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Implementation of advanced micropollutants removal technologies in wastewater treatment plants (WWTPs) - examples and challenges based on selected EU countries

Klaudia Kosek [1,*], Aneta Luczkiewicz [1,*], Sylwia Fudala-Książek [1], Katarzyna Jankowska [1], Małgorzata Szopińska [1], Ola Svahn [2], Jens Tränckner [3], Alena Kaiser [3], Valdas Langas [4], Erland Björklund [2]

[1] Faculty of Civil and Environmental Engineering, Gdansk University of Technology, 11/12 Narutowicza St., Gdansk 80-233, Poland; e-mails: klaudia.kosek@pg.edu.pl, aneta.luczkiewicz@pg.edu.pl, sylwia.fudala-ksiazek@pg.edu.pl, katarzyna.jankowska@pg.edu.pl, malgorzata.szopinska@pg.edu.pl
[2] Faculty of Natural Science, Kristianstad University, SE-291 39 Kristianstad, Sweden; e-mails: ola.svahn@hkr.se, erland.bjorklund@hkr.se
[3] Department of Water Management, University of Rostock, Satower Straße 48, 18059 Rostock, Germany; e-mails: jens.traenckner@uni-rostock.de, alena.kaiser@uni-rostock.de
[4] Study Centre of Social Geography and Regional Sciences and Marine Research Institute, Klaipeda University, ˙ LT-92294 Klaipeda, Lithuania; e-mail: valdas.langas@apc.ku.lt
* Correspondence: klaudia.kosek@pg.edu.pl; aneta.luczkiewicz@pg.edu.pl

Abstract: The accumulation of micropollutants (MPs) and their increasing concentration in the aquatic environment are an emerging issue for water quality in the world. The complex web of exposure pathways, as well as the variety in the chemical structure and potency of MPs, represents enormous challenges for researchers and policy initiatives. In order to manage MPs, it has to be decided which of them have to be reduced and to what extent, where in the water cycle this would be the most efficient and which technical means that should be applied to be sustainable. All of these aspects require a knowledge of MPs abundance, properties, fate and impact in the environment, which is essentially determined by two related features: the sources and the physico-chemical characteristics of MPs. Micropollutants including pharmaceuticals, antibiotics and hormones can enter the aquatic environment through both diffuse and point sources, but in urbanised regions wastewater treatment plants (WWTPs) play a crucial role in their dissemination. Conventional WWTPs are effective in removal of macropollutants (e.g. nutrients, suspended solids and some trace elements), while MPs may go through the treatment unchanged or be removed at different rates. Most of the EU countries are convinced that the presence of MPs in the environment poses a serious problem, particularly in highly populated regions where surface water resources serve as a source of potable water. Presently, various technical solutions are available and have been proven possible to integrate with existing treatment processes in an

expedient manner. The solutions that have been evaluated are mainly based on ozonation and/or activated carbon treatment technologies which may definitely be considered the most effective compared to the costs incurred.

Keywords: micropollutants, pharmaceuticals, ozonation, granular activated carbon, powdered activated carbon, wastewater treatment

1. Introduction

Water bodies have a variety of functions such as for drinking and irrigation purposes, hydroelectric reservoirs and leisure area reserves. Thus, to supply all current and future needs, these water reserves have to be effectively and sustainably managed. There are some directives that describe water management in the European Union (EU). The Urban Wastewater Treatment Directive (UWWTD), the Water Framework Directive (WFD) and the Drinking Water Directive are the most important pieces of legislation introduced into the EU water sector. Because of them, 'water' is currently seen as a heritage (rather than a commercial product), which must be protected, defended and treated as such [1]. To reach a good or high ecological status of water bodies [2], the WFD introduces a 'catchment-based approach' and an 'integrated river basin management'. It requires a deep and holistic understanding of ecosystems complexity, including the local human-nature interdependencies. Even a single human activity can be considered as a source of disturbance and contributes to ecosystem degradation [3].

Currently, of special concern, are chemical compounds regularly used in industrial, commercial and/or domestic applications, including over 30 000 different substances such as human and veterinary pharmaceuticals, plant protection products, biocides, personal care products, household chemicals and detergents [4]. Most of them enter the wastewater system, and may thus finally end up in the surrounding water bodies (receivers). Wastewater treatment plants have already been recognised as a key pathway and chief point sources of chemical compounds to the environment [5]. Usually chemical compounds are detected at very low concentrations, and many compounds such as pharmaceuticals, antibiotics and hormones are present at concentration levels below μg/L, and are considered micropollutants

(MPs). These MPs may trigger unwanted ecological effects by exerting stress to aquatic life. Since our daily usage of chemical compounds increases, MPs represent an important challenge for WWTP operators and for our water resources [5]. In response to the emerging problem of increasing pharmaceutical concentration levels in the wastewater systems, and thus in the environment, in 2019 the Communication from the Commission (regarding EU Strategic Approach to Pharmaceuticals in the Environment) to the European Parliament, the Council and the European Economic and Social Committee, was established [6].

In 2001, Decision 2455/2001/EC [7] established a list of 33 priority substances, among which 13 were identified as "priority hazardous substances". This first list was replaced by Annex II of the Directive on Environmental Quality Standards (Directive 2008/105/EC) [8] (EQSD), which limits the concentrations of 33 priority substances (Annex II) and 8 other pollutants (Annex III) in surface waters. The Commission subsequently reviewed this list. In 2013, Directive 2013/39/EU [9] amended both WFD and the EQSD and established a Watch List mechanism. The Watch List indicates potential water pollutants that should be temporarily monitored in surface waters to obtain a high-quality Union-wide dataset. It is crucial to properly assess the risk that such pollutants pose to the aquatic environment. The Watch List 1, was published in 2015 [10], and included ten substances or groups of substances (Table 1). Reviewing Watch List 1 resulted in the Watch List 2 [11] where the Commission removed five substances or groups of substances (diclofenac, the herbicides oxadiazon and triallate, the sunscreen ingredient 2-ethylhexyl-4-methoxycinnamate and the industrial compound 2,6-di-tert-butyl-4-methylphenol) and included three new substances (the pesticide metaflumizone and the two antibiotics amoxicillin and ciprofloxacin). The inclusion of the antibiotics on Watch List 2 is consistent with the European One Health Action Plan against Antimicrobial Resistance (AMR) [12], which, among others, supports the use of the Watch List to improve knowledge and to evaluate the risks to human and animal health posed by the presence of antimicrobials in the environment. The updated Watch List 2 was published in 2018 (Table 1).

From the above it can be concluded that the removal of pharmaceuticals from wastewater today is not required within the European Union, but their monitoring has been included in EU Watch List 1 and Watch List 2. Multidisciplinary efforts are also expected to develop new tools for detection and new technologies to prevent water resources from antimicrobials and antimicrobial-resistant microorganisms. In response to the EU legal basis and recommendations, some EU countries have already suggested/introduced indicator compounds, which should be monitored at a national level [12].

Table 1. Micropollutants, including pharmaceuticals, antibiotics and hormones included in EU Watch List 1 and Watch List 2

Watch List 1 2015	Compounds	Watch List 2 2018
	Pharmaceuticals	
X	Diclofenac	-
-	Ciprofloxacin	X
-	Amoxicillin	X
X	Macrolide antibiotics (Erythromycin, Clarithromycin, Azithromycin)	X
	Synthetic and natural hormones	
X	Estrone (E1)	X
X	17-Beta-estradiol (E2)	X
X	17-Alpha-ethinylestradiol (EE2)	X
	Sunscreen ingredients	
X	2-Ethylhexyl 4-methoxycinnamate	-
	Pesticides	
X	Methiocarb	X
	Herbicides	
X	Tri-allate	-

X	Oxadiazon	-
Insecticides		
X	Neonicotinoids (Imidacloprid, Thiacloprid, Thiamethoxam, Clothianidin, Acetamiprid)	X
-	Metaflumizone	X
Industrial compounds		
X	2,6-ditert-butyl-4-methylphenol	-

To sum up, the substances included in the Watch Lists are selected to produce and store high-quality monitoring data to assess the risks they pose at EU level. Furthermore, a substance can be taken out of the Watch List if enough high-quality, EU-wide monitoring data has been collected to allow an appropriate risk assessment, otherwise it has to remain on the list. The precise criteria that have to be fulfilled for the removal of substances from the Watch Lists are described in JRC Technical Reports - Review of the 1st Watch List under the Water Framework Directive and recommendations for the 2nd Watch List [13].

2. Strategies required to limit MPs from entering the aquatic environment

To reduce the impact of MPs (including pharmaceuticals) on the environment, a complex and coordinated strategy is required, including mitigation at the source and user side, to end-of-pipe strategies (Figure 1). According to the precautionary principle, applied in EU legislation, pollution and pollutants should rather be prevented and controlled at the source than be removed during the wastewater treatment process. Addressing the sources of MPs (production and imports), a responsibility of the producers is expected for preventing or reducing MPs input into the aquatic environment. Consequently, assessment of the environmental risks connected with the discharge of MPs into aquatic ecosystems, should be studied and transparently communicated e.g. by accurate information on labels. Consumers (householders and professionals) provided with such recommendations, can make responsible decision and take appropriate actions. Informational and educational campaigns should also be used to raise public awareness about proper usage, handling and disposal of chemical substances, which in water bodies behave as MPs [12].

It is also recommended that, if possible, the chemical compounds acting as MPs should be substituted by environmentally friendly ingredients, which are less/not persistent, bioaccumulative, and less toxic to the environment. Unfortunately, not all inputs can be prevented by these strategies. For example, many pharmaceuticals are absolutely essential in our healthcare systems and cannot easily be replaced by more environmental friendly alternatives. One group of pharmaceuticals of special concern are antibiotics. According to the WHO, antibiotic resistance is one of the biggest threats to global health today and new antibiotics are highly required to be developed. Thus, it is not realistic to put additional constraints, such as environmental factors, on the development of effective antibiotics [14].

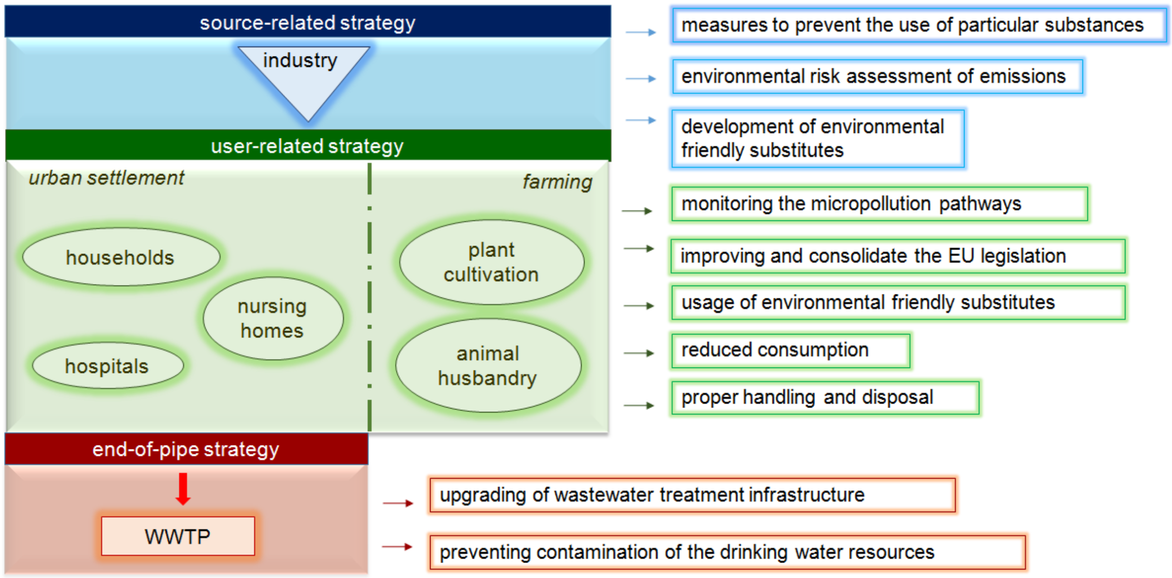

Figure 1. Strategies required to reduce the dissemination of MPs, including pharmaceuticals, into the aquatic environment

Currently, the EU strategy of preventing and reducing the fate of MPs in the environment is based on a multi-stakeholder dialogue (Figure 2) and guided by 'the precautionary principle' and 'the polluter pays principle' [15]. All above are essential (I) to introduce and integrated the relevant policies, (II) to implement more effective source-control measures, (III) to propose indicator substances for monitoring the MPs pathways at the local, national and EU-wide level, (IV) to develop practical and viable technological solutions (end-of-pipe technologies) for MPs removal and (V) to establish efficient financial programmes for investments in new infrastructure [15].

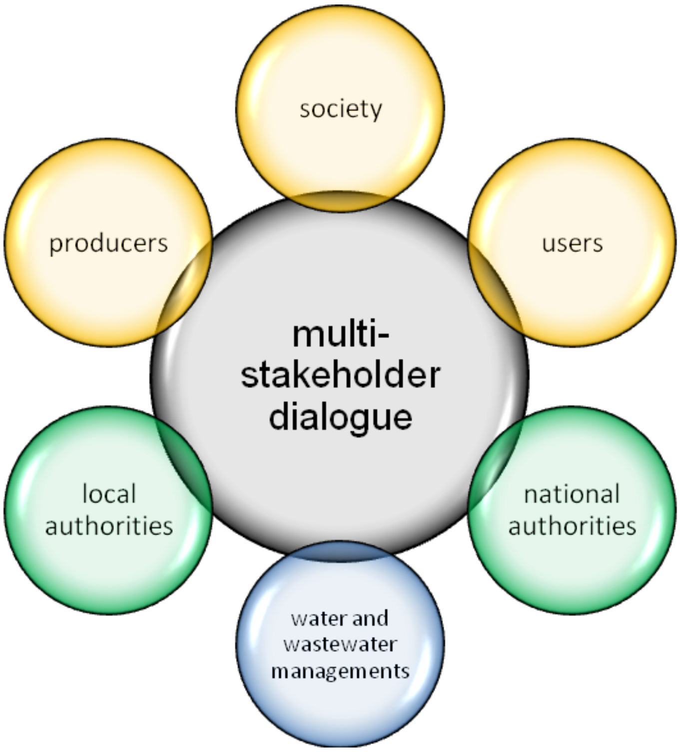

Figure 2. Multiple stakeholders involved in the dialogue on removal of MPs [14]

The actions mentioned above can create measurable reductions of MPs production and consumption, but it requires both time and large resources. Therefore, wastewater treatment processes by use of end-of-pipe technologies are today a major protective barrier in the water pollution control. They are very effective in organic matter, nitrogen and phosphorous removal but need to be upgraded in order to reduce trace substances [16].

3. Monitoring and removal of MPs, including pharmaceuticals, in selected EU countries - legal and recommended basis

The selection of four European Union countries for this study was based on the location of these countries in the South Baltic Sea, which is a common element of all described countries. In addition, it is important to supplement knowledge about monitoring and available techniques for MPs removal in countries in central-eastern Europe. The basis for monitoring and removal of MPs in Sweden, Germany, Lithuania and Poland are discussed below.

3.1. Sweden

According to the Swedish EPA, there are more than 1000 active pharmaceutical ingredients (APIs) in use on the Swedish market today [17]. The ability to analyse and identify these in complex environmental water samples depends on the availability of advanced technologies based on e.g. liquid-chromatography coupled to single or tandem mass spectrometry (LC-MS, LC-MS/MS) [18,19]. Additionally, specific methods directed towards the analysis of a variety of compounds must be developed or improved to increase significantly the knowledge about the presence of MPs in wastewater, surface water, ground water and the surrounding seas. The backside of including a large variety of compounds to the methods is an increased complexity which may hamper both the quality and the interpretation of data. Additionally, this complexity leads to increased costs of analysis. Thirdly, the results comparability might also be poor when the applied methods differ [19].

From a Swedish perspective, the Swedish Medical Products Agency identified the problems associated with analysis of pharmaceuticals, antibiotics and hormones in water and stated that there was a need for coordinated national analyses. In 2015, they issued a report in Swedish named *'Miljöindikatorer inom ramen för nationella läkemedelsstrategin (NLS) 2015'* - *'Environmental indicators in the scope of the national pharmaceuticals strategy (NLS) 2015'* [20]. In the report they stated: *'The working group considered the indicator 'measure levels of pharmaceutical substances in environment' to be of the very highest priority. This is because, besides it being of major importance to monitor the development of drug residues in the environment over time to evaluate the effect of implemented measures, the working group felt that there is considerable potential to optimise the use of the public resources through a better coordination of measurements in the environment. Many measurements have been taken historically by different public actors without any coordination.'* Furthermore, the Swedish Medical Products Agency writes: *'The working group's continued work came to focus on preparing proposals on substances that should be* *monitored in the environment, i.e. measurement of the occurrence of pharmaceutical substances in water, sludge, inlet and outlet water of treatment plants, biota, etc.'*. In total 25 pharmaceuticals were finally suggested; carbamazepine, ciprofloxacin, citalopram, clarithromycin, diclofenac, erythromycin, estradiol, ethinylestradiol, fluconazole, ibuprofen, ketoconazole, levonorgestrel, losartan, metoprolol, metotrexat, naproxen, oxazepam, sertraline, sulfamethoxazole, tramadol, trimethoprim and zolpidem.

In 2013, the Swedish Agency for Marine and Water Management published statutes containing regulations on classification and environmental quality standards for surface water [21]. These included assessment grounds for specific pollutants in inland surface water as well as in coastal water. This list of compounds included the pharmaceutical diclofenac. To conclude that surface water has good environmental status, the maximum concentration of diclofenac was set at 100 ng/L, expressed as a yearly average. The corresponding concentration for coastal water was ten times lower, with a maximum of 10 ng/L. These values may be further revised, as diclofenac has received much attention from different stakeholder groups lately, due to its high consumption as an over-the-counter drug, and for its harmful effects on the environment [22].

3.2. Germany

Since many years, the German environmental sector is aware of MPs, which is directly shown in the established national regulations, for example the Surface Water Ordinance. This Ordinance implements the European Requirements concerning EQS and regulates the emissions of so-called priority substances. However, this list of substances does not yet include pharmaceuticals even though suggestions for EQS have been made already by the Federal Environment Agency (UBA) in Germany. Another research funding program concerning risk management of new pollutants and pathogens in the water cycle (RiSKWa) developed a guideline for a list of indicator substances with the purpose to identify the sources of the pollution, indicate anthropogenic changes in water quality and control/monitor natural and technical treatment processes [23]. These chemical indicator substances included several pharmaceuticals but with the mentioned functional purpose – they do not represent the degree of pollution or water quality standards itself, human and ecotoxicological criteria were not included. Until now, these indicators are not adopted to legal regulations.

There have also been activities at the source of pharmaceutical products within admission of new substances. Since 1998, it is obligatory to perform an environmental risk assessment (ERA) within the admission procedure of human and veterinary pharmaceuticals in Germany according to the German Medical Products Act [24]. The European Medicines Agency published a guideline for the ERA of human pharmaceuticals where in the potential ecotoxicological risk has to be evaluated [25].

On a political level, a MP strategy is currently under discussion, and some federal states, in particular North Rhine Westphalia and Baden-Württemberg, have already upgraded some WWTPs with a fourth wastewater treatment step for removal of MPs. These regional activities are mainly a consequence of a pressure to act: The Rhine-Ruhr river basin is driven by a high population density. Besides the resulting large consumption loads of pharmaceuticals, the high population also leads to a high wastewater ratio inflow into the natural water bodies. Hence, the issue on MPs, including pharmaceuticals, first arose in the affected regions of Germany. Nevertheless, both regional and national research is now ongoing and a common national strategy is on the way to harmonize the concept and measures to reduce the discharge of pharmaceuticals into the aquatic systems [25].

3.3. Lithuania

In Lithuania long- and mid-term environmental approaches and water management policies are determined by two strategic documents adopted in recent years. In 2015, Lithuanian Seimas (Parliament) approved the National Environmental Protection Strategy. The Strategy define the priority areas of the environmental protection policy, long-term objectives up to 2030 and a vision for the Lithuanian environment, including water resources management up to 2050. For the reduction of dangerous chemicals in water bodies, the Strategy emphasizes the importance of applying innovative technologies and well-balanced use of plant protection substances [26].

To prevent harmful effects of wastewater discharges, these key implementing directions have been identified by the Strategy: *to raise public awareness about the aquatic environmental impact of wastewater; ensure that enterprises control priority hazardous substances that may be released into wastewater and that all generated wastewater is collected and managed in conformity with the established requirements; ensure the development and modernisation of wastewater management infrastructure through the efficient use of EU financial instruments.* More attention must also be paid to strengthened control of economic facilities, as well as implementation of an integrated pollution prevention and control system [26]. Specific indicators and measures are set in the medium-term Water Sector Development Programme for 2017–2023 approved by the Lithuanian Government and the Programme implementation Action Plan endorsed by Ministries of Environment and Agriculture in 2017 [27].

3.4. Poland

After accession to the European Union in 2003, Poland was obliged to harmonize the Polish law with European law, also in the wastewater sector. But environmental protection has been and still is one of the greatest challenges to EU integration, especially implementation of the EU directives such as the Water Framework Directive (2000/60/EC), the Drinking Water Directive (98/83/EC) and the Urban Wastewater Treatment Directive (91/271/EEC) (UWWTD). It was clear that besides the financial support for new investments, also know-how, new procedures, and new approaches are needed. Therefore, during the accession negotiations, the transitional period was prolonged for Poland in some cases, e.g. Poland was obliged to implement the UWWTD before the end of December 2015. To tackle the situation, the Polish government developed the National Programme for Urban Wastewater Treatment (NPUWWT), which entered into force in 2003. In the first version, the NPUWWT contained 1 378 agglomerations (> 2 000 PE), where 21 000 km of sewage networks and 1 163 of WWTPs had to be built, extended or modernized [3]. The NPUWWT was periodically revised and updated four times, but there is still no legal basis related to the fate of pharmaceuticals in the wastewaters and receivers. The WWTPs' discharges have to fulfill the Polish Regulations, set out in the Regulation of the Ministry of Environment from 2014 on conditions of discharges into water and soil and on substances particularly hazardous to the aquatic environment. This regulation came into force on the 1st of January 2016. These values match those of the Urban Wastewater Treatment Directive (91/271/EC) except for the Biological Oxygen Demand, for which the limit of the Polish regulation (15 mg/l) is actually more stringent than in the Directive (25 mg/l). The local authorities, however, require higher efficiency of treatment, depending on the receiver status [3].

Currently in Poland the total number of municipal WWTPs is about 3 000, but still 25% of the households have no access to the sewerage system, especially in rural areas, where construction of centralized WWTPs is often considered as too expensive. Polish regulations, in fact, allow the use of small/individual WWTPs, but only if there is no existing sewerage system in the area. In Poland, however, in rural areas septic tanks are more common than small WWTPs. A septic tank is used as part of a sewage network, for temporal wastewater accumulation, and cannot be regarded as a treatment system (only settlement of large solids occurs there). This type of wastewater is delivered to WWTPs by septic vacuum trucks and is usually higher contaminated than municipal wastewater reaching WWTPs via sewerage system. Additionally, in Poland it is often observed that the septic tanks are overflowing or leaking, causing contamination of groundwater resources.

It should be mentioned, that the legal basis for the national regulations imposing the need to assess the priority substances is the Water Act of 20 July 2017 [28]. In 2016, the Ministry of Environment (Journal of Laws No. 2016, item. 681) [29] published a list of 45 priority substances in accordance with Directive 2013/39/EU. Currently the Polish Inspection for Environmental Protection is monitoring priority substances for which environmental quality standards have been specified in flora and fauna, priority substances that tend to accumulate in sediments, and substances particularly harmful to the aquatic environment included in the Watch List. Additionally, pursuant to Art. 45 Section 1 Point 1 of this binding Water Act, the Ministry of the Environment is required to issue the Ordinance on the conditions to be met when introducing wastewater into water or soil and on substances particularly harmful to the aquatic environment (amending the current legal basis: Journal of Laws. No. 2014, item 1 800 [30]). Up to now, no changes have been made in the existing legislation. Poland, however, has indicated in the National Environmental Monitoring Programme for the years 2016–2020, the need to continue existing tasks (and to implement new ones) connected with EU requirements for the environmental monitoring system, especially the implementation of the Directive 2013/39/EU of the European Parliament and of the Council regarding priority substances in the field of water policy [27,28]. In the aquatic environment, the monitoring of harmful substances (including priority substances) should be carried out annually on water bodies at representative points.

3.5. Switzerland

To put the situation in the above four countries into a broader context, they can be compared to Switzerland, which already has regulations on both monitoring and removal of MPs in place. In January 2016 Switzerland imposed legislation that entailed large-scale and extensive WWTP expansion. The implementation of advanced wastewater treatment should be completed by 2040 involving 100 out of 750 WWTPs. This was the result of extensive research conducted for 10 years showing that MPs had negative effects on the aquatic environment downstream WWTPs, along with a risk of contamination of drinking water resources. There are various reasons why WWTPs of different sizes must be upgraded. WWTPs with a load of 80 000 persons or more should be upgraded since these WWTPs cover more than 50% of the Swiss population. Overall this will result in a large reduction of the total MP load to Swiss recipients. WWTPs dimensioned for 24 000 persons or more and that have a discharge to lakes should be upgraded to protect drinking water. Finally, WWTPs with a load of more than 8 000 persons discharging to recipients with insufficient dilution (>10% wastewater) must be upgraded protect sensitive recipients.

4. Current wastewater treatment system and MPs removal

The efficiency of conventional WWTPs varies depending on the pollutant's characteristics and on the treatment process employed. The main mechanisms for removal of micropollutants occurring during the secondary treatment at WWTPs are biological and/or chemical transformation and sorption [31]. Some MPs with low sorption coefficient, high water solubility and/or persistence to biodegradation may act as inert contaminants in the wastewater treatment process, passing unaltered through the WWTPs [32]. There are several factors that determine the effectiveness of MPs removal from wastewater - the most important are the properties of the MPs which are:

- molecular weight
- molecular size
- charge
- adsorption
- hydrophobicity
- biodegradability
- volatility

It should be noted that pharmaceuticals, antibiotics and hormones are a very diverse and inhomogeneous group of MPs and they cover a broad variety of differing properties. This may include, for example, size, charge and hydrophobicity. Apart from the properties of the MPs, other factors, such as plant configuration and operating conditions at the WWTP, are also important. This includes, for example, sludge age, hydraulic retention time (HRT), pH and temperature of the wastewater, and high differences in existing wastewater treatment systems (each WWTP is unique) [33]. It means that there is a large number of possibilities for combining them with advanced technologies. Thus, it is not possible to devise a single general solution for removal of all MPs such as pharmaceuticals at all WWTPs. If removal of MPs is planned as a complementary treatment by WWTPs, this decision should be preceded by:

- MPs monitoring in treated wastewater and receivers to prioritise the MPs of concern
- defining the requirements for MPs removal rate
- performance of comparable, pilot-scale and on-site tests to confirm the effectiveness of MPs removal by specific technology in existing WWTPs and to estimate the costs of this

technology, as well as to check whether this technology affects the existing wastewater treatment process and sewage sludge management

- knowledge transfer, by e.g. study visits at other WWTPs, where the same technology has already been implemented

Various technical solutions effective in micropollutants removal are available and have been proven to be possible to integrate with existing treatment processes in an expedient manner [33] (Figure 3).

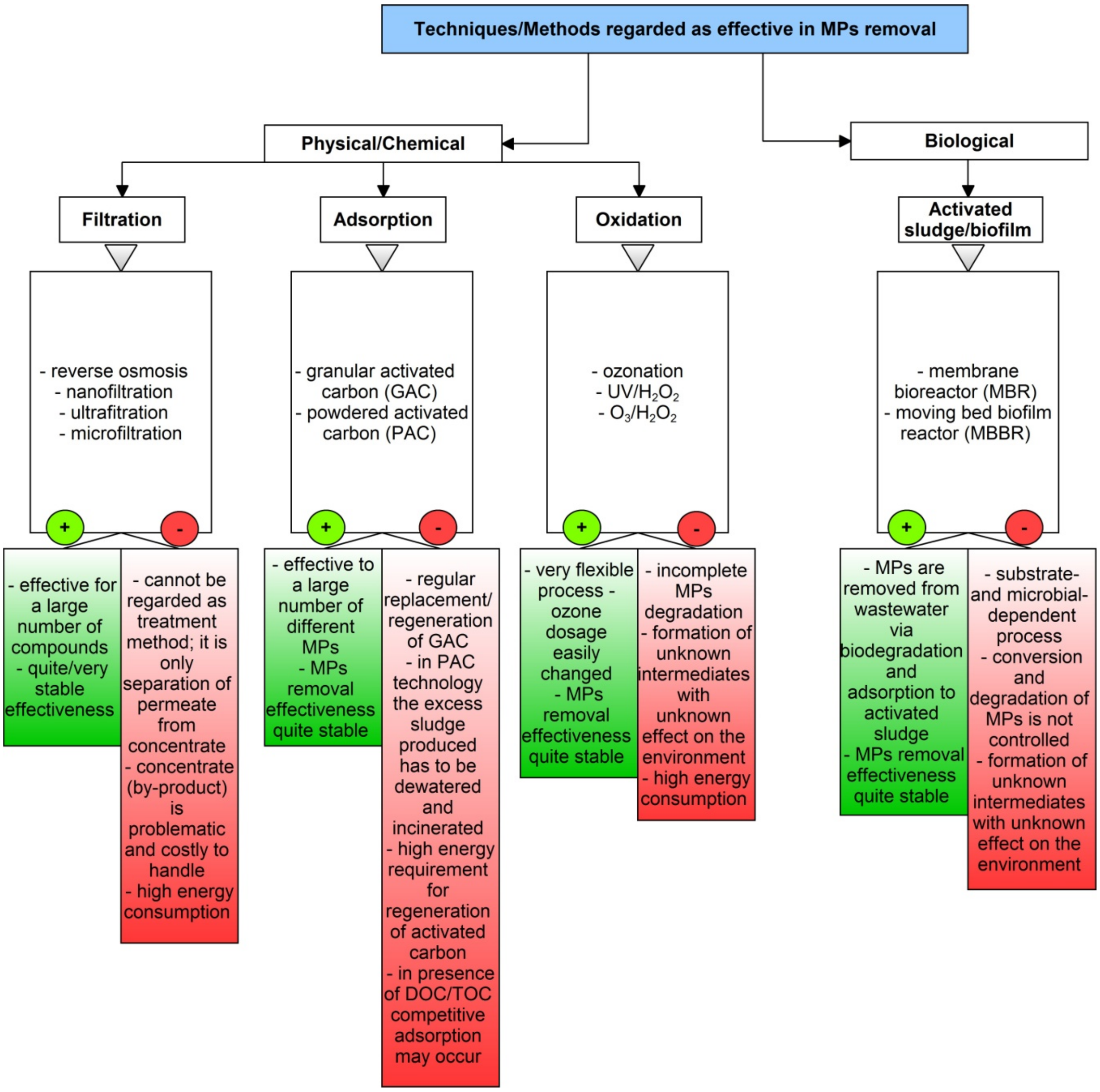

Figure 3. Technical solutions regarded as effective in micropollutants removal (including advantages and disadvantages of each technique)

Up to now, several processes such as filtration and adsorption to granular and powdered activated carbon (PAC and GAC), membrane systems (sometimes combined with biological degradation) and advanced oxidation processes (such as O_3, UV/H_2O_2 or O_3/H_2O_2) have been developed for the removal of MPs from wastewater. Various lab-scale, pilot-scale and full-scale studies have been conducted in several countries (e.g. Switzerland and Germany) in the past decade-and-a-half in order to investigate the application of advanced treatment in WWTPs [33].

5. Advanced technologies for pharmaceutical removal

Although, there are many possibilities of MPs removal, a large number of studies have concluded that primarily two technologies are capable of eliminating a broad range of micropollutants at reasonable costs: ozonation and activated carbon treatment [34,35], and thus, these two technologies will be the main focus of this paper.

4.1. Ozonation

Ozonation is in general an effective technology to reduce MPs in WWTPs. One of the benefits of using ozonation in aqueous solutions is that the hydroxyl ($OH^{\cdot}$) radicals, which are generated through the self-decomposition of ozone in water, react non-selectively with pharmaceuticals and other MPs. Ozonation additionally reduces some ecotoxicological effects, especially estrogenic activity. The disinfectant properties of ozone are also considered important advantages of this method in some cases [36].

There are also some disadvantages to this method. For instance, in an acidic environment, there is no spontaneous decomposition of ozone to free radicals. In a neutral environment, this decay is only partial - the processes take place in two ways - through a direct reaction with ozone and with radicals. Another issue is that in the production of potable water, the usage of ozone is limited if the concentration of natural bromide (Br^-) is significant, due to the formation of the carcinogenic bromate (BrO_3^-) in treated water. US EPA [37] and EU [38] quality standards limit the concentrations of BrO_3^- in drinking water to 10 μg/L. In wastewater technology, it is suggested that several MPs are not completely mineralised under the ozone dosages applied today, which are about 0.6–1.0 g O_3 per g DOC (dissolved organic carbon) and hydraulic retention times of about 20–30 minutes. Consequently, during ozonation, the MPs are transformed into other compounds, which may not be completely removed from the effluent. This formation of intermediates, which can be more toxic than the parent compounds, is a critical, extensively studied topic of ozonation [36]. Up to now, however, significant production of toxic by-products in full-scale WWTPs has not been noted. Nonetheless, for example in Germany, it is advised to implement ozonation with a post-biological or sand-filtration step, to remove any biodegradable transformation by-products before release into the recipient. The effectiveness of sand filtration in removing reactive compounds is, however, not fully recognized. Germany's suggested general design criteria for MPs removal are given in Table 2 [35].

Table 2. General design criteria for removal of MPs from biologically treated wastewater by ozonation unit in Germany [35]

Subject	Unit	Value
	Ozonation	
Dosage	g O_3 / g DOC	0.6–0.9
	mg O_3/l [a)]	4–14
Hydraulic Retention Time Contact Tank	minutes	15–30 (reactor 10–25 min; removing remaining ozone 5 min)
Power consumption	kWh/kg O_3 × h	10

	W/treated m^3	45
	Sand filtration after ozonation [b)]	
Upflow velocity	m/h	12
Backwash water	% of incoming flow	5–10
Power consumption	W/treated m^3	15

[a)] based on dissolved organic carbon (DOC) content in WWTP effluent of 7–15 mg/L

[b)] similar criteria for sand filtration after PAC

From a technological point of view, it can be noted that ozone is unstable, and thus cannot be stored on site, but must be produced directly prior to its application. The ozone is generated from pure oxygen or air through electrical discharge. After ozone has been generated it is mixed by injectors or diffusers with the effluent water of the WWTPs in a contact basin. It has been noted that energy consumption is slightly higher for the injectors and no increased removal of MPs was found compared to the use of diffusers [35]. Therefore, the latter are regarded as the better solution. The ozone–wastewater contact basin is air-tight, as the remaining ozone in gaseous form has to be treated. The effluent of the contact basin is then passed through a sand filter to remove any biodegradable metabolites.

It should be noted that during ozone generation, oxidation of nitrogen can also take place, which in the presence of moisture may form nitric acid. In order to avoid corrosion of the ozonator, the air or oxygen must be moisture-free, which is achieved by cooling or drying the gas. Additionally, it is important to note that a concentration of ozone in air greater than 1 ppm is considered unsafe for prolonged human exposure [39]. A diagram of a typical ozonation system at a WWTP is shown in Figure 4.

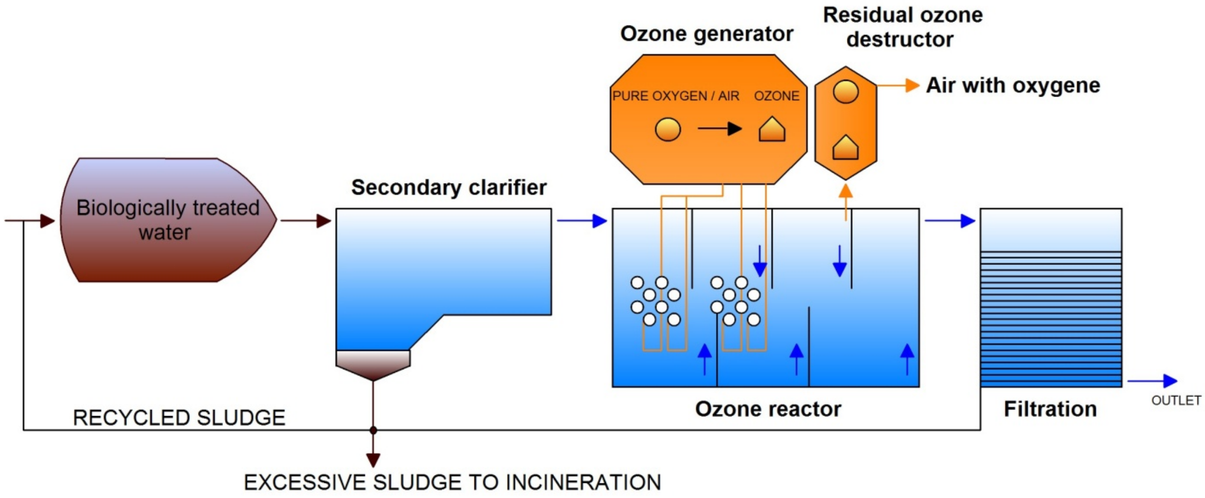

Figure 4. Example of ozonation system at WWTP - based on [39]

In legislation developed by the EU concerning air quality standards, the maximum allowable concentration of ozone in air should not exceed 120 µg/m^3 for continuous human exposure for 8 hours or 180 µg/m^3 for one-hour exposure [40]. WHO standards set a value of 100 µg/m^3 exposure per 8 hours [41]. Consequently, ozone detectors and warning systems should be present in buildings and other places where ozone is produced and used.

4.2. Activated Carbon

Activated carbon is commercially available in granular (GAC) and powdered (PAC) form, and is widely used as an adsorbent in many industrial processes due to its microporous, homogeneous structure. GAC typically has a particle size diameter ranging between 1.2 and 1.6 mm, while PAC has a particle size diameter smaller than 0.2 mm, typically in the range of 5–50 µm. The surface area of activated carbon is very large, normally ranging from 500 to 1.400 m^2/g [42]. Activated carbon properties depend on the surface area, pore volume and distribution of pore size, and the material used for production (Figure 5). Currently, activated carbons can be produced from a variety of materials of high carbon content that are activated at high temperatures (>700°C). Common raw materials are coals (anthracite, bituminous and lignite), coconut shells, wood, peat and petroleum residues [43].

The effectiveness of activated carbon in organic matter removal, including MPs, is generally connected to the physical properties of the compounds. Commonly, hydrophilic compounds are less adsorbed than hydrophobic substances. However, the charge of the MPs is also of great importance, where negatively charged pharmaceuticals bind less hard than those positively charged. Neutral pharmaceuticals bind more strongly than negative ones, but less so than positive pharmaceuticals. Additionally, the removal rate of hydrophilic MPs is greatly influenced by the presence of other organic matter, especially hydrophobic contaminants, due to the competitive adsorption (hydrophobic compounds are usually more easily and strongly adsorbed to activated carbon) [43].

4.2.1. Granular Activated Carbon technology

The advantages of MPs removal by GAC technology include its simple application, operation and maintenance. GAC treatment in WWTPs is often applied as a single filtration step by a fixed bed filter as exemplified in Figure 5.

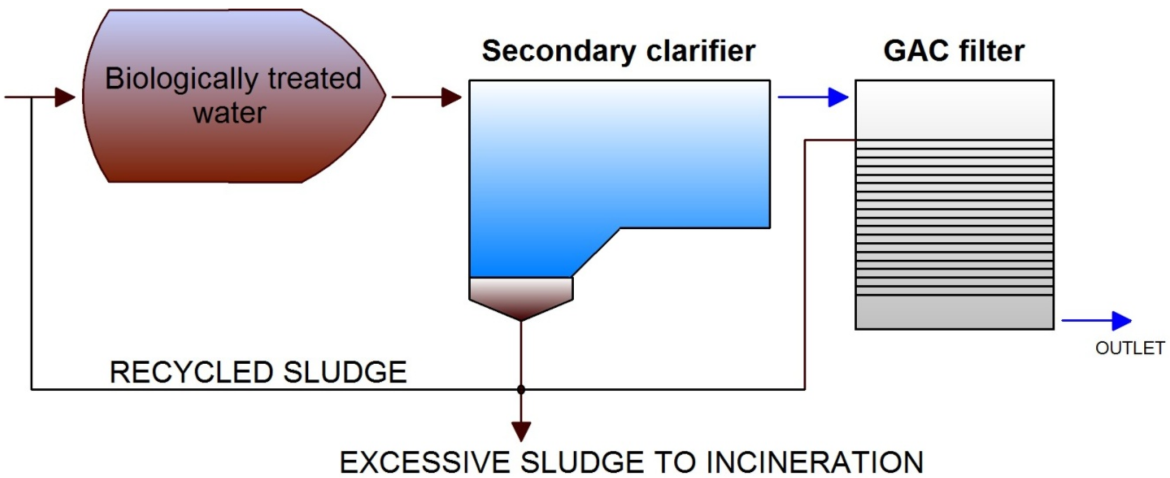

Figure 5. Example of a GAC system at a WWTP – based on [39]

The incoming water flows downward under the force of gravity through the GAC medium, which is usually placed in a cylindrical tank. It should be noted that the presence of organic matter may reduce the effectiveness of MPs removal due to competitive adsorption..

A schematic illustration of the adsorption of small and large organic molecules onto an activated carbon particle is shown in Figure 6 [43]. Additionally, the blocking of GAC filter pores may become faster, when high amount of organic material is present. As a consequence, if the settling tank at a full-scale WWTP does not function well, and the incoming water to the GAC filter contains suspended solids at higher than 10 mg/L, the GAC-filter should be bypassed [35]. To avoid clogging of the GAC filters, they are therefore sometimes preceded by sand filters which improves the situation by removing/changing some of the organic material before entering the GAC filter. Parameters important in MPs removal are presented in Figure 6.

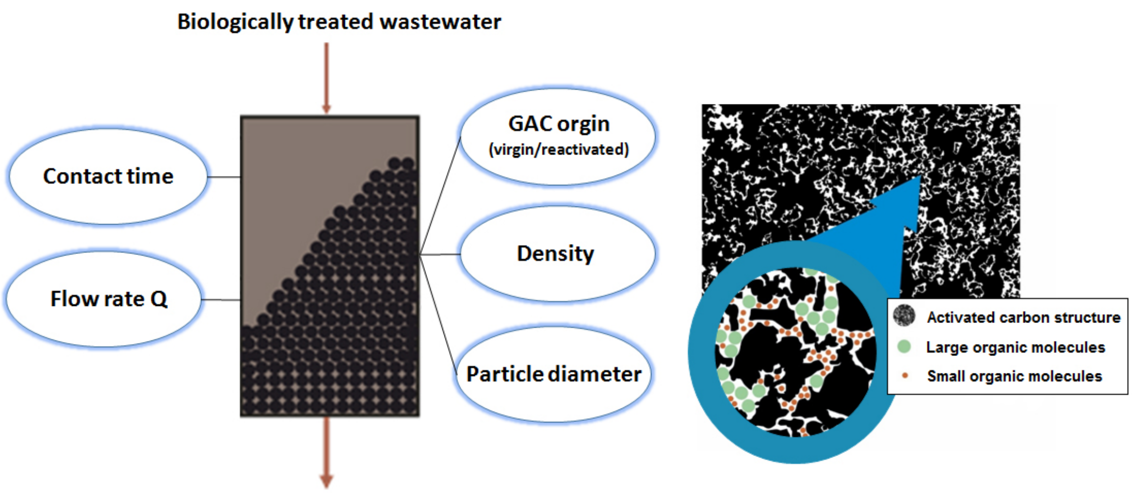

Figure 6. Parameters important in MPs removal by a GAC treatment step – based on [38] including illustration of organic molecules adsorption onto an AC particle - based on [43]

Periodically, to remove organic matter and prevent blockage, the GAC filters may have to be flushed backwards with clean water, and this 'backwashed' water is then directed back to the WWTP. If backwashing with water does not solve the problem, the GAC filter can be flushed with pressurised air. Additionally, the GAC filters have to be replaced once the effectiveness of the targeted compounds removal begins to drop. This means that all adsorption sites on the activated carbon are filled with contaminants. Reduced performance of the GAC filters is a signal to refill the filter with new or reactivated GAC. The advantage of GAC technology over PAC technology is connected with the possibility of GAC thermal regeneration. During this process, all adsorbents are volatilised and/or degraded (mineralised) and the adsorption capacity is completely restored. However, during regeneration, about 10% of the GAC mass is lost [35,39]. Some general design criteria suggested in Germany for MPs removal by GAC are given in Table 3.

Table 3. General design criteria for removal of MPs from biologically treated wastewater by GAC units in Germany [35]

Subject	Unit	Value
	GAC	
Empty Bed Contact Time	Minutes	20–40
Upflow velocity	m/h	6–10
Backwash water	% of incoming flow	5–15
Power consumption	W/treated m^3	40
Replacement coal	-	After 7000–15000 bed volumes (standing time 4 months to 1 year)

4.2.2. Powdered Activated Carbon technology

In PAC technology, the effluent of the WWTPs is treated in a separate system, consisting of a contact tank, a settling tank and a filter [35]. The PAC system is usually located after the existing biological stage (Figure 7A). To the contact tank, together with PAC, flocculants and coagulants are dosed (e.g. Al/Fe solutions). Due to the small size of PAC, its particles remain in the effluent, and a post-treatment is thus needed, mainly as a filtration step: sand, membrane, activated carbon filtration. The sludge from the PAC system is usually partly recycled to the contact tank but, optionally, can also be recycled to the biological step, e.g. to the aeration zone. Alternatively, the PAC can be dosed directly into the aeration tank of the activated sludge step (Figure 7B) or to the inlet of existing sand filters. Direct dosing to biological treatment may significantly reduce the investment costs but this solution is still under investigation, since it is still not clear how either final or direct PAC dosing influences the effectiveness of MPs removal or the effectiveness of the existing treatment system [39].

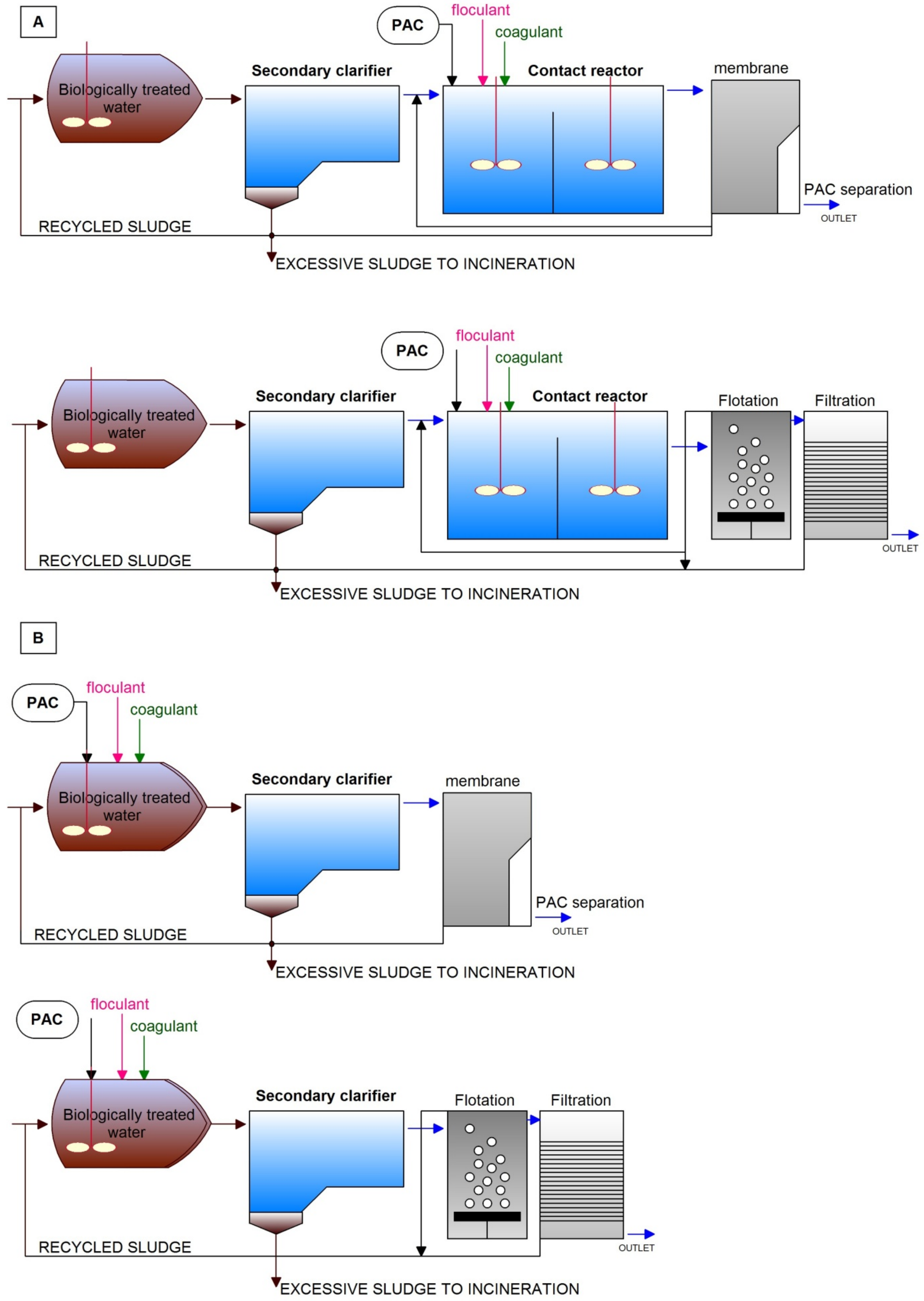

Figure 7. A - two PAC systems with separation by membrane and flotation (PAC system located after the existing biological stage); **B** - two PAC systems with separation by

membrane and flotation (PAC system dosing directly into the aeration tank of activated sludge stage) – based on [39]

The dosages of PAC normally applied to WWTP effluents vary from 10 to 20 mg PAC/L. It should be noted that PAC dosage increase the amount of routinely dosed polymers and precipitation solutions by approximately 10% to 20%. Several PAC storages and feeding systems into wastewater are currently commercially available. The installations usually consist of a storage module, gravimetric feeding devices and a dissolving/mixing unit, which provides the optimal dose, depending on the volume and quantity of wastewater. Importantly, high feeding accuracy reduces operating costs. Additional PAC can react with oxygen, releasing heat [35, 44]. Additionally, PAC dosage systems should be designed in accordance with special regulations dealing with occurrences of sparks, since PAC can be explosive in the form of dust. Other disadvantages of PAC treatment for MPs removal is the clogging of the carbon slurry transport systems. This is mainly the result of undersized piping systems, short and sharp radius bends, insufficient velocity, and lack of cleaning. Additionally, abrasion of pipes transporting the slurry is a common problem. Those problems can be solved by increasing the size of the piping, using glass- or rubber-lined-steel or coated-cast-iron pipes [39].

Additionally, MPs removal requires a certain PAC dosage. Spent PAC is continuously removed from the system, and usually dewatered, dried and finally incinerated, which limits the further dissemination of the pollutants into another environment. Some general design criteria suggested in Germany for MPs removal by PAC are given in Table 4.

Table 4. General design criteria for removal of MPs from biologically treated wastewater by PAC units in Germany [35]

Subject	Unit	Value
	PAC	

Dosage	g PAC / g DOC	0.7–1.4
	mg PAC /L [a)]	10–20
Dosage coagulant	mg/L	4–6
Dosage polymer	mg 100% active /L	0.2–0.3
Hydraulic Retention Time Contact Tank	Minutes	30–40
Surface load settler	m/h	2.0
Recycle factor PAC	-	0.5–1.0
Power consumption	W/treated m^3	45
Sand filtration after PAC [b)]		

[a)] based on dissolved organic carbon (DOC) content in WWTP effluent of 7–15 mg/L

[b)] similar criteria as for sand filtration after ozonation (see Table 3)

4.3. Process control and automation

The process parameters and effectiveness of MPs removal should be easily controlled by process automation both in GAC and PAC technologies. In the case of GAC filters, the important issue is the accuracy of velocity through the filter bed volume. Another important issue is the clogging of the GAC filter [45], which can be measured through pressure changes. Pressure increase beyond a threshold value should automatically start the back-flushing of the filter bed with air or water. To estimate the lifetime of a GAC filter and the need to replace the activated carbon, break-through curves need to be established by periodical MPs measurement in the quaternary treated effluent [39].

For both ozone and PAC technology, accurate dosage is an important issue in MPs removal. Currently, ozone and PAC can be easily over- or under-estimated because dosages are adjusted to the flow of biologically treated wastewater, while the effectiveness of those technologies is strongly linked to the DOC concentration. At full-scale installations direct online measurement of DOC concentration is rather difficult and not accurate enough. For this reason, at several applications, indirect measurement was applied based on the loss of UV light absorption at 254 nm (UVA254) [46]. Besides the dosage, the contact time is also important in MPs removal, and further optimization of ozone and PAC dosage is needed.

6. Cost estimation of ozonation and activated carbon treatment technologies

The ozonation and activated carbon treatment technologies may be considered the most effective compared to the costs incurred. In estimating the costs of MPs removal from wastewater, several parameters need to be taken into account, as shown in Figure 8.

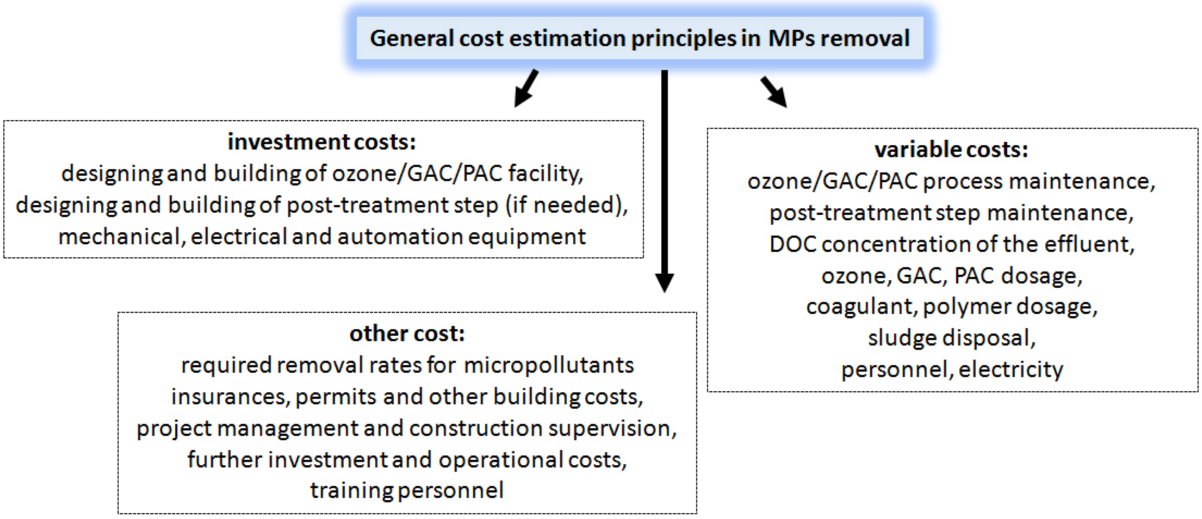

Figure 8. Parameters to consider in calculating the cost of MPs removal from wastewater [35]

In general, it can be concluded that the overall investments costs for ozonation and PAC treatment are similar, while for GAC treatment they are usually significantly lower, due to the simplicity of this technology's installation [35]. In energy consumption, ozone technology usually requires double the energy of PAC treatment and up to 12 times more than GAC treatment. However, the overall maintenance costs of ozonation are the lowest, since PAC treatment requires continuous dosage of PAC, coagulants and polymers, as well as sludge treatment (usually dewatering and thermal processing), while for GAC treatment the maintenance costs are connected with the periodical need of activated carbon exchange/regeneration. When comparing the viable costs of GAC and PAC treatment, it may be assumed that GAC technology is more expensive per m^3 of treated wastewater than PAC technology [35], including both GAC replacement and the sludge processing with a post-treatment step in PAC treatment. However, a very recent 4-year Swedish study in the FRAM project (Full-scale treatment of micropollutants) [3] performed at Kristianstad University showed that the cost of GAC filtration can be reduced significantly by an appropriate protection of the GAC filter from unwanted organic material using a common sand filter as a pre-filtering step.

There are also trials to reduce the costs of this technology by using biochar instead of GAC. For example, the System Läk project (Systems for the purification of pharmaceutical residues and other emerging substances) [47] conducted adsorption tests using different types of biochar, and some could reduce pharmaceutical residues from particular treated wastewater with a capacity comparable to that of commercially available activated carbon.

In cost calculations one has also to be aware that when costs are presented in the literature, the MPs removal is sometimes separated from post-treatment, so costs connected with the design, building and maintenance of post-treatment installations may not be included in the overall costs needed to build and maintain MPs removal technology. It was concluded, that depending on the technique, the costs of post-treatment steps for MPs removal may vary from 0.16 to 0.33 EUR/m^3 in treated effluent [35]. Between countries, differences in, for example, electricity and labour expenditure may also influence the final cost calculations for advanced technologies.

Based on the relatively large number of studies performed primarily on ozonation and PAC, there are several documents available that make it possible to compare the two technologies. An overview of system costs and design criteria for ozonation and PAC is shown in Table 5, based on experiments performed in German.

Table 5. Overview of system costs and design criteria for ozonation and PAC during implementation in German WWTPs [48,49,50]

Criteria	Ozonation	PAC
Primary Energy demand	0.09–0.37 kWh/m³	0.05–0.08 kWh/m³
Primary Energy demand (production and transport)	0.03–0.09 kWh/m³	0.36–0.72 kWh/m³
CO_2 emission	60–130 g CO_2/m³	150–240 g CO_2/m³
Yearly costs	0.02–0.14 €/m³	0.04–0.20 €/m³
Operation	High degree of automation	Low degree of automation
Space requirement	Low	High for contact and sedimentation tanks, low for dosing into filters or activated sludge reactor
Advantages	Slight disinfection	Loaded carbon coal (incinerator)
Disadvantages	Transformation products, biological follow-up treatment required)	Expanding the sludge amount, potentially PAC-drifting into receiving water, increasing CO_2 emissions

7. Possibilities of MPs reduction or/and removal strategies implementation in EU countries, and experiences from Switzerland

The decision to start reducing MPs dissemination via WWTPs, in Switzerland, was preceded by a 10 year long preparation and testing phases which included technical, ecological and socioeconomical aspects with special attention given to the environmental burden. In the preparation phase the crucial step is to define the local objectives and criteria for advanced treatment, while at the testing phase critical analysis of the most promising alternatives should be conducted by lab-, and or pilot-scale studies. It is important to correctly estimate the on-site effectiveness of the tested advance treatment as well as the costs of implementation and maintenance (Figure 9).

According to the obtained results it was decided that both WWTPs and technical innovations should follow the below criteria [51]:

<u>Selection of WWTPs for upgrading</u> has to be based on:

- the anticipated MPs load of WWTPs serving > 80 000 people, upstream responsibility
- dilution capacity of wastewater receiver if wastewater consists of > 10% of dry-season stream flow
- protection of sensitive areas and water bodies feeding drinking water reservoirs

<u>Selection of technical innovations</u> has to be based on:

- effectiveness on as broad a range of micropollutants as possible
- flexibility and accessibility for implementation in existing infrastructure without disturbing existing processes
- acceptable cost/benefit ratio

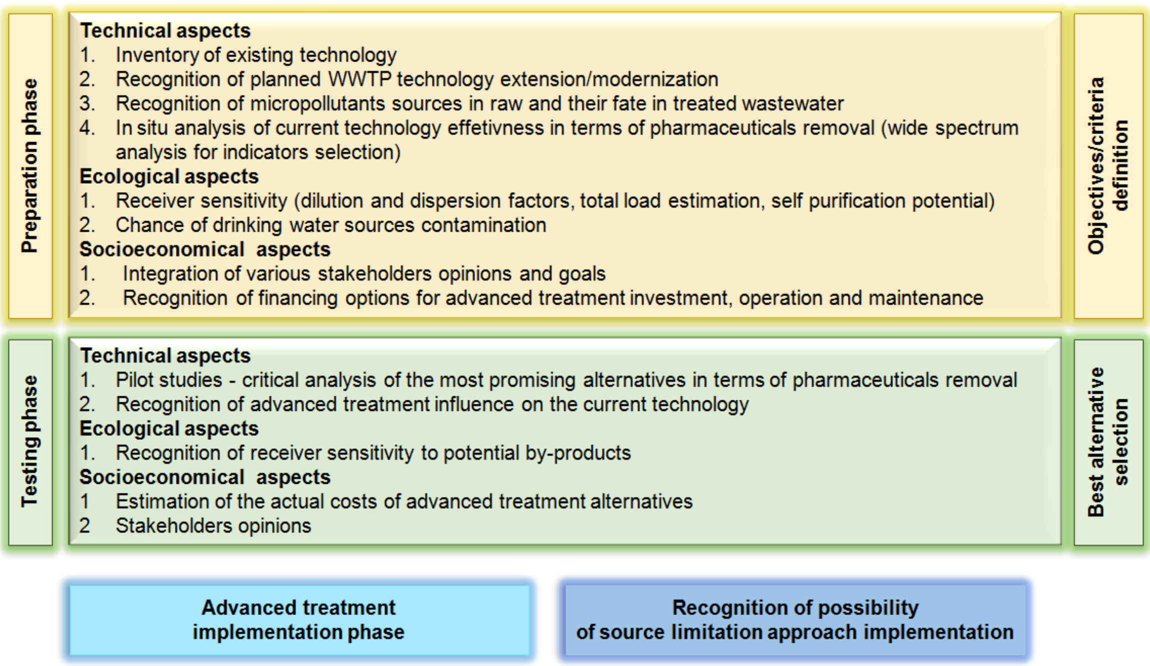

Figure 9. Decision making criteria for implementation of advanced treatment at wastewater treatment plants

This article mainly focuses on two technologies (ozonation and activated carbon treatment) for the removal of micropollutants, including pharmaceuticals, from wastewater in the four selected EU countries Sweden, Germany, Lithuania and Poland, all located in the South Baltic Sea region. The various technological implementation strategies in these four selected countries are presented below.

7.1. Sweden

In Sweden, a number of research and development projects dealing with pharmaceuticals in the environment have been carried out since 2005. From 2005 to 2009, a local but large Swedish project was run by Stockholm Water called 'Pharmaceuticals - occurrence in the aquatic environment, preventive measures and possible treatment methods'. The final report was published in 2010 [52]. In the project various complementary methods were tested based on either biological, oxidizing or separating principles and it was found that several of these methods worked well for removing pharmaceuticals from wastewater. However, ozone or activated carbon were the most promising technologies from a holistic perspective. It is noteworthy that already in 2009, from a Swedish perspective, it was stated that activated carbon had both physicochemical and ecotoxicological benefits. This is probably due to the different mechanisms by which the two technologies reduce the presence of pharmaceutical residues in water. Activated carbon adsorbs the substances so that they can no longer be detected by the analytical chemical technique (LC-MS/MS) since they are physically separated from the water phase. Ozone, on the other hand, converts the substances into new 'unknown' chemical compounds with 'unknown' effects on organisms. Admittedly, the pharmaceuticals can no longer be detected with LC-MS/MS after treatment with ozone (which is chemically very reactive), but this is not the same as there being no active molecules left in the water after this oxidative process. Despite several advantages of activated carbon and an uncertainty associated with ozonation from an ecotoxicological perspective, ozone became the technology that was ultimately recommended in the report by Stockholm Water 2010 [52]. Between 2008 and 2015, the Swedish Foundation for Strategic Environmental Research funded a project called MistraPharma. The projects resulted in a large number of scientific articles on a great variety of topics, and a final report published in 2016 [53]. The key outcome of the research was a policy brief with ten recommendations for improving environmental risk assessment [54], but part of the research was devoted to wastewater treatment technologies. According to the final report various pilot plants for ozonation and activated carbon (GAC and PAC) were constructed. The key finding for ozone was that with an appropriate ozone dose of 5–7 g O^3/m^3, ozonation a removal efficiency of 85–95% was reached, with lower biomarker responses than today's effluent. However, sand filter treatment after ozonation did not improve the removal of pharmaceuticals. Key findings for activated carbon was that PAC and GAC systems showed the highest removal of pharmaceuticals, 95–98%. The dose of activated carbon was in the range of 15–70 g prod./m^3. In PAC systems, the activated carbon consumption was typically one half to one third that of GAC systems [53].

Between 2014-2017 the Swedish Agency for Marine and Water Management financed a number of projects aimed at developing technologies for advanced treatment of wastewater from pharmaceuticals and other hazardous substances e.g. activated carbon and ozonation. These projects should be done in close collaboration between universities and municipalities, shifting focus from being mainly scientific projects into being more society-based projects by performing treatment of wastewater at selected WWTPs in pilot-scales or larger. The final report summarizing the results from these projects were published in 2018 and concluded that both activated carbon and ozonation could be used in Sweden to remove MPS [55].

In December 2015, the Swedish Government commissioned the Swedish Environmental Protection Agency to investigate the possibility of implementing advanced treatment of wastewater to remove pharmaceuticals from wastewater in order to protect the aquatic environment. The final report was published in 2017 [56] and it was concluded that emission of pharmaceuticals can be reduced by equipping Swedish WWTPs with more advanced technology, such as carbon filters or ozone treatment. The Swedish EPA stated that the next step was to investigate which WWTPs that primarily should be upgraded with advanced treatment, but found out that this could not be specified with the limited number of occurrence data in Swedish. The Swedish EPA concluded that several factors are of major significance to prioritise where the first installations should be made. This included local conditions such as:

- The amounts of pharmaceuticals that are discharged into the recipients
- The water turnover of the recipient
- The number of WWTPs that discharge into the same recipient
- The sensitivity of the recipient
- Yearly variations
- Variations in discharged amounts from the WWTP

Presently the Swedish EPA is financing several pre-studies and full-scale installations for *Treatment of pharmaceutical residues* various selected WWTPs to support Swedish municipalities who are at the edge of upgrading their WWTPs with advanced treatment and wants to try out these new technologies.

7.2. Germany

The micropollutants removal strategy in Germany is in progress and will be developed at both the national and the regional level by involved actors such as the Federal Environmental Agency and the German Working Group on water issues of the Federal States and the Federal Government, represented by the Federal Environment Ministry (abbrev. LAWA) [57]. The main goal of the German micropollutants strategy is to harmonise both regional and national approaches and data of finalised studies. In November 2016 a multi-stakeholder dialogue was launched as part of the preparations for a federal-government strategy to mitigate MPs in the aquatic environment [16]. Currently, the following main criteria are suggested for use:

- ecological sensitivity of receiving water body

- conservation of water resources for potable and leisure uses (bathing waters) – upstream responsibility
- efficiency and cost-effectiveness criteria (such as the size and state of wastewater management facilities)
- pollution charge of receiving waters

In Germany, 16 full-scale treatment plants in North Rhine-Westphalia and in Baden-Württemberg have currently been upgraded with a fourth treatment step, 6 installations are currently under construction, and such treatment is planned for another 11 WWTPs. There are plans also for WWTPs in other federal states (e. g. Berlin, Bavaria and Hesse). It is supposed that the experiences gained with these plants can be compared with the results of various lab-scale research projects [16]. Various advanced processes are available for MPs removal, but in Germany so far only two methods are regarded as technically feasible on a larger scale: I) oxidation with ozone, and II) adsorption onto activated carbon (PAC or GAC), or a combination of these two methods. They are feasible for plant operators, and it is assumed that, appropriately equipped and managed, WWTPs may obtain a reduction of 80% of many MPs. The elimination rate is, however substance-specific and depends on the treatment technology. It is also expected that besides the MPs, ozonation and activated carbon give opportunity to enhance the removal of other organic compounds and/or to improve the hygienic quality of the WWTPs effluents [16].

7.3. Lithuania

In Lithuania, long- and mid-term environmental approaches in water management policies have been determined by two strategic documents adopted in recent years: the National Environmental Protection Strategy and the Water Sector Development Programme for 2017–2023 [26]. To improve the status of surface and groundwater bodies and to achieve and maintain good environmental status of the Baltic Sea, the studies carried out mainly involve reducing agricultural pollution (diffuse sources) and increasing treatment efficiency in 12 WWTPs (point sources). However, in Lithuania, no measures are integrated in the mentioned strategic planning documents to implement advanced treatment for the removal of

MPs, including pharmaceuticals, in wastewater. Nevertheless, pilot investments in technological solutions for removing pharmaceuticals and other micropollutants are introduced in Kretinga town WWTP. Substances from the Watch List are included in the Lithuanian State Environmental Monitoring Programme for 2018-2023 [26].

7.4. Poland

Currently in Poland, no large-scale systems aiming to limit the discharge of pharmaceuticals and other MPs into the aquatic environment are being implemented at municipal WWTPs. However, in 2015 the National Environmental Monitoring Programme for the years 2016–2020 was established, and adapts the current European strategic documents in water monitoring, in particular the need to monitor priority substances in the field of water policy as provided by Directive 2013/39/EU [10]. There is also an interest in this topic, as expressed by the participation of Polish institutions in European projects aiming to test new, cost-effective technological solutions for the removal of pharmaceuticals and other micropollutants by upgrading existing wastewater treatment systems e.g.: the two Interreg South Baltic projects MORPHEUS (http://www.morpheus-project.eu) and LESS IS MORE (https://www.swedenwaterresearch.se/projekt/less-is-more/).

To sum up, in Germany, the national micropollutants strategy is currently in the consulting period and in the process of defining new regulations for advanced wastewater treatment. However, there are not yet any legal requirements, neither for the application of technologies removing MPs, nor for pharmaceutical thresholds. Some federal states, in particular North Rhine Westphalia and Baden-Württemberg have already equipped several WWTPs with a fourth treatment stage on a voluntary basis. The technologies for MPs removal in Germany are mainly based on ozonation and activated carbon.

In Sweden, the government has already funded several projects related to MPs removal from wastewater (mainly pharmaceuticals). Currently, the knowledge and operating experience of various technical solutions are completed and available as a foundation for the full-scale introduction of advanced treatment at WWTPs. Primarily ozonation and activated carbon have been tested and suggested as realistic alternatives for upgrading Swedish WWTPs at the national scale.

In Poland and Lithuania, there is neither a legal basis, nor other documents related to monitoring and/or removal of pharmaceuticals from wastewater. However, both countries are introducing national regulations imposing the need to assess priority substances. In Poland, for some substances particularly harmful to the aquatic environment, the maximum permissible values of pollution indicators have been specified for industrial wastewater.

8. Conclusions

The access to clean water is of ever-increasing significance. Worrying is the fact that the number of anthropogenic substances in waters grows daily due to newly introduced products, and this threatening situation becomes increasingly clear due to improved analytical technologies and methodologies [10,11]. Of growing concern are MPs, such as pharmaceuticals and personal care products as well as steroid hormones, surfactants, industrial chemicals and pesticides [58]. MPs, such as pharmaceuticals, easily enter surface waters following discharge from WWTPs since many of them are notcompletely removed during the conventional wastewater treatment processes. MPs in surface waters can be detected at trace levels. Since some of the are semi-persistent in the environment they can spread through water and soil as well as accumulate in plants or wildlife, where may pose a risk due to their potential toxicity [6]. The accumulation of MPs in the environment is the cause of many environmental problems, and for pharmaceutical this may include drug resistance and fish feminization among others [59]. An essential legal EU obligation to mitigate MPs arises from the European Water Framework Directive (WFD) and the environmental quality standards (EQS) for priority substances. Currently, besides listing priority substances (45 compounds or groups of compounds), Directive 2013/39/EU and Commission Decision 2018/840/EU have also implemented so-called Watch Lists including selected pharmaceuticals which should be temporary monitored in surface water to obtain high-quality data sets. The monitoring of pharmaceuticals is important not only to determine the risk posed by them to the aquatic environment, but also, in the case of antimicrobial agents, to support the European One Health Action Plan against antimicrobial resistance. Tangible existing evidence regarding pharmaceuticals in the environment includes the results of several completed and running projects in EU. More information is still needed to understand and evaluate certain pharmaceuticals as regards to their environmental concentrations and the resulting levels of risk. One reason is that many pharmaceuticals put on the market several years ago, were not subject to an environmental risk assessment as part of the authorisation process. Another reason is that monitoring of pharmaceuticals in the environment is still very limited. Although selected substances are monitored in surface and groundwaters under the Water Framework Directives, monitoring could be extended to better cover certain parts of the environment, where involving cooperation with stakeholders is necessary [6].

Moreover, it has already been pointed out that besides monitoring, new treatment technologies efficient in degrading or removing antimicrobials in wastewater to reduce the spread of antimicrobial resistance, are of high concern. But the lack of EU recommendation on effluent standards for MPs has postponed the implementation of new technologies, e.g. ozonation and activated carbon treatment, in the wastewater sector. Additionally, in principle the EU policy says that the polluter pays, but in terms of MPs the subject is very complex. It is not clear who the polluter is, since stakeholders for example can be producers of chemicals, the pharmaceutical sector, hospitals and various consumer groups. Therefore, two approaches need to be developed simultaneously:

- substitute critical MPs production and usage (source and user measures)
- mitigate the dissemination of MPs by WWTPs (end-of-pipe measures). Since not all of the substances, particularly pharmaceuticals, can be replaced with harmless alternatives, end-of-pipe technologies seem to be an essential part of the solution

It can be concluded that there are no requirements to remove pharmaceuticals from wastewater within the European Union, but there is a need, posed by the European Commission and other organisations, to monitor them at a European level. Additionally, we need to develop methods to investigate the feasibility of upgrading selected urban WWTPs to more advanced treatment technologies capable of eliminating a broad range of MPs at reasonable costs.

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