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Fate of selected emerging contaminants in wastewater treatment systems

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ABSTRACT

Currently, the occurrence and fate of emerging contaminants in the environment is one of the most studied subject mainly due to their not fully understood ecological effect. This review concerns the occurrence of selected emerging contaminants in raw and treated wastewater and their fate in wastewater treatment plants. The following classes of emerging contaminants were included in these studies: antibiotics, antimicrobial agents, anticolvusnats, nonosteroidal anti-inflammatory drugs, artificial sweeteners, lipid regulating drugs, steroidal hormones, X-ray contrast media, stimulants, insect repellents, plasticizers, and nanoparticles. It was found that the concentration of ECs in influent and effluent of wastewater treatment plants depends on many factors such as geographic location, weather, population density, water supply, treatment system, sampling, and analytical methods. A higher concentrations of most studied contaminants were higher in Asian countries than in European and North America regions. Because it is not possible to remove most of emerging contaminants during conventional treatment process, application of additional treatment method in third treatment step in wastewater treatment plants is required.

Keywords: Emerging contaminants; Wastewater treatment plant; Biodegradation

1. Introduction

Currently, one of the most important environmental problems to be solved is the monitoring of emerging contaminants (ECs) in various environmental matrices and reduction their negative impact on animals and humans health. ECs is a group of various compounds and substances such as pharmaceuticals, artificial sweeteners (ASs) and other food additives, endocrine disrupting chemicals (EDCs), pesticides, industrial by-products, veterinary products, nanoparticles that occur in the environment in low concentrations and can cause an undesirable ecological effect [1]. Several routes are known through which ECs get into the environment such as hospitals, direct discharge of raw municipal wastewater or effluent from wastewater treatment plants (WWTPs) [2], industrial WWTPs, landfill leachate [3], server overflow, and surface runoff from agricultural and urban areas [4] (Fig. 1).

Due to low emerging contaminants removal efficiency in the WWTPs they are still present in the water environment. The concentration of ECs in treated wastewater ranging from ng/L to μg/L and depends on many different parameters such as the structure of ECs and their concentration in the influent, treatment method, and geographical regions [5]. It is unchanging that the main source of ECs in the environment are human living and economic activities.

Although most of these substances occur in the aquatic environment in very low concentrations, they are dangerous because they are characterized by resistance to biodegradation or toxic effects on living organisms. For instance, it is well-known that pharmaceutical compounds manufactured in order to produce a biological response in a pathogenic

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organism can affect the same way non-target entities [6]. Moreover, presence of antimicrobial agents (e.g., Triclosan and Triclocarban) and antibiotics may accelerate development of antibiotic resistance genes (ARGs) and bacteria resistant to antibiotics (ARBs) which shade health risks to humans.

In recent years, more and more publications show that the continues discharge of ECs into the environment by wastewater treatment plants has a negative impact on the environment. For example, numerous scientific studies reported that steroidal estrogens estrone (E1), 17-estradiol (E2), (natural hormones), and 17-ethinyl estradiol (EE2) (synthetic hormone) with unchanged form are discharged into surface water together with effluent from WWTPs. Those hormones have very high biological activity and cause reproductive toxicity at the population level [7]. It was found that that EE2 can modulate the activity of enzymes responsible for neurotransmission and detoxification [8]. Due to documented adverse effects on sensitive aquatic species those steroidal estrogens were included in a European Union (EU) Water Framework Directive (WFD) "watch-list" [9–11]. The aim of the watch-list mechanism was mainly collection of high quality monitoring data of ECs concentration. Results obtained during monitoring period could be used for future substance prioritization and assessment of environmental risk of ECs in European countries [12]. The research also contributed to the development of ECs detection methods in environmental matrices. Many authors emphasize the need for optimization the chromatography methods for detection of ECs representing extremities of physicochemical composition [13,14]. Another concern is that ECs do not appear individual in the environment, so it is not possible to predict unwanted synergistic effect of their mixture.

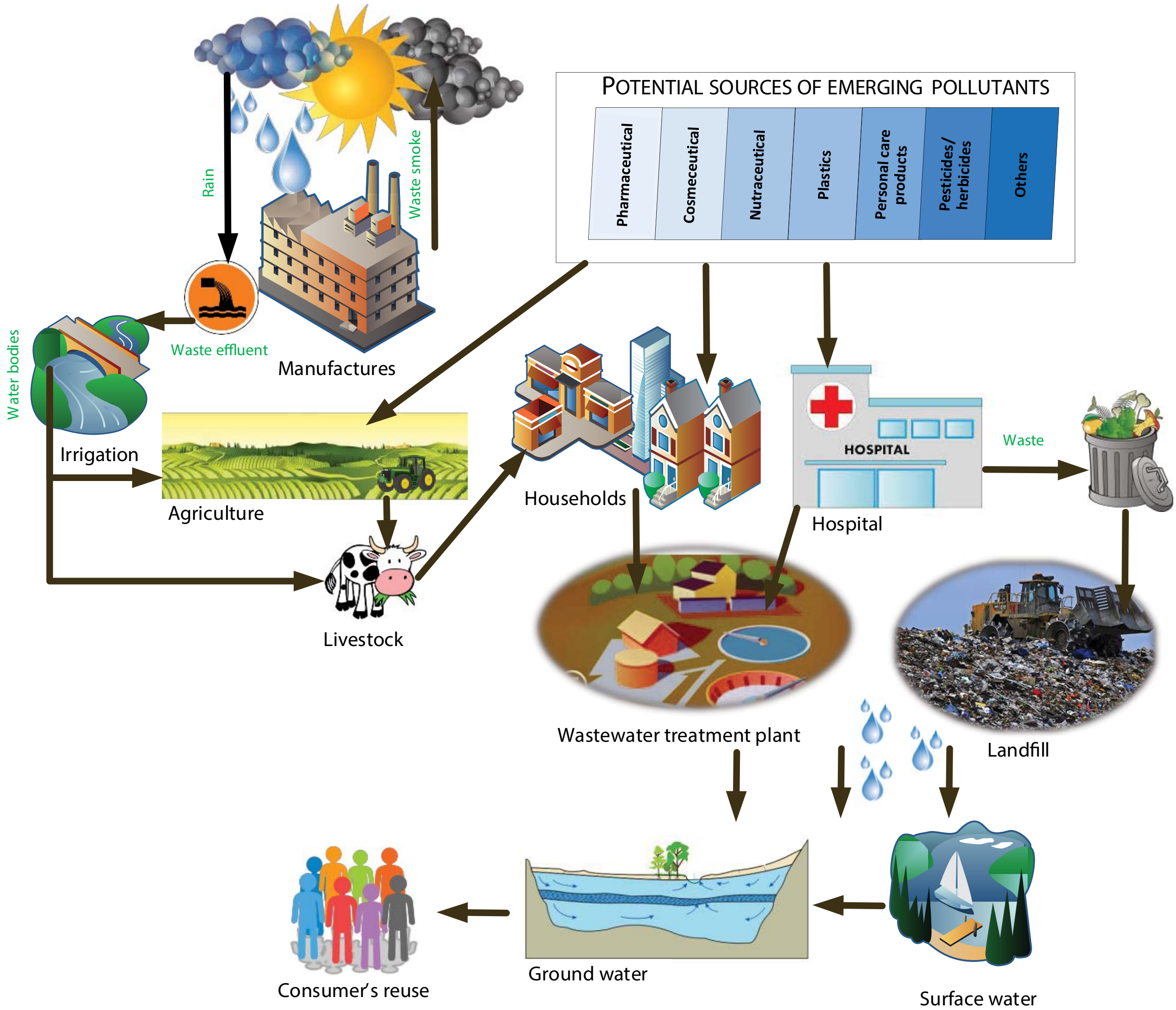

Fig. 1. Potential routes of ECs introduction into the aquatic environment.

Huge variety and amount of substances called emerging contaminants in wastewater and their still not fully known toxicity requires a better understanding of their fate in wastewater treatment systems and ecological impact. Therefore, the objective of this review is summarize current knowledge about occurrence and fate of selected ECs in WWTPs. The occurrence of studied emerging contaminants in influents and effluents of WWTPs is categorized according to each geographical region, that is, Asia, Europe, and North America. The article presents the transformation paths of ECs into wastewater treatment plant and their removal efficiency. Several knowledge gaps and recommendations for further research were also presented in this article.

2. Occurrence of ECs in raw and treated wastewater

Many compounds included to emerging contaminants are not new, but their detection in raw and treated wastewater was possible only in the last 20 y with the development of analytical techniques. The environmental risk of many ECs is not yet know because monitoring technique are still in development [15]. Most of the monitoring data come from WWTPs in Europe, North America, and Asia, mainly from Japan, China, and South Korea, while very limited data was available for other continents [5].

2.1. Occurrence of antibiotics and antimicrobials in WWTPs

Numerous studies indicate that WWTPs are important reservoir of resistance genes due to the presence of bacteria resistant to antibiotics, including human and animal pathogens, as well as antibiotics in the wastewater [16]. The sources of antibiotics are municipal, hospital and industrial wastewater from pharmaceutical industry or from the slaughterhouse. The biological reactor has favorable conditions for bacterial reproduction, therefore the number of bacteria is very large, which promotes the exchange of genetic material through the horizontal gene transfer (HGT) route [17]. Most antibiotics are not degraded in this condition and maintain their activity for a long time. Resistant and multi-drug resistant bacteria, as well as the antibiotics themselves, together with treated wastewater get into the river or soil, and from there they can spread further, posing a real threat to human and animal health [18]. It was found that approximately 50%–90% of antibiotics are excreted with urine and feces, and carried to wastewater treatment plants. Some of antibiotics or their metabolic forms are partially degraded during treatment processes but part of them pass the process unchanged [5,18].

Figs. 2A and B, 3A and B show the concentration of selected antibiotics in influent and effluent of WWTPs located in different geographical regions. Those antibiotics belong to nine classes commonly used by humans and animals, and include: β-lactams, linocosamides, fluoroquinolones, sulfonamides, macrolides, tetracycycline group, reductase inhibitors, amphenicols, and glycopeptides. Interpretation of the data presented on figures is difficult because they are achieved from different treatment systems, and are based on different type of wastewater samples and detection methods. Nevertheless, some trends are noticeable, for example macrolides, trimethoprim, fluoroquinolones, and sulfonamides were detected in raw wastewater as well in effluent of WWTPs worldwide. Many authors reported that the main factors affecting the concentration of antibiotics in wastewater are [5]: usage patterns in each country, water consumption, sewer system, and degradation efficiency in WWTPs.

For example, although β-lactam antibiotics are very widely used, their concentration in effluent is on a very low level because their degradation in wastewater treatment system is high [19]. Taking into account geographical regions it could be assumed that in most Asian countries the concentration of antibiotics in treated wastewater tend to be higher than in North America and Europe [5,20].

Antifungal and antimicrobial agents (thiabendazole, miconazole, triclosan, and triclocarban) have the same influence on development of ARGs in aquatic systems as antibiotics. Those agents are widely used in household products such dermal creams, shampoos, shower gels, toothpaste, soaps, and therapeutic products for elimination fungal infection. The concentration of selected antifungal and antimicrobial agents in influent and effluent of WTTP in different geographic regions is shown in Figs. 2B and 3B. It can be observed that concentration of triclosan and triclocarban are very often higher than considered as predicted no effect concentration (PNECs) for aquatic organisms and is generally higher in Asian region than in North Americans and European countries.

2.2. Concentration of nonsteroidal anti-inflammatory drugs (NSAIDs) in raw and treated wastewater

NSAIDs are one of the most investigated class of emerging contaminant, including among others diclofenac, codeine, fenoprofen, naproxen, acetaminophen, ibuprofen, ketoprofen, salicylic acid, and indomethacin. They are very commonly used pharmaceuticals as painkillers and anti-inflammatory drugs and their concentration in raw wastewater can reach several 100 μg/L [21,22]. Figs. 2C and 3C show significant fluctuations in concentration of NSAIDs in influent and effluent of WWTPs depending on the geographical region. It can be associated with difference in use patterns in individual countries, size of population, climate condition, and sampling procedure. It was found that concentration of NSAIDs in raw wastewater is high mostly in a highly urbanized region [23]. Analyzing the data in the figures, it can be seen that the concentration of some ECs in raw and treated wastewater was higher than PNECs for aquatic systems, therefore potential long-term risk on this environment can be expected.

2.3. Occurrence of anticonvulsants/antipsychotic drugs and artificial sweeteners in WWTPs

In the years 2005–2008 antidepressant drugs were the most frequently drugs used by person age 18–44 y, and third most popular drug taken by American of all ages [24]. Drugs from this group such as gapapentin, sulpride, and carbamazepine are most often detected in WWTPs [25–28]. The concentration of those ECs varied from below detection limit to 1,000's of ng/L (Figs. 2D and 3D). Very often their concentrations detected in effluent were upper PNECs to aquatic organisms.

Wastewater treatment plants are also the main recourses of artificial sweeteners used in food additives and personal care products [29,30]. As shown in Figs. 2D and 3D the concentration of artificial sweeteners varied from several to several thousand ng/L. Due to a good biodegradation,

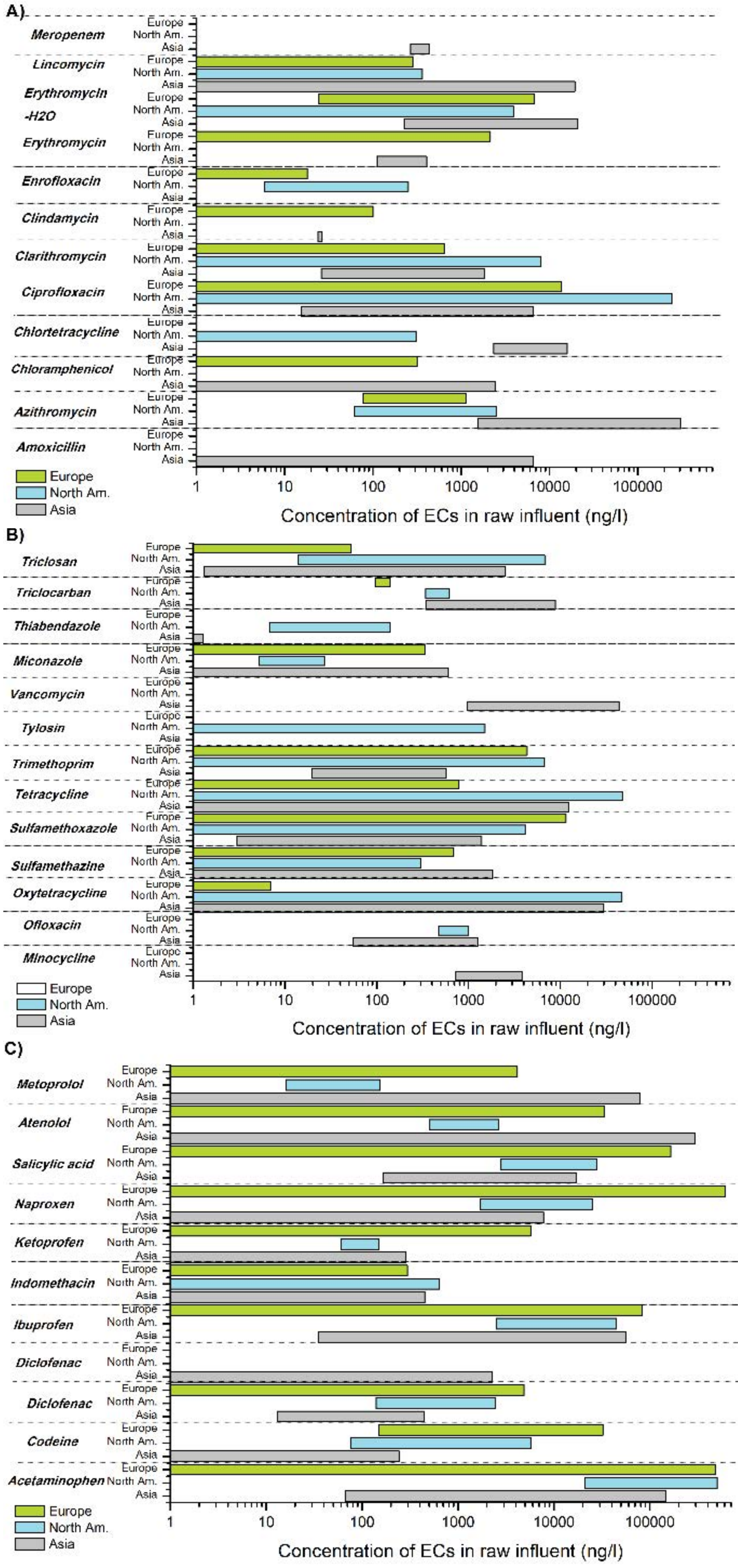

Fig. 2. *Continued*

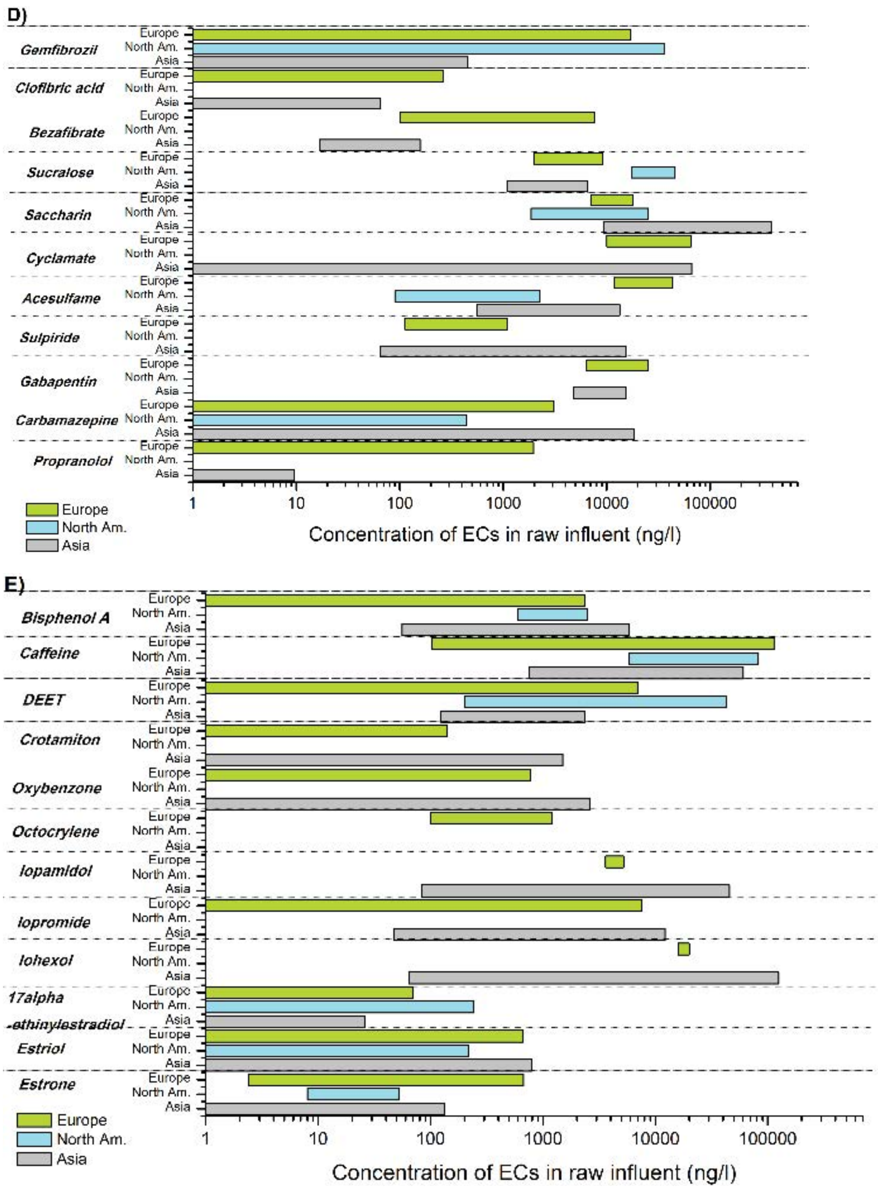

Fig. 2. Concentration range of selected ECs in WWTP influent in Europe, North America, and Asia: (A) Antibiotics, (B) Antibiotics and Antimicrobials, (C) NSAIDs, (D) Anticolvulsants, artificial sweeteners and lipid regulators, and (E) X-ray contrast media, UV-filters, hormones, and other pharmaceuticals and personal care products (adopted from Tran et al. [5]).

cyclamate, and saccharine are practically no detected in effluent, while acesulfame and sucralose are presence in treated wastewater because those compound are resistant to degradation in the wastewater treatment system.

2.4. Occurrence of selected hormones, X-ray contrast media, UV filters, stimulant, anti-iching, insect repellent, and plasticizer in WWTPs

Figs. 2E and 3E show concentration of selected ECs in raw and treated wastewater taking into account geographical regions. One of the most frequently detected ECs in raw wastewater are X-ray contrast media (ICM), which are excreted by human mainly in unchanged forms [31–33]. Large fluctuations in concentrations of ICM in WWTPs are observed, depending on different factors such as land use patterns, sampling procedure, size of population etc. Generally, those factors have influence on the concentration of all ECs in wastewater. Because most of ICM are bio-transformed with high efficiency their concentration in effluent is rather low.

In addition, seasons are an additional factor influencing the presence of UV filters in wastewater. For example, it was found that concentration of octocrylene and oxybenzone in WWTPs is higher in hot weather, which is logical because they are more widely used then in cold weather. Other classes of ECs shown in the above figures, such as bisfenol A, caffeine, and DEET are occurrence both in raw and treated wastewater, and very often the concentration were higher than their PNECs to aquatic organisms [34–36].

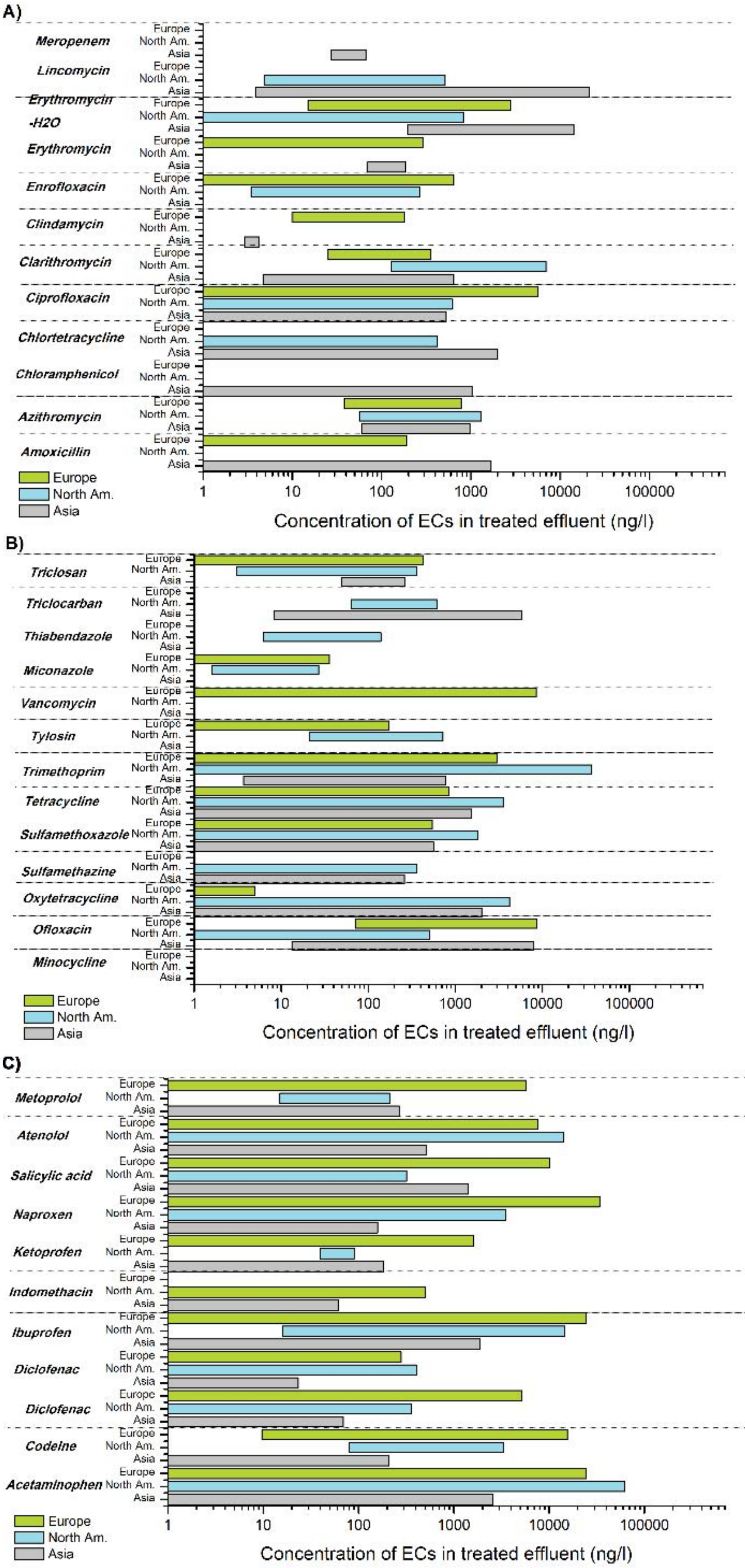

Fig. 3 *Continued*

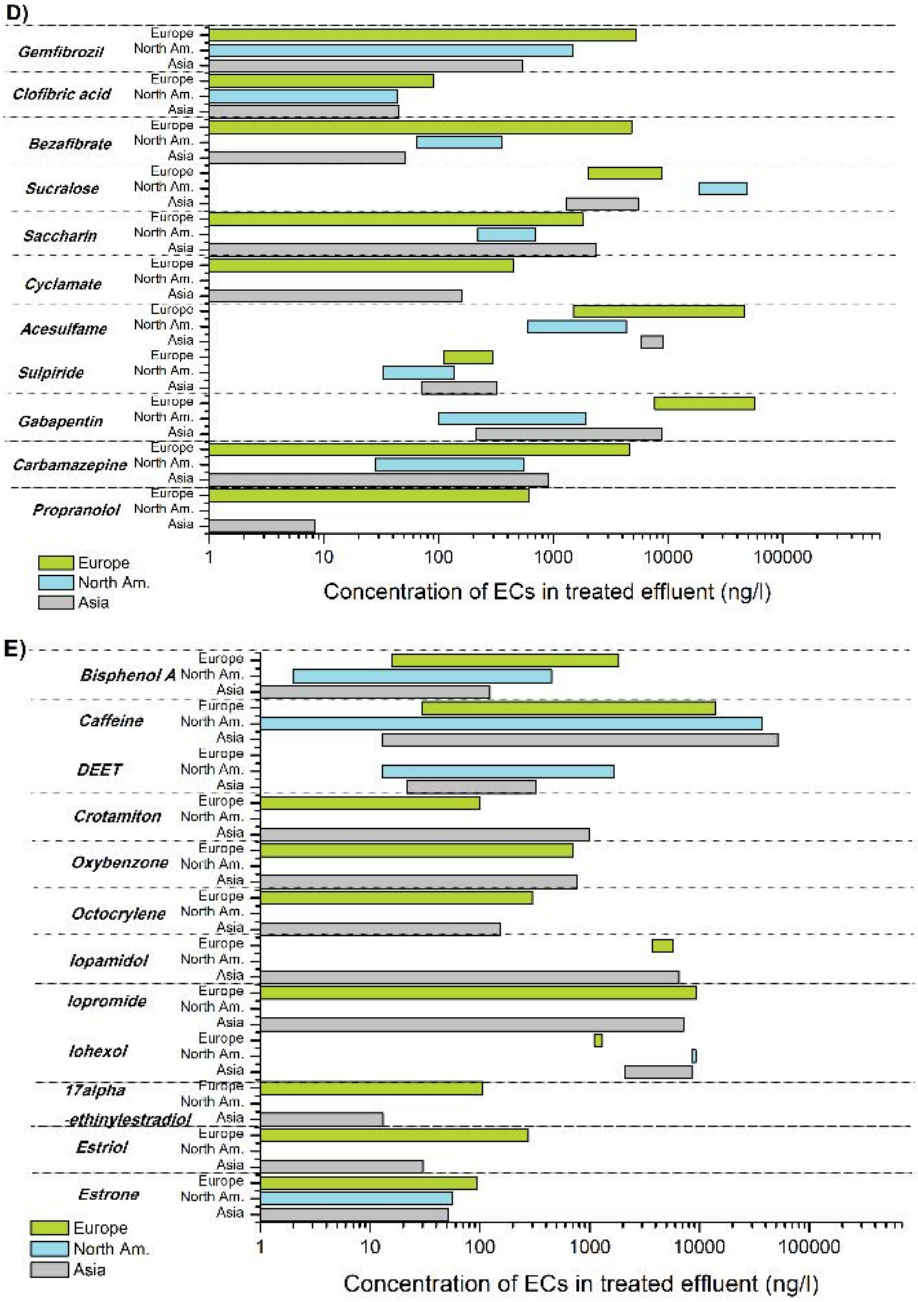

Fig. 3. Concentration range of selected ECs in WWTP effluent in Europe, North America, and Asia: (A) Antibiotics, (B) antibiotics and antimicrobials, (C) NSAIDs, (D) anticonvulsants, artificial sweeteners, and lipid regulators, and (E) X-ray contrast media, UV-filters, hormones, and other pharmaceuticals and personal care products (adopted from Tran et al. [5]).

2.5. Occurrence of nanoparticles in WWTPs

Nanomaterials are specific compounds which are characterized by their regular structure at a molecular level. It is well-known that these are materials which in at least one external dimension are expressed in nanometer, that is, not more than 100 nm, but this varies depending on the material characterization and exhibit some special features often unavailable to traditional materials. Therefore, nanomaterials have been used in many fields of science and industry, and their spectrum usage are very wide and diverse. Particles in the nano-sized range have been present on earth for millions of years. However, recent decades have seen the emergence of manufactured nanoparticles, and the substantial advantages of NPs are now widely recognized. Nanoparticles are used in various fields such as medicine, computers, electronics, the automotive industry, pharmacy, cosmetics, chemical industry, and more than 1,800 consumers products [37]. Commercially important nanoparticles include mainly metal oxide nanopowders or iron oxides. Nanosilver materials are the most widely used NPs while copper NPs seems to be most promising during their low price. With increasing production and application of NPs, their concentration increase in influent and effluent of WWTPs is observed. WWTPs are important for preventing NPs from entering the natural environment. Wastewater treatment process have been observed to remove the majority of NPs in aqueous effluents [38–40]. There is not much information on the concentration of NPs in wastewater treatment plants. For example, it was found that the concentration of TiO_2-NPs in effluents of 10 municipal WWTPs in the USA varied from 52 to 20 mg/L [41]. Other studies have found that in USA and Europe the concentrations of nano-TiO_2, nano-ZnO, nano-Ag in treated wastewater were 1.75–4.28, 0.3–0.441, and 21.0–42.5 ng/L, respectively [42].

3. Fate of ECs in WWTPs

During wastewater treatment process a different efficiency of ECs removal in aqueous effluents have been observed. The fate of a large part of emerging contaminants in WWTPs is still unknown and has been identified as one of the major knowledge gaps for accurate environmental risk assessment. Although ECs may undergo transformation, the primary process of ECs removal from wastewater will be associated with biosolids (biosorption), and the removal by sedimentation and/or filtration [5,43]. Thus, the released part of ECs mainly ended in sewage sludge, which might affect anaerobic digestion of sludge, and then its disposal and reuse. An examples of the fate of NPs in a wastewater treatment plant are shown in Fig. 4. However, reports on the fate of NPs during wastewater treatment process has been scarce in the literature. Most of the studies have focused on the effect of NPs on the microbial growth activity, change in the bacterial community structure, and decrease in the chemical oxygen demand and nitrogen removal. For example, as NPs enter wastewater streams and end up at the treatment plants, they inhibit some bacterial species in the activated sludge and result in a reduction in the efficiency in biological wastewater treatment [44]. During wastewater treatment process and anaerobic digestion of sewage sludge NPs are transformed. The way of transformation depends on NPs properties and place of conversion [45]. For example, AgNPs can be transformed into Ag ion, Ag_2O, or Ag_2S. Several author showed that nanoparticles associate quickly with the particles present in wastewater and then transformed, in the case of AgO, via oxidation and sulfidation. As shown in Fig. 4 more than 80% of NPs are associated with the solid phase of sewage sludge.

It should be emphasized that wastewater treatment plants have been designed primarily to remove organic pollutants and nutrients and not to eliminate ECs. Generally, most of the NCs pass through primary step of treatment process (primary treatment on Fig. 4) without effective elimination from wastewater. Only hydrophobic ECs can be adsorbed onto primary sludge and partially eliminated from wastewater [46–48]. During the secondary treatment ECs are biology degraded (i.e., aerobic or anaerobic) with different efficiency. This degradation can provide to mineralization or incomplete degradation to transformation products via metabolisms and co-metabolism mechanisms. Many studies indicate co-metabolism as the main path for biodegradation of ECs and their resistant or toxic impact on microorganisms [49–53]. It means that energy obtained during biodegradation of most of ECs is not sufficient for microbial growth and generation of enzymes involved in biodegradation processes. Moreover, degradation efficiency is correlated with process parameters like carbon and nutrient sources as well as presence of microorganism which provide co-metabolic degradation. Some of the ECs can be removed from wastewater also via volatilization

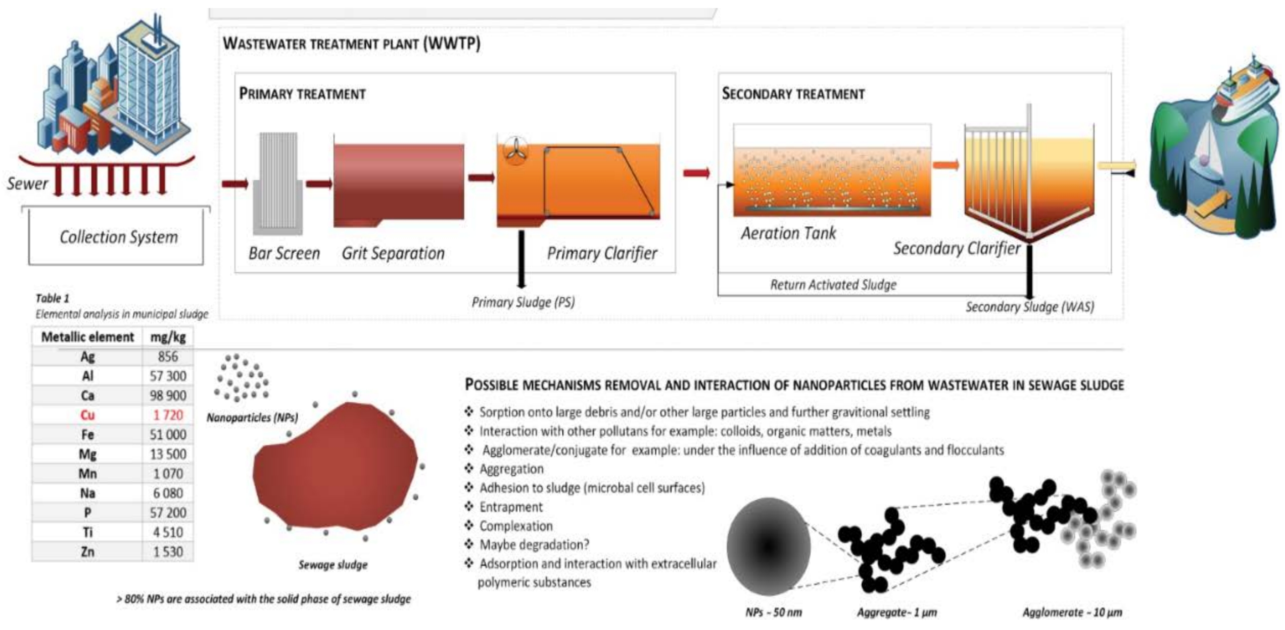

Fig. 4. Fate of selected NPs in WWTP.

where volatile emerging contaminants are transferred to gaseous phase [54].

4. ECs removal efficiency

As can be seen in Fig. 5 as well as in Table 1, a removal efficiency of selected ECs varied significantly, from 0% to 100% in full-scale WWTPs and depending mainly on the type of contaminant and treatment system. In the case of such tested emerging contaminants as meropenem, chloretetracycline, amoxicillin, ciprofloxacin, minocycleine, oxytetracycline, tertracycline, ibuprofen, naproxen, salicylic acid, estrone, estrriol, DEET, caffeine, saccharin, and bisfenol, a median removal was over 80% [5]. At the same time, in the case of many ECs, the median removal does not exceed 40% and, as in the case of some antibiotics, NSAIDs, lipid regulators, beta-blokers a negative median can be observed. As many authors emphasize, the effectiveness of these processes depends on many factors, including the types of bioreactors used and process parameters. Membrane bioreactors (MBRs) are recognized as a treatment systems that can remove ECs to a high degree, which is due to better biomass retention efficiency than a conventional activated sludge (CAS) system, retention of contaminants on the membrane alone and longer sludge age (significant effect of nitrifying bacteria) [55,56]. The main process parameters which have influence on ECs removal efficiency in activated sludge process are: age sludge, HRT, share of anoxic,

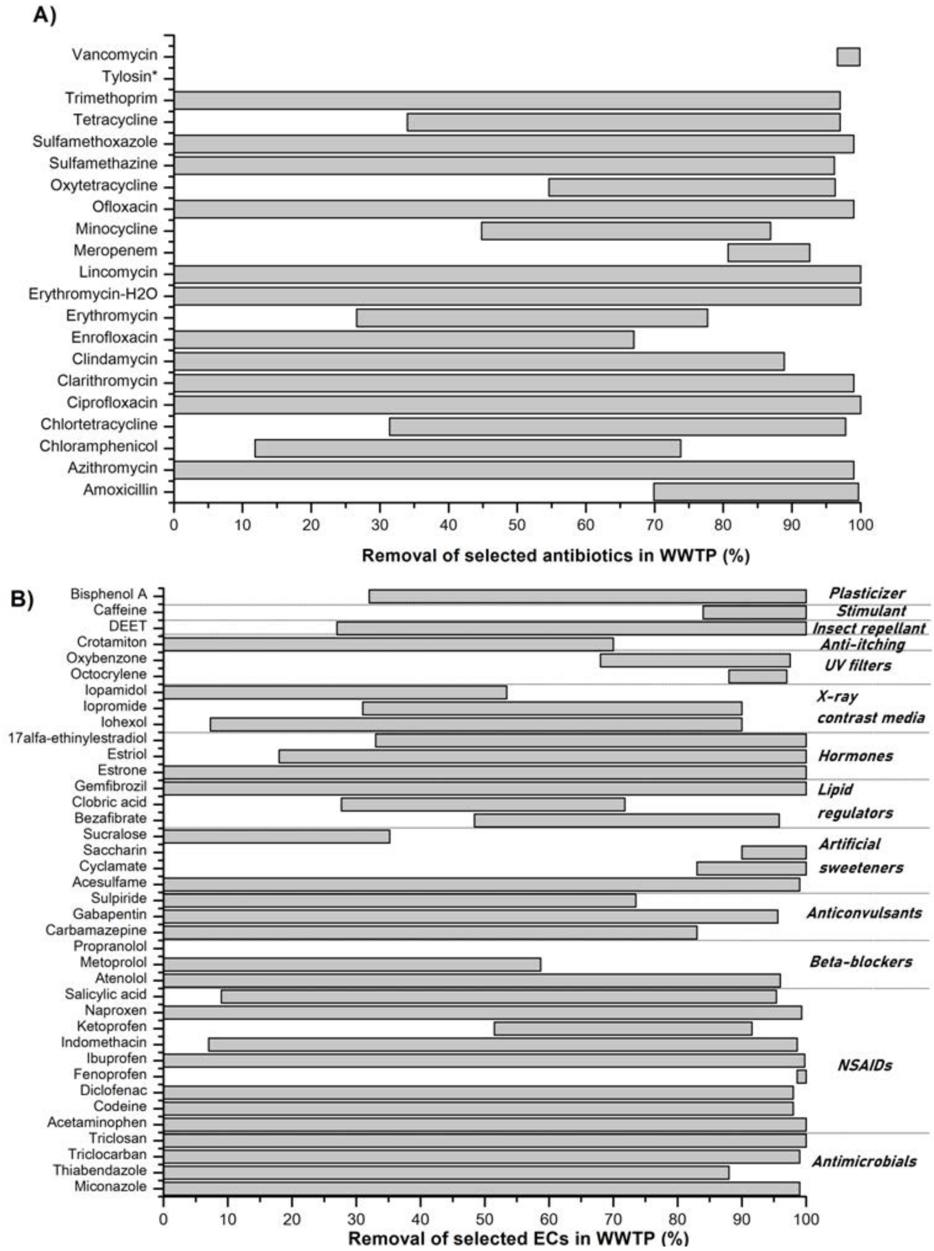

Fig. 5. Removal of selected (a) antibiotics and (b) emerging contaminants in full-scale WWTPs (adopted from Tran et al. [5]).

Table 1
Removal of selected engineering nanomaterials in WWTP (based on [63])

NPs	Characteristic of NMs	Removal (%)	Removal process
Ag	S: 68 nm, 34 nm, 41 nm; C:Y	90	Mixed liquor (batch)
Ag	S: 13 nm; C:N	97	Activated sludge (batch)
Ag	S: 3 nm; C:Y	39	Activated sludge (batch)
Ag	S: 5 nm/30 nm; C:Y	88	Sequencing batch reactors
Ag	S: NA; C:Y	99	Sequencing batch reactors
Ag	S: 21 nm; 29 nm; C:Y	60–90	Mixed liquor (batch)
Au	S: 7 nm (TA), 11 nm (PVP); C:Y	90 (TA); 55 (PVP)	Activated sludge (fresh) (batch)
C_{60}	S: 88 nm; C:NA	88	Activated sludge (batch)
C_{60}	S: 40/90 nm; C:NA	95	Sequencing batch reactors
C_{60}	S: 35; C:NA	90	Activated sludge (fresh) (batch)
CeO_2	S: 50 nm; C:NA	97	Activated sludge (batch)
SiO_2	S: 56 ± 12 nm; 110 ± 17 nm; 65 nm; C:Y	71	Simulated primary (settling) wastewater treatment
SiO_2	S: 56 ± 12 nm; 110 ± 17 nm; 65 nm; C: N	0	Simulated primary (settling) wastewater treatment
SiO_2	S: 50 nm; C:NA	23	Activated sludge (batch)
TiO_2	S: 40 nm; C:Y	91	WWTP
TiO_2	S: 20/1,700 nm; C: NA	91	Sequencing batch reactors

S – size; C – coating; Y – coated; N – uncoated; NA – not available (lack of information); TA – tannic acid; PVP – polyvinylpyrrolidone.

and oxygen zones, and additional chemical reagents used during mechanical treatment processes [57].

Since most xenobiotics are not completely degraded during biological treatment processes, it is often suggested to use other methods to remove these substances from aquatic environment, in particular those based on physico-chemical processes. This group includes coagulation, adsorption on activated carbon photolysis, ozonolysis and advanced oxidation processes (AOPs) [58–60]. Many authors report high efficiency of ECs removal from wastewater using AOP. For example, a highly removal (up to 97%) of endocrine disrupting compounds from WWTP effluent with UV/chlorine AOP system wastewater was observed [61].

However, many factors such as the social and economic parameters, engineering, and environmental impact should be taken into account when assessing the potential for using AOP to remove ECs from wastewater. In studies that took into account all mentioned factors, it was shown that in the case of the studied AOPs, preozonation (H_2O_2/O_3) presented the highest average ranking as compared to

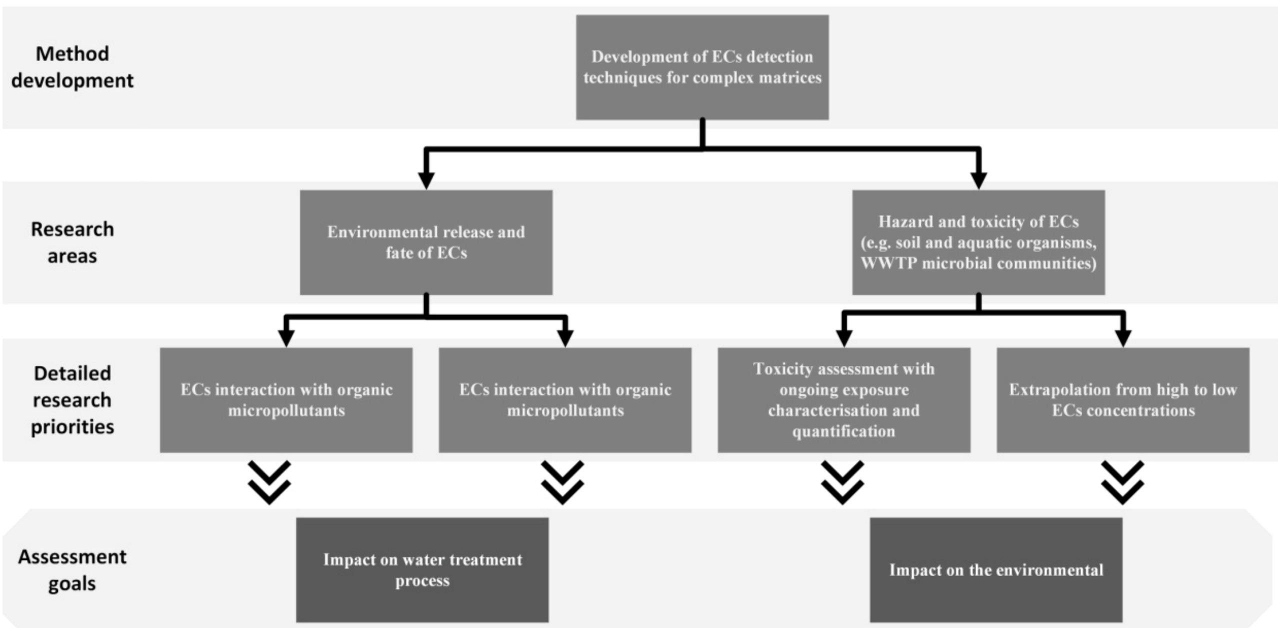

Fig. 6. Recommendation for development of ECs detection method, research areas and priorities.

other analyzed processes (among others UV irradiation, ozonation, photocatalysis, Fenton reaction, and hybrid processes) [62].

5. Research recommendation

Although there are more and more publications considering the fate of EC in wastewater treatment systems, knowledge on this subject is still limited. Improvement understanding of ECs fate in WWTPs is mainly limited by detection method, lack of knowledge about interaction of ECs with other contaminants in wastewater, and their impact on microbial community at bioreactors. Fig. 6 shows the directions of further research development in this area.

The following actions are recommended to better understand the impact of ECs on wastewater treatment systems and the environment:

- Identification and validation appropriate detection methods for ECs for complex matrices, such wastewater and sewage sludge
- Identification of potential sources of environmental release of ECs
- Better understanding of ECs transformation in wastewater treatment systems including the sewer pre-WWTP and different staged of WWTP.
- Assessment the interaction between ECs and others inorganic and organic contaminants in wastewater
- Assessment of ECs toxicity including the toxicokinetics and toxicodynamics

Increasing knowledge in the above-mentioned areas will allow to develop the technologies for removing ECs from wastewater. Moreover, it may helps to reduce their negative impact on the environment.

6. Conclusion

This review concerns the occurrence of selected emerging contaminants in raw and treated wastewater and their fate in wastewater treatment systems. The concentration of ECs in influent and effluent of WWTPs depends on many factors such as geographic location, weather, population density, water supply, treatment system, sapling, and analytical methods. The removal efficiency of selected ECs can vary considerably, from 0% to 100% in full-scale WWTPs and depends mainly on the type of contaminant and treatment system. For example, for such emerging contaminants as meropenem, chloretetracycline, amoxicillin, ciprofloxacin, minocycleine, oxytetracycline, tertracycline, ibuprofen, naproxen, salicylic acid, estrone, estrriol, DEET, caffeine, saccharin, and bisfenol, a median removal could achieve value over 80%. At the same time, many ECs such as some antibiotics, NSAIDs, lipid regulators, and beta-blokers are slightly biodegradable.

The most important factors affecting the ECs removal efficiency in activated sludge process are reactor type, age sludge, HRT, share of anoxic and oxygen zones, and additional chemical reagents used during mechanical treatment processes. The best results were observed for membrane bioreactors mainly due to very high biomass retention efficiency and long sludge age.

Because municipal wastewater treatment plants are primary designed to remove organic carbonaceous and nutrients it is obvious that we will not remove all emerging contaminants from wastewater during primary and secondary treatment. To achieve this purpose, it is necessary to introduce a third step of wastewater treatment and use more sophisticated methods, for example, AOPs. For this purpose, such methods are most often used as (H_2O_2/O_3), UV irradiation, ozonation, photocatalysis, Fenton reaction, and hybrid processes (e.g., UV/chlorine).

It is also very important to monitor concentration of ECs in biosolids, due to a large group of this micropollutants are absorbed into sludge during treatment process. Therefore, there is a risk of negative impact of biosolids on animal and human health after their land application.

Research priorities and knowledge gaps outlined in this article may help to steer future research on improvement ECs treatment efficiency and reduction their negative impact on environment.

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