in the PTA wastewater (Kim, Woo et al., 2012).

Oil spills and refinery wastewaters are the major source of hydrocarbon contamination in water resources. Jin, Kim et al. (2012) identified microbial players influencing the environmental fate of released polycyclic aromatic hydrocarbons (PAHs) after an oil spill in South Korean tidal flats. They found that *Alteromonas* sp. SN2 and its relatives having the naphthalene dioxygenase enzyme are the key players in PAH degradation and adapted to winter conditions in these contaminated sea-tidal-flat sediments.

Treatment of petrochemical industry wastewaters with high organic matter and aromatic compounds in integrated fluidized bed reactor under anaerobic and aerobic conditions was studied. The system was operated at HRT of 2.7 and 2.2 h in the anaerobic and aerobic reactor, respectively. COD removal in the anaerobic reactor was close to 85% and removals of the COD up to 94% were obtained in the aerobic reactor. High removals of benzene, toluene, ethylbenzene, xylenes, styrene, 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene and naphthalene were achieved in this study (Estrada-Arriaga et al., 2012).

Presence of polyaromatic hydrocarbons and benzene, toluene, ethyl benzene, and xylene (BTEX) compounds in produced water, which is the liquid waste that is generated during coal bed methane and oil explorations, is of particular concern because these contaminants are recalcitrant and slow to degrade. Biological treatment of naphthalene and BTEX in laboratory-scale batch and membrane bioreactors was investigated. It was shown that the removal efficiencies of naphthalene and BTEX were more than 95% with initial concentrations of about 10mg/L in batch reactors. The membrane bioreactor also showed more than 90% COD removal consistently and enabled good effluent quality in terms of solids removal (Li and Goel, 2012).

Removal of naphthalene from water and wastewater by an aquatic plant, the water hyacinth (*Eichhornia crassipes*) devoid rhizospheric bacteria was studied in batch systems. Water hyacinth coupled with natural rhizospheric bacteria was able to remove all naphthalene within 9 days whereas only water hyacinth failed to remove 100% of the naphthalene within the same time period. Therefore, rhizospheric bacteria enhanced the degradation of naphthalene in water and wastewater by aquatic plants (Nesterenko-Malkovskaya et al., 2012).

Olive Mill Wastewater. Olive mill wastewater is characterized by its high phenolic content. Photocatalytic degradation of two phenolic compounds, p-coumaric acid and caffeic acid in olive mill wastewater in a suspended mixture of TiO₂ and PAC (at pH = 3.4 and 8) was investigated. A higher removal rate of contaminants by either adsorption or photocatalysis was obtained at a low

pH (pH 4). Fast removal of the pollutants from the solution over three sequential runs was achieved only when both TiO₂ and PAC were present (Baransi et al., 2012).

Anaerobic treatment of raw oil mill wastewater in was investigated two reactors operated under influent concentrations from 5 to 48 g COD/L and HRT between 10 and 5 days. With an intermittent feeding whenever the reactors showed a severe decay in the methane yield, mineralization of oleate and palmitate improved and removal of resilient phenolic compounds was promoted reaching remarkable removal efficiencies of 81%. The supplementation of a nitrogen source increased the methane yield approximately 5 times (Goncalves, Costa et al., 2012).

Oil mill wastewater was treated in sequential reactor system employing anaerobic digestion followed by electrochemical oxidation. The electrochemical oxidation over RuO₂ based DSA after anaerobic treatment yielded 99% COD, 100% phenols and color removal from the wastewater (Goncalves, Marques et al., 2012).

Ntougias et al. (2012) investigated the treatment of olive mill wastewater by white-rot fungi. Among 39 strains tested 4 of them, *Ganoderma australe*, *Ganoderma carnosum*, *Pleurotus eryngii* and *Pleurotus ostreatus*, were selected due to their comparably higher treatment efficiency. Significant decolorization (by 40-46% and 60-65% for *Ganoderma* and *Pleurotus* spp. respectively) and reduction of phenolics (by 64-67% and 74-81% for *Ganoderma* and *Pleurotus* spp. respectively) from wastewater were achieved by the selected fungal strains.

With the target of achieving the parametric requirements for public waterways discharge or even for reuse in the production process, a bench scale study was undertaken to evaluate the feasibility of a thin-film composite RO membrane (polyamide/polysulfone) for the purification of OMW. Combining operating parameters adequately in a semi-batch operating regime ensured high and sustainable permeate flux, yielding over 99.4% and 98.5% removal efficiencies for the chemical oxygen demand and ionic content respectively, as well as complete rejection of phenols, iron and suspended solids (Ochando-Pulido et al., 2012).

Coking Wastewater. The occurrence, behaviors and fate of 18 PAHs in a coking WRRF were investigated. It was found that the target compounds occurred widely in raw coking wastewater, treated effluent, and sludge. In raw coking wastewater, high molecular weight (MW) PAHs were the dominant compounds, while 3-6 ring PAHs predominated in the final effluent. Fluoranthene, pyrene, chrysene and benzo[k]fluoranthene were dominant in sludge. The process achieved over 97% removal for all the PAHs, 47-92% of eliminations of these target compounds in liquid phase were achieved in biological stage. Adsorption to biomass and biotransformation were the main mechanisms of PAH removal (Zhang, Wei et al., 2012).

In another study, the treatment of coking wastewater by an advanced Fenton oxidation process using iron powder and hydrogen peroxide was investigated. The results showed that higher COD and total phenol removal rates were achieved with a decrease in initial pH and an

increase in H₂O₂ dosage. At an initial pH of less than 6.5 and H₂O₂ concentration of 0.3 M, COD removal reached 44-50% and approximately 95% of total phenol removal was achieved at a reaction time of 1 h. In addition, biodegradation of the coking wastewater after this treatment was significantly improved. Authors indicated that this process may be combined with a classical biological process to achieve effluent of high quality (Chu et al., 2012).

Change of hazardous materials in coking wastewater at different treatment stages (anaerobic, anaerobic/aerobic, anaerobic/aerobic/photo degradation, anaerobic/aerobic/ozone oxidation treatment) and the effects of them on the development of maize embryos and the activity of amylase and protease in maize seeds were investigated. All of the above processes except the one with photodegradation resulted in generation of less toxic effluents (Wei, Zhang et al., 2012).

Nanomaterials

The study by Peng et al. (2012) compared the adsorption of ofloxacin (OFL) and norfloxacin (NOR) on carbon nanotubes (CNTs) to evaluate the role of structural and hydrophobic properties in regulating their adsorption. They observed a significant relationship between single-point adsorption coefficients (K_d) and specific surface area (highly hydrophobic), but not between K_ds and oxygen content. This result suggested that site-specific adsorption was not important but hydrophobic effect may have an important contribution to OFL and NOR adsorption on CNTs.

The effect of sodium dodecylsulfate (SDS) concentration on single-walled carbon nanotubes (SWNT) suspension stability was investigated by Bouchard et al. (2012) with time-resolved dynamic light scattering (TRDLS) initial aggregation studies utilizing both monovalent (Na^+) and divalent (Ca^{2+}) cations. The critical coagulation concentration (CCC) values increased with SDS concentration for the Na^+ treatments, but the Ca^{2+} treatments were less sensitive to SDS concentration changes. Overall, these studies demonstrate that low levels of surfactant were effective in stabilizing and mobilizing SWNTs in environmental media.

Bosch et al. (2012) provided evidence for the capability of *Thiobacillus denitrificans* to anaerobically oxidize a putatively nanosized pyrite particle fraction with nitrate as electron acceptor. In their experiments, nanosized pyrite was readily oxidized to ferric iron and sulfate. The mass balance of pyrite oxidation and nitrate reduction revealed a closed recovery of the electrons.

The antimicrobial activity of Cu_2O , ZnO and NiO nanoparticles supported onto natural clinoptilolite was investigated in the secondary effluent under dark conditions by Hrenovic et al. (2012). They observed that the 24 h of contact the Cu_2O and ZnO nanoparticles reduced the numbers of viable bacterial cells of *E.coli* and *Staphylococcus aureus* in pure culture for four to six orders of magnitude and showed consistent 100% of antibacterial activity against native *E. coli* after 1 h of contact during 48 exposures. Authors concluded that metal oxide nanoparticles could find a novel application in the disinfection of secondary effluent and removal of

pathogenic microorganisms in the tertiary stage of wastewater treatment.

To better understand their fate and toxicity in aquatic environments, Unrine et al. (2012) compared the aggregation and dissolution behavior of gum arabic (GA) and polyvinylpyrrolidone (PVP) coated Ag nanoparticles (NPs) in aquatic microcosms. Their results highlighted the potential complexities of nanomaterial behavior in response to biotic and abiotic modifications in ecosystems, and may help to explain differences in toxicity of Ag observed in realistic exposure media compared to simplified laboratory exposures. Further, Bone et al. (2012) studied the effects of complex environmental media on silver nanoparticle (AgNP) toxicity. They found the presence of plants in the microcosms (with and without sediments) reduced both the concentration of Ag in the water column and toxicity for all Ag treatments. Reductions in toxicity were related to decreased water column concentrations as well as changes in the surface chemistry of the particles induced by organic substances released from the plants.

Badireddy et al. (2012) introduced a novel methodology based on hyperspectral imagery with enhanced Darkfield microscopy for detection, characterization, and analysis of engineered nanoparticles in both ultrapure water and in complex waters, such as simulated-wetland ecosystem water and wastewater. Hyperspectral imagery and Raman spectral analyses of the silver nanoparticles (AgNPs) revealed that the apparent hydrodynamic size of the particle increased while the primary size remained unchanged in the presence of coatings, which is further confirmed by dynamic light

scattering measurements. Similar in size, AgNPs with different coatings exhibited similar spectral color (or peak position) but a red-shift in the peak positions by same amount relative to Bare AgNPs was observed. The review by Levard et al. (2012) discussed the major transformation processes of Ag-NPs in various aqueous environments, particularly transformations of the metallic Ag cores caused by reactions with (in)organic ligands, and the effects of such transformations on physical and chemical stability and toxicity.

Chowdhury, Cwiertny et al. (2012) investigated the contributions of NOM and bacteria to the aggregation and deposition of TiO₂ nanoparticles (TNPs) in aquatic environments. Results revealed that interactions among TNPs, NOM, and bacteria exhibited a significant dependence on solution chemistry (pH 5 and 7) and ion valence (K⁺ and Ca²⁺), and that these interactions subsequently affected TNPs deposition. NOM and *E. coli* significantly reduced deposition of TNPs, with NOM having a greater stabilizing influence than bacteria.

In the study by Lv et al. (2012) the influence of phosphate on the dissolution and microstructural transformation of ZnO NPs was investigated. Transmission electronic microscopy observation showed that the morphology of the particles changed from structurally uniform nanosized spherical to anomalous and porous material containing mixed amorphous and crystalline phases of ZnO and zinc phosphate in the presence of phosphate. Authors concluded that the wide existence of phosphate contamination in water and its strong metal-complexation capability, phosphate-induced

transformations may play an important role in the behaviors, fate, and toxicity of many other metal-based nanomaterials in the environment.

Wang, Westerhoff et al. (2012) operated SBRs with HRT and SRT representative of full-scale biological WRRFs for several weeks to study fate and biological effects of engineered nanomaterials (NMs) including silver, titanium dioxide, and C-60 (fullerene) nanomaterials. Under environmentally relevant NM loadings and biomass concentrations, NMs had negligible effects on ability of the wastewater bacteria to biodegrade organic material. Based on the results, Authors suggested that efficient particle separation processes (sedimentation or filtration) could further improve removal of NM from effluent.

Lombi et al. (2012) investigated the chemical transformation of two ZnO-NPs and one hydrophobic ZnO-NP commercial formulation (used in personal care products), during anaerobic digestion of wastewater. The results indicated that "native" Zn and Zn added either as a soluble salt or as NPs was rapidly converted to sulfides in all treatments. The hydrophobicity of the commercial formulation retarded the conversion of ZnO-NP; however, at the end of the anaerobic digestion process and after post processing of the sewage sludge (which caused a significant change in Zn speciation), the speciation of Zn was similar across all treatments.

The impact of UVA irradiation and humic acid (HA) on deposition of aqueous fullerene nanoparticles (nC(60)) on a silica surface as a surrogate for natural sediments was studied by Qu et al. (2012) using packed column experiments and quartz crystal microbalance with

dissipation monitoring under various solution conditions. Surface oxidation of nC(60) induced by UVA irradiation greatly retarded its deposition due to the increased negative surface charge and hydrophilicity. Dissolved HA, once adsorbed onto the nC(60) surface, also hindered its deposition mainly through steric hindrance forces. Rottman et al. (2012) performed batch studies to show that the NPs were partitioning onto the cellular surface. Their results suggested that electrostatic interactions play a major role in determining NP association with biosolids used in municipal WRRFs.

Chowdhury, Duch et al. (2012) investigated the fate and transport of single walled carbon nanotubes (SWCNTs) from synthesis to environmentally relevant conditions in the aquatic environment. They found that addition of NOM increased the transport of all SWCNTs primarily due to electrosteric repulsion. It was also observed that the synthesis methods resulted in distinctive breakthrough trends for SWCNTs, which were correlated to metal content.

Kiser et al. (2012) evaluated the use of freeze-dried, heat-treated (FDH) activated sludge (AS) in batch experiments for quantifying NM removal from wastewater. While soluble pollutants sorbed equally to fresh and FDH AS, fullerene, silver, gold, and polystyrene nanoparticles' removals with FDH AS were approximately 60-100% less than their removals with fresh AS in their experiments. The study indicated that natural or engineered processes (e.g., anaerobic digestion, biosolids decomposition in soils) that result in cellular degradation and matrices rich in surfactant-like materials (natural organic matter, proteins,

phospholipids, etc.) may transform nanoparticle surfaces and significantly alter their fate in the environment.

Eckelman et al. (2012) employed the USEtox model, an integrated multimedia fate, transport, and toxicity model covering large classes of organic and inorganic substances, to quantify and compare aquatic ecotoxicity impacts over the life cycle of carbon nanotubes (CNTs). The results indicated that the ecotoxicity impacts of nanomaterial production processes were roughly equivalent to the ecotoxicity of CNT releases under the unrealistic worst case scenario, while exceeding the results of the realistic scenario by 3 orders of magnitude. Ecotoxicity from production processes was dominated by emissions of metals from electricity generation.

Yang, Chen et al. (2012) determined the impact of Ag NPs on anaerobic glucose degradation, sludge digestion and methanogenic assemblages. At ambient (22 degrees C) and mesophilic temperatures (37 degrees C), there was no significant difference in biogas and methane production between the sludge treated with AgNPs and the control. The results suggested that AgNPs at moderate concentrations had negligible impact on anaerobic digestion and methanogenic assemblages because of little to no silver ion release.

During simulated primary clarification study by Hou et al. (2012), over 90% of the Ag-NPs remained in the wastewater, indicating that the majority of silver nanoparticles in sewage would enter the subsequent treatment units. During sequencing batch reactor processes, silver nanoparticles were effectively removed in each cycle throughout the 15-d experimental duration. Continuous

input of silver nanoparticles into the wastewater did not significantly alter chemical oxygen demand (COD) removal while NH_4 removal was reduced at the beginning of the SBR experiment but quickly recovered at the later stage of the experiment.

Metals

The cathodic reduction of complex-state copper(II) was investigated by Zhang, Tao et al. (2012) in a dual chamber microbial fuel cell (MFC) for recovery of ammonia-copper(II) complexes from wastewater. The inner resistance of MFC system could be reduced in the presence of ionizing NH_4^+ , however, mass transfer was hindered at higher ammonia concentration. Thermodynamic and electrochemical analyses indicated that the processes of complex dissociation and copper reduction were governed by the ratio of $\text{T}[\text{Cu}]:\text{T}[\text{NH}_3]$ and the pH of solution.

Sanchez et al. (2012) tested domestic wastewater (DW) as a cost-effective carbon-source for the remediation of acid mine drainage (AMD). Higher biodiversity was found in the bioreactors compared with that of the inoculum in their studies. Dominant species found belong to two metabolic groups: fermentative (*Clostridium* spp., *Delftia* spp., *Paludibacter* spp. and *Pelotomaculum* spp.) and sulfate-reducing bacteria (*Desulfomonile* spp., *Desulfovibrio* spp., *Desulfosporosinus* spp. and *Desulfotomaculum* spp.).

Cheng et al. (2012) demonstrated the high potential for the application of a novel self-assembled positively charged nanofiltration membrane, PA6DT-C, in processes such as the recovery of valuable cationic

macromolecules in the bioprocess and pharmaceutical industries or removal of multi-valent cations such as dyes and heavy metals in the paper and pulp, textiles, nuclear, and automotive industries. The PA6DT-C membrane was successful in removal of methylene Blue dye from synthetic dye house wastewaters achieving 98% rejection. Thus, Authors concluded that this new membrane can be used for the recovery of other multi-valent cations including heavy metals.

The study by Lorenz et al. (2012) investigated the release of silver from eight different commercially available silver-textiles during a washing and rinsing cycle. Their results showed that different silver textiles release different forms of silver during washing and that among the textiles investigated AgCl was the most frequently observed chemical form in the washwater.

The effects of the combined coagulation-disk filtration (CC-DF) process on the fouling characteristics and behavior caused by interactions between EfOM and the membrane surfaces of the ultrafiltration (UF) and RO membranes in a pilot plant for municipal wastewater reclamation (MWR) were investigated by Chon et al. (2012). They found that fouling of the UF and RO membranes was dominated by hydrophilic fractions of EfOM (e.g., polysaccharide-like and protein-like substances) and inorganic scaling (e.g., aluminum, calcium and silica). The desorbed UF membrane foulants included more aluminum species and hydrophobic fractions than the desorbed RO membrane foulants, which was presumably due to the residual coagulants and aluminum-humic substance complexes.

Fibbi et al. (2012) investigated the fate of hexavalent and trivalent chromium in a full-scale subsurface horizontal flow constructed wetland planted with *Phragmites australis*. They found that chromium in water-suspended solids was in the trivalent form, thus indicating that its removal from wastewater was obtained by the reduction of hexavalent chromium to the trivalent form, followed by accumulation of the latter inside the reed bed

Vaiopoulou and Gikas (2012) reviewed the published literature on chromium effects on activated sludge, and on the operation of WRRFs. They found that clear conclusions cannot be derived about the critical chromium concentrations that affect activated sludge growth, as the latter is a function of a number of factors. Also, a number of reports from the literature supported that Cr-(VI) was toxic to activated sludge at concentrations above 5 mg/L, while others reported growth stimulation at concentrations up to 25 mg/L.

Mahmoud and Hoadley (2012) investigated the ability of the hybrid ion exchange electro dialysis (IXED) technique for the treatment of acidified copper sulfate solutions simulating rinsing water of copper plating lines. Their experimental results showed that the performance at steady-state of the IXED operation with a layered bed remained modest, because of the small dimension of the bed and notably the metal removal efficiency varied from 25 to 47% depending on the conditions applied. The feasibility of using the IXED in operations for removal of heavy metals from moderately dilute rinsing waters was successfully demonstrated in their study.

Thirteen pilot-scale sludge drying reed bed (SDRB) units were constructed and operated under various settings by Stefanakis and Tsihrintzis (2012) to study fate of heavy metals. They observed that the Heavy metal (HM) accumulation in the residual sludge layer was negligible or low, and was found to increase with sludge layer depth. Plant uptake was low; the below-ground biomass accumulated significantly more HMs compared to the above-ground biomass.

Ke et al. (2012) used a sonication-cryptic growth technique in SBR to reduce sludge yield and then analyzed the accumulation of eight typical heavy metals in sludge, namely, Hg, Cr, Ni, Cu, Zn, As, Cd, and Pd. Their results showed that the addition of heavy metals in influent caused the latter to accumulate in sludge and that each metal exhibited different behavior. Heavy-metal addition dropped sludge bioactivity by 53% and decreased the effluent chemical oxygen demand (COD).

The study by Clara et al. (2012) investigated the occurrence of heavy metals, including cadmium (Cd), nickel (Ni), copper (Cu), selenium (Se), zinc (Zn), in municipal WRRF effluents. They observed that all compounds were removed at least partially during wastewater treatment and for most substances the removal via the excess sludge was the major removal pathway.

In the study by Mokone et al. (2012), the effect of strategies used to manage supersaturation was investigated during copper and zinc sulfide precipitation reactions. Their studies showed the origin of sulfide (biological or chemical) had no significant effect on the efficiency of zinc sulfide precipitation. The results also showed that the

extremely high supersaturation prevalent during metal sulfide precipitation was difficult to control using conventional approaches and that the seeded fluidized bed reactor was not suitable for this application.

Matamoros et al. (2012) analyzed the concentrations of six heavy metals (Pb, Ni, Cu, Cd, Zn and Cr) in the accumulated sludge in a 20-year old sludge treatment reed bed (STRB) in Denmark in order to assess the degradation and fate of these contaminants in a STRB and the relation to sludge composition. Their results showed that the deposited sludge was dewatered to reach a dry matter content of 29%, and that up to a third of the organic content of the sludge was mineralized. The concentrations of heavy metals generally increased with depth in the vertical sludge profile due to the dewatering and mineralization of organic matter.

Jagadevan et al. (2012) tested the degradation ability of a semi-synthetic metalworking fluid (MWF) wastewater by employing a novel hybrid approach to combine the synergistic effects of nano zerovalent iron (nZVI) induced oxidation, followed by biodegradation, specifically for the remediation of recalcitrant components of MWF effluent. Their results showed 78% reduction in chemical oxygen demand (COD) by nZVI oxidation at pH 3.0 and 67% reduction in neutral pH (7.5), and 85% concurrent reduction in toxicity. Reduction in metal toxicity made the nZVI treated effluent more amenable for a second stage biological oxidation step.

The study by Weng and Wu (2012) investigated the adsorption characteristics of Cu(II) onto a low-cost biosorbent, pineapple leaf powder (PLP). Results of batch

adsorption experiments from the study showed that PLP could effectively remove Cu(II) from aqueous solution. They found that the copper adsorption was highly dependent on solution pH, and the removal efficiency was greater at a higher pH and low ionic strength.

In the study by Qiu, Zhang et al. (2012), both Cr(VI) reduction and phenol oxidation induced by polymer-sensitized TiO₂ were investigated under visible light. Study of the reaction mechanism indicated that poly(flourene-co-thiophene) (PFT) acted as a semiconductor and was by itself able to reduce Cr(VI) under visible light irradiation. When coupled with TiO₂, PFT served not only as the electron donor for Cr(VI) reduction, but also as a sensitizer.

Biological Agents

Pathogens. Pathogens in the water environment have long been a concern due to their implications on human health. Therefore, almost every country has very strict regulations to control pathogens. Current studies mainly focus on the development of cost effective and efficient new technologies to control pathogens in water environment. In this section we aim to give a brief summary on novel technologies proposed for pathogen control in 2012.

Chemical aided solar radiation is one of the most studied technologies for cost effective disinfection. Chemicals used to enhance solar disinfection include H₂O₂ (Bichai et al., 2012), sodium percarbonate in combination with either citric acid or copper plus ascorbate (Fisher et al., 2012) and Fenton's reagent (Fe²⁺ + H₂O₂) (Ortega-

Gomez et al., 2012; Polo-Lopez et al., 2012). The outcomes of these studies suggested that chemical aided solar disinfection is promising in the control of pathogens in the water environment. In addition, semiconductor photocatalysis was also suggested as an efficient technology for disinfection of water and wastewater (Robertson et al., 2012). Moreover, Pecson et al. (2012) demonstrated that iron oxide coated sand enhanced the separation of viruses from the water column, and additionally provided a photocatalytic mechanism to promote inactivation of viruses in constructed wetlands. This technology may be used in sand filters to enhance wastewater disinfection in WRRFs.

Han et al. (2012) showed that Fenton oxidation in a sequential combination with coagulation/flocculation can achieve 100% pathogen and organics removal from biohazard wastewater.

Kollu and Ormeci (2012) indicated that presence of particles is the main factor decreasing the efficiency of UV disinfection of wastewater. He et al. (2012) implied that use of high pressure water jet for the pretreatment of wastewater enhances UV disinfection by 1 log unit by decreasing particle content of the wastewater.

Antibiotic Resistance Genes. Residual antibiotic and disinfectants in the wastewater resulted in the development and dissemination of antimicrobial resistance in the environment.

Now, antibiotic resistance genes (ARGs) in the environment are the most serious emerging contaminants in the water environment. An extensive review by Rizzo et al. (2013) has attributed WRRFs as the hotspots for antibiotic

resistant bacteria and genes spread into the environment (Rizzo et al., 2013). Many antibiotic resistant microorganisms identified in WRRFs contain multiple ARGs associated with mobile genetic elements such as integrons, plasmids and transposons which facilitate the dissemination of resistance among microorganisms very easily. Therefore, release of effluents into natural aquatic and terrestrial environments can serve as reservoirs of ARGs, which can be horizontally transferred to human-associated bacteria through water and food webs, and thus contributes to antibiotic resistance proliferation.

In a study, about 2000 strains of the family *Enterobacteriaceae* were isolated from different stages of a municipal WRRF, 12.1% were integron positive. All of the integron positive strains were resistant to at least three unrelated antimicrobials. Regarding the diversity of resistance genes and the number of multiresistant bacteria in the final effluent, municipal sewage may serve as a reservoir of integron-embedded ARGs (Mokracka et al., 2012).

The diversity and distribution of drug-resistant *Escherichia coli* (*E. coli*) strains and their integrons in hospital wastewater and river water were compared. As expected, the prevalence of antibiotic resistance in each *E. coli* subgroup from hospital wastewater was higher than in subgroups from river water. Integrons were found in 78.2% the *E.coli* strains isolated. The prevalence of drug-resistance gene cassettes and diversity of arrays further proved that integrons were important contributors to the widespread occurrence of antibiotic resistance in *E. coli* in aquatic environments (Zhang, Su et al., 2012).

Amaya et al. (2012) found high level of resistance to common antibiotic drugs in the *E.coli* isolates obtained from hospital wastewater and groundwater wells. Genes conferring resistance to beta-lactams were most predominant genes.

Prevalence of multidrug resistant bacteria and resistance genes in the samples from hospital and municipal raw sewage, treated effluent from WRRF as well as lake water and sediment samples obtained close to the WRRF outlet pipe and a remote site close to a drinking water pump were evaluated. It is found that wastewater treatment reduced the total bacterial load up to 78% but the remaining bacteria were extremely multiresistant. ARGs were predominant in the lake and sediment close to the outlet of the WRRF (Czekalski et al., 2012).

Occurrence and identification of extended-spectrum beta-lactamase (ESBL)-producing *Enterobacteriaceae* in environmental water samples were investigated in Congo. About 10% of the 194 isolates were identified as ESBL-producers most of which were co-resistant to aminoglycoside and/or fluoroquinolone antibiotics (De Boeck et al., 2012). In addition, Diwan et al. (2012) identified *E.coli* strains carrying genes conferring resistance to commonly used beta-lactams and quinolones in hospital wastewater in central India.

Comparative genomic analysis made for the IncN plasmids, plasmid groups that are often associated with drug resistance, obtained from a final effluent of a WRRF revealed that, IncN plasmids likely contribute to the dissemination of resistance determinants between

environmental bacteria and clinical strains (Eikmeyer et al., 2012).

Gao, Munir et al. (2012) found significant correlations between numbers of resistant bacteria and genes and antibiotic concentrations in raw influent and final effluent of a WRRF in a study whereas correlations of the quantities of antibiotic resistance genes and antibiotic resistant bacteria with corresponding concentrations of antibiotics in sludge samples were found to be considerably weak.

ARGs of *Pseudomonas* species isolated from freshwater and mixed liquor environments in the Eastern Cape Province of South Africa were assessed. Disk diffusion antibiogram of the *Pseudomonas* isolates from the two locations showed 100% resistance to penicillin, oxacillin, clindamycin, rifampicin and 100% susceptibility to ciprofloxacin and gentamicin with varied percentage resistances to cephalothin, nalidixic acid, tetracycline, and ampicillin. Moreover, most of the strains contained integrons. This study suggests *Pseudomonas* species as important reservoirs of multidrug resistance genes in the environment (Igbinosa et al., 2012). Similarly, prevalence of antibiotic-resistant *Aeromonas* species isolated from two different WRRFs in the Eastern Cape Province of South Africa was evaluated. *Aeromonas* isolates from both locations were 100% resistant to penicillin, oxacillin, ampicillin, and vancomycin. Class 1 integron was present in 20.8% of *Aeromonas* isolates. This study further supports wastewater as potential reservoirs of antibiotic resistance determinants in the environment (Igbinosa and Okoh, 2012).

Li, Wang et al. (2012) investigated the distribution of plasmid-mediated quinolone resistance genes from swine feedlots and their surrounding environment. They found that quinolone resistance genes were abundant in the wastewater and the surrounding wastewater-irrigated soils. Results suggested that swine feedlot wastewater may be a source of plasmid-mediated quinolone resistance genes that could facilitate the spread of antibiotic resistance.

Microbial community change upon the exposure to high levels of OTC antibiotic in activated sludge was investigated in a bench-scale aerobic biological treatment system. After OTC exposure, *Alphaproteobacteria* and *Betaproteobacteria* became the dominant bacteria. In addition, the relative abundance of nine tetracycline resistance genes and four mobile elements (class 1 integron, class 2 integron, transposon Tn916/1545, and pattern 1 insertion sequence common region) significantly increased upon exposure to OTC (Liu, Zhang, Ding et al., 2012).

In another study, Liu, Zhang, Yang et al (2012) showed that tetracycline resistance genes and mobile genetic elements harboring these genes were very abundant in an OTC antibiotic production wastewater treatment system compared to treatment systems.

Treated wastewater irrigation is becoming increasingly prevalent in arid regions of the world, due to growing demand and decline in freshwater supplies. The release of residual antibiotic compounds, AR bacteria, and ARGs from wastewater effluent may result in proliferation of AR in irrigated soil microcosms. Impact of treated

wastewater irrigation on soil antibiotic resistant bacterial and ARG reservoirs was investigated. High levels of bacterial antibiotic resistance were detected in both freshwater and treated wastewater irrigated soils which suggested that antibiotic resistant bacteria were native AR associated with the natural soil microbiome (Negreanu et al., 2012).

Paes et al. (2012) identified a Gram-positive aerobic bacterial population preliminary composed of *Bacillus* spp. in the chlorine tank of a WRRF. In addition to chlorine resistance, all isolates obtained from the community were found to be resistant to at least one of five antibiotics tested. This study showed the potential risk of spreading antibiotic resistance genes in the environment by chlorine-resistant strains of *Bacillus*.

The water environment may represent a key dissemination pathway of resistance elements to and from humans. A study investigating the correlations between ARGs and putative upstream anthropogenic sources in the watershed highlighted that abundance of ARGs in water environment is correlated with the type and amount of anthropogenic activity such as animal feeding operations, WRRFs, and fish hatchery, affecting that watershed (Pruden et al., 2012).

Not only antibiotics but also nanomaterials in water, such as nanoaluminum which can cause oxidative stress on bacteria, can significantly promote the horizontal conjugative transfer of multidrug-resistance genes mediated by the RP4, RK2, and pCF10 plasmids. These findings are important in assessing the risk of nanomaterials to the environment, particularly from water and wastewater

treatment systems, and in the estimation of the effect of manufacture and use of nanomaterials on the environment (Qiu, Yu et al., 2012).

A study investigating the resistance profile in *Pseudomonas* species isolated from untreated hospital effluents revealed that most of the species were multidrug resistant through efflux pumps and carry class 1 integrons. Untreated hospital effluents could be a source of environmental contamination due to discharge of antimicrobial resistant bacteria which can carry integron class 1 and act as a reservoir of resistance genes and have efflux pump systems (Spindler et al., 2012).

About 3456 *E.coli* isolates obtained from 38 sampling sites of the Dongjiang River catchment in the dry and wet seasons were evaluated for antibiotic susceptibility and presence of integrons and cassette array. About 90% of the isolates carried integrons and conferred resistance to at least 3 antibiotics. This study also indicated that bacterial resistance in the catchment was seriously influenced by human activities, especially discharge of wastewater (Su et al., 2012).

Disinfection of WRRF effluent may be an important barrier for limiting the spread of antibiotic-resistant bacteria and ARGs. Potential of UV disinfection to damage four ARGs, *mec(A)*, *van(A)*, *tet(A)*, and *amp(C)*, both in extracellular form and present within a host microorganism was examined. The findings showed that UV is limited in its potential to damage ARGs (McKinney and Pruden, 2012).

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