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Eco design LCA of an innovative lab scale plant for the production of oxygen-enriched air. Comparison between economic and environmental assessment

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A B S T R A C T

Here we developed an early-stage attributional LCA of an innovative production process of oxygen-enriched air by water desorption. We compared LCA's results with a rigorous economic model, in an eco-design optimization perspective. We developed the life cycle analysis using primary data from the lab-scale plant. We used SimaPro 8.1.1.16 for the LCA analysis and the ReCiPe Endpoint V 1.12 E/A method for the interpretation of the results. The functional unit was the amount of enriched air produced by the plant in 24 h in Milano (Italy). A steady state chemical plant simulation software (PRO/II 9.3) calculated the reference flow and we input it in SimaPro with a parametric function. We considered a "cradle-to-grave" analysis. Temperature, pressure and water flowrate were varied to minimize environmental burdens and plant costs. Uncertainty analysis revealed that there is no difference between operating the degassing unit at 20 °C or 30 °C. SimaPro and PRO/II presented different optimum conditions. However, taken as a whole, we individuated the best process parameters from an eco-economic prospective. The results obtained are related to a lab-scale plant and thus no comparisons between industrial processes are possible, but the methodology we propose will improve the design of any process.

1. Introduction

Processes involving air as an oxidizing agent are the most common in industrial chemistry (Franz, 2005). Considering oxidation reactions, 79 % of air contains inert compounds (mainly nitrogen) (Seinfeld and Pandis, 2006). Inert gases require larger apparatuses and more energy to heat them, thereby increasing costs (US Department of Energy, 1999). Oxygen-enriched air, i.e. air with a percentage of oxygen greater than 20.95 %, is a cost-reducing solution because it operates well with lower volume reactors (Gollan and Kleper, 1984).

Cryogenic distillation (Belaissaoui et al., 2014) and membrane separation of oxygen from nitrogen (Bernardo et al., 2009) are the current industrial processes for EA production. New less energy demanding technologies have been proposed in recent years, e.g. Eriksson and Kiros designed and built a portable device to produce EA which employs zeolite as an adsorbent and exploits the technology of pressure swing adsorption (Eriksson and Kiros, 2014) while Habib et al. optimized a hollow-fiber polymeric membrane and simulated a multi-stage unit capable to reach 91.4 % of oxygen with competitive energy consumption (Habib et al., 2017).

Manenti and Pirola published a patent (Manenti and Pirola, 2014a) and an article (Manenti and Pirola, 2014b) concerning a new process for the synthesis of EA. EA desorbs from water since oxygen has a higher solubility in this medium compared to nitrogen (Wilhelm et al., 1976). This method relies on Henry's Law, a thermodynamic equation which describes the solubility of gas in liquids in function of partial pressure and temperature. The lower the temperature and the higher the pressure, the greater the gas solubility. Therefore, EA can be extracted from any kind of water (seawater, tap water) by varying the operative parameters in order to decrease oxygen solubility. This new technology is interesting when it is coupled with a plant that recycles excess heat.

We also verified experimentally the process at the lab-scale. We set up and ran a plant for the continuous production of EA to show the feasibility of this new unit operation. Gathering early-stage data

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is the first step towards the scale-up of the process, which requires modelling and a rigorous optimization study (Dimian, 2003).

Engineers design chemical processes by minimizing the total annual costs of the plant (Towler and Sinnott, 2013). This analysis provides good results in terms of the process synthesis but lacks the environmental aspect. On the other hand, the LCA of a process focuses on environmental issues. Hetherington et al. (2014) suggest the application of LCA to the early stage development of a process. However, eco-design could lead to unfeasible operative parameters, which in turn creates unwanted expenses. Thus, it is crucial to calculate the economic potential and the environmental impact of a process to balance costs and environment preservation. Kniel et al. (1996) combined environmental-economic analysis on existing plants, whose technology is well known and developed. As a case study, Park et al. (2016) studied the eco-design of the production of 7-aminocephalosporanic acid for carbohydrate waste using a 15 ton y^{-1} production. Peuportier et al. (2013) applied LCA to energy efficient buildings to study their use phase impacts. This kind of assessment should be applied during the early-stage of the process scale-up to stress the economic and environmental hot spots. For example, Barton et al. (2002) applied LCA at the conceptual stage for technologies to assess the best available techniques in the sector of Integrated Pollution Prevention and Control (IPCC). He concluded that the use of LCA provided useful insight to the research program (Barton et al., 2002).

To face the lack of independent economic and environmental analysis typical of the chemical and environmental engineering fields respectively, we propose a new approach that considers the optimization of a lab-scale process in a holistic point of view, i.e. joining these aspects and obtaining optimal operative parameters. We studied a laboratory scale plant for the continuous EA production from these two aspects, i.e. applying an LCA analysis to the process and, simultaneously, calculating its economic potential using PRO/II, a steady state simulation software.

This work represents a first evaluation of the technology proposed. It is not possible at the moment to propose a similar LCA-economic assessment for the industrial EA production, due to the lack of consolidated and verified scheme of enriched air industrial processes (Piccinno et al., 2016).

2. Material and methods

2.1. Experimental plant

Fig. 1 reports the lab-scale plant flowsheet divided into the two main sections.

Before the start-up operation, helium purged all the units of desorption section. This line was never used again. Tap water equilibrated with compressed air (5 bar) in tank S1 at ambient conditions, i.e. 1.01 bar and 20 °C. This section guaranteed that oxygen and nitrogen concentrations reached equilibrium. An online portable model Hanna Instruments oximeter 9146, equipped with a probe (HI 76407/4F) monitored oxygen content in S1. Through a rubber pipe, water reached the degassing unit (R1). Valve (V4) regulated its flowrate, which was measured using a rotameter (K1). The degassing unit is a jacketed cylindrical glass container, 350 mm height and with a diameter of 145 mm, heated by hot water provided by a thermostat (Falc FA-90, mod SB5). A thermocouple (Pt-100 Delta Ohm, HD 9010) monitored the temperature inside R1. A peristaltic pump (G1, KNF CH-6210) withdrawn the degassed water, whose flowrate was measured by a second rotameter (K2). An Edward 2 Two stage pump generated vacuum, and a micrometric valve (V6) regulated it. A micro-GC Agilent 3000A analyzed the EA produced. It was equipped with a MOLSIEVE column, kept at 45 °C, and a TCD detector. The line ended in a Ritter TG01/5 gas meter, that measured the amount of EA produced. We performed 25 experiments varying water inlet flow between 10 and 30 L h^{-1} (set by a peristaltic pump), degassing pressure between 600 mmHg and 400 mmHg and temperature between 28 °C and 63 °C. We kept the R1 pressure over 400 mmHg to avoid water boiling inside the unit. A peristaltic pump set water flowrate. R1 temperature was constrained because water was the thermostatic fluid. The plant ran for one year.

2.2. LCA analysis

We considered a “cradle-to-grave” life cycle analysis. The boundaries of the system include the manufacture of the plant, the transportation of the material, the production of oxygen-enriched air and plant disposal (Fig. 2). The location of the plant is Milan (Italy).

Since the operative parameters influence EA production rate and composition, we chose the volume of EA produced in 24 h of plant operation in Milan, Italy as the functional unit. Indeed, we adopted the Italian energetic mix in LCA calculations. We assumed a 20-years lifespan for the lab-scale plant. For the waste scenario, we considered the average recycling data of Northern Italy, i.e. assuming the average landfill, recycling and incineration percentage for the main constituents of the plant, glass (Assovetro, 2009), plastic (Eurostat, 2017) and metals (Giliberto, 2015). Moreover, we did not account for the environmental burden the helium purging may have caused, since we employed it only at the reactor start-up.

SimaPro 8.1.1.16 by PRé Consultants modeled the LCA study. We adopted Ecoinvent v3.1 and ELCD v3.1 libraries to account for the secondary data. ReCiPe Endpoint V 1.12 E/A evaluated the environmental impact.

We modeled EA production as depending on three main parameters, i.e. water flowrate, temperature and pressure. We varied these parameters and MATLAB 2015b regressed the EA volume and the oxygen concentration with the following equation (Eq. (1)):

$$f(F,T,P) = k_1 \cdot F + k_2 \cdot T + k_3 \cdot P + k_4 \cdot F \cdot T + k_5 \cdot F \cdot P + k_6 \cdot T \cdot P + k_7 \cdot F \cdot T \cdot P \tag{1}$$

where k_i are the adjustable variables calculated by MATLAB built-in nonlinear regressions routine and *F*, *T* and *P* are the three main parameters chosen. We ran the same regression considering the EA price as calculated function (see Section 2.3).

A voltmeter-ammeter measured the power consumption of each instrument. After the plant reached the stationary conditions, we carried out these measurements for 3 h to correlate the power consumption with the operative parameters.

We modeled the dependence of the electrical consumption with a second order polynomial of *T* (Eq. (2)).

$$f(T) = a \cdot T^2 + b \cdot T + c \tag{2}$$

where *a-c* are the adjustable parameters and *T* the operative temperature.

We weighed all the plant parts (Table 1). In SimaPro, the plant was divided into two sections: the water equilibration with atmospheric air and the enriched air desorption (Fig. 1).

We collected the primary data several times to calculate the standard deviation. For instruments, e.g. pumps and thermostat, we used the INPUT-OUTPUT database available in SimaPro (Input Output\USA 2002). We entered the actualized economic value for these instruments. We considered a lognormal distribution of the

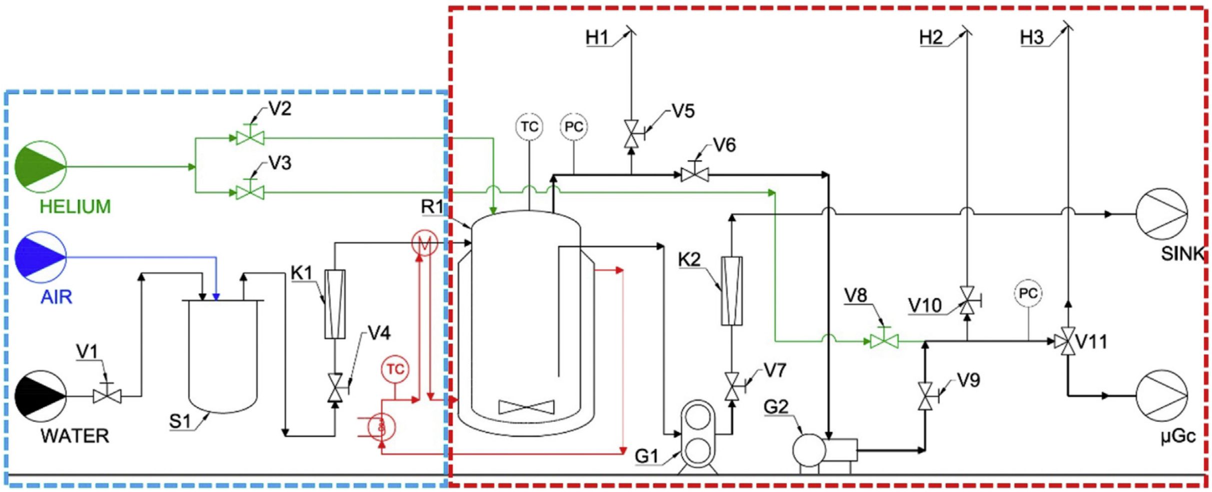

Fig. 1. Flowsheet of the experimental setup, V1-11: valves, S1: equilibrating vessel, R1: degasser, K1-2: liquid rotameters, G1: liquid pump, G2: vacuum pump, H1-3: purge. Blue dotted line indicates the water enrichment section while red dotted line the EA desorption one. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

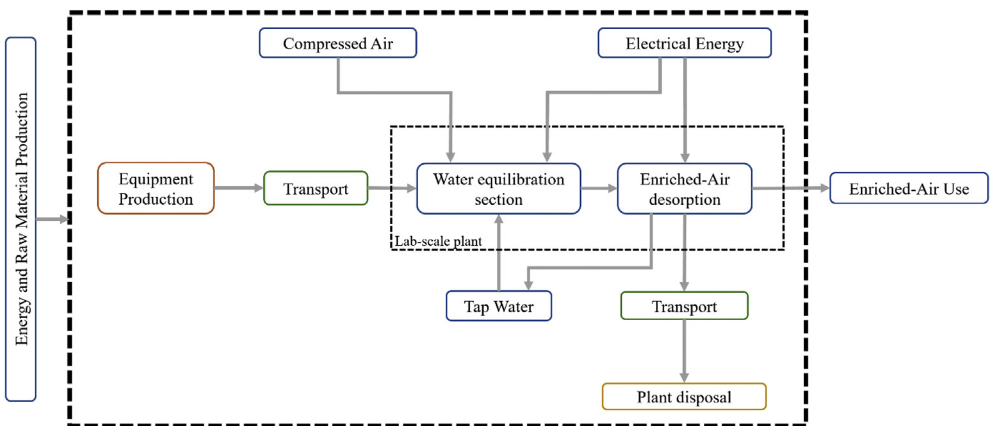

Fig. 2. Scheme of the system boundaries of the LCA project.

transportation distance of the plant's parts with a SD of 2 (Goedkoop et al., 2016). We accounted for the transportation of all plant parts from the manufacturer to the laboratory facility and the transportation to the disposal site (Fig. 2). We tested 30 scenarios (Table 2) to identify the best parameter sets that minimize the environmental impact and performed a Monte Carlo simulation for the uncertainty analysis (Hung and Ma, 2009).

Table 1
Summary of the materials constituting the lab-scale plant.

Material	Mass [kg]	Comments
	Water equilibration section	
Steel, unalloyed	1.244	Steel constituent pipe and valves
Rubber	0.284	Connection pipes
Glass tube	0.300	Rotameter glass cylinder
Polypropylene	1.321	Water equilibrating tank
	Enriched air desorption	
Steel, unalloyed	2.739	Steel constituent pipe and valves
Rubber	0.474	Connection pipes
Teflon	5.461	Teflon constituent pipe and gaskets
Polyurethane	0.544	Thermal insulation pipe
Silicone	0.028	Gaskets
Iron	0.008	Magnetic stirrer

2.3. Economic analysis

Tank and degassing units are modeled as flash unit operations. Operative conditions of these two units are set using experimental temperatures and pressures. Henry's law calculated the solubility of nitrogen and oxygen in water. NRTL model estimated the activity of oxygen and nitrogen in water phase. We expressed Henry's constants as function of temperature (Eq. (3))

$$\ln(H) = C_1 + \frac{C_2}{T} + C_3 lnT \quad (3)$$

whose coefficients are reported in Table 3 for N_2 and O_2 in water.

Capital costs (CAPEX) are the ones of the equipment, i.e. degasser and pumps (Supplementary material, Table S1). Operating costs (OPEX) include utilities and consumables like tap water, electricity and compressed air (Guthrie, 1974, 1969). We calculated the price of EA by assuming that the plant will last 20 years and have a depreciation time of 9.5 years. Eq. (1) correlated the price with the three operative parameters. We reported a detailed description of the model elsewhere (Galli et al., 2017).

Table 2
Parameters for each scenario.

Parameter scenario	Water flowrate [Lh^{-1}]	Temperature [°C]	Pressure [mmHg]	Parameter scenario	Water flowrate [Lh^{-1}]	Temperature [°C]	Pressure [mmHg]
1	10	20	350	16	20	20	350
2	10	30	450	17	20	30	450
3	10	40	550	18	20	40	550
4	10	50	350	19	20	50	350
5	10	60	450	20	20	60	450
6	10	20	550	21	30	20	550
7	10	30	350	22	30	30	350
8	10	40	450	23	30	40	450
9	10	50	550	24	30	50	550
10	10	60	350	25	30	60	350
11	20	20	450	26	30	20	450
12	20	30	550	27	30	30	550
13	20	40	350	28	30	40	350
14	20	50	450	29	30	50	450
15	20	60	550	30	30	60	550

Table 3
Parameters used to compute Henry's constants (databank: PROII_9.3).

	T_{min} [K]	T_{max} [K]	P_{min} [kPa]	P_{max} [kPa]	C_1 [-]	C_2 [K]	C_3 [-]
Oxygen	200	500	1	10,000	155.5533	−7442.29	−20.2359
Nitrogen	200	500	1	10,000	158.2643	−7260.14	−20.7005

Refer to supplementary material for all the equations parameters (Table S2).

3. Results

The lab-scale plant successfully produced EA (Fig. 3a–b).

Table 4 reports the values of k for Eq. (1). Only S1 temperature influences electrical power consumption. The optimum adjustable values for Eq. (2), are $a = 0.14$ Wh°C^{-2}, $b = -3.90$ Wh°C^{-1}, $c = 1036.5$ Wh. We inserted these values in the SimaPro model to evaluate the environmental impacts of the production process. The volumes of EA produced after 24 h of operation (functional unit) in each scenario are reported in the Supplementary material (Table S3).

4. Discussion

Environmental impact depends on operating temperature. The higher the temperature, the greater the impact (Fig. 4) since the plant's need for electricity was exclusively for heating. The treatment of waste water also contributes to the environmental impact but to a lower degree.

Our environmental and economical results (Fig. 5) show the single score impact points (calculated with the Europe ReCiPe E/A Endpoint method) together with the uncertainty (calculated by Monte Carlo analysis) and the simple moving average (step 3) of EA price.

The Europe ReCiPe E/A method considers long-term ecological effects and is based on precautionary principle thinking. The single score (Fig. 5) evaluates the environmental impact under three aspects: human health, ecosystems and resource depletion.

Even if we cannot apply these results to larger EA production plants, the proposed methodology could be applied to any other process at the R&D stage.

The best scenario, only considering LCA, is the first (T = 20 °C, F = 10 Lh^{-1} and P = 350 mmHg). Another parameter that influences the final impact is the water flowrate. This could be noticed by the different impact between the scenarios with the same temperature and pressure but different water flowrate (for example 4 and 24). Pressure influences the quantity of EA produced and does not affect the environment. Monte Carlo analysis revealed that scenarios calculated at 20 °C and at 30 °C are not significantly different. Economic analysis shows that EA price depends mainly on water flowrate, because at higher flowrate, a higher amount of EA is

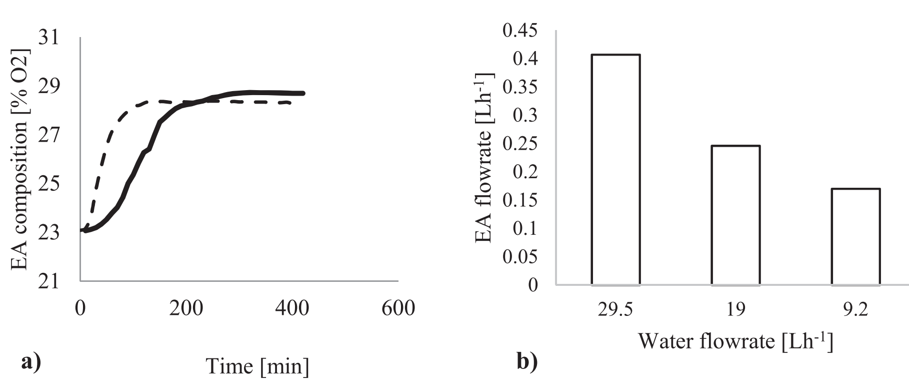

Fig. 3. Experimental results: a) EA oxygen composition at different pressures, 500 mmHg (dotted line) and 300 mmHg (full line) and b) EA flowrate at different inlet flowrates (data obtained at T = 50 °C and P = 300 mmHg).

Table 4
K values, Eq. (1).

	k_1	k_2	k_3	k_4	k_5	k_6	k_7
EA oxygen Molar fraction [-]	6.09E-01 [L^{-1}]	4.46E-02 [$°C^{-1}$]	4.60E-01 [$torr^{-1}$]	−1.29E-03 [$L^{-1}°C^{-1}$]	−1.29E-02 [$L^{-1}torr^{-1}$]	−6.70E-04 [$torr^{-1}°C^{-1}$]	2.73E-05 [$L^{-1}torr^{-1}°C^{-1}$]
EA volume [L]	6.25E-03 [–]	8.98E-05 [$L°C^{-1}$]	1.14E-03 [$Ltorr^{-1}$]	−1.30E-05 [$°C^{-1}$]	−2.62E-04 [$torr^{-1}$]	−6.27E-06 [$Ltorr^{-1}°C^{-1}$]	8.10E-07 [$torr^{-1}°C^{-1}$]
EA price [EUR]	6.20E-03 [$EURL^{-1}$]	3.70E-04 [$EUR°C^{-1}$]	4.15E-03 [$EURtorr^{-1}$]	−1.19E-05 [$EURL^{-1}°C^{-1}$]	−1.19E-04 [$EURL^{-1}torr^{-1}$]	−7.98E-06 [$EURtorr^{-1}°C^{-1}$]	2.55E-07 [$EURL^{-1}torr^{-1}°C^{-1}$]

produced. The best operative parameters for what concerns the economic evaluations are scenarios 23–30. Temperature and pressure do not influence the costs significantly because electricity (OPEX) is inexpensive compared to capital expenses. Scenario 26 is the best when we consider the economic and environmental aspects.

We report the midpoint categories of scenario 26 in Fig. 6.

The highest impact category is that of fossil depletion in which

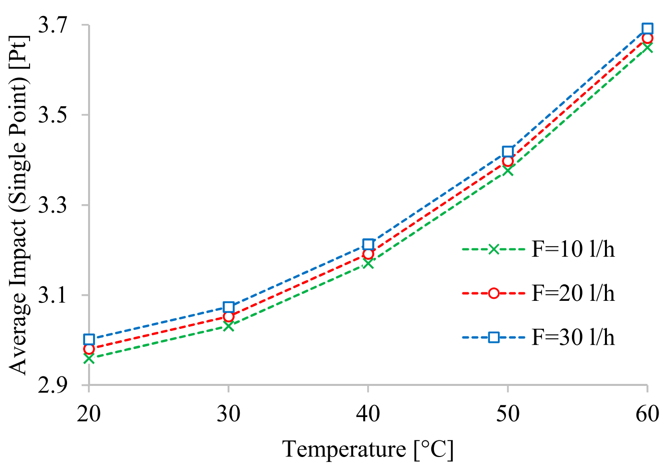

Fig. 4. Environmental impact of lab-scale plant in function of temperature and water flowrate.

electricity (low voltage) accounts for 84 %. This high electric consumption is typical of a lab-scale plant and is not comparable to any industrial scale data, where the main heating medium is steam. On a larger scale plant, we expect the impacts of metal and fossil fuel depletion to decrease since higher volume per unit operation are achieved and heat recovery units are implemented.

The land consumption, together with terrestrial and water pollution are negligible. Freshwater contamination categories are not giving the highest impact since water acts as an absorbent and it does not need to be treated at the end of the process. The weight of the impact categories was similar for all scenarios. Fossil and metal depletions were the most important.

5. Conclusion

In this work, the environmental and economic evaluation of an innovative production process of oxygen-enriched air was performed using two different commercial software, SimaPro 8.1 and PRO/II 9.3. Even though the proposed technology is not industrial-grade, significant results on the assessment methodology could be extrapolated. The results obtained using the two methods, independently, gave different optimal operative conditions. Compared to the economic impact only, the combined optimization leads to a single point impact reduction of 19 % (from 3.69 to 3.00 Pt). On the other hand, considering only the environmental burden, we design a process whose costs are 350 % greater with respect to the eco-economic one.

Any other process can be improved with this new methodology

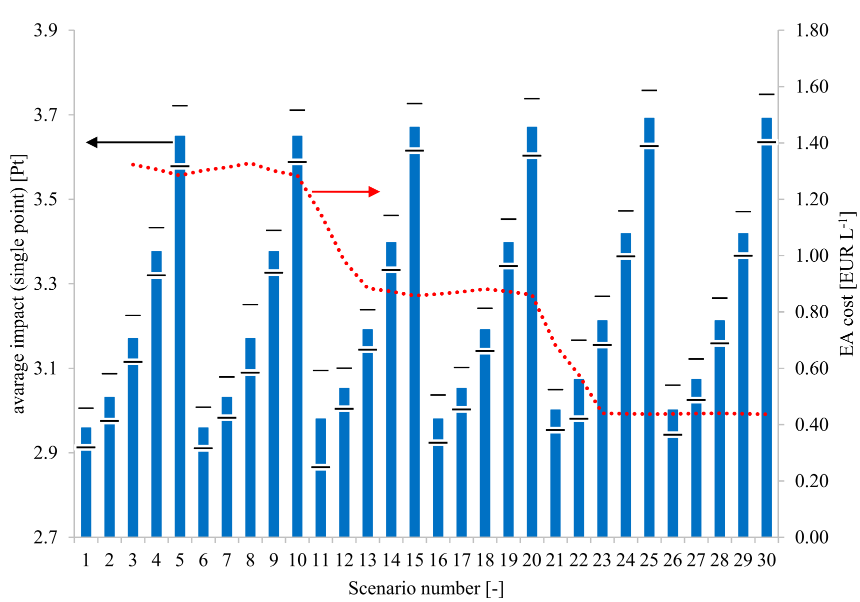

Fig. 5. Economic and environmental results. Average impact (bars, single point with ReCiPe Endpoint E/A) with respective uncertainty, calculated by Monte Carlo analysis, and [-] average EA price per liter.

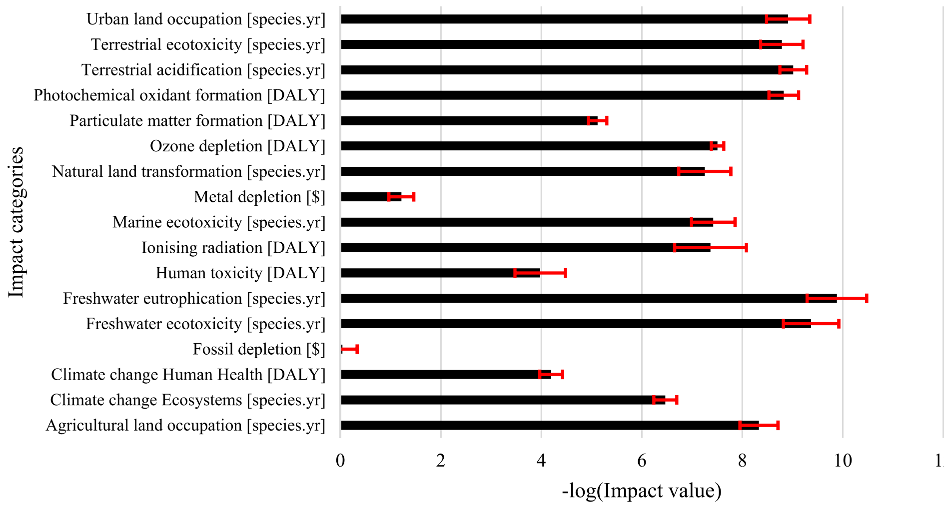

Fig. 6. Midpoint impact results of the LCA analysis performed with the parameters of scenario 26 (water flowrate = 30 Lh^{-1}, T = 20 °C and P = 450 mmHg). The X axis is a negative logarithm, i.e. the higher the value, the lower the impact.

and we recommend the application of this approach at the R&D stage to have a direct and immediate idea of the best process variables to user to avoid environmental and economic hot spots. Low pressures, temperatures and high water flowrates are the best design parameters for the production of EA.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jclepro.2017.09.268.

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