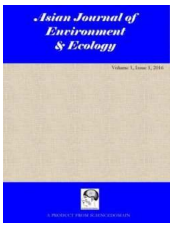

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Pharmaceuticals in Surface Water and Waste Water Treatment Plant Effluent around the World – A Review

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Authors' contributions

This work was carried out in collaboration between all authors. Author KD wrote the final draft and approved the final manuscript.

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ABSTRACT

Pharmaceuticals and personal care products (PPCPs) are "Emerging Contaminants" which widely exist around the word in trace amounts. Evidence by researchers showed that PPCPs can have potential risk on humans and the environment. This paper reviews the occurrence of nine PPCPs for North America, Europe, Asia and Australia based on published literature. The study revealed that industrialized countries in North America and Europe have a higher concentration of PPCPs and with advanced techniques like GC/MS, LC-MS/MS, HPLC/UV and UPLC/MS/MS can precisely analyze the PPCPs from the surface water and waste water effluent.

The paper also reviews technologies for the treatments of removal of those PPCPs. To remove PPCPs in wastewater and surface water, conventional physiochemical methods were not suitable. Advanced methods like reverse osmosis, nano-filtration and constructed wetlands can effectively remove PPCPs. Advanced techniques such as reverse osmosis, nano-filtration and constructed wetlands showed great removal efficiency.

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Keywords: Pharmaceutical personal care products; analytical methods; trace; removal.

1. INTRODUCTION

Pharmaceuticals and personal care products (PPCPs) are one group of "Emerging Contaminants" in the world a name that has been used by the United States Environmental Protection Agency (USEPA) since the 1990s [1]. PPCPs include a diverse collection of thousands of chemical substances, including prescription and over the counter therapeutic drugs, veterinary drugs, fragrances, cosmetics, sun-screen products, diagnostic agents, and nutraceuticals [2].

PPCPs have been present in ecosystems for decades at a trace level concentration i.e., at the µg/l or even ng/l) [3]. They were not studied at first because of the technology limitation for detection. Now with advances in technology, that provide the ability to detect and quantify these "undetectable" chemicals, they can be identified and their effects on humans and the environment can be studied.

PPCPs have been a serious problem as they are widely used and some of them are harmful to the environment and humans. From 1999 to 2000, Barnes et al. [4] conducted a nationwide survey in the US, and identified the occurrence 95 different pharmaceuticals, hormones, and other Organic Wastewater Contaminants (OWCs) in 139 streams in 30 states. The survey showed that at least one OWC was detected in 80% of the streams sampled, with 82 of the 95 analyzed OWCs determined in this study detected in at least one sample. Also, PPCPs can easily enter the environment, for instance through manufacturing plants, effluents of sewage treatment plants, hospital waste water household waste, agricultural and landfill effluent and sludge deposited in landfills and agricultural fields [5-8]. In 2009, Cheri Garvin, CEO of the Leesburg Pharmacy, made a statement about secure and responsible drug disposal that U.S. manufacturers had legally released 271 million pounds of drugs into the environment, and an estimated 250 million pounds of pharmaceuticals and contaminated packaging were discarded by hospitals and long-term care facilities [9]. The objective of this research is to review, document and summarize available data on PPCPs contamination for North America, Europe, Asia and Australia in order to create awareness of an emerging problem that humanity will have to face in the near future.

2. METHODS USED FOR PPCPS ANALYSIS

2.1 Extraction and Concentration

PPCPs appear in the environment at trace concentrations at the ng/l scale, and most analytical methods are not designed to detect compounds at the ng/l scale, so an extraction step is first necessary. Table 1 shows some extraction techniques used for the removal of contaminants. Solid-Phase Extraction (SPE) is the most common method that is used today for the extraction of contaminants.

The basic approach for SPE involves passing the liquid sample through a column, a cartridge, a tube or a disk containing an adsorbent that retains the analytes. After all samples have been passed through the sorbent, retained analytes are subsequently recovered upon elution with an appropriate solvent [16].

2.2 Gas Chromatography

Gas Chromatography (GC) [17] works by injecting a liquid sample into a heated injection chamber, where the compounds are quickly vaporized and swept by a carrier gas (usually helium) onto a silica chromatography capillary column internally coated with a stationary phase. The column is located in an oven where the temperature can be changed. The individual compounds will separate as they flow through the column based on their boiling point, affinity for the stationary phase, and temperature profile

Table 1. Extraction techniques with references

Extraction techniques	Liquid-liquid	Soxhlet	Steam distillation	Solid-phase extraction
References	Yook et al. 1994 [10] Holm et al. 1995 [11]	Bennie et al. 1997, [12] Pryor et al. 2002 [13]	Kubeck and Naylor 1990 [14] Fowler et al. 1998 [15]	Camel 2003, [16]

of the GC oven. A detector then monitors the compounds as they leave the column. Mass Spectrometry (MS) is one of the most useful and common detectors due to its sensitivity and superior selectivity. MS detects [3] analytes based on mass to charge ratio (m/z). Analytes first must be ionized. After ionization, they can be identified based on their fragmentation patterns.

2.3 Liquid Chromatography

Liquid Chromatography (LC) is also a wildly used analytical technique which consists of a solvent pump, sample injector, analytical column, and a detector. Unlike the GC, the separation of analytes in LC occurs in the liquid phase. Usually LC [3] is coupled with mass spectrometry to be more sensitive and selective.

Modern LC-Mass Spectrometers (LCMS) methods use High Pressure Liquid Chromatography (HPLC) instrumentation. In the HPLC instrument, the sample is forced as a liquid at high pressure (the mobile phase) through a column that is packed with a stationary phase composed of irregularly or spherically shaped particles chosen or derivatized to accomplish particular types of separations.

3. CURRENT OCCURRENCE OF PPCPS AROUND THE WORLD

Nine common PPCPs (Carbamezapine, Trimethoprim, Diclofenac, Iopromide, Ibuprofen, Naproxen, Sulfamethoxazole, Gemfibrozil) were selected based on their frequencies showed in the literatures. Table 2 gives the general information of these PPCPs' type, extraction methods, and analytical techniques used for the individual studies. Major data were compiled from the following references: [18-29]. Supplemental data are available from [30-38].

Table 2. Methods for selected PPCPs

Target compound	Type	Extraction	Chromato-graphy	Citation
Carbamazepine	Anticonvulsant	SPE	Derivatize GC/MS HPLC/UV LC/MS LC-MS/MS	Benner et al. 2008, [24], Benotti et al. 2008 [25]. Gomez et al. 2008, [21]. Kim et al. 2007, [22]. Loos et al. 2009, [17], Melo et al. 2013, [24]. Miège et al. 2008, [25], Pal et al. 2010, [26]. Petrović et al. 2003, [28]. Radjenović et al. 2009, [29].
Trimethoprim	Pyrimidine antibiotic	SPE	Derivatize GC/MS UPLC/MS/MS HPLC/UV LC-MS/MS	Benner et al. 2008, [24], Boleda et al. 2011, [25]. Benotti et al. 2008, [25], Gomez et al. 2008, [21]. Kim et al. 2007, [22], Loos et al. 2013, [23]. Miège et al. 2008, [25], Pal et al. 2010, [26] Radjenović et al. 2009, [29]
Diclofenac	Analgesic	SPE	Derivatize GC/MS UPLC/MS/MS HPLC/UV LC-MS/MS LC-MS	Benner et al. 2008, [24], Boleda et al. 2011, [25]. Benotti et al. 2008, [25], Gomez et al. 2008, [21]. Kim et al. 2007, [22], Loos et al. 2009, [23]. Melo et al. 2013, [24], Miège et al. 2008, [25]. Pal et al. 2010, [26], Petrović et al. 2003, [28]. Radjenović et al. 2009, [29].

Target compound	Type	Extraction	Chromato-graphy	Citation
Iopromide	X-ray Contrast Media	SPE	UPLC/MS/MS HPLC/UV LC-MS/MS	Benner et al. 2008, [24], Boleda et al. 2011, [26]. Kim et al. 2007, [22], Loos et al. 2013, [23]. Miège et al. 2008, [25].
Ibuprofen	Analgesics	SPE	Derivatize GC/MS UPLC/MS/MS HPLC/UV LC-MS/MS LC-MS	Benner et al. 2008, [24], Boleda et al. 2011, [26]. Gomez et al. 2008, [21], Kim et al. 2007, [22]. Loos et al. 2009, [23], Melo et al. 2013, [24]. Miège et al. 2008, [25], Pal et al. 2010, [26]. Pérez et al. 2000, [27], Petrović et al. 2003, [28]. Radjenović et al. 2009, [29].
Naproxen	non-steroidal anti-inflammatory drug	SPE	GC/MS UPLC/MS/MS HPLC/UV LC-MS/MS LC-MS GC/ECD	Benner et al. 2008, [24], Boleda et al. 2011, [26], Benotti et al. 2008, [25], Gomez et al. 2008, [21]. Kim et al. 2007, [22], Loos et al. 2009, [23]. Miège et al. 200, [25], Pal et al. 2010, [26]. Pérez et al. 2000, [27], Petrović et al. 2003, [28]. Radjenović et al. 2009, [29].
Sulfamethoxazole	Sulfonamide antibiotic	SPE	Derivatize GC/MS UPLC/MS/MS HPLC/UV LC-MS/MS	Benner et al. 2008, [24], Boleda et al. 2011, [26]. Benotti et al. 2008, [25], Gomez et al. 2008, [21]. Kim et al. 2007, [22], Loos et al.2013, [23]. Miège et al. 2008, [25], Pal et al. 2010, [26]. Radjenović et al. 2009, [29].
Gemfibrozil	Antilipemic	SPE	Derivatize GC/MS UPLC/MS/MS LC-MS/MS LC-MS GC/ECD	Boleda et al. 2011 [26], Benotti et al. 2008, [25]. Gomez et al. 2008, [21], Kim et al. 2007, [22]. Loos et al. 2013, [23], Melo et al. 2013, [24]. Miège et al. 2008, [25], Pal et al. 2010, [26]. Pérez et al. 2000, [27], Petrović et al. 2003, [28], Radjenović et al. 2009, [29].
Ketoprofen	Analgesic	SPE	Derivatize GC/MS LC-MS/MS LC-MS	Gomez et al. 2008, [21], Melo et al. 2013, [24]. Miège et al. 2008, [25], Pal et al. 2010, [26]. Petrović et al. 2003, [28], Radjenović et al. 2009, [29].

The following figures show the occurrence and the concentration ranges in waste water effluents and surface water of the selected PPCPs in various regions of the world during the publication period of the cited literature between 2006 to 2013.

Data from gathered from the literature search show that PPCPs are widely existing around the world at trace concentrations. But some of the PPCPs appear at a very high concentration levels in certain regions of the word. A amount of 0 in the graphs indicate that no data for the particular PPCPs were found in the literature.

Fig. 1a & b shows the range of carbamazepine concentration founded in the research in different area.

Carbamazepine concentration in effluent was reported between 111.2 ng/L to 187 ng/L in North America, 130 ng/L to 290 ng/L in Europe, and 9 ng/L to 157 ng/L in Asia and Australia. The highest concentration was reported to be up to 1240 ng/L in the effluent in Belgium and France shown in Fig 2b, while in the other regions, it was much lower in either effluent or surface water. It was 2.7 ng/L to 113.7 ng/L in North America, 9 ng/L to 157 ng/L in Europe, and 25 ng/L to 34.7 ng/L in Asia and Australia in surface water. For Spain, Belgium and France, it was not reported in the surface water.

The range of Trimethoprim concentration founded in the publicized research in different area areas of the world is shown in Fig. 2a & b.

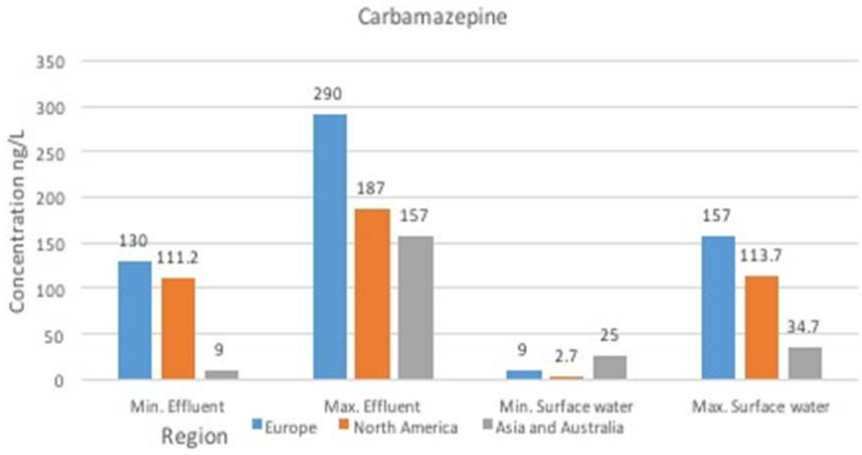

Fig. 1a. Occurrence and concentration of carbamazepine in effluent and surface water around world 2006-2013

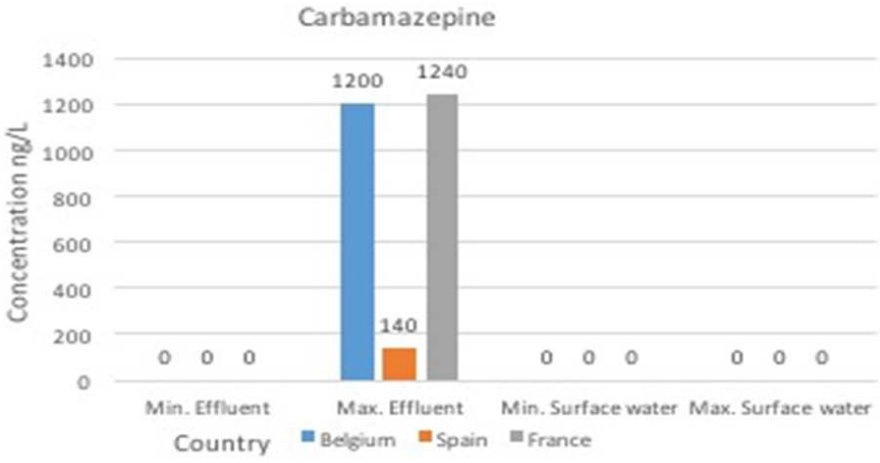

Fig. 1b. Occurrence and concentration of carbamazepine in effluent and surface water around Europe 2006-2013

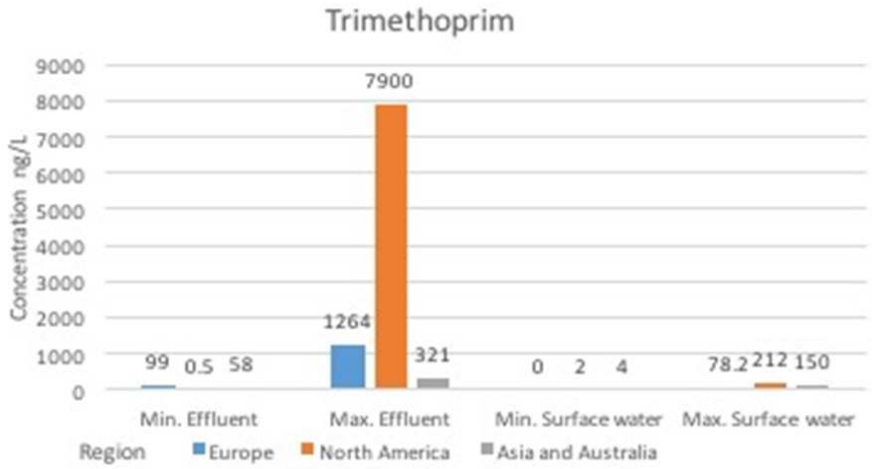

Fig. 2a. Occurrence and concentration of trimethoprim in effluent and surface water around world 2006-2013

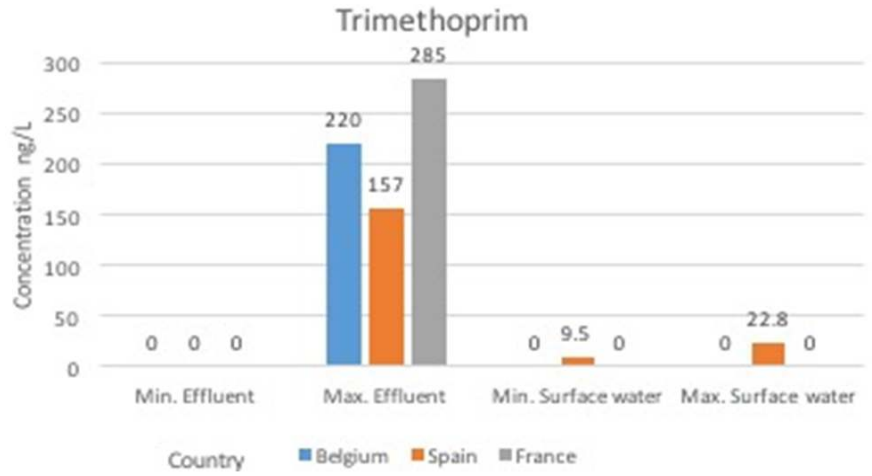

Fig. 2b. Occurrence and concentration of trimethoprim in effluent and surface water around Europe 2006-2013

Trimethoprim concentration in effluent was reported between 0.5 ng/L to 7900 ng/L in North America, 99 ng/L to 1264 ng/L in Europe, and 58 ng/L to 321 ng/L in Asia and Australia. Trimethoprim concentrations were found to be lower in surface water than the concentration in effluent waste water which was between 2 ng/L to 121 ng/L in North America, zero ng/L to 78.2 ng/L in Europe, and 4 ng/L to 150 ng/L in Asia and Australia. For Belgium and France no data was reported.

The Fig. 3a & b shows the range of the Diclofenac concentration founded in the publicized research for the different areas of the world.

Diclofenac concentrations were very low and were not reported in surface water in the countries of Belgium and France (Fig. 3b).

Diclofenac concentration in effluent (Fig. 3a) was reported between 0.5 ng/L to 177.1 ng/L in North America, 460 ng/L to 3300 ng/L in Europe, and 8.8 ng/L to 127 ng/L in Asia and Australia. It was 11 ng/L to 82 ng/L in North America, 21 ng/L to 41 ng/L in Europe, and 1.1 ng/L to 6.8 ng/L in Asia and Australia in surface water. In Spain, the highest concentration detected was 810 ng/L in surface water (Fig. 3b).

Fig. 4 shows the range of Iopromide concentration founded in the research in different

areas of the world. Iopromide was reported in relatively fewr countries. The concentration of Iopromide in the effluent water was much higher than in the surface water. The highest concentration reported was 4775 ng/L in France and 1380 ng/l in Belgium. For surface water a concentration of 74.4 ng/l to 505 ng/l was reported in Spain.

Fig. 5a & b shows the range of sulfamethoxazole concentration founded in the research in different areas of the world.

Sulfamethoxazole was widely existed in the world. Diclofenac concentration in effluent was reported between 5 ng/L to 2800 ng/L in North America (Fig. 5a). concentrations of 91 ng/L to794 ng/L in Europe, and 3.8 ng/L to 1400 ng/L in Asia and Australia. It was 7 ng/L to 211 ng/L in North America, 0.5 ng/L to 41 ng/L in Europe, and 1.7 ng/L to 2000 ng/L in Asia and Australia in surface water. Concentrations in Europe were not reported for surface water for Belgium. The highest in surface water concentration reported was 149 ng/L in Spain (Fig. 5b).

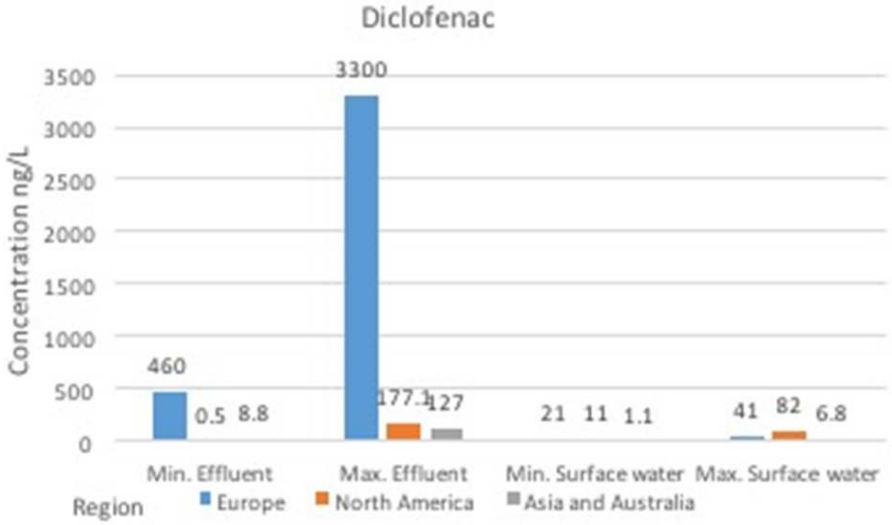

Fig. 3a. Occurrence and concentration of diclofenac in effluent and surface water around world 2006-2013

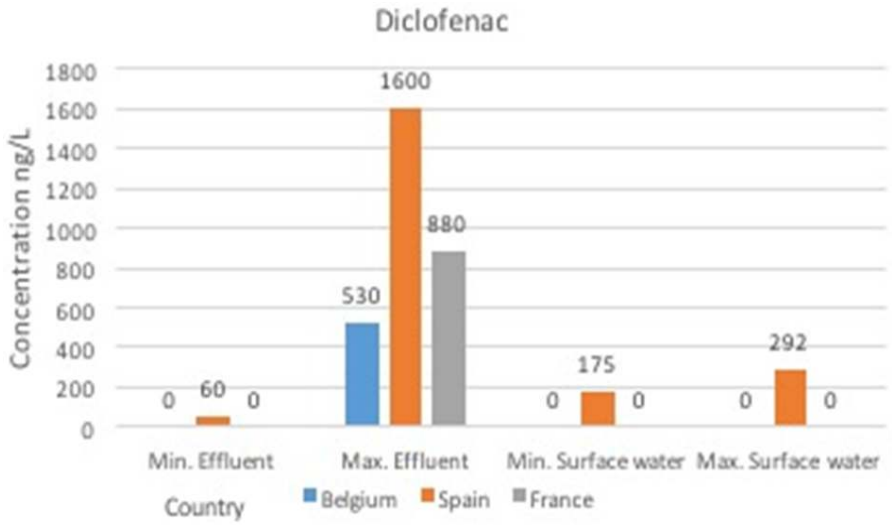

Fig. 3b. Occurrence and concentration of diclofenac in effluent and surface water around Europe 2006-2013

In Fig. 6a & b the range of gemfibrozil concentration founded in the research in different area is shown.

Gemfibrozil in surface water was much lower than in effluent. Many places were not reported in surface water like France and Mexico. Gemfibrozil concentration in effluent was reported between 47.2 ng/L to 180 ng/L in North America, 2 ng/L to 28571 ng/L in Europe, and 3.9 ng/L to 17 ng/L in Asia and Australia. It was 5.4 ng/L to 16 ng/L in North America and 22 ng/L to 248 ng/L in Asia and Australia in surface water.

Fig. 7a & B shows the range of Ketoprofen concentration founded in the research in different areas of the world.

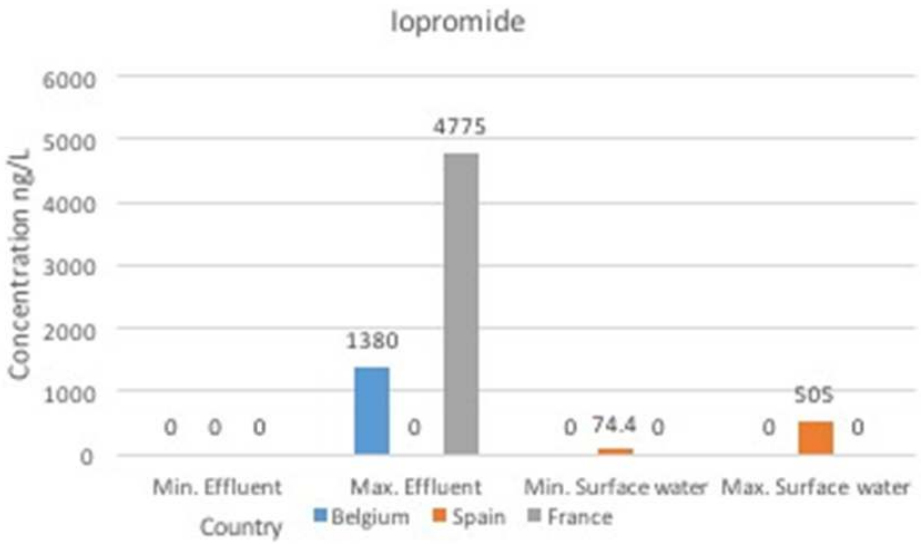

Fig. 4. Occurrence and concentration of Iopromide in effluent and surface water around world 2006-2013

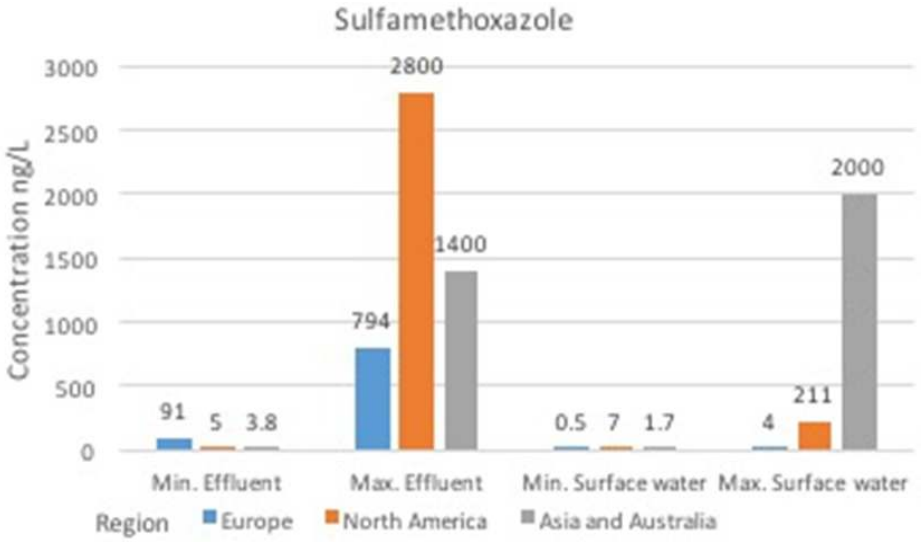

Fig. 5a. Occurrence and concentration of sulfamethoxazole in effluent and surface water around world 2006-2013

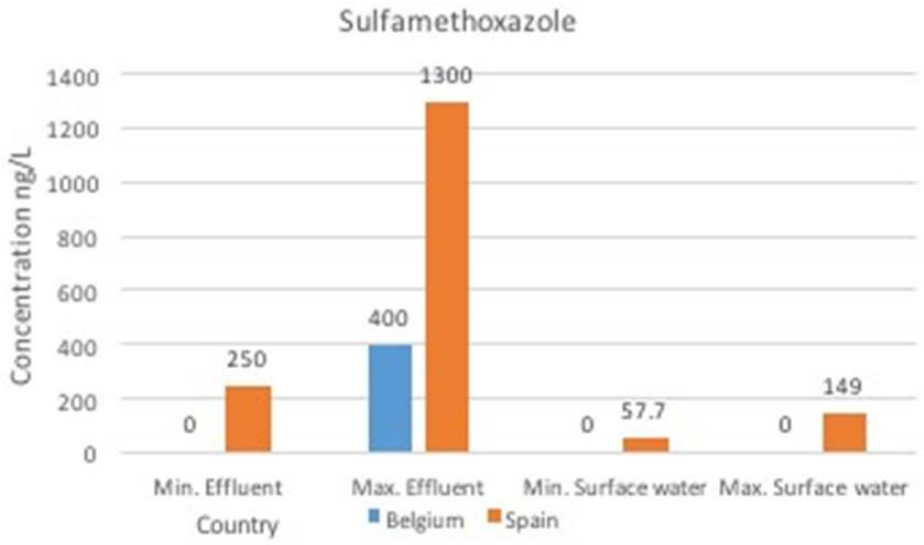

Fig. 5b. Occurrence and concentration of sulfamethoxazole in effluent and surface water around Europe 2006-2013

Ketoprofen in the surface water was very low and not reported in many places such as Spain, France and Mexico. In effluent waters, the highest concentration was in Spain 590 ng/L and in Europe 954 ng/L.

The range of ibuprofen concentration founded in different areas of the word is shown in Fig. 8a &b.

Ibuprofen widely exists in the world. Its concentration in effluent was relatively higher compared to other PPCPs. Ibuprofen concentration in effluent was reported between 220 ng/L to 3600 ng/L in North America, 134 ng/L to 7100 ng/L in Europe, and 65 ng/L to 1785 ng/L in Asia and Australia. The concentration of ibuprofen was much lower in surface water with ranges from 0 ng/L to 34 ng/L in North America, 14 ng/L to 44 ng/L in Europe, and 28 ng/L to 360 ng/L in Asia and Australia in surface water. It was not reported in the surface water of Belgium and France.

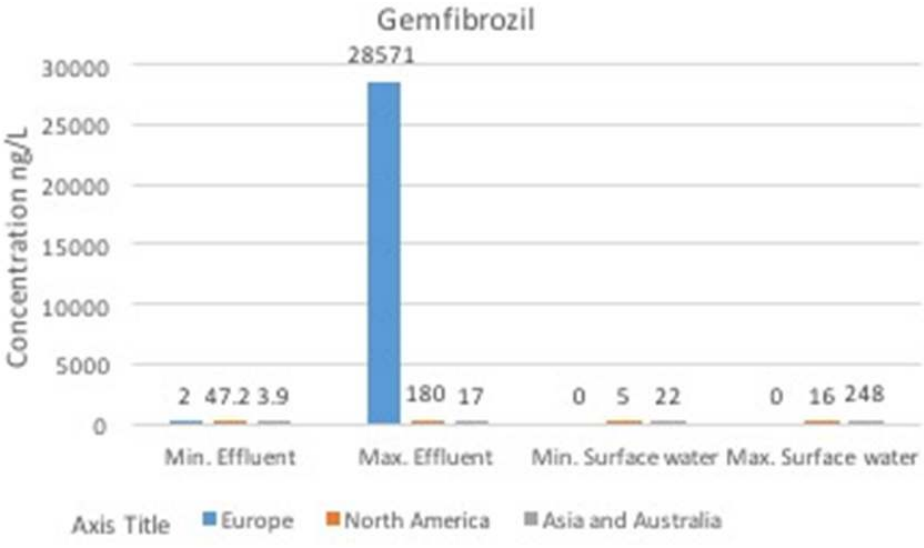

Fig. 6a. Occurrence and concentration of gemfibrozil in effluent and surface water around world 2006-2013

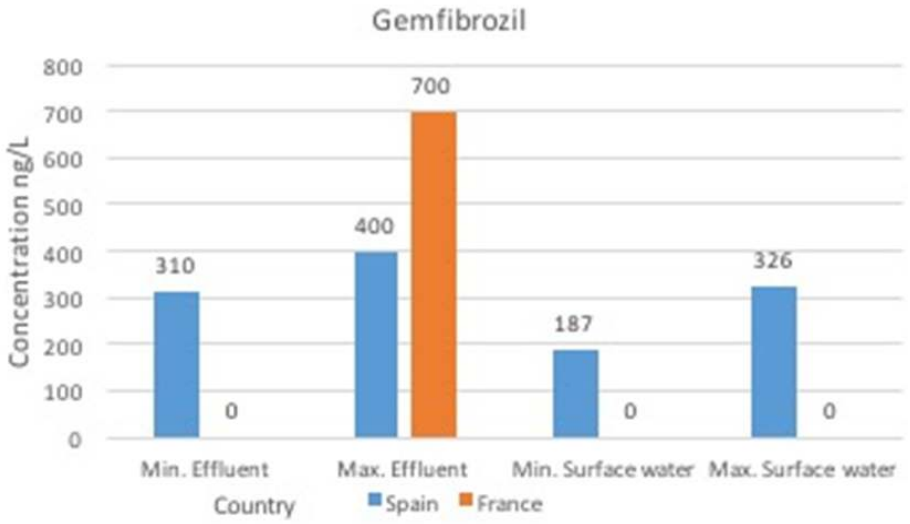

Fig. 6b. Occurrence and concentration of Gemfibrozil in effluent and surface water around Europe 2006-2013

Fig. 9a & b shows the range of naproxen concentration founded in the publicized research in different areas of the world.

Naproxen concentration in effluent was reported between 1 ng/L to 5100 ng/L in North America, 450 ng/L to 1840 ng/L in Europe, and zero ng/L to 135.2 ng/L in Asia and Australia. The highest concentration detected in effluent was in France 17035 ng/L. But the concentration of naproxen was measured in surface water was much lower. It was zero ng/L to 34 ng/L in North America, 0.3 ng/L to 146 ng/L in Europe, and 11 ng/L to 181 ng/L in Asia and Australia. In surface water of Belgium it was not reported.

4. TREATMENT TECHNOLOGIES FOR REMOVAL OF PPCPS

Traditionally treatment processes are not designed to remove the trace amount PPCPs. New methods and technologies should be applied to treat the wastewater containing such substances.

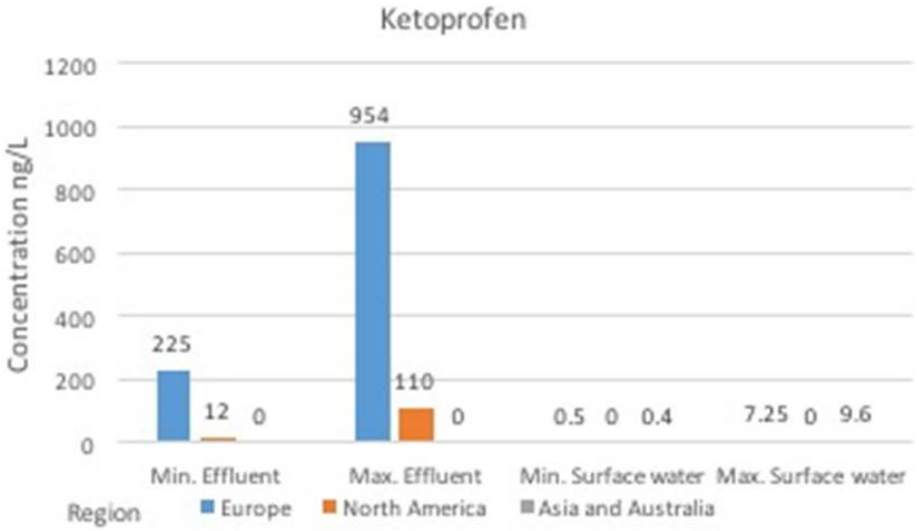

Fig. 7a. Occurrence and concentration of Ketoprofen in effluent and surface water around world 2006-2013

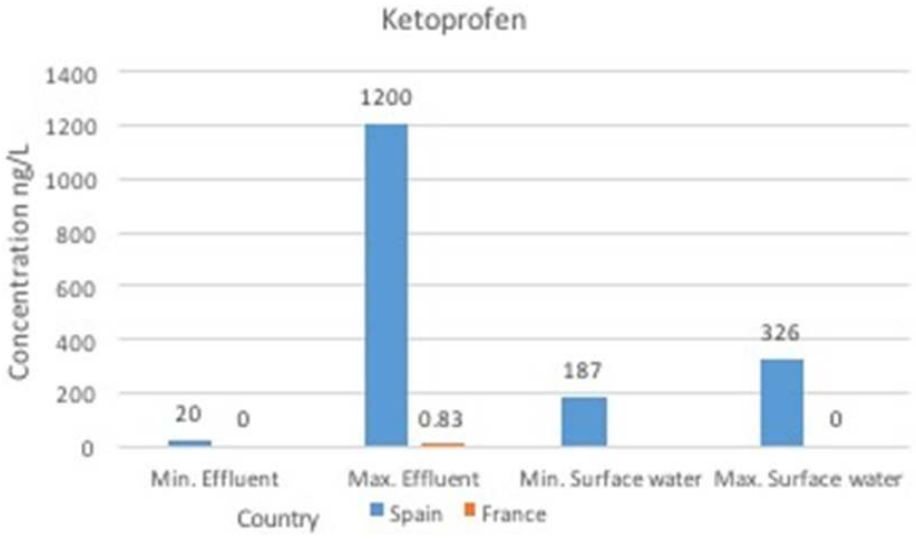

Fig. 7b. Occurrence and concentration of Ketoprofen in effluent and surface water around Europe 2006-2013

4.1 Physicochemical Treatment

Physicochemical treatment as a coagulation–flocculation process was generally found to be unable to effectively remove PPCPs. Addition of activated carbon power, water treatment by chlorination and ozonation can be more effective to remove PPCPs. In 2005, a study conducted by Westerhoff et al. [39], using a bench-scale simulation water treatment plant model and natural waters spiked with 30 pharmaceuticals to 80 different Endocrine disruptors (EDCs) to test the removal efficiency by different treatment processes. 90% of the PPCPs and EDCs were removed through chlorination and ozonation. 50-98% of PPCPs and EDCs were removed by adding 5ml/L powder activated carbon for a four-hour contacting. The treatment processes like coagulation and chemical lime softening only remove less than 25% of the PPCPs and EDCs. Table 3 shows the results for removal performance with different processes.

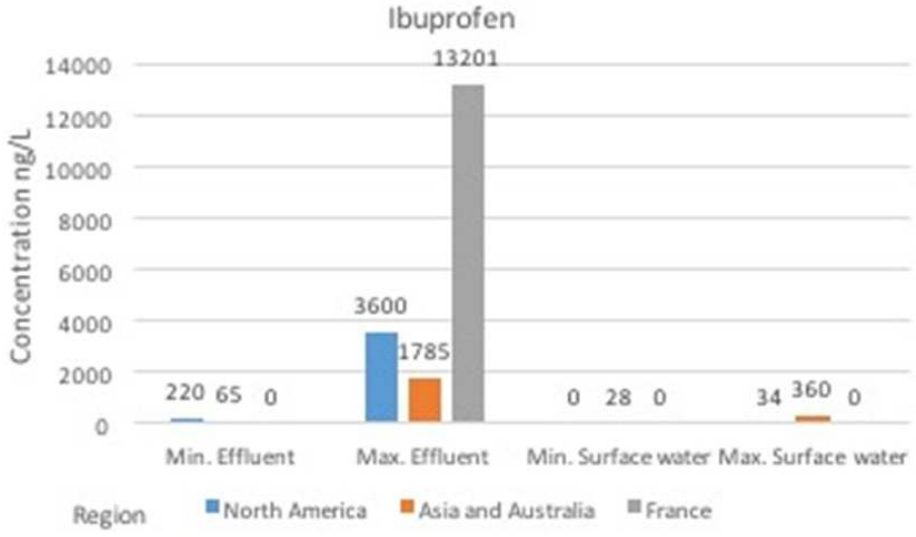

Fig. 8a. Occurrence and concentration of ibuprofen in effluent and surface water around world 2006-2013

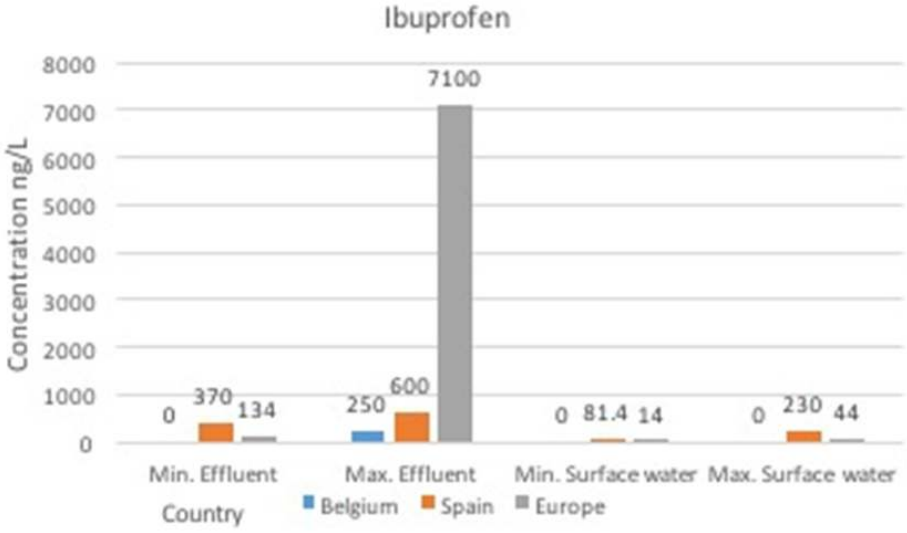

Fig. 8b. Occurrence and concentration of ibuprofen in effluent and surface water around world 2006-2013

4.2 Biological Treatment

Biological treatment removes only a part of a wide range of PPCPs, while some of them are much more resilient to degradation especially, polar ones which are discharged with the effluent [3,22].

In 2005, Urase and Kikuta [40] tested the removal of 10 PPCPs like ibuprofen by activated sludge. At the neutral pH condition, PPCPs showed little tendency for adsorption in the sludge because they appeared as ions and remained in the water phase. While there is an increasing tendency of absorption observed in the lower pH conditions, biological treatment was weak because the limiting stage for removal was not biodegradation, but the transfer of substances from the water phase to the sludge phase.

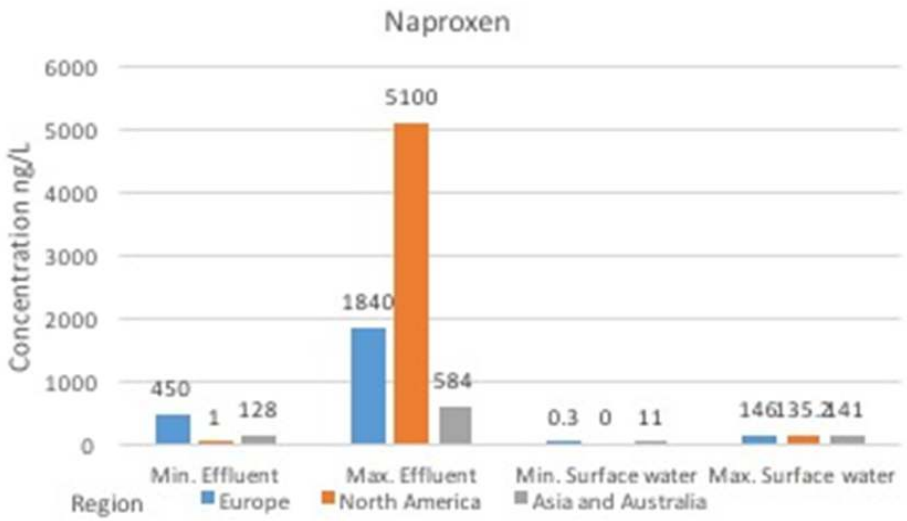

Fig. 9a. Occurrence and concentration of naproxen in effluent and surface water around world 2006-2013

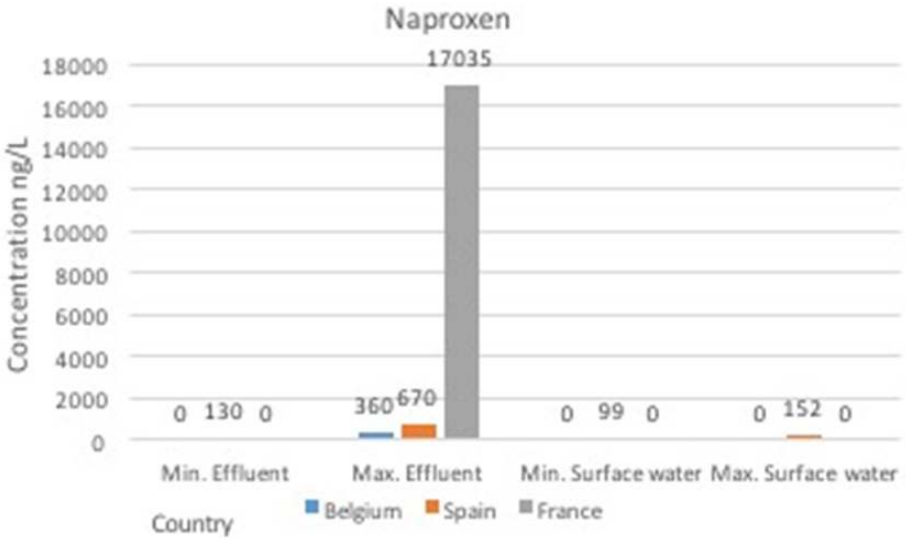

Fig. 9b. Occurrence and concentration of naproxen in effluent and surface water around Europe 2006-2013

Table 3. Removal performance of EDCs by selected treatment processes

Treatment process	Removal performance
Coagulation Add aluminum sulfate and ferric chloride as coagulant	<25% of EDCs/PPCPs removed
Chemical lime softening Add calcium hydroxide and soda ash simulated chemical softening treatments	<25% of EDCs/PPCPs removed
Powder activated carbon (PAC) 5 mg/L of (PAC) with a 4-h contact time.	50-98% removed Higher PAC dosages improved EDC/PPCP removal
Chlorination Add 1200 mg/L chlorine solution	Able to remove >90% for more reactive compounds containing aromatic structures with hydroxide functional groups Not suitable because it produces chlorine by-product.
Ozonation Add 40-50 mg O_3/ L liquid ozone solution	Oxidized methods is similar to chlorination but at slightly higher removal rates Addition of hydrogen peroxide during ozone addition slightly increased the EDCs/PPCPs removal.

4.3 Advanced Treatments

Advanced treatment for removing PPCPs include ultra-violet (UV) photolysis, ion exchange and membrane filtration [41].

UV and ion-exchange can improve the removal of PPCPs, but they were insufficient to be considered as feasible removal options for future application because the removal efficiency was depending on the dosages: the higher the better [28,42]. Membrane filtration technology such as reverse osmosis (RO), or Nano-filtration (NF) has demonstrated itself as a promising alternative for eliminating micro-pollutants [41].

In the study of Urtiaga [43], the Ultrafiltration (UF) removal efficiency was less than 20% for the

majority of the micro-pollutants. Excellent removal efficiencies were achieved with the RO treatment.

4.4 Constructed Wetlands

Constructed wetlands (CWs) for wastewater treatment were engineered designed, constructed to utilize natural processes. They mimic natural wetland systems, utilizing wetland plants, soil, and associated microorganisms to remove contaminants (Like solids, nitrogen, phosphorous, heavy metals, etc.) from wastewater effluents [44].

But with the more concern of PPCPs, CWs are also improved for removing PPCPs. Matamoros and Bayona [45] used subsurface horizontal flow constructed wetlands (SSFWs) to remove he PPCPs, and found that caffeine, salicylic acid, methyl dihydrojasmonate, and carboxyl-ibuprofen were 80% removed, ibuprofen, hydroxy-ibuprofen, and naproxen were 50−80% removed, ketoprofen and diclofenac were difficult to remove. A similar study conducted by Matamoros et al. at 2007 [46] using vertical subsurface-flow constructed wetland (VFCW). Caffeine, salicylic acid, methyl dihydrojasmonate, CA-ibuprofen, hydrocinnamic acid, oxybenzone, ibuprofen, and OH-ibuprofen were more than 95% removed, naproxen, diclofenac, galaxolide, and tonalide were 70 to 90% removed, and carbamazepine were less than 30% removed.

Prado Wetlands, covering 425 acres located at southern California. In 2004, a study [47] of these wetlands found that the site helped reduce levels of ibuprofen and organic chemicals found in pesticides and flame retardants. And new project [48] was conducted at Prado Wetlands channels river water through three ponds. Sunlight and bacteria are used to degrade residues of antibiotics, anti-inflammatories, sex hormones, and other drugs and man-made chemicals.

5. FUTURE WORK AND DIRECTIONS

More studies should be done on PPCPs' risks on human since these risks to humans in both short-term and long-term exposure at low concentration remain poorly understood [5].

Study more of the fate of the PPCPs through animals, plants or even human bodies.

Build a more complete environmental monitoring system including a database to collecting the data and track PPCPs, globally if possible.

6. CONCLUDING REMARKS

In this review, PPCPs exist widely around world at trace level. Some regions show very high level of one of the investigated PPCPs such as Carbamazepine up to 1240 ng/L in Belgium, Trimethoprim, Diclofenac and Sulfamethaxaole of up to 7900 ng/L, 3300 ng/L, and 2800ng/L respectively in North America, Iopromide, Ibuprofen and Naproxen of up to 4775 ng/l, 13201 ng/L and 17035 ng/L respectively in France, Gemfibrozil of 28571 ng/L in Europe, Ketoprofen of 1200 ng/L in Spain. Currently there are many useful techniques for the analysis of PPCPs, for instance, GC/MS, LC-MS/MS, HPLC/UV and UPLC/MS/MS. The EPA method uses a high-performance liquid chromatography combined with tandem mass spectrometry (HPLC/MS/MS) method. All technologies require expensive equipment, highly educated and trained persons for operation, are not standardized and at a high cost of over $150 per sample and compound. In addition PPCPs are not regulated and therefore there is no requirement for testing. For removing the PPCPs in the surface and wastewater, conventional physicochemical treatment as a coagulation–flocculation process is not efficient enough. While some advanced techniques like reverse osmosis (RO), nano-filtration (NF) and constructed showed great removal efficiency in studies and in the application of many regions.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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