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Treatment of micropollutants in wastewater: Balancing effectiveness, costs and implications

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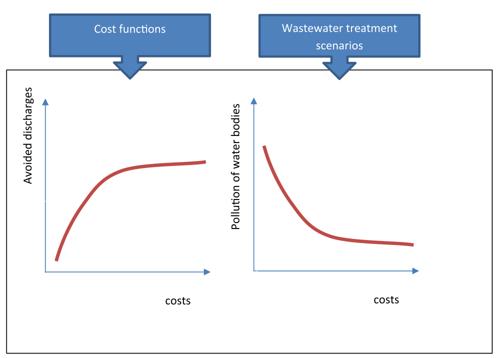
HIGHLIGHTS

- Cost/effectiveness of advanced treatment scenarios for wastewater micropollutants.
- Pollution down by 75 % with advanced treatment everywhere, but costs ~4 bn €/y
- Acceptably protective, "compromise" scenario costs about 1.5 bn €/y.
- Large plants should treat, risk assessment to decide on treatment at a small plant.

GRAPHICAL ABSTRACT



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ABSTRACT

In this contribution, we analyse scenarios of advanced wastewater treatment for the removal of micropollutants. By this we refer to current mainstream, broad spectrum processes including ozonation and sorption onto activated carbon. We argue that advanced treatment requires properly implemented tertiary (nutrient removal) treatment in order to be effective. We review the critical aspects of the main advanced treatment options, their advantages and disadvantages. We propose a quantification of the costs of implementing advanced treatment, as well as upgrading plants from secondary to tertiary treatment when needed, and we illustrate what drives the costs of advanced treatment for a set of standard configurations. We propose a cost function to represent the total costs (investment, operation and maintenance) of advanced treatment. We quantify the implications of advanced treatment in terms of greenhouse gas emissions. Based on the indicators of total toxic discharge, toxicity at the discharge points and toxicity across the stream network discussed in

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Pistocchi et al. (2022), we compare costs and effectiveness of different scenarios of advanced treatment. In principle the total toxic load and toxicity at the points of discharge could be reduced by about 75 % if advanced treatment processes were implemented virtually at all wastewater treatment plants, but this would entail costs of about 4 billion euro/year for the European Union as a whole. We consider a "compromise" scenario where advanced treatment is required at plants of 100 thousand population equivalents (PE) or larger, or at plants between 10 and 100 thousand PE if the dilution ratio at the discharge point is 10 or less. Under this scenario, the length of the stream network exposed to high toxicity would not increase significantly compared to the previous scenario, and the other indicators would not deteriorate significantly, while the costs would remain at about 1.5 billion Euro/year. Arguably, costs could be further reduced, without a worsening of water quality, if we replace a local risk assessment to generic criteria of plant capacity and dilution in order to determine if a WWTP requires advanced treatment.

1. Introduction

In the past decades, urban wastewater treatment plants (WWTPs) have been mostly designed for the removal of organic carbon, nitrogen and phosphorus with effluent standards depending on the permitting rules in place. In the European Union (EU), these conform to the Urban Wastewater Treatment Directive (UWWTD) 91/271/EEC. Although WWTPs are able to remove many pollutants through sorption and biodegradation, they remain an important point of entry into the aquatic environment for the more persistent and mobile contaminants. A demand has grown over time to address in particular organic trace compounds of emerging concern such as pharmaceuticals, biocides and pesticides, and other household and industrial chemicals, collectively referred to as micropollutants (MPs), besides constituents such as microplastics, engineered nanoparticles and pathogens.

An appreciable removal of MPs at a WWTP usually requires more advanced treatment processes than conventional pollution, and may imply higher resource intensity and associated greenhouse gas (GHG) and other emissions. Most obviously, an advanced treatment implies higher investment and operational costs, and may require more skilled operators compared to a conventional treatment.

Good conventional treatment (including at least effective nitrification) is *de facto* a prerequisite for advanced treatment (see Supplementary information (SI), Note 1 for a more extensive discussion). Establishing an appropriate level of wastewater treatment entails balancing the reduction of MP concentrations in effluents, on the one side, and resource intensity, emissions and costs, on the other. As the marginal removal of MPs decreases with the intensity of treatment, arguably it would not be convenient to push the treatment beyond a certain level. Finding a trade-off between costs, impacts and benefits is key to sustainable wastewater treatment.

In this contribution, we consider the mainstream options currently available for the removal of MPs from wastewater beyond the performance of conventional WWTPs. We focus on two broad families of processes, namely activated carbon (AC, in powdered (PAC) or granular (GAC) form) and chemical oxidation with ozone (O_3). Other advanced treatment processes, such as membranes (see SI, Note 2), are substantially less used for this purpose, also because of their higher costs. Constructed wetlands may be useful to reduce certain MPs in effluents, but may struggle with the most recalcitrant ones (see SI, Note 2).

A plethora of specific processes has been developed to address particular classes of contaminants. Moreover, disinfection processes already applied in WWTPs discharging to water bodies used for bathing or aquaculture, not falling into one of the above categories, may also have in principle an effect on MPs (see SI, Note 3). All these processes have usually shown a capability to remove contaminants to a similar extent, but at higher costs or with more severe constraints (e.g. regeneration of alternative adsorbents, security problems with the storage of hazardous reagents, by-products, energy consumption, availabilities of resources or catalysts etc.) in comparison with AC and O_3 (Abegglen and Siegrist, 2012; Miklos et al., 2018). Therefore, solutions at the full scale are mainly limited to the above-mentioned processes alone or in combination.

Applying AC or O_3 to the effluents of conventional biological treatment plants enables in principle a substantial removal of the majority of MPs from wastewater. However, we need to compare this positive effect with the costs of investment and operation, increased consumption of energy and other resources, and associated greenhouse gas (GHG) emissions, both directly during the process, and embedded in the process input (e.g. Jekel et al., 2015; Meier and Remy, 2020).

Moreover, advanced treatment of MPs may have unintended negative effects on the effluents due to the formation of by-products (e.g. bromates during O_3 if bromide is appreciably present in the influent (Soltermann et al., 2016)), or undermine the reuse of sludge in agriculture (e.g. when PAC is applied in the activated sludge bioreactor and cannot be separated from the excess sludge) (VSA, 2021).

This paper addresses the technical and economic conditions that make advanced urban wastewater treatment for MP removal a cost-effective solution.

Assuming the preconditions for biological treatment are met before implementing advanced treatment (see SI, Note 1), we examine the different options available, the costs of their implementation depending on the desired removal of MPs, their energy and GHG emissions and subsequent implications, and other factors that may hamper or facilitate the incorporation of advanced treatment processes in existing WWTPs. In this, we take into account the initial conditions of a given WWTP and the possible synergies with disinfection processes for the removal of MPs. On this basis, we explore the trade-offs between costs and benefits of implementing advanced wastewater treatment in the European context, with the aim of identifying an acceptable, cost-effective strategy for the end-of-pipe control of MPs in wastewater. Throughout this contribution, we regard end-of-pipe treatment as complementary to the control of micropollutants at the source. Consequently, our analysis does not endorse end-of-pipe treatment as the only, or even the most important component of an overall approach to MP pollution control.

2. Options for the advanced treatment of wastewater micropollutants

2.1. Removal processes based on oxidation

Ozonation entails a direct pathway consisting of the reaction of the MP with ozone (O_3) and a pathway by radical reactions, mostly driven by hydroxyl radical ($^{\bullet}OH$) (Von Sonntag and von Gunten, 2012 and von Gunten, 2018). The dosage of O_3 needed to abate MPs depends largely on the dissolved organic carbon (DOC) of the influent (Antoniou et al., 2013; Lee and von Gunten, 2016; Bourgin et al., 2018), as well as on NO_2^- and to a lesser extent the suspended solids. Besides NO_2^-, other chemical species found in wastewater can react with ozone and/or $^{\bullet}OH$, including bromide (Br^-) and iodide (I^-) (Gottschalk et al., 2009). Other constituents consume $^{\bullet}OH$ (e.g. carbonate (CO_3^{2-}) and dissolved organic matter (DOM) (Von Sonntag and von Gunten, 2012 and Gottschalk et al., 2009)) resulting in a limitation of the $^{\bullet}OH$-induced pathway of oxidation. The reactivity of a compound with O_3 depends on the presence in it of specific functional groups rather than the entire structure: activated aromatic moieties, olefins, neutral amines and reduced sulfur moieties make compounds particularly reactive to ozone (Von Sonntag and von Gunten, 2012; Lee and von Gunten, 2016; von Gunten, 2018). The extent of transformation of MPs in ozonation strongly depends on their chemical structure. Many MPs are abated by >80 % for an average ozone dose of 0.5–0.9 $gO_3.gDOC^{-1}$ (e.g. Penru et al., 2018; Hollender et al., 2009; Lee et al., 2013; Bourgin et al., 2018; El-taliawy et al., 2017).

A typical configuration of an ozonation process entails equipment for on-site generation of O_3, a reactor where O_3 comes into contact with the influent, and an ozone quenching (destruction) unit to treat the off-gas. The feedstock for O_3 generation may be pure oxygen or air, and the process most commonly used in wastewater treatment plants is electrical discharge (Metcalf & Eddy, 2014). When using air, the generation of O_3 requires 1.5 to 2 times as much energy as from pure oxygen. The latter, though, needs to be prepared at another industrial facility and transported in liquefied form, so the impact of both feedstocks is arguably comparable and the choice between the two depends on practical factors, such as the availability of space, plant size, safety requirements on the site, etc. (Abegglen and Siegrist, 2012).

The contact time with O_3 and the configuration of the reactor play an important role. It is generally recommended to work at low ozone dosage with a contact time of 10–14 min for maximum flow.

Not every wastewater is suitable for treatment with ozone. In some cases, harmful non-degradable reaction products can form by oxidation (for instance, high concentrations of bromide in the influent yield potentially carcinogenic bromate (Soltermann et al., 2017)). The safety of ozonation has to be tested in an early stage of planning, and specific procedures exist on purpose (VSA, 2021).

Moreover, oxidation may cause only a partial oxidation of compounds, generating some transformation products that can be potentially harmful. Many studies indicate that oestrogenicity is efficiently abated during ozonation and with other oxidants (Huber et al., 2004; Lee et al., 2008; Escher et al., 2009; Schindler Wildhaber et al., 2015). In some cases, though, biological essays have shown that a reduced concentration of certain target MPs in ozonation effluents from real wastewater did not correspond to a reduction of oestrogenicity, which could be attributed to transformation products (Bertanza et al., 2011, 2013). It has been also shown that effects such as antibiotic activity and algal toxicity can be removed (von Gunten, 2018). In order to limit risks from transformation products, it is good practice to polish ozonation effluents via sand filtration or GAC and/or biological treatments (see e.g. Tang et al., 2020; Bourgin et al., 2018).

The main effect of the biological post-treatment is the degradation of the oxidation by-products formed from the dissolved organic matter matrix, as transformation products are usually not biodegradable (Gulde et al., 2021).

2.2. Removal processes based on adsorption

Adsorption of hydrophilic MPs occurs when the treated effluent is put in contact with solids of very high specific surface, and is primarily due to electrostatic interactions (Metcalf & Eddy, 2014). AC is widely adopted for the purification of water and other fluids in many industrial applications. While alternative sorbents have been developed, such as zeolites or expanded clay (e.g. Tahar et al., 2014), AC remains to date the most cost-effective option for the removal of wastewater MPs. AC is produced from an appropriate thermal processing of a carbonaceous feedstock, yielding a carbon structure with very high micro-porosity, hence very large specific surface. The latter enables the capture of dissolved MPs from the influent, until the available adsorption sites are saturated. The feedstock may be virtually any material containing carbon, including hard coal, lignite, charcoal of wood or other ligno-cellulosic materials, or biochar (Gu et al., 2018). The typical configurations of AC include powder (PAC), fluidized granular grains (micro-grains), or a granular fixed bed (GAC) Metcalf & Eddy, 2014).

PAC can be dosed directly into or after the biological reactor. Direct dosing of PAC has been studied mainly in suspended sludge processes (Serrano et al., 2011; Boehler et al., 2012; Meinel et al., 2016), with our without membranes for biomass separation, but the possibility has also been demonstrated for biofilm processes (Cimbritz et al., 2019). Dosing after the biological treatment entails separate contact reactors and various separation methods (Boehler et al., 2012, Altmann et al., 2015; Margot et al., 2011; Löwenberg et al., 2013; Kårelid et al., 2017). It is possible to dose the PAC directly before a sand filter to keep the system compact (Löwenberg et al., 2016; Boehler et al., 2011).

When the process configuration allows, PAC can be recirculated in order to maximize its utilization (counter current principle).

After treatment, PAC must be separated from the effluent either through settling and/or through filtration (e.g. membrane ultra-filtration, UF, sand filtration or cloth filters).

When the ambition is to remove 80 % of certain indicator substances, a HRT of 30 min should be ensured according to DWA-M 285-2, 2021. As for oxidation, the quality of the influent for an AC process is critical. DOC, that has affinity with AC, must be therefore as low as possible in order to reduce the dose of sorbent, hence costs. A typical dosage is 1.5 g PAC/gDOC to treat a secondary effluent, which must be increased (2–2.5gPAC/gDOC) if the PAC is added directly in the biological treatment.

PAC added into the biological system makes the quality of the sludge unfit for reuse through spreading on farmland due to the presence of the adsorbed MPs. It also increases the volume of sludge produced by 20–30 % depending on the dosage. On the contrary, PAC is beneficial for sludge incineration, as it increases its energy content. When PAC is obtained from biogenic feedstock, its combustion is also climate-neutral, but this is not the case when PAC derives from fossil feedstocks.

The appraisal of PAC as a solution requires a broader consideration of the sludge handling strategy applied. At present, the agricultural use of sludge is heavily debated, and some countries are virtually phasing it out (e.g. Germany's regulation, BGB I Nr. 65 S. 3465 ff., 2017) also considering that part of the agricultural value of sludge (particularly phosphorus) may be recovered through specific processes compatible with incineration. However, in many other contexts sludge application in agriculture remains of interest as a cost-effective option.

GAC entails a separate reactor where the influent comes into contact with the granules forming a fixed bed. This avoids the need for settling, contrary to the case of PAC. However, not only requires GAC a low influent DOC content, but also lower concentrations of suspended solids (usually <20 mg/L) to enable a smooth operation by limiting clogging and reduced backwash frequency, hence costs. For an effective removal of MPs, the hydraulic retention time (or "empty bed contact time", EBCT) of the reactor should be of 20–30 min (Böhler et al., 2020; Fundneider et al., 2020) for the maximum flow that has to be treated. Shorter contact times were also found effective in some cases (Besnault et al., 2015). When the GAC filter is saturated, it requires regeneration of the sorbent. The time before a GAC bed requires regeneration, usually expressed as the volume of influent treated divided by empty bed volume, i.e. number of bed volumes (BV), is a critical parameter driving the costs and impacts associated to a GAC treatment. Swiss recommendations (Böhler et al., 2020) indicate a GAC duration of 20,000–30′000 BV when treating urban wastewater, although the variation observed in practice is very large (e.g. Benstoem et al., 2017). This is dependent on the concentration of DOC and the general composition of the wastewater.

Regeneration requires transport of used GAC to a factory, and its exposure to high temperature (1200 °C). During this step, a loss of 10 to 20 % of GAC occurs requiring the addition of new GAC, the so called "make up". The possibility of regeneration is an advantage of GAC over PAC, potentially reducing GHG emissions (Meier and Remy, 2020).

GAC can be used also in a fluidized bed system. This type of solution uses a smaller grain size of GAC compared to a fixed bed, and leaves the grains fluidized in a reactor. Fresh GAC is added regularly (e.g. once a day) and used GAC is removed about once a week and stored for regeneration. So the dosage is more flexible compared with a conventional GAC filter and the smaller grain size allows higher reaction rates. The GAC stays in the system for about 100 days. This enables installations with lower reactor volume compared to conventional GAC filtration, and lowers risks of clogging.

For both PAC and GAC, the production or regeneration of the sorbent accounts for a significant part of the treatment costs and impacts because of the energy and chemicals use it entails.

3. Life-cycle impacts of advanced treatment

The removal of MPs from wastewater using PAC, GAC or O_3 implies a more complex treatment plant with a potentially larger use of materials, process inputs, and energy. All this entails supply chains with the associated impacts, and calls for a life-cycle analysis (LCA) perspective (Risch et al., 2021). LCA must consider greenhouse gas (GHG) and other emissions, such as those causing acidification, ozone depletion, toxicity and eutrophication, and resource use of all material and energy flows associated to the inputs, processes and outputs of wastewater treatment. In this way, it can unveil the environmental implications of alternative treatment options in a holistic perspective. Application of LCA to wastewater treatment can be very challenging. Reliance on existing databases for the so-called characterization factors, representing parametric impacts of wastewater inputs (such as the energy and chemicals used for a treatment process) on various environmental dimensions, may not provide a correct account of the specific conditions of each plant, e.g. when energy or chemicals are derived from circular processes and may have a peculiar impact profile. Process innovation and optimization may also play a substantial role in mitigating the impacts of advanced treatment. For instance, using energy from renewable resources may significantly reduce the GHG emissions of O_3, and use of non-fossil feedstock for the sorbent may reduce the GHG emissions of GAC and PAC (Joseph et al., 2020).

Several studies have addressed the LCA of solutions for the removal of MPs from wastewater. Rahman et al. (2018) and Risch et al. (2021), consider GAC, O_3 and other advanced treatment options for a list of usual wastewater MPs in the American and French contexts, and conclude that the reduced ecological and human toxicity due to lower discharges of MPs may be comparable with, or even outweighed by the toxicity caused by additional treatment. However, GAC and O_3 have lower life-cycle impacts than other solutions (membranes or other AOPs). While MP removal by GAC and O_3 cause slight increases in other impact categories (eutrophication, global warming potential, acidification and ozone depletion), Rahman et al. (2018) show that the effects attributable to GAC or O_3 alone are small compared to the overall impacts of wastewater treatment (including nutrient removal).

Tarpani and Azapagic (2018) show better performances for GAC than for O_3 under the point of view of greenhouse gas emissions, mainly associated to energy demand, but the differences among the two are relatively small.

Pesqueira et al. (2020), in their systematic review note that LCA studies on MP removal processes are relatively consistent and show that energy and chemicals used for MP removal drive the bulk of the impacts. The production of both energy and chemicals may cause impacts in terms of all types of emissions. However, Pesqueira et al. (2020) point out that such studies may generally suffer from an underestimation of the benefits associated to the removal of MPs, because they consider a reduction of toxicity with reference to a limited set of contaminants. As broad-spectrum advanced treatment processes typically remove a range of chemicals, the reduction of effluent toxicity is expected to exceed by far the additional toxicity associated to reagents and energy for GAC and O_3, contrary to the findings of Rahman et al. (2018). A more realistic picture of the extent to which advanced treatment reduces effluent toxicity may stem from the deployment of bioassays, providing a more appropriate characterization than a few indicator chemicals (Pedrazzani et al., 2018; Escher and Leusch, 2011; Gonzalez-Gil et al., 2016; Papa et al., 2016; Pedrazzani et al., 2020; see also Phan et al., 2021). Use of results of bioassays in LCA requires their conversion into equivalent pollutant mass flows (as suggested by Pedrazzani et al., 2018 and Pedrazzani et al., 2019; see also Papa et al., 2013, Papa et al., 2016).

The GHG emissions associated to advanced treatment, due to the use of energy and reagents, remain an important issue to consider. Recent LCA studies (Tarpani and Azapagic, 2018; Li et al., 2019) indicate a relatively narrow range of emissions between 0.15 and 0.3 kg CO2e/m^3 of wastewater treated for AC and O3. Georges et al. (2009) also suggest emissions from sand filters around 0.1 kg/m^3.

4. Modelling the costs of advanced wastewater micropollutant removal

The implementation of an advanced treatment level for wastewater MPs entails (1) costs for the retrofitting of the existing biological (secondary and tertiary) treatment in order to ensure sufficient effluent quality, and (2) costs for the implementation of new treatment processes. The costs of retrofitting of existing plants depend on their initial operating conditions and margins of improvement, on the available space and reactor volumes, and several other site-specific factors. In this section, we propose a schematic quantification of the implementation of advanced treatment only, with the sole purpose of a screening-level calculation to compare strategic investment alternatives at the continental scale. The SI (Note 4) illustrates an additional calculation including an estimate of the costs to retrofit the existing plants, when these do not yet perform nutrient removal.

The costs of new treatment processes depend largely on the type of process and design configuration adopted, and in any case the specific conditions of a plant may cause a considerable variation. Therefore it is difficult to correctly compare the costs of projects at different WWTPs.

The costs of advanced treatment include the capital expenditure (CAPEX) for additional infrastructure, as well as the maintenance and energy costs, and the costs of consumables employed in the processes (i.e. the operational expenditure, OPEX). For ozonation, the infrastructure includes the contactor as well as the ozone generation and quenching equipment. Moreover, ozonation needs post-treatment, e.g. with a GAC, sand filter or polishing biological treatment in order to remove by-products. If a filtration stage is already in place, ozonation can be quite convenient, whereas if a new filtration is needed, the costs for advanced treatment are considerably higher. Also PAC added directly in the biological treatment may require a filtration to minimize AC loss. PAC may entail a lower CAPEX if it can be dosed directly in the bioreactor, whereas GAC requires a dedicated contactor. The OPEX of ozonation is driven by the energy demand of O_3 generation, while that of PAC and GAC is driven by the sorbent and/or its regeneration. The costs of advanced treatment may vary considerably depending on the initial conditions of a plant. Indicative ranges of costs and cost functions proposed in the literature in various European contexts are summarized in Table 1 and Table 2, typically highlighting highest costs for GAC, followed by PAC and ozonation, also rather consistently with Rizzo et al. (2019). These authors report a CAPEX in the range of 0.035–0.05 Euro/m^3 of treated wastewater (30 years depreciation for civil works and 15 years for mechanical equipment, 10 for electrical

Table 1
cost functions from Switzerland (CH) and Germany (DE). O_3 = ozonation; SF = sand filter; PAC, GAC = powdered, granular activated carbon; PE = population equivalents.

Type of treatment	Source	Context	Expenditure function	Costs included and units
PAC + SF	BG Ingenieure und Berater AG, 2012	CH	$48,234.9679*PE^{-0.4786}$	Investment costs CHF/PE
PAC + SF	BG Ingenieure und Berater AG, 2012	CH	$179.4029*PE^{-0.2225}$	Operation cost CHF/PE/Y
O3 + SF	BG Ingenieure und Berater AG, 2012	CH	$26,889.5261*PE^{-0.5078} + 21,424.4602*PE^{-0.5733}$	Investment costs CHF/PE
O3 + SF	BG Ingenieure und Berater AG, 2012	CH	$367.5990*PE^{-0.3528}$	operation cost CHF/PE/Y
O3	Türk et al., 2013	DE	$2261.9*V^{0.4417}$	investment cost before VAT (approximate function); V = water volume treated, m^3/a
O3	Türk et al., 2013	DE	0.0147*V + 46,081	operation cost; V = water volume treated, m^3/a
O3	Türk et al., 2013	DE	0.0073*V + 9322	energy cost; V = water volume treated, m^3/a
O3	Antakyali, 2017, cit. in Rizzo et al., 2019	DE	$5.68\ PE^{-0.38}$	Investment cost Euro/m^3
PAC	Antakyali, 2017, cit. in Rizzo et al., 2019	DE	$6.23\ PE^{-0.36}$	Investment cost Euro/m^3
GAC	Antakyali, 2017, cit. in Rizzo et al., 2019	DE	$143.23\ PE^{-0.63}$	Investment cost Euro/m^3
Ensemble curve	Herbst et al., 2016	DE	$10.861\ PE^{-0.424}$	Total cost Euro/m^3

Table 2
Ranges of costs of advanced treatment for Switzerland (CH), the Netherlands (NL) and Sweden (SE). O_3 = ozonation; SF = sand filter; PAC, GAC = powdered, granular activated carbon; BAF = biologically active filter; CAS = conventional activated sludge; UF = ultrafiltration.

Type of treatment	Source	Context	Units	For 10,000 PE	For 20,000 PE	For 50,000 PE	For 100,000 PE	For 500,000 PE
O_3 + SF	Baggenstos, 2019	CH	Euro per PE per year	25.38	22.50	13.72	12.60	
PAC in CAS + SF	Baggenstos, 2019	CH	Euro per PE per year	27.26	24.72	17.11	16.07	
PAC + SF	Baggenstos, 2019	CH	Euro per PE per year	30.08	26.88	17.30	15.79	
GAC	Baggenstos, 2019	CH	Euro per PE per year	31.96	29.09	20.49	17.40	
O3[b]	Baresel et al., 2017	SE	Euro per m^3	0.04	0.03		0.02	0.01
BAF(GAC)[b]	Baresel et al., 2017	SE	Euro per m^3	0.09	0.07		0.05	0.04
O3 + BAF(GAC)[b]	Baresel et al., 2017	SE	Euro per m^3	0.11	0.08		0.05	0.04
PAC-UF[b]	Baresel et al., 2017	SE	Euro per m^3	0.21	0.16		0.13	0.12
UF-BAF(GAC)[b]	Baresel et al., 2017	SE	Euro per m^3	0.21	0.16		0.11	0.09
GAC	STOWA, 2017	NL	Euro per PE per year		14.80		13.70	12.60[a]
PAC + SF	STOWA, 2017	NL	Euro per PE per year		13.80		10.50	8.40[a]
O_3 + GAC	STOWA, 2017	NL	Euro per PE per year		15.90		10.00	9.50[a]
O_3	STOWA, 2017	NL	Euro per PE per year		4.80		3.20	2.60[a]
O_3 + SF	STOWA, 2017	NL	Euro per PE per year		10.60		8.90	7.90[a]

[a] Capacity 300,000 PE.
[b] Referred to specific flow of 150 m^3 per PE per year.

equipment), and an OPEX of 0.04 Euro/m3 of treated wastewater (including electric energy, maintenance, additional analyses and workload, oxygen input), practically independent of the process adopted. The operational cost of O_3 and AC depends on the dose, in turn very sensitive to NO_2 and DOC concentration in the water to treat. Other contexts may entail higher costs. For instance, a technical and economic assessment of ozonation in France (Choubert et al., 2017) calculated a cost of 0.1–0.2 Euro/m^3 for ozonation and 0.2–0.3 Euro/m^3 for activated carbon, depending on the size of the plant and its initial and operating conditions and the supply chain of reagents.

The variability of costs among different treatment options seems to be comparable with the variability within a certain treatment option depending on the specific plant conditions. Consistently, some authors have suggested that the specific costs of advanced treatment (including CAPEX and OPEX) could be represented as a first approximation by a single expenditure function (e.g. Herbst et al., 2016).

In order to define an appropriate expenditure function for advanced wastewater MP removal in Europe, we compiled a dataset of actual or accurately estimated costs for advanced treatments in European WWTPs (the dataset is provided and documented as Supplementary information). By plotting the levelized costs of advanced treatment as a function of plant capacity, we show (Fig. 1) that indeed the variability among technical solutions is comparable with the variability within plants with a given solution. While the heterogeneity of the data hampers a conventional statistical analysis, we observe that costs can be captured within a factor 2 by the simple function:

$$C_{adv} = 1000 \ PE^{-0.45} \tag{1}$$

where C represents the levelized cost (Euro/PE/year) of advanced treatment and PE is the count of population equivalents to be treated (plant capacity). This expenditure function, plotted in Fig. 1, should not be regarded as a statistical model, but as a practical working assumption for a preliminary assessment of aggregated costs, at the European scale, taking into account the capacity of the plant.

In order to better understand the drivers of the total cost of advanced treatment, let us consider a set of nine typical treatment process

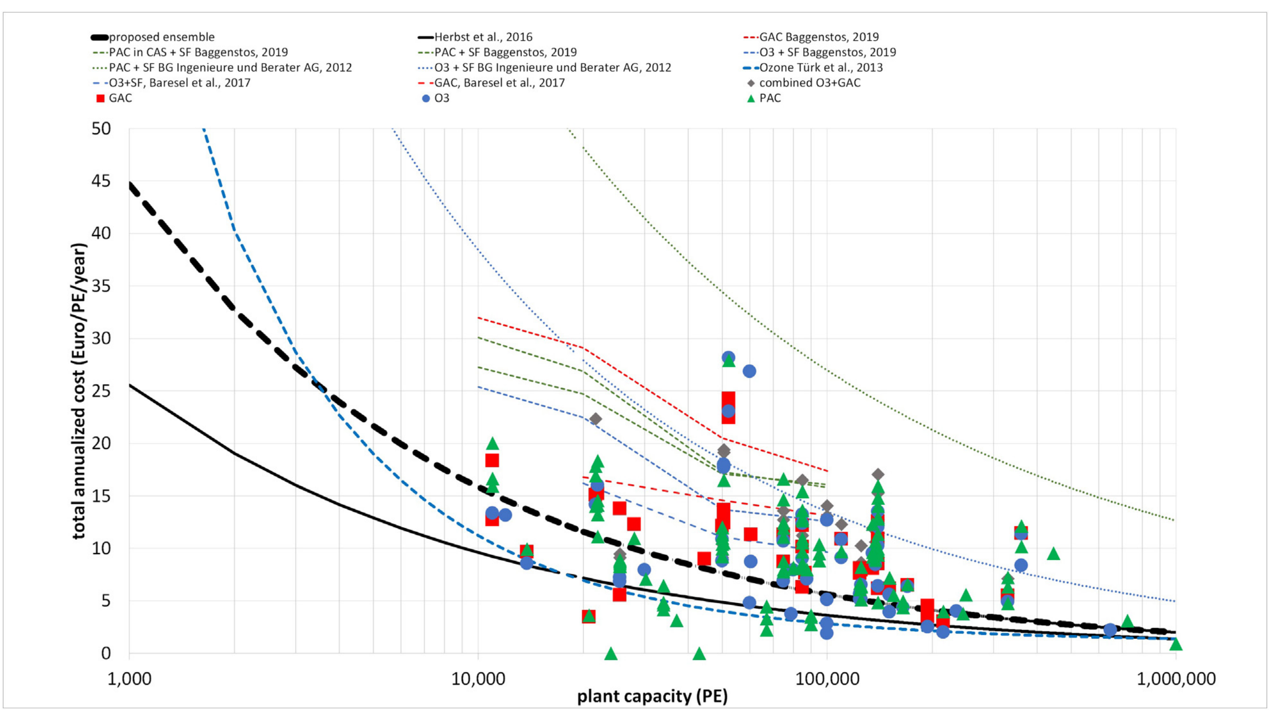

Fig. 1. – Comparison of cost functions and case-specific data. The proposed ensemble expenditure function is superimposed to the data.

Table 3
Design and operating parameters for 9 typical configurations of advanced treatment for wastewater MPs.

Code	PAC1	PAC2	GAC	O31	O32	O33	O3GAC	PAC + UF
	Advanced treatment processes, PAC, after sec. Treatment, 1.5 mg PAC/mg DOC	Advanced treatment processes, PAC, into CAS, 2–3 mg PAC/mg DOC	Advanced treatment processes, GAC, EBCT >20 min; v < 9 m/h > 20,000 BVT	Advanced treatment processes, Ozone, <0.4 mg O_3/mg DOC	Advanced treatment processes, Ozone, 0.4–0.6 mg O_3/mg DOC	Advanced treatment processes, Ozone, >0.7–1.0 mg O_3/mg DOC	Advanced treatment processes, Ozone, 0.2–0.3 mg O_3/mg DOC combined with GAC, EBCT > 20 min; v < 9 m/h; BVT < 20,000	Membrane filtration, UF-PAC, 15 mg/L
PAC (kg/PE/y)	1.095	2.19						1.095
O_3 (kg/PE/y)				0.292	0.438	0.584	0.1825	
GAC replacement (kg/PE/y)			1.314				1.314	
PAC contactor volume (m^3/PE)	0.008333							
Ozone contactor volume (m^3/PE)				0.0014	0.0014	0.0014	0.0014	
GAC contactor volume (m^3/PE)			0.002778					
Energy use for O_3 production from pure O_2 (kWh/kgO_3)				12.5	12.5	12.5	12.5	
Energy use for external pure O_2 production kWh/kgO_3				8.2	8.2	8.2	8.2	
Pumping and other energy use kWh/m^3	0.05	0.05	0.03	0.06	0.06	0.06	0.07	0.2
Cost of UF membrane modules (Euro/(m^3/day))								150
Cost of GAC Euro/kg			2.5				2.5	
Cost of PAC Euro/kg	2.5	2.5						2.5
Cost of pure oxygen Euro/kg				0.2	0.2	0.2	0.2	
Investment for contactor vessels Euro/m^3	500		500	500	500	500	500	
Investment for sand filters Euro/PE	8	8		8	8	8		
Investment for O_3 production equipment (Euro/PE)				6.5	6.5	6.5		
Cost of GAC regeneration Euro/kg			0.5				0.5	

configurations with AC, O_3 or a combination thereof, as shown in Table 3. We refer to a wastewater treatment plant of medium capacity (50,000 PE) with a dry weather flow of 0.2 m^3 day^{-1} PE^{-1}. We assume a concentration of DOC in secondary effluents equal to 10 mg/L. We design the PAC and ozonation treatment configurations with a contact time of 60 and 10 min respectively, and the GAC configuration with an EBCT of 20 min. We further assume a GAC mass equal to 0.625 kg/PE and GAC losses in regeneration/reactivation equal to 20 %. Maintenance and operation costs other than energy are assumed to be equal to 3 % of the investment every year, and we assume a write-off time of 20 years with a discount rate of 3 % for investments. We further assume a cost of other unspecified chemical additives of 0.01 Euro/m3 for all treatment processes. All other design and costing parameters are calculated or provided in Table 3. Based on the assumptions made, the total costs of the various configurations would be broken down as shown in Fig. 2.

It appears that the various configurations reflect trade-offs between energy and process inputs. Due to the variability of cost items depending on the context, the ranking of alternatives may completely change compared to this example. Arguably, all configurations are rather similar to each other, although ozonation features consistently lower costs.

5. Energy requirements

The removal of micropollutants with O_3, AC or membrane filtration entails an increased use of energy, either directly in the process, or indirectly as embedded in the process input. Production of ozone, in particular, is more energy intensive compared to AC, requiring about 0.3 kWh per m^3 of treated wastewater (Rizzo et al., 2019) as the sum of energy use for ozone generation from oxygen (about 0.05 kWh per m^3), and the preparation of oxygen (about 0.25 kWh per m^3). Those figures are confirmed by the findings of Bertanza et al. (2018), who, for an ozone dose of 8 g/m^3, report an energy consumption for a tertiary ozonation stage (all equipment included) of 0.05–0.08 kWh/m^3 of treated wastewater (excluding the energy required for liquid oxygen production). Hansen et al., 2010, find an overall energy demand of about 0.1 kWh/m^3 per g of O_3 used in the process. For comparison, membrane nanofiltration requires about 0.5 to 1 kWh/m3 (Rizzo et al., 2019). A sand filter downstream of ozonation usually requires 0.01–0.05 kWh/m3 according to Büeler et al. (2018).

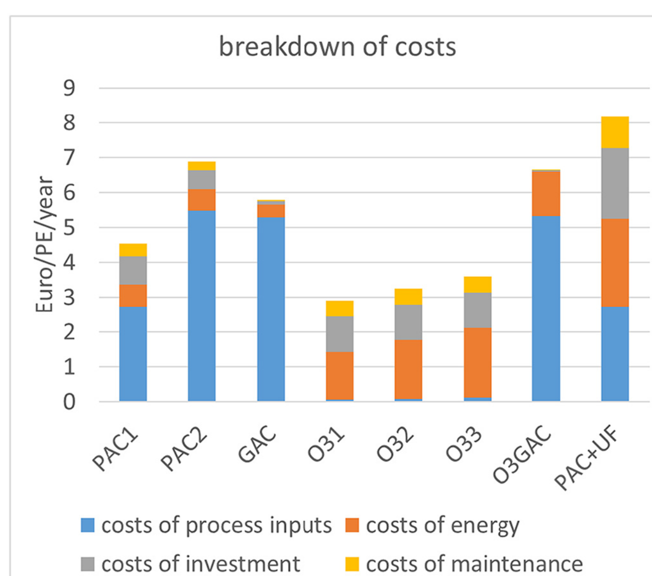

Fig. 2. - Breakdown of costs of the nine representative configurations of advanced treatment, based on the specific assumptions of Table 3. Labels of the categories axis correspond to the codes of the treatment configurations shown in Table 3.

Compared to a typical energy use for conventional wastewater treatment <0.5 kWh/m^3, the additional energy consumption represents an increase in excess of 10 %. Experience in Switzerland shows an increased power demand of 10–30 % in WWTPs upgraded with ozonation (Rizzo et al., 2019). Data from the Sophia-Antipolis plant in France (Choubert et al., 2017) indicate that ozonation has caused a 25 % increase of the total energy consumption due to under-load and thermal destruction of O_3 in excess. The destruction of ozone in excess contributes to the overall energy demand of ozonation, and can be made less energy-intensive using a catalytic rather than thermal excess O_3 quenching.

While the AC process in itself requires only limited energy (see Risch et al., 2021), the production of activated carbon is also energy intensive, and so is the process of GAC regeneration (Metcalf & Eddy, 2014). Among the case studies that we have compiled for the present analysis, energy use ranges from <1 to >12 kWh per PE per year (Fig. 3). Although ozonation alone or in combination with GAC shows higher energy intensity compared to GAC and PAC, the patterns are rather complex and not clear, anyway not suggesting any economy of scale.

6. Identification of optimal treatment scenarios at the European scale

The considerations presented above enable an appraisal of advanced wastewater treatment scenarios for the removal of MPs at the European scale. In this section, we systematically explore scenarios of advanced wastewater treatment in order to identify cost-effective combinations enabling the highest removal of pollution at the lowest cost or GHG emission. Pistocchi et al. (2022) present an analysis of three indicators of impact by MPs at the European scale, namely:

- The total load of MPs, L, defined as the toxicity-weighted sum of mass discharges of a list of MPs representing a proxy of "total pollution".
- Near field pollution at the discharge point LT, defined as the cumulative toxicity of the same list of MPs, computed at the discharge points of wastewater treatment plants (this indicator is discussed in the supplementary information of Pistocchi et al. (2022)).
- Far field pollution CT, defined as the cumulative toxicity of the same list of MPs, computed for the whole European stream network.

Indicators LT and CT are computed as the toxic units or risk quotients (sum of concentrations normalized by appropriate reference risk thresholds) resulting from the discharge of the WWTP effluents in the European

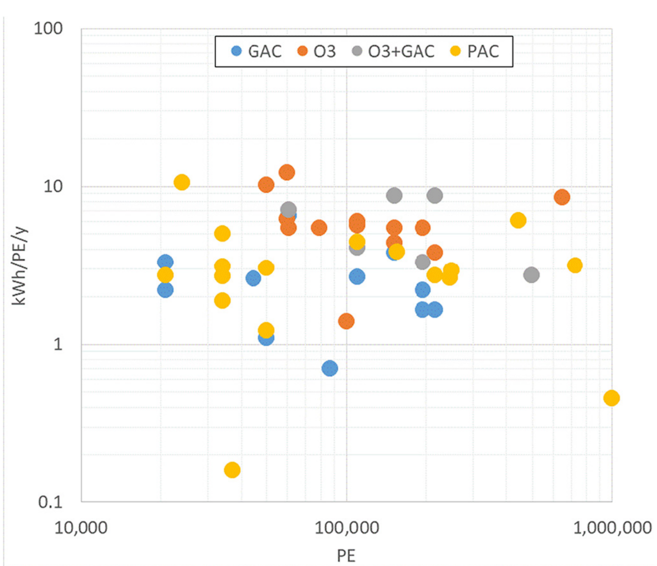

Fig. 3. Energy use for advanced treatment of MPs at the plants whose costs are displayed in Fig. 1, plotted as a function of the treated population equivalents – PE. Data in detail are provided as Supplementary information.

surface waters. They depend on the concentration of contaminants in the effluents and, unlike indicator L, on the dilution occurring in the ambient waters.

Details on the three indicators are described in Pistocchi et al. (2022). The indicators are computed for various scenarios: in each scenario, it is assumed that all plants above a certain capacity or treated volume of wastewater (P, expressed in population equivalents: PE), and/or discharging in water bodies with a dilution ratio D below a certain threshold are required to implement an advanced treatment process. D is the ratio of effluent to ambient concentrations, assuming the WWTP discharge is the only source of pollution, i.e. the receiving water body is initially free of pollution. In Pistocchi et al. (2022) the dilution ratio is evaluated under average flow conditions in the water body.

While the removal efficiency of each MP varies from one process configuration to another, the cumulative toxicity of the effluents is shown to depend only weakly on the type of advanced treatment, but changes with the level of wastewater treatment (conventional or advanced) that is required under a given scenario.

Here we compare L, LT and CT, with an estimation of the corresponding costs and GHG emissions.

We consider the WWTPs existing in the EU as per the 10th UWWTD Implementation Report (EC, 2020). When a plant is required to perform advanced treatment, we estimate the corresponding cost based on Eq. 1. Fig. 4 plots as a function of annual treatment costs the reduction, compared to a baseline scenario of full compliance with the UWWTD, of the total load indicator L achieved with various combinations of D and P. For a given threshold D, L is progressively reduced at lower thresholds P, with apparent marginally decreasing effectiveness. This indicates that it is more cost-effective to require advanced treatment at larger plants than at smaller plants. The higher the dilution threshold D, the more cost-effective it is to implement advanced treatment. When we consider no threshold for D, i.e. an advanced treatment is required for all plants above a threshold P irrespective of the dilution ratio in the receiving water bodies, we obtain the highest reduction of L for a given total annual cost. Conversely, while advanced treatment at all plants with $P \geq 100{,}000$ PE allows a reduction of L of about 38 % and a cost of about 840 million Euro per year, the same reduction can be obtained with advanced treatment at all plants of 50,000 PE if $D = 100$, or 5000 PE if $D = 20$, at higher costs. This means that, when it comes to the total load, it is more cost-effective to implement advanced treatment only at larger plants than based on the dilution of the receiving water bodies.

The indicator of near field pollution, LT, is reduced mainly by lowering the plant threshold P, and the improvements are marginally decreasing with increasing dilution threshold D (Fig. 5). Typically, there is no significant additional benefit beyond $D = 10$.

The indicator of cumulative pollution in the stream network (CT) may be used to map how the percentages of the stream network in a given risk category change as a consequence of implementing advanced treatment. Pistocchi et al. (2022) propose to classify the European stream network in four categories with high, medium-high, medium-low and low risk of exceeding risk thresholds. In Fig. 6, we plot the percentage of the length of the European stream network falling in each of these categories under each of the scenarios of dilution D and plant capacity P considered here, as a function of the cost of the corresponding scenario. Each point in the graphs is therefore a scenario. It is apparent that advanced treatment may reduce by almost a half the percentage of the stream network at high risk (Fig. 6A), and that at medium-high risk (Fig. 6B) at increasing costs. Usually the reduction of the high and medium-high risk causes an increase of the percentage at medium-low risk. When advanced treatment is implemented at most plants, though, there can be a significant increase of the percentage of the network at low risk, while that at medium-low risk decreases (Fig. 6C, D) when we consider scenarios of more widespread advanced treatment (hence higher costs). The decrease of the percentage at high risk and, less apparently, of the percentage at medium-high risk show a decreasing marginal effectiveness of the investments. In particular, significant increases in the percentage at low risk require high investments.

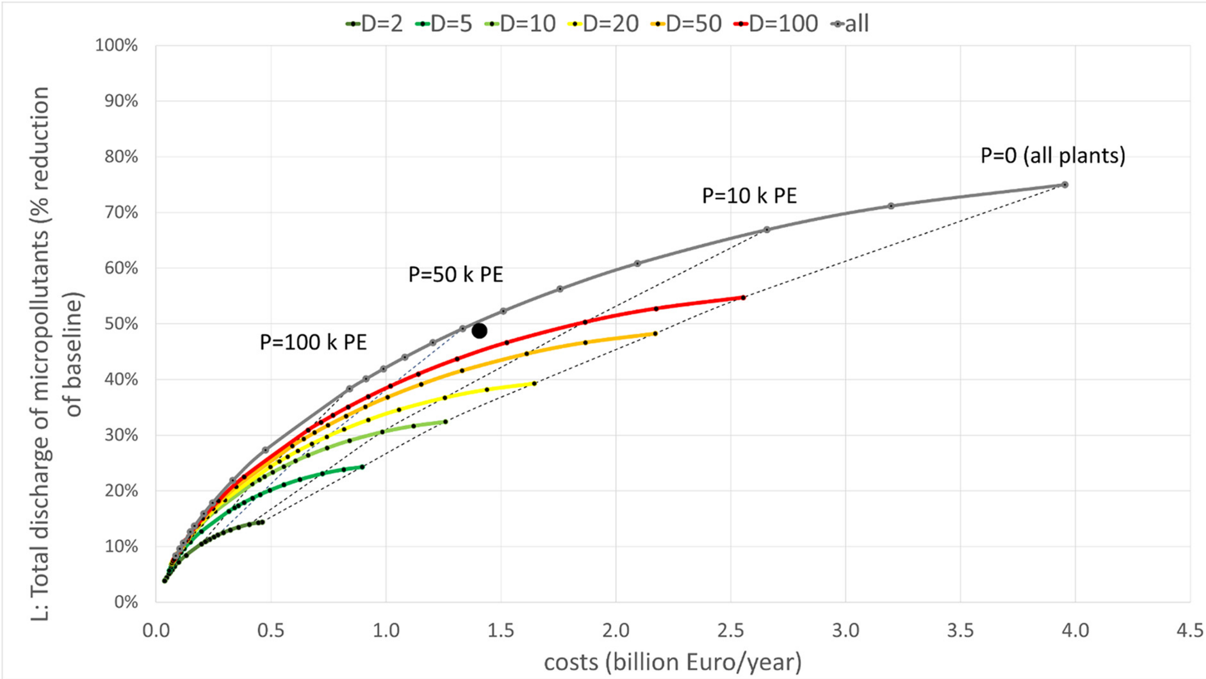

Fig. 4. – Plot of indicator L as a function of the implementation cost (total annual costs as per Eq. 1), for different combinations of D and P. The continuous lines in colour correspond to a given value of dilution at the point of discharge, D, while the data points along these lines represent each a threshold plant capacity P (increments of 100,000 PE above 100,000 PE, and 10,000 PE below). The dashed black lines connect points with the same value of P. "All" denotes the case of D set to infinity (all plants irrespective of D). The black dot represents the "compromise" scenario.

The three indicators describe the response of MP pollution to investments in advanced wastewater treatment. Looking at the total toxic load discharged in the environment (indicator L), it is most cost-effective to require advanced treatment at larger plants irrespective of the dilution in the receiving water bodies. Looking at the near field toxicity (indicator LT) it appears that requiring advanced treatment where dilution exceeds 10 brings little marginal improvements. Looking at the overall conditions of the stream network (indicator CT), most of the reduction in high and medium-high risks would be achieved with annual costs below 1.5 billion Euro/year, indicating that advanced treatment at larger plants and/or at plants with low dilution is cost-effective. However, if the target is to bring the whole stream network to low risk conditions, it is necessary to target

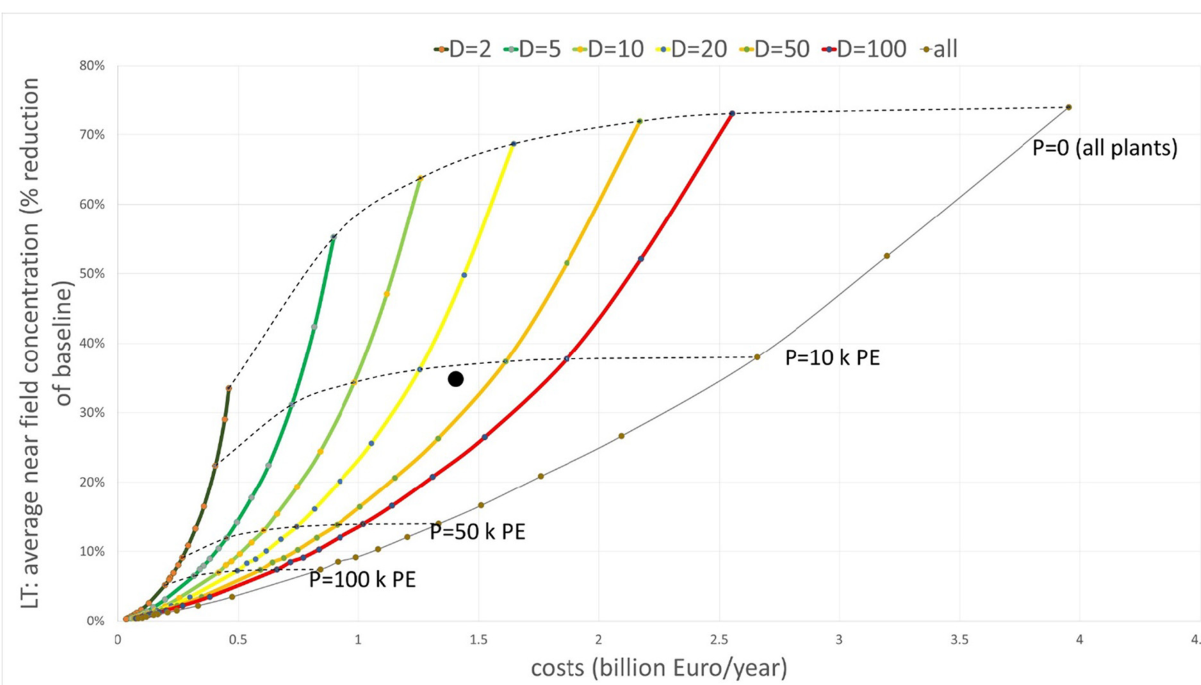

Fig. 5. – average across the EU of indicator LT as a function of the implementation cost (total annual costs as per Eq. 1), for different combinations of D and P. The continuous lines in colour correspond to a given value of dilution at the point of discharge, D, while the data points along these lines represent each a threshold plant capacity, P (increments of 100,000 PE above 100,000 PE, and 10,000 PE below). The dashed black lines connect points with the same value of P. "All" denotes the case of D set to infinity (all plants irrespective of D). The black dot represents the "compromise" scenario.

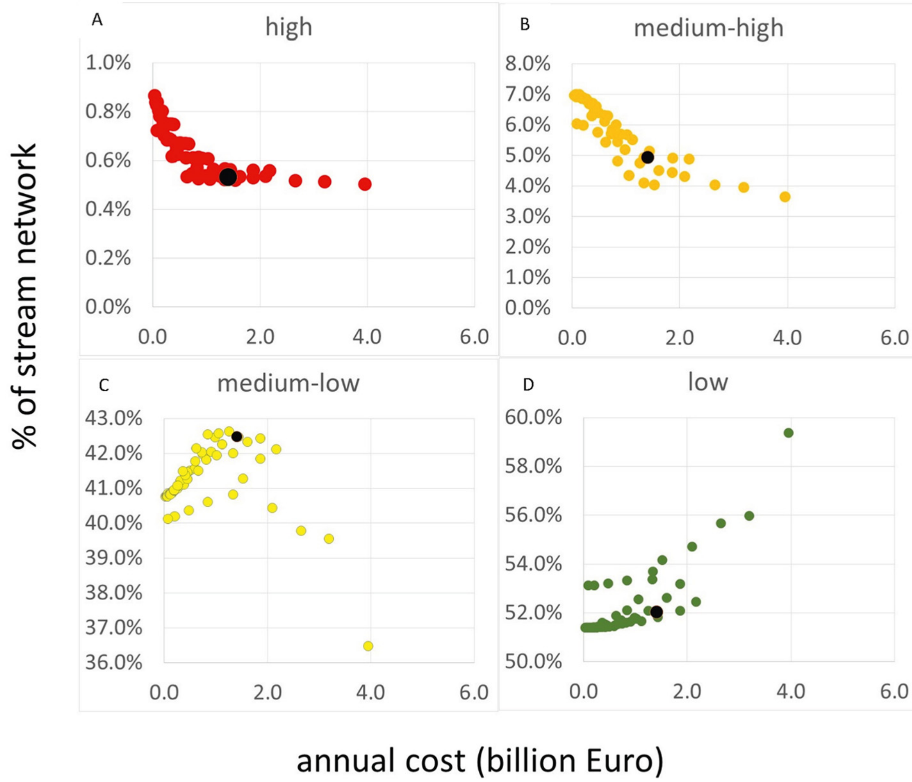

Fig. 6. –percentage of the stream network under four toxicity risk categories (high, medium-high, medium-low and low) under each advanced treatment scenario, as a function of the corresponding costs. Each point in the graph is a scenario (not labelled for the sake of readability). The black dot represents the "compromise" scenario discussed in Pistocchi et al. (2022) (see text below for details).

as many plants as possible, including those with lower capacity and/or discharging at higher dilution. Based on these considerations, we can consider a "compromise" scenario where advanced wastewater treatment is prescribed at all plants above 100,000 PE irrespective of dilution, and at all plants above 10,000 PE when the dilution ratio at discharge is 10 or less. This scenario is represented by a black dot in Fig. 4, Fig. 5 and Fig. 6. The graphs show how this scenario performs reasonably well in terms of all three indicators, although it may not be strictly optimal according to each of them.

The costs discussed above do not include the upgrade of WWTPs from carbon removal to denitrification, which is de facto a prerequisite of advanced treatment. We estimate the costs of upgrade for the above "compromise" scenario (Fig. 7), following the conservative approach illustrated in the SI (Note 4). Under these assumptions, the costs may be up to about four times as high as the costs of advanced treatment, on a PE basis. Hence even if the total capacity of plants requiring an upgrade to nitrogen removal is relatively small, the total cost of upgrade may be comparable to the total cost of advanced treatment alone in many cases. It should be also stressed that the costs of upgrade may vary very significantly from case to case, and in various circumstances denitrification could be implemented at plants presently required to remove only carbon, through relatively inexpensive improvements in the operation of the process, particularly based on instrumentation, control and automation. Our estimated cost of upgrade varies significantly among countries, depending on the current level of wastewater treatment. Countries where virtually all WWTPs already perform nitrogen removal (such as Germany and Austria) are bound to face lower overall costs. On the contrary, in countries like Spain, Italy, Ireland, Portugal and, to a lesser extent, France and Hungary, the upgrade costs may be large. In any case, even accounting for these differences, they would remain below 0.5 % of the annual income even for the less privileged population in the lowest-income countries of the EU, as shown in Fig. 7.

In the estimation of costs, we neglected the presence of advanced treatment/disinfection in existing plants. When disinfection is present, advanced treatment for MPs could offer opportunities for synergies and economies.

We can also estimate the corresponding GHG emissions from the population equivalents (PE) undergoing advanced treatment under that scenario. As a first approximation, we may consider GHG emissions proportional to cumulative the PE subject to advanced treatment. We assume an emission of 0.225 kg CO2e/m3 of treated wastewater (midrange of literature values) and a wastewater discharge of 0.2 m3/day per PE for a first quantification, obtaining for the above "compromise" scenario the GHG emissions by country as shown in Fig. 8.

The average GHG emissions vary by country depending on the cumulative PE subject to advanced treatment. Emissions represent always <1 % (and mostly between 0.2 and 0.3 %) of the total national emissions of GHG for 2019, and are highest in countries with a less carbon-intensive electric system.

The GHG emission factors estimated here do not take into account the improvement of GHG emissions due to the upgrade of existing plants (potentially lower CH_4 and NO_2 emissions, see Parravicini et al., 2022), which are expected even net of possible additional energy requirements for aeration.

7. Conclusions

MPs used in urban areas and conveyed by wastewater pose a concern, and discharges of WWTPs are, and will likely remain, one of their key entry points to the environment. Therefore, removing MPs during wastewater treatment is an essential component of pollution control. This does not diminish the importance of other actions, such as measures at the source or a mitigation of storm overflows (whose relative contribution to pollution increases when removal of MPs at the WWTP is effective). Control at the source is usually the most cost-effective measure when addressing a specific substance or class of substances (e.g. perfluorinated compounds). However, it is very likely that a broad spectrum of substances will continue to leak

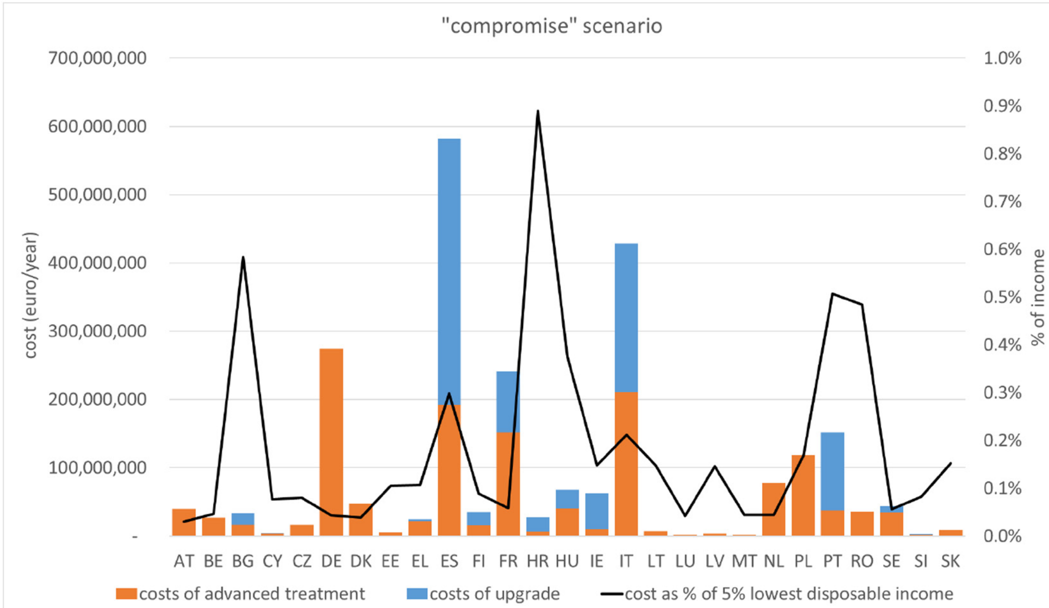

Fig. 7. – Costs of advanced treatment (absolute and as a % of the 2019 annual income of the poorest 5 % of the population). Source for income data: EUROSTAT.

from the technosphere and flow through our WWTPs in the short and mid-term, before we can complete a full ecological transition in our production and use of chemicals. In this perspective, we need a strategic approach to the treatment of MPs at WWTPs. Among the available options for the removal of MPs, mainstream cost-effective solutions are limited to activated carbon, ozonation or their combination in various process configurations. These are largely equivalent in terms of costs, energy use and GHG emissions, and a decision on the solution to adopt necessarily depends on the specific conditions of each case. In specific cases, membrane filtration processes may prove a cost-effective alternative, especially when effluent quality standards need to be high (e.g. for direct or indirect potable reuse of waters). Nature-based solutions such as constructed wetlands (CW) are attractive because of their low energy demand, their operational simplicity and many possible co-benefits including on landscape and biodiversity. On the other hand, CW require spaces that may not be available for WWTPs, particularly in urban contexts, and may struggle with the most recalcitrant chemicals, which makes them an option for effluents polishing, but not an alternative to advanced treatment of wastewater. The suitability and sustainability of an advanced treatment process must be evaluated based on many criteria, such as plant reliability, process flexibility, need of skilled personnel, administrative constraints, availability of space, residual capacity and adaptability of existing biological treatment, etc. Procedures for the integration of these items in a general evaluation and decision making framework have been proposed e.g. for MBR (Bertanza et al., 2017) and for sludge disposal routes (Bertanza et al., 2016).

In this contribution, we have shown that widespread implementation of advanced treatment with O_3, AC or their combination can reduce total toxic discharge and toxicity at the point of discharge by about 75 % (Fig. 4). The percentage of the length of the stream network exposed to medium-high toxicity, based on the indicators defined in Pistocchi et al. (2022), can be reduced from about 7 % to about 3.5 % (Fig. 6B), and the percentage exposed to high toxicity from about 0.9 % to about 0.5 % (Fig. 6A). However, achieving these objectives would cost about 4 billion euro/year for the whole EU. Implementing advanced treatment at all plants larger than 100,000 PE, or between 10,000 and 100,000 PE when the dilution rate is below 10, enables a reasonable compromise, in the range of solutions with the highest effectiveness while preventing costs to increase excessively. At the same time, the capacity of these target plants is arguably sufficient to support the management complexity of advanced treatment.

Under this "compromise" scenario of advanced treatment, we would achieve a reduction by about 50 % of the total toxic discharge (Fig. 4), 40 % of the toxicity at the point of discharge (Fig. 5), a percentage of the stream network exposed to high toxicity of about 0.5 %, and a percentage exposed to medium-high toxicity of about 5 % (Fig. 6). The costs would remain at about 1.5 billion euro/year for the whole EU. Limiting the total PE undergoing advanced treatment enables a proportional limitation of GHG

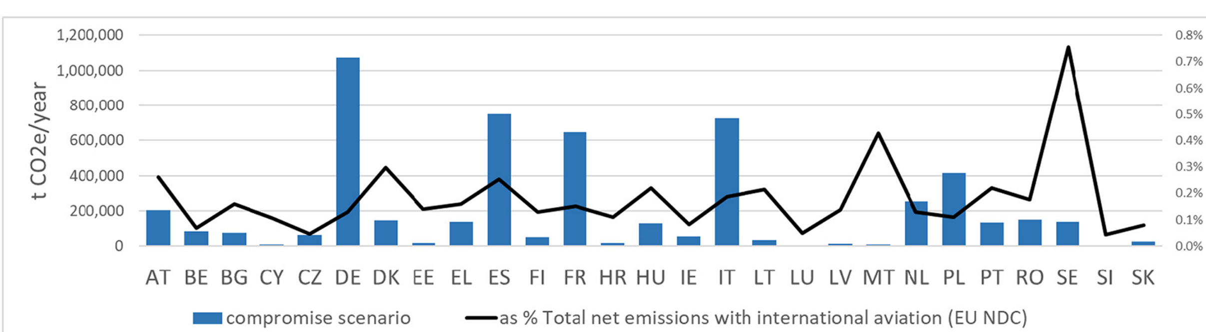

Fig. 8. GHG emissions of advanced treatment under a "compromise" scenario where all plants above 100,000 PE have advanced treatment irrespective of dilution, and all plants above 10,000 PE but below 100,000 PE have advanced treatment if dilution is 10 or less. National GHG emission totals are referred to the year 2019 (source: https://www.eea.europa.eu/data-and-maps/data/data-viewers/greenhouse-gases-viewer/).

emissions. Under the above “compromise” scenario, the estimated total GHG emission is about 5.4 million tonnes CO2e/year, approximately 0.15 % of the EU total GHG emissions in 2019. The increasing decarbonisation of power generation and the development of low-carbon AC, e.g. from biochar including from circular industrial processes (Thompson et al., 2016; Kozyatnyk et al., 2020), may contribute to further reduce, or even neutralize the GHG footprint of advanced treatment.

The costs of the “compromise” scenario appear affordable even compared to the standards of the lowest incomes in the European context, and part of those costs could be required anyway for reasons of nutrient removal or disinfection. Opportunities for synergistic solutions may particularly arise in this respect from fixing non-performing WWTPs, achievement of full compliance with the UWWTD, replacement of existing disinfection/filtration etc. In some cases, a site-specific risk assessment may unveil opportunities for more cost-effective solutions (e.g. source control of hazardous releases from industrial facilities). Therefore the costs estimated for the “compromise” scenario could be further reduced while achieving the same water quality standards.

Further research on the drivers of effluent toxicity, and a wider adoption of advanced treatment processes, may significantly improve the trade-offs between reducing effluent toxicity and controlling the costs of wastewater treatment as well as the negative impacts of treatment processes, including use of energy and consumables.

CRediT authorship contribution statement

A. Pistocchi: Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **H.R. Andersen:** Conceptualization, Data curation, Validation, Writing – review & editing. **G. Bertanza:** Conceptualization, Data curation, Validation, Writing – review & editing. **A. Brander:** Conceptualization, Data curation, Validation, Writing – review & editing. **J.M. Choubert:** Conceptualization, Data curation, Validation, Writing – review & editing. **M. Cimbritz:** Conceptualization, Data curation, Validation, Writing – review & editing. **J.E. Drewes:** Conceptualization, Data curation, Validation, Writing – review & editing. **C. Koehler:** Conceptualization, Data curation, Validation, Writing – review & editing. **J. Krampe:** Conceptualization, Data curation, Validation, Writing – review & editing. **M. Launay:** Conceptualization, Data curation, Validation, Writing – review & editing. **P.H. Nielsen:** Conceptualization, Data curation, Validation, Writing – review & editing. **N. Obermaier:** Conceptualization, Data curation, Validation, Writing – review & editing. **S. Stanev:** Conceptualization, Data curation, Validation, Writing – review & editing. **D. Thornberg:** Conceptualization, Data curation, Validation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.157593.

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