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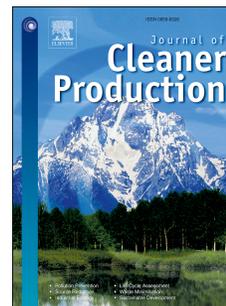
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# Accepted Manuscript

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## Ecodesign of microbial electrochemical technologies for the production of waste-based succinic acid thanks to a life cycle assessment

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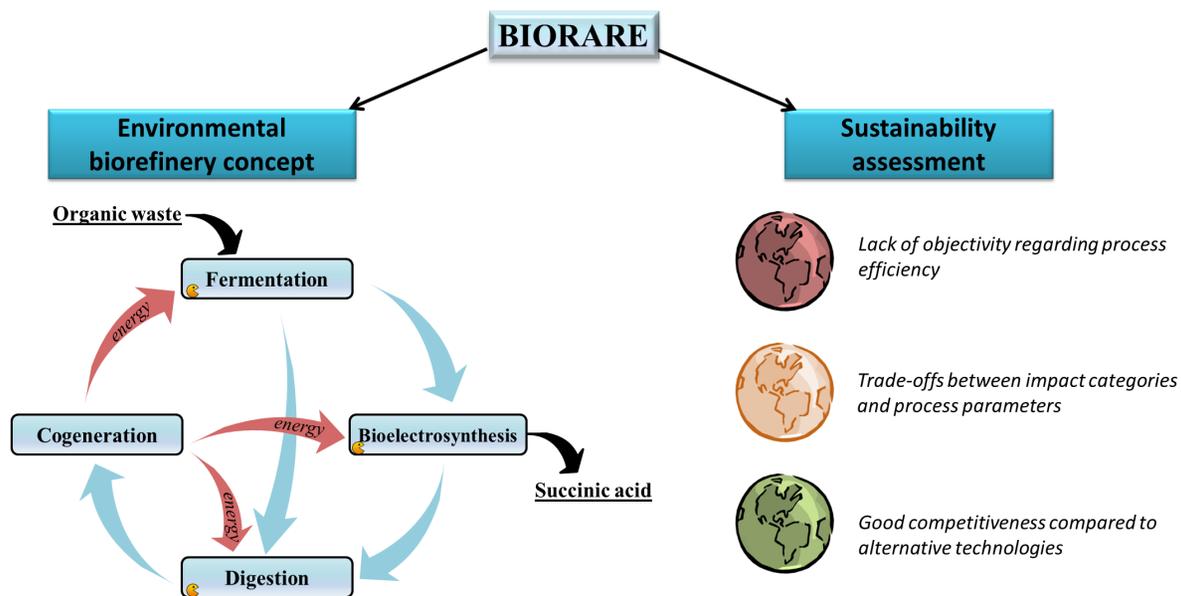
## Abstract

To face up abiotic resource depletion and other environmental issues as climate change due to usual fossil-based chemical production technologies, some alternative strategies have been developed using renewable resources. To produce such bio-based chemicals, renewable raw materials such as cereal crops or vegetables are currently used. To promote an environmental responsible practice, organic waste could be a relevant alternative to these dedicated crops. BIORARE technology is an innovative concept based on coupling an anaerobic digestion plant processing with bioelectrosynthesis in order to produce a range of chemicals from organic waste. Even if bioelectrosynthesis processes are not yet technologically mature; it is appropriate to consider the credibility of this emerging technology in environmental terms thanks to an eco-design approach.

This eco-design approach is based on the life cycle assessment (LCA) methodology. A LCA of biosuccinic acid production thanks to BIORARE technology has been carried out and has been combined with sensitivity analysis. The aim of this strategy is to ensure that sensitive parameters are identified and adjusted in order to make the technology the more eco-friendly possible whilst maintaining good economy efficiency. The present study describes the sensitivity analysis of the key parameters of the BIORARE technology applied for the production of succinic acid. These key parameters and their range of variation are chosen according to a realistic strategy allowing the control of the BIORARE technology on an industrial scale.

The results show that the current density applied during the bioelectrosynthesis and the hydrolysis yield during the pre-treatment of the waste stream are key parameters in the optimisation between production efficiency and the environmental footprint. The environmental efficiency of the process was studied by applying the eco-efficiency ratio. When the production of biosuccinic acid using the BIORARE technology was compared to a reference scenario, better overall eco-efficiency was shown despite some environmental penalties. In parallel, when the same study was performed for bioethanol production a low efficiency was revealed without environmental penalties.

**Keywords:** anaerobic digestion, LCA, eco-design, sensitivity analysis, eco-efficiency, biorefinery



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## 1. Introduction

The management of natural resources is a major topic of interest because it affects economic, social and strategic decision making. The way resources are being consumed generates substantial environmental concerns (Barnett and Morse, 2013). To lower mankind's environmental footprint, alternative energy sources have received special attention in recent decades. Resource depletion remains a major environmental issue which can be reduced by using waste streams as raw materials (European Commission, 2015). Anaerobic digestion (AD) is a renewable energy production following this approach. It is a natural biological process which biologically degrades organic matter in anaerobic environment (Nallathambi Gunaseelan, 1997). The products are methane (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>), also called biogas which can be burned to produce heat and electricity. AD is therefore recognized as an environmental friendly approach to produce green energy. Likewise, because of economic and environmental advantages, biological treatment of biowaste is of great interest for the production of a range of raw materials (Reddy et al., 2018).

In a further development, AD might be usefully combined with some emerging technologies (Yan et al. 2010; Ras et al. 2011; Foulland et al. 2014; Escamilla-Alvarado et al. 2017) and especially with microbial electrosynthesis (also called bioelectrosynthesis or BES) (Beegle and Borole, 2018; Bhatia et al., 2018; De Vrieze et al., 2018; Foulet et al., 2018). This concept is at the heart of the BIORARE project which seeks to both generate electricity and produce useful chemical products from biowaste. This biorefinery concept employs a synergistic approach comprising waste treatment (applying anaerobic digestion) and chemical production (applying BES). By combining the two processes, BIORARE technology seeks to overcome disadvantages linked to the conventional production methods through single anaerobic fermentation, and to produce a useful chemical product and electricity while processing organic wastes. The BIORARE study is based on the theoretical coupling of BES to existing AD plants. Therefore, the actual system as considered does not yet exist. Consequently, since the process is based on largely theoretical knowledge (supported by limited study made at the laboratory scale), there is a need to establish its likely environmental credibility at a future full scale installation. To do so, a methodology based on process engineering and coupled with Life Cycle Assessment (LCA) is necessary. This approach, referred to as eco-design, enables the introduction of an environmental dimension in the design process which can help direct new processes towards a sustainable system.

LCA is a methodology that allows the quantification of the potential environmental impacts of a product or process through its life cycle, following ISO 14040 (2006a) and ISO 14044 standards (2006b). The multicriteria aspect of LCA when applied in research and development relating to a new product helps define the technological challenges with respect to the environment. Published data regarding the sustainability of bioreactors technology is rare. However, Foley et al. (2010) and Pant et al. (2011) demonstrate the usefulness of LCA on bioelectrochemical systems when applied to wastewater treatment. This scarcity of LCA case studies concerning such innovative technologies is due to the difficulty to reconcile the needs of LCA (robust industrial data and system modelling) and the characteristics of an emerging technology that is not yet fully developed (lack of robust industrial data and knowledge about the behaviour of the technology). However, this early development of an innovative technology is the right moment to perform a LCA in order to improve its environmental performances because it is still possible to make strategic environmental choices to finalize the technological development (Azapagic et al., 2006; Tsang et al., 2014). Therefore, the issues of such LCA are to succeed the system modelling with few data and knowledge and to produce useful LCA results to identify some strategic environmental choices. Concerning the BIORARE process, a LCA should be useful to identify the most environmentally friendly technological parameters and produced biomolecules to ensure the environmental relevance of this technology in a sustainable context. Some challenges will have to be met as design issues due to stream constraints to produce the targeted molecule and sensitivity analysis of relevant parameters at BES

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scale and at AD plant scale. In biomolecule production context, the design of the system should be based on the production constraints (nature and quantity of biomolecule). Thus the difficulty is to model the whole system with these constraints – it is not usual in LCA of AD plants with a modelling most often based on the quantity of treated waste. To carry out sensitivity analyses, the range of values of each tested parameter is needed. However, this knowledge of the technology's behaviour is missing for emerging technology. How to perform a robust LCA for BIORARE process? How to obtain useful LCA results to determine some strategic technological and environmental choices?

In this paper, the LCA approach for the environmental assessment of BIORARE process will be described in the section Method in highlighting the interest and the relevance of applying LCA through especially sensitivity analyses to support the development of the BIORARE process. Results of sensitivity analyses will be then presented and discussed to identify strategic choices of the intrinsic parameters value for BES (such as current density and inputs streams) as well as parameters relating to the AD plant, such as the waste hydrolysis efficiency and CO<sub>2</sub> emissions management. Economical aspects will be brought in to the analysis by studying the economic performance of the technology as a function of its environmental performances by applying eco-efficiency estimation.

## 2. Methods

In this section, the BIORARE process is described by considering its components and their possible synergies. Amongst the possible application of BIORARE technology, is the production of alcohols (such as ethanol or 1,4-butanediol), organic acids, (such as succinic and formic acid), and caproate (Rabaey and Rozendal, 2010). For this study, succinic acid was chosen because of its role as a widely used intermediate product, (such as coatings, food and pharmaceuticals), also because of the increasing attention given to this product by the scientific community (Bechthold et al., 2008; Cao et al., 2013; Delhomme et al., 2009; Du et al., 2008; Lam et al., 2014; McKinlay et al., 2007).

### 2.1. How to deal with a prototype system like BIORARE process using the relevant LCA

#### 2.1.1. LCA of innovative technology issues

The advancement of a technology can be measured against a scale such as that set out by the NASA Technology Readiness Levels (TRL) (Mankins, 1995). When a system is considered as early in its development, this is reflected by a low TRL (below 6). In the case of the BIORARE process, the basic principles have been established (TRL 1), the technology concept has been formulated (TRL 2), the experimental proof of concept has been validated (TRL 3), as well as laboratory tests (TRL 4), as set out by definitions used by the European Commission (European Commission, 2014).

In its early stages of development, pursuing a novel technology carries few risks. However such systems are not validated by demonstration nor an evaluation phase in a relevant and operational environment which means a lack of reliable data. This deficiency can be partly fulfilled with laboratory validation tests supported by relevant literature that strengthens the feasibility of the project. Based on theoretical studies, a new process should undergo different modelling approaches in order to simulate reality. This procedure also allows the identification of sensitive parameters which are difficult to analyse without experimental testing or LCA. Once such parameters are identified, a sensitivity analysis using LCA is important in order to understand their influence on the overall environmental profile of the process. By studying the environmental profile of a process prior to implementation, eco-design strategies can be applied at an early stage in order to limit the potential environmental burdens of a badly configured process (Azapagic et al., 2006). However it is difficult to select test values for such parameters and uncertainties come along this process (Gargalo et al., 2017). At this point, the opinion of reliable experts is necessary to define an appropriate range of values for all sensitive parameters.

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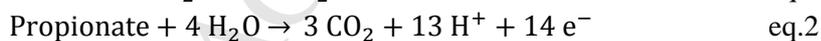
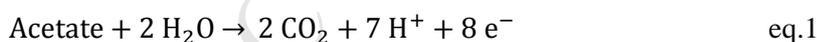
Most LCA studies evaluating innovative biotechnology have been conducted using lab scale data. It is important to scale up these results to an industrial operation because the experimental conditions can oversimplify the system. Such extrapolation is a difficult procedure that seeks to move from a scale of 1/10 m<sup>3</sup> to one of several m<sup>3</sup> or even a hundred m<sup>3</sup> or more. Several studies in the LCA field propose strategies to scale up from lab or pilot scale to the industrial scale (Shibasaki et al., 2007). Other research proposes methodology for the use of LCA as a development tool within the early stages of research (Hetherington et al., 2014). LCA results based on lab-scale data can still be used to improve the technology early in its developmental phase, particularly when comparing options on its process set up (Tsang et al., 2014). However, when compared with processes assessed using industrial-scale data, the lab-scale evaluation will be inferior and less reliable. Thus the choice of representative input and output data for such a studied system is especially important, as well as the careful choice of a reference scenarios for comparison.

### 2.1.2. Overview of the BIORARE process

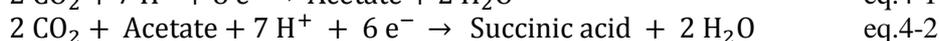
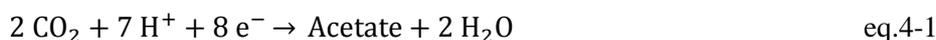
The BIORARE process relies on a new technology: microbial electrosynthesis. This is based on the principle of stimulating microbial activity in an electrolytic cell in order to catalyse microbial reduction of organic molecules leading to the synthesis of useful products. The consequence is the direct production of bio-based chemicals from organic waste.

A BES concept is based on an electrolytic cell set up with two distinct compartments separated by a membrane: (see Fig.1). Each compartment includes a bio-electrode made of covered by a biofilm. In the bio-cathode compartment, a carbon source, (carbon dioxide in this case), is needed while a carbon-rich input has to be provided to the anodic compartment. As shown in Fig. 1, each of the individual unit steps making up the system requires inputs and generates outputs. Because some outputs can become inputs for other process steps, a synergistic approach is appropriate and described here. The first step, hydrolysis, is a catabolic process in which bacteria breaks down (by hydrolysis) complex organic molecules into simpler ones such as carboxylic acid, organic fatty acids, hydrogen gas and carbon dioxide (McCarty, 1964). This pre-treatment step (preceded by sanitisation at 70 °C (Smith, 2015)) leads to the production of CO<sub>2</sub>, (resulting from organic matter degradation), and, a liquid waste stream which is enriched in volatile fatty acids (VFAs) such as acetate, propionate and butyrate. The pre-treated effluent is then passed through a screw press producing a liquid stream that contains most of the VFAs and a concentrate. The liquid stream rich in VFA is then injected in the anodic compartment and the concentrate is sent to the digestion unit in order to produce a biogas comprising 60 v.% of methane (CH<sub>4</sub>) and 40 v.% of CO<sub>2</sub>.

Within the anodic compartment of the BES, the stimulated microorganisms breakdown the VFAs (eq. 1-3) with the extracted electrons flowing to the bio-cathode (Lovley, 2006; Moscoviz et al., 2017);



The CO<sub>2</sub> produced from the previous hydrolysis step can be directly injected into the cathodic compartment of the BES unit in which the biomolecules are produced. A BES cell can produce a range of organic molecules depending on the reduction reactions taking place at the bio-cathode. Since the reduction reactions depend on electrical potentials, specific biomolecules can be targeted. The production of succinic acid follows the eq.4-1 and eq.4-2 at the bio-cathode:



Moreover, the BES unit generates a VFAs-depleted effluent but which still constitutes a carbon-rich resource useful for the production of biogas from the AD process. Electricity and heat for the process comes from the cogeneration unit fuelled by the biogas produced.

**Fig. 1. Simplified representation of the BIORARE process studied**

### 2.1.3. Proposal of a LCA-based methodology

The BIORARE process comprises three main parts as illustrated by Fig. 1. Between these parts, there are interactions and dependencies of note because downstream flows production depends on the operating conditions of the upstream flows production and *vice versa*. The technical aspect of these dependencies is explained in the following section (2.2.).

The common LCA methodology is based on four main steps widely applied respecting ISO standards and recommended guidelines (2006a, 2006b, European Commission, 2010). The first step is the definition of the goal and scope, followed by the inventory, the impact assessment and the interpretation, as shown in Fig. 2. The compilation of the inputs and the outputs of a simple system, meaning no existing interactions between flows, can be performed by using this methodology as it is. However, it is necessary to adjust the methodology for complex and low-TRL systems, such as the BIORARE process. The very design of the BES is the subject of current research and there are thus currently no industrial existing processes to confirm the selected values nor the strength of the flow dependencies. In order to design the BIORARE process, the sensitive parameters and dependencies should be identified and analysed. As shown in Fig. 2, the functional unit is constrained by the inventory step due to the streams dependencies. For this reason, we propose an adjusted LCA methodology which synchronizes the first and the second step of the standard methodology (Fig. 2). At last, the interpretation step comprises the study of the production efficiency within environmental and economic aspects, in other words the eco-efficiency of the BIORARE process. The eco-efficiency aims to identify the targeted biomolecules presenting the best environmental interest which is a trade-off between environmental performances of the BIORARE process and the potential market share of targeted molecules (see section 2.2.4.)

**Fig. 2. Adjusted LCA methodology for the BIORARE process**

## 2.2. LCA-based methodology applied to the BIORARE process

### 2.2.1. The goal, scope and boundaries of the study

The goal of this study is to identify the sensitive parameters that specify the BIORARE process and their influence on the subsequent environmental impact of its application. Fig. 3 illustrates the interdependency of the AD system and the BES system. In most LCA studies based on waste reuse, no burden is allocated to the waste input and thus it is excluded from the boundary of the BIORARE system as well. The raw materials used for the electrodes and the membrane, as well as any other chemicals used in the BIORARE system, are included in the system boundary. On the other hand, fabrication material (for example: hydrolysis and AD digestion tanks, BES reactor except for the electrodes and the membrane) are all omitted due to their long lifetime.

The functional unit (FU), which quantifies the primary purpose of the BIORARE technology and which allows for a comparative assessment, is defined as the production of 1,000 tonnes of the targeted product (succinic acid) by treating the corresponding amount of organic waste. The latter has been determined to be up to 35,000 tonnes for the production of 1,000 tonnes of succinic acid.

The design specification of the BES unit remains an objective of the BIORARE project. The linking up of such a unit with an anaerobic digestion plant has not yet been done, neither theoretically nor in practice. A relevant and consistent model for the BES unit and its implementation in conjunction with an anaerobic

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digestion plant are central issues covered by this paper: its application will also address the sensitivity analyses already discussed.

**Fig. 3. System boundaries for the LCA analysis of the BIORARE system**

### 2.2.2. Inventory

The life cycle scenarios presented here all were modelled using the GaBi 7.2.1 LCA software package (Thinkstep, 2016). The foreground life cycle inventory (LCI) data were compiled from experimental tests, detailed documents and previous work from each of the author's respective institutions ("Biorare project," 2018). Special attention was given to the inventory of the anaerobic digestion process, that it respected existing guidelines on biowaste management (Manfredi et al., 2011). The background life cycle inventory data (e.g. with respect to electricity, heat, graphite, steel, cationic membrane, and all chemicals cited) is provided by the ecoinvent database (Wernet et al., 2016) and the PE international (former name of Thinkstep) database ("GaBi Databases: GaBi Software," n.d.). Even with good data, the scaling up BIORARE technology depends upon appropriate calculations. To perform this LCA, the behaviour of the BES has to be understood, as well as its ability to produce succinic acid and the necessary operating parameters. Since no BES coupled with anaerobic digestion currently exists at even the pilot scale, an innovative calculation methodology is proposed here. In the following section the inventories of the BES unit and the anaerobic digestion system are described.

#### 2.2.2.1. Stream constraints

An important characteristic that quantifies the strength of organic wastes is its capacity to consume oxygen. This value can be determined in different ways but most relevantly in this case by the Chemical Oxygen Demand (COD) property. The quantity of succinic acid produced depends on the capacity of the BES to process COD taking into account the cathodic coulombic efficiency ( $CE_{cathode}$ ) and the current density (J), as summarized by equation 5:

$$m_{succinic\ acid} = \frac{J \times CE_{cathode}}{m_{COD/succinic\ acid} \times f_{Q_{COD}}} \quad (\text{eq. 5})$$

where  $m_{COD/succinic\ acid}$  is the quantity of COD to produce one unit mass of succinic acid and  $f_{Q_{COD}}$  is the quantity of electric charge the BES requires to breakdown a unit mass of COD (expressed in A.day/g<sub>COD</sub>). The volume of the BES depends on the output quantity of the product, thereby setting the required surface area of the electrode ( $S_{electrodes}$ ):

$$S_{electrodes} = 2 \times \frac{m_{succinic\ acid} \times F \times x_{e-}}{t \times J \times CE_{cathode} \times M_{succinic\ acid}} \quad (\text{eq. 6})$$

where  $m_{succinic\ acid}$  is the quantity of succinic acid produced,  $F$  is the Faraday constant,  $x_{e-}$  the number of electrons required at the cathode,  $t$  the time expressed in seconds and  $M_{succinic\ acid}$  the molar mass of the molecule of succinic acid. It is expected that the size of the BES unit increases with the production rate resulting in an increased total energy demand. One of the consequences is that the biogas combustion might not then produce enough energy to supply the BES unit and the AD system. It is further assumed that another consequence is the increase of the CO<sub>2</sub> and VFAs demand of the BES unit. The production rate of the succinic acid thus depends on the quality of the organic waste and also on the electrochemical parameters and the BES framework design.

The amount of energy produced from biogas is constrained by the substrate input to the AD digestion unit. Clearly, the methanogenic bacteria generate more biogas when they receive more substrate. The substrate is provided to the digestion unit by two other units: the BES unit and the separation unit (Fig. 1). This substrate composition depends on the performance of the hydrolysis step which hydrolyses the organic matter in the incoming sanitised waste. The efficiency of the hydrolysis phase is subject to various parameters such as raw waste composition, inoculums, pH and temperature (He et al., 2012). This

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efficiency is expressed as the hydrolysis yield which corresponds to the COD fraction of the total biodegradable COD of waste that is effectively solubilised. The quantity of waste required by the BIORARE process will be high when the hydrolysis yield is low, in order to meet the VFAs demand of the BES unit. However, as a consequence, the substrate loading of the AD tank then increases leading to an increased yield of biogas.

Since each step of the process influences each other via several conflicting relationships between the various stages of the process, it is difficult to anticipate the environmental outcomes as a result of the variation of a single parameter. LCA is thus the most appropriate approach to estimate those global outcomes by taking into account of all the streams involved and to set out their respective dependencies.

### 2.2.2.2. Inventory of the BES process

The electrochemical performances of the BES drive the calculation methodology. The required VFA ( $m_{VFA}$ ) and  $CO_2$  ( $m_{CO_2i}$ ) inputs depends on the criteria set out in Table 1 and are calculated according to equations 7 and 8, respectively, where  $m_{COD}$  and  $DM$  is respectively the COD value and the dry matter content of the hydrolysed waste handled annually by the BES unit,  $C_{COD}$  is the COD value of the VFA content,  $m_{CO_2/COD_{acetate}}$  is the amount of  $CO_2$  equivalent to one gram of COD from acetate decomposition and  $m_{COD_{acetate}}$  is the COD value of acetate.

$$m_{VFA} = m_{COD} \times DM / C_{COD} \quad \text{eq. 7}$$

$$m_{CO_2i} = m_{CO_2/COD_{acetate}} \times m_{VFA} \times m_{COD_{acetate}} \quad \text{eq. 8}$$

Table 1. Data used for the inventory of the BES process

Parameter	Origin of data	Quantity	Unit
Current density ( $J$ )	Experimental tests	20	A.m <sup>-2</sup>
Faradaic yield ( $FY$ )	Experimental tests	65	%
Electronic charge borne by one gram of COD ( $Q_{COD}$ )	$Q_{e^-} \times n_{e^-} / n_{COD} / M_{O_2}$ (1)	0.14	A.day.g <sup>-1</sup> <sub>COD</sub>
COD supply ( $m_{COD}$ )	$365 \times J / (Q_{COD} \times FY)$	4.02	kt.yr <sup>-1</sup>
Dry matter content of the hydrolysed waste ( $DM$ )	Experimental tests	0.25	kg.l <sup>-1</sup> <sub>hydro. waste</sub>
Total COD in the hydrolysed waste ( $C_{COD}$ )	Experimental tests	22.6	g.l <sup>-1</sup> <sub>hydro. waste</sub>
COD of acetate in the hydrolysed waste ( $m_{COD_{acetate}}$ )	Experimental tests	13.1	g.l <sup>-1</sup> <sub>hydro. waste</sub>

(1)  $Q_{e^-}$  is the charge borne by one mole of electrons,  $n_{e^-} / n_{COD}$  is the number of moles of electrons for one mole of COD, and,  $M_{O_2}$  is the molar mass of oxygen.

### 2.2.2.3. Inventory of the anaerobic digestion process

All of the streams are specified using the waste characteristics set out in Table 2. Knowing the dry matter content of the substrate, methane production ( $m_{CH_4}$ ) from digestion can be estimated by applying the formula (eq. 9):

$$m_{CH_4} = V_{CH_4/VM} \times BMP \times r_{VM/DM} \times \rho_{CH_4} \times DM \quad \text{eq. 9}$$

Where  $V_{CH_4/VM}$  is the volume of methane produced per kilogram of volatile matter (VM) and is assumed to be at 450 liters/kg; BMP is the methanogenic potential of the substrate and is assumed to be at 91 %;  $r_{VM/DM}$  is the ratio between volatile matter and dry matter;  $\rho_{CH_4}$  is the volumetric mass density of methane; and  $DM$  is the dry matter mass of the substrate.

From the methane production, carbon dioxide production can be estimated knowing their volume ratio (60 v.%  $CH_4$ , 40 v.%  $CO_2$ ). Gas leaks are also taken into account and are assumed to be 5 wt.%. The

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biogas is fed to a cogeneration unit, which produces heat and electricity that goes to meet some of the input required of the BIORARE process. For the rest of electricity and heat demand, system expansion rule of the boundaries is used for their recovery (ISO, 2006b). Additional electricity and heat come from the French power grid and from the production of steam from natural gas.

Table 2. Data used for the inventory of anaerobic digestion

Parameter	Origin of data	Quantity	Unit
Dry matter content of waste	Experimental tests	25.0	%
Hydrolysis yield	Experimental tests	19.7	
Volume content of CO <sub>2</sub> in the hydrolysis gas	Experimental tests and experts opinion	80.0	%
Volume content of H <sub>2</sub> in the hydrolysis gas	Experimental tests and experts opinion	20.0	%
Gas leak	Experimental tests and experts opinion	5.0	%
Volatile matter and dry matter ratio	Experimental tests and experts opinion	85.0	%
Volume of CO <sub>2</sub> produced for one kilogram of volatile matter input	Experimental tests and experts opinion	25.0	1

This methodology applied follows the “bottom-up” approach (Silveira et al., 2017; Swan and Ugursal, 2009), which focuses on the sub-systems (here the BES performance) to arrive at the whole system (here the production of succinic acid and carbon dioxide). This approach is appropriate for LCA studies on bioelectrochemical systems because it is essentially based on the COD measurement. In most cases dealing with biowaste sludge, this parameter is the dominant factor which is widely used to characterise the waste streams.

#### 2.2.2.4. Sensitivity analysis

This subsection presents parameters of the BIORARE technology which could influence the environmental impacts of the complete process including the AD step. In the following sections, current density, hydrolysis yield and management of the CO<sub>2</sub> stream are discussed as possible sensitive parameters.

#### Current density

The BES design has to satisfy various conditions to ensure its productivity. Some parameters are fixed such as electrode component and membrane thickness. To optimize the BES, its electrochemical performance has to be considered. The current density (J) greatly influences the efficiency of a BES unit. Laboratory scale experiments have been performed and revealed that the BES can produce succinic acid at different current densities. The lowest operational value is 5 A.m<sup>-2</sup> and the highest 300 A.m<sup>-2</sup>. In between, current densities of 20 A.m<sup>-2</sup> and 100 A.m<sup>-2</sup> were used in this study. The set of scenarios are thus labelled J5, J20, J100 and J300, corresponding to the current density. As a consequence of varying the current density, the electrical potential difference ( $\Delta V$ ) between the two electrodes also changes as illustrated by equation 10:

$$\Delta V = V_{bio-anode} - V_{bio-cathode} + J \times R \quad \text{eq. 10}$$

where  $R$  is the electrolyte resistance,  $V_{bio-anode}$  is the electrical potential at the anode, and,  $V_{bio-cathode}$  is the electrical potential at the cathode. The electrical potential difference is a key variable because the electrical charge input of the BES is determined by it. Indeed this input is the result of the product of the electrical potential difference, the current density, the electrode surface area and the production time.

By varying the current density, the quantity of succinic acid produced varies also. Indeed, the amount produced generally increases with the current density. As a consequence, the surface area of the electrodes has to be adjusted in order to produce the same amount of succinic acid for all current density values tested, which is necessary in the interest of the comparative LCA. In Table 3 the adjusted electrode surface, mass and energy demand are presented for the 4 scenarios studied.

**Table 3. Influence of current density (J) variation on BES inputs**

Scenario	J (A.m <sup>-2</sup> )	ΔV (V)	S <sub>BES</sub> (m <sup>2</sup> ) <sup>(1)</sup>	m <sub>elec</sub> (kg) <sup>(2)</sup>	E (J) <sup>(3)</sup>
J5	5	0.88	8.54x10 <sup>4</sup>	1.54x10 <sup>4</sup>	1.19x10 <sup>13</sup>
J20	20	1.14	2.14x10 <sup>4</sup>	3.84x10 <sup>3</sup>	1.54x10 <sup>13</sup>
J100	100	2.5	4.27x10 <sup>3</sup>	7.69x10 <sup>2</sup>	3.37x10 <sup>13</sup>
J300	300	5.90	1.42x10 <sup>3</sup>	2.56x10 <sup>2</sup>	7.95x10 <sup>13</sup>

(1) surface area of electrode; (2) mass of electrode; (3) electricity input of the BES

### Hydrolysis yield

The principle step of pre-treatment is the hydrolysis of the organic matter of the sanitised waste stream. This process encourages the solubilisation of the COD content to produce VFAs which can then be used in the BES unit at the bio-anode. The biochemical methane potential (BMP) assay is a common analytical method used to estimate the biodegradability of organic substrates under anaerobic conditions. The degradable COD can be calculated from the observed specific methane yield and the theoretical 350 ml of methane (at STP) per gram of COD stabilized (McCarty, 1964). In the context of this study, BMP assays were conducted on biowaste and resulted in a degradable COD of 3.2 g<sub>COD</sub>/g<sub>DM</sub>. Not all the degradable COD can be broken down by hydrolysis. Hydrolysis yield represents the dissolved COD over the initial degradable COD in the biowaste. The biowaste used in the BIORARE project had a dry matter content of 25 % w/w. Theoretical hydrolysis yields of 10 % (scenario Y10), 20 % (scenario Y20), 50 % (scenario Y50) and 80 % (scenario Y80) were considered. This has a very strong influence on the VFAs content of the hydrolysed waste sent to the BES unit. As shown in Table 4, this increases with the hydrolysis yield thus changing the amount of waste needed downstream. The total amount of CO<sub>2</sub> produced from the hydrolysis stage decreases if the hydrolysis yield increases because the required amount of organic waste feed then decreases. On the other hand, a poor 10 % yield would require eight times more waste than in the case of a 80 % yield, resulting in eight times more CO<sub>2</sub> available for the BES biocathode. When the CO<sub>2</sub> produced from the hydrolysis stage is not enough to meet the demand of the biocathode, CO<sub>2</sub> produced at the bioanode (see eq.1, eq.2 and eq.3) is diverted to the biocathode.

**Table 4. Analysis of the hydrolysed waste (and CO<sub>2</sub> production) as a function of the hydrolysis yield**

Scenario	Hydrolysis yield	COD of the hydrolysed waste (g.l <sup>-1</sup> )	Organic waste required (kg)	Required CO <sub>2</sub> input of the BES (at the bio-cathode) (kg)	CO <sub>2</sub> production from hydrolysis (kg)	Additional CO <sub>2</sub> to send to the biocathode
Y10	10 %	28	6.83x10 <sup>7</sup>	3.05x10 <sup>5</sup>	5.43x10 <sup>5</sup>	-
Y20	20 %	56	3.41x10 <sup>7</sup>		2.71x10 <sup>5</sup>	From BES bioanode
Y50	50 %	141	1.37x10 <sup>7</sup>		1.09x10 <sup>5</sup>	From BES bioanode
Y80	80 %	226	8.54x10 <sup>6</sup>		6.78x10 <sup>4</sup>	From BES bioanode

### CO<sub>2</sub> stream management

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Reducing and/or recovering CO<sub>2</sub> emissions from industrial processes is a large part of environmental policy (Bygrave and Ellis, 2003; Jos et al., 2016). CO<sub>2</sub> emissions arise in the application of BIORARE technology: from hydrolysis, electrolysis (BES) and cogeneration (Table 5). The values correspond to the scenarios Y20 and J20.

Table 5. Summary of carbon dioxide emissions

Unit process	Purpose of the unit	CO <sub>2</sub> production mechanism	Succinic acid production (1,000 t)	
			Quantity of CO <sub>2</sub> produced (kg)	Quantity of CO <sub>2</sub> required as input (kg)
<b>Hydrolysis Unit</b>	Pre-treatment of waste	Hydrolysis of waste	2.76x10 <sup>5</sup>	-
<b>BES Unit</b>	Functional biomolecules production	Breakdown of VFA at anode	6.73x10 <sup>5</sup>	3.06x10 <sup>5</sup>
<b>AD Unit</b>	Anaerobic biotransformation of substrate	Biogas production (no losses)	2.16 x10 <sup>6</sup>	-
<b>Cogeneration</b>	Biogas recovered into heat and electricity	From methane combustion	5.22x10 <sup>6</sup>	-

The hydrolysis unit emits CO<sub>2</sub> via the mechanism of organic matter degradation, which can also be responsible for H<sub>2</sub> emission. The quantity of CO<sub>2</sub> is directly estimated from the volatile and dry matter content of waste. Thus CO<sub>2</sub> emission from hydrolysis is determined by the nature and composition of the organic waste used. Among the four unit processes (Table 5), the hydrolysis unit and the BES unit generate the smallest amounts of CO<sub>2</sub>. The cogeneration unit and the digestion unit are responsible for, respectively, 63 % and 26 % of the total production of CO<sub>2</sub> from the BIORARE process. The cogeneration is the biggest source of CO<sub>2</sub> because of CO<sub>2</sub> from biogas and supplementary CO<sub>2</sub> production due to methane combustion reaction. The BES unit is the only part of the process that also requires CO<sub>2</sub> as input. The objective to recover CO<sub>2</sub> emissions could be achieved by means of streams management strategies that are presented below. Sending all of the CO<sub>2</sub> streams into the BES bio-cathode would not increase the amount of succinic acid produced because of the limiting effect of VFAs on biomolecule production. Three different management scenarios for the input of CO<sub>2</sub> into the BES unit are studied and presented in Fig. 4:

Fig. 4. CO<sub>2</sub> streams management scenarios (A, B and C)

- **Management A:** the CO<sub>2</sub> comes mostly from hydrolysis. 276 tonnes of CO<sub>2</sub> from hydrolysis are sent to the BES unit for the production of succinic acid. The hydrolysis process does not produce enough CO<sub>2</sub> to cover the BES unit input. Therefore, some of the CO<sub>2</sub> produced in the cathodic compartment (approximately 10 % of the BES CO<sub>2</sub> demand) of the BES unit is recycled to the anodic compartment. The rest of the anodic CO<sub>2</sub> is sent to the purification unit.
- **Management B:** CO<sub>2</sub> is provided by the biogas stream. The biogas output of the digestion unit is directly sent to the BES unit. The bacteria in the BES unit removes from the biogas the required amount of CO<sub>2</sub>, thus allowing the subsequent injection of a methane enriched biogas (composed of 37 v.% CO<sub>2</sub> and 63 v.% CH<sub>4</sub>) into the cogeneration unit. This hypothesis has yet to be verified. All the CO<sub>2</sub> from the bio-anode and all that from the hydrolysis unit are sent to the purification unit.
- **Management C:** the BES unit is not balanced in terms of CO<sub>2</sub> input/output. The anodic compartment produces more CO<sub>2</sub> than the cathodic compartment requires. Around 45 % of the

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CO<sub>2</sub> generated at the anode is sent to the cathodic compartment while the remaining 55 % goes to the purification unit.

Once purified, CO<sub>2</sub> might be used as a feedstock in the food industry, pharmaceuticals or analytical chemistry. Several purification techniques can be applied for CO<sub>2</sub> recovery and purification. Of these techniques, pressure swing adsorption (PSA) appeared to be the most suited to BIORARE technology (Overcash et al., 2007; Thambimuthu et al., 2002). This process operates on a repeated cycle with the basic steps being adsorption of CO<sub>2</sub> on adsorbent beds and then regeneration through pressure reduction. CO<sub>2</sub> recovery efficiency relies on many factors such as the initial purity of the CO<sub>2</sub> stream. In the case of BIORARE technology, the CO<sub>2</sub> streams arising from the hydrolysis unit, the BES unit, the AD digestion unit and cogeneration make up 80 v.%, 40 v.%, 40 v.% and 11 v.%, respectively. To limit the potential environmental impacts of the CO<sub>2</sub> purification unit, only that CO<sub>2</sub> produced by the hydrolysis and BES units is recovered.

When CO<sub>2</sub> is recovered, the BIORARE process becomes a dual-output system that is the production of both succinic acid and CO<sub>2</sub>. In such a case, it is appropriate to partition the related environmental burdens (ISO, 2006b). In LCA, this approach is called allocation. The inputs and outputs of a system can be partitioned between the different products on the basis of their underlying physical relationships, for example their mass ratio or their energy content ratio. In the case of BIORARE technology, the physical relationship between the succinic acid and CO<sub>2</sub> cannot be directed to favour the production of one product over the other. In this specific case, the allocation can be performed in proportion to the economic value of each product (Ardente and Cellura, 2012). The allocation methodology and inventory collection of the purification system are described in the annexe, "Supporting Material".

### 2.2.3. Environmental impact assessment

A procedure is required to calculate the potential environmental impact of each described scenario. This is the link between the released (or consumed) substance and its potential environmental impact. The January 2016 revision of the CML-IA method is used in this study (Heijungs, R. et al., 2001). There are ten impact categories listed that determine potential impact transfers. The categories are: abiotic depletion, acidification, eutrophication, global warming, ozone depletion, photochemical ozone creation, human toxicity and ecotoxicity categories (freshwater, marine and terrestrial). The scenarios to be tested against these impacts (described above) are summarised in Table 6.

**Table 6. Summary of scenarios evaluated**

Parameter	Scenario label	Tested values or description	Methodology	Results
Current density of the BES	J5	5 A.m <sup>-2</sup>	Section 2.4.2.1.	Section 3.1.
	J20	20 A.m <sup>-2</sup>		
	J100	100 A.m <sup>-2</sup>		
	J300	300 A.m <sup>-2</sup>		
Hydrolysis yield of the fermentation step	Y10	10 %	Section 2.4.2.2.	Section 3.2.
	Y20	20 %		
	Y50	50 %		
	Y80	80 %		
CO <sub>2</sub> emission and reuse management	A	BES CO <sub>2</sub> partial feeding from fermentation unit	Section 2.4.2.3.	Section 3.3.
	B	BES CO <sub>2</sub> feeding from digestion unit		
	C	BES CO <sub>2</sub> partial auto-feeding		

### 2.2.4. Eco-efficiency of biosuccinic acid production

When talking about eco-efficiency, the World Business Council For Sustainable Development (WBCSD) refers to a philosophy based on creating more goods and services with ever less use of resources, waste and pollution (Helminen, 2000). By applying the eco-efficiency criterion, it is possible to compare two processes, which have the same purpose but operate in different ways, taking into account the environmental point of view but bound to an economic aspect. In the BIORARE project, several chemical

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products can be produced as explained in section 2.1. Targeting a different product to succinic acid would result in treating a different amount of biowaste because the cathodic reaction is unlikely to be the same. The eco-efficiency criterion can be represented by the equation 11 (Kuusmanen and Kortelainen, 2005):

$$EE = \frac{\text{Economic value added}}{\text{Environmental damage}} \quad \text{eq. 11}$$

This measurement requires that the economic value added should be known or could be calculated from available data. However, the BIORARE process is at an early stage of development and data on investment, production cost as well as product sale price is unavailable. The eco-efficiency criterion is thus revised as follows (eq. 12):

$$EE = \frac{\% \text{ market share}}{\Delta I} \quad \text{eq. 12}$$

The market share that the BIORARE process might claim is based on the current French and European market shares of biosuccinic acid.  $\Delta I$  is the difference between the impact value of the BIORARE scenario and that of a reference scenario, divided by the sum of the impact values of both scenarios. The reference scenario of BIORARE succinic acid has been discussed separately (see the section, Supporting Material).  $\Delta I$  can range from -1 to 1. A negative value indicates environmental burdens whereas a positive value means environmental benefits with respect to the reference scenario. Conversely, a positive value shows that the technology is environmentally better than the reference scenario. Data on the environmental benefit/burdens and the reference scenario are given separately in the section, "Supporting Material".

The production of succinic acid through the BIORARE technology can be assimilated to an environmental biorefinery. Depending on the electrochemical conditions of the BES, several biomolecules can be produced (Rabaey and Rozendal, 2010). Because the stock of organic is deterministic (Gargalo et al., 2017) and would be moderate for mass production, it is necessary to identify the production of the biomolecule which is part of the proper market along with good environmental performances. In order to go towards the proper market, the production of succinic acid is compared to the production of bioethanol because the latter is part of an important energy market whereas succinic acid is considered an intermediate commodity. Indeed, the French and European markets for succinic acid are still in their infancy. As the study takes place in France, it is necessary to first evaluate the potential volumes of biowaste in France that could be treated by AD. In metropolitan France, nine existing AD units were identified. By adding up their operating capacities, approximately 701,000 tonnes of biowaste could be treated annually ("SINOE® déchets," n.d.). If a BIORARE process is to be implemented at each French AD site, 9,000 tonnes of bioethanol or 20,000 tonnes of succinic acid could be produced annually. Considering these production rates, succinic acid produced using the BIORARE technology would represent 87 % of the French market and 46 % of the European market ("Chemical industry awaits for bio-succinic acid potential," n.d.; Cok et al., 2014; Weastra, 2012), whereas the production of bioethanol would only represent 0.98 % of the French market and just 0.27 % of the European market (Flach et al., 2016).

### 3. Results and discussion

In this section, the results of the sensitivity analysis are presented. Firstly, the importance of the parameters describing the BES process is discussed in order to show their effect on performance. Section 3.2. deals with the constraints on the AD digestion brought about by the preceding hydrolysis step which influences all the waste-related streams in the BIORARE system. For instance, the quantity of greenhouse gases emitted during AD digestion and subsequent cogeneration, especially CO<sub>2</sub>, varies with the quantity of input substrate in the feed stream. In order to limit such emissions, CO<sub>2</sub> management strategies in the

context of LCA are presented in section 3.3. The outcome of all life cycle impact assessment (LCIA) is discussed in a final discussion.

### 3.1. The effect of current density in the BES on the environmental impact of the process

Changing the current density results in resizing the BES unit in terms of the electrode surface area in order to produce the same quantity of product. The quantity of electrode material increases with surface area. By contrast, the electricity consumed by the BES unit increases when the electrode surface decreases. This is due to the fact that a higher current density implies a higher voltage and thus a higher energy demand.

Thus increasing the BES unit production efficiency results in a trade-off between the need of electrode material and the electricity consumption. Through a comparative LCA, the consequences of this trade-off on potential environmental impacts are investigated, with the main results shown in Fig. 5. Electrode material is the highest contributing fraction of the BIORARE technology in the J5 scenario when considering the process contribution to acidification, eutrophication, climate change, ecotoxicity categories and photochemical oxidation. The contribution of the electrodes decreases as the current density increases, as shown also by Table 3. Meanwhile the electricity cost contribution increases given that the BES requires more and more energy to work at higher current densities. The environmental burdens of electricity production (based on the sources used to power the French grid) seem to be more significant than the burdens of the electrodes production since the scenario J300 is the least beneficial scenario.

**Fig. 5. Comparative analysis of LCIA of the variation of current density and the process impact on the environment**

The difference in contribution for electricity cost between scenarios J5 and J100 is too great to allow identification of the precise point from which the electricity contribution makes the scenario more environmentally damaging. A more precise comparative LCA study is therefore conducted. New current densities of 10, 15 and 25 A.m<sup>-2</sup> were tested and the consequences on the BES unit inputs presented in Table 7. The environmental burdens caused by the process as a function of current densities are represented in Fig. 6 with narrower band current densities of 5 to 25 A.m<sup>-2</sup>.

**Table 7. Additional data on the influence of the current density (J) variation on BES inputs**

Scenario	J (A.m <sup>-2</sup> )	ΔV (V)	S <sub>BES</sub> (m <sup>2</sup> ) <sup>(1)</sup>	m <sub>elec</sub> (kg) <sup>(2)</sup>	E (J) <sup>(3)</sup>
J10	10	0.97	4.27x10 <sup>4</sup>	7.69x10 <sup>3</sup>	1.31x10 <sup>13</sup>
J15	15	1.06	2.85x10 <sup>4</sup>	5.13x10 <sup>3</sup>	1.42x10 <sup>13</sup>
J25	25	1.23	1.71x10 <sup>4</sup>	3.08x10 <sup>3</sup>	1.65x10 <sup>13</sup>

(1) surface of electrode; (2) mass of electrode; (3) electricity input of the BES

**Fig. 6. Refined LCIA of the effect of current density variation by taking into account only the burdens of the BES materials and the electricity cost**

Depending on which impact category is considered, the contribution to environmental impacts of the BIORARE technology starts to increase either at a current density of 10 A.m<sup>-2</sup> (in the case of abiotic depletion, freshwater ecotoxicity, human toxicity), or at a current density of 15 A.m<sup>-2</sup> (in the case of acidification, eutrophication, climate change, marine ecotoxicity and photochemical oxidation), or at a current density exceeding 25 A.m<sup>-2</sup> (in the case of ozone layer depletion and terrestrial ecotoxicity). Under the last category, the environmental burdens are mostly related to the electrode production cost. Considering all these factors, the appropriate balance would be to set the current density of the BES between 10 A.m<sup>-2</sup> and 20 A.m<sup>-2</sup> depending on the dominant pollution concern.

### 3.2. The influence of the hydrolysis yield in the pre-treatment of the biowaste

The hydrolysis yield represents the capacity of pre-treatment to breakdown the organic matter present in the biowaste feed stream. When this yield is relatively high (> 50 %), the hydrolysis of waste is

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considered efficient. Consequently, less waste volume is needed to obtain the same required amount of VFA (see Table 4). However, at a low 10 % yield meaning a low VFA content ( $28 \text{ g}_{\text{DCO}}/\text{l}$ ), the hydrolysed subst from the BES bio-cathode (1,353 tonnes), (which is sent to the digestion unit), is almost eight times higher than that under the 80 % yield conditions (169 tonnes). The potential environmental impact resulting from different hydrolysis yields are showed in Fig. 7. Scenario Y10, (10 % yield), presents the highest environmental burden in all impact categories. This decreases as the hydrolysis yield increases. This trend is due to the contribution of the AD digestion system. Compared to the scenario Y80, more waste is needed thus more substrate has to be treated in the digestion unit under scenario Y10, which generates more digestate to be spread but more biogas is produced for the cogeneration unit. As a result, higher air emissions linked to digestion, combustion and spreading processes are expected which fall when the hydrolysis yield increases.

On the other hand, the difference between the options is far less marked when considering the following categories: abiotic depletion, human toxicity and marine ecotoxicity. The main reason is that scenarios Y50 and Y80 require more electricity input than scenarios Y10 and Y20 because less energy is produced internally resulting from less biogas production.

The hydrolysis yield is a parameter which depends on the pre-treatment conditions and the raw biowaste composition, it is thus difficult to predict. From a LCA perspective, any hydrolysis yields above 50 % would be of special interest in terms of the mitigation of environmental impacts.

**Fig. 7. Comparative LCIA of the system as a function of the variation of hydrolysis yield**

### 3.3 Is it better to emit or to recycle CO<sub>2</sub>?

The BIORARE process includes organic matter degradation in three distinct process units: the hydrolysis step, BES synthesis and AD digestion. These processes also generate a large amount of respiration gases, especially CO<sub>2</sub>. A CO<sub>2</sub> recovery unit was added to the BIORARE scenario and the comparison of the latter with and without this option shown in Fig. 8.

**Fig. 8. LCIA of succinic acid production compared with bioethanol production a) without and b) with CO<sub>2</sub> recovery**

Even at a low significance threshold, the recovery of CO<sub>2</sub> does not provide obvious environmental benefits since the relative contribution of both scenarios is very similar. The biggest difference, (although still not significant), is observed for the option Management B with respect to climate change. Before any CO<sub>2</sub> recovery, management B contributes the most amongst all three scenarios but it is the one showing a slightly reduced burden when CO<sub>2</sub> recovery is added in. In the “management A” scenario, 6,430 tonnes of CO<sub>2</sub> are recovered whereas the management scenarios B and C allow the recovery of 9,490 tonnes and 6,430 tonnes respectively. When a system generates more than one product, allocation is necessary in a LCA study. Economic allocation is applied for the BIORARE scenario (see Supporting Material) in order to apportion the environmental impacts between succinic acid and CO<sub>2</sub> production. Consequently, the relative contribution of each product increases with the amount produced. As more CO<sub>2</sub> is recovered in “management B” scenario, the CO<sub>2</sub> allocation is higher in this case than in “management A” and “management C” scenarios, thus leading to a smaller allocation to succinic acid production.

### 3.4. Eco-efficiency through biomolecule choice

When the environmental benefit of a given scenario is less than zero, then it is considered to show a low eco-efficiency (EE) to no EE, as shown in Fig. 9. The production of succinic acid using BIORARE technology displays a low EE close the “no EE” zone with respect to the following impact categories: terrestrial ecotoxicity, abiotic depletion and ozone layer depletion. On the other hand, the French market share and  $\Delta I$  value of BIORARE succinic acid make the BIORARE technology quite eco-efficient for six impact categories. The production of bioethanol using the BIORARE approach has no negative environmental impacts unlike the comparable production of biosuccinic acid. However, the market share of the BIORARE technology used to produce bioethanol would be very low (below 1 %). For this reason,

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the eco-efficiency of the BIORARE technology is considered low with a promising progress to a better EE.

The European market share case is not displayed here because the observations and conclusions are the same as for the French market share.

**Fig. 9.** Eco-efficiency comparison of the production of succinic acid and bioethanol on the French market if the BIORARE BES were to be implemented on all French AD plants treating biowaste (ADP: abiotic depletion potential; AP: acidification potential; EP: eutrophication potential; FEP: freshwater ecotoxicity potential; CC: climate change; HTP: human toxicity potential; MEP: marine ecotoxicity potential; ODP: ozone layer depletion potential; PCOP: photochemical oxidation; TEP: terrestrial ecotoxicity potential)

### 3.5 Discussion

Enhancing the productivity of the BES unit by increasing the current density also changes the environmental profile of the BIORARE technology. When the current density is at  $5 \text{ A.m}^{-2}$  (scenario J5), the BES is considered as a low cost process because it requires less energy input compared to the other three scenarios. However the high quantity of electrode material makes it less beneficial to the environment regarding resource preservation. The LCIA results might change in either direction if the electrode material was changed, for example using stainless steel instead of graphite. The electrode material was not revealed to be the most significant factor in the study of current density variation. Indeed, the sensitivity analysis did point out that the energy efficiency of the BES system was a real issue when the current density had been increased. Therefore, the choice of the external electricity source has to be relevant in the study. In this study, power from the French grid is used. There are alternative options to avoid purchasing electricity from grid, such as the integration of solar panels in to the BES process (Nevin et al., 2010). Overall the sensitivity of the current density parameter shows that to ensure good environmental performance, the productivity may have to be compromised, thus revealing a conflict between environment and economic objectives.

The sensitivity analysis of the current density parameter showed certain variability in the BES unit. The latter is part of a whole system which includes the anaerobic digestion. The importance of the waste transformation was considered through a sensitivity analysis of the hydrolysis yield of biowaste within the hydrolysis unit. The experimental yield is approximately 20 % which does not allow the sufficient production of biogas in the AD unit to supply the whole system in terms of electricity. In order to increase the electricity production, more substrate has to be sent to the digestion unit thus the hydrolysis yield has to be lower than 20 %. With this setup, less succinic acid is produced because of a lower provision of VFA content to the anodic compartment of the BES. Increasing the amount of succinic acid produced results from increasing the hydrolysis yield. However, this configuration implies producing less electricity, resulting in purchasing some from external sources. As observed in the case of the current density parameter, the sensitivity of the hydrolysis yield shows two conflicting effects. If the yields higher than 20 % can be achieved, the type of efficiency (energy or production) should be well considered in advance. Thus strategies to enhance the BIORARE technology may boost its eco-efficiency by lowering the contribution to certain environmental impact categories such as abiotic depletion, terrestrial ecotoxicity and ozone layer depletion.

As well as testing the sensitivity of process input parameters, the sensitivity with respect to outputs was also considered. Indeed, high emissions of  $\text{CO}_2$  were identified during the inventory step. The benefit of avoiding such  $\text{CO}_2$  emissions to the atmosphere is modest in the case of BIORARE technology because the quantity of  $\text{CO}_2$  to be recovered is relatively small. It might not be worthwhile to recover this small amount of  $\text{CO}_2$  based on the argument that the technical requirements for capturing and purifying  $\text{CO}_2$  could be considerable. A contribution analysis was performed and is set out separately in the Supporting Material annexe. The environmental impact of the  $\text{CO}_2$  purification unit is barely noticeable when compared to that of the other process steps. If the amount of  $\text{CO}_2$  to be emitted or recovered was larger, it would be of interest to see if the benefit of recovery would then be significant considering the increased burdens of biogenic  $\text{CO}_2$  emissions and of the purification unit inputs/outputs. The interest of BIORARE

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technology is to convert carbon (contained in biowaste) into energy through the production of biogas during the anaerobic digestion process and into high added value molecules in the microbial fuel cell. This sensitivity analysis focuses on the ways to reuse CO<sub>2</sub> emissions to supply microbial fuel cell in one hand and to recover CO<sub>2</sub> emissions for industry in the other hand. In these cases, the carbon is used and not sequestered. Actually, it is necessary to produce a soil enricher like compost or biochar to sequester carbon in soil. The philosophy of BIORARE is simultaneously the use of carbon to improve the economic value of the product (energy or biomolecules) and the production of a digestate as an organic fertilizer.

The assessment of the eco-efficiency of the biomolecule choice shows that the best combination of environmental and economic performances is obtained by the production of biosuccinic acid for an intermediate market. This conclusion is the result of a theoretical approach for the calculation of eco-efficiency of the BIORARE technology. Indeed, this calculation should consider the quality of biowaste which influences the technological performance of the disintegration process, and the flexibility of the market for the acceptance of waste-based molecules which influences the economical performances. Despite these limitations, the production of waste-based molecules is promising for intermediate markets from an environmental point of view.

#### 4. Conclusions

Many strategies and technologies have been developed to recycle organic wastes fitting in the concept of bio-economy. The concept of biorefineries that rely on bioelectrochemical systems are an attractive concept since it aims at producing useful chemical products from waste sources. Such a technology coupling a bioelectrosynthesis and an anaerobic digestion plant was modelled using a LCA methodology, in order to identify key environmentally sensitive design parameters and to select best implementation strategies, thus demonstrating how LCA approaches could be used to improve environmental performances of innovative processes. Sensitive parameters identified were the current density of the bioelectrosynthesis process and the hydrolysis yield during an initial pre-treatment of the waste. By testing the effect of these parameters, a balance between environmental benefit and good productivity was revealed. Separately, the recovery of CO<sub>2</sub> emissions, turned out to be inconsequential. The environmental efficiency of the technology was tested by studying the eco-efficiency ratio which is based on the possible market share of the product and the environmental benefit. In the case of succinic acid production, good eco-efficiency was shown despite some trade-offs on environmental impacts: if production was switched to bioethanol, there was a poorer efficiency with no impacts trade-offs. This eco-design approach of an innovative process based on LCA and eco-efficiency calculation shows the potential of environmental assessment to be useful for the improvement of the environmental, technical and economical performances of a technology with a low TRL. This systemic approach could be performed to guide the choice of targeted biomolecules which should be produced and the key parameters of the innovative technologies.

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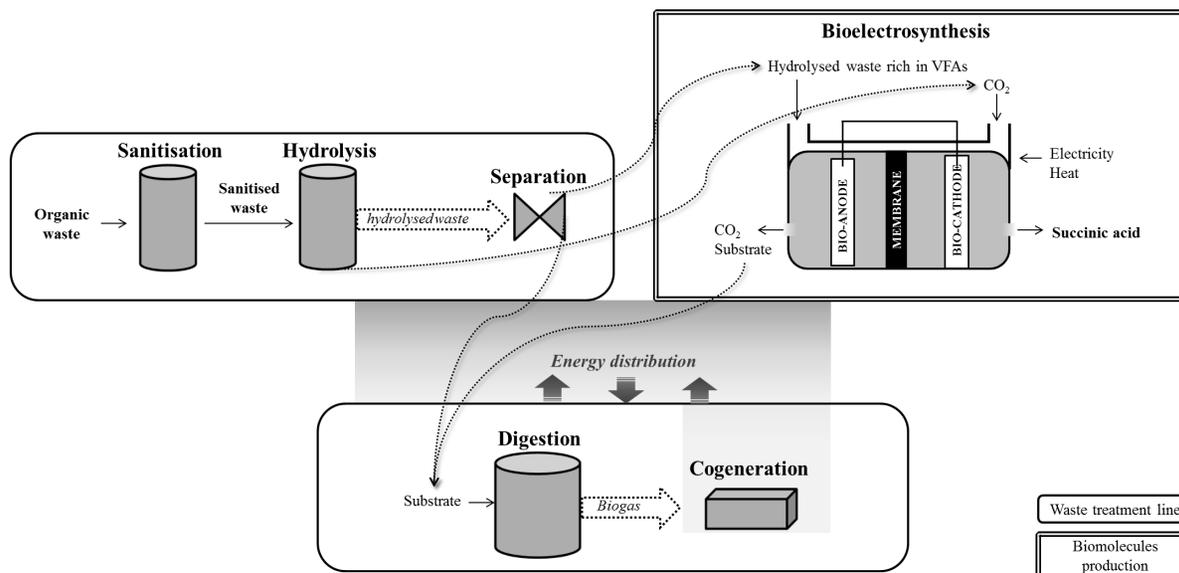
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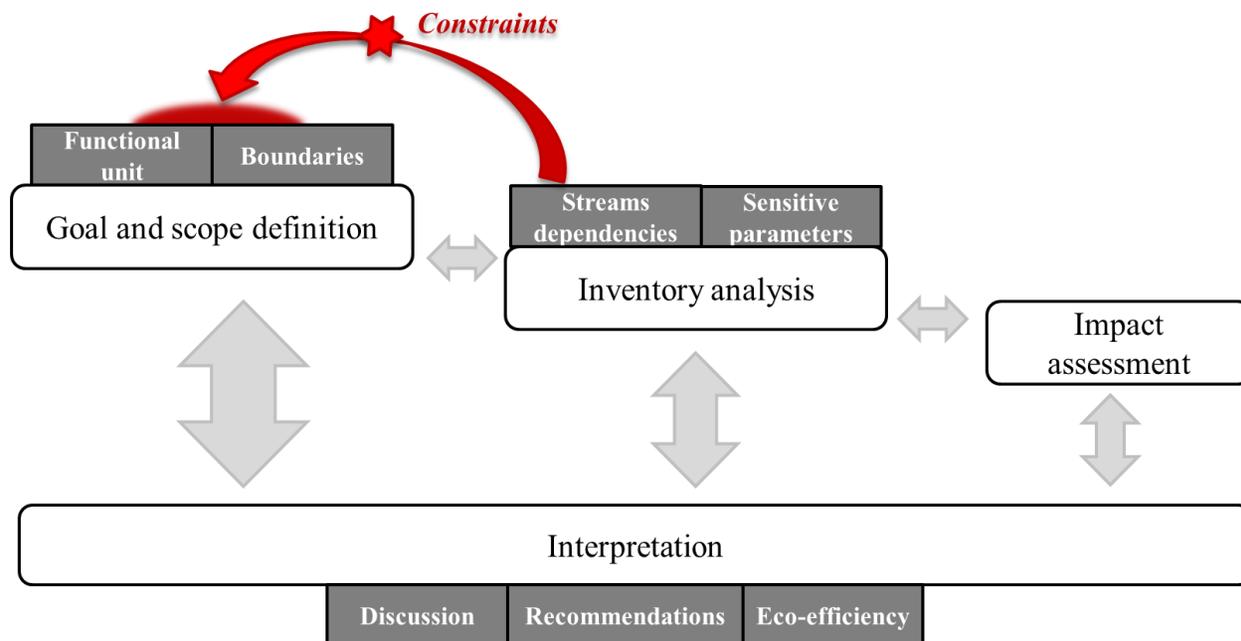
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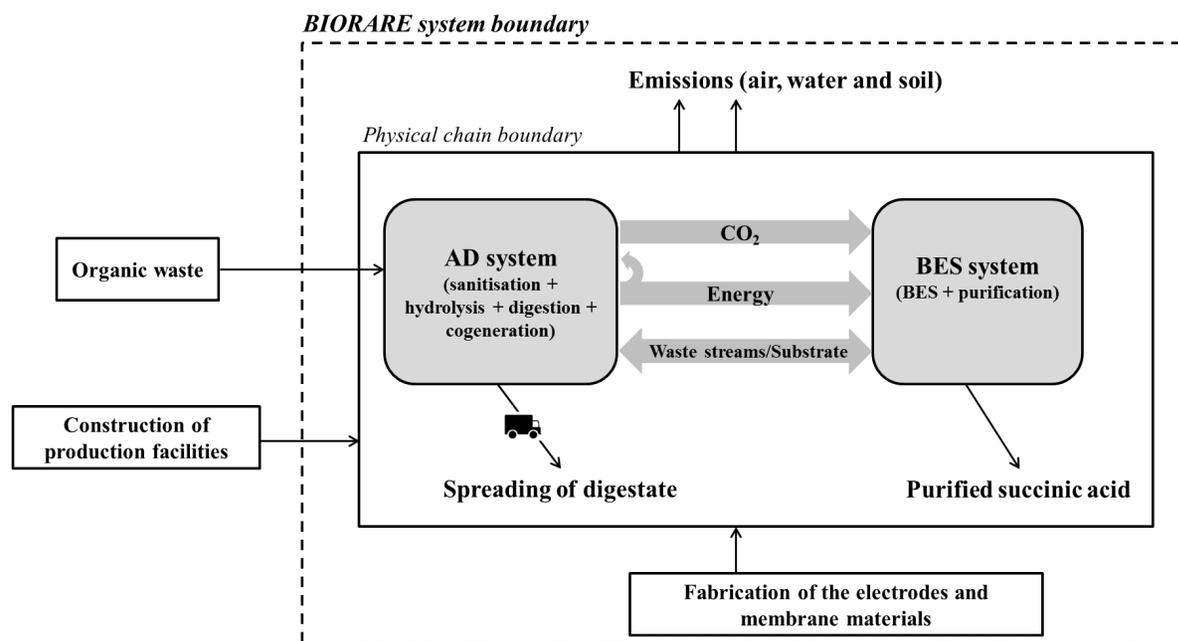
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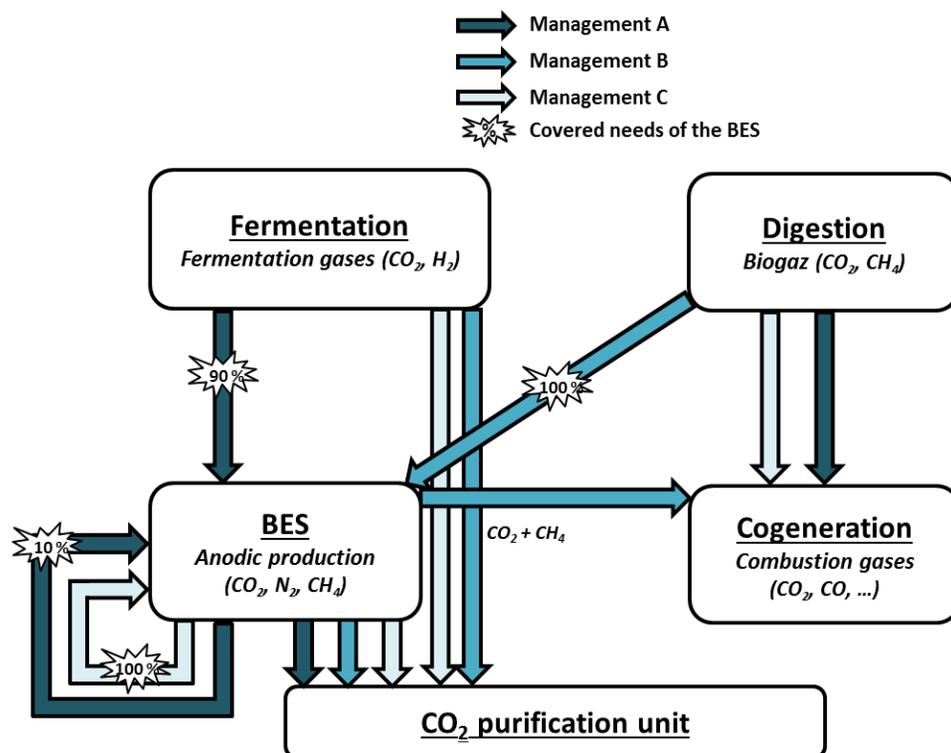
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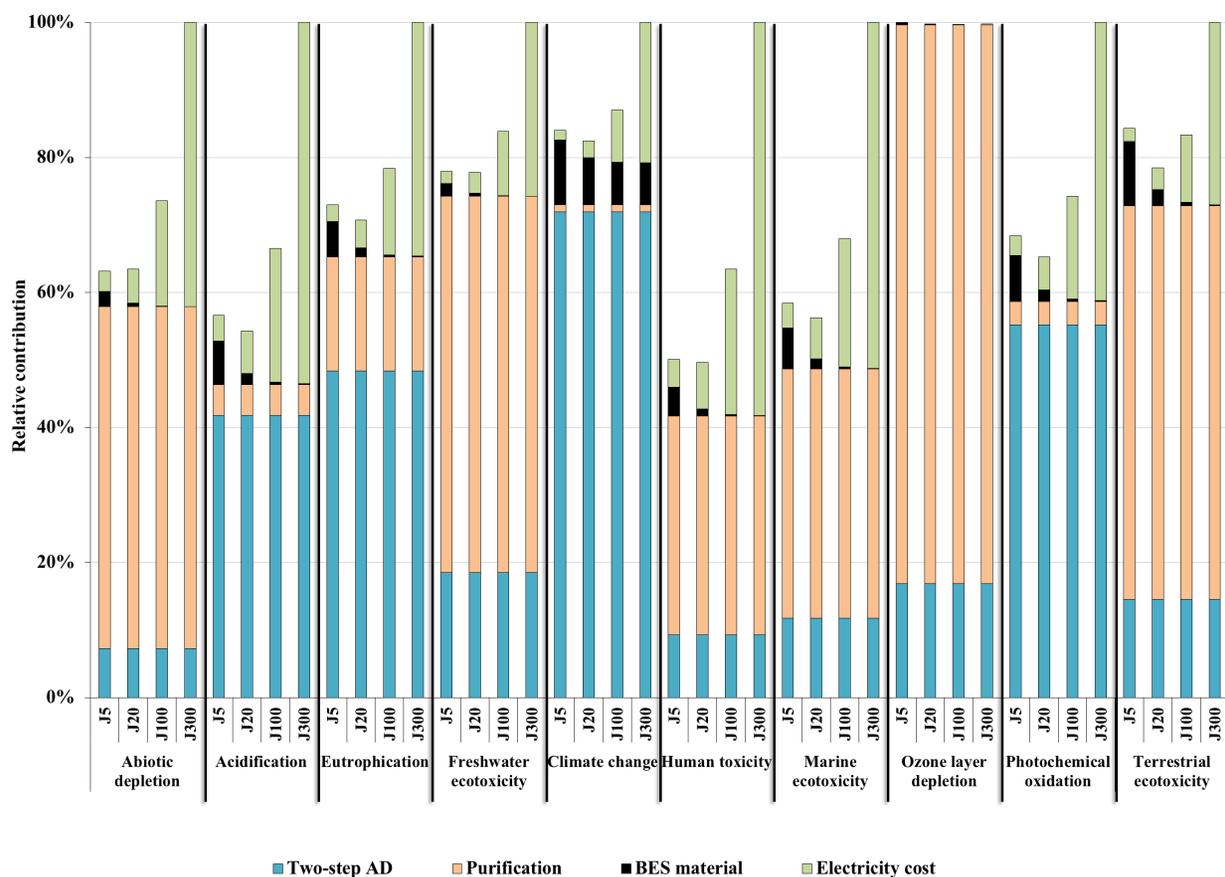
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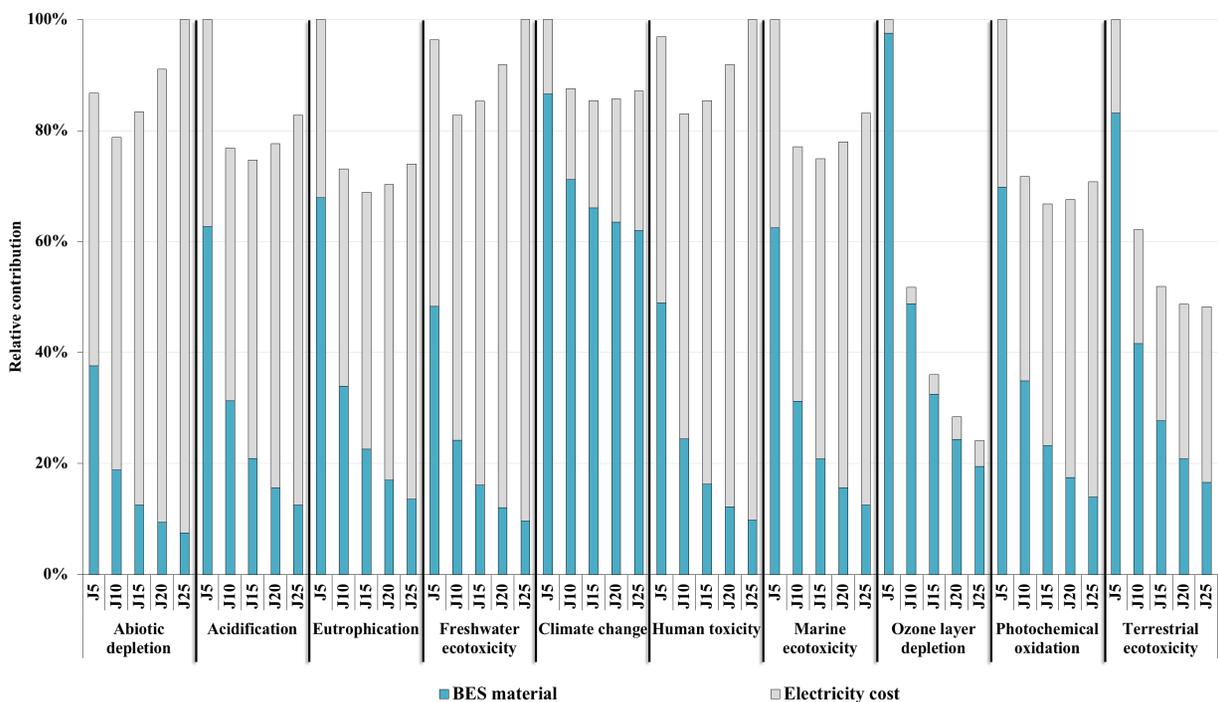
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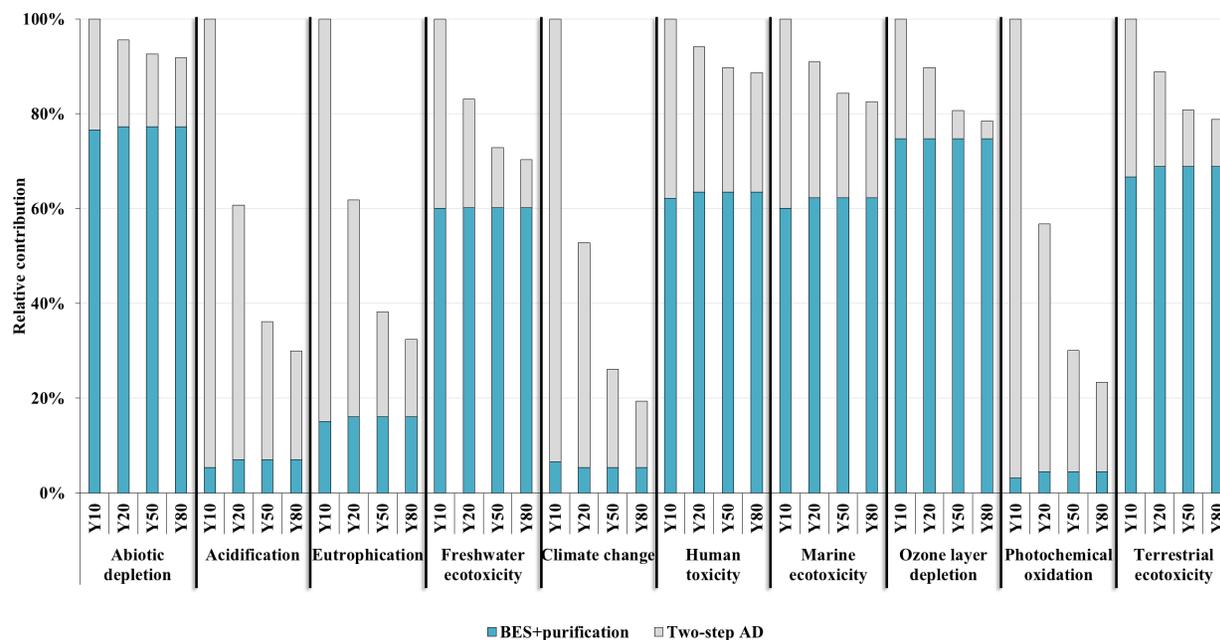
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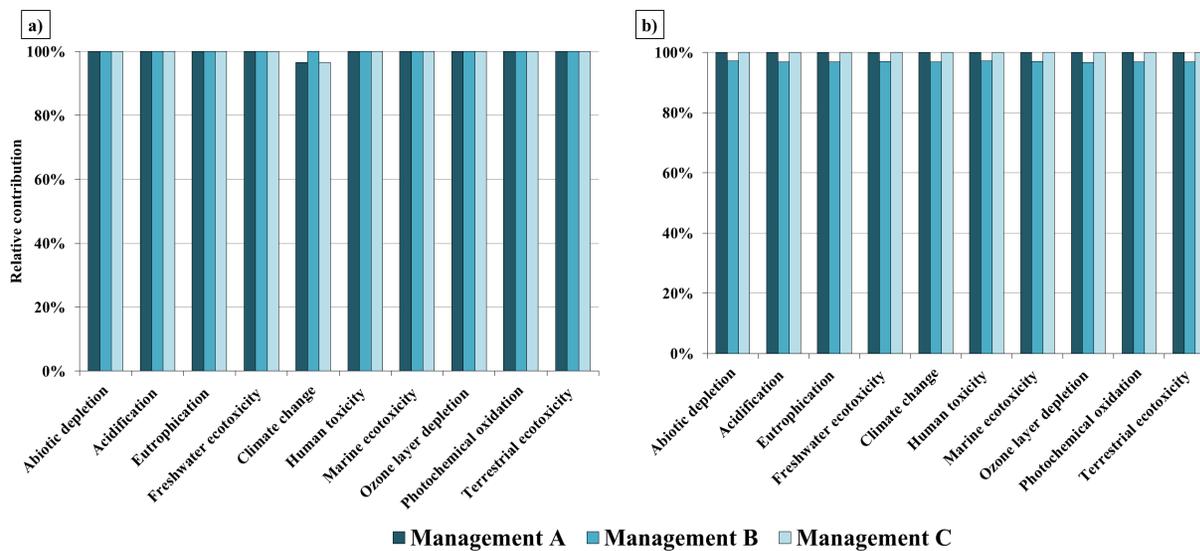


■ BES+purification ■ Two-step AD

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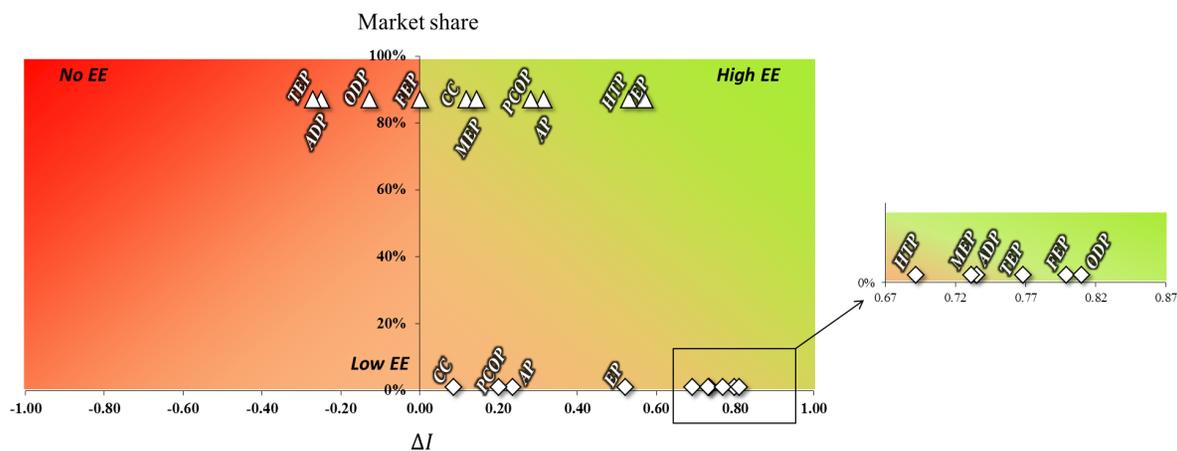
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△ Production of biosuccinic acid through the BIORARE BES unit coupled with the French AD plants

◇ Production of bioethanol through the BIORARE BES unit coupled with the French AD plants

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## Highlights

- Environmental biorefinery can produce bio-succinic acid from organic wastes
- Eco-design through life cycle assessment helps identifying sensitive parameters
- Trade-offs exist between production efficiency (set of technical parameters to optimize the production) and environmental burdens

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