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Sustainable water purification: life cycle trade-offs in carbon-based catalyst design

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This study evaluates the environmental performance of carbon-based macrostructured catalysts developed for advanced drinking water treatment applications. A novel two-stage catalytic system, comprising ozonation and hydrogenation, was assessed using the Life Cycle Assessment (LCA) methodology, considering three synthesis approaches: traditional Chemical Vapor Deposition (CVD), and two washcoating methods using Triton X-100 or sodium alginate as dispersants. While all catalysts achieved similar pollutant removal efficiencies, LCA results revealed significantly lower environmental impacts for washcoated catalysts compared to CVD, with the TX-100 method showing the lowest total burden. Key impact contributors included energy-intensive thermal treatments and raw material choices, particularly palladium for the synthesis of bimetallic catalysts. This work represents the first LCA applied to an integrated water treatment process focused on heterogeneous catalysis, as well as the first comparative LCA of different methodologies for the synthesis of macrostructured catalysts. The combination of LCA with experimental lab-scale synthesis data and the Safe-and-Sustainable-by-Design (SSbD) framework makes this study unique and among the first to adopt such an approach in this research area. The analysis highlights opportunities to optimise synthesis protocols for improved sustainability and supports the integration of SSbD principles in water treatment technologies.

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1. This work presents the first life cycle assessment of an innovative water treatment technology for the simultaneous removal of organic and inorganic contaminants. The system showed high efficiency, in removing both classes of contaminants, with relevance for the conversion and mitigation of bromate formed during the ozonation step of the integrated treatment. The system improves sustainability by using catalysts immobilized on macroscopic supports, preventing powder release and avoiding mass transfer limitations.
2. An LCA compared macrostructured catalysts differing only in preparation. Washcoating significantly reduced life cycle impacts *versus* chemical vapor deposition while maintaining similar performance, lowering total impact by ~30–50% per liter of treated water.
3. To further enhance the sustainability of the system, future studies should focus on optimizing the thermal treatment steps applied during catalyst synthesis, as these steps were identified as the main contributors to the overall environmental impact of the process.

1. Introduction

Freshwater contamination remains a pressing issue and still lacks an effective solution due to the complexity related to the water matrices and the emergence of increasingly resistant compounds that are immune to the currently applied treatment systems. The urgent need for innovative water treatment strategies has propelled the development of heterogeneous catalysis to the forefront as a promising approach for the effective degradation or transformation of diverse classes of pollutants.[1,2]

In the context of applying such systems for water treatment, an essential step is immobilising the powder catalysts into robust macrostructured frameworks (to ensure stable process performance while mitigating the risk related to free nanomaterials in water).[3,4]

Carbon and/or metal-based catalysts immobilised on macroscopic supports have been applied in a wide range of heterogeneous catalytic processes.[5–8] Recent studies have highlighted that the catalytic performance of carbon-based materials in advanced oxidation processes is strongly influenced by electron-transfer mechanisms occurring at carbon–metal–oxidant interfaces. Structural features of carbon materials, such as defect density, heteroatom doping and confined architectures, can steer radical and non-radical pathways, thereby influencing catalytic selectivity and stability. These

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characteristics may also contribute to suppressing metal leaching and reducing the formation of secondary pollutants during water treatment processes.[9]

Generally, the most conventional methodology for the synthesis of carbon-coated supported catalysts is through chemical vapour deposition (CVD), which enables the direct growth of carbon nanofibers in the supported structure, allowing the synthesis of highly stable, resistant and active macrostructured catalysts.[10–13] These catalysts have proven efficient in different heterogeneous catalysis reactions;[13–15] however, the greatest disadvantage of this methodology is the impossibility of taking advantage of most mechanical/chemical modifications performed in powder catalysts (which are usually associated with improved results).[4,9,16]

The synthesis of macrostructured catalysts through the washcoating procedure allowed overcoming many of the disadvantages associated with CVD process, by performing the dispersion of pre-formed catalysts in an aqueous solution with the aid of adequate dispersants.[16–21] Additionally, in what concerns mono/bimetallic catalysts synthesis, the most conventional thermochemical processes for catalysts synthesis are impregnation/co-impregnation and precipitation/co-precipitation, which are known to be highly energetically dependent phases involving thermal treatments and metal precursor utilisation.[22]

The development of novel macrostructure catalyst systems presents both significant challenges and promising benefits. However, their environmental performance and sustainability implications must be thoroughly investigated to ensure the advancement of eco-friendly technologies, considering their entire life cycle. The synthesis of these catalysts introduces several challenges, including potential environmental impacts and economic considerations,[23] which require careful evaluation to optimise their viability and minimise adverse effects.

One significant issue is the use of rare or toxic raw materials, which can lead to mineral resource depletion and environmental contamination during extraction and processing of raw materials.[24,25] Additionally, the energy-intensive nature of catalyst synthesis methods, such as high calcination temperatures or complex chemical treatments, can contribute to greenhouse gas emissions and high operational costs.[24,26–28] Additionally, the production process may generate hazardous by-products that require proper handling and disposal, further increasing the environmental footprint.[28] As a result, developing sustainable synthesis routes, such as employing green chemistry principles and renewable materials, is crucial to mitigate these impacts and ensure that catalytic processes remain economically viable and environmentally friendly. In addition, the Safe and Sustainable by Design (SSbD) framework, developed by the European Commission, promotes the design of chemicals and materials that are safe, functional, and environmentally sustainable across their life cycle,[29] the last aspect of this approach being addressed by the globally accepted Life Cycle Assessment (LCA) methodology. This framework has been addressed in recent works, like the recent one, which has emphasized that the sustainability of carbon-based materials used in environmental remediation should be evaluated within circular treatment frameworks that integrate pollutant capture, regeneration, and end-of-life management. Such approaches highlight the importance of life-cycle assessment and system-level evaluation to properly assess the environmental implications of carbon-based technologies for water treatment.[30]

In this vein, to ensure the sustainability of these advanced materials, this study conducts an LCA to evaluate the environmental performance of these novel catalysts across their life-cycle, from raw material extraction to end-of-life management. This research aims to optimise sustainable catalyst design from an environmental perspective to minimise toxicity and reduce resource consumption, thereby contributing to the development of eco-friendly water treatment technologies. The ISO 14040/44:2006[31,32] guidelines support the LCA methodology for conducting a thorough assessment of the environmental effects of products, processes or services across their entire life cycle: from the raw material extraction, production/manufacturing, transportation/distribution, use phase and End-of-Life (EoL).[31,33,34]

In this study, the life cycle assessment (LCA) methodology was applied to evaluate and compare the performance of a novel catalytic technology for surface water treatment intended for human consumption, considering different structured catalyst systems. The technology involves the sequential application of catalytic ozonation and catalytic reduction to simultaneously degrade and convert organic and inorganic pollutants in water, employing immobilised catalysts in macroscopic frameworks to ensure efficiency and operational safety. Experimental trials using surface water have demonstrated promising results, with the effective degradation of organic micropollutants while maintaining bromate (BrO_3^-) and nitrate (NO_3^-) concentrations within regulatory limits for drinking water. Although still at an early developmental stage (laboratory scale), this work represents the first LCA applied to an integrated water treatment process focused on heterogeneous catalysis, as well as the first comparative LCA of different methodologies for the synthesis of macrostructured catalysts. The combination of LCA with experimental lab-scale synthesis data and the Safe-and-Sustainable-by-Design (SSbD) framework makes this study unique and among the first to adopt such an approach in this research area. The LCA aims to identify environmental hotspots across the life cycle stages of the technology (from raw material extraction, system construction, and catalyst synthesis to the operation of the treatment system for drinking water production), providing valuable insights to guide future research, improve system design, and support optimisation during scale-up to larger applications.

2. Methodology

2.1 Macrostructured catalysts synthesis

Cordierite honeycomb monoliths (1 cm diameter, 6 cm length and with 100 channels per square inch (cpsi)) from Corning were used as a macroscopic framework for catalyst immobilisation.

For the macrostructured catalysts synthesis, three different methodologies were studied and are acutely described in previous works.[16,21,35–37] The methodologies employed include one based on the conventional approach commonly used for the synthesis of macrostructured catalysts (CVD), and two others focused on the application of the washcoating technique (WCP). The latter were developed to improve the design of macrostructured catalysts while addressing common issues typically associated with the CVD method. A brief description of each synthesis method is provided below.

2.1.1 Powder catalysts synthesis. Firstly, the powder catalysts (intended to be immobilised) were synthesised. Commercial multiwalled carbon nanotubes (CNT, Nanocyl 3100) were used as starting material and, also, for the synthesis of composite CNT:TiO_2 (commercial TiO_2, Evonik Degussa Ibérica SA), catalysts to be immobilised in the macrostructured catalysts applied under the scope of the present work.

Modification of commercial CNT was performed by ball-milling (Retsch MM200) using a frequency of 15 Hz for 4 h, yielding the sample labelled CNT(BM 4 h). The composite materials (CNT:TiO_2) were synthesised using the same method, combining TiO_2 with CNT(BM 4 h) during 30 min at a vibration frequency of 10 Hz.[38] The resulting sample was labelled CNT:TiO_2.

2.1.2 Washcoating procedure using Triton™ X-100 (WCP_550 and WCP(CNT:TiO_2)_550). WCP_550 and WCP (CNT:TiO_2)_550 were prepared through washcoating methodology, adhering the previously synthesised powder catalysts (CNT(BM 4 h) and CNT:TiO_2, respectively). WCP_550 is the macrostructured catalyst to be used in the ozonation process, while WCP(CNT:TiO_2)_550 are the catalyst to be used in the hydrogenation process (used as support for bimetallic nanoparticles). To this end, a coating solution was prepared by performing adequate dispersion of CNT(BM 4 h) or CNT:TiO_2 catalyst in water with the aid of Triton X-100™ (labelled TX-100). For the adequate immobilization of CNT:TiO_2, a mixture of NYACOL® (commonly called nyacol) and TX-100 was necessary to promote the efficient dispersion of both phases of the catalyst.

The prepared coating solution was immobilised on the cordierite structure, through washcoating, using a Bungard RDC21 K coater. The monolith coating was performed five times with a drying step (in an oven at 100 °C) between each coating. Finally, the prepared catalyst was calcined in a vertical oven for 2 h, under N_2 atmosphere, at 550 °C, with a heating rate of 10 °C min^{-1}, to ensure the surfactant's complete decomposition.

For bimetallic catalysts synthesis (hydrogenation catalysts – WCP(CNT:TiO_2)_550_Pd_200_Cu), metal nanoparticles incorporation (Pd and Cu) was achieved through a staged impregnation methodology, using aqueous solutions of each of the metal precursors. An aqueous solution of the respective metal precursors ($PdCl_2$ and $Cu(NO_3)_2 \cdot 3H_2O$) was put in contact with the previously coated structure, at different stages. First, the macrostructure catalyst was contacted with an aqueous solution (5% HCl, 37% from Sigma-Aldrich) of $PdCl_2$ (99% from Sigma-Aldrich), in a shaking water bath (VWR) for 8 h followed by a drying step at 100 °C and a reductive thermal treatment (200 °C, 1 h N_2 and 3 h H_2) for metal nanoparticles formation. A second adsorption step of an aqueous solution of $Cu(NO_3)_2 \cdot 3H_2O$ (99% from Sigma-Aldrich) was performed, under the same conditions.[35,39]

2.1.3 Washcoating procedure using sodium alginate (WCP$_{SA}$_550 and WCP(CNT:TiO_2)$_{SA}$_550). Similar to the previous section, sodium alginate (SA) was applied as dispersant for both CNT(BM 4 h) and CNT:TiO_2 dispersion in water to produce WCP$_{SA}$_550 and WCP(CNT:TiO_2)$_{SA}$_550 macrostructured catalysts, respectively.[37] This dispersant was tested as a greener alternative to TX-100 application.

The prepared coating solution was applied using the washcoating methodology described in the previous section; however, the coating process was carried out manually, without the use of a dip coater, due to limitations associated with coating solution viscosity.

For the specific case of this preparation, the cordierite monolith was, first, coated with an aqueous solution of colloidal alumina (nyacol : water, 1 : 1, v/v) in order to increase the cordierite porosity and enhance the coating adhesion.[37] This coating was performed two times (with a drying step in an oven, at 100 °C, between each coating). Finally, a calcination step was performed at 550 °C, under an air atmosphere with a heating rate of 2 °C min^{-1}.

The immobilization of the dispersed catalyst was performed through dip-coating, with a drying step between each coating and a final calcination treatment was performed in a vertical furnace under N_2 atmosphere at 550 °C, with a heating rate of 2 °C min^{-1} to ensure the complete sodium alginate decomposition and avoid coating cracking due to high temperature rates.

The bimetallic macrostructured catalyst (WCP(CNT:TiO_2)$_{SA}$_550_Pd_200_Cu), metal deposition was performed through the methodology described in section 2.1.2.

2.1.4 Macrostructured catalysts synthesis through chemical vapor deposition (CVD). Chemical vapor deposition emerged as the most conventional technique for the synthesis of carbon macrostructured catalysts. For the CVD preparation, the cordierite monolith was dipped, first into a nyacol solution, and then in a nickel suspension, with a thermal treatment at 450 °C under N_2 atmosphere (50 cm^3 min^{-1}) between each stage. The final phase of preparation consisted on the growth of the carbon nanofibers on the monolithic structure through a thermal treatment until 650 °C, first with a $N_2 : H_2$ mixture (25 : 25) and finally with $C_2H_6 : H_2$ (30 : 30).[14]

For the bimetallic macrostructured catalyst (CVD_Pd_200_Cu), the metal deposition was performed through the methodology described in section 2.1.2.

2.2 Integrated catalytic system for water treatment

A two-stage bubble column system was designed and constructed for the simultaneous treatment of organic and inorganic species, consisting of catalytic ozonation followed by catalytic reduction (International Patent Application No. PCT/IB2024/058128). In this work, catalytic ozonation was first applied for organic compounds degradation, followed by the application of a catalytic reduction treatment to convert the inorganic species that were formed during the previous stage. A schematic representation of the developed system can be found in the SI (Fig. S1). Each column is 2.2 cm in diameter and 60 cm in length. For each experiment, the centre section of each column was packed with three macrostructured catalysts, covering an approximate height of 16.5 cm.[40]

Each column included a liquid inlet and outlet, a liquid recirculation loop, and a gas inlet and outlet. The liquid entered near the top of column 1, at 10 mL min^{-1}, while liquid recirculation in both columns was maintained at 100 mL min^{-1}. The liquid outflow from the bottom of column 2 was controlled with a needle valve.

Ozone (O_3), the only reactive gas in column 1, was generated from oxygen (O_2) using a BMT 802X O_3 generator at a constant flow of 200 cm^3 min^{-1} and an inlet ozone concentration of 45 g m^{-3}. O_3 concentration was monitored with a BMT 964 O_3 analyzer. In column 2, a mixture of hydrogen and carbon dioxide ($H_2:CO_2$) (1 : 1) was fed at 200 cm^3 min^{-1}. All gas flows were precisely controlled with calibrated mass flow controllers, and gases were vented at the top of each column.

Two sampling points were incorporated in the setup: one just before the liquid inlet of the hydrogenation column, and another at the system outlet, following the complete treatment process. Samples were collected from these points at specific intervals for analysis.

The used reaction parameters and conditions were kept constant for all the experiments and only the combination of the used catalysts (in each reaction) was varied.

2.3 Life cycle assessment methodology

The LCA methodology is the tool used in this study to assess the impacts associated with the application of the developed integrated system for drinking water treatment, considering different scenarios related to the macrostructured catalysts combination. LCA is a systematic procedure that addresses the life cycle stages of a product, process, or service's life cycle going from raw materials extraction, through manufacturing, distribution, use, and disposal/recycling. The goal of an LCA is to identify areas where environmental improvements can be made, helping organizations to make informed decisions to reduce their ecological footprint.

LCA is recognised by using four interconnected phases, as outlined by international standards ISO 14040 and ISO 14044,[33] as briefly described as follows:

(1) **Goal and scope definition**: define the purpose of the study, the product or process being assessed, and the boundaries of the system (*e.g.*, which stages of the life cycle will be included).

(2) **Life cycle inventory (LCI)**: gather data on all inputs (*e.g.*, materials, energy) and outputs (*e.g.*, emissions, waste) for each stage of the life cycle.

(3) **Life cycle impact assessment (LCIA)**: analyse the inventory data to assess potential environmental impacts, such as greenhouse gas emissions, water consumption, or resource depletion.

(4) **Interpretation**: draw conclusions and make recommendations based on the results, ensuring alignment with the study's goals.

Given its interconnection, the LCA methodology is an iterative process, and its holistic approach is widely applied in sectors such as energy, construction and consumer goods to evaluate the environmental performance of products and technologies. It helps identify key environmental hotspots and guides research methodologies for improvement; while using LCA, decision-makers can make data-driven choices that align with sustainability goals and minimize unintended consequences.

2.4 Environmental LCA of the integrated catalytic system application

2.4.1 Goal and scope.

In line with LCA principles, the study begins with the goal and scope definition, focusing on the environmental performance of a novel catalytic water treatment system across its life cycle stages. The system employs sequential catalytic ozonation and reduction for drinking water treatment, using immobilised macrostructured catalysts. The analysis is divided into two stages: (i) synthesis of the macrostructured catalysts, assessed with a cradle-to-gate approach, and (ii) application of the integrated treatment system, assessed with a cradle-to-grave approach. The functional unit is defined as 1 L of treated water. This functional unit was chosen because the catalyst configurations under evaluation exhibited similar treatment performance within the experimental conditions of this study (as it is going to be presented later in the present section). The system boundaries are illustrated in Fig. 1.

Three different scenarios were considered for the application of the integrated treatment, considering different combinations of the applied catalysts, with the main difference being the synthesis methodologies employed. Table 1 describes the three different scenarios considered for the integrated treatment application.

The three catalysts combinations were applied to the treatment of surface water collected at the inlet of a drinking water treatment plant (DWTP) and were published in a previous work.[41] The initial characterization of the used water was performed by UHPLC-MS/MS, targeting 34 compounds of high environmental concern. From these, 13 compounds were detected in the raw sample, including pharmaceuticals, personal care products, industrial additivies and pesticides. Most of the detected micropollutants were effectively removed after the oxidative stage of the integrated catalytic system with similar degradation results regardless of the catalyst used in the oxidation step (WCP_550 or WCP_{SA}_550), indicating comparable performance during this phase.

After oxidation, the treated water was directly subjected to a second stage based on catalytic hydrogenation, aiming to convert inorganic by-products formed during the oxidative

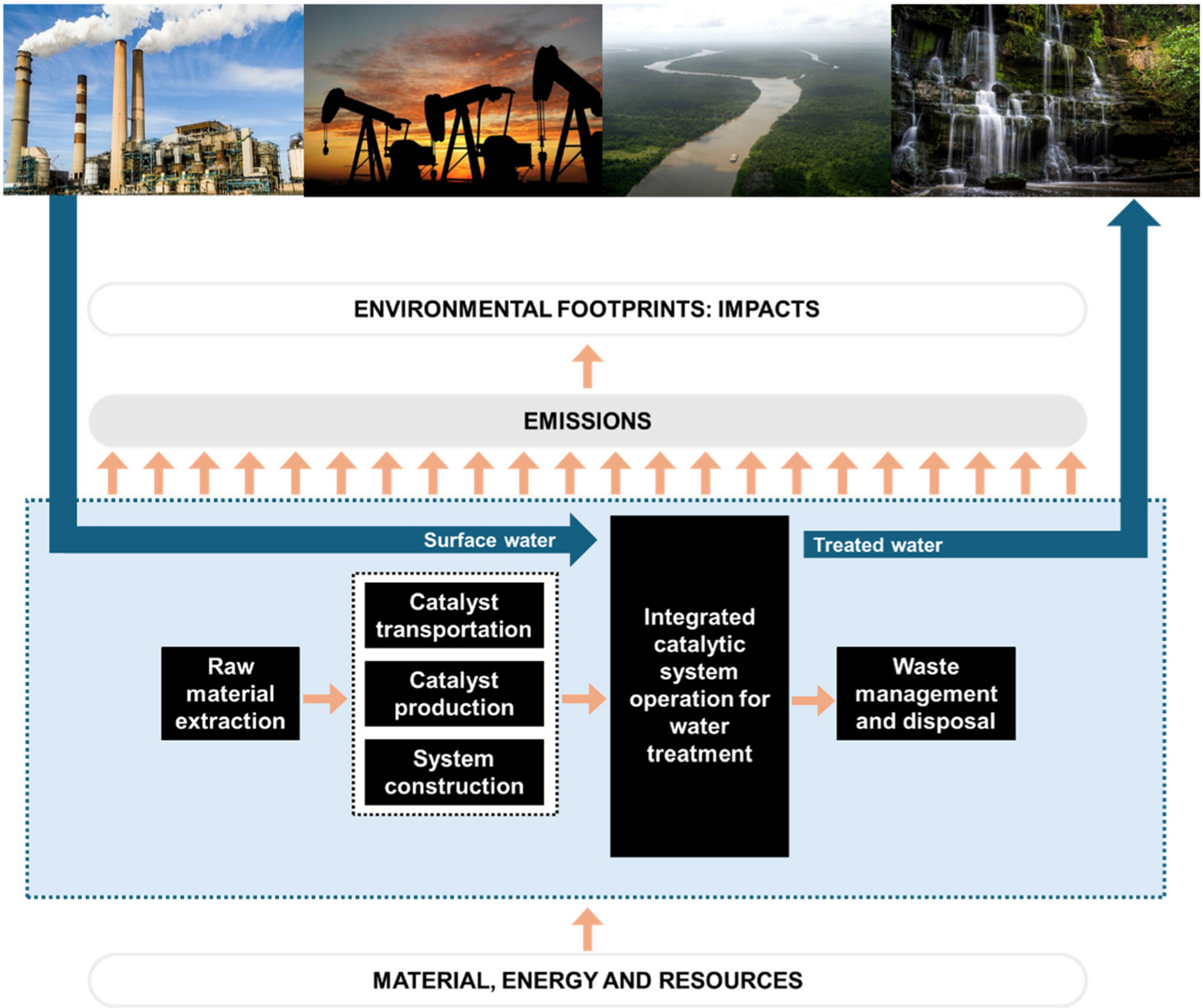

Fig. 1 System boundaries of the system for the LCA study covering raw material extraction, catalyst synthesis, system construction, operation, and end-of-life treatment.

Table 1 Catalyst system and reaction conditions for each scenario studied

Scenario	Ozonation catalyst	Hydrogenation catalyst	Treatment gases	Liquid flow
1	WCP_550	WCP(CNT:TiO_2)_550_Pd_200_Cu	45 g m^{-3} O_3 (using 200 cm^3 min^{-1} of O_2)	10 mL min^{-1}
2	WCP_550	CVD_Pd_200_Cu	200 cm^3 min^{-1} $H_2 + CO_2$ (1 : 1)	
3	WCP_{SA}_550	WCP(CNT:TiO_2)$_{SA}$_550_Pd_200_Cu		

process. At this stage particular attention was given to BrO_3^-. The initial surface water contained, approximately 5 µg L^{-1} of BrO_3^- (which is below the recommended legal limit for the presence of this compound in drinking water intended sources). However, after the oxidative treatment, BrO_3^- concentrations increased to values, around 15 µg L^{-1} as a consequence of the oxidation of the organic micropollutants and Br^- species present in the water source, exceeding the recommended limit (10 µg L^{-1}). The subsequent hydrogenation step successfully reduced BrO_3^- concentrations to values below this limit, independently of the catalyst used in this stage (WCP(CNT:TiO_2)_550_Pd_200_Cu, WCP(CNT:TiO_2)$_{SA}$_550_Pd_200_Cu or CVD_Pd_200_Cu), confirming the effectiveness of the integrated treatment approach.

The similar performance of the catalysts in degrading the target pollutants present in water allows to define the scope of the life cycle environmental assessment in the next section, which focuses exclusively on the production and synthesis stage of the catalysts, since they fulfil the same functionality during the use phase.

In what concerns the applied macrostructured catalysts, they have been previously adequately studied in later works.[16,21,36,37,41] Generally, metal-free carbon macrostructured catalysts (WCP_550 and $\mathrm{WCP_{SA}\_550}$) showed good mechanical resistance (considering the adherence tests performed using a US bath during 30 min) and excellent durability under the considered ozonation conditions. For the specific case of the experiments performed with the integrated catalytic system for surface water treatment, no coating detaching was detected during the considered reaction time. In what concerns the bimetallic macrostructured catalysts applied during the hydrogenation stage ($\mathrm{WCP(CNT{:}TiO_2)\_550\_Pd\_200\_Cu}$, $\mathrm{WCP(CNT{:}TiO_2)_{SA}\_550\_Pd\_200\_Cu}$ and CVD_Pd_200_Cu), the bimetallic systems showed no detectable Pd or Cu leaching over 30 h of operation, confirming their suitability for water treatment applications.[41]

2.4.2 Life cycle inventory analysis (LCI)

2.4.2.1 Bill of materials and energy: production and catalyst testing. The inventory in a Life Cycle Inventory (LCI) typically comprises foreground data, collected directly from the main process or system under study, and background data, related to upstream and downstream processes. Accordingly, all material and energy flows, along with SI for the foreground system, were obtained through laboratory experiments focused on the synthesis and application of the catalysts considered in the model. Data gaps were addressed using assumptions and calculations based on balanced chemical equations and thermodynamic principles, or through additional experimental measurements. The inventories for each scenario are presented in the SI, in Tables S1–S3, which provide detailed accounts of the materials, equipment, and energy consumption associated with the chemical processes for catalyst preparation and testing in each respective scenario.

2.4.2.2 Transportation system. The environmental impacts associated with the transportation of raw materials were estimated following the ecoinvent methodology. For suppliers located within Europe, transportation was modelled using average distances of 130 km by freight van, 240 km by train, and 270 km by freight ship. For suppliers outside Europe, longer transport routes were considered, involving either 1000 km by freight van followed by 18 000 km by transoceanic ship, or alternatively, 10 000 km by cargo plane.[42]

2.4.2.3 End of life – disposal. The amount of waste products (mainly related to the macrostructured catalysts synthesis process) was calculated considering the synthesis of 3 macrostructured catalysts for each preparation methodology. These waste products were estimated assuming that, after preparation, the liquid and solid waste were properly sent to a waste treatment system, where they would be properly disposed of, and gaseous emissions were discharged into the atmosphere. Considering the laboratory limitations that prevent the detection and quantification of the gases released during the thermal treatment applied in the synthesis of the macrostructured catalysts, the impacts associated with these emissions were not accounted for in the scope of the present life cycle assessment study.

After synthesis, the remaining coating solution, consisting of water, the powder material, and the respective dispersant (TX-100 or SA) could be adequately stored (in a glass container) to be used again in the next synthesis. For the solution to be properly reused, it needs to be re-dispersed in an ultrasonic bath, for around 15 min, before being re-used to perform the coatings. If the solution is not reused, it will be disposed of as a liquid waste solution. For the impregnation phase, the starting solutions of the metal phase become liquid waste solutions (metal salts diluted in water with traces of HCl).

2.4.2.4 Background data. The LCA model for impact calculations primarily focused on material and energy inputs, using recognised datasets such as ecoinvent and LCA for Experts (Sphera). Emission outputs, including those declared as foreground data and released to air, water, or soil during the preparation and operation of the catalyst system, were not included in the impact calculations due to simplifications made by using Excel spreadsheets.

During thermal treatment processes for surfactant decomposition or metal nanoparticles formation, various gaseous emissions (currently unidentified and unquantified) are released into the atmosphere. These emissions are typically modelled within specialised software as part of the life cycle inventory (LCI), where they are quantified as specific pollutants or substances released during process stages. This step may be critical to ensure a comprehensive inventory that reflects the full environmental footprint. The authors are aware that the omission of these emissions in this study introduces uncertainty, potentially underestimating the environmental burden across key impact categories such as global warming potential, acidification, and eutrophication.

Liquid and solid waste generated in this study were properly managed through a waste treatment system, which helps mitigate some of the potential impacts. Additionally, the remaining coating solution, identified as a potential liquid emission, is proposed for reuse. Regarding the material/reagent and transportation, generic scenarios were assumed to calculate their impacts.

To address the underestimation associated with excluded foreground emissions, we estimated emission-specific impact contributions using EF 3.1 midpoint characterisation factors. These results were added to the original LCA outputs for each relevant impact category. This combined approach provides a more accurate estimate of the total environmental burden of the process, especially for categories such as global warming, acidification, and eutrophication.

2.4.2.5 Life cycle impact assessment (LCIA). Considering the compiled data analysis for the life cycle inventory, a life cycle impact assessment was conducted to evaluate the environmental implications of macrostructured catalysts synthesis (support and bimetallic) and the application of the novel integrated catalytic system for water treatment intended for human consumption. This means that the inventory data, including background and foreground data obtained from laboratory experiments on the synthesis and application of all catalysts, were mathematically processed to calculate the environmental impact categories. To this end, the inventory was pre-calculated for inputs such as materials (per kg or L) and energy consumption (kWh) using recognised datasets from ecoinvent v3.9 and LCA for Experts (Sphera Solutions GmbH) for converting the flows to category impacts. This approach enabled the creation of an auxiliary database containing pre-calculated impacts per material, which was subsequently used in the calculation model implemented in open-source Python software. In this context, the impact of entry considered in the life cycle inventory was calculated, considering the 31 impact categories selected for this study, as shown in Table S4, which are present in the SI.

This study employed the Environmental Footprint (EF) and Primary Energy Demand (PED) methods, both of which are based on midpoint indicators used in Life Cycle Impact Assessment (LCIA).[43–46] These indicators quantify environmental impacts at an intermediate stage, offering detailed insights before final damage occurs. The EF method, developed by the European Commission, assesses various impact categories, while PED measures the total raw energy required to produce a product or service.

2.4.2.6 Limitations and assumptions. For the assessment of the applied treatment, some assumptions were made:

- All the applied catalysts were considered active for at least 30 h of reaction, the equivalent of producing 18 000 L of treated water. Taking into consideration that, at this stage, longer experiments were not considered, these assumptions were used to define the functional unit as 1 L of treated water.
- The impacts related to the system construction were calculated taking into consideration the entire lifetime of the system *i.e.* considering the total period in which the system maintained its integrity and functioning without requiring any alteration or replacement. To address this, parameters were calculated based on the scalability of each stage. It was assumed that approximately 300 experiments, each lasting 5 h, were conducted over a 5 year period using the same system without any modifications. In this vein, considering the scalability process, the impact distribution related to the system construction only accounts for around 10 to 20% of the total impact attributed to the integrated system (construction + application).
- All O_3 was considered to be converted to oxygen after exiting the system by means of contact with KI (2%) solution, so it is not considered any release of O_3 to the atmosphere.
- Gaseous emissions potentially generated during catalyst thermal treatment (*e.g.*, CO, light hydrocarbons or VOCs from dispersant decomposition) were not explicitly quantified due to the absence of exhaust gas monitoring during furnace operation. Consequently, these emissions were not included in the life-cycle inventory and the environmental impacts of this stage were modelled based on the measured electricity consumption of the furnace. This represents a limitation of the present assessment.

2.4.3 Sensitivity analysis. A sensitivity analysis was performed for the inputs that showed a higher environmental impact contribution. Parameters whose changes significantly affect the result, or the variance of the output, are considered sensitive.

For this analysis, the energy intensity in different countries (Portugal, Spain, France, Italy, and an average value for Europe) was varied. Additionally, different European electricity sources (photovoltaic and wind power) datasets mentioned above (ecoinvent and LCA for Experts) were considered.

3. Results and discussion

3.1 Overall LCA of the complete integrated treatment application

The overall impact of the technology – evaluated across the different scenarios defined in Table 1 – was calculated with reference to the production of 1 L of treated water. The obtained results are presented in Fig. 2, while a comprehensive analysis of all calculated impacts is provided in Table S5, presented in the SI.

As the great difference among the three scenarios lies in the implemented catalysts, the variations in the overall impacts are primarily attributed to the synthesis processes associated with these catalysts. Overall, the impacts per liter of treated water indicate that the scenario with the highest impact is Scenario 2 (corresponding to the WCP_550 + O_3|CVD_Pd_200_Cu + (H_2 + CO_2) system), followed by Scenario 3 (WCP$_{SA}$_550 + O_3|WCP(CNT:TiO$_2$)$_{SA}$_550_Pd_500_Cu + (H_2 + CO_2)), and finally Scenario 1 (WCP_550 + O_3|WCP(CNT:TiO$_2$)_550_Pd_500_Cu + (H_2 + CO_2)). This outcome is particularly noteworthy, as it demonstrates that the washcoating-based preparation methods developed in this project result in significantly lower environmental impacts compared to the more

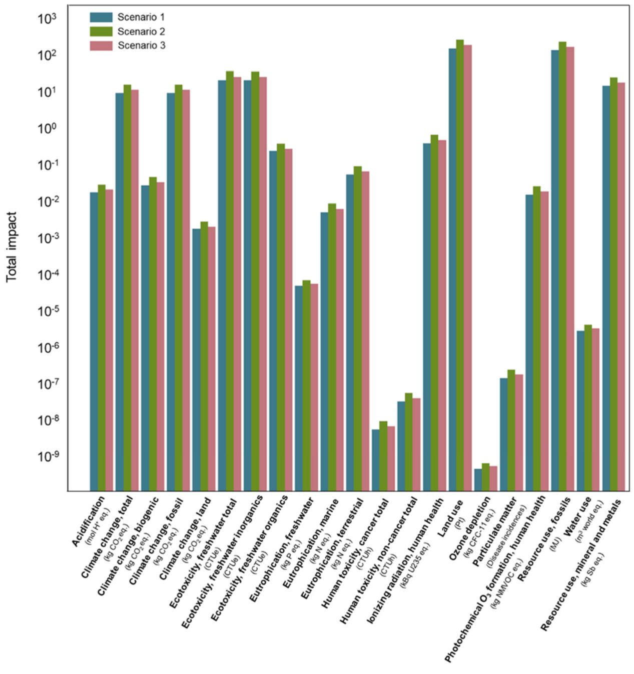

Fig. 2 Normalised overall impacts categories per 1 L of treated water to compare scenarios.

conventional CVD-based approach commonly used for producing macrostructured carbon catalysts.[12,47]

Scenarios 1 and 3, both employing washcoating techniques for macrostructured catalysts preparation, exhibit comparable environmental impacts. However, Scenario 3, which uses sodium alginate as a surfactant, has a slightly higher (around 5% higher across most of the indicators) associated impact compared to Scenario 1, where the catalyst is prepared using TX-100.

Fig. S2 presents the category impacts by life cycle stages—transport of raw materials and reagents, macrostructured catalyst synthesis, and system implementation—for each scenario. The macrostructure catalyst synthesis is the dominant contributor in 85% of the indicators studied. This behaviour is not followed in ozone depletion, resource use and human toxicity cancer, in particular these last two categories, where the integrated operating treatment has a higher contribution. In contrast, the transportation system shows the lowest life cycle contributor in all category impacts, which means raw-materials transportation is not an environmental hotspot in this study.

These results show that, although all catalyst systems are effective in producing water suitable for human consumption, variations in synthesis methods and materials significantly influence their environmental life cycle performance. In particular, indicators related to water use and ecotoxicity are notably affected by the catalyst synthesis process. Therefore, the purification of water for human consumption should be balanced with life cycle considerations to ensure greater sustainability. Advancing more sustainable catalyst synthesis strategies is essential to reducing the overall environmental impact of the process.

Regarding the contributions of each phase to the overall impact of the system, the synthesis of the macrostructured catalysts accounts for approximately 97–99% of the impacts associated with the system's use. In contrast, the impacts related to the transportation of raw materials and gases, as well as the construction and application of the system, collectively represent only about 1–3% of the total impacts.

3.2 Life cycle analysis of the macrostructured catalysts synthesis

Since the macrostructure catalysts synthesis shows higher impacts in most of the considered impact categories, this section shows the LCA results for the synthesis processes involved in the production of macrostructured catalysts, intended for application in ozonation and hydrogenation reactions. To better understand how the total environmental impact is distributed across the different stages of catalyst preparation, heatmaps were generated for each scenario. These heatmaps illustrate the impact distribution associated with each synthesis method and are available in the SI (Fig. S3–S5). Based on these results, the following sections provide a detailed analysis of the LCA results, organised by synthesis method and by the specific components or inputs used in the catalyst production.

3.2.1 Triton™ X-100-based macrostructured catalysts (Scenario 1). In Scenario 1, the catalysts WCP_550 and WCP(CNT:TiO_2)_550_Pd_200_Cu were prepared using TX-100 as dispersant for CNT(BM 4 h) and CNT:TiO_2 powders, respectively. The synthesis process comprises three main stages: washcoating solution preparation (WSP), washcoating procedure (WP), and thermal treatment for surfactant decomposition (TTSD). For the bimetallic catalyst used in the hydrogenation stage (WCP(CNT:TiO_2)_550_Pd_200_Cu), an additional metal impregnation step (MI) is included.

According to the results shown in Fig. S3, TTSD stage is the main contributor to the overall environmental impact across all impact categories. This stage includes both the application of thermal treatment and the use of nitrogen gas, with energy consumption being the primary driver of the associated impacts. In both WSP and WP stages, electricity use is the main source of impact. Specifically, ball-milling (for CNT(BM 4 h) and CNT:TiO_2 synthesis) and ultrasonic treatment (to disperse the powder catalyst) are the most energy-intensive processes associated with the WSP stage, while the drying step in an oven is the most impactful part of the WP stage.

Fig. 3 shows the results, by inputs, for WSP, WP and TTSD stages. Since WP and TTSD are common to both catalysts (ozonation and hydrogenation), Fig. 4 focuses on the WSP and MI stages for WCP(CNT:TiO_2)_550_Pd_200_Cu, which requires a modified dispersion due to the use of a composite material as support for the active metal phase.

From Fig. 3 and 4, it can be observed that the environmental impact associated with electricity consumption during the thermal treatment, drying between coatings, and US-Probe stages is the main contributor, accounting for 40 to 100% of the total impact. This variation is directly related to the type of energy source used in the process (fossil or renewable).

Fig. S6 presents the primary energy demand across the synthesis stages of WCP_550 and WCP(CNT:TiO_2)_550_Pd_200_Cu. In both cases, renewable energy accounts for a larger share than fossil sources throughout the process (741 MJ *vs.* 593 MJ for WCP_550). The WCP(CNT:TiO_2)_550_Pd_200_Cu catalyst includes an additional metal impregnation (MI) stage, which increases total primary energy demand by approximately 254 MJ (from both renewable and non-renewable sources). However, in both cases, the thermal treatment stage (TTSD) remains as the most significant contributor to energy consumption.

Although renewable energies significantly contribute to the process, their whole life cycle—including raw material extraction, infrastructure development, and operation—still involves environmental impacts.[48,49] These impacts, while generally lower than those from fossil energy, should not be overlooked. Nevertheless, the overall results are encouraging, as renewable energy use helps reduce the environmental burden of the three most energy-intensive stages.

Graphs constructed for each stage (WSP, WP and TTSD) by inputs can be found in Fig. S7–S9 in SI for both, WCP_550 and WCP(CNT:TiO_2)_550_Pd_200_Cu, macrostructured catalysts. A stage-by-stage analysis of the energy source distribution reveals

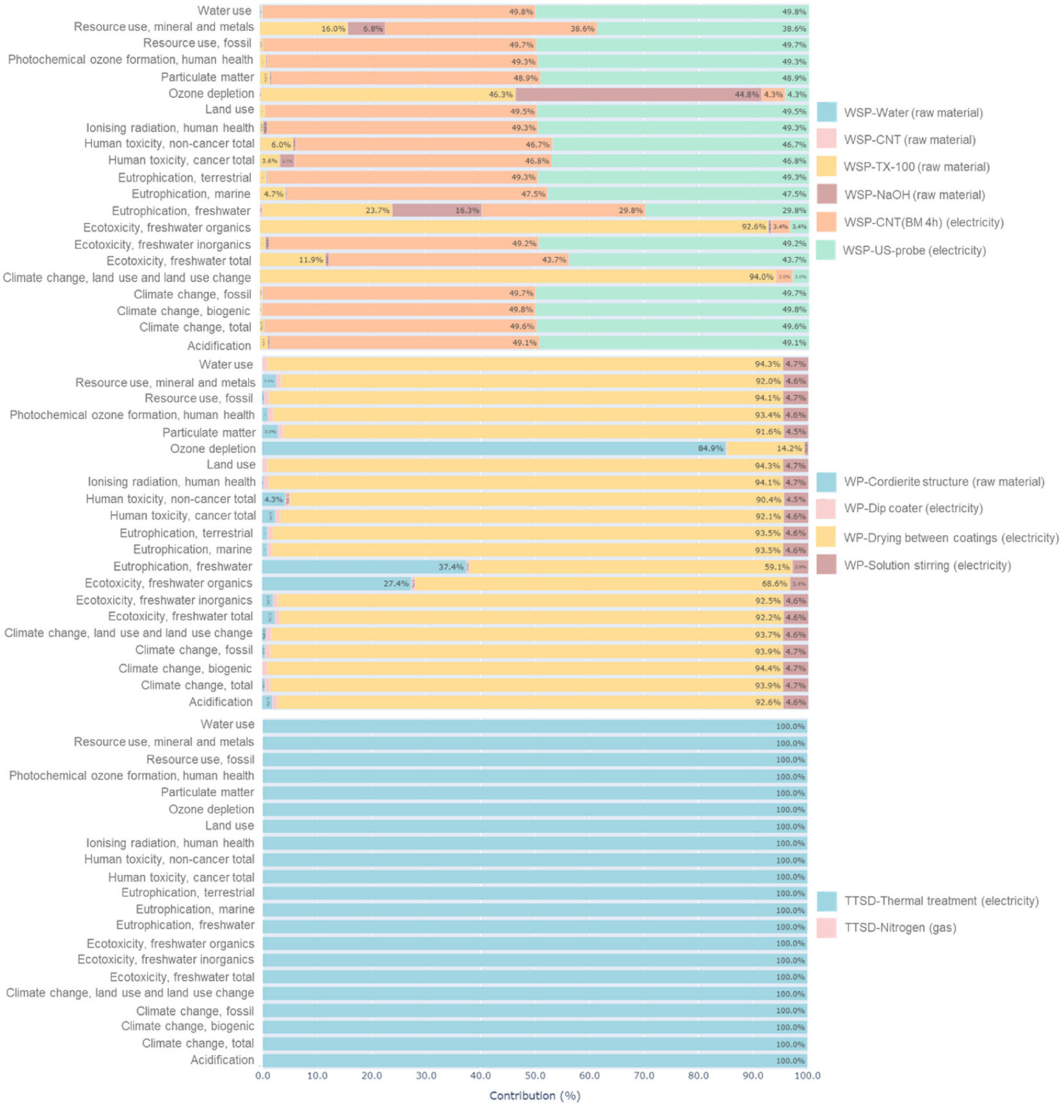

Fig. 3 Percentage contribution by inputs of WSP, WP and TTSD stages in Scenario 1 across impact categories.

a significant reliance on non-renewable inputs, particularly during stages involving raw materials and gases. The production of these inputs typically requires mining, refining, and chemical processing (activities that heavily depend on non-renewable primary energy sources such as fossil fuels). These energy sources are crucial for achieving the high temperatures and pressures needed in processes like nanoparticle synthesis, metal extraction, and chemical purification.[50] Consequently, this dependence on non-renewable sources significantly contributed to the environmental impact of catalyst production, highlighting the urgent need for more sustainable alternatives in both material sourcing and energy use. Metal impregnation (MI) stage is highly dependent on non-renewable energy use, which is specially pronounced due to the involvement of palladium chloride—a key material. The extraction, processing, and use of palladium chloride are closely tied to considerable environmental impacts, with non-renewable energy consumption being a major factor throughout its entire lifecycle.

Some reduction of energy-intensive impacts could be achieved by modifying specific synthesis steps. For example, replacing oven drying between coatings with open-air drying would lower energy consumption; however, this change would considerably extend the synthesis time, as each layer must be fully dried before the next can be applied. Regarding the thermal treatment for surfactant decomposition (TTSD), mitigation is limited, as this step is essential to ensure catalyst performance. Therefore, impact reduction would depend on optimizing thermal treatment conditions, particularly through improved furnace and reactor design to enhance heat transfer efficiency.

Beyond energy-related impacts, Fig. 2 shows relevant impact contributions related to the overall environmental impacts. In the WP phase of both macrostructured catalysts

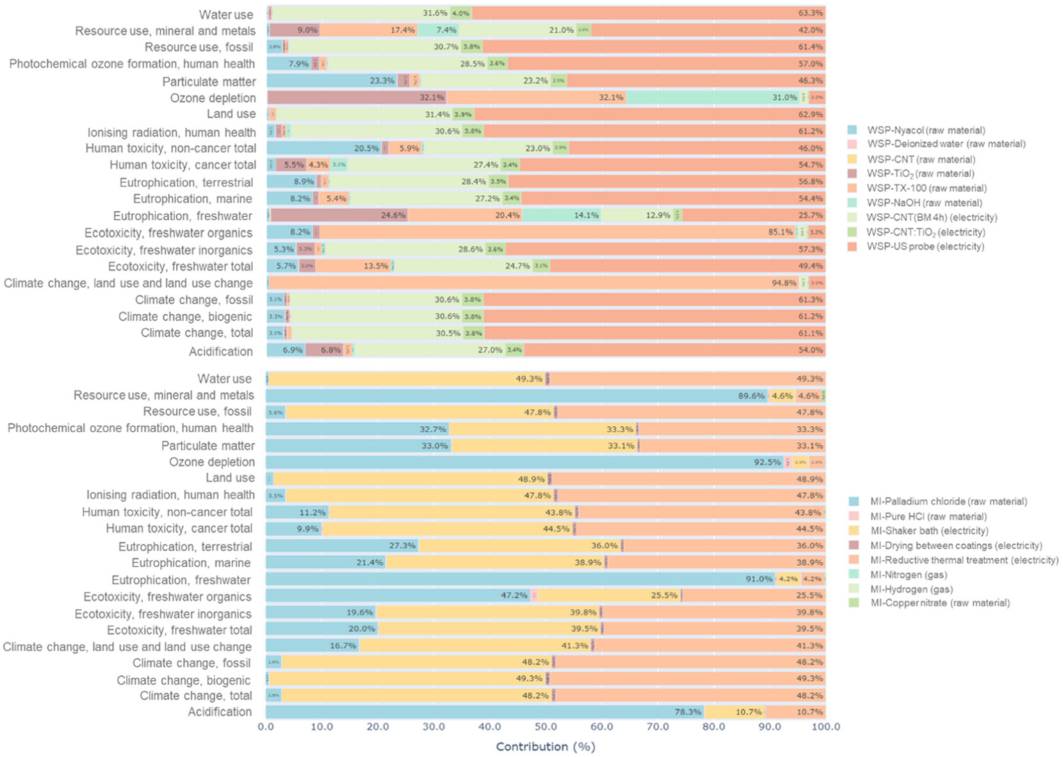

Fig. 4 Percentage contribution by inputs of WSP and MI stages for the synthesis of WCP(CNT:TiO_2)_550_Pd_200_Cu.

synthesis, the cordierite structure emerges as a major contributor—particularly to ‘Ozone depletion’, ‘Freshwater eutrophication’, and ‘Freshwater ecotoxicity (organics)’. These impacts are likely linked to cordierite composition, which primarily consists of MgO, SiO_2, and Al_2O_3, and goes from the extraction, to processing, and application of these raw materials.[51] The cordierite structure accounts for approximately 85% of the ‘Ozone depletion’ impact in the WP stage, mainly due to the production and processing of metal oxides. These processes involve mining, chemical treatments, and thermal operations that can release ozone-depleting substances. Additionally, runoff containing phosphorus or nitrogen from these activities contributes to freshwater eutrophication and ecotoxicity.

Although the environmental impact of the washcoating solution preparation (WSP) stage is relatively minor, the use of TX-100 as a surfactant for CNT dispersion stands out as a notable contributor across all impact categories (excluding electricity-related impacts). TX-100 (petroleum-based origin) is associated with fossil fuel depletion, greenhouse gas emissions, and toxicity to aquatic ecosystems.[52] Its life cycle—from production to disposal—also contributes to acidification, eutrophication, and potential human health risks. These combined effects highlight TX-100 as a critical point in the environmental performance of macrostructured catalyst synthesis, underscoring the importance of exploring more sustainable alternatives.

In the specific case of WCP(CNT:TiO_2)_550 synthesis, beyond the impact associated with TX-100 and NaOH use, significant contributions also arise from the use of TiO_2, in the composite catalyst synthesis, and the application of Nyacol to ensure effective dispersion of the composite material.

TiO_2 production involves energy-intensive processes and the extraction of raw materials, which significantly contributes to resource depletion – particularly in the minerals and metals category.[53] Extraction and refinement stages also result in emissions of substances contributing to ozone depletion and acidification.[54,55] Furthermore, fine TiO_2 particles can leach into water bodies, contributing to eutrophication in freshwater systems due to their potential to disrupt aquatic ecosystems.[25,56] In what concerns to Nyacol use, the associated impacts (mainly on categories such as ‘Ozone formation’, ‘Particulate matter’, and ‘Human toxicity (non-cancer related)’) may be related to alumina nanoparticles production, involving processes that emit volatile organic compounds (VOCs) and fine particulate matter.[57,58] Such emissions contribute to ozone depletion and particulate matter emissions, impacting both human health and atmospheric quality. Additionally, the ecotoxicity and eutrophication effects observed in terrestrial, marine, and freshwater ecosystems can be attributed to the potential release of residual nanoparticles and chemicals during production or disposal.[59]

The detailed analysis of the MI stage highlighted the significant environmental impact associated with the use of palladium chloride, employed in the formation of palladium nanoparticles. This procedure proved to be quite impactful in the categories of resource use (89.6%), ozone depletion (92.5%), freshwater eutrophication (91.0%) and acidification (78.3%). The environmental impact associated with palladium chloride is more significant than that associated with energy-intensive steps, such as shaker treatment and reductive thermal processing. Palladium is a rare and precious metal, and its extraction involves significant environmental costs, including large-scale mining, energy consumption, and the generation of toxic waste.[60,61] These processes can also lead to the emission of pollutants that contribute to ozone depletion, especially through the use of industrial chemicals, like solvents or reagents, that may release harmful substances such as CFCs.[60]

Palladium chloride may also contribute to freshwater eutrophication, primarily through industrial runoff containing heavy metals and synthesis-related chemicals that degrade water quality and promote algal growth. Finally, the production and use of palladium compounds, particularly in mining and refining, can release acidic gases, such as sulphur dioxide and nitrogen oxides, leading to soil and water acidification. These environmental impacts highlight the need for more sustainable practices in the mining, production, and disposal of palladium-based materials. This will be one of the main problems associated with the synthesis of bimetallic macrostructured catalysts, as all of them (independent of the scenario) are Pd–Cu based catalysts.

To overcome this phenomenon, some optimization experiments could be performed, mainly focused on the optimization of metal nanoparticles synthesis and the impregnation stage. The shape, size, and distribution of supported metal nanoparticles on a catalyst play a crucial role in its catalytic performance.[62,63] Smaller, uniformly distributed nanoparticles provide a higher surface area for reactions, enhancing activity and selectivity.[64] Additionally, nanoparticles shape can influence their active facets exposure, directly affecting the efficiency of catalytic processes.[62,64] Optimizing the impregnation and synthesis methods for mono, bi, or trimetallic nanoparticles is crucial for enhancing catalytic activity while simultaneously addressing both economic and environmental challenges. Advanced techniques, such as precise control of precursor concentrations, deposition parameters, and thermal treatments, allow for better control over nanoparticle size, shape, and distribution.[65–67] These improvements maximize the exposure of active sites, thereby increasing the efficiency and selectivity of the catalyst. Additionally, such optimization reduces the number of expensive metals required (such as palladium, platinum, and gold), as higher activity can be achieved with smaller quantities of these materials. This reduction not only lowers the overall synthesis cost but also allows for the minimisation of chemical waste and by-products, which are often associated with traditional catalyst preparation processes.

3.2.2 Sodium alginate based macrostructured catalysts (Scenario 3). In an effort to reduce the environmental impacts of the synthesis procedure, the washcoating method was adapted to include 'greener' alternatives for dispersant.

One of the main concerns in the synthesis of macrostructured catalyst using surfactants to promote powder materials immobilization is the potential release of undecomposed surfactant residues into water during the reaction, even after applying a surfactant decomposition process. In the specific case of TX-100, this compound can be toxic even at very low concentrations and has a significant impact on aquatic life.

As a safer alternative, sodium alginate (a natural polysaccharide known for its excellent properties such as biosafety, biocompatibility, and renewability) was employed to disperse the powder catalysts and ensure their proper immobilization onto cordierite supports. Sodium alginate was selected as the main dispersant due to its efficiency in CNT dispersion.[37] Nevertheless, achieving highly stable and active macrostructured catalysts required procedural modifications, which may also influence the overall environmental performance of the process.

The main synthesis methodology can be divided into 3 different stages (for WCP_{SA}_550 and $WCP(CNT{:}TiO_2)_{SA}$_550 synthesis): cordierite preparation before coating (CP), washcoating solution preparation for immobilization in the cordierite structure (WSP) and the thermal treatment to perform the dispersant degradation (TTSD). Similar to what was described in section 3.1.1 an additional phase, regarding metal nanoparticles impregnation (MI) was performed for $WCP(CNT{:}TiO_2)_{SA}$_550_Pd_200_Cu synthesis. In addition to the use of a greener dispersant, the main novelty of this methodology is the efficient dispersion of a composite catalyst ($CNT{:}TiO_2$) using sodium alginate, without the necessity of adapting the methodology to achieve proper dispersion of both phases of the catalyst. The synthesized catalyst (WCP_{SA}_550 and $WCP(CNT{:}TiO_2)_{SA}$_550_Pd_200_Cu) was used as the main catalyst for ozonation and hydrogenation treatment in Scenario 3.

The constructed heatmap in Fig. S5 reveals similar results to those observed for TX-100 based catalysts, identifying TTSD phase as the most impactful stage, mainly due to the high energy demand of the thermal treatment required for sodium alginate decomposition.

At this stage, the TTSD phase is similar for both TX-100 and sodium alginate-based catalysts, with the main difference being the applied heating ramp (10 °C min^{-1} for TX-100 and 2 °C min^{-1} for sodium alginate to prevent coating cracks), resulting in higher energy consumption.

The impact distribution related to CP and WSP phases (for both, CNT(BM 4 h) and $CNT{:}TiO_2$ dispersion) is depicted in Fig. 5. Similar to Scenario 1, the electricity consumption involved in the different processes of the preparation procedures has a significant contribution, in particular the cordierite preparation stage (CP) accounting for almost 100% in all indicators.

On the other hand, the life cycle associated with the production/use of the cordierite structure still presents a relevant impact for the category of ozone depletion (similar to what was observed for WCP_550 and $WCP(CNT{:}TiO_2)$_550_Pd_200_Cu synthesis), but in this case, we have a contribution of around 95% for this indicator attributed to sodium alginate as raw material.

The main difference between this synthesis method and the previously described approach using TX-100, as the primary dispersant, lies in the need for an additional step

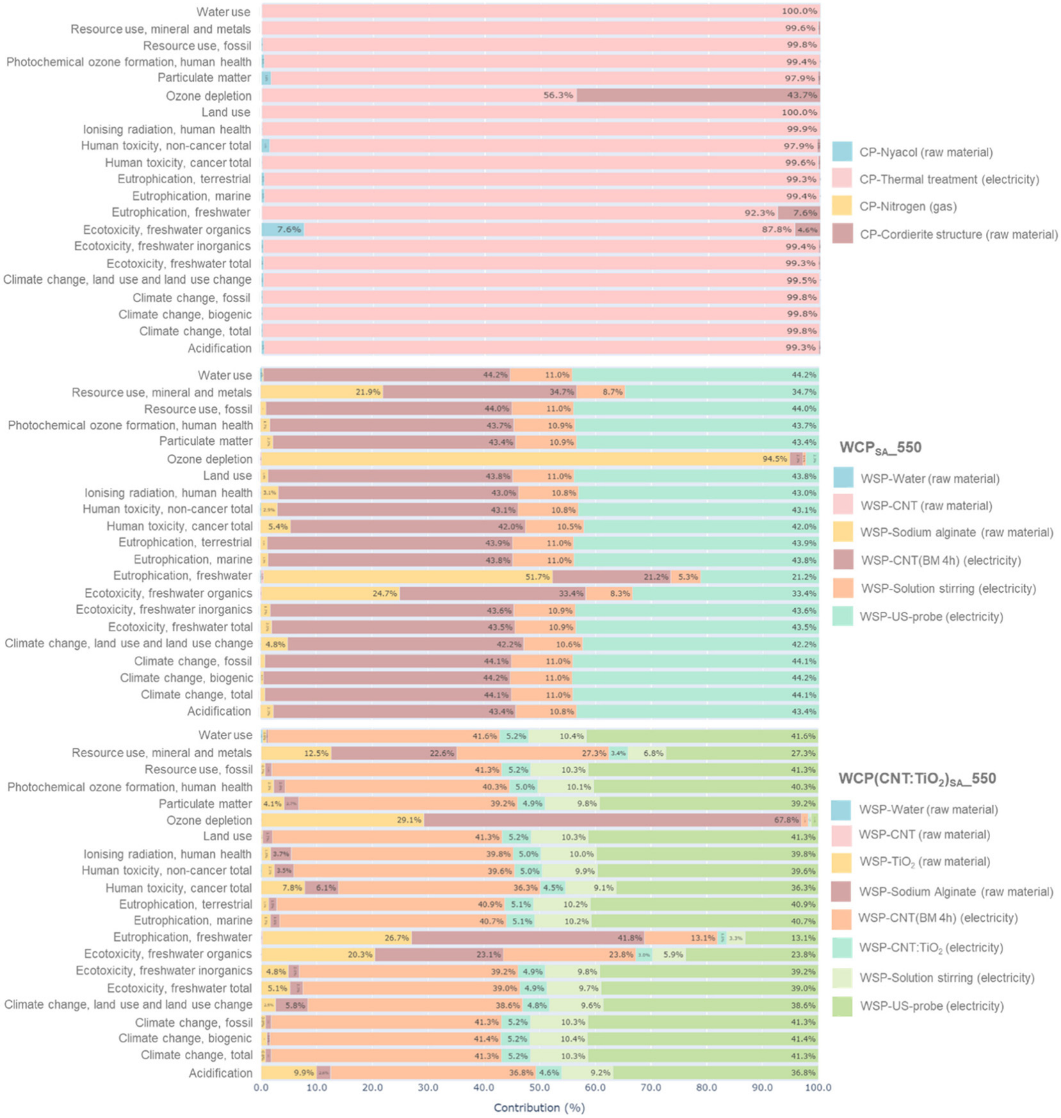

Fig. 5 Percentage contribution of CP and WSP stages, considering all the inputs, for the synthesis of WCP$_{SA}$_550 and WCP(CNT:TiO$_2$)$_{SA}$_550_Pd_200_Cu catalyst.

specifically dedicated to preparing the cordierite structure prior to coating. This preparatory phase is essential to ensure proper coating adhesion, contributing to the catalyst stability and it involves the application of an additional thermal treatment process.[37]

Additionally, in this new preparation method, a WP (washcoating procedure) does not exist since the coating of the structure is not performed using a dip-coater (it is done manually) and the drying process is carried out in open air (without the use of heated ovens to prevent "cracking" of the coating and ensure stability during the coating process).

As conclusion, the washcoating methodology, using sodium alginate as a greener dispersant, presents an innovative and effective approach for dispersing CNT:TiO$_2$ composite catalysts without requiring major modifications for phase compatibility. While this method reduces reliance on hazardous chemicals and simplifies certain process steps, it introduces new environmental trade-offs, particularly in the thermal treatment phase (TTSD), which shows increased energy demand due to slower heating ramps. Additionally, the cordierite preparation step (CP) remains a dominant contributor to environmental impacts, especially in terms of electricity use and ozone depletion, the latter strongly influenced by sodium alginate sourcing. Despite the environmental challenges associated with thermal treatment and raw-material production, this methodology represents a step forward in developing more sustainable catalyst preparation techniques for advanced water treatment applications.

Fig. S10 shows the results of CP and WSP phases without considering the impacts related to electricity consumption to better understand the contribution of raw materials and gases in this preparation method.

The use of sodium alginate has significant impacts, particularly in the categories of resource use (around 21.9% of total contribution), ozone depletion (around 94.5%), freshwater ecotoxicity due to organics (around 24.7%) and freshwater eutrophication (around 51.7%) categories. In terms of resource consumption—especially minerals and metals—the production of sodium alginate typically involves chemical agents such as sodium hydroxide (NaOH) and calcium salts, which are employed during the extraction and conversion processes.[68,69] These compounds are sourced from mining and industrial operations that significantly contribute to environmental degradation through energy consumption, emissions, and the depletion of finite mineral resources.[70] The extraction and processing of sodium and calcium, in particular, raise concerns regarding the long-term sustainability of sodium alginate production at an industrial scale.

Regarding ozone depletion, although the sodium alginate molecule itself does not contain ozone-depleting substances, its production processes may involve the use or release of volatile organic compounds (VOCs) and industrial solvents. Depending on the specific practices employed, these compounds can indirectly contribute to ozone layer degradation.[71,72] Moreover, when the energy required for alginate synthesis is derived from fossil fuels, additional indirect effects—such as emissions of nitrogen oxides and other pollutants—may further impact atmospheric chemistry and contribute to ozone depletion.

Freshwater eutrophication is another relevant concern. While sodium alginate is derived from seaweed, the large-scale cultivation of macroalgae can lead to nutrient runoff, particularly when fertilizers are applied in farming operations. Runoff from nearby agricultural activities can also contribute to the nutrient load in adjacent water bodies.[73] Additionally, industrial effluents containing residual chemicals from alginate processing, if inadequately treated, may release nitrogen and phosphorus compounds into freshwater systems, exacerbating eutrophication and threatening aquatic ecosystems.

In summary, although sodium alginate is a naturally derived biopolymer, its industrial production entails substantial environmental implications. The reliance on mineral-based reagents, potential emissions linked to ozone depletion, and the risk of freshwater nutrient loading underscore the importance of implementing more sustainable and controlled production practices within the alginate industry.

Metal nanoparticles impregnation (MI) stage, for WCP(CNT:TiO_2)$_{SA}$_550_Pd_200_Cu synthesis, proved to be quite similar, in terms of impacts, to the ones described for WCP(CNT:TiO_2)_550_Pd_200_Cu (Fig. 4 section 3.2.1).

Regarding primary energy demand, the synthesis using sodium alginate showed a clear increase in energy consumption compared to catalysts prepared with TX-100. The comparison between WCP_550 and WCP$_{SA}$_550 shows differences of around 575 MJ (1275 MJ against 1850 MJ, respectively), which may be related to the longer thermal treatment (with lower heating rate) required for sodium alginate decomposition process. The obtained results are presented in Fig. S11 in the SI.

3.2.3 Chemical vapor deposition macrostructured catalysts (Scenario 2).

In Scenario 2, the catalyst CVD_Pd_200_Cu was employed as a hydrogenation catalyst, representing the most conventional approach for macrostructured catalyst synthesis. The environmental impact associated with the production of this catalyst was evaluated to identify the most impactful stages of the process and to compare its overall burden with that of the alternative washcoating method developed in this study.

The conventional synthesis route was divided into four main stages: (i) coating of the cordierite substrate with a diluted Nyacol solution (NC), (ii) coating with a nickel precursor solution (NicC), (iii) carbon nanotube (CNT) growth (GS), and (iv) metal nanoparticle impregnation (MI). This breakdown allowed for a detailed life cycle assessment of each step, facilitating a more comprehensive comparison between the conventional and washcoated catalyst preparation methods.

The heatmap constructed for Scenario 2 (regarding catalysts synthesis) is shown in Fig. S4(B) and it is possible to see that the stages related to the synthesis of the macrostructured catalyst by CVD (NC, NicC and GS) generally have a greater impact than the metal phase impregnation (MI) stage, which was previously identified as one of the most impactful phases during the synthesis of the WCP(CNT:TiO_2)_550_Pd_200_Cu catalyst. The main reason for this result is related to the fact that all synthesis phases culminate in a high-temperature thermal treatment with very low and controlled heating ramps, leading to significant energy consumption. At this stage, MI phase is identical to the one reported for WCP(CNT:TiO_2)_550_Pd_200_Cu catalyst synthesis (see Fig. 5).

The detailed analysis of each preparation stage (Fig. S12) of the process allows to conclude that the impacts associated with energy consumption (mainly related to the thermal treatment necessary for synthesis) are the dominant factor in all phases (excluding the MI phase). If the impacts related to energy consumption are not considered (see Fig. S13), the impacts associated with the use of Nyacol and the nickel solution stand out as the most significant in each impact category for the NC and NicC stages, respectively. These impacts were shown to be considerably more relevant than the use of gas throughout the entire thermal treatment, in each stage, mostly related to the impacts associated with raw-materials extraction, production and purification, as previously discussed.

A comprehensive analysis of the energy consumption associated with the CVD_Pd_200_Cu synthesis process (see Fig. S14 and S15) reveals that its total energy demand is, approximately, four times higher than that of the catalysts produced *via* the washcoating method. Specifically, the energy requirements for synthesizing three macrostructured catalysts are 1526 MJ for WCP(CNT:TiO_2)_550_Pd_200_Cu and 2122 MJ for WCP(CNT:TiO_2)$_{SA}$_550_Pd_200_Cu, whereas the CVD-based process exhibits substantially greater energy consumption (around 5000 MJ).

3.3 Summary of the environmental impacts of methodologies used for macrostructured catalysts synthesis

Considering the different methodologies used for macrostructured catalyst synthesis (washcoating with TX-100, washcoating with sodium alginate, and direct growth *via* CVD), the overall environmental impact values were normalised and compared across the selected impact categories to allow for a consistent comparison. The resulting data is presented in Fig. 6.

Results are promising, as the washcoated catalysts developed in this study exhibited significantly lower environmental impacts compared to the conventionally used CVD catalysts. The catalyst synthesized using TX-100 as a surfactant achieved an approximate 50% reduction across impact categories, while those synthesized with SA as a dispersant showed a reduction of around 30%. Notably, this decrease in environmental impacts did not compromise catalytic performance—the washcoated catalysts maintained excellent activity, and the quality of the treated water remained within standards for human consumption when tested under the experimental conditions.

Although sodium alginate is generally considered a ‘greener’ and less hazardous dispersant, the synthesis of alginate-based catalysts resulted in important environmental impacts compared to those prepared using TX-100 as a surfactant. This difference mainly results from the higher energy demand of sodium alginate-based catalyst synthesis, which involves two separate thermal treatments at different stages (CP and TTSD) with lower heating rates, unlike TX-100-based catalysts, which require only a single heat treatment for surfactant removal. Furthermore, additional differences in the WSP stage between the two preparation methods may also contribute to the overall impact variation, although their influence appears to be less significant than that of energy consumption. Fig. S16 shows the comparison between the two washcoating synthesis methodologies, considering only the inputs related to WSP stage (A) and considering all the stages excluding inputs related to electricity consumption (B). Overall, when disregarding energy-related inputs, the distribution of impacts between the two methodologies becomes somewhat more balanced. Nevertheless, the synthesis of macrostructured catalysts using sodium alginate still presents higher impact values across most categories studied. However, focusing on coating solution step preparation (WSP), which represents the primary difference between the two methods, a noticeable shift in impact distribution is observed. At this stage, the catalyst prepared with TX-100 proves to be significantly more impactful (except for two impact categories related to ‘Freshwater eutrophication’ and ‘Ozone depletion’). This difference is most likely linked to the changes in the dispersant used and the treatment performed to achieve an adequate dispersion of the powder catalysts.

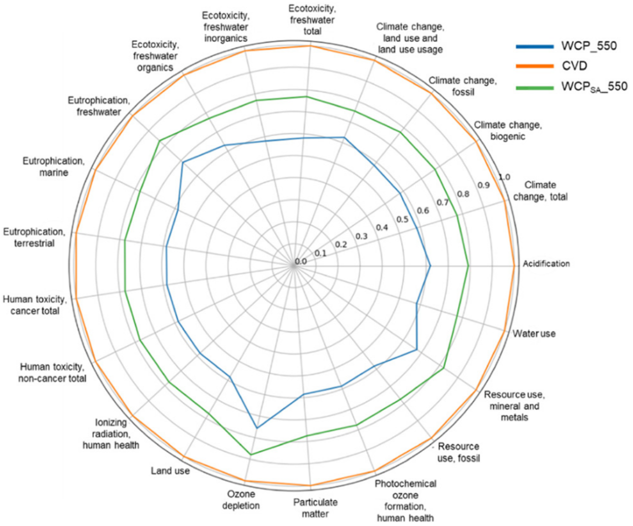

Fig. 6 Normalised radar chart comparing the impacts across impact categories for the three macrostructured catalyst preparations (WCP_550, CVD and WCP$_{SA}$_550).

The use of sodium alginate plays a significant role in reducing the environmental impacts associated with the washcoating catalyst preparation process. One of the key advantages of the method developed with sodium alginate is its versatility—it enables effective and straightforward dispersion of a wide range of carbon-based materials, including carbon composites. In contrast, the use of TX-100 presented certain limitations, requiring modifications to the synthesis procedure to achieve adequate material dispersion.

In cases where dispersion is readily achieved with sodium alginate, the minor differences observed in impact values are primarily linked to the material itself, as the methodology remains consistent. Additionally, in both CNT and CNT:TiO_2 dispersions, the coating solution preparation step using sodium alginate was approximately 30–40% less impactful than when TX-100 was used as a surfactant. Thus, sodium alginate emerges as a promising dispersant to mitigate the environmental impacts of washcoating technology for the synthesis of macrostructured catalysts. However, further optimisations are needed to reduce the energy dependency of the process. The results of these analyses can be found in Fig. S17, presented in the SI.

3.4 Reducing synthesis burden through catalyst regeneration and washcoating solution reuse

Throughout the process, residual materials were identified, which are primarily associated with the coating solution preparation phase. In this context, residual materials refer to raw materials not fully consumed during catalyst synthesis or operation of the integrated system, which have the capacity of being reused, specifically the remaining coating solution after each catalyst batch.

Within the scope of this analysis, macrostructured catalyst regeneration and coating solution reutilization are key strategies for reducing the overall environmental impact of the integrated catalytic system at long term. Previous life-cycle results indicate that catalyst synthesis is the dominant contributor for the registered environmental burdens. Therefore, approaches aimed at extending catalyst lifetime and minimizing the need for new catalyst production were explored. These strategies include catalyst regeneration to restore activity and reuse, as well as the conservation and reuse of the prepared coating solutions to mitigate synthesis-related impacts. At this stage, it is important to highlight that this discussion constitutes a complementary reflective analysis, as the environmental analysis associated with coating solution reuse were not included in the previously presented LCA model.

The present analysis is framed within a capture → treatment → regeneration → reuse framework which promoted a transition from a linear consumption model toward a circular management strategy.[30] In this framework, contaminants are first captured and treated within the integrated catalytic system, after which the materials are regenerated to restore the performance and extend the life-time of a given process. This approach aims to minimize primary resource demand, reduce waste generation, and lower the overall life-cycle environmental burden of the system, thereby enhancing its long-term sustainability.

In the context of the present study, the coating solution (and its intermediate solutions for the preparation of CVD catalysts, such as the Nyacol and nickel solutions) can be stored for reuse in subsequent synthesis processes. To ensure proper reuse of the coating solution, it must undergo treatment in an ultrasonic bath for approximately 1 h prior to use. This process is essential to promote the re-dispersion of any catalyst that may have deposited during storage and to ensure the mixing of all the components within the coating solution.

When comparing the impacts associated with the recovery method (ultrasound treatment) to those associated with the preparation of a new coating solution, for each of the considered scenarios (per liter of coating solution recovered/prepared), it becomes evident that the reuse method accounts for, maximum, approximately 0.13% of the impacts related to the synthesis of a new coating solution in the case of catalysts synthesized using TX-100, and about 0.23% for catalysts synthesized using sodium alginate. In terms of the sustainability of structured catalyst synthesis processes, this represents a clear advantage of washcoating methodologies over the CVD technique. The latter, while being considerably more demanding in terms of both environmental and energy impacts, does not incorporate any step for the reuse or recovery of materials to offset the impacts associated with synthesizing a new batch of catalysts.

In what concerns the solution containing the metallic precursors ($PdCl_2$ and $Cu(NO_3)_2{\cdot}3H_2O$ used for the bimetallic catalysts synthesis), those are not susceptible of being recovered or reused and will therefore be discarded through appropriate waste management procedures. As these metals are linked to energy-intensive and costly extraction/purification processes, exploring alternative strategies for their recovery from solution could be valuable. Nevertheless, such recovery methods often involve significant environmental impacts as well.[74] However, the investigation of these methodologies falls beyond the scope of the present work and it will not be further addressed in this discussion.

Either way, based on the high environmental impact associated with the synthesis of new catalysts and considering the significant drawbacks associated with the disposal of metallic precursors solutions used in catalyst preparation, it is evident that extending the lifetime of the catalysts through the application of regeneration processes emerges as a viable alternative.

In previous studies, regeneration methodologies were investigated, focused on the application of thermal treatments similar to those used during the synthesis of the bimetallic catalysts (200 °C, 1 h under N_2 followed by 3 h under H_2, or alternatively just 1 h under N_2).[75] The results demonstrated a recovery of catalytic activity ranging from approximately 60% to 80%, depending on the catalyst studied, for catalysts that had experienced deactivation levels between 40% and 70%. For ozonation catalyst, which consisted in metal-free carbon catalysts, the considered regeneration methodology was 1 h under N_2 flow until 500 °C.[76]

Through the performed analysis, and considering the three studied scenarios separately, regeneration procedures accounted for a substantial portion of the total environmental impact for the ozonation catalysts: up to 94% in Scenario 1, 80% for Scenario 2 and 75% in Scenario 3. This is consistent with the data related to the synthesis of these catalysts, where the thermal treatment step represents the largest contributor to the associated environmental impact due to its high energy demand. In contrast, the regeneration impact associated with the hydrogenation catalysts, is significantly lower, accounting for a maximum of 30% in the case of Scenario 1, 15% in Scenario 2 and 26% in Scenario 3. Despite involving a 4 h thermal treatment, implementing regeneration remains environmentally preferable to producing new macrostructured catalysts. This is largely because the metal phase impregnation step, during bimetallic macrostructured catalysts synthesis greatly contributes to the total impact, primarily due to the energy intensive and resource-demanding metal precursors used. Avoiding or optimizing this stage could, therefore, substantially reduce overall impacts.

Additionally, considering that the integrated system explored in this study is intended as a complementary solution for drinking water treatment, high organic and inorganic loads are not expected. Therefore, significant catalyst deactivation is unlikely, and when it does occur, it could be effectively mitigated through the proposed regeneration process.

3.5 Life cycle analysis of the integrated treatment system

The applied integrated system consists of a self-developed catalytic setup. The associated impacts can be classified into two main categories: (i) the construction of the integrated system (BM phase – Building Materials) and (ii) its operation (RE phase – Reaction Experiments). The operation phase encompasses all processes involved in the 30 hour water treatment reaction, whereas the construction phase refers to the use of all raw materials for system assembly. Overall, as shown in Fig. S18, the impact of applying the integrated system for water treatment represents only a very small fraction of the total impact. The synthesis of the macrostructured catalysts is by far the largest contributor to the treatment process impact across nearly all impact categories. Therefore, despite the relevance of the topic, the discussion of the impacts associated with the construction and operation of the system is provided in detail in the SI.

3.6 Sensitivity analysis

As previously discussed, for the developed catalytic treatment, electricity plays an important role in the total impacts generated by the overall process. Electricity mix strongly varies between different countries (even within Europe), and these changes are reflected in the total impact of the process. Fig. 7 presents a heatmap showing the variation, in each of the considered impact categories, regarding the application of different electricity grids. This heatmap was constructed using the impact values obtained with Portugal's energy grid as comparison (the grid considered throughout the entire life cycle analysis implemented in this section).

Considering the previously presented results, it is possible to observe a significant variation, regarding the impacts associated with energy usage, depending on the energy mix of the grids utilized.

Overall, there was a clear improvement in all three scenarios when altering the energy grid used. For the energy grids of Spain and France, the results obtained, although more advantageous (as they overall showed a reduction in impacts for most of the categories considered), were quite similar to those obtained for the Portuguese energy grid. However, in this context, it is necessary to highlight the sharp increases in impacts associated with the category of 'Ionising Radiation, human health' caused by the different grids.

The significant increase in impacts associated with the ionising radiation category when switching from the Portuguese energy grid to the Spanish, French, or MIX Europe grids can be attributed to the substantial reliance on nuclear power within these grids. In France, nuclear energy dominates the electricity sector, accounting for around 70% of total production in 2024.[77] Nuclear power generation, while associated with low indirect greenhouse gas emissions, is linked to ionising radiation due to the extraction, processing, and disposal of radioactive materials, as well as the operation of nuclear reactors.[78,79] Similarly, in Spain, while over 82% of power

	Scenario 1					Scenario 2					Scenario 3				
	Spain	France	MIX Europe	MIX Europe | solar	MIX Europe | wind	Spain	France	MIX Europe	MIX Europe | solar	MIX Europe | wind	Spain	France	MIX Europe	MIX Europe | solar	MIX Europe | wind
Water use	-5.2	-92.5	-84.4	-98.3	-99.2	-5.2	-92.5	-81.6	-98.4	-99.2	-12.0	-93.1	-82.9	-98.5	-99.3
Resource use, mineral and metals	10.5	-32.3	-8.3	415.7	83.6	12.3	-37.9	0.0	487.7	98.0	-86.9	-92.4	-88.2	-35.6	-77.6
Resource use, fossils	45.9	92.4	61.9	-90.7	-95.1	45.9	92.5	51.5	-90.8	-95.2	35.6	76.9	40.7	-91.4	-95.5
Photochemical ozone formation, human health	-31.2	-65.9	-27.1	-75.5	-88.7	-31.5	-66.6	-13.8	-76.3	-89.6	-36.0	-68.8	-19.7	-77.4	-89.7
Particulate matter	-26.3	-57.5	-5.3	-32.0	-82.2	-26.4	-57.8	12.7	-32.1	-82.7	-99.5	-99.7	-99.2	-99.5	-99.9
Ozone depletion	14.6	-1.2	14.7	806.8	-24.3	18.0	-1.4	25.0	999.3	-30.0	-100.0	-100.0	-100.0	-100.0	-100.0
Land use	-65.4	-79.5	-52.2	-97.2	-98.3	-65.4	-79.5	-42.8	-97.2	-98.3	-67.9	-81.1	-46.9	-97.4	-98.4
Ionising radiation, human health	609.9	5191.5	2281.0	-93.3	-90.6	612.3	5212.3	1281.1	-93.7	-90.9	564.4	4803.9	1188.1	-93.9	-91.3
Human toxicity, non-cancer total	-22.0	-67.3	21.3	-37.1	-84.1	-22.1	-67.5	52.1	-37.2	-84.3	-99.9	-99.9	-99.7	-99.9	-100.0
Human toxicity, cancer total	-20.6	-63.0	-44.3	-87.3	87.0	-21.0	-64.0	-38.5	-89.0	88.5	-100.0	-100.0	-100.0	-100.0	-99.9
Eutrophication, terrestrial	-32.6	-63.1	-19.4	-76.0	-89.1	-32.8	-63.6	-4.3	-76.6	-89.8	-37.3	-66.1	-10.8	-77.8	-90.0
Eutrophication, marine	-32.5	-60.6	-18.0	-76.3	-89.8	-32.6	-61.0	-3.3	-76.7	-90.3	-37.3	-63.9	-10.2	-78.1	-90.6
Eutrophication, freshwater	-26.7	-28.4	10.8	-53.2	-55.9	-32.3	-34.4	29.4	-64.4	-67.6	-49.5	-56.5	-10.3	-70.0	-72.0
Ecotoxicity, freshwater organics	-11.1	-31.7	1.0	-49.2	-73.8	-12.1	-34.4	13.4	-53.4	-80.1	-16.3	-39.3	7.7	-55.2	-80.3
Ecotoxicity, freshwater inorganics	2.5	-10.3	136.1	-79.6	-89.1	2.4	-10.2	186.2	-79.4	-88.8	-4.7	-17.4	167.0	-81.2	-90.0
Ecotoxicity, freshwater total	2.3	-10.5	134.6	-79.3	-88.9	2.3	-10.5	184.4	-79.1	-88.7	-4.8	-17.6	165.3	-80.9	-89.9
Climate change, land use and land use change	-37.8	-63.4	-21.5	-33.1	-62.9	-40.6	-68.2	-7.6	-35.5	-67.7	-46.0	-73.0	-14.7	-41.1	-71.6
Climate change, fossil	-14.7	-72.7	-13.5	-88.1	-94.7	-14.8	-72.8	6.9	-88.2	-94.8	-20.8	-74.9	-0.7	-89.0	-95.1
Climate change, biogenic	-61.4	1.2	173.5	-94.4	-96.1	-61.4	1.2	233.0	-94.4	-96.1	-64.2	-7.0	209.0	-94.8	-96.3
Climate change, total	-14.9	-72.5	-13.0	-88.1	-94.7	-14.9	-72.5	7.6	-88.2	-94.8	-20.9	-74.7	-0.1	-89.0	-95.1
Acidification	-22.6	-49.6	5.0	-57.0	-74.7	-24.0	-52.7	25.3	-60.5	-79.2	-28.0	-54.5	16.9	-61.2	-78.2

Fig. 7 Heat map showing the percentage differences in the system's overall impact when comparing various energy grids from European countries (Spain, France, and a European average, including solar and wind power) to Portugal's electricity grid used throughout the study.

comes from low-carbon sources, nuclear energy remains a significant component of the mix (around 20%), alongside wind (23%), hydropower (19%), and solar energy (18%).[80]

For the MIX Europe grid, nuclear power contributes over 20%, combined with a substantial share of fossil fuels (38.6%), including gas and coal.[81] Fossil fuel combustion can, also, contribute to ionising radiation impacts by releasing naturally occurring radioactive materials found in coal and other fuels, further amplifying this impact category.

In contrast, the Portuguese energy grid likely relies less on nuclear energy, thus presenting lower contributions to ionising radiation impacts. The higher nuclear share in the French, Spanish, and MIX Europe grids inherently increases the associated impacts in this category, as nuclear power has a distinct and pronounced effect on ionising radiation indicators, despite its advantages in reducing greenhouse gas emissions.

An additional increase was detected for 'Resource use, fossils' category, which is mainly attributed to the notable presence of fossil fuels in the Spanish, French, and MIX Europe grids. This is directly associated with the lifecycle processes involved in the extraction, processing, and combustion of fossil fuels. The "Resource use, fossils" impact category quantifies the resource depletion by assessing the consumption of these non-renewable resources relative to their limited availability.[82]

For instance, the MIX Europe grid includes 38.6% fossil fuels, with gas (19.6%) and coal (15.8%) being the primary contributors. Fossil fuel use involves significant resource extraction and consumption, leading to higher impacts in the "Resource use, fossils" category. While Spain and France have cleaner energy mixes compared to MIX Europe, they still depend on fossil fuels to a certain extent, which likely exceeds the reliance on such sources in the Portuguese grid. This shift to grids with higher fossil fuel contributions explains the observed increase in impacts in this category.

Additionally, the high reliance on fossil fuels and nuclear energy, associated with the MIX Europe grid may be one of the principal reasons for the evident increases in the impact for the categories of 'Climate change, biogenic' and 'Ecotoxicity, freshwater'. The combustion of fossil fuels like coal and gas, which comprises 38.6% of the MIX Europe grid, releases significant amounts of CO_2 and methane, contributing to global warming and increasing impacts in the 'Climate change, biogenic' category.[83] Additionally, the extraction and processing of fossil fuels release toxic pollutants, such as heavy metals and other contaminants, which affect freshwater ecosystems. Nuclear energy, which accounts for over 20% of the grid, also poses risks of radioactive contamination of water.[84] In contrast, the Portuguese grid, with lower reliance on fossil fuels and nuclear power, results in lower impacts in these categories.

The most significant differences were observed when considering energy grids focused on the use of renewable energies (MIX Europe, solar, and wind), with the most impact reductions in the range of 80–90%.

In conclusion, the impact of the catalytic treatment process is significantly influenced by the energy mix of the electricity grids used. Switching from Portugal's grid to those of Spain, France, or a European mix often results in increased impacts, especially in categories like ionising radiation and resource use, due to a higher reliance on nuclear and fossil fuels. However, grids with a higher share of renewable energy, such as wind and solar, show substantial reductions in environmental impacts, highlighting the importance of cleaner energy sources for minimizing negative outcomes.

4. Conclusions

This study demonstrates the feasibility and environmental benefits of using washcoated carbon-based macrostructured catalysts for drinking water treatment, offering a sustainable alternative to conventional CVD-based synthesis. All catalyst configurations achieved effective degradation of organic micropollutants and nitrate/bromate levels within regulatory limits.

Life Cycle Assessment revealed that thermal treatments and precious metal use (especially palladium) are major environmental hotspots. Washcoated systems, particularly those using TX-100, showed significantly lower overall environmental burdens—approximately 30–50% less than CVD catalysts. While sodium alginate offers a greener alternative, its synthesis introduces specific environmental trade-offs, particularly in energy demand and ozone depletion.

Optimizing thermal treatment steps, energy sources, and metal nanoparticle synthesis can further reduce their impacts. Future work should explore process-intensification strategies aimed at reducing the environmental impact associated with catalyst thermal treatment, which was identified as the main hotspot in the present life-cycle assessment. Potential approaches may include alternative heating methods (*e.g.*, rapid thermal, infrared or microwave heating), shortened thermal profiles or modified binder chemistries that could allow lower-temperature curing while maintaining catalyst stability and catalytic performance. These findings will support the development of more sustainable water treatment materials aligned with SSbD criteria and offer guidance for upscaling eco-innovative catalyst technologies.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data that support the findings of this study are available in the supplementary information (SI). Supplementary information is available. See DOI: https://doi.org/10.1039/d5gc06949a.

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