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Contaminants of Emerging Concern: a Review of Risk Assessment and Treatment Strategies

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Abstract

Contaminants of emerging concern (CECs) such as pharmaceutically active compounds (PhACs), personal care products (PCPs), pesticides, artificial sweeteners (ASWs) in aquatic ecosystems entail a potential risk for the environment, due to their persistent behavior and adverse effect on living organisms during a long-term exposition, even at residual concentrations. Conventional Wastewater Treatment Plants (WWTPs) are not designed to eliminate CECs properly because the treatment technologies are not enough to remove these contaminants, which generates environmental and technological challenges. In this review, the sources of CEC contaminants to aquatic environments have been discussed in detail. Understanding the occurrences and pathways of CECs, their adverse effects on the environment, and removal techniques is a valuable key for the proper maintenance of global ecological health. This scenario was more explored through the harmful impacts of CECs on the environment, including their toxic effects and permissible limits. This review gathers information about CECs occurrences from a global perspective compiling information about their ecotoxicological effects, conventional and advanced treatment methods towards their mitigation. Advanced hybrid treatment techniques such as membrane bioreactor with ozonation, reverse osmosis, and ultrafiltration have shown to be a promising alternative for CECs removal. New advanced oxidation processes with assisted and non-assisted UVC/H_2O_2 systems with TiO_2 photocatalysis were also demonstrated as a good approach to be implemented in the CECs mitigation strategies.

Author Keywords. CECs, Statutory Guidelines, Hybrid Wastewater Treatment, Risk Quotient, Water Management.

Type: Review Article

1. Introduction

Water is a vital resource responsible for maintaining life on earth. Manufacturing industries and agricultural activities need to synchronize with the population continuous growth rate by suppressing their demands which naturally increases water usage. Nevertheless, the wastewater generation also increases, increasing the responsibility of the wastewater treatment plants (WWTPs) to remove the contaminant species and pathogens from the wastewater matrix, thus recovering the quality of the water for its reuse or return to the environment. Over the last decade is become understandable how water scarcity is a danger to the sustainability of human society because of this growing demand scenario (Kleiner 1999; Jéquier and Constant 2009; Bond et al. 2019).

Pharmaceutically active compounds (PhACs), personal care products (PCPs), pesticides, artificial sweeteners (ASWs), X-ray contrast media, flame retardants, and stimulant/illicit drugs are more detected in the aqueous environment in recent decades, as they are crucial for the general needs of the global population and thus keep pace with the growth rate of society (Saidulu et al. 2021). The presence and constant release of those substances in the aquatic bodies seems to be an environmental issue and an emerging concern (Taheran et al. 2018; Wan et al. 2020). The concentration of those substances in the water depends on the usage pattern, per capita water consumption, population density, land use, sewer conditions, environmental persistence, among other reasons (Ali Gamal Al-Kaf et al. 2017; Patel et al. 2019; García et al. 2020). Usually, the average detected concentration of those substances is considerably low (e.g., in the range of ng/L to µg/L) and long-period of exposure to them can cause harmful effects on the ecosystem's health balance (Barbosa et al. 2016). These substances are also called contaminants of emerging concern (CECs) due to the lack of substantial guidelines and toxicological data (Shah et al. 2020).

The hazardous potential of emerging contaminants has been attracting the researchers' attention for a while, due to their impact on freshwater, groundwater, marine environment and atmosphere (Petrisor 2010; Kapelewska et al. 2018; Bilal et al. 2019). To date, an official regulatory methodology, assessing CECs potential toxicity with quantitative measurements, still hasn't been established yet. Only a limited number of studies have been conducted towards a comparative analysis between maximum CECs concentrations reported in specific environment locations, overall harmful impact on selected environments and species and local anthropological data on daily water intake (Nakada et al. 2017; Parida et al. 2021). The Risk Quotient (RQ) methodology (Nika et al. 2020) presents a good estimation on CECs toxic potential and therefore was calculated for a selected group of CECs in this review, considering their lethal concentration (LC_{50}) or 50% effective concentration (EC_{50}) on selected aquatic organisms from different studies.

The conventional wastewater treatment techniques are not enough to remove the CECs, so several studies using different treatment techniques are being carried out (Deegan et al. 2011; Phoon et al. 2020; Mohapatra et al. 2016). A comparative analysis was performed between the concentrations of CECs after a conventional treatment technique has been applied on the wastewater matrix and the maximum CEC concentration stated by the provisional international statutory guidelines (Radley-Gardner, Beale, and Zimmermann 2016; USEPA 2016; Herschy 2012; NHMRC Australian Guidelines 2008).

This review aims on establishing the global occurrences of CECs and their toxic potential against aquatic environment, showing the conventional, consolidated and advanced wastewater treatment techniques towards CECs removal.

2. CEC occurrence in the environment

Contaminants of emerging concern can be introduced into the environment via direct point sources, i.e., industrial effluents, hospital and household sewerage system, or indirectly via a wide variety of other sources, i.e., leaching and runoff during agricultural activities, dumping sites, atmospheric deposition and landfill leachate (Saidulu et al. 2021; Parida et al. 2021; Akhbarizadeh et al. 2020).

Direct point source effluents pass through a wastewater treatment plant in order to remove the contaminants below the regulatory limits before discharge in the environment. Nevertheless, CECs have been reported persisting in the compositions of such treated effluents as well as in drinking water, groundwater and surface water (Samaras et al. 2013; L. Rizzo et al. 2013; Krzeminski et al. 2019; Shah et al. 2020; Tran, Reinhard, and Gin 2018).

The persistence of CECs in the treated effluents are due to the limited design of conventional WWTPs, which are unable to mitigate or eliminate the CECs and their metabolites, inevitably leading to their release as sewage effluents into streams or rivers which have an active biodiversity (Grandclément et al. 2017; Tran, Reinhard, and Gin 2018; Patel et al. 2019).

In addition to conventional wastewater treatment plants releasing CEC through liquid discharges, the sludge formed during the removal of organic matter from the effluent also generates a considerable amount of these contaminants. Without a proper sludge management, CECs can access the groundwater through the leaching phenomenon and severely contaminate it over time (Wu et al. 2010; Buerge et al. 2009).

A collection of CECs concentration data was gathered in the literature showing their presence in raw influent and effluent streams of WWTPs worldwide, see (**Table 1**).

Class	CEC	Region	Concentration range (µg/L) in raw influent	References	Concentration range (µg/L) in effluent	References
PhACs	Paracetamol	India, South Korea, Singapore, Spain, UK, US	1.13 - 172.00	[1], [2], [3], [4]	0.13 – 44.65	[41], [42]
	Erythromycin		0.63 - 10.41	[2], [3], [8]	0.19 - 7.30	[42], [43]
	Ibuprofen	Germany, UK, Canada, Portugal, Singapore	0.26 - 61.27	[5], [6], [7]	1.89 - 14.60	[42], [43], [45]
	Tetracycline	China, Portugal, Spain, US, UK, Mexico	0.06 - 7.84	[1], [9], [10]	1.53 - 3.60	[43], [44], [46]
	Carbamazepine	China, Switzerland, Spain, Italy, Greece, Germany, Canada, Australia, South Africa	0.20 - 3.98	[12], [13], [14], [15]	0.55 - 0.72	[42], [46], [48]
	Atenolol	South Korea, Singapore, Saudi Arabia, Switzerland, Spain, Portugal, Germany, South Africa	0.28 - 28.00	[10], [15], [17]	0.52 - 7.36	[43], [46], [47]
	17 β - estradiol	Mexico, South Africa	0.04 - 0.08	[16], [17]	0.01 - 0.05	
Pesticides	Diazinon	Switzerland, Spain, US	0.04 - 0.28	[18], [19]	0.05 - 0.16	[50], [51], [52]
	Malathion	Spain, Italy	0.07 - 2.62	[18], [20]	0.06 - 0.8	[50], [51], [52]
	Diuron	Germany, Spain	0.08 - 1.01	[18], [19], [21]	0.12 - 0.36	[49], [50], [51]
	Mecoprop	Switzerland, Germany, Spain	0.23 - 0.48	[19], [21], [22]	0.02 - 0.38	[49], [50], [51]
ASWs	Saccharin	China, Vietnam, Switzerland, Germany, Greece, US	15.00 - 77.74	[21], [22], [23], [24]	0.70 - 2.37	[43], [48]
	Sucralose	India, China, Vietnam, Switzerland	1.84 - 12.63	[24], [25], [26]	1.30 - 10.1	[43], [48]
	Acesulfame	Vietnam, Switzerland, Spain, Greece, US	13.00 - 27.00	[26], [27]	5.84 - 9.15	[43], [48]

Class	CEC	Region	Concentration range (µg/L) in raw influent	References	Concentration range (µg/L) in effluent	References
X-ray contrast media	Iohexol	Switzerland, Germany	44.50 - 80.12	[9], [15], [29], [30]	2.10 - 8.7	[30], [43], [54]
	Iopromide	South Korea, Vietnam, Switzerland, US	3.45 - 36.52	[15], [28], [29]	0.05 - 8.14	[30], [53]
	Iopamidol	Vietnam, Singapore, Switzerland	1.46 - 14.28	[28], [29], [30]	4.70 - 6.52	[43], [54]
Flame Retardants	TCEP	China, South Korea, Germany, Spain, Sweden, US, Australia	0.22 - 20.76	[31], [32]	0.20 - 10.20	[55], [56]
	TnBP	China, Germany, Sweden, US	0.45 - 50.70	[31], [33]	0.3 - 37.00	[56], [57]
Stimulants/illicit drugs	Caffeine	India, China, South Korea, Vietnam, Greece, Brazil	72.00 - 165.55	[21], [35]	0.01 - 51.70	[42], [43]
	Codeine	India, Vietnam, Spain, Germany, Sweden, UK, US, Canada, Australia	0.47 - 9.89	[34], [35], [36]	0.21 - 3.30	[42], [59]
UV filters	Benzophenone-3	India, China, Singapore, Switzerland, Germany, Spain, Australia	0.63 - 398.40	[37], [38], [39]	0.01 - 0.03	[58], [61]
	Octocrylene	China, Germany	0.16 - 4.25	[37], [38], [39], [40]	0.15 - 1.20	[43], [60]

Table 1: Presence of CECs in WWTPs influent and effluent streams worldwide.

[1] (de Jesus Gaffney et al. 2017); [2] (Chen et al. 2016); [3] (Lutterbeck et al. 2020); [4] (Barbosa et al. 2016); [5] (Petrie, Barden, and Kasprzyk-Hordern 2015); [6] (Villarín and Merel 2020); [7] (J. L. Santos et al. 2009); [8] (Kasonga et al. 2021); [9] (Karthikeyan and Meyer 2006); [10] (Rathi, Kumar, and Show 2021); [11] (Varma et al. 2021); [12] (Sim et al. 2011); [13] (Chaturvedi et al. 2021); [14] (Saidulu et al. 2021); [15] (Pena-Pereira et al. 2021); [16] (Gani et al. 2021); [17] (Mohapatra et al. 2016); [18] (Corcoran et al. 2012); [19] (Nie et al. 2013); [20] (Thomaidi et al. 2015); [21] (Subedi et al. 2015); [22] (Tran et al. 2015); [23] (van Stempvoort et al. 2020); [24] (Sharma et al. 2019); [25] (Subedi and Kannan 2014); [26] (Buerge et al. 2009); [27] (Gan et al. 2013); [28] (García-López, Rodríguez, and Cela 2010); [29] (Akao et al. 2020); [30] (Pérez and Barceló 2006); [31] (Loos et al. 2013); [32] (U. J. Kim, Oh, and Kannan 2017); [33] (Pantelaki and Voutsa 2019); [34] (Kasprzyk-Hordern, Dinsdale, and Guwy 2009); [35] (Shah et al. 2020); [36] (Nika et al. 2020); [37] (Gilbert et al. 2013); [38] (Krause et al. 2012); [39] (Kunisue et al. 2012); [40] (Kupper et al. 2006); [41] (Ali Gamal Al-Kaf et al. 2017); [42] (M. J. Gómez et al. 2007); [43] (Tran, Reinhard, and Gin 2018); [44] (Kumar, Singh, and Ambekar 2021); [45] (Blair et al. 2015); [46] (Tran et al. 2016); [47] (Boleda, Galceran, and Ventura 2011); [48] (Yang et al. 2017); [49] (Westlund and Yargeau 2017); [50] (Köck-Schulmeyer et al. 2013); [51] (Firouzsalari et al. 2019); [52] (Sutton et al. 2019); [53] (Kormos, Schulz, and Ternes 2011); [54] (Zemann et al. 2014); [55] (Xu et al. 2021); [56] (Liang and Liu 2016); [57] (Margot et al. 2015); [58] (Magi et al. 2012); [59] (Dey, Bano, and Malik 2019); [60] (Gago-Ferrero et al. 2013); [61] (Kapelewska et al. 2018).

Household sewage is one of the head leaders in which comes to the releasing source of the pharmaceutically active compounds. Camotti Bastos et al. (2020) and Margot et al. (2015) found primary health care products in concentration ranges from 264 to 7620 µg per kilogram of WWTP influent wastewater matter.

Despite the reported levels of PhACs in **Table 1**, paracetamol was found in Portugal in a concentration higher than 620 µg/L and, in the UK, concentrations higher than 510 µg/L from WWTP influent (Lutterbeck et al. 2020; Chen et al. 2016; de Jesus Gaffney et al. 2017). Spain presented a greater level of ibuprofen concentration in WWTP influent (603 µg/L) (J. L. Santos et al. 2009; Tran, Reinhard, and Gin 2018).

This is because, during the winter season, the use of pharmaceuticals tends to increase due to seasonal illnesses such as cold, fever, and severe acute respiratory syndrome. Mohapatra et al. (2016) found that pharmaceuticals concentration in WWTPs influent increased over 24% in winter season, compared with summer season. In addition, maximum levels for tetracycline concentration (48 µg/L) were accounted in WWTPs influent from North America, 40 to 480 times higher than Asia and Europe levels (Barbosa et al. 2016; Kasonga et al. 2021; Agüera, Martínez Bueno, and Fernández-Alba 2013).

Pesticide use follows the population growth rate ensuring large scale crop availability by optimizing the agriculture practice. Asia and Americas lead the pesticides to use per area of cropland, overcoming 3.5 kg/ha which is 34% higher than the world's average pesticides use. Europe, Oceania and Africa meet between 0.4 and 2 kg/ha of pesticide use. The most used class of pesticide is herbicides, with a worldwide average of 40% occurrence (FAO 2021). Pesticides diazinon, malathion, diuron and mecoprop were found in WWTPs in a few European countries, with concentration values in the order of 2.5 µg/L (Firouzsalari et al. 2019). Such pesticide occurrence can represent a serious threat to environmental biota due to the exponential bioaccumulation properties found in the pesticides over time (Katagi 2010; Li 2020).

Artificial sweeteners are part of a sector of society and one of the largest ones in the food industry. Across various WWTPs in India, considering influent, effluent and sludge samples, concentrations above 300 µg/L for saccharin and concentrations above 1.8 µg/L for sucralose were registered (Subedi et al. 2015). In the USA and China's WWTPs samples, sucralose concentration levels were detected above 20 µg/L, over 2 times higher than the other artificial sweeteners concentration levels (Subedi and Kannan 2014; W. Guo et al. 2021). On the other hand, Switzerland detected concentrations of acesulfame up to 46 µg/L in untreated wastewaters and concentrations above 12 µg/L in treated wastewaters. Untreated wastewaters return to the environment and can access the groundwaters due to the hydraulic conductivity property of the wastewater through the porous feature of the soil (Qian, Chen, and Howard 2020). Persistence levels of artificial sweeteners were also detected in North America and Western Europe around 5 µg/L (Buerge et al. 2011; van Stempvoort et al. 2020).

X-ray contrast media are widely used in daily hospital exams. Administered intravascularly, X-ray contrast media can be consumed from 200 to 725 g in a daily routine of exams (Weissbrodt et al. 2009; Kormos, Schulz, and Ternes 2011). Iodinated X-ray contrast media type are the most commonly used, e.g. Iohexol, Iopromide and Iopamidol. Considering their high biochemical stability, they are excreted via urine and feces mainly in non-metabolized forms and consequently enter in the sewage system (Tran, Reinhard, and Gin 2018). Iohexol and iopamidol have been reported in wastewater influents of the Southeast Asia region, with a maximum concentration of 124.9 µg/L and 45.6 µg/L, respectively. On the other hand, Portugal’s WWTPs influents presented a maximum iopromide concentration of 164 µg/L (Tran and Gin 2017; Patel et al. 2019).

As a prevention mechanism, flame retardants are incorporated into combustible materials to delay their ignition point and prevent flame propagation by interrupting or hindering the combustion process (Pantelaki and Voutsa 2019). Organophosphate flame retardants (OPFRs) were detected in airborne particles over the ocean, which may suggest that these compounds can be transported within a long-range distance in atmospheric medium over the ocean towards Arctic and Antarctic areas (Möller et al. 2012). (Ma et al. 2017) detected OPFRs in ocean sediments over Arctic areas, inferring the long transport of these compounds also via the aquatic medium. Gustavsson et al. (2018) found traces of OPFRs in northern Swedish rivers without a source point. The authors suggested the long-range atmospheric transport could be the source pathway. **Table 1** shows concentrations levels for tri(2-chloroethyl) Phosphate (TCEP) and tri-n-butyl phosphate (TnBP) found in worldwide WWTPs influent and effluent streams.

Other types of substances considered CECs are the alkaloids, caffeine and codeine. Caffeine makes the population's daily diet with consumption of more than 170 mg per day in consumers over 23 years of age (Mitchell et al. 2014; Mahoney et al. 2019). Levels above 200 µg/L of caffeine were found in Portugal and Spain WWTPs influent samples (Tran, Reinhard, and Gin 2018; Ramírez-Malule, Quiñones-Murillo, and Manotas-Duque 2020). Higher caffeine concentrations were observed in WWTP influents located in the UK, with more than 540 µg/L registered (Nakada et al. 2017). US WWTP influents presented up to 300 µg/L of caffeine and Asia, average levels above 100 µg/L (Chaturvedi et al. 2021; Patel et al. 2019; Deegan et al. 2011). Codeine is one of the most used opioids against pain, cough, cold and flu, when combined with other antihistamines and decongestants; reaching an average above 25 consumed tablets per person (Schaffer et al. 2020). Maximum values concentration of codeine were found above 32 µg/L in US and UK wastewater effluents (Parida et al. 2021).

Sunscreens, lotions, and shampoos incorporated with UV filters are the most commonly used PCPs in European countries, resulting in the highest concentration of UV filters in different environmental matrices (Brausch and Rand 2011).

3. Harmful impacts of CECs in environment

One of the main concerns about emerging contaminants lies in their accumulation potential over time or their retention into the organism structure. That phenomenon is denominated bioaccumulation factor, ordinarily calculated as the ratio of the compound of interest concentration in the biota sample (plant, animals) to that in the surrounding media, e.g. soil or water (Zenker et al. 2014; Shenker et al. 2011). Although this definition of bioaccumulation factor seems to be a straight way calculation, there are some specific models and criteria to take in consideration depending on the substances that are being investigated. One known criterion is the use of octanol–water partition coefficient (K_{ow}) being > 5 as listed in Annex D of the Stockholm Convention (Gobas et al. 2009). Bioaccumulation can occur via direct exposure, when humans and animals drink contaminated water, or via trophic levels succession along the food chain (Majumder, Gupta, and Gupta 2019).

Emerging contaminants generally present a high molecular weight and unique chemical structure, in which some specific functional groups are combined, e.g. benzene, amine, amide fluoride, carboxyl, ketone, among others. The physical, chemical and toxicological properties of a compound are related to its molecular structure. Therefore, the harmful potential of CECs may be evaluated based on the reactivity of the attached functional groups and their decomposition products (Barratt 2000; Parida et al. 2021). An extended exposure can cause adverse effects on the aquatic biota and, in some cases, may impact the hormonal and metabolic activity of animal and human beings (Bolong et al. 2009; Tran, Reinhard, and Gin 2018; Rout et al. 2021).

Contaminants of emerging concern containing nitrogen groups (amine, amide, cyclic amine), such as paracetamol, erythromycin, norfloxacin, ciprofloxacin, tetracycline, carbamazepine, saccharin, iohexol, caffeine, diuron, among others, can form toxic fumes of nitrogen oxides when decomposed (Parida et al. 2021). Hydrogen fluoride gas is also formed from the decomposition of fluoride-based compounds for example ciprofloxacin and norfloxacin (Parida et al. 2021). The presence of ketone groups (e.g. saccharin, erythromycin and benzophenone molecular structures) in a high concentration may cause toxic to living beings organism (Schultz and Yarbrough 2007). X-ray contrast media substances like iohexol, iopromide and iopamidol finds their toxicity mainly due to the presence of the benzene group, which is similar to the pesticides. In addition, Iopromide is a dicarboxylic acid diamide, which acts as a radiopaque medium, a nephrotoxic agent, and a xenobiotic substance (Kaller and An 2021). Many phenolic compounds act as carcinogen, causing damages to the red blood cells and liver, even in low concentrations. When they interact with microorganisms, other organic or inorganic substances in water, has a chance to produce substituted chemical species, which may be as toxic as the original phenolic compounds (Anku, Mamo, and Govender 2017). CECs have a significant impact on the aquatic biota, where a proper understanding of their environmental implications should be established.

Quantitative risk assessment of CECs toxic behavior can be estimated by the Risk Quotient (RQ) value, calculated using (**Equation** (1) (Nika et al. 2020).

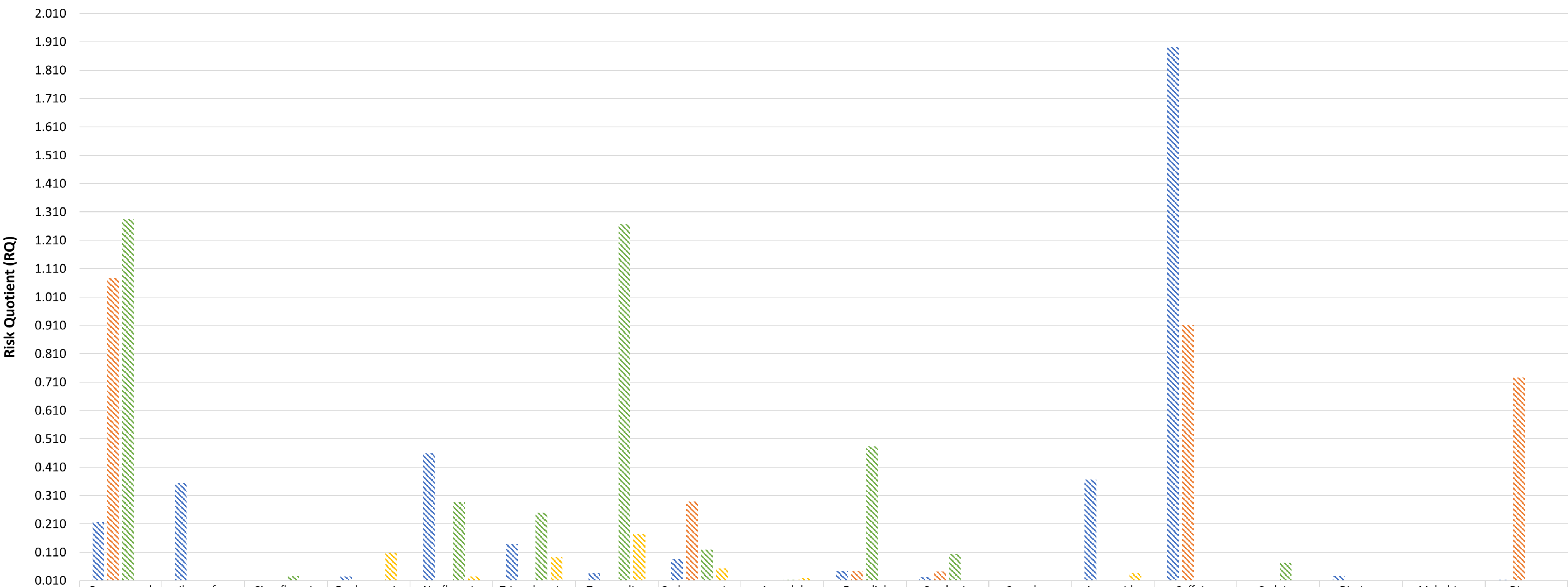
$$RQ = \frac{MEC}{PNEC_{aq}} \tag{1}$$

where MEC (mg/L) is the average of the highest measured concentrations of a compound detected in wastewater from different locations; $PNEC_{aq}$ (mg/L) is the Predicted Non-Effect Concentration estimated according to the (**Equation** (2), which an assessment factor of 1000 was used to adjust the PNEQ values and have been done this way because risk assessment was carried out using only acute toxicity data, due to scarce chronic toxicity data available (Nika et al. 2020).

$$PNEC_{aq} = \frac{EC_{50} \text{ or } LC_{50}}{1000} \tag{2}$$

where EC_{50} and LC_{50} are the 50% Effective Concentration and 50% Lethal Concentration of the CEC respectively (mg/L). These two parameters are described by the European Commission Directive (EU Directive 98/8/EC 1998) as a concentration causing 50 % inhibition of a given parameter, e.g. growth of an aquatic organism).

Values of RQ > 1 have been reported as a potential high risk to aquatic life. On the other hand, values of RQ between 1 and 0.1, represent a medium risk and for values below 0.1, represents a low risk to the aquatic environment (Gani et al. 2021; Parida et al. 2021). **Figure 1** shows the calculated Risk Quotient values considering four different types of aquatic organisms: fish, daphnia, algae and crustaceans taking into account the values of EC_{50} or LC_{50} found in the literature for the following CECs: paracetamol (Yamamoto et al. 2007; Y. Kim et al. 2007; Henschel et al. 1997), ibuprofen (Ginebreda et al. 2010; Sharma et al. 2019), ciprofloxacin (Sharma et al. 2019), erythromycin (González-Pleiter et al. 2013; J. W. Kim et al. 2009), norfloxacin (Eguchi et al. 2004; Y. Kim et al. 2007), trimethoprim (Y. Kim et al. 2007; de Andrés, Castañeda, and Ríos 2009; Minguez et al. 2016), tetracycline (Brausch and Rand 2011; Wollenberger, Halling-Sørensen, and Kusk 2000; González-Pleiter et al. 2013), carbamazepine (Ferrari et al. 2004; Sharma et al. 2019; J. W. Kim et al. 2009; Y. Kim et al. 2007), atenolol (Sharma et al. 2019; de Andrés, Castañeda, and Ríos 2009), 17- β estradiol (Lin et al. 2020), saccharin, sucralose (Sharma et al. 2019), iopromide (J. Guo 2015; L. H. M. L. M. Santos et al. 2010), caffeine (Sharma et al. 2019), codeine (J. Guo 2015), diazinon , malathion and diuron (Munn et al. 2001).



	Paracetamol	Ibuprofen	Ciprofloxacin	Erythromycin	Norfloxacin	Trimethoprim	Tetracycline	Carbamazepine	Atenolol	Estradiol	Saccharin	Sucralose	Iopromide	Caffeine	Codeine	Diazinon	Malathion	Diuron
Fish	0.215	0.354	0.004	0.025	0.459	0.139	0.036	0.087	0.002	0.046	0.021	0.005	0.365	1.892		0.027	0.009	0.012
Daphnia	1.075		0.009					0.289	0.004	0.044	0.042	0.001		0.910				0.725
Algae	1.284		0.025		0.287	0.249	1.265	0.119	0.012	0.484	0.103		0.004		0.074			
Crustaceans				0.109	0.025	0.094	0.175	0.052	0.019				0.037					

Types of Contaminant of Emerging Concern (CECs)

Figure 1 - Risk quotients of different CECs for different aquatic organisms (fish, daphnia, algae and crustaceans) with respect to acute toxicity for each of them.

Through a similar methodology, Xiang et al. (2021) calculated the RQ values for PhACs and personal care products with respect to different aquatic species and found that ciprofloxacin, erythromycin, triclosan, and diclofenac have shown high risk magnitudes with RQ > 10. (Thomaidi et al. 2015) investigated 30 CECs, which most of the were PhACs. Authors obtained a RQ > 1 were calculated for the 30 compounds concentration in secondary treated wastewater, considering the lethal concentration for fish, daphnia and algae. Even higher values of RQ has also been reported for ibuprofen, concerning the acute toxicity to daphnia and algae groups [6.76 and 15.32 respectively] (Kovalova et al. 2013; Song et al. 2018). Paracetamol RQ levels of 5.72 has been found against the crustacean species (Dolar et al. 2012). Despite the low risk values of RQ for diazinon and malathion, showed in**Error! Reference source not found.**, Pelaez et al. (2012) estimated lethal concentrations for daphnia species which resulted in extremely high RQ values of 308.8 and 204.7 respectively. Other CEC considered very toxic to fish, daphnia and algae species is the plasticizer, bisphenol-A, found to present RQ levels of 109.8, 26.7 and 105.6 (Chowdhury, Viraraghavan, and Srinivasan 2010). Nevertheless, more research is necessary on the selection of aquatic species and $PNEC_{aq}$ values, in order to evaluate the toxicity on the aquatic environment.

Nie et al. (2013) discovered that some antibiotics can prevent the photosystem II electron transport chain during the ATP synthase process, thereby inhibiting the photosynthesis process. Antibiotics can also affect prokaryotic cells through a complex mechanism, such as inhibition of nucleic acid (DNA/RNA) synthesis, protein synthesis, and cell envelope synthesis in aquatic organisms (Patel et al. 2019). Iodinated x-ray contrast substances and their metabolites were found to present some toxic level influence in animals and humans (Waqas et al. 2020; E. F. Gómez and Michel 2013). Reproductive hazards like spermatogenesis, teratogenesis or other reproductive impairment have been detected in animal and human organism, caused by diazinon (Daniela A et al. 2015; Harchegani et al. 2018). Malathion exposure in rats induced a degradation process of membrane lipids, involving the deterioration of the cellular integrity (lipid peroxidation LPO), reported by (Rezg et al. 2008). Other pesticides as mecoprop and diuron have shown to cause organ damage and erythropoiesis irregularity (Domingues et al. 2011; Food Safety Authority et al. 2017). In the case of ASWs, it has been found that acesulfame, saccharin and sucralose have a potential to decrease the microbial diversity, parallel to an increase of proinflammatory species inside the body and alterations in the composition of gut microbiota and intestinal epithelial barrier (Ruiz-Ojeda et al. 2019).

4. Treatment methods towards CECs mitigation

Several different biological treatment techniques are well consolidated where removal of organic matter and nutrients from wastewater are the main concern. Processes like activated sludge process (ASP), constructed wetland (CWs), trickling filters (TFs), sequential batch reactor (SBR), oxidation ditches, facultative lagoons, etc., are wide investigated due to their cost-effectiveness operation (Miège et al. 2009; van Stempvoort et al. 2020; Subedi et al. 2015; Chen et al. 2016). Numerous studies have already demonstrated those mentioned treatment techniques removing up to 90% of the organic matter from targeted wastewater matrices (Salter et al. 2000; Zieliński, Zielińska, and Debowski 2013; Showkat and Najar 2019; Luo et al. 2020; Waqas et al. 2020; Varma et al. 2021). However, when those conventional wastewater treatment techniques are applied to mitigate CECs concentration (see **Table 2**), such good efficiency is limited to a few specific compounds and treatment processes.

Another important factor to be considered during the treatment process of wastewaters, are the regulatory guidelines. **Table 2** also describes a few existing regulatory guidelines to deal with the CECs from European Union (EU), United States Environmental Protection Agency (US EPA), Australian Drinking Water Guidelines (ADWG) and World Health Organization (WHO) (Radley-Gardner, Beale, and Zimmermann 2016; Carvalho et al. 2016; Korkaric et al. 2019; NHMRC Australian Guidelines 2008; Seibert et al. 2020; Careghini et al. 2015). Comparing the regulatory guidelines' max limits and the treated effluent concentration of the selected CECs **Table 2**, it is possible to see that most of the CECs extrapolate the concentration limits, which could indicate the level of limitation faced by those treatment techniques. However, sucralose, saccharin, acesulfame, tetracycline, norfloxacin, atenolol, iohexol, iopromide, iopamidol and mecoprop were satisfying the guideline values due to the high permissible discharge limits.

On the other hand, there are some well consolidated wastewater treatment techniques, which can mitigate the CECs in a more efficient and economically feasible way, such as pressure-driven processes (e.g. reverse osmosis (RO) and nanofiltration membranes (NF)), ozonation, and activated charcoal (AC) treatment (Shah et al. 2020). Ozonation treatment can result in the formation of by-products (e.g. N-nitrosodimethylamine (NDMA) and bromate), therefore a post-treatment step with a biological active sand filter is recommended to mitigate those unwanted compounds (Hollender et al. 2009), as they can severely damage the human and animal organs (Klein et al. 1991; WHO 2005). Wastewater treatment using AC offers no by-product formation and low energy consumption, although the CECs adsorbed from the wastewater matrix placed on the surface of the charcoal, generally are considered hazardous to the environment, and hence require proper disposal strategies (Rajasulochana and Preethy 2016; Luigi Rizzo et al. 2019). According to some studies, ozonation treatment for CECs removal usually is more efficient for certain compounds such as diclofenac, gabapentin, and sulfamethoxazole, while AC treatment was found to be effective for some CECs (benzotriazole, fluconazole, valsartan) (Margot et al. 2013; Kovalova et al. 2013; Jekel et al. 2015; Luigi Rizzo et al. 2019). Both ozonation and AC demonstrated inefficacy in the removal of negatively charged iodinated contrast media, however AC performed a slightly better removal of iopromide compared with ozonation (Knopp et al. 2016). In the other hand, steroid hormones like estrone (E1), 17β-estradiol (E2) and 17α-ethinylestradiol (EE2) are effectively removed by both ozonation (Sun et al. 2017) and AC (Margot et al. 2013) treatment techniques.

Method	Type of wastewater	Concentration of contaminant	CEC removal efficiency	Reference	Statutory guidelines	Reference
ASP	Municipal, industrial	Ibuprofen: 2640 - 5700 ng/L 17β-estradiol: 2 - 3 ng/L	Ibuprofen: < 70% 17β-estradiol: 65%	[62]	Ibuprofen: 11 ng/L 17β-estradiol: 1 ng/L	[78, 79]
ASP	Municipal, industrial	Caffeine: 6170 ng/L Ibuprofen: 93600 ng/L Carbamazepine: 480 ng/L	Caffeine: 44 - 75% Ibuprofen: 80 - 88% Carbamazepine: 8 - 15%	[63]	Caffeine: 500 ng/L Ibuprofen: 11 ng/L Carbamazepine: 10 ng/L	[78, 79, 81]
ASP	Municipal	Paracetamol: 1629 ng/L Carbamazepine: 566 ng/L	Paracetamol: 76.5% Carbamazepine: 32.7%	[64]	Paracetamol: 1100 ng/L Carbamazepine: 10 ng/L	[81]
ASP	Domestic, industrial and hospital	Acesulfame: 360 ng/L Iopromide: 100 ng/L	Acesulfame: 47.2% Iopromide: 75 %	[65]	Acesulfame: $1x10^8$ ng/L Iopromide: 750000 ng/L	[81]
ASP	Residential, hospital	Iohexol: 115436.5 ng/L Iopamidol: 34887 ng/L Carbamazepine: 323.8 ng/L Bisphenol A: 5733.5 ng/L	Iohexol: 50.6% Iopamidol: 14.4% Carbamazepine: 4.6% Bisphenol A: 85.8%	[66]	Iohexol: 720000 ng/L Iopamidol: 400000 ng/L Carbamazepine: 10 ng/L Bisphenol A: 80 ng/L	[78, 80, 81]
ASP	-	Diazinon: 316 ng/L Diuron: 2526.1 ng/L	Diazinon: 57.2 % Diuron: 5.3%	[67]	Diazinon: 0.4 ng/L Diuron: 90 ng/L	[78, 81 - 83]
ASP	-	Mecoprop: 252 ng/L	Mecoprop: 19.4%	[68]	Mecoprop: 2100 ng/L	[78, 79]
ASP	Municipal	Sucralose: 5289 ng/L Acesulfame: 3863 ng/L TCEP: 439 ng/L Carbamazepine: 188 ng/L	Sucralose: 24% Acesulfame: 4% TCEP: 21% Carbamazepine: 17%	[69]	Sucralose: $1x10^8$ ng/L Acesulfame: $1x10^8$ ng/L TCEP: 1000 - 4000 ng/L Carbamazepine: 10 ng/L	[78, 81]
ASP	Municipal	TCEP: 1430 ng/L	TCEP: 23%	[70]	TCEP: 1000 ng/L	[81]
ASP	Municipal	TnBP: 74.4 ng/L	TnBP: 61.3%	[71]	TnBP: 100 ng/L	[79]
ASP, Trickling filters	Communal, industrial	Erythromycin: 1609 ng/L Codeine: 10321 ng/L Atenolol: 12913 ng/L	Erythromycin: 13.9% Codeine: 48.9% Atenolol: 77.7%	[72]	Erythromycin: 100 ng/L Codeine: 1000 ng/L Atenolol: 90000 ng/L	[78, 81]

Method	Type of wastewater	Concentration of contaminant	CEC removal efficiency	Reference	Statutory guidelines	Reference
SBR activated sludge	Domestic, industrial	Sucralose: 2100 ng/L Acesulfame: 16000 ng/L Saccharin: 7200 ng/L	Sucralose: 14.3% Acesulfame: 6.2% Saccharin: 96.2%	[73]	Sucralose: $1x10^8$ ng/L Acesulfame: $1x10^8$ ng/L Saccharin: $1x10^8$ ng/L	[84]
Facultative lagoon	Industrial, commercial	Acesulfame: 12900 ng/L Sucralose: 15300 ng/L	Acesulfame: 43% Sucralose: 56.5%	[74]	Acesulfame: $1x10^8$ ng/L Sucralose: $1x10^8$ ng/L	[84]
Primary, chemical assist, ultraviolet disinfection	Industrial, commercial	Acesulfame: 17700 ng/L Sucralose: 26000 ng/L	Acesulfame: 11.8% Sucralose: 19.1%	[74]	Acesulfame: $1x10^8$ ng/L Sucralose: $1x10^8$ ng/L	[84]
Constructed wetlands	Municipal	Paracetamol: 180000 ng/L	Paracetamol: 91%	[75]	Paracetamol: 1100 ng/L	[81]
Anaerobic fluidized bed reactor	Municipal	Paracetamol: 2695 ng/L Ciprofloxacin: 157 ng/L Erythromycin: 319 ng/L Caffeine: 3470 ng/L	Paracetamol: 87.8% Ciprofloxacin: 89.4% Erythromycin: 58.7% Caffeine: 86.7%	[76]	Paracetamol: 1100 ng/L Ciprofloxacin: 100 ng/L Erythromycin: 100 ng/L Caffeine: 500 ng/L	[79, 81]
Biological nutrient removal	Municipal	Paracetamol: 2695 ng/L	Paracetamol: 84%	[77]	Paracetamol: 1100 ng/L	[81]

Table 2 – Removal efficiency of CECs by different conventional wastewater treatment techniques.

[62] (Carballa et al. 2004); [63] (J. L. Santos et al. 2009); [64] (Stamatis and Konstantinou 2013); [65] (Watanabe et al. 2016); [66] (Tran and Gin 2017); [67] (Masiá et al. 2013); [68] (Wick, Fink, and Ternes 2010); [69] (Ryu et al. 2014); [70] (U. J. Kim, Oh, and Kannan 2017); [71] (Liang and Liu 2016); [72] (Kasprzyk-Hordern, Dinsdale, and Guwy 2009); [73] (Gan et al. 2013); [74] (van Stempvoort et al. 2020); [75] (Vymazal et al. 2017); [76] (Dutta et al. 2014); [77] (Park et al. 2017); [78] (Korkaric et al. 2019); [79] (Carvalho et al. 2016); [80] (Careghini et al. 2015); [81] (NHMRC Australian Guidelines 2008); [82] (Radley-Gardner, Beale, and Zimmermann 2016); [83] (Seibert et al. 2020); [84] (Parida et al. 2021).

Covering a wider number of CECs removal, advanced treatment techniques such as usage of membrane bioreactor (MBR) integrated systems have been shown a good performance in some studies (Qin et al. 2018; Rodriguez-Sanchez et al. 2018; Iorhemen et al. 2018; Dhangar and Kumar 2020). However, MBR treatment is inefficient against biologically persistent and hydrophobic compounds, motivating researchers to combine MBR with other techniques such as ultrafiltration (UF) (Holloway et al. 2014), reverse osmosis (RO) (Dolar et al. 2012), activated charcoal (AC) (Kovalova et al. 2013), advanced oxidation process (AOPs) (Nguyen et al. 2013), membrane distillation (Song et al. 2018), etc., towards the removal of recalcitrant CECs. Parida et al. (2021) gathered 78 combinations of advanced treatment techniques to remove CECs from wastewater, which 38 were combinations of MBR with aforementioned techniques or other types of advanced treatment. Between the 38 combinations, only BioMnOx-MBR presented CEC removal efficiency lower than 80% for iohexol and ioprimide (Forrez et al. 2011), all other MBR combinations lead the CEC removal efficiency around 95%. (**Table 3**) presents a review on hybrid systems that combine advanced treatment techniques to remove CECs. Most of the hybrid treatment techniques presented showed a high efficiency (≥ 90%) removal with exception for the BioMnOx (biogenic metals manganese oxides) - Membrane Bioreactor process, which removal efficiencies did not reach 80% for iohexol and iopromide (Forrez et al. 2011), nevertheless their non-treated and treated concentration were below the Statutory Guideline level, what shows to be difficult to analyze this hybrid treatment efficacy in this specific case. For the iodinated contrast media compounds, the authors (Forrez et al. 2011) found a removal efficiency higher than 90% using a bio-palladium (Bio-Pd) - Membrane Bioreactor hybrid process. From the (**Table 3**) data analysis, it is clear that treatment process integrating ozone (O_3), membranes (ultrafiltration, microfiltration, osmotic MBR or reverse osmosis) and Ultraviolet stands in the front in what comes to the CECs removal performance, but further feasibility analysis should take place on those treatment techniques, due to their relatively high energy consumption and maintenance cost (Agustina, Ang, and Vareek 2005).

New advanced technologies are being investigated towards CECs mitigation with promising results (Castellanos et al. 2020; Vilar et al. 2020). A hybrid technique of hydrogen peroxide (H_2O_2) oxidant and UV radiation with membrane technology functioning as a multiple point oxidant doser were combined. This combination gave rise to a tube-in-tube membrane contactor that has already been used for CECs removal from synthetic and real wastewater matrices, assisted with heterogeneous TiO_2 photocatalysis (Castellanos et al. 2020) and using a UVC/ H_2O_2 system (Vilar et al. 2020). The highlight point of those investigations on tube-in-tube technology were the good removal efficiency considering a very short residence time (4.6 s) and a low UVC fluence (30 to 45 mJ/cm^2). Castellanos et al. (2020) achieved E2 and EE2 removal percentages of 51/32% and 48/30%, respectively, for both synthetic and real municipal wastewater matrix, using 45 mJ/cm^2 of UVC fluence. Vilar et al. (2020) achieved oxytetracycline (OTC) removal efficiencies of 36% and 7% for the synthetic and real urban wastewater composition respectively, using 34 mJ/cm^2 of UVC fluence. The authors conclude that the tube-in-tube membrane contactor has the advantage of an easy upscaling into a real WWTP by implementing multiple parallel membranes into a single shell. In terms of removal percentage, CECs mitigation using the tube-in-tube technology may appear to have a low efficiency comparing it to any other treatment technique presented in this review. However, it must be considered that the treatment techniques presented (traditional and advanced), authors carried out their investigations in a pilot or full municipal/industrial scale, which the contact time between the contaminant and the removal agent is in the order of hours or days.

Contaminants	Hybrid treatment technique	Concentration	Removal Efficiencies	References
Paracetamol	Ultrafiltration membrane - Osmotic Membrane Bioreactor	61200 ± 18216 ng/L	100%	[85]
	Microfiltration membrane- Granular Activated Carbon	114 ng/L	> 96%	[86]
	Aerated lagoon – Constructed Wetland – Ultraviolet	39300 ng/L	>99%	[87]
Ibuprofen	Constructed Wetland - Ozonation	15000 ng/L	97.2%	[88]
	Ultrafiltration - Osmotic Membrane Bioreactor	15680 ± 3444 ng/L	100%	[85]
	Anaerobic Membrane Bioreactor – Membrane Distillation	2000 ng/L	> 95%	[86]
	Aerated lagoon – Contructed Wetland - Ultraviolet	9922 ng/L	>99%	[87]
	Sequential Batch Reactor +Nanofiltration	10000 ng/L	99 ± 0.4%	[89]
Ciprofloxacin	Sponge Membrane Bioreactor - Ozonation	7280 ng/L	83 ± 7%	[90]
	Membrane Bioreactor + Powdered Activated Carbon	15700 ng/L	100 ± 0%	[91]
Erythromycin	Sponge Membrane Bioreactor - Ozonation	1070 ng/L	90 ± 1%	[90]
	Membrane Bioreactor + Reverse Osmosis	49 ng/L	>99%	[92]
Norfloxacin	Sponge Membrane Bioreactor - Ozonation	16680 ng/L	92 ± 4%	[90]
	Membrane Bioreactor + Powdered Activated Carbon	3140 ng/L	> 99%	[91]
	Nanofiltration - Ultraviolet/O_3	221 ng/L	> 99%	[93]

Contaminants	Hybrid treatment technique	Concentration	Removal Efficiencies	References
Trimethoprim	Sponge Membrane Bioreactor - Ozonation	1280 ng/L	97 ± 2%	[90]
	Ultrafiltration - Osmotic Membrane Bioreactor	13 ± 22 ng/L	100%	[85]
	Anaerobic Membrane Bioreactor – Membrane Distillation	2000 ng/L	> 95%	[86]
	Microfiltration-Granular Activated Carbon	974 ng/L	> 99%	[86]
Tetracycline	Sponge Membrane Bioreactor - Ozonation	730 ng/L	100 ± 0%	[90]
Carbamazepine	Membrane Bioreactor + Powdered Activated Carbon	235 ng/L	100 ± 0%	[91]
	Sequential Batch Reactor +Nanofiltration	14500 ng /L	93 ± 3%	[89]
	Membrane Bioreactor + Reverse Osmosis	83 ng/L	>99%	[92]
	Microfiltration-Granular Activated Carbon	2240 ng/L	> 99%	[86]
Atenolol	Ultrafiltration - Osmotic Membrane Bioreactor	61200 ± 18216 ng/L	100%	[85]
	Microfiltration-Granular Activated Carbon	466 ng/L	> 99%	[86]
	Sequential Batch Reactor +Nanofiltration	10000 ng /L	99 ± 0.8%	[89]
	Membrane Bioreactor + Reverse Osmosis	1820 ng/L	>99%	[92]
	Aerated lagoon – Contructed Wetland – Ultraviolet	1442 ng/L	>99%	[87]

Contaminants	Hybrid treatment technique	Concentration	Removal Efficiencies	References
17 β -Estradiol	Membrane Bioreactor – Reverse Osmosis and Membrane Bioreactor – Ultraviolet	5000 ng/L	> 90% for (MBR - RO), > 90% for (MBR - UV)	[94]
	Membrane Bioreactor - Granular Activated Carbon	5000 ng/L	100%	[95]
	Flocculation-Activated Sludge-Ultrafiltration	14 ng/L	> 90%	[96]
	Osmotic Membrane Bioreactor +Microfiltration	5000 ng/L	> 95%	[97]
Sucralose	Sequential Batch Reactor +Nanofiltration	13500 ng/L	88 ± 11%	[89]
	Ultrafiltration - Osmotic Membrane Bioreactor	42366 ± 12586 ng/L	100%	[85]
Acesulfame	Sequential Batch Reactor +Nanofiltration	13500 ng/L	81 ± 16%	[89]
	Ultrafiltration - Osmotic Membrane Bioreactor	12236 ± 9155 ng/L	100%	[85]
Iohexol	BioMnOx (biogenic metals manganese oxides)- Membrane Bioreactor	224 ng/L	72%	[98]
	Activated Sludge - Slow sand filter	3280 ng/L	91 ± 8%	[99]

Contaminants	Hybrid treatment technique	Concentration	Removal Efficiencies	References
Iopromide	Sequential Batch Reactor +Nanofiltration	10000 ng/L	90 ± 8%	[89]
	Activated Sludge - Slow sand filter	2900 ng/L	91 ± 6%	[99]
	Membrane Bioreactor + Ultraviolet	118000 ng/L	92 ± 0%	[91]
	BioMnOx (biogenic metals manganese oxides)- Membrane Bioreactor	454 ng/L	68%	[98]
	Membrane Bioreactor + Powdered Activated Carbon	118000 ng/L	91 ± 0%	[91]
Iopamidol	Membrane Bioreactor + Powdered Activated Carbon	3353000 ng /L	80 ±2%	[91]
	Membrane Bioreactor + Powdered Activated Carbon	3353000 ng /L	92 ± 1%	[91]
Octocrylene	Osmotic Membrane Bioreactor +Microfiltration	5000 ng/L	90%	[97]
	Osmotic Membrane Bioreactor – Reverse Osmosis	5000 ng/L	> 90%	[100]
Benzophenone-3	Ultrafiltration - Osmotic Membrane Bioreactor – Reverse Osmosis	3442 ± 1232 ng/L	100%	[85]
	Osmotic Membrane Bioreactor – Reverse Osmosis	5000 ng/L	> 90%	[100]
	Osmotic Membrane Bioreactor +Microfiltration	5000 ng/L	> 95%	[97]

Contaminants	Hybrid treatment technique	Concentration	Removal Efficiencies	References
TCEP	Ultrafiltration - Osmotic Membrane Bioreactor – Reverse Osmosis	810 ± 687 ng/L	> 95%	[85]
	Sequential Batch Reactor +Nanofiltration	10000 ng/L	89 ± 7%	[86]
	Anaerobic Membrane Bioreactor – Membrane Distillation	2000 ng/L	> 90%	[86]
Caffeine	Anaerobic Membrane Bioreactor – Membrane Distillation	2000 ng/L	> 95%	[86]
	Microfiltration-Granular Activated Carbon	1410 ng/L	> 99%	[86]
	Ultrafiltration - Osmotic Membrane Bioreactor	82457 ± 16903 ng/L	100%	[85]
	Aerated lagoon – Constructed Wetland - Ultraviolet	25567 ng/L	>99%	[87]
Codeine	Membrane Bioreactor + Reverse Osmosis	152 ng/L	>99%	[92]
	BioMnOx (biogenic metals manganese oxides)- Membrane Bioreactor	151 ng/L	>93%	[98]
Bisphenol-A	Osmotic Membrane Bioreactor – Reverse Osmosis	5000 ng/L	> 90%	[100]
	Flocculation-Activated Sludge-Ultrafiltration	857 ng/L	> 90%	[96]
	Membrane Bioreactor – Granular Activated Carbon	5000 ng/L	> 90%	[95]
	Sequential Batch Reactor +Nanofiltration	13000 ng/L	99 ± 0.4%	[89]
Diazinon	Anaerobic Membrane Bioreactor – Membrane Distillation	2000 ng/L	> 95%	[86]

Contaminants	Hybrid treatment technique	Concentration	Removal Efficiencies	References
Malathion	Ni doped TiO_2 nanoparticles	200 ng/L	94%	[101]
Mecoprop	BioMnOx (biogenic metals manganese oxides)- Membrane Bioreactor	55 ng/L	> 81%	[98]
Diuron	BioMnOx (biogenic metals manganese oxides)- Membrane Bioreactor	67 ng/L	> 94%	[98]
	Anaerobic Membrane Bioreactor – Membrane Distillation	2000 ng/L	> 95%	[86]

Table 3 – Hybrid treatment performances of different advanced techniques towards CECs removal from wastewater.

[85] (Holloway et al. 2014); [86] (Shanmuganathan et al. 2017); [87] (Conkle, White, and Metcalfe 2008); [88] (Lancheros et al. 2019); [89] (Wei et al. 2018); [90] (Vo et al. 2019); [91] (Kovalova et al. 2013); [93] (Liu et al. 2014); [92] (Dolar et al. 2012); [94] (Nguyen et al. 2013); [95] (Nguyen et al. 2013); [96] (Melo-Guimarães et al. 2013); [97] (W. Luo et al. 2015); [98] (Forrez et al. 2011); [99] (Escolà Casas and Bester 2015); [100] (W. Luo et al. 2017); [101] (Surendra et al. 2020).

5. Conclusions

The growing use of pharmaceutically active compounds, personal care products, artificial sweeteners, UV-filters, X-ray contrast media, pesticides, etc., contributes to the maintenance of those compounds in the wastewater matrices. CECs occurrence is a global matter, although the most prominent occurrence is through Europe and Asia. Without an adequate treatment system and limitations in the regulatory guidelines to deal with such compounds, reinforces their persistence in the aquatic environment which can become harmful to the biota due to their toxic potential, turning them into compounds of emerging concern. The toxicity of CECs on aquatic life, such as fish, algae, daphnia and crustaceans were measured by calculating the RQ value for different compounds of emerging concern. Paracetamol presented the highest risk against daphnia and algae (1.075 and 1.284 respectively), on the other hand, caffeine RQ value was found to have the highest toxic influence on fish (1.89) and tetracycline with a RQ of 1.26 against algae, thus showing a severe threat to those species by those compounds. Crustaceans showed an overall better resistance against the selected CECs, however further investigations on CECs ecotoxicity potential with a wide range of aquatic biota should be established.

Most of the conventional treatment techniques were not efficient in mitigate the CECs concentrations down to the limits stated by the international statutory guidelines. Overall average of CECs removal efficiency for ASP, CWs, SBR, etc., was found to be less than 70%. The combined treatment techniques such as MBR-RO, MBR-NF, MBR-AOP, etc., showed almost complete removal of CECs. Other new AOP technologies to remove CECs, such as the tube-in-tube membrane contactor have been showing promising results with potential for a scaling up to urban WWTP. Further research should be performed on the costs of the hybrid treatment processes and its optimization, allowing easy and feasible upscaling options.

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