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How to choose the best tertiary treatment for pulp and paper wastewater? Life cycle assessment and economic analysis as guidance tools

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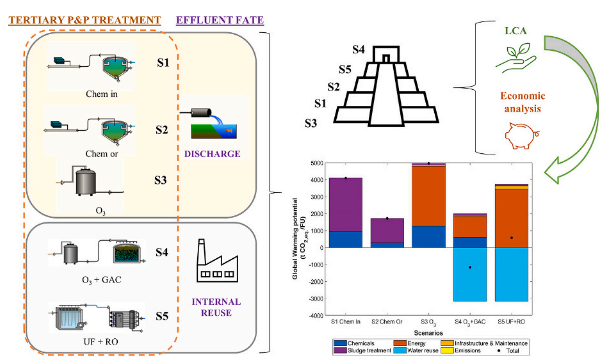
HIGHLIGHTS

- LCA and economic analysis were used to rank different P&P tertiary treatments.
- Coagulation-flocculation, O_3, O_3+GAC, and UF+RO were compared.
- Effluent reuse strongly reduces the environmental impacts for UF+RO and O_3+GAC.
- Energy use is very high for O_3 alone, while it is reduced for UF+RO and O_3+GAC.
- O_3+GAC is the preferable solution from economic and environmental standpoints.

GRAPHICAL ABSTRACT



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ABSTRACT

Pulp and paper wastewater (P&P WW) often requires tertiary treatment to remove refractory compounds not eliminated by conventional biological treatment, ensuring compliance with high-quality effluent discharge or reuse standards. This study employs a life cycle assessment (LCA) methodology to compare alternative tertiary treatment technologies for P&P WW and rank them accordingly. The evaluated technologies in the scenarios include inorganic (S1) and organic (S2) coagulation-flocculation, ozonation (O_3) (S3), O_3+granular activated carbon (GAC) (S4), and ultrafiltration (UF)+reverse osmosis (RO) (S5). The analysis focuses on a P&P wastewater treatment plant (WWTP) in Northeastern Italy. The LCA is complemented by an economic analysis considering each technology's capital and operating costs, as well as potential revenues from internal effluent reuse. Results indicate that S4 (O_3+GAC) outranks all the other scenarios in terms of both environmental performance and economic viability, primarily due to the advantages associated with effluent reuse. S5 (UF+RO), which also involves reuse, is limited by the high energy consumption of UF+RO, resulting in increased environmental impacts and costs. The physicochemical scenario S2 (Chem Or), currently utilized in the WWTP under study, remains the best-performing technology in the absence of effluent reuse. In contrast, S3 (O_3 alone) exhibits the poorest environmental and economic outcomes due to substantial energy requirements for O_3 generation and

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the inability to reuse the treated effluent directly. Lastly, a sensitivity analysis underscores the strong influence of chemical dosages in S1 and S2 on environmental and economic impacts, which is more significant than the impact of water reuse percentages in S4 and S5. The high electricity cost observed during 2022 negatively affects the energy-intensive scenarios (S3-S5), making coagulation-flocculation (S1-S2) even more convenient.

1. Introduction

The demand for freshwater is experiencing an exponential surge in conjunction with global population growth, while the effects of climate change make freshwater sources increasingly vulnerable and less predictable, compromising both their quantity and quality. In light of these challenges, the imperative to boost treated industrial wastewater (WW) reuse arises not only as a means to address water scarcity but also to attain overall water sustainability (Rathoure, 2020), implementing circular economy principles.

Among the industrial sectors, the pulp and paper (P&P) industry stands out as a prominent water consumer, discharging approximately 3 billion m^3 of WW annually, contributing to roughly 30 % of the global industrial WW generation (Hou et al., 2020). The extent of water utilization within the P&P industry depends on various factors, including the specific production processes, the type of paper being manufactured, the facility size, and the extent of water reclamation and reuse. On average, the water consumption of the P&P sector ranges from 10 to 300 m^3 per ton of product, amounting to a global annual consumption of 4000 to 120,000 million m^3 (Esmaeeli and Sarrafzadeh, 2023).

Hence, the implementation of a closed-loop industrial water system has the potential to yield a substantial reduction (or even complete elimination) of liquid discharges (e.g., through the zero liquid discharge concept) (El-Awady et al., 2019), as well as a decrease in freshwater consumption through process water and treated WW reuse (Rathoure, 2020). Despite the associated increase in operational expenses and initial capital costs, opting for treated WW reuse within the same P&P industry offers noteworthy advantages, including savings associated with freshwater purchase and positive environmental impacts (Pizzichini et al., 2005).

In addition to the water directly utilized in P&P production, encompassing activities ranging from raw material washing to finished product cooling, significant water volumes are employed for cleaning and maintenance. The resultant P&P WW consists of a complex mixture of organic and inorganic compounds, including lignin, cellulose, and hemicellulose, which pose challenges in terms of biodegradability. The organic matter content, quantified as chemical oxygen demand (COD), is considerably high, reaching levels up to 9000 mg/L (Han et al., 2021). Moreover, many COD components exhibit poor biodegradability, including chlorinated compounds, unsaturated fatty acids, ethylenediaminetetraacetic acid (EDTA), and diethylenetriaminepentaacetic acid (DTPA) (Roudier et al., 2015). Such organic matter features present significant hurdles in P&P WW treatment, particularly when considering the increasing stringency of effluent quality standards required to protect the environment or allow effluent reuse (Mauchauffee et al., 2012).

As a result, after secondary treatment, which is often conducted by means of conventional activated sludge (CAS) (Mainardis et al., 2020), tertiary treatment becomes imperative to comply with strict discharge limits, particularly concerning COD and total suspended solids (TSS), while nutrient concentration (N, P) appears not to be an issue, due to very limited concentrations in raw P&P WW (Mainardis et al., 2022b).

Tertiary treatment commonly involves physical and/or chemical processes (flotation, adsorption, membrane filtration, coagulation-flocculation, ion exchange, ozonation, and advanced oxidation), targeting the removal of residual TSS, colloids, color, toxic substances, and recalcitrant COD (Teng et al., 2014). Among these techniques, coagulation-flocculation is still the prevalent method at full-scale level, involving the addition of an organic or inorganic coagulant to the WW to destabilize TSS and colloidal particles, followed by the dosage of a flocculant (typically a high-molecular-weight polymer) to facilitate the formation of large flocs that can be easily separated by means of conventional sedimentation (Metcalf, and Eddy, Inc., 2015). This technique is efficient, relatively straightforward to operate, and effective in removing a wide range of contaminants, depending on the chemicals' characteristics and the operational conditions. However, chemicals' consumption and chemical sludge generation pose significant concerns (Mainardis et al., 2022b; Mehmood et al., 2019), suggesting the exploration of alternative approaches to achieve economic and environmentally sustainable treatment processes. Several other technologies (including filtration and ozonation), classified as Best Available Technologies (BAT) by the European Joint Research Center (JRC), have been identified, although their full-scale implementation remains at present limited (Roudier et al., 2015).

Membrane-based processes, such as ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO), produce high-quality effluents amenable to reuse by employing advanced filtration levels (Mänttäri et al., 2006). However, their notable energy requirements, limited membrane lifespan, and maintenance challenges, together with the need for pre-treatment, still limit their widespread application (Kumar et al., 2021). Ozonation, while effective in removing organic contaminants and pathogens thanks to the powerful oxidation process pursued by ozone and OH• radicals, demands substantial electricity levels for ozone production (Mainardis et al., 2020), although it results in limited waste generation (Arzate et al., 2019). Adsorption through granular activated carbon (GAC) beds is another promising technology widely used as a tertiary WW treatment, offering versatility, high separation efficiency, ease of operation, and micropollutants removal capability (Altmann et al., 2016). However, GAC performances deteriorate under high pollutant loads, necessitating frequent adsorbent regeneration which increases the operating costs (Hou et al., 2020). Combining multiple technologies can provide an effective solution (Esmaeeli et al., 2023): e.g., GAC filtration can be applied as a post-treatment of ozonated effluents (Kreetachat et al., 2009).

Therefore, the selection of the most appropriate tertiary WW treatment technology depends on various factors, including the desired effluent quality for the specific reuse applications, the environmental impact, and the capital and operating costs, which can be challenging to evaluate simultaneously (Mainardis et al., 2022b). Life cycle assessment (LCA) coupled with economic analysis serves as a valuable tool for thoroughly evaluating these options, giving useful insight to decision-makers (Mainardis et al., 2021). LCA is a comprehensive method to assess the environmental impacts throughout the entire life cycle of a product or service, encompassing production, use, and disposal phases. LCA application to WW has been constantly growing in the literature with a special focus on toxicity-related impact categories (Corominas et al., 2020). LCA enables the identification of opportunities for improvement and the evaluation of trade-offs between different environmental aspects (Bui et al., 2022). The usefulness of LCA is apparent both at the planning/design level and for WWTP operations and retrofitting (Corominas et al., 2020); however, still, a lack of standardization emerges in the scientific literature, requiring further effort by researchers toward its wider application.

LCA has been recently applied to quantify the benefits linked to municipal WW reuse for industrial uses, such as cooling towers, through the implementation of tertiary treatment (filtration), showing remarkable environmental advantages due to reduced freshwater consumption (Bui et al., 2022; Pintilie et al., 2016). The importance of tertiary treatment (e.g., ozonation and ozone+hydrogen peroxide) to reduce ecotoxicity in biologically treated municipal effluents has been highlighted by Muñoz et al. (2009) through LCA modeling with the purpose of agricultural effluent reuse. At the same time, a relevant reduction in global warming potential (GWP) has been proved when comparing tertiary-treated WW to desalinated water utilization, suggesting that priority must be given to WW reuse rather than to desalination, whenever possible (Muñoz et al., 2009).

As specifically concerns P&P WW, chemicals and electricity consumption were shown to be the main contributors to the WWTPs' environmental impact (Bui et al., 2022), while the importance of WW reuse has been highlighted by Moosavi et al. (2021). However, no specific study has been found in the scientific literature dealing with LCA application to alternative tertiary treatment technologies of P&P WW, providing useful insights to decision-makers.

Thus, the aim of this study is to compare by using a standardized approach, consisting of LCA coupled with economic analysis, the environmental and economic impact of several alternative scenarios for tertiary P&P WW treatment (inorganic and organic physicochemical coagulation-flocculation; ozonation; ozonation+GAC filtration; UF + RO). The developed model was applied to a P&P WWTP located in Northeastern Italy, which currently employs organic coagulation-flocculation as a tertiary treatment. Previous experimental studies at laboratory and pilot scale, together with full-scale data, were used to build up the model; literature data were considered as well. The coupled environmental and economic assessment is expected to be a useful guidance tool for decision-makers and stakeholders to plan WWTP revamping in the circular economy framework, boosting WW reuse. More generally, this modeling approach can be of interest to all industrial WWTPs that require advanced tertiary treatment solutions, due to the presence of poorly biodegradable compounds in the WW, with the overall aim to minimize both the environmental impacts and the economic burdens.

2. Materials and methods

2.1. Wastewater treatment plant description

The investigated P&P WWTP, located in the Northeast of Italy, has a design capacity of 143,000 population equivalent (PE), and treats three distinct P&P WW lines (condensate, bleaching, and process WW), together with a municipal WW stream. The P&P WW lines contribute together about 128,000 PE to the total plant load, while 15,000 PE are ascribable to the municipal WW. Detailed physicochemical and hydraulic characteristics of each influent WW stream are reported by Mainardis et al. (2022b).

Regarding the treatment scheme, briefly, the WW treatment line (Fig. S1) includes pretreatment (grit removal for municipal WW, up-flow anaerobic sludge blanket -UASB- process for condensate WW), pre-aeration and neutralization (pH correction and mixing of the four WW streams), CAS treatment with secondary sedimentation, tertiary physicochemical treatment of coagulation-flocculation (Mainardis et al., 2022b; Mainardis and Goi, 2019). The sludge line includes aerobic digestion, dewatering through a filter press, and final disposal.

2.2. Tertiary treatment scenarios

The investigated scenarios for tertiary P&P WW treatment include inorganic (S1_ Chem in) and organic (S2_ Chem or) physicochemical treatment with coagulation-flocculation, ozonation (S3_O_3), ozonation followed by GAC filtration (S4_O_3 + GAC), and ultrafiltration followed by reverse osmosis (S5_UF + RO). The scenarios are schematically represented in Fig. 1. Considering that the WW treatment up to the secondary clarifier is the same for all the investigated scenarios, only the tertiary treatment was considered in the LCA and economic analysis.

S1 involves the utilization of aluminum oxide as a chemical agent to remove the residual colloidal organic matter from biological treatment, while S2 forecasts the dosage of formaldehyde and polyacrylamide respectively as coagulating and flocculating agents. The mean pollutant removal obtained from S1 and S2 is 50 % and 70 %, as regards respectively COD and TSS (Mainardis et al., 2022b). Due to the aggregation of colloidal matter and the dosage of chemicals, a significant amount of chemical sludge is produced. A life cycle of 30 yr is considered for the equipment in S1 and S2. The data used for all the successive analyses come from the scientific literature (S1) and full-scale plant data (S2), as detailed in Table S1 in the Supplementary Materials.

In S3, the biologically treated effluent is sent to the ozonation reactor, which partially mineralizes the residual organic matter, showing COD and TSS removals respectively of 60 % and 70 % (Mainardis et al., 2020). Ozone is produced from oxygen in cylinders by using electricity and a stainless-steel ozonation reactor is used (contact time of 40 min). A negligible amount of sludge is generated by this process. A life cycle of 30 yr of the ozonation equipment is considered. All the data referred to in S3 are derived from the pilot experimental campaign reported by Mainardis et al. (2020)) and are detailed in Table S2.

S4 involves coupling ozonation and GAC filtration; the process is capable of enhancing COD and TSS abatement respectively up to 85 % and 90 % (Roudier et al., 2015). The applied ozone dosage is 30 % of that utilized in S3. The energy consumption for GAC regeneration is considered as well, together with the amount of GAC that needs to be used initially to fill the bed and successively to compensate for the losses during regeneration. A life cycle of 30 yr and 10 yr is considered respectively for the ozonation reactor and the GAC filter. The data used for S4 modeling include both experimental (Mainardis et al., 2020, 2022b) and literature data, as detailed in Table S2.

S5 forecasts a tertiary treatment train including UF and RO. The full-scale membrane area to be installed was calculated starting from the mean flux through the membranes (Metcalf, and Eddy, Inc., 2015). In this scenario, higher COD and TSS removals can be obtained (respectively up to 85 % and 100 %), thanks to the excellent membrane retention. A life cycle of 5 yr is supposed for the UF and RO membranes. Besides membrane installation and the energy required for operations, the chemicals used for membrane cleaning were considered as well. All the inventory data are summarized in Table S3.

2.3. Life cycle assessment

The environmental sustainability of the five investigated scenarios (Section 2.2) was assessed using the LCA methodology in accordance with ISO (International Organization for Standardization) requirements (ISO 14040; ISO 14044) (Corominas et al., 2020). The main aim of the LCA was the comparison of the environmental performances of the advanced tertiary treatments (namely ozonation, S3; ozonation+GAC, S4; UF+RO, S5) with those of the conventional inorganic (S1) and organic (S2) coagulation-flocculation, to identify the most sustainable solution. In fact, often an improved effluent quality implies a higher resource consumption.

The most commonly used functional unit (FU) in LCA studies in the WW treatment field is a WW volume (Corominas et al., 2020; Pasciucco et al., 2023), usually expressed as an amount of m^3 of raw WW (Li et al., 2013; Maniakova et al., 2023; Pintilie et al., 2016; Sheikholeslami et al., 2022).

Therefore, following the literature, the FU of the study was defined as 10.68×10^6 m^3/yr, which is the quantity of secondary effluent (with the composition detailed below) annually treated in the investigated WWTP. Considering that the assessed scenarios only differ for the type of tertiary treatment, the upstream WW treatment phases (primary and secondary treatment) were excluded from the analysis, being the same for all scenarios. Consequently, the system boundaries of the study, shown in Fig. 2, were defined to go from the tertiary treatment inlet (i.e., the secondary treatment outlet) until the final discharge/reuse of the treated effluent, including sludge treatment and disposal.

Secondary effluent characteristics show mean COD and TSS

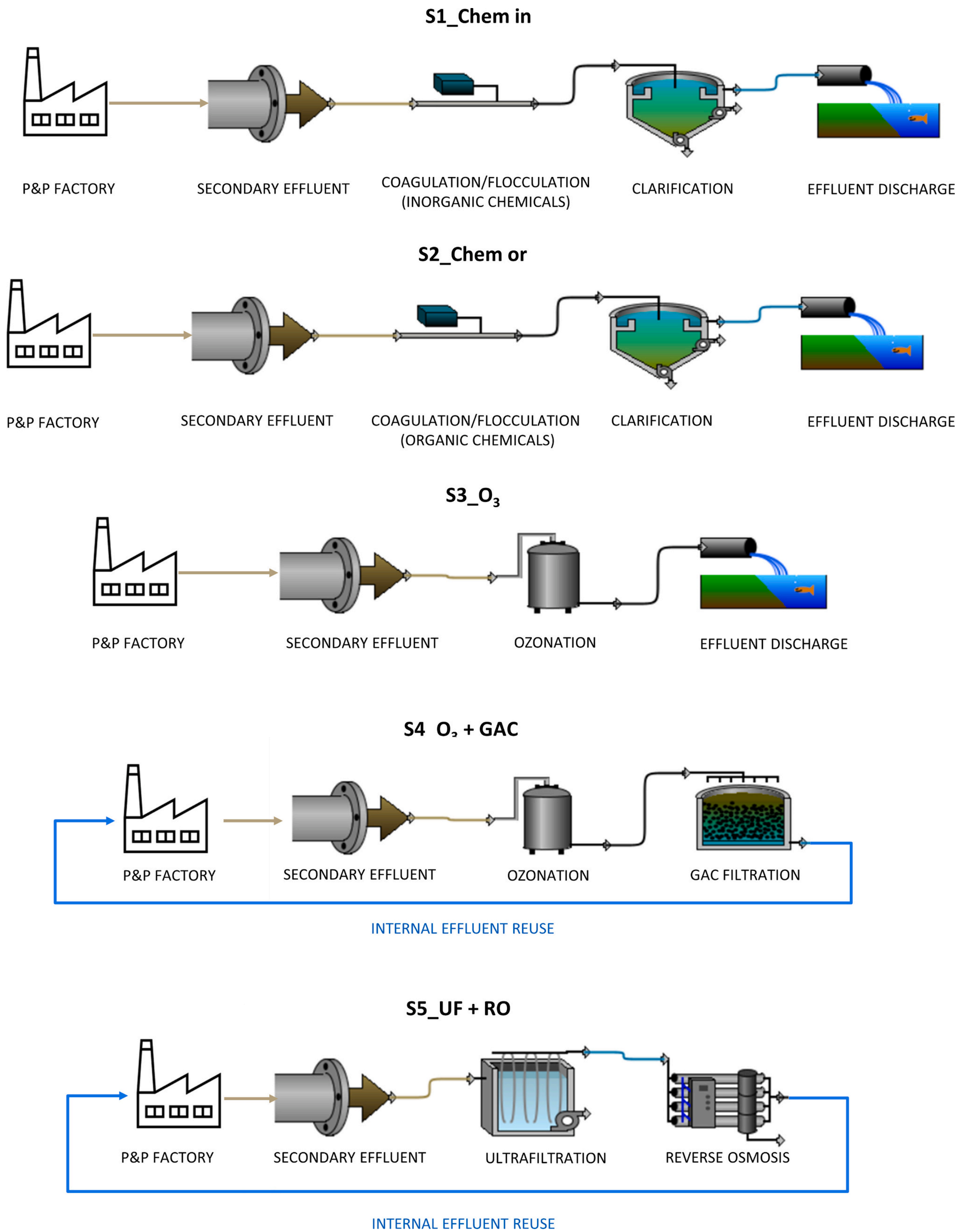

Fig. 1. Schematic representation of the alternative scenarios for tertiary pulp and paper wastewater treatment (S1: inorganic coagulation-flocculation; S2: organic coagulation-flocculation; S3: ozonation; S4: ozonation+granular activated carbon filtration; S5: ultrafiltration+reverse osmosis).

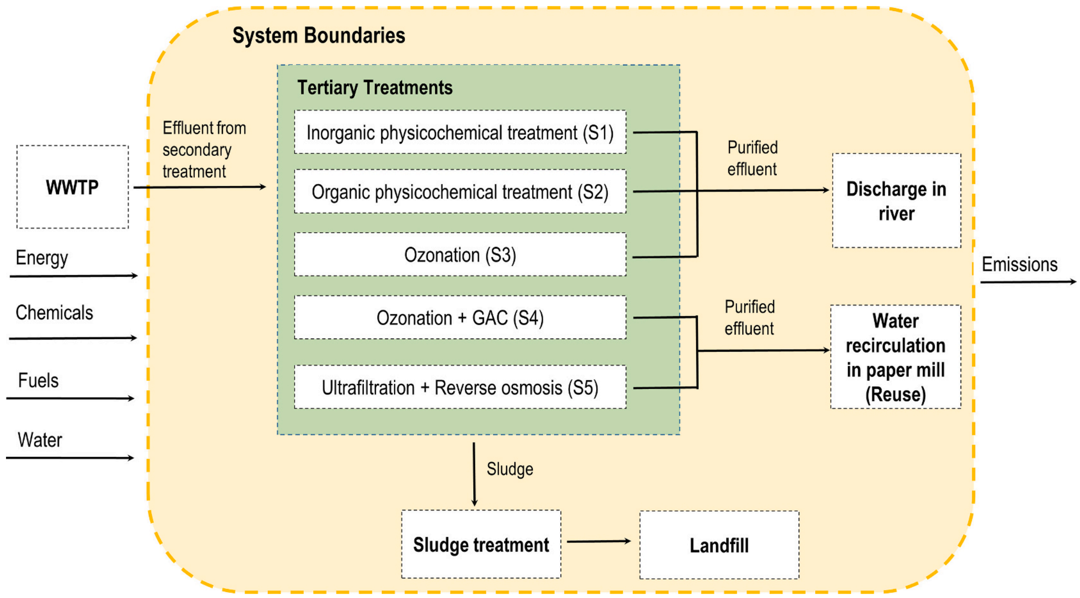

Fig. 2. System boundaries of the study.

concentrations respectively of 143 mg/L and 25 mg/L; nutrients (expressed as total nitrogen, TN, and total phosphorus, TP) are substantially absent due to their low concentration in the influent WW streams, having mean concentrations respectively of 0.48–0.60 mg N/L and 0.22–0.40 mg P/L. A limited variability of secondary effluent strength is observed throughout the year, due to the holiday periods of the P&P factory and some episodes of unwanted biomass entrainment from the secondary clarifiers (Mainardis et al., 2022b).

More in detail, for each scenario the construction and operational phases were considered, including i) production and transportation of chemicals and energy consumed during tertiary WW treatment and maintenance operations; ii) production of capital goods (infrastructure and equipment); iii) sludge treatment and subsequent transportation and landfilling; iv) pollutant emissions in air and water. Regarding the final destination of the treated effluent, for scenarios S1, S2, and S3 its discharge in the downstream river was assumed, while, for scenarios S4 and S5, the internal reuse of the purified effluent was assumed through its recirculation in the P&P mill, thanks to the higher pollutant removal.

The modeling of the developed scenarios (Fig. 1) was carried out using SimaPro 9 software (Pre Consultants, Amersfoort, The Netherlands). The processes included in Ecoinvent v.3 databases were the main source of the background data (infrastructure and equipment, vehicles, Italian energy mix, extraction and processing of raw materials and fuels). The primary inventory data (chemicals and energy consumption and equipment maintenance, pollutant emissions, sludge production and treatment, and subsequent transportation and landfilling), were collected through interviews with experts, laboratory analysis, and analytical calculations, as well as from relevant literature studies. The inventory data are reported in Table S1- S3 of the Supplementary Materials for each modeled scenario.

Infrastructure and equipment were modeled by adopting data from Ecoinvent database processes, considering a lifetime of 30 yr for ozonizers and GAC filter structure, 10 yr for GAC, and 5 yr for UF and RO membrane modules.

Water reuse through purified effluent recirculation in the P&P mill, forecast in scenarios S4 and S5, allows for avoiding water consumption produced by an alternative source, which was modeled as tap water production in Italy by using data provided by the processes of Ecoinvent v.3 databases.

In accordance with other LCA studies on WW treatment (Anastasopoulou et al., 2018; Arzate et al., 2019; Carré et al., 2017), the environmental impacts of all scenarios were assessed using the ReCiPe 2016 evaluation method with hierarchist perspective (H), adopting both midpoint and endpoint levels (Huijbregts et al., 2017). ReCiPe 2016 is one of the most used evaluation methods in the WW treatment sector thanks to the high number of impact categories that can be considered (Corominas et al., 2020). The midpoint level (problem-oriented approach) of ReCiPe 2016 contains 18 impact categories: Global Warming Potential (GWP); Stratospheric Ozone Depletion (SOD); Ionizing Radiation (IR); Ozone Formation- Human Health (OF-HH); Fine Particulate Matter Formation (FPMF); Ozone Formation- Terrestrial Ecosystems (OF-TE); Terrestrial Acidification (TA); Freshwater Eutrophication (FE); Marine Eutrophication (ME); Terrestrial Ecotoxicity (TEcotox); Freshwater Ecotoxicity (FEcotox); Marine Ecotoxicity (MEcotox); Human Carcinogenic Toxicity (HCTox); Human non Carcinogenic Toxicity (HnCTox); Land Use (LU); Mineral Resource Scarcity (MRS); Fossil Resource Scarcity (FRS); Water Consumption (WC). The endpoint level (damage-oriented approach), instead, considers three macro-categories: damage to human health, damage to ecosystems, and resource consumption.

2.4. Economic analysis

The input data used for the economic assessment, including specific data sources, are reported in Table S4. The operativity of all the proposed tertiary treatment technologies (320 d/yr) forecasts a yearly 40-day period for maintenance. The chemical sludge disposal cost corresponds to current landfill disposal fees. The capital cost of physicochemical treatment (S1- S2), despite being reported in Table S4, is nil in the specific case-study due to the fact that it is the currently installed solution in the investigated WWTP.

Considering that the P&P factory has its own pumping wells for freshwater, the economic value of the treated effluent in case of internal reuse (S4- S5) was estimated by assigning to this stream a marginal economic value (equal to the agricultural water cost).

2.5. Sensitivity analysis

Due to the uncertainty in selected input parameters having a significant impact on the environmental and economic assessment, a sensitivity analysis was later performed by varying i) the chemicals' dosage used for the inorganic and organic physicochemical treatment

(S1 and S2), ii) the percent of treated effluent reuse for scenarios where reuse was modeled (S4 and S5), and iii) the electricity cost. According to the existing literature, the estimated range of chemicals dosage was 33.5–335 mg/L and 10–100 mg/L respectively for inorganic (S1) and organic (S2) physicochemical treatment (Ahmad et al., 2008; Wang et al., 2011; Wong et al., 2006). The chemical sludge production was recalculated for each dosage starting from the baseline scenario, considering an increase in TSS abatement, according to the specific chemical dosage.

As for effluent reuse percent (S4 and S5), the hypothesized purified effluent range was 70–95 % of the secondary WW effluent (Pizzichini et al., 2005; Rathoure, 2020; Yang et al., 2021). Finally, minimum and maximum electricity costs for industrial users with a yearly consumption of 500–2000 MWh/yr in the period 2014–2022 were considered for the sensitivity analysis on electricity prices (ARERA, 2023; EUROSTAT, 2023), given the significant fluctuation in this important parameter due to market volatility and the current geopolitical situation.

The environmental and economic impact assessment was calculated for each of these situations by running again the LCA and economic models under the modified operating conditions, and the results are presented in Section 3.3.

3. Results and discussion

3.1. Life cycle assessment

A comparison between the environmental performances of the five treatment scenarios adopting the endpoint categories of the ReCiPe 2016 H method is depicted in Fig. S2 of the Supplementary Materials, while Fig. 3 summarizes the results related to the mid-point impact categories.

Overall, the best environmental performances were obtained by S4 followed by S5, where a higher energy and resources consumption was compensated by the achievement of a high-quality purified effluent that could be internally reused in the P&P factory. The negative values observed in S4 and S5 in terms of Human health and Ecosystems endpoint categories were due to the significant environmental benefits given by water reuse that allows for avoiding tap water consumption. However, adopting the Resources endpoint category, S5 showed worse environmental performances than all the other alternatives except S3, due to the high energy consumption for UF and RO membrane operations.

The ozonation scenario (S3) was the worst treatment solution in environmental terms because the purified effluent could not be reused, showing a comparable effluent quality to that obtained from physicochemical treatment (Mainardis et al., 2020). In S3, a higher treatment level causes a higher impact without pursuing significant environmental benefits. Comparing the two physicochemical treatment scenarios (S1 and S2), the results highlight that S2 generated lower environmental burdens than S1, mainly due to the lower chemicals' dosage.

By focusing on the mid-point impact categories (Fig. 3), in most impact categories (10 out of 18) the highest impact is obtained in S3, while S1 shows the worst environmental performances in 7 impact categories, and S5 is the major contribution only concerning the "stratospheric ozone depletion" category (Fig. 3). Overall, S4 clearly appears to be the most favorable solution from an environmental perspective.

To better determine the key aspects that affect the environmental sustainability of the alternative treatment scenarios, Fig. 4 shows, for each scenario, the contribution provided by the main hotspots to the total impacts by considering the most relevant midpoint impact categories of ReCiPe 2016 H.

The first aspect that clearly emerges is the significant role of energy influencing the performances of the advanced treatment scenarios (S3-S5), especially in terms of GWP, particulate matter formation, and fossil resource scarcity. An improved effluent quality, indeed, can be obtained only with higher energy and resource consumption. However, for S4 energy influence is lower than for S5, because the presence of GAC filters allows for achieving an improved effluent quality with a significantly lower ozone dosage than S3, implying a lower energy consumption. S4 emerges as the most environmentally sustainable treatment option: the high negative impact (i.e., environmental benefit) given by water reuse provides the greatest contribution to the total impact for all midpoint categories (except freshwater eutrophication). Thus, water reuse is a sensitive parameter that significantly affects the results of the study. Recovery of 90 % of the treated effluent as permeate was initially

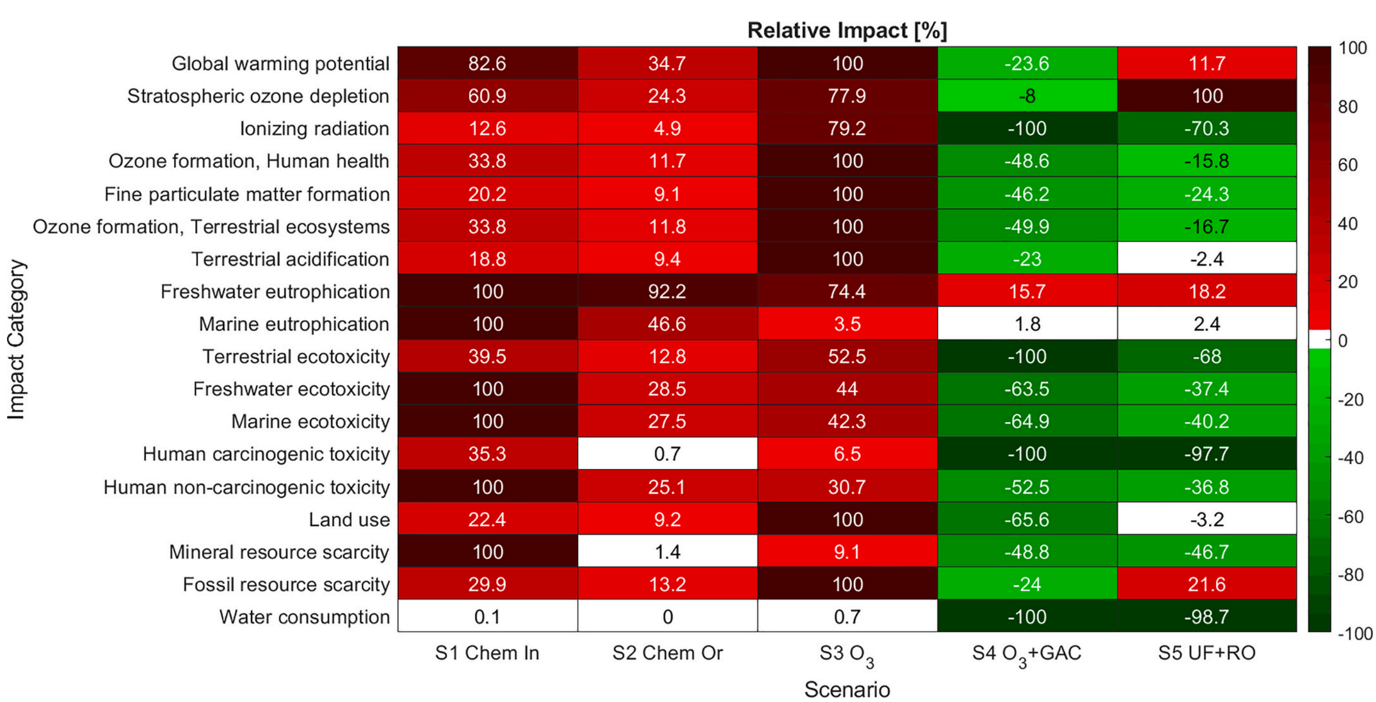

Fig. 3. Environmental comparison of the five investigated treatment scenarios with the midpoint approach of the ReCiPe 2016 H method.

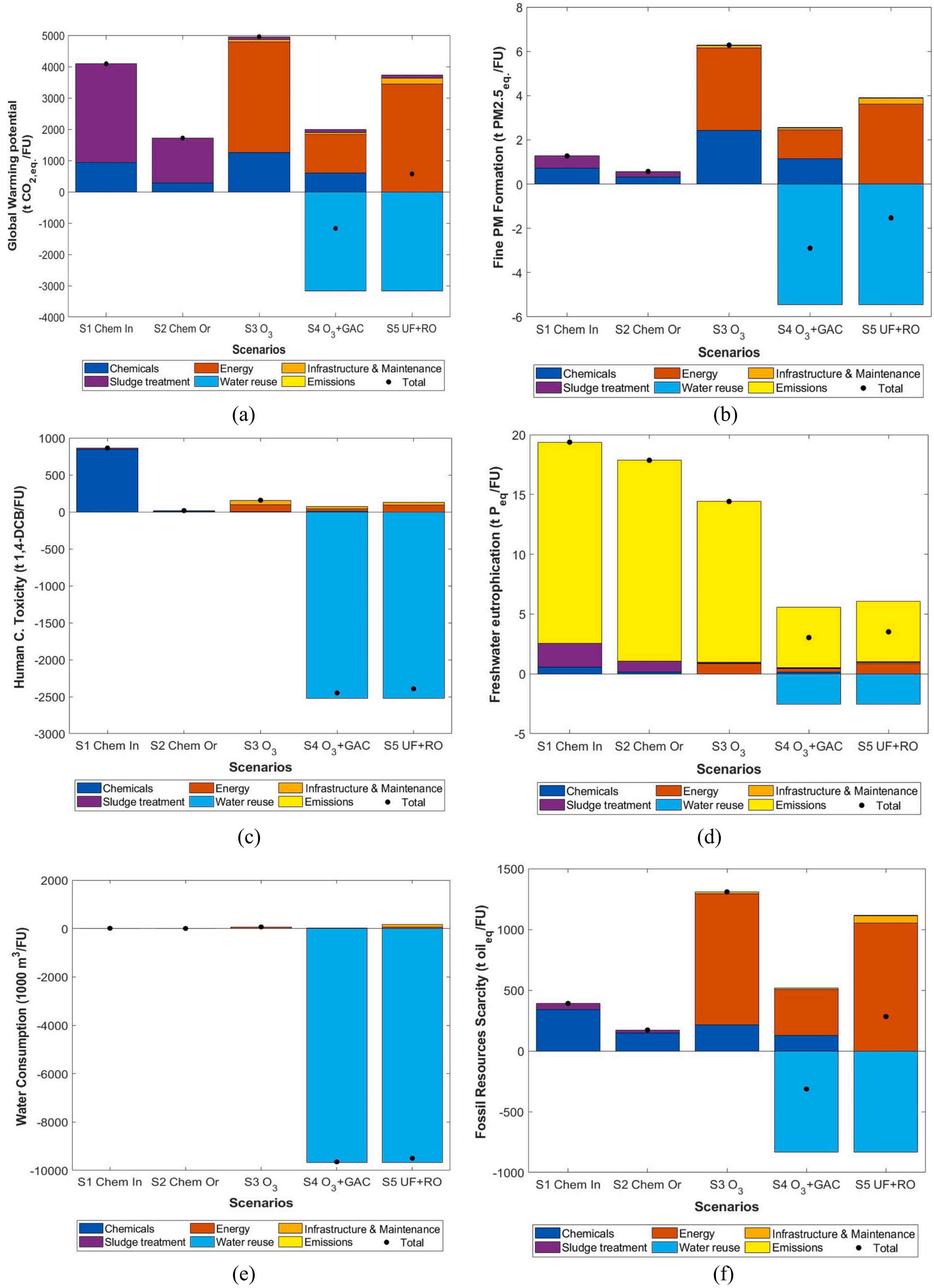

Fig. 4. Impact contributions of the main hotspots to the total life cycle impacts of each investigated treatment scenario. The impacts were estimated with the following impact categories of ReCiPe 2016 H midpoint level: Global warming potential (a); Fine particulate matter formation (b); Human carcinogenic toxicity (c); Freshwater eutrophication (d); Water consumption (e) and Fossil resources scarcity (f).

supposed, however, a high uncertainty was observed in this parameter, thus a sensitivity analysis was later performed varying the effluent reuse in a range of 70–95 % (Section 3.3). Although S1-S3 generally show compatible characteristics for agricultural effluent reuse, according to the recent EU Directive 741/2020 (Mainardis et al., 2022a), no reuse was forecast due to the following considerations: i) the residual persistent pollutants present in the P&P effluent may accumulate in the receiving soils and irrigated crops, altering their natural equilibrium (Al-Hazmi et al., 2023) and resulting in unintended cross-contamination and human-health risk (Penserini et al., 2023); ii) agricultural reuse is particularly adapt to effluents with residual nutrient concentrations, to couple water and nutrient recovery through fertigation practices (Khan et al., 2022; Mainardis et al., 2022a), while P&P effluents include no valuable nutrients (Mainardis et al., 2022b); iii) the selected WWTP is not located in an agricultural-intensive area (Mainardis et al., 2022b); iv) alternative water sources, e.g., treated municipal effluents or freshwater, should be preferred (whenever possible) to industrial effluents for agricultural reuse.

Analyzing the impacts in terms of freshwater eutrophication, pollutant emission in water is the main contributor to the total impact for all investigated scenarios, due to the residual COD in the purified effluent. Therefore, physicochemical treatments (S1 and S2), which show a lower COD removal than advanced solutions (S4 and S5), obviously generate higher impacts, while the scenarios characterized by a higher treatment level are responsible for lower impacts, due to their higher COD removal (up to 85 % for S4 and S5).

For most impact categories, another important hotspot is chemicals' production and consumption. For the physicochemical scenarios, such impacts were due to the inorganic (S1) or organic (S2) chemicals needed for coagulation-flocculation. In terms of human toxicity, chemicals' consumption provides the highest contribution to the total impact of the inorganic physicochemical treatment (S1), mainly because of the impact generated by aluminum oxide production.

However, also for S3 and S4 chemicals' consumption plays a significant role; for these scenarios, ozone generation and GAC production are the main ones responsible for the impacts.

Focusing on the impacts in terms of GWP, sludge treatment and landfill disposal is the aspect that mostly affects the environmental performances of the two physicochemical scenarios. This occurs because of the higher amounts of sludge produced during these treatments (especially in S1) compared to the other three alternative scenarios. The higher chemicals' dosage applied in S1 generates greater sludge amounts than S2, where the chemicals' dosage is continuously tailored on-site to reduce both chemicals' consumption and sludge generation. Ozonation has the advantage of significantly lowering the amounts of generated sludge, leading to lower transportation and disposal costs. Also, membrane treatment generates lower sludge amounts than S1 and S2.

A comparison with the scientific literature can be fruitful, as in the last 20 years a relevant interest has been dedicated to LCA application to WW treatment (Corominas et al., 2020). The importance of conducting pilot-scale studies to get representative results for the LCA modeling was highlighted by Carré et al. (2017), who also showed that low energy intensity processes (e.g., sand filtration + ultraviolet- UV- disinfection, or UF alone) generate lower environmental impacts than energy-intensive processes (e.g., UF coupled with UV disinfection). Consistently, in the present study, most of the inventory data were obtained from full-scale, pilot-scale, or laboratory data referred to the specific P&P WW composition (Table S1-S3) to provide robust indications to decision-makers.

The preponderant effect of electricity consumption on the environmental impacts in tertiary WW treatment was highlighted again by Akhoundi and Nazif (2020), coherently with what is shown in Fig. 4 for energy-intensive processes, including ozonation and RO. Innovative treatment solutions, such as solar photo-Fenton, which are currently being tested at the pilot scale, still show higher environmental burdens than the technologies studied in the present work (e.g., ozonation), due to the necessity of pH adjustment, effluent storage, and chemicals' consumption (Arzate et al., 2019). Source separation of the different P&P effluents may enhance the applicability of these advanced treatments, by applying a dedicated treatment technology to each stream according to its biodegradability and peculiar physicochemical characteristics, treating only a fraction of the overall flowrate (Esmaeeli et al., 2023).

Furthermore, advanced treatment solutions (including membranes and advanced oxidation processes) show the general feature that the environmental impacts generated by the infrastructure are much lower than those given by energy and chemicals' consumption during operations (Arzate et al., 2019), as clearly emerges from Fig. 4. A relatively higher impact of the infrastructure is observed in S5, due to the limited membranes lifetime, when compared to ozonation reactor (S3 and S4) and GAC filters (S4).

3.2. Economic analysis

The results of the economic analysis are summarized in Table 1. A substantial overlap between the environmental and economic outcomes emerges: S4 appears again as the most favorable solution, thanks to the internal P&P WW reuse, with total yearly expenses of about 500 k€/yr. However, a significant capital cost (8.0 M€) has to be initially sustained. S2 comes next, with significantly better economic performances than S1, due to the lower applied chemicals' dosage, which is coupled with reduced chemical sludge production. S3 is penalized by the high energy consumption, while the relatively negative performances of the membrane-based scenario (S5) are linked to the significant electricity requirement, especially if compared to S4. As for S5, the internal reuse is not sufficient to obtain an overall economic balance more favorable than physicochemical treatment (S1 and S2). In addition, the estimated capital costs (10.9 M€) are higher than those forecast for S4. The capital cost of physicochemical treatment (S1-S2), instead, is comparable to that of ozonation (S3), however, in the specific case-study it can be considered nil due to the fact that the process is already in operation.

As regards coagulation-flocculation, recent literature studies showed that the main impact on operating costs is linked to chemicals' usage (Wang et al., 2018). Significant fluctuations in operating costs can arise due to specifically applied dosages (e.g., between S1 and S2): normally, bench-scale tests are performed at each WWTP location to select the best-performing coagulant and flocculant agents (Chen and Horan, 1998), optimizing the overall process efficiency and reducing the related economic burdens. Current market uncertainty and chemicals' price fluctuations may negatively affect the future economic balance of coagulation-flocculation. Finally, the high disposal cost of the generated chemical sludge is another known limit of this technology (Mainardis et al., 2020). Thus, alternative solutions with reduced chemicals' consumption (e.g., O_3 + GAC and UF + RO) may further enhance their competitiveness in the near future. On the other hand, the advantages of

Table 1
Results of the economic analysis for each scenario.

Scenario	Capital cost (M€)	Lifetime (yr)	Costs (M€/yr)	Revenues (M€/yr)	Yearly economic balance (M€/yr)
S1_Chem In	1.8	30	0.803	0	−0.803
S2_Chem Or	1.8	30	0.598	0	−0.598
S3_O_3	2.0	30	2.469	0	−2.469
S4_O_3 + GAC	8.0	30 (O_3 reactor) 10 (GAC filter)	1.357	0.843	−0.514
S5_UF + RO	10.9	5	1.876	0.843	−1.033

coagulation-flocculation are linked to its maturity, flexibility, and efficiency (Mainardis et al., 2022b).

As concerns the proposed technological alternatives for tertiary P&P WW treatment, it was recently demonstrated that the activated carbon process outperforms from an economic standpoint both ozonation and membrane filtration when spent activated carbon is regenerated (Peyrelasse et al., 2021), coherently with the present outcomes. Furthermore, the critical aspects of O_3 and membrane filtration are shown to be respectively energy consumption and retentate management.

Combined processes often lead to an improvement both in pollutant removal efficiency and economic balance, as shown in the present study when comparing O_3 alone (S3) and O_3 + GAC (S4). Accordingly, O_3 and UF combination has been recently demonstrated as an effective post-treatment of biologically treated effluents, enhancing their reuse feasibility with possible economic valorization (Clem and Mendonça, 2022).

As concerns membrane-based processes, it was shown that UF and RO combination as tertiary WW treatment can be economically sustainable if treated effluents are reused in industrial processes, as advanced treatment costs are comparable to current freshwater tariffs (Pérez et al., 2022), thus the economic valorization of the treated effluents appears as a key for the profitability of UF + RO process. Again, the importance of the end-use destination of effluents treated through RO has been highlighted by Kehrein et al. (2021), when comparing irrigation, potable, and industrial reuse: agricultural reuse is not cost-effective, due to the low water tariff, while industrial reuse can lead to a superior income. Considering the specific case study, the high-quality water obtained from RO in S5 may be used not only for internal P&P reuse but also for other industrial processes (e.g., automotive sector), as the WWTP is located in an industrial-intensive area. Selling the treated effluent to local industries at current market prices (about 1.0 €/m^3) may further improve the economic balance of S5, leading to significantly higher profits (Mainardis et al., 2022b).

3.3. Sensitivity analysis

Given that LCA models are built on huge amounts of data coming from different sources with various precision, a significant uncertainty can be observed in the results, which can affect the final ranking of the different treatment solutions (Sheikholeslami et al., 2022). Thus, in this work, the effect of variable chemicals' dosage (S1 and S2), effluent flowrate for reuse (S4 and S5) and electricity cost, which are affected by a high uncertainty level (connected to their dependency on specific case-study and market conditions), was later analyzed. The environmental and economic aspects, respectively investigated through LCA and economic analysis, were both analyzed. The obtained results are summarized in Fig. 5.

Regarding the sensitivity on chemicals' dosage (S1-S2), in the most

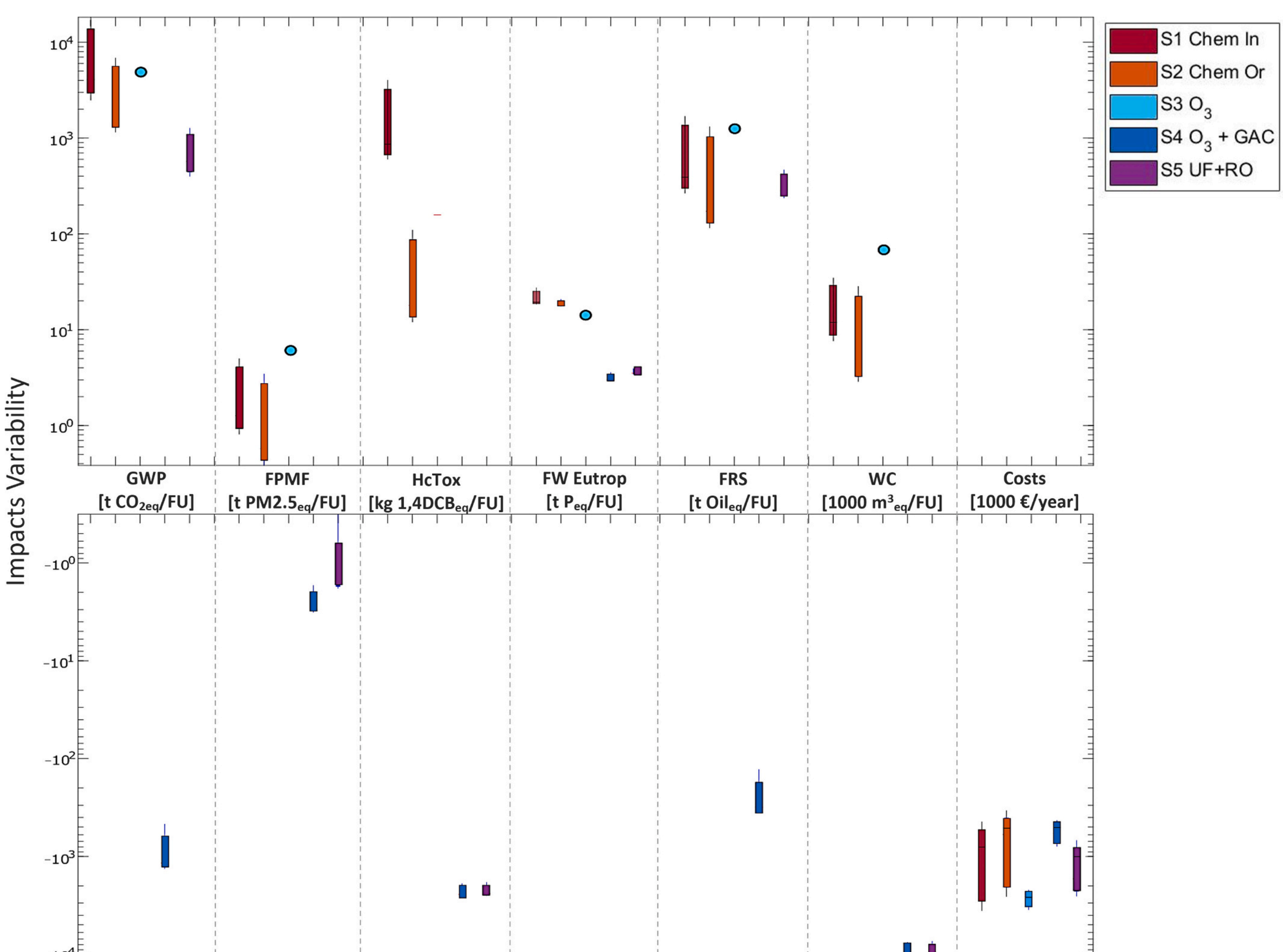

Fig. 5. Boxplot of the sensitivity analysis results on the following impact categories of ReCiPe 2016 H midpoint level: Global warming potential (GWP); Fine particulate matter formation (FPMF); Human carcinogenic toxicity (HcTox); Freshwater eutrophication (FW Eutrop); Fossil resources scarcity (FRS); Water consumption (WC) and Economic Balance (Costs).

unfavorable cases (highest chemical dosage), both S1 and S2 appear to be the least preferable solutions, even when compared to the most energy-intensive scenario (S3, ozonation alone). On the other hand, the minimum chemicals' dosage (i.e., most favorable case) allows for significantly reducing the environmental impacts of both S1 and S2, yielding an economic balance comparable to S4. Thus, the importance of properly tailoring chemicals' dosage appears again as a fundamental aspect to get favorable environmental and economic indicators when applying coagulation-flocculation as a tertiary treatment.

As for the amount of purified effluent for reuse, despite having a moderate impact both on the environmental and economic outputs (Fig. 5), the tested range does not modify the final ranking of the scenarios, which always forecasts S4 as the preferred alternative. As regards electricity cost, instead, the minimum cost makes S5 more convenient than S1, leaving S4 and S2 respectively as the first and second choice, while the highest electricity cost makes S2 and S1 the preferred solutions overall, due to their negligible energy consumption. It should be noticed that while the minimum energy cost is very similar to the mean value (0.17 versus 0.20 €/kWh), the higher cost (0.36 €/kWh) is significantly higher than the mean value, due to the exponential surge in energy prices recently observed on the market.

In the literature, LCA has been shown to be an effective decision-support tool when comparing multiple scenarios for advanced WWTP design (Baskurt et al., 2017), and the present study confirmed its suitability to rank alternative treatment solutions for tertiary P&P WW remediation. A multi-criteria decision-making approach was recently proposed to tackle conflicting objectives and different uncertainties to prioritize WW reuse (Akhoundi and Nazif, 2018). After identifying the potential alternatives, several criteria were quantified, including technological, environmental, economic, and cultural aspects. The alternatives were finally ranked through recursive algorithms that aggregated multiple criteria (Akhoundi and Nazif, 2018).

The conducted environmental and economic assessment proves that coagulation-flocculation is still the most convenient full-scale alternative as tertiary P&P WW treatment, despite its well-known drawbacks (chemicals' consumption, sludge generation, impossibility of directly reusing the effluent). The coupling of innovative solutions, such as ozonation and GAC, appears more promising than single technologies (e.g., ozonation alone) as a retrofit of existing P&P tertiary treatment plants, especially in case the effluent can be properly monetized or dedicated incentives are provided to water utilities, leading to a real circular economy implementation. Further amelioration of effluent characteristics, e.g., through RO, appears technically and economically feasible only in the case the effluent can be sold at a higher price to industrial users that require highly pure water (e.g., the automotive sector). Effluent reuse will become mandatory in the near future, due to the reduced availability of conventional water sources, linked to accelerating climate change, thus the establishment of local circular economies will be imperative; this study demonstrated that thorough techno-economic and environmental assessments are required for conscious decision-making. Further studies on the topic may include, besides environmental and economic impacts, the assessment of the social aspects to provide a complete framework for sustainable WW treatment (Padilla-Rivera and Güereca, 2019), focused on virtuous reuse practices.

4. Conclusions

This study compared alternative tertiary treatment solutions for P&P WW (S1 - inorganic coagulation-flocculation, S2 - organic coagulation-flocculation, S3 - ozonation, S4 - O_3 + GAC, S5 - UF + RO) through LCA and economic analysis, providing valuable insights for decision-makers regarding the environmental and economic impact of the different technologies. The findings revealed that S4 (O_3 + GAC) exhibited the lowest environmental impact across most end-point and mid-point categories, outperforming both S5 (UF + RO) and coagulation-flocculation (S2 and S1). The reduced ozone dosage in S4 compared to S3 (O_3 alone) resulted in a significant decrease in electricity demand, successfully addressing the major ozonation drawback. Physicochemical treatment remained a favorable option when reuse was not considered, owing to its substantial environmental benefits.

The economic assessment demonstrated that O_3 + GAC was economically more convenient than coagulation-flocculation when the treated effluent was internally reused, despite significant initial capital costs, while membranes (UF + RO) remained relatively costlier. The economic valorization of the treated effluent could be further enhanced by selling it to local industries, leading to net profits for both S4 and S5, leading to a rapid recovery of the initial investment. The sensitivity analysis highlighted substantial variability in the environmental and economic impacts of S1 and S2, emphasizing the critical importance of tailoring chemicals' dosage to optimize the treatment process. Conversely, the variability was lower for the amount of treated water for internal reuse in S4 and S5. Electricity cost was shown to have a strong influence too, with advanced treatment scenarios (S3-S5) being severely penalized by the high electricity prices recently observed on the market due to the current geopolitical situation.

Future studies should delve into social aspects, beyond environmental and economic impacts, by analyzing stakeholders' acceptance of the proposed reuse scheme. Additionally, establishing fruitful collaborations between P&P factories, water utilities, public authorities, and local industries would promote a sustainable approach in the sector. Giving proper information to all involved stakeholders will be fundamental to boosting circular economy implementation in the P&P sector, providing relevant economic and environmental benefits, as shown in the present study.

CRediT authorship contribution statement

Matia Mainardis: Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Carmen Ferrara:** Conceptualization, Methodology, Formal analysis, Investigation, Software, Validation, Writing – original draft, Visualization. **Beatrice Cantoni:** Conceptualization, Investigation, Data curation, Writing – review & editing, Visualization. **Camilla Di Marcantonio:** Conceptualization, Investigation, Data curation, Writing – original draft, Visualization. **Giovanni De Feo:** Software, Writing – review & editing. **Daniele Goi:** Resources.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.167598.

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