Comparison and predesign cost assessment of ozonation, membrane filtration and activated carbon for the treatment of recalcitrant organics, a conceptual study

Christine Peyrelasse (christine.peyrelasse@apesa.fr)
APESA
Matthieu Jacob
Total SA: TotalEnergies SE
Audrey Lallement
APESA

Research Article

Keywords: Ozone treatment, granular activated carbon, membrane filtration, wastewater treatment, organic pollutant

Posted Date: August 19th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-802348/v1

Abstract

The presence of micropollutants in the environment is today of major concern. These pollutants could have long-term impacts on the environment and on population health. Biological treatment of wastewater is generally insufficient to allow their complete elimination. The establishment of efficient treatments is then needed to degrade the refractory organic matter. Activated carbon adsorption, membrane filtration and oxidation processes are common suitable solutions. All of them have advantages and are effective to treat wastewaters but drawbacks are well known such as waste production, energy consumption or by-products formation. This study aims at defining a strategy to choose the best option according to the nature of the wastewater and the treatment objectives. A methodology was designed for the rating of theses processes to choose the best strategy regarding environmental, technical and economic criteria. A simulation of three wastewater treatment scenarios was carried out to compare the costs of ozonation, adsorption and reverse osmosis. According to the result obtained, a decision tree is proposed to define the best option for a tertiary treatment to reach reuse or discharge objectives.

Highlights

- Micropollutants are of major concern on surface water quality and population health.
- Tertiary treatments remove recalcitrant organics depending on wastewater nature and objectives.
- An optimization of the processes is needed to limit environmental issues.

1. Introduction

The presence of micropollutants in today's environment is of major concern. Research has permit to create new chemical compounds for medicine, chemistry, cosmetics and agriculture (phytosanitary) uses. These compounds belong to the family of surfactants, flame retardants, pharmaceuticals, cosmetics, petrol additives, biocides, pesticides and all of their degradation products. The fate of these new compounds in the environment need to be measured as their behavior, their fate and their (eco)toxicological effects are not very well known (Norman, 2018).

They can have toxic effects (carcinogenic, mutagenic or reprotoxic) or even interfere with the hormonal system of living beings (endocrine disruptors). They could also have health and environmental effects in short or long terms. Furthermore, the potential risk of a "cocktail" effect due to a mixt of these compounds must be considered. Over 1,000 substances have been referenced as emerging environmental substances (Norman, 2019). Since 2000, the European Directive 2000/60/EC (European commission, 2000) sets the reduction target of hazardous substance emission in water. A list of substances or group of substances concerned by the emission reduction (defined as priority substances) or elimination (considered as hazardous priority substances) by 2021 has been defined. The first list, published in 2001, included 33 priority substances (metals, pesticides, hydrocarbons) and evolved with the Directive 2008/105/EC (2008/105/EC, 2008) amended by the Directive 2013/39/EC (2013/39/EU, 2013).

The Industrial Emissions Directive (IED), adopted on 24 November 2010, defines an integrated approach to prevent and reduce the industrial emissions (integrated pollution prevention and control) (European Commission, 2010). The IED provides emission thresholds associated with the performant technic available (BAT-AELs) presented in the BREF documents. BAT-AELs exist for direct emissions of Total Organic Carbon (TOC) and Chemical Oxygen Demand (COD) for a receiving water. For COD, the BAT-AELs threshold for emissions are between 30 to 300 mg L^{-1} (Brinkmann et al., 2016; Chronopoulos et al., 2019; Pinasseau et al., 2018).

With the tightening of regulatory constraints, releases of some pollutants are decreasing, such as pesticides, however other substance releases are increasing, like pharmaceutical substances (Metz & Ingold, 2014). The organic pollutants including emerging pollutants in urban or industrial effluents are not completely biodegradable and a part of these compounds are refractory to biological treatments. Consequently, pollutants are measured at the output of activated sludge treatments (Baalbaki et al., 2017; Coquery et al., 2008; Du et al., 2014; Margot et al., 2013; Ruel et al., 2011), which are the most conventional treatment for urban wastewaters. The main mechanisms involved in the elimination of micropollutants in biological treatments are mainly biodegradation but also sorption onto sludge, air stripping and photo-transformation (Gusmaroli et al., 2020). The implementation of tertiary treatments may be required to allow the elimination of these refractory pollutants and to increase the water quality discharged to surface waters.

Adsorption on activated carbon, Reverse Osmosis (RO) and oxidation are the common treatment solutions implemented as tertiary treatment. Activated carbon is widely employed to remove organic compounds in industrial or drinking waters. This treatment presents various advantages: easily to implement and operate, no chemical needs and no generation of by-products. However, the replacement of the spent activated carbon is necessary and the cost involved limits its use on low COD concentration effluents. RO presents also very high treatment performances but requires efficient pre-treatment and high opertational costs due to energy consumption, elimination of the retained pollutants (retentate) and a qualified staff to operate and control the process. Only the oxidation processes make it possible to really degrade polluants but formation of unknown and potential toxic by-products can occur due to partial oxidation of the compouds (Hamdi El Najjar et al., 2014; Wu et al., 2019).

Studies comparing the costs of tertiary treatments to reduce the discharge of micropollutants are generaly available for urban wastewater treatment plants (Bui et al., 2016; Wahlberg et al., 2010). Few works exist on the field of tertiary treatments employed for industrial wastewaters. This study aims to achieve a comparison and a predesign cost assessment of ozonation, reverse osmosis and activated carbon for the treatment of an industrial effluent with a COD concentration higher than values commonly found in urban discharges. Attempts have been made to estimate the capital and annual operating and maintenance cost (O&M cost) for a 2,000 m^3 d^{-1} capacity treatment plant with a COD concentration of 500 mg L^{-1}. Secondly, a multi-criteria approach is proposed to compare the performance of the processes. Finally, a decision tree is designed to select the most appropriate treatment for the elimination of dissolved organics.

2. Predesign Cost Assessment

2.1 Costs of the tertiary treatments

2.1.1 Ozonation cost

Ozone generators are supplied with air for low capacities (maximum of 75 kgO_3 h^{-1} per ozone generator) and with oxygen for larger needs (200 kgO_3 h^{-1} maximum by ozonator) (Baig and Mouchet 2017). Ozone generators operating with air have higher energy consumption than those operating with oxygen, 13 to 20 kWh $kg^{-1}O_3$ compared to 7 to 13 kWh $kg^{-1}O_3$ (Baig & Mouchet, 2017). The oxygen consumption of ozone generators supplied with oxygen are around 8.3 kgO_2 $kg^{-1}O_3$ (Xylem, 2017) with an oxygen cost estimated at 0.1 € kg^{-1} (besnault et al., 2015).

The first cost of investment is the ozone generators. RECORD (2006) established this cost (excluding engineering) between 150 € $g^{-1}O_3$ h and 30 € $g^{-1}O_3$ h for the highest capacities, greater than 10 kg h^{-1} (RECORD, 2006). Similarly, Mendret et al. (2019) considered a ratio of 100 \$ $g^{-1}O_3$ h for a capacity of 1.15 kgO_3 h^{-1}, based on feedback from suppliers and manufacturers of ozone generators, and Landry Carter (2017) gave a ratio of 54 \$ $g^{-1}O_3$ h corresponding to an investment cost (Ex-works) of \$ 2,500,000 for an ozone generator of 46 kgO_3 h^{-1}. This cost, including the injection, agitation and residual ozone destroyer system, was communicated by the company Primozone (Sweden) (Landry Carter, 2017).

2.1.2 Granular Activated Carbon (GAC) cost

GAC is implemented in a filter bed for the adsorption of organics on granular carbon. Process energy requirements are low for GAC and include both supply and backwash pumping (Hansen et al., 1979). The pollutants are eliminated by adsorption due to their affinity with the activated carbon and its high specific surface area of this adsorbent. The consumption of activated carbon can be first estimated from the COD load treated, typically in the range of 250 to 500 g COD kg^{-1}AC or higher (Truc, 2007). In first approach, a consumption of 250 to 300 g COD kg^{-1}AC is generally considered. Performances of this treatment are dependant of the organics compounds to be adsorbed and in particular the polarity, molecular weight, solubility and concentration. This can be evaluated in laboratory by adsorption isotherms. After the saturation, activated carbon must be replaced and reactivated in high temperature ovens. In France, the reactivation of coal is done in specialized centers, it is too expensive to be carried out on user sites (Bui et al., 2016).

The reactivation yield is dependent on the type of carbon and the nature of the molecule adsorbed. For charcoal made from softwood (pine) this yield is relatively low (70-90%), while for charcoal made from coconut it can reach 98% (information obtained from Chemviron 2019). Treatment of the spent GAC in a reactivation center will require a prior acceptance certificate with limits to be respected for certain parameters such as sulfur, chlorine and fluorine.

The cost of GAC is generally between 1 and 4 € kg^{-1} and the cost for the reactivation is 0.6-0.7 € kg^{-1} (excluding transport). The reactivation cost is slightly higher than the elimination cost (0.4-0.5 € kg^{-1}), but leads to savings on the purchase of new GAC until it cannot be reactivated. Activated carbon treatment should not be used when treated fluxes have too high COD due to the costs associated with the carbon reprocessing. The investment costs were identified and estimated by the company IRH as part of a study carried out for the Rhône Mediterranean Corsica Water Agency. This preliminary design approach is exclusive of taxes and fees and doesn't include supply (cost and mankind), the contracting authority staff and the project management mission (IRH, 2010). The investment costs are very dependent on the capacity and are here established at 50 € m^{-3} d and 625 € m^{-3} d for the highest and lowest capacities respectively (60 and 1 m^3 h^{-1}). Significantly higher costs have been estimated by Guo et al. (2014): 350 € m^{-3} d and 960 € m^{-3} d for the same highest and lowest capacities. For them, the investment cost covers the process, initial charge of activated carbon, piping, control and instrumentation and is linked to the flow quantity treated:

$$\text{Log (Capital cost (\$))} = 0.722 \times \log(\text{flow rate } (m^3\ d^{-1})^{1.023} + 3.443)$$ Guo et al. (2014).

This equation was mainly defined using a simulation tool created by the Water Research Foundation and the USEPA.

Altogether, the investment costs depend on the flow treated and on its composition influencing the nature of the material used, the kinetic of filtration end the number of filters used.

2.1.3 Membrane filtration cost assessment

Reverse osmosis and Nanofiltration (NF) are implemented for water reuse or very strict constraints on discharges (very low threshold or low water flow rate authorized for discharge). They require more efficient pre-treatment than adsorption and ozonation. Energy consumption is higher and the elimination of retentate (10 to 30% of the initial volume of water treated) remains problematic impacting the operating fees. NF and RO investment costs, identified by the company IRH, are estimated near 1,500-1,750 € m^{-3} d (for units treating 200-300 m^3 d^{-1}) and 7,500 € m^{-3} d (for a capacity lower than 10 m^3 d^{-1}). In addition, an additional cost of 25 to 30% must be taken into account for the assembly, commissioning, etc. (IRH, 2010). In another study, Plumlee et al. (2014) have proposed an equation for capital cost estimation linking the overall investment (process and auxiliary costs) to RO and NF:

$$\text{Capital cost } (\$M\ MGD^{-1}) = 7.14 \times \text{flow rate (MGD)}^{-0.22}$$ Plumlee et al. (2014)

Where, capital cost is in \$M MGD^{-1} and the flow rate is in MGD (Millions of Gallons per Day) with 1 US MGD = 3,785 m^3 d^{-1}. This equation can be used for flows higher than 1 MGD.

The overall capital cost was thus estimated at 1,500 € m^{-3} d for a unit treating 3,800 m^3 d^{-1}. This cost could be lower for the treatment of slightly polluted unsalted water.

For our simulations, the equation of Plumlee et al. (2014) will be used for a simulated unit with a flow at 0.5 MGD (assumption supported by the supporting information of (Plumlee et al., 2014) publication).

2.2 Cost assessment of the different wastewater treatment scenarios used for the comparison of costs

Studies comparing the costs of tertiary treatments are generally carried out for urban wastewater treatment plant applications (Bui et al., 2016; Choubert, 2018; Wahlberg et al., 2010). Only few works exist on these tertiary treatments on industrial wastewaters. The economic considerations drive the selection of the process implementation. Here, a simulation of different scenarios for is proposed to treat a conceptual effluent with a COD concentration at 500 mg L^{-1} (higher than urban waste concentration). A comparison of the 3 different treatment processes (ozonation, adsorption and membrane filtration) is carried out on a cost basis. Treatment channels considered are presented in **Fig. 1**. The channel with membrane filtration (ultrafiltration / reverse osmosis) is considered to study a scenario with water reuse.

In the different scenarios tested, the three post processes are described as follow:

- Membrane channels: wastewater is first filtered by ultrafiltration and reserve osmosis and retentate is post-treated by ozonation. Two scenarios are proposed here with permeate directly discharged (Channel 1) or reused (Channel 1 bis).
- Activated carbon channels: wastewater is treated by AC post-treatment, the obtained water can directly be discharged. Two scenarios here are considered using either reactivated AC (Channel 2) or new AC (Channel 2 bis).
- Finally, wastewater can be processed directly by ozonation treatment allowing direct discharge.

2.2.1 Hypothesis used in the simulation

General assumptions used for simulation are:

- A flow rate at 2 000 m^3 d^{-1}, 24h/24, 365 days per year;
- An electricity cost at 0.1 € kWh^{-1};
- A staff cost fixed at 50 € h^{-1}.

The capital cost is amortized over 20 years (n) (Besnault et al., 2014) considering an interest rate (r) of 4.5% y^{-1} (Margot et al., 2013). The amortized capital cost (A) is given by the following equation (Mahamuni & Adewuyi, 2010):

$$A = \frac{\text{Total capital} \times r}{1 - \left(\frac{1}{1 + r}\right)^{n}}$$

Specific hypotheses of each scenario are listed in Table 1.

Table 1. Assumptions for cost estimation (APESA, RECORD 2020).

Item	UF + RO + O_3, Channel 1 and 1 bis			GAC, Channel 2 and 2 bis		O_3, Channel 3
	UF	**RO**	**O_3**	**GAC Elimination**	**GAC Reactivation**	**O_3**
Performances	10% COD reduction 95% recovery ratio	95% COD reduction 80% recovery ratio (f)	> 90% COD reduction 90% O_3 for transfer efficiency (l)	300 gCOD kg^{-1}GAC		> 90% COD reduction 90% O_3 for transfer efficiency (l)
Maintenance	0.026 \$ m^{-3} x conversion rate €/\$ (a)	3% capital cost y^{-1} (g)	1.5% of the process capital cost y^{-1} for the annual part replacement cost (m)	1.5% of the process capital cost y^{-1}		1.5% of the process capital cost y^{-1} for the replacement (m)
Operating and maintenance staff	21h month^{-1} (b)	0.2 € m^{-3} (h)	150 h month^{-1} (n)	22 h month^{-1} (t)		170 h month^{-1} (n)
Consumables (chemicals, GAC, membrane)	0.025 \$ m^{-3} x conversion rate €/\$ for membrane replacement and 2% of capital cost y^{-1} for the cleaning agents (a,c)	0.1 \$ m^{-3} for membrane replacement and 0.1 \$ € m^{-3} for the cleaning agents ^{3} x conversion rate \$/€ (g,i)	2.5 gO_3 g^{-1} COD (o) 8.3 kgO_2 kg^{-1}O_3 (p) 0.1 € kg^{-1}O_2 (q)	2 € kg^{-1} for a new one and 0.45 € kg^{-1} for the elimination	0.7 € kg^{-1} with a regeneration efficiency of 95% and 2 € kg^{-1} for a new one	2.5 gO_3 g^{-1}COD (o) 8,3 kgO_2 kg^{-1}O_3 (p) 10 kWh kg^{-3}O 0,1 € kg^{-1}O_2 (q)
Electrical consumption	0.3 kWh m^{-3} permeate (d)	2.8 kWh m^{-3} permeate (j)	10 kWh kg^{-1} O_3 (r)	0.019 kWh m^{-3} (u)		10 kWh kg^{-1}O_3 (r)
Water saving		1.45 € m^{-3} (k)				
Capital cost (€)	Conversion rate €/\$ x Flow rate (MGD) x 3.57 x flow rate (MGD)$^{-0.22}$ x 10^6 (e)	Conversion rate €/\$ x Flow rate (MGD) x 7.14 x flow rate (MGD)$^{-0.22}$ x 10^6 (e)	Process (30 € g^{-1}O_3 h) + piping (30%) + site work (10%) + contractors (15%) + engineering (15%) + contingencies (20%) (s)	Conversion rate €/\$ x $10^{(0.722 \times \log(\text{flow rate}^{1,023} + 3.443))}$ + site work (10%) + contractors (15%) + engineering (15%) + contingencies (20%) (a,s)		Process (30 € g^{-1}O_3 h) + piping (30%) + site work (10%) + contractors (15%)+ engineering (15%)+ contingencies (20%) (s)

Reference used: (a) - (Guo et al., 2014); (b) - (Margot et al., 2011); (c) - (Andrade et al., 2015); (d) -(Guo et al., 2018) ; (e) - (Plumlee et al., 2014); (f) - (Bick et al., 2012); (g) - (Shouman et al., 2015); (h) - (Koroneos et al., 2007); (i) - (Andrade et al., 2017); (j) - (Burn et al., 2015) ; (k) (Pedro-Monzonis et al., 2016); (l) - (Margot et al., 2013), (m) - (Mahamuni & Adewuyi, 2010); (n) - (Mundy et al., 2018) ; (o) - (de Franceschi, 2018); (p) - (Xylem, 2017); (q) - (besnault et al., 2015); (r) - (Baig & Mouchet, 2017): (s) - (G.Melin (Ed.), 1999; Mahamuni & Adewuyi, 2010); (t) - (Hansen et al., 1979) ; (u) - (Nijdam et al., 1999).

2.2.2. Results of cost calculation and discussion

The O&M (Operating and Maintenance cost) consists of maintenance, operating and maintenance staff, consumables and electrical costs. The consumables include the membrane replacement costs, the chemical costs and the GAC replacement with elimination or reactivation of the spent activated carbon. It also took in consideration the water saved (channel 1 bis). The annual O&M and capital costs for all the scenarios have been calculated based on the aforementioned assumptions and are presented in Supplementary material**.**

The highest cost corresponds to the channel 1 which implements membrane filtration and ozonation for retentate treatment. This result confirms those of Wahlberg et al. (2010) and Choubert (2018) who showed that the reverse osmosis channels have the highest costs. The treatment cost can be revised downwards for the membrane filtration channel with water reuse. Considering that the water treated by RO is of very good quality and can be recycled, purchase of water and payment fees for withdrawals and rejections are saved. Indeed, the RO has the advantage of removing salts. Here an economy of 1.45 € m^{-3} of water recycled is considered (cost of surface water for industrial used (Pedro-Monzonis et al., 2016)). This cost can be higher or lower depending on the origin of water. Pedro-Monzonis et al. (2016), for example, considered for industrial water a cost of 0.18 € m^{-3} by employing groundwater. Thereby, RO with water saving can be a very interesting option depending on the regulatory of the sites and the price of water. When the activated carbon can be reactivated, the overall treatment cost is the lowest simulated. However, if GAC is eliminated and not reactivated, cost increases from 1.5 € m^{-3} to 4.2 € m^{-3} impacting the overall cost. In the case of ozonation, a cost of 3.6 € m^{-3} has been calculated.

This simulation provides a comparison of the costs for the different tertiary treatment plants and shows the impact of the various assumptions on the overall result. However, estimations are carried out on the basis of average treatment performance assumed for a fictitious effluent and not from available data for a real effluent. The treatment rates necessary for ozonation, the consumption of activated carbon as well as the performances of membrane filtration are specific to each effluent and could differ significantly from the assumptions made for this conceptual study.

3. Comparison Of Processes And Strategy Of Treatment

3.1 Environmental impact

A comparison of the environmental impacts of ozonation, GAC and membrane filtration has been carried out by Li et al. (2019). The tertiary treatments studied are positioned at the outlet of urban treatment plants. The toxicological impact linked to the rejection of hazardous products and pharmaceutical residues is included in the assessment. This study showed that the scenario with reverse osmosis (RO) has the highest environmental impact due to the consumption of energy and material during operation. Bayer et al. (2005) performed a Life Cycle Analysis (LCA) to compare the impact of a new GAC with that of a reactivated GAC. The results showed that (thermal) reactivation allows to avoid impacts linked to mining and transformation of raw coal, to natural gas consumption used (reactivation represents 40% of the gas consumption needed for the AC production), to toxicity risk (by 25%) and to CO_2 emissions (by almost 90%) (Bayer et al., 2005). The implementation of a tertiary treatment is generally effective in reducing the residual toxicity of an effluent if the potential generation of by-products generated by oxidation processes (ex. ozonation) is controlled. However, the tertiary treatment requires additional consumption (chemicals, electricity, construction materials) which may affect the overall environmental impact. This is the conclusion reached by Rahman et al. (2018) for the treatment of emerging pollutants leaving urban wastewater treatment plants. Membrane filtration is the most impacting process due to the high energy consumption implemented unlike reactivated carbon filtration. In the future, an optimization of tertiary treatments is necessary to reduce their environmental impact and in particular their energy consumption (Margot, 2014).

3.2 Performances of the tertiary treatments

A comparison of the different processes studied is complex because the mechanisms for removing pollutants are very different. Membrane filtration and adsorption are separative processes while oxidation processes are destructive processes. Indeed, pollutants retained by RO or GAC are effectively eliminated from the treated water while the oxidation processes generally produce oxidation by-products leading to potential incomplete mineralization of pollutants. Moreover, performances are dependent on the wastewater quality (salinity, type and pollutant concentrations), the technology used and the operation conditions (type of membrane, pressure applied, nature of the activated carbon, kinetics, etc.).

Despite these points of attention, a summary of process performances according to category of pollutants is proposed in **Table 2**.

Table 2. Yields of elimination of priority and emerging pollutants by oxidation, adsorption and membrane filtration processes (Compilation APESA, RECORD 2020) adapted from (Besnault et al., 2014; Ruel et al., 2011).

	Membrane filtration (RO)	Granular activated carbon	Ozone
Organometallics	> 90% (a)	40-70% (a)	<40% (a)
Beta blockers	> 70% 30-70% for oxyprenolol (b)	> 99% at 6 months (k) 30-70% for timolol and nadolol (b)	> 98% (k)
Antibiotic drugs	> 70% (b)	> 73% (k)	> 72% (k)
Other drugs (carbamazepine, diazepam, diclofenac and erythromycin)	> 70% (except erythromycin) (b)	> 85% at 6 months (k)	> 99% and 75% for diazepam (k)
Surfactants / detergents	(a)	(a)	(a)
Polycyclic Aromatic Hydrocarbon	59-72% (c) 50-91% (d) 80% Naphtalene (e) > 90% Acenaphtylene, Phenanthrene, Pyrene (f) > 90% Benzo(a)pyrene (g)	Partially absorbed less and less efficient (k) 72% Benzo(a)pyrene, 94% fluorene (g)	> 90% for acenaphtylene, acenaphtene and pyrene our partial elimination (k)
Perfluorinated compounds	> 90–99% (m) (h)	64% PFOS and 45% PFOA (l) to > 90% (m) > 90% PFOA et PFOS (n)	< 10% for PFOA 10-50% for PFOS (m)
Pesticides (Atrazine, diuron, simazine, isoproturon)	> 70% Simazine and Diuron (b) 84–97% Atrazine (i)	> 85% (k)	> 55% except for diuron, (91%) (k)
Pesticides (glyphosate, AMPA)	< 30% AMPA (b) > 70% glyphosate (b) > 90% glyphosate and > 95% AMPA (j)	Elimination occurred only for 3 months (k) Non reliable (j) Low performance (o)	> 70% glyphosate (b), 63% AMPA (b), 60-99% glyphosate and 25-95% AMPA (j)
Color reference for the performances	< 30%	30– 70%	> 70%

Reference used: (a) (Fono & McDonald, 2008), (b) - (Ruel et al., 2011), (c) - (Smol et al., 2015), (d) - (Gong et al., 2017), (e) (Dupont, 2020), (f) - (Zhu, 2015), (g) - (Snyder et al., 2007), (h) - (Flores et al., 2013), (i) - (Brinkmann et al., 2016), (j) - (Jönsson et al., 2013), (k) - (Besnault et al., 2014), (l) - (Flores et al., 2013), (m) - (AWWA), (n) - (Cummings et al., 2015), (o) - (Lenntech).

Membrane filtration reach the better performances for the elimination of priority and emerging pollutants. Granular activated carbon is efficient on a large part of the pollutants studies but organometallics and some pesticides can’t be eliminated. In the same way, ozonation is not o good option for organometallics and perfluorinated compounds but it can reach good performances on some pesticides, drugs and Polycyclic Aromatic Hydrocarbon.

Performances of ozonation and activated carbon were compared in the framework of the MicroPoll project and showed that ozonation followed by a sand filter and Powder Activated Carbon (PAC) is effective in removing the majority of micropollutants with similar average removal rates. Ozone is very effective for some types of pollutants while PAC (Powder Activated Carbon) acts on a wider range of substances (but with lower yields) (Margot et al., 2013). Ozone and PAC significantly reduce the effluent toxicity with comparable costs (PAC separated by sand filter) (Margot et al., 2013) or higher (ultrafiltration used for PAC recovery). Combination of ozone and activated carbon allow to avoid the risk of toxic oxidation by-product rejection, adsorbed by AC. This coupling is implemented on urban wastewater treatment plants in Switzerland (Grelot et al., 2017). The French project Ampère has shown that RO has very high micropollutants abatement performance. Molecules hardly eliminated by activated carbon and ozonation were retained (Ruel et al., 2011).

3.3 Comparison of the processes

The three processes studied have specific characteristics with their own performances and degradation or separation efficiency. A comparison of the tertiary processes is proposed in **Table 3** considering the main criteria that impact the choice of the treatment.

Table 3. Comparison of the main tertiary treatments: ozonation, membrane filtration and activated carbon. TSS (Total suspended solids).

	Ozonation	Membrane filtration NF/RO	Activated Carbon
Maturity	Standard process for the micropollutant treatment	Poorly used for micropollutant treatments, suitable for water reuse	Standard process for the micropollutant treatment
Performance	Very high	Very high	Effective (depending on AC and regeneration frequency)
Pretreatment need	Elimination of biodegradable COD and TSS	Filtration of TSS	Elimination of biodegradable COD and TSS (< 10 mg/L)
Chemicals	Oxygen	Scale inhibitor and soda (for membrane cleaning)	None
Electrical consumption	High	High	Low (except for reactivation – high)
Staff qualification	Medium	High	Low
Implementation	Needs safety equipment	Available in modules	Easy. Possibility of renting.
Environnemental impact	Intermediate potential toxicity (degradation by-products)	High. (coupling transport and treatment of retentate, energy and water recovery)	Low (Transport for the treatment of spent GAC)
Advantages	Strong oxidizing power Effective for a wide number of pollutants Numerous feedbacks Disinfection	Very high performance Salt removal Possibility to reuse treated water	Ease of implementation and management modular Low investment cost, rental possibility Numerous feedbacks
Limits	High costs Generation of potentially toxic oxidation by-products Ineffective on certain substances	Non-destructive process (10-30% of retentate to be eliminated) Energy consumption Risk of fouling Maintenance : cleaning	Periodic replacement (elimination/reactivation) Risk of clogging Drastic performance reduction with an improper AC

To complete this first comparison, a multi-criteria approach is proposed to assess and compare the processes. Adapted from the methodology reported by Fast et al. (2017), this approach is based on the evaluation of process performance according to technical, environmental and economic criteria. Different key points have been defined with a rating base proposed allowing the reproducibility of this approach presented. The rating is carried out by applying a score ranging from 1 to 5. The highest scores correspond to the best performances. Technical and environmental criteria rating are based on the data listed previously and regrouped in topics detailed by method in Supplementary material. This ranking is not an absolute indication indeed, all the criteria were considered without weighting according to the method used by Fast et al. (2017). This could be discussed, in the same way other criteria could have been considered. The scoring results obtained are shown in **Table 4** and **Fig. 2**.

Table 4. Results of the scoring of the processes (APESA, RECORD 2020).

Criteria	Quotation basis	Reverse Osmosis	Reverse Osmosis with water reuse	GAC Reactivation	GAC Elimination	Ozonation
Technical criteria		3.6	3.6	4.4	4.4	4.2
Level of development	Nbr of industrial units	4.0	4.0	5.0	5.0	5.0
Technical reliability	Material reliability	3.0	3.0	4.0	4.0	4.0
Performances	High efficiency and constant over time	5.0	5.0	4.0	4.0	4.0
Flexibility / adaptability	Variation of flow and load to be treated	3.0	3.0	4.0	4.0	4.0
Operational Complexity	Nbr of processes and their innovative aspect	3.0	3.0	5.0	5.0	4.0
Environmental impact		3.5	3.5	4.3	3.8	4.0
Energy consumption		3.0	3.0	5.0	5.0	3.0
Toxicity of the water treated	Generation of oxidation by-products or chemical releases	5.0	5.0	4.0	4.0	4.0
Wastes (production and transport)		2.0	2.0	4.0	2.0	5.0
Possibility of recycling water		4.0	5.0	4.0	4.0	4.0
Economic criteria		2.8	3.3	4.3	2.9	3.2
O&M cost (€ m^{-3})		1.7	2.8	3.6	0.9	2.2
Capital cost (€ m^{-3})	Amortized annual	3.8	3.8	4.9	4.9	4.2
Average Score		**3.3**	**3.6**	**4.3**	**3.7**	**3.8**

First, all the process analyzed have a ranking between 3.3 and 4.3 showing globally good performances. Activated carbon adsorption obtains the best results when reactivation is applied on the spent carbon. On the contrary the RO presents the lower score due to the complexity of operation with the risk of fouling and scaling. Moreover the production of wastes (retentate) is a major drawback that increases the O&M cost. On contrary the possibility of reusing water is a great advantage that can lead to choose this solution. It can solve problems of availability of freshwater or impossibility to discharge wastewater.

The results of this ranking have to be considered only for the scenarios studied. Other results could have been found for other wastewaters with lower flowrate or organic concentration. The choice of the process will be made according to the effluent to be treated (flow rate, load, nature of pollutants) and the local context. Moreover, the quotation applied may differ depending on the person carrying out this quotation with a subjective part on the weighting parameters for example. However, the proposed methodology makes it possible to define technical, environmental and economic criteria and to balance it according to country legislation.

3.4 Strategy defined for the estimation of the tertiary treatment best option

These different processes can be complementary and must be chosen according to the effluent nature to be treated, its flow rate and the treatment objectives to be achieved.

Biological processes not evaluated in this study have the lowest costs and are preferred when pollution is biodegradable. They are widely used for the treatment of urban and industrial wastewater. Partial oxidation can be necessary to increase the biodegradability of an effluent before a bio-treatment in presence of recalcitrant pollutants. This option is generally less expensive than total oxidation when the residual COD is greater than 350 mg/L. Membrane filtration will be useful for water reuse projects and/or when the effluent is refractory to oxidation. Adsorption will be implemented for the treatment of low flows of organics or pollutants refractory to oxidation. The adsorption can also make it possible to eliminate oxidation by-products. A simplified decision tree has been designed to choose the best option according to the effluent composition for the treatment of organic pollutants from industrial or urban effluents **(Fig. 3)**.

As described previously, the choice of the best wastewater treatment scheme will be a combination of processes on a case-by-case basis depending on the effluent to be treated and the treatment objectives. However, by coupling this decision tree and the ranking methodology presented, an impartial and customizable choice can be made.

4. Conclusion

This study aims at establishing the better course for effluent remediation with dissolved organic matter load with refractory compounds. Different wastewater treatment strategies can be studied depending on the nature of the wastewater, the water flow rate and the treatment objectives.

A methodology was designed for the rating of tertiary processes to choose the best strategy on environmental, technical and economic criteria.

From this tool, we have compared the most effective tertiary treatments for recalcitrant COD. The best result was obtained by the activated carbon when reactivation is applied on the spent carbon which allowed to reduce the O&M cost and the environmental footprint. On the contrary the reverse osmosis presents the lower score due to the complexity of operation and the production of retentate which is a major drawback that increases the O&M cost and the environmental footprint. However, the possibility of reusing water is a great advantage that can lead to reduce the O&M cost and solve problems of availability of freshwater or impossibility of discharge of wastewater. Ozonation obtained intermediate result. It is very effective but costs can be high when the organics concentration of the wastewater is high. Despite their performance, oxidation processes generally do not completely mineralize organic matter and the formation of potentially toxic oxidation by-products can be problematic. A post-treatment (sand filter, adsorption on activated carbon or bio-treatment) may be necessary to eliminate these by-products. Generally, ozonation is very effective, however, it can't oxidize all the molecules. Advanced Oxidation Processes (AOPs) could be a solution in this case. They are able to generate non-selective and highly reactive hydroxyl radicals, which are effective on the majority of organic compounds. They can be chemical, photochemical, catalytic, electrochemical, sonochemical and physical. The AOPs will be reserved for the treatment of the most problematic effluents with difficult compounds. Generally, adsorption on activated carbon and membrane filtration (NF/RO) can deal with effluents refractory to oxidation processes. When the refractory COD concentrations are high, other methods will be more suitable. Evapo-concentration or thermal oxidation processes (wet oxidation, supercritical oxidation) can manage effluent concentrations up to 100 g L^{-1} of COD, above these concentrations incineration is generally the only treatment solution.

A decision tree was designed to summarize the results of this study and facilitate the choice of the processes to implement for the treatment of organic (non-particulate) pollutants in low concentration. The choice of the treatment process or the combination of processes will be made on a case-by-case basis depending on the effluent to be treated and the treatment objectives.

Declarations

Acknowledgements

Authors are grateful to the French Industry–University Cooperative Research Network on environment and Waste (RECORD – www.record-net.org) for its contribution to the funding of this work and for providing industrial orientations and scientific supervision to the research.

Funding – This project was fund by the French Industry–University Cooperative Research Network on environment and Waste (RECORD).

Conflicts of interest/Competing interests – Authors do not known on any conflicts of interest or competing interests.

Ethics approval – No experimentations have been performed in this publication.

Consent to participate & Consent for publication – All of the authors have been consulted before submission of this paper.

Availability of data and material – A supplementary material is available, all the calculations performed are explained and any supplementary questions can be address at email address given.

Code availability – No software were used in this publication.

Authors' contributions – Peyrelasse performed the calculations and write the publication; Lallement, & Jacob review the publication.

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Figures

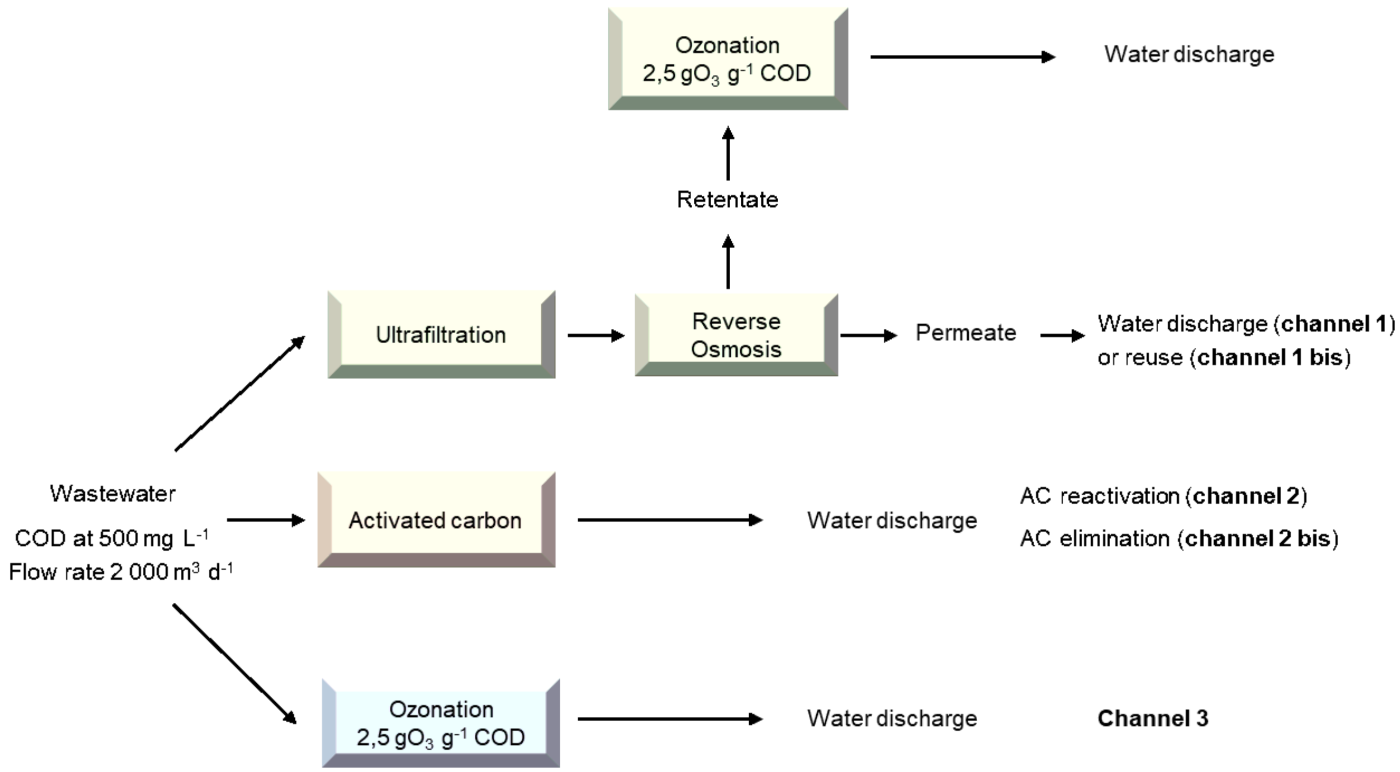

Figure 1

Wastewater treatment scenarios used for the cost evaluation. The COD concentration of the water discharge is less than 50 mg L-1.

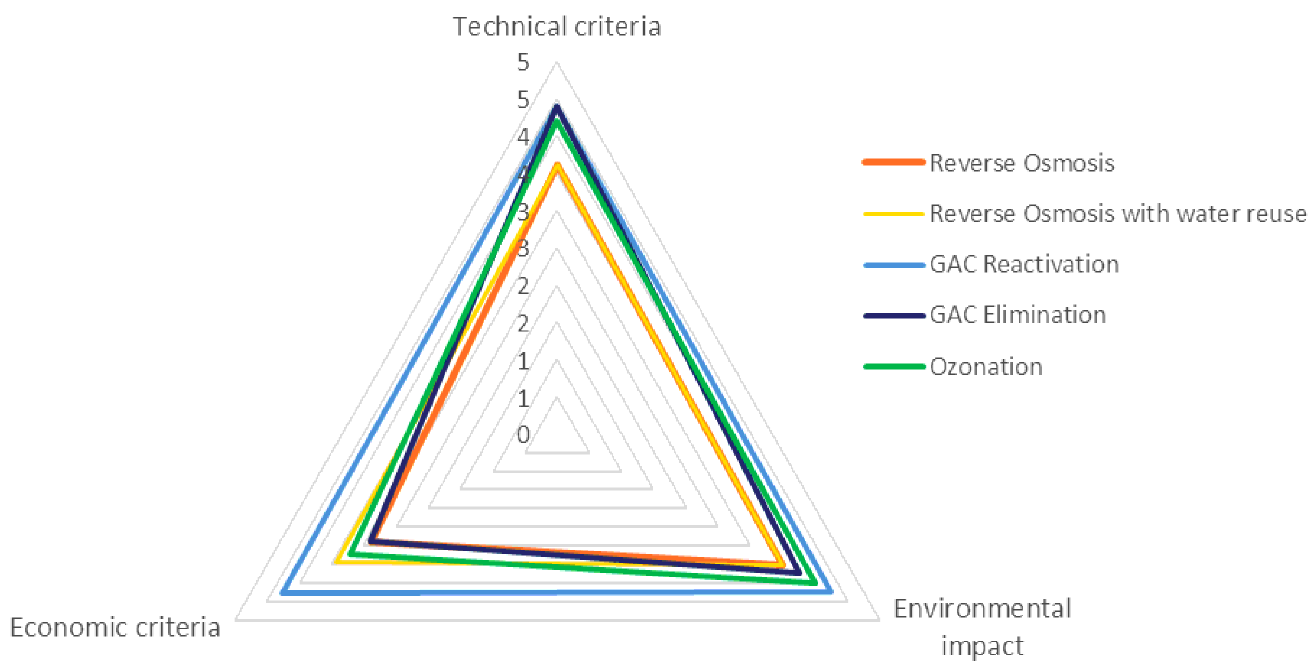

Figure 2

Comparison of ranking of the three technologies (ozonation, RO with or without water reuse and GAC with reactivation or elimination) according technical, economic and environmental criteria.

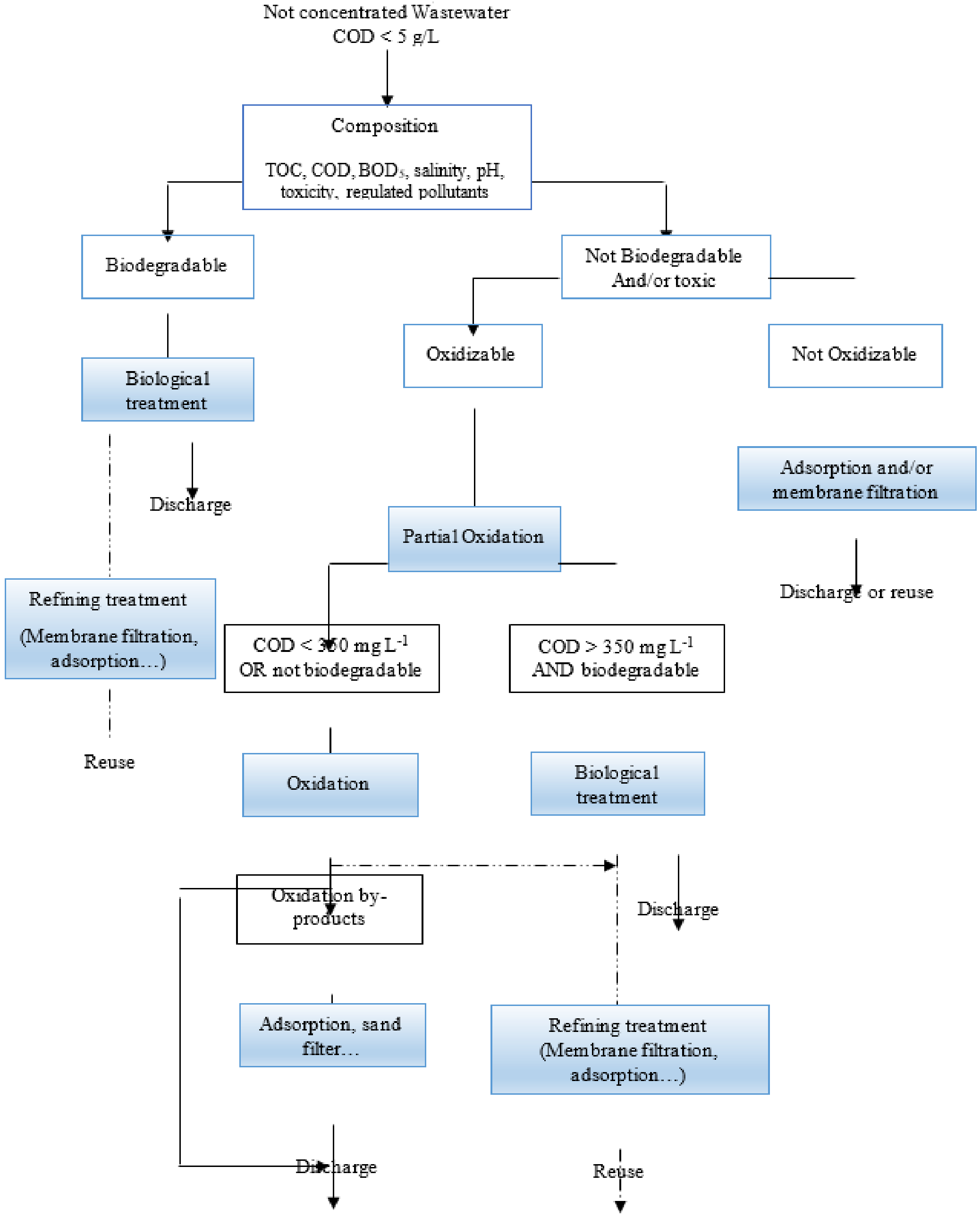

Figure 3

Decision tree designed for the treatment of organic (non-particulate) pollutants in low concentration.

Supplementary Files

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