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A multiple case study of brine effluents

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Application of LCA and LCC in the early stages of wastewater treatment design: A multiple case study of brine effluents

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1. Introduction

The circular economy and industrial symbiosis are recognised as ways to address sustainability challenges by focusing on closing resource loops through recovery, reuse, remanufacturing and recycling of resources (Niero and Kalbar, 2019; Elia et al., 2017; Martin and Harris, 2018). One of the most common wastewater sources from industry is brine, a high concentration of salt between 35,000 and 260,000 ppm, which can also contain valuable elements and compounds. The chemical industry in Europe alone produces 1.15 million tonnes/year (Xevgenos et al., 2018) and its disposal to water bodies can result in environmental impacts to the local flora and fauna (European Commission, 2014). In addition, industry accounts for 22% of the global water demand and hence reuse and recovery can play a significant role in reducing water stress. Brine is produced in industry by adding salt to water in order to regenerate resins used in chemical processes, water treatment or textile dying (Roskill, 2018). Its treatment is challenging and risks creating alternative impacts. For instance, traditional evaporation requires large amounts of energy and being subject to fouling (Xevgenos et al., 2016). Three promising technologies developed within the framework of EU-funded projects are the forward-feed evaporator (FF-MED) for brine concentration, and Eutectic Freeze Crystallisation (EFC) with ion exchange membranes (CRIEM) for brine crystallisation (Xevgenos et al., 2016). Combining these technologies in different configurations with established technology units such as reverse osmosis and nanofiltration has the potential to treat brine and recover valuable components such as magnesium and calcium salts.

However, quantifying the impacts of potential treatment systems at the design stage is also challenging as the available experimental data may be inadequate for methods such as Life Cycle Assessment (LCA). LCA is the leading method to quantify the environmental impacts of...
Nomenclature

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
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<tbody>
<tr>
<td>CAPEX</td>
<td>Capital Expenditure</td>
</tr>
<tr>
<td>CrEM</td>
<td>Crystallisation with ion exchange membranes</td>
</tr>
<tr>
<td>DWP</td>
<td>Demineralised water plant</td>
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<tr>
<td>ED</td>
<td>Electrolysis</td>
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<tr>
<td>EFC</td>
<td>Eutectic Freeze Crystallisation</td>
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<tr>
<td>eLCC</td>
<td>Environmental life cycle costing</td>
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<tr>
<td>fLCC</td>
<td>Financial life cycle costing</td>
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<tr>
<td>FF-MED</td>
<td>Forward-feed evaporator</td>
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<tr>
<td>IEX</td>
<td>Ion exchange</td>
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<td>LCA</td>
<td>Life cycle assessment</td>
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<td>LCC</td>
<td>Life cycle costing</td>
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<tr>
<td>MD</td>
<td>Membrane distillation</td>
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<tr>
<td>MED</td>
<td>Multi-effect Distillation</td>
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<tr>
<td>NF</td>
<td>Nanofiltration</td>
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<tr>
<td>OPEX</td>
<td>Operating Expenditure</td>
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<tr>
<td>RCE</td>
<td>Remote Component Environment</td>
</tr>
<tr>
<td>RO</td>
<td>Reverse Osmosis</td>
</tr>
<tr>
<td>TOC</td>
<td>Total organic carbon</td>
</tr>
<tr>
<td>UA</td>
<td>Unified Approach</td>
</tr>
</tbody>
</table>

products, processes and services and can be used to aid design (Baumann and Tillman, 2004). Integrating LCA early in the technology development has the potential to optimise the development and increase the understanding of design implications and compromises on the environmental performance (Buyle et al., 2019). Recent literature has highlighted prospective LCA as an approach to model the future full-scale performance of technology at an early stage of development (Arvidsson et al., 2018; Bartolozzi et al., 2019). Other similar terms used are anticipatory (Wender et al., 2014) or ex-ante LCA’s (Buyle et al., 2019; Cucurachi et al., 2018) predictive LCA (Karka et al., 2019), “process simulation based LCA” (Rathnayake et al., 2018), eco-design and product design evaluation (Suhariyanto et al., 2018). Prospective LCA’s have the common need to utilise a range of data to model the future scenario of the foreground system such as scientific articles, patents, expert interviews, lab results and process simulations (Arvidsson et al., 2018). The major methodological challenges in conducting LCAs of emerging technologies are comparability, data availability and quality, scaling, and uncertainty (Monti et al., 2019).

To quantify costs, Life Cycle Costing (LCC) is a related accounting technique that calculates all of the costs that an owner or producer of an asset will incur over its life cycle, including both capital expenditure (CAPEX) and operating expenditure (OPEX) (Swarr et al., 2011). LCC has become aligned with LCA and developed into a code of practice (Hunkeler, 2008; Swarr et al., 2011). Four types of LCC are generally distinguished to address financial (fLCC) (conventional), environmental (eLCC), full-environmental (fLCC) and societal (sLCC) (Pinkbeiner et al., 2010). Whereas fLCC focuses on a specific actor and internal costs connected to a specific product (Hoogmartens et al., 2014), eLCC assesses all costs incurred by actors linked to a specific functional unit and includes internal and external costs (Kambanou and Sakao, 2020; Hunkeler et al., 2008). fLCC builds on this cost structure, but with monetised environmental impacts, and sLCC extends this to also include costs borne by society now and in the future (Hoogmartens et al., 2014). These terms are not always used consistently in literature, with Gluch and Baumann (2004) identifying more than ten LCC techniques that address environmental concerns at varying degrees.

In relation to brine and wastewater treatment, LCA has been used to highlight the dominant role of energy demand and compare water supply options (Zhou et al., 2013) or disposal and reuse alternatives (Muñoz et al., 2008; Meneses et al., 2010). Fernández-Torres et al. (2012) compared eutectic freeze crystallisation and evaporative crystallisation for the treatment of saline water. There are several instances where prospective LCA has been used to assess wastewater treatment. Fang et al. (2016) use LCA to aid the development of wastewater biotechnology for resource recovery using simulation of the full scale. Meanwhile, Muñoz et al., 2019 theoretically scale-up a pilot plant for the integration of a solar assisted heat pump to recovery energy from wastewater, using both LCA and LCC for assessment. By comparing the integrated system to a conventional treatment plant, they demonstrated clear environmental benefits in all impact categories and lower costs. In the design of thermal cracking technology, Gear et al. (2018) LCA was combined with ternary diagrams that provide guidance for design, which avoided the need for high quality data. Other studies using prospective LCA in design include the recovery of metals using bioleaching (Villares et al., 2016), municipal wastewater treatment (Baresel et al., 2015), microalgae treatment and valorisation of by-products (Siez et al., 2015) and volatile fatty acid production from dairy wastewater finding (Elginoz et al., 2020).

Other papers that combine LCA and LCC include the economic feasibility of 22 wastewater treatment plants in Spain (Lorenzo-Toja et al., 2016), anaerobic membrane bioreactor for resource recovery from domestic wastewater (Harclerode et al., 2020) and strategies for wastewater management systems (Abdallah et al., 2020). The studies show that chemical and energy use are two key factors related to environmental impact of the potential wastewater treatment systems (Harclerode et al., 2020; Elginoz et al., 2020). Some authors have denoted a lack of consistency when LCC and LCA are jointly applied (Bachmann, 2012; Guinée, 2016). Further challenges include the lack of a standardized methodology for the allocation of LCC cost factors and obtaining reliable and adequate data from companies with confidentiality concerns (Heijungs et al., 2012). Other research is continuing to develop a framework to link the approach of LCA and LCC and improve the consistency of their joint application (Hoogmartens et al., 2014).

Within the EU Zero Brine project three wastewater treatment units (FF-MED, EFC and CrEM) are being developed from Technology Readiness Level (TRL) 5 to 9. These are combined in four separate case studies, with established unit technologies (e.g. reverse osmosis) to form the case specific water treatment and recovery systems, herein referred to as the Zero Brine system (or ZB system; note that the textile ZB system does not use any of the ZB units under development). This paper presents the use of prospective LCA and LCC at the first stage of a three staged analysis in the development of the ZB systems, with the objectives to:

- Better understand the ZB systems, environmental implications and challenges in data limitations.
- Inform the design process by highlighting hotspots and key issues.
- Develop a project based unified approach for LCA and LCC to increase consistency across three teams of researchers, for subsequent assessment in the development process.

2. Materials and methods

2.1. Case study descriptions

The Zero Brine project consists of four case studies that utilise various configurations of established technologies such as reverse osmosis (RO) and nanofiltration (NF), combined with three technologies developed within the project: EFC, CrEM and FF-MED evaporator. The case studies and ZB systems are described in the following subsections.

2.1.1. Demineralised water plant in the Netherlands

The demineralised water plant (DWP) based in Rotterdam, produces demineralised water for the local chemical industry. Approximately 2.5 million m$^3$ year of brine is produced from the use of ion-exchange (IEX) and RO technology and is discharged into the local sea. The IEX requires large volumes of vacuum salt for regeneration, which can potentially be...
recovered with the proposed ZB systems, along with organic carbon material and clean water. Two separate ZB systems have been designed to treat the two brine discharges of the DWP that derive from the IEX and RO. The IEX configuration is shown in Fig. 1 A. The concentrate from the NF undergoes a double crystallisation stage to recover the by-product salts. Evaporation recovers the NaCl and clean water from the NF permeate and MC effluent. The configuration in Fig. 1B is designed to recover clean water and salts from the RO brine using IEX, NF, EFC, RO and evaporation to recover salts and clean water. The by-products are reused internally with the clean water replacing lake water input and the NaCl used for regeneration of the IEX units.

2.1.2. Coal mine discharge in Poland

The coal mine case study focuses on saline water discharged from a Polish coal mine. The current treatment is limited to a settling pond to remove the large suspended solids, followed by dilution with industrial wastewater. This ensures the discharge conforms to regulatory thresholds to enable the discharge to a nearby river. A main driver for improved treatment is regulatory pressure to decrease the salt discharge. The proposed ZB system is illustrated in Fig. 2, showing the two-stage NF, RO, electrodialysis and crystallisation, to produce clean water. In addition, the system aims to recover the by-products magnesium hydroxide (Mg(OH)_2), salt and gypsum. The clean water will be reused in the mining operations whilst the recovered compounds will be sold externally.

2.1.3. Textile industry in Turkey

The textile case study located at Büyükkarşışiran- Lüleburgaz, Kırklareli, examines the integration of a ZB system into a textile manufacturing plant to treat brine effluent and recover by-products for reuse. Brine is produced due to the use of salt in both the dyeing (325 tons/year of refined salt) and water softening processes (275 tons/year). The proposed ZB system is designed to treat brine that derives from the RO unit of the current wastewater treatment plant, as shown in Fig. 3. Both cationic and anionic resins are used in the IEX, followed by ozonation to oxidise the remaining carbon material, and RO to recover clean water and concentrated brine. Both the water and concentrated brine will be reused in the textile plant operations.

2.1.4. Silica industry in Spain

The silica case study located in Zaragoza, involves a chemical company producing silica derivatives which results in approximately 438,000 m^3/year of brine effluent. This is currently sent to the municipal wastewater treatment plant before being discharged to the local river. The proposed ZB system shown in Fig. 4 consists of physicochemical pretreatment (including pH modification, chemical addition and sand filtering), NF and EFC. Regenerated RO membranes are used in the NF to maximize the recovery and modulate the rejection. The recovered clean water will be reused in the silica production (greatly reducing extracted groundwater use), and sodium sulphate (Na_2SO_4) will be marketed externally.

2.2. LCA and LCC methodology

The LCA methodology used herein is consistent with ISO 14040 and 14044 standards (ISO14044, 2006; ISO14040, 2006) and the International Reference Life Cycle Data System (ILCD) (JRC-IES, 2010).
Fig. 4. ZB system for precipitated silica plant, treatment of saline wastewater (brine). Source: Tsalidis et al. (2020).

The main differences applied to the case studies are outlined in Table 1. Three separate LCA teams used Simapro version 8 software to model the life cycles. The coal mine and textile case studies were modelled by the same team, whereas the DWP and chemical production were assessed by separate teams. An environmental LCC of the ZB system was conducted by accounting for the life cycle costs associated with the main actor (i.e. the company where the systems are located) as described by Hunkeler et al. (2008). The LCA and LCC were aligned using the same functional unit, allocation and system boundaries, etc. At this stage in the development process, partial consequential LCC is used (full consequential is used by Mutioz et al., 2019 in a similar study) as our aim is to identify the environmentally relevant physical flows in and out of the life cycle (Finnveden et al. 2009). Systems expansion is used to incorporate the reduction of overall environmental burdens that result from the recovery of by-products and clean water. It is therefore assumed that they reach markets and displace equal products.

As discussed in the introduction, this paper represents the first stage of a 3-stage process that uses LCA and LCC in the design and development of the ZB systems. A project unified approach is developed to guide this process and bring increased consistency and robustness (this is discussed in section 2.3). The aims of the initial stage and this paper were presented in section 1, whilst the eventual and final goal of the project (and unified approach) is to compare each of the ZB systems with the reference case. However, due to data limitations, the only case study where a comparison with the reference system was possible is the DWP. The coal mine study focuses on four different configurations of using either single or two-stage NF, with recycling of the NF retentate and ED dilute. Whereas, the textile and silica case studies provide an assessment of the ZB systems and a hotspot analysis, supplemented with further analysis of the use of RO membranes for the latter case.

2.2.1. Data collection for life cycle assessment

Background data was obtained from Ecoinvent v3.2 and primary data was provided in most cases by the technology providers who are partners in the project. Additional data is based on bench scale data and simulation using PHREEQC software. PHREEQC is simulation software capable of calculating a wide range of geochemical reactions between water and minerals, ion exchangers, solid solutions, and gases (Charlton and Parkhurst, 2011). The technology units used in the different cases studies and the technology providers from which data was obtained are shown in Table 2.

For the DWP, site data on inflows for Site 1 and Site 2 are based on three separate LCA teams used Simapro version 8 software to model the life cycles.

Table 1

<table>
<thead>
<tr>
<th>LCA aspect</th>
<th>Case Study</th>
</tr>
</thead>
<tbody>
<tr>
<td>Goal (initial LCA stage):</td>
<td>Assess performance of the DWP with and without ZB system.</td>
</tr>
<tr>
<td>Functional unit Allocation</td>
<td>Treatment of 1 m³ of total brine.</td>
</tr>
<tr>
<td>System boundaries</td>
<td>DWP, with and without ZB system, including all inputs and outputs. (see section 2.1)</td>
</tr>
<tr>
<td>Data quality</td>
<td>Primary data obtained from bench scale tests for ZB system.</td>
</tr>
</tbody>
</table>

Table 2

<table>
<thead>
<tr>
<th>ZB Process/input</th>
<th>Relevant to which case study</th>
<th>Technology provider/data source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ion-exchange</td>
<td>DWP, Textile plant</td>
<td>Lenntech</td>
</tr>
<tr>
<td>Nanofiltration</td>
<td>DWP Site 1, Coal mine</td>
<td>Lenntech</td>
</tr>
<tr>
<td>Reverse osmosis unit</td>
<td>DWP, Coal mine</td>
<td>UNIPA</td>
</tr>
<tr>
<td>Membrane crystallisation</td>
<td>DWP, Coal mine</td>
<td>UNIPA</td>
</tr>
<tr>
<td>Evaporator unit</td>
<td>DWP</td>
<td>NTUA/Sealeau</td>
</tr>
<tr>
<td>TOC removal unit</td>
<td>DWP</td>
<td>Arvia</td>
</tr>
<tr>
<td>Physio-chemical pretreatment</td>
<td>Silica plant</td>
<td>TYPISA -primary data</td>
</tr>
<tr>
<td>Membrane regeneration</td>
<td>Silica plant</td>
<td>TYPISA and Eurecat-primary data</td>
</tr>
<tr>
<td>EFC</td>
<td>DWP, Silica plant.</td>
<td>TU Delfi – primary</td>
</tr>
<tr>
<td>Reverse osmosis/UF/MF</td>
<td>DWP, Coal mine</td>
<td>Lenntech</td>
</tr>
<tr>
<td>Electrodialysis</td>
<td>Coal mine</td>
<td>Fujifilm/mega</td>
</tr>
<tr>
<td>Chemical types and quantities</td>
<td>All.</td>
<td>Project partners, technology providers and simulation in PHREEQC</td>
</tr>
</tbody>
</table>

Table 1 Overview of the main LCA components, choices and differences for the initial evaluation.

Impact categories

Global warming potential (GWP); Acidification; Eutrophication; Freshwater aquatic toxicity; Resource depletion.

*DWP = demineralised water plant. IEX = ionic exchange unit.*
results from bench scale tests (for NF and membrane crystallisation) and PHREEQC simulations, respectively. In the coal mine and textile studies, foreground data is derived from bench scale experiments combined with PHREEQC simulation. For the silica plant, primary data was derived from the technology developer TYPSA for membrane generation. IQE (the silica plant operator) provided silica and wastewater production quantities (primary data). Secondary data was obtained from IQE’s Environmental Operation License (publicly available) (PRTR, 2019). In addition, “Large Volume Inorganic Chemicals - Solids and Others industry” BREF document (EC, 2007) has also been consulted.

2.3.2. Life Cycle Costing

This paper is limited to a Life Cycle Costing (LCC) of the main actor and includes costs related to acquisition (CAPEX), operational costs, such as consumption of energy and other resources, maintenance and repair costs (OPEX), and end of life costs, such as collection and recycling costs. In addition, revenues from the recovered by-products are included as negative costs. For the DWP and silica plant the life of the ZB systems are 20 years. We do not apply any discount rates and only include cost by the main actor involved. Nonetheless, in the next stages the project aims to perform a full environmental LCC and include both internal costs and monetised external environmental costs, such as those linked to the contribution to global warming.

At this stage the approach varies to some extent across the individual case studies due to data availability, as shown in Table 3. For example, the coal mine and textile case studies only include OPEX. Inclusion of aspects such as replacement of equipment, maintenance and end-of-life costs are only included where stipulated. Costs are derived from the literature, technology developers and the technology providers involved. Data collection for the technology units is derived from estimates and calculations of the individual technology providers. The staff costs were estimated from company reports for the DWP and silica plant, and consumables were derived from industrial supplier websites (see supplementary material 1 for further information).

2.3. Development of a project unified approach

The LCA and LCC’s are performed by three teams of researchers and coordinated by a fourth. Therefore, a unified approach (UA) is key to bring consistency to the approach and assessments. The approach is prospective with the intention of aiding the design and development process. Therefore, the UA embodies a staged approach to apply LCA and LCC at different phases of development. The UA is seen as a learning process throughout the project, with continuous improvement until the final analysis and has a three staged approach as shown in Fig. 5. This paper represents the first stage, with the development of the initial unified approach and an initial LCA and LCC. Each of the three stages are outlined below.

2.3.1. Stage 1: initial unified approach and LCA and LCC

The objectives of this stage are to: i) agree on the overall approach and main LCA components to align, ii) developing an understanding of the case studies, technologies, data and simulation challenges; and iii) provide an initial LCA and LCC and identify the main hotspots and iv) identify preliminary design implications and where additional efforts to improve data quality might be required.

The methodology started with several workshops with the LCA coordinator and the three LCA teams. This resulted in agreeing on the three-stage approach in the development of the unified approach and to provide continued analysis to aid design and development process of the ZB systems. Agreement was also reached on the functional units, system boundaries and to compare the ZB systems to the individual reference case. The LCA approach can be described as partial consequential LCA, as systems expansion is used to provide credits for avoided products that result from the recovered by-products. This was followed by design workshops for each case study that involved all key stakeholders: the LCA teams, LCA coordinator, technology developers/providers and representatives from the participating companies. The initial LCA and LCC were then performed using initial system calculations and simulations, supplemented with validation or modification from bench scale results. The initial LCC’s do not involve externalities, but focus on obtaining data and the identification of hotspots.

2.3.2. Stage 2: refined unified approach and second LCA and LCC

The objective of the second stage is to improve the LCA and LCC using improved data and increasing the scope so that the reference systems are compared in all cases. This will involve using empirical data from pilot plants (increasing the representativeness from bench scale data) to improve full-scale modelling. Modelling is based on simulation and increasingly it is anticipated that remote component environment (RCE) software will be used. The RCE software is an open source software that can be used to model complex systems. In another work package of the Zero Brine project the case studies have been implemented in Python and integrated into the RCE simulation platform to enable techno-economic analysis (Miciari et al., 2020). The LCC is expanded to include environmental externalities with consistency across the case studies and no gaps in scope.

2.3.3. Stage 3: final unified approach and LCA and LCC

The final sustainability evaluation aims to provide a robust LCA and LCC that makes a full comparison with the current reference systems of the case studies. The evaluation will be based on updated and optimised data from the pilot and demo site (where applicable). Lessons from the design process will be documented and discussed in a final report. The Unified Approach will be finalised so that LCA and LCC can be robustly applied in the continued development of the case study systems, as well as in future replication efforts where ZB systems are developed to treat new industrial brine sources.

3. Results

This section presents the results of the four case studies, identifying key hotspots and challenges for the development of the ZB systems.

3.1. DWP in the Netherlands

The initial LCA analysis for the DWP, shown in Fig. 6, suggests that the environmental performance of the ZB system has a higher impact than the current system for the chosen impact categories. The GHG emissions per 1 m³ of generated brine for the DWP is 3.5 kgCO₂eq, but this rises to 15.6 kgCO₂eq with the ZB system. Mineral and resource depletion is also significantly higher in the ZB system. Fig. 7 shows that electricity use forms a large part of the GWP, particularly in the production of chemicals (related to the sodium hydroxide used in the TOC removal process), but also due to an increase in the plant’s electricity use. The impact of sulphuric acid is also significant for the acidification impacts (also resulting from upstream processes). Fig. 7 also illustrates
**Fig. 5.** Flowchart showing staged development and assessment approach.

**Fig. 6.** Percentage comparison (and quantities in table) of impacts for 1 m$^3$ of brine for selected representative impact categories, with absolute values in table.

**Fig. 7.** Contribution analysis of climate change of ZB system.
that the credits derived from the by-product recovery are small compared to the impacts from the ZB systems. The increase in impact will be carefully considered alongside the environmental benefits of reduced salt emissions in the next stages of analysis and technology development. Onsite impact tests on benthic marine species are ongoing and will inform the final evaluation.

The economic performance of the DWP is compared to ZB system and DWP together in Fig. 8. It suggests a significant increase in costs with the ZB system, rising from 2.8 €/m³ to 10.3 €/m³, with raw materials accounting for a large share. The total costs of raw materials per m³ of brine rises from 0.15 €/m³ to 4.06 €/m³ with the ZB system. This is primarily due to the use of sodium hydroxide, sulphuric acid and anti-scalant (Vitec 4000) in the ZB system at Site II. Meanwhile, the LCC shows that the costs of energy are insignificant.

3.2. Coal mine discharge in Poland

The LCA analysis for the coal mine, shown in Fig. 9, provides mixed results for the four configurations of the technology units, with none of the configurations performing best for all categories. However, on average the configurations 1 and 2 perform slightly better. (The configurations are as follows: 1) Two stage NF, with electrodialysis diluate recycled to NF; and 2) Single-stage NF, with ED diluate recycled to NF; further information and results on other impact categories are provided in the supplementary material 1). Configuration 4 (single stage nanofiltration, 75% of NF retentate is recycled, ED diluate is recycled back before the RO) is the poorest performer for several categories, but one of the best for climate change. In addition, due to the high performance in recovery of by-products, the configuration produces the highest credit for abiotic resources.

Most of the environmental impacts of the system configurations resulted from the energy consumption of the technology units and the use of dolime in the magnesium recovery process. Further analysis in Fig. 10 illustrates that the recovery of sodium chloride and magnesium hydroxide strongly counteracts the impact of the ZB system for human toxicity, freshwater ecotoxicity and “mineral and fossil resource depletion”. It should be noted that, at the time of analysis, reliable energy consumption data was only available for the complete system and not for the individual unit processes. Total climate change impacts for the configurations 1,2,3 and 4, are 10.2, 10.8, 17.3, and 11.7 kgCO₂e/m³ respectively.

The preliminary LCC results shown in Fig. 11 show that the total costs for the different configurations have a wide range from €5.5/m³ to €42.4/m³. This is highly dependent on the quantities of chemicals used and the by-products recovered. This is particularly dependent on the cost of dolomite and the potential revenue of magnesium hydroxide that could generate between 60 and 287 Euros per m³ (based only on market price and not a packaged and distributed product) if markets were found for the products and high quality was attained.

3.3. Textile industry in Turkey

Fig. 12 shows that the main impact (80–90%) of the ZB system is derived from the reverse osmosis unit for all of the impact categories due to its relatively high energy consumption. However, Fig. 13 shows that the recovery of the brine solution for reuse in the dyeing process (therefore saving mined salt) provides overall benefits for all of the impact categories. However, the transport of recovered materials has not been included as this stage of the assessment. The preliminary LCC results suggest that there will be an overall cost for the ZB system of almost €25/m³ as shown in Fig. 14.

3.4. Silica industry in Spain

The impact of the silica ZB system by unit process, for selected impact categories, is presented in Fig. 15. It shows that the RO consistently accounts for 80% of the total impact across the categories. The physico-chemical treatment is the next highest with 10% of the impacts, followed by the UF with 8%. These are primarily caused during the operation, as opposed to the raw material or production stage and is closely related to the energy consumption. Related analysis highlights that the use of regenerated membranes reduces the impact by 90%
compared to using new RO membranes (please see supplementary material).

The LCC showed that the costs are divided into 20% CAPEX and 80% OPEX, as shown in Fig. 16. The R&D costs associated with the research project contributed 70% of the CAPEX. The main OPEX comes from the labour costs (51%) and energy (20%).

3.5. Sensitivity analysis

For each case study a contribution analysis was performed for the LCA’s and LCC’s, which is a recognised form of sensitivity analysis (Clavreul et al., 2012). This provided an overview of the main variables which contribute to impacts, costs and revenue. Table 4 shows the results of performing an analysis on the sensitivity of each of these on the climate change impact for a 25% increase and decrease of input (similarly performed by Muñoz et al., 2019). It shows that the climate impact is very sensitive to changes in these variables, especially for the textile plant.

Correspondingly, Table 5 shows the sensitivity results for the main contributing variables of the LCC.

It shows that a decrease revenue for the DWP would increase the overall cost from 0.26 to 2.26 Euro/m³, but a 20% increase in revenue leads to a profit of 2.15 Euro/m³. Similarly, it shows that a 25% decrease in the cost of dolomite for the coal mine ZB system would result in a revenue of 18.6 Euro/m³ but a 25% increase would result in a cost of 62.3 Euro/m³. Therefore, there is also a high sensitivity for these variables to the costs of the systems. Only the DWP and silica plant included CAPEX at this stage, which shows that OPEX is more important due to energy and chemical usage.

4. Discussion

The lessons of this paper are three-fold: insights from the LCA and LCC on the performance of the ZB systems, implications for the design process; and lessons for the Unified Approach. These are discussed in the following sections. The use of LCA and LCC together is both complementary and able to provide insights from different perspectives, in agreement with previous research (Calado et al., 2019). The task of aligning the LCA and LCI inventory elements was reasonably straightforward. One challenge however was finding reliable commodity prices and some company costs which are deemed confidential. Although the analysis is based on bench scale data, other research has shown that LCA based on data from laboratory scale experiments, can identify hotspots (Piccinno et al., 2016).

4.1. Performance of the ZB systems

The first stage of applying LCA throughout the development process has highlighted the importance of energy and chemical consumption in the ZB systems. This aligns with other research on wastewater treatment systems e.g. Harclerode et al. (2020) and Elginoz et al. (2020). In addition, it has highlighted that reverse osmosis is a key concern in terms of operational energy use, dominating impacts in the textile and silica plants. The recovery of by-products can potentially compensate for some of the increased impacts of the ZB system but is dependent on the quantities available in the feedwater, as well as the recovered quantity and quality. For instance, in the DWP and coal mine case studies, the
recovery of by-products does not compensate for the impact of the processes (one exception is for resource depletion in the coal mine). However, the recovery of brine solution in the textile plant system does compensate for the impacts of the ZB system, particularly for resource depletion. The silica plant analysis does not consider the by-products at this stage. An important consideration that was not included in the initial evaluation of the DWP and coal mine, was the benefits of stopping brine discharge to the local water courses. Its inclusion is a remaining challenge as LCA datasets do not currently exist. Options being explored are to include data from local environmental impact assessments or assessing via LCC by costing the externalities.

Similarly, the LCC showed that the ZB systems can potentially be cost neutral or profitable depending on the value of the recovered by-products. However, this only occurred for the coal mine case where the recovery of magnesium hydroxide almost covers the cost of the ZB system. The analysis of the four configurations showed that increased

Fig. 11. LCC for the coal mine by commodity showing total costs of the configurations. RP = Recovered products; AM = Auxiliary Materials; EC = Energy Consumption.

Fig. 12. Characterisation results for selected impacts of the proposed ZB system at the textile plant Credits due to by-products are not included in the analysis.
recovery leads to greater potential revenue but also greater costs. However, it should also be noted that the efficiencies of the system are expected to increase with further development and refinement at full scale. For the textile plant the cost of the treatment is over €25 euro/m³, which is not compensated by the recovery of the brine solution. The silica plant has a lower cost of €2.6 euro/m³ and is dominated by operating costs (specifically staff costs).

However, the contribution and sensitivity analysis show that the LCC and LCA are highly sensitive to one or two of the variables in each case study. Due to the prospective nature of the analysis, there is also high uncertainty in the quantities of materials used and the value of the recovered by-products. For instance, the cost of the coal mine ZB system
is highly affected by the price of dolomite. Nonetheless, this shows the value of considering both LCA and LCC at an early stage. The sensitivity analysis showed that the systems are highly sensitive to these variables for economic and environmental viability. In contrast Muñoz et al. (2019) did not experience such a high sensitivity in comparing a new solar heat pump for energy recovery with a reference system. There needs to be a firm focus in the next stages on these aspects in the design, evaluation and UA development to improve the performance and reduce uncertainty of the costs. This is further discussed in the next sections.

4.2. Design implications

Fazeni et al. (2014) stated that within life cycle process design, LCA is used to identify environmental hotspots and LCC identifies economic concerns (including both qualitative and quantitative information). Together with the process design team, these allow the identification of the technical feasibility issues in an iterative process. The next sections highlight the main lessons of the LCA and LCC for consideration in the development process.

4.2.1. Netherlands DWP

Much of the impact of the ZB system in the DWP case study are due to impacts that occur upstream during the production of chemicals (sulphuric acid and sodium hydroxide), which are used in the removal process of the total organic compounds. An increase in electricity of the plant is also a large contributor. This is supported by the LCC analysis which shows that the cost of raw materials increases from 0.38 €/m³ to 4.39 €/m³ of brine, with the ZB system. Therefore, the consumption of chemicals firstly needs to be reduced by lowering the dosage and optimising the process. Secondly, options to explore include recovery and reuse of chemicals, and using lower impacting chemicals, such as those from production sources that have lower GHG emissions. Any such action would also have to consider the impacts of transportation.

4.2.2. Polish coal mine

The main design recommendations are to reduce the electricity use
Sensitivity analysis for the LCC variables that have a strong contribution to climate change in the case study systems, showing the effect of a 25% increase and decrease of input on the total climate change impacts.

<table>
<thead>
<tr>
<th>Case</th>
<th>Variable</th>
<th>% contribution to total climate change impact in ZB system</th>
<th>Effect on climate change of ±25% increase or decrease</th>
</tr>
</thead>
<tbody>
<tr>
<td>DWP</td>
<td>Electricity:</td>
<td>47%</td>
<td>-12%</td>
</tr>
<tr>
<td></td>
<td>chemical prod</td>
<td></td>
<td>+12%</td>
</tr>
<tr>
<td></td>
<td>Plant electricity</td>
<td>36%</td>
<td>-10%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>+9%</td>
</tr>
<tr>
<td>Coal mine</td>
<td>Electricity</td>
<td>71%</td>
<td>-18%</td>
</tr>
<tr>
<td></td>
<td>Magnesium recovery</td>
<td>46%</td>
<td>-12%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>+12%</td>
</tr>
<tr>
<td>Textile</td>
<td>Reverse osmosis</td>
<td>93%</td>
<td>-23%</td>
</tr>
<tr>
<td></td>
<td>Recovered brine solution</td>
<td>98% (of benefits)</td>
<td>+24%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>-36%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>+35%</td>
</tr>
<tr>
<td>Silica plant</td>
<td>RO</td>
<td>80%</td>
<td>-20%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>+20%</td>
</tr>
</tbody>
</table>

* Results here are for configuration 1.

of the ZB system or investigate options to utilise renewable energy. This is critical because of the high carbon intensity of Polish electricity. In addition, the use of dolime in the magnesium recovery process is another cause of environmental impact. Therefore, the design should seek to optimise this process and reduce the use of dolime. The LCC showed a cost of 22.9% on reducing the energy consumption of the RO process and investigate options for using renewable energy. The LCC showed an overall cost of 12% to generate revenue from the magnesium hydroxide, so this should also be optimised, although increased recovery was correlated to increased costs in the current designs. However, the result is sensitive to both the price of inputs and the value of by-products, so further analysis should seek to accurately determine potential markets for by-products.

4.2.3. Turkish textile
For the textile ZB system the main design recommendation is to focus on reducing the energy consumption of the RO process and investigate options for using renewable energy. The LCC showed an overall cost of 25% per m³, primarily due to the use of hydrochloric acid and sodium hydroxide. Whilst the recovered by-products of brine solution and deionised water only generate a small potential value. Hence, the main design consideration is to optimise the use of chemicals or use alternatives.

4.2.4. Silica plant
The RO stage was highlighted as having high energy use and dominant across all other impact categories accounting for 80%. However, the use of regenerated membranes has a much lower impact than using new RO membranes. As energy use is the main concern, design improvements include consideration of potential heat recovery for the crystallisation step and the use of renewable energy sources.

4.3. Implications for the unified approach
The results obtained, help inform the next version of the UA and which aspects to develop. The approach of each of the three teams to the LCA and LCC was different, due to the availability of data, and the interest to investigate different aspects of the individual systems. The main lessons for the development of the UA are:

1. The teams require more specific instructions to guide them in performing the LCA and LCC, due to differences in approach, experience and knowledge.
2. The coverage and approach of the LCC’s needs to be made consistent.
3. A comparison of the reference (current) system needs to be made for all case studies.
4. The use of data, transparency and tracking and method used to upscale data needs to be improved. Also, a sensitivity and uncertainty analysis must be performed.

The coverage of the LCC’s varied considerably as shown in Table 3 with the coal mine and textile plant not able at this stage to account for the capital expenditure. However, the DWP and silica plant demonstrated that CAPEX is of less significance to life cycle costs than other factors such as consumables and staff. In the next stage, all case studies will cover each of these factors, in addition to environmental externalities. The coal mine examined four different process configurations and demonstrated a wide range of costs and revenues from the varying quantities of recovered by-products. This approach allowed more focus to be placed on the configurations and the recovery of by-products, showing the sensitivity of the cost of inputs and the value of recovered by-products. Similarly, the textile case also highlighted the importance of by-product value, as the value is too low to compensate for treatment costs.

The second stage of the UA also aims to include a comparison of the reference system for all case studies. Therefore, it is critical that data is collected on the current system, which includes local environmental impacts (primarily the impacts on waterways) as well as potential costs (externalities) to the regions (e.g. loss of amenity or fishing etc). For data and transparency, the latest version of the UA (see supplementary material) starts to lay out more stringent requirements for the LCA and LCC, including data preferences and ways to handle data gaps. To support this a hierarchy of preferences has been proposed in the UA and this will be finalised in the next stage. A further aspect introduced into the UA is communication. This is to ensure that both the LCA and LCC is communicated and reported in the same way, so that there can be more cross comparison and learnings across the case study.

5. Conclusion
The use of LCA and LCC at an early stage of the design and development process has been complementary and provided early insights into environmental and economic issues. The objectives of this initial

Table 5
Sensitivity analysis for the LCC variables that have a strong contribution to costs and revenues in the case study systems showing the effect on total costs or revenues of a 25% increase and decrease of the variable ("~" represents a revenue, whereas "~" represents a cost).

<table>
<thead>
<tr>
<th>Case study</th>
<th>System total cost/revenue (Euro/m³)</th>
<th>Variable</th>
<th>Cost/revenue of variable (Euro/m³)</th>
<th>Total cost/revenue with ±25% of variable (Euro/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DWP</td>
<td>+0.26</td>
<td>Revenue · demi water</td>
<td>-9.66</td>
<td>+2.26 ± 2.15</td>
</tr>
<tr>
<td>Coal mine</td>
<td>+24.11</td>
<td>Raw materials (chemicals)</td>
<td>+4.39</td>
<td>-0.83 ± 1.35</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Dolomite</td>
<td>+162</td>
<td>-18.6 ± 62.3</td>
</tr>
<tr>
<td>Textile</td>
<td>+24.9</td>
<td>Magnesium hydroxide</td>
<td>-141</td>
<td>+57.0 ± 13.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Hydrochloric acid</td>
<td>+12.3</td>
<td>+21.8 ± 28.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Sodium hydroxide</td>
<td>+8.2</td>
<td>+22.9 ± 27.0</td>
</tr>
<tr>
<td>Silica plant</td>
<td>+2.6</td>
<td>Staff</td>
<td>1.31</td>
<td>+2.46 ± 2.73</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Energy</td>
<td>0.53</td>
<td>+2.3 ± 2.9</td>
</tr>
</tbody>
</table>

* Results here are for configuration 1.
evaluation were to assess the ZB systems, inform the design process and provide lessons to develop the unified approach for the next evaluation stages.

The LCA showed that common hotspots are chemical use (with an increase in GHG emissions from 3.5 to 15.6 kgCO₂e/m²), with the addition of the ZB system (Munoz et al., 2019) in the DWP and energy use (in the coal mine), particularly in the RO process (for the textile and silica case studies). The ZB systems impact (in environmental and cost terms) are dependent on whether the benefits from by-product recovery counteract the increased chemical and energy use. The LCC highlighted a wide range of costs from £2.6/m² to £24.9/m². However, there is a large sensitivity of total costs at this stage and costs are expected to be lower at full-scale. The textile case study showed that the recovery of the brine solution for reuse in the plant leads to environmental benefits. The coal mine example could be profitable due to the value of the by-products, if the cost of chemicals could be reduced or revenues from by-products increased.

Design implications include a focus on the reduction and reuse of the use of chemicals (replacement with lower impacting ones), reduction of energy use, particularly in the RO units (and/or use of renewable energy sources) and maximising the recovery of by-products. The preliminary evaluation as a first step in a three-stage process was critical in the development of, a unified approach across the case studies. This may offer some guidance to other similar research projects that seek to develop a consistent LCA and LCC approach across multiple case studies.

CRediT authorship contribution statement

Steve Harris: Conceptualization, Methodology, Writing – original draft, Writing – review & editing. George Tsalidis: Investigation, Writing – original draft. Joan Berzosa Corbera: Investigation, Writing – original draft. Jose Jorge Espí Gallart: Writing – original draft, Writing – review & editing. Fredrik Tegstedt: Methodology, Writing – original draft.

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.

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Appendix A. Supplementary data

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References


