To link to this article : doi:10.1016/j.enconman.2013.03.037
URL : http://dx.doi.org/10.1016/j.enconman.2013.03.037

To cite this version : Ouattara, Adama and Pibouleau, Luc and Azzaro-Pantel, Catherine and Domenech, Serge Economic and environmental impacts of the energy source for the utility production system in the HDA process. (2013) Energy Conversion and Management, vol. 74 . pp. 129-139. ISSN 0196-8904

Economic and environmental impacts of the energy source for the utility production system in the HDA process

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Keywords:
Hydrodealkylation of toluene
Multi-objective genetic algorithm
Energy source
Environmental burdens
Life Cycle Assessment

A B S T R A C T

The well-known benchmark process for hydrodealkylation of toluene (HDA) to produce benzene is revisited in a multi-objective approach for identifying environmentally friendly and cost-effective operation solutions. The paper begins with the presentation of the numerical tools used in this work, i.e., a multi-objective genetic algorithm and a Multiple Choice Decision Making procedure. Then, two studies related to the energy source involved in the utility production system (UPS), either fuel oil or natural gas, of the HDA process are carried out. In each case, a multi-objective optimization problem based on the minimization of the total annual cost of the process and of five environmental burdens, that are Global Warming Potential, Acidification Potential, Photochemical Ozone Creation Potential, Human Toxicity Potential and Eutrophication Potential, is solved and the best solution is identified by use of Multiple Choice Decision Making procedures. An assessment of the respective contribution of the HDA process and the UPS towards environmental impacts on the one hand, and of the environmental impacts generated by the main equipment items of the HDA process on the other hand is then performed to compare both solutions. This "gate-to-gate" environmental study is then enlarged by implementing a "cradle-to-gate" Life Cycle Assessment (LCA), for accounting of emission inventory and extraction. The use of a natural gas turbine, less economically efficient, turns out to be a more attractive alternative to meet the societal expectations concerning environment preservation and sustainable development.

1. Introduction

Utility production largely contributes to energy consumption in process plants and consequently to the operating cost in a scenario of increasing fuel costs. In that context, significant reductions in the consumption of fossil fuels can be achieved by the simultaneous reduction of the combustion emissions in the steam and power generation plant, mainly carbon dioxide helping to comply with Kyoto Protocol (for instance El-Halwagi [1]). In many cases, the dual requirements of power and heating in industrial processes are treated separately: power is purchased from an off-site energy provider and heating is produced on-site through fossil fuel combustion. More precisely, process plants require energy in several forms (mechanical energy, electricity, steam, hot water etc.), which are provided by a variety of sources such as gas-turbine generators, steam-turbine generators, exhaust gas boilers, and fuel-burning boilers. In addition, the utility network serves as a source of additional electricity if needed, or as a sink when excess electricity is produced. The design and operation of utility plants have been tackled by the Process Systems Engineering community for long, particularly with stochastic optimization procedures: for instance, genetic algorithms were successfully applied to the optimization of the operation of a cogeneration system which supplies a process plant with electricity and steam at various pressure levels [2]. For illustration sake, energy management has become an increasingly important component for some kinds of process industries such as the pulp and paper industry. For instance, an analysis of the mill steam production and distribution system has been performed by simulation of various configurations including the incorporation of a back-pressure steam turbine and a condensing steam turbine either alone or in combination [3]. Significant work has been carried out on the synthesis of utility system (for instance, Shang and Kokossis [4,5]). This issue is generally tackled solely from an economic and energy efficiency perspective without considering environmental criteria. More recently, both economic and environmental considerations are included in the general optimization methodology of the synthesis of utility systems.

It must be emphasized that the efforts to limit energy-related environmental emissions lies beyond the process industries. For instance, GSHP systems (also referred to as geothermal heat pump systems, earth energy systems and Geo-Exchange systems) have received major attention as an alternative energy source for

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http://dx.doi.org/10.1016/j.enconman.2013.03.037

Nomenclature

AP	Acidification Potential (t SO_2 equivalent/y)
EP	Eutrophication Potential (t PO_4^{3-} equivalent/y)
FUCA	Faire Un Choix Adéquat – Make an Adequate Choice
GA	Genetic Algorithm
GWP	Global Warming Potential (t CO_2 equivalent/y)
HTP	Human Toxicity Potential (t C_6H_6 equivalent/y)
HDA	hydrodealkylation of toluene
LCA	Life Cycle Assessment
MOOP	Multi-Objective Optimization Problem
MCDM	Multiple Choice Decision Making
NSGA	non dominated sorting genetic algorithm
POCP	Photochemical Ozone Creation Potential (t C_2H_4 equivalent/y)
TOPSIS	Technique for Order Preference by Similarity to Ideal Solution
UPS	Utility Production System

residential and commercial space heating and cooling applications [6,7].

The objective of this paper is to take into account the potential environmental impacts of the energy consumed in a chemical process since energy will have both an environmental impact as well as an economic impact on process design and analysis. For this purpose, the system boundaries must be extended to encompass the power plant which supplies the energy being consumed by the process and incorporating the environmental effects of the power plant into the analysis. This issue has been tackled in the pioneering work of [8]: it involves the development of the WAR algorithm, a methodology for determining the potential environmental impact (PEI) of a chemical process, that was extended to account for the PEI of the energy consumed within that process. But no optimization procedure was embedded in the framework proposed by these authors.

In this work, a particular emphasis will be focused on the antagonist behaviour of the various environmental impacts that may be encountered and to their simultaneous consideration in the resulting optimization problem, thus leading to a multi-objective optimization formulation. This contribution is thus devoted to the presentation of an eco-design approach for process design combining process and utility production modelling, multi-objective optimization, multiple criteria decision aid tools and Life Cycle Assessment.

To support the methodology, this paper deals with the choice of the source of energy either fuel oil or natural gas for the utility production system (UPS) of the classical benchmark HDA (hydrodealkylation of toluene to produce benzene) process [9] by implementing multi-objective optimization.

In a first step, the basic principles of multi-objective optimization are recalled, and the genetic algorithm implemented for this study, namely NSGA IIb, which is an upgraded version of the well-known NSGA II of Deb et al. [10], is presented.

Then, after the Pareto front (set of non dominated solutions) is identified, a subset of good solutions has to be identified among them. This Multiple Choice Decision Making (MCDM) is carried out by implementing two procedures: TOPSIS (Technique for Order Preference by Similarity to Ideal Solution) algorithm [11] and the FUCA (Faire Un Choix Adéquat – Make an Adequate Choice) procedure [12].

The following section concerns the presentation of the HDA process and its utility production system (UPS). The HDA process is modelled according to the principles proposed by Douglas [9], while the UPS and the furnace are modelled as a bi-fuel turbine fed with either natural gas or fuel oil by use of the software ARIANE™ [13]. Furnace and process emission modelling is also carried out by means of ARIANE™ [14,15].

Then, two studies concerning respectively the UPS fed with either fuel oil or with natural gas are performed for a fixed benzene production. In each case, the multi-objective optimization problem involving the total annual cost of the process, and five environmental burdens, namely Global Warming Potential (GWP in t CO_2 equivalent/y), Acidification Potential (AP in t SO_2 equivalent/y), Photochemical Ozone Creation Potential (POCP in t C_2H_4 equivalent/y), Human Toxicity Potential (HTP in t C_6H_6 equivalent/y), Eutrophication Potential (EP in t PO_4^{3-} equivalent/y), is solved. The best solution, identified by means of TOPSIS and FUCA, is then studied both in terms of the respective contributions of the HDA process and of the UPS on environmental impacts as well as of the environmental impacts of the main equipment items of the HDA process. This "gate-to-gate" environmental study is then enlarged by performing a "cradle-to-gate" Life Cycle Assessment (LCA), for accounting of emission inventory and extraction.

Finally, the choice between fuel oil and natural gas turbines is performed according to economic objective, environmental impacts and LCA analysis.

2. Multi-objective optimization

When dealing with process optimization, the current trend is to consider additional objectives to the traditional economic criterion, which means criteria related to sustainability, concerning more precisely environment and safety. In many engineering fields, most of process optimization problems became multi-objective optimization problems (MOOPs).

A MOOP can be formulated as:

$$\text{Min } F(x) = \left[f_1(x), f_2(x), \ldots, f_p(x)\right]^T \tag{1}$$

$$\text{where } x \in X \subset R^n \tag{2}$$

The subspace X is defined by a set of equality-inequality constraints (linear, nonlinear, differential) and bounds on variables:

$$X = \{x \in R^n / g_i(x) \leqslant 0, i = 1 \text{ to } r; h_j(x) = 0, j = 1 \text{ to } s; l(i) \leqslant x(i) \leqslant u(i)\} \tag{3}$$

In a MOOP, the concept of optimality is replaced by efficiency or Pareto optimality. The efficient (or Pareto optimal, non dominated, non-inferior) solutions are the solutions that cannot be improved in one objective function without deteriorating their performance in at least one of the rest. The mathematical definition of an efficient solution is the following: a feasible solution x^* of a MOOP is efficient (non dominated) if there is no other feasible solution x such as:

$$f_i(x) \leqslant f_i(x^*) \forall i \in \{1, \ldots, p\} \tag{4}$$

with at least one strict inequality.

According to de Weck [16], there is general consensus that multi-objective optimization methods can be broadly decomposed into two categories: scalarization approaches and evolutionary methods. From a popular classification, scalarization methods, where the multi-objective problem is transformed into a mono-objective one, apply in well mathematically defined problems with explicit formulations of objectives and constraints, while evolutionary methods are mainly used in black box problems, where objectives and/or constraints are evaluated by an external computer code for each value of the optimization variables. In evolutionary methods, the elements of the objective function vector are kept separate throughout the optimization process; these approaches typically use the concept of dominance to distinguish between dominated and non-dominated solutions.

Indeed, black box problems are classical situations in chemical engineering applications where heat and mass balances lead to complex sets of nonlinear equations; furthermore, energy balances may produce ordinary differential equations. Besides the black box problem feature, the possibility to mutate out of a local optimum and the ability to compute the entire Pareto front in one run, make also this type of methods attractive; this explains why they have been considered in this study.

The literature survey [17–20] reveals that evolutionary algorithms, derived by observing the process of biological evolution in nature, have proven to be a powerful and robust optimizing technique in many cases. Among evolutionary methods, genetic algorithms are generally attractive methods in the chemical engineering community, particularly when the evaluation functions are computed by a flowsheeting software tool.

Indeed, the use of evolutionary algorithms for simultaneous structural and parameter optimization in process synthesis in a modular program environment [21] was identified as particularly interesting. In the abovementioned work, the commercial simulator ASPEN PLUS™ was integrated for the determination of the target function value. The simulations and the cost calculations embed the complete process modeling accuracy without the necessity of simplifications due to restrictions imposed by the optimization method.

The application of a multi-objective genetic algorithm with constraints concerns other classical problems in chemical enginering, for instance, the optimization of Petlyuk sequences in distillation [22]. A multi-objective genetic algorithm (GA) with constraints was formulated and coupled with the Aspen Plus process simulator to obtain each data point during the search process. In addition to providing more energy-efficient designs than some reported structures, the analysis highlights first that the feed location in the prefractionator can be expressed as a function of the mixture properties, and second the optimal structures requires four interconnecting stages instead of the two normally used for Petlyuk sequences. The GA exhibited a robust performance, and was practically independent on the initial values for the search variables.

Another interesting contribution [23] is relative to the implementation of an optimization framework for the synthesis and design of complex distillation sequences, based on a modified genetic algorithm (GA) coupled with a sequential process simulator. The use of a simulator facilitates the formulation of rigorous models for different process alternatives, while the genetic algorithm allows the solutions of the complex non-convex mathematical problem, involving discrete and continuous decisions. The GA strategy succeeds in problems where deterministic mathematical algorithms had failed.

One of the most efficient genetic algorithms is NSGA II [10], an upgrade of NSGA which estimates the density of solutions surrounding a particular one. From Coello Coello and Becerra [24], its performance is so good, that it has gained a lot of popularity in the last few years.

3. Algorithm NSGA IIb

The well-known NSGA II (Non Sorted Genetic Algorithm) of Deb et al. [10] developed for multi-objective continuous problems was used as the basis case for further algorithmic development of NSGA IIb [25]. This elitist procedure lies on a ranking procedure. The population is sorted based on non-domination into each front. The first front being completely non-dominant is placed into the current population and the second front is dominated by the individuals in the first front only and the front goes so on. A rank is thus assigned to each individual in each front, that is to say that the rank is based on the front the individual belongs to. In other words, this elitist procedure lies on a ranking procedure, where the rank of each solution represents the number of times that a solution is dominated (rank one corresponds to non dominated solutions, rank two corresponds to the solutions that are only dominated once and so on).

A crowding distance factor defined as the size of the largest cuboid enclosing a given solution without including any other one, guarantees the genetic diversity of the generated solutions.

3.1. Initial population generation in NSGA IIb procedure

Two options are provided for the generation of an initial population. The classical one, based on a purely random generation, may produce over-crowded or under-crowded zones in the search space. Another more efficient solution consists in meshing the range of bounded variables, and randomly generating the same number of points into each cuboid of the grid in order to ensure a uniform overlapping of the search space [25]. A forced mutation is activated for clones in each cuboid, so that all the initial solutions are different.

3.2. NSGA IIb procedure

This algorithm uses the same SBX crossover (Simulated Binary Crossover, Deb and Agrawal [26]) operator as in NSGA II, but when the crossover generates two children identical to the parents, a forced mutation of children occurs. The goal is to avoid unnecessary calculations of both objective functions and constraints of clone solutions that have been already evaluated. All the solutions generated by the reproduction scheme are different.

3.3. Constraint handling

The strategy proposed by Deb et al. [10] is used for inequality constraint handling. The procedure consists in comparing the sum of violated constraints for establishing the first domination ranking. This step is performed first, before comparing the objective function values in order to determine the final ranking.

For a problem involving n variables and m ($m < n$) equality (either linear or nonlinear) constraints, the analysis of degrees of freedom gives n–m independent variables. After scrutinizing the constraint set, these n–m decision variables can be chosen. For each evaluation of an objective function, the system of m equations must be solved. It must be highlighted that about 70% of CPU time is spent in solving the equality constraints.

3.4. Numerical procedure implementation

The VBA/MATLAB platform was a constraint imposed by our industrial partner (CEA, Commissariat à l'Energie Atomique – French Agency for Nuclear Energy) for NSGA IIb implementation [25]. In the following examples, the sets of linear/nonlinear equality constraints due to balance equations are solved at each move of the genetic algorithm by the Newton–Raphson procedure FSOLVE of the MATLAB toolbox.

The GA parameters that were used are the following ones: 200 individuals per generation, 200 generations, a SBX crossover procedure with probability of 0.75 and a mutation probability of 0.2. As the GA is a randomly initialized search, each problem is run 20 times. Among the generated Pareto fronts, the most "rich" front corresponding to the highest number of points is conserved. Indeed, the choice may be sometimes quite difficult. Another strategy would consist in merging the 20 fronts, and performing a Pareto sort on the final front. This strategy was implemented on each numerical example, and no significant difference exists between both solutions. The first strategy, which is less greedy in computational time, was finally adopted.

4. Choice of the best solutions

Once the complete set of solutions of the multi-objective optimization problem (i.e. the Pareto front or set of efficient solutions) is found, the next step consists in identifying the best ones. The MCDM (Multiple Choice Decision Making) issue is a complex problem, mainly because of its more subjective nature, than the multi-objective optimization problem itself.

Some generic tools, like the TOPSIS (Technique for Order Preference by Similarity to Ideal Solution) algorithm [11] or the FUCA (Faire Un Choix Adéquat – Make an Adequate Choice) procedure [12] are used.

The two procedures are described in Ouattara et al. [14,15] and will not be presented here by the sake of brevity. However, for industrial problems, the practitioner may make his final decision according to some specific internal features of his company.

5. The HDA process and the utility production system (UPS)

A classical method of manufacturing benzene from the distillation of light oils is the hydrodealkylation (HDA) of toluene [9]. HDA process has been used intensively both in education and research to illustrate fundamental issues in Process Systems Engineering, such as process synthesis and energy integration, as well as in integrating design and control [27–29]. This can be considered as an asset since there is no limitation due to process data.

This process involves two reactions: the conversion of toluene to benzene and the equilibrium between benzene and biphenyl.

$$\text{Toluene} + H_2 \rightarrow \text{Benzene} + \text{CH}_4$$

$$2\ \text{Benzene} \leftrightarrow \text{Biphenyl} + \text{H}_2$$

This well-known benchmark problem for process design and synthesis studies, was first extensively studied by Douglas [9] using a hierarchical design/synthesis approach, and Turton et al. [30,31]. The hydrogen feed stream has a purity of 95% and involves 5% of methane; this stream is mixed with a fresh inlet stream of toluene, recycled toluene, and recycled hydrogen. The feed mixture is heated in a furnace before being fed to an adiabatic reactor. The reactor effluent contains unreacted hydrogen and toluene, benzene (the desired product), biphenyl, and methane; it is quenched and subsequently cooled in a high-pressure flash separator to condense the aromatics from the non-condensable hydrogen and methane. The vapour steam from the high-pressure flash unit contains hydrogen and methane that is recycled. The liquid stream contains traces of hydrogen and methane that are separated from the aromatics in a low-pressure flash drum. The liquid stream from the low-pressure flash drum consisting of benzene, biphenyl and toluene is separated in two distillation columns. The first column separates the product, benzene, from biphenyl and toluene, while the second one separates the biphenyl from toluene, which is recycled back at the reactor entrance. Energy is saved by using the outlet stream leaving the reactor as its temperature is in the range of 620 °C, to preheat the feed stream coming from the mixer, via a heat exchanger (Fehe), so some energy integration is achieved [32] (see Fig. 1).

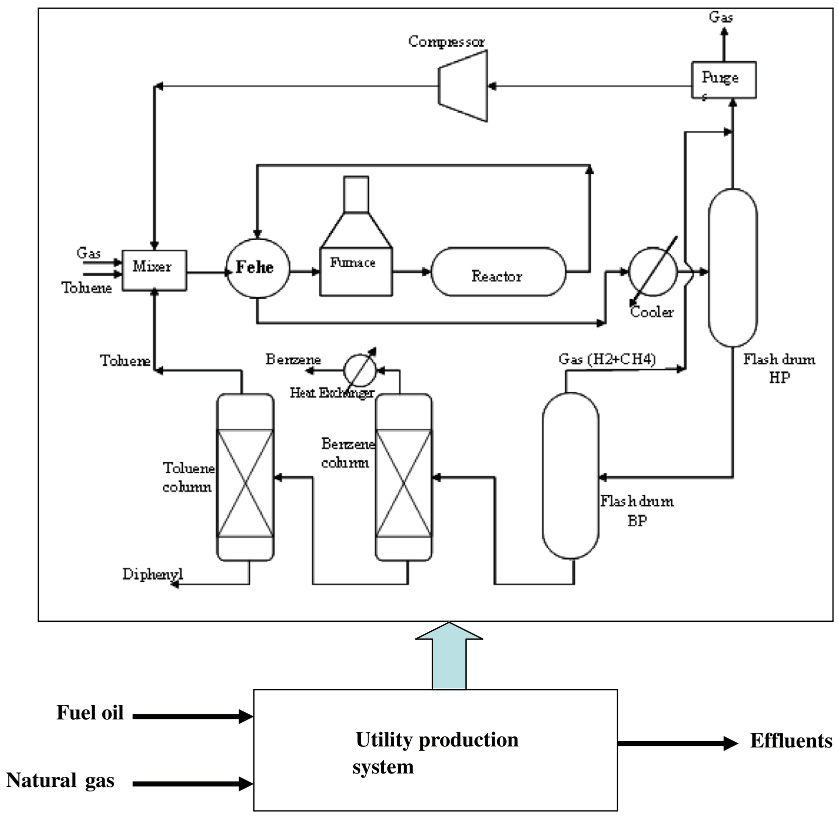

Fig. 1. HDA process coupled with the UPS.

Both the UPS and the furnace are modelled as a bi-fuel turbine fed with either natural gas or fuel oil by using the software ARIANE™ [13], which has been developed by ProSim Company (French Chemical Engineering Software Company) for designing optimal operation of power plants. Furnace and process emission modelling is also carried out by means of ARIANE™ [6].

6. Multi-objective optimization of the HDA process – fuel oil turbine

6.1. Problem formulation

For a fixed benzene production (300 kmol/h), the multi-objective optimization problem is defined as:

$$\text{Min (annual cost)} \tag{5}$$

$$\text{Min } (\text{EI}_i), i = 1, 5 \tag{6}$$

s.t.
Mass and energy balances (Excel® and ARIANE™).
Bounds on decision variables.

Among the environmental impacts (EIs) proposed by IChemE [33] and Azapagic et al. [34], five representative environmental impacts have been considered: Global Warming Potential (GWP in t CO_2 equivalent/y), Acidification Potential (AP in t SO_2 equivalent/y), Photochemical Ozone Creation Potential (POCP in t C_2H_4 equivalent/y), Human Toxicity Potential (HTP in t C_6H_6 equivalent/y), Eutrophication Potential (EP in t PO_4^{3-} equivalent/y). A more detailed discussion can be found in [6].

Based on the works of Douglas [9] and Turton et al. [30], the bounds on decision variables are indicated in Table 1.

The environmental burdens GWP, POCP and HTP can be expressed as multilinear functions of annual cost (AC), EP and AP in [14]. A set of 200 values of decision variables was randomly generated between the bounds defined in Table 1, and the corresponding objectives AC, EP, AP, GWP, HTP and POCP were computed. A multilinear regression was carried out using the Excel toolbox between independent objectives AC, EP and AP and dependent ones, GWP, HTP and POCP. As it is shown by the coefficient correlation values of Table 2, the multi-linear expression is very good.

So the initial six-objective problem can be reduced to a tri-objective one solved by use of NSGA IIb. The problem formulation is detailed in [14].

Table 1
Bounds on decision variables.

Decision variables	Lower bound	Upper bound
Toluene conversion rate	0.5	0.9
Purged hydrogen flow rate (kmol/h)	31	308
HP flash pressure (bar)	30	34
LP flash pressure (bar)	4	10
Pressure column 1 (bar)	2	4
Pressure column 2 (bar)	1	2
Ratio fuel flowrate/gas flowrate at the furnace	0.1	0.9

6.2. Problem solution

The Pareto front provided by NSGA IIb and reported in [14] is displayed in Fig. 2. The flat portion of the cloud of points near annual cost ≈ 205 M$/y, EP ≈ 10,000 t PO_4^{3-}/y and AP ≈ 5000 t SO_2/y suggests that good solutions may exist in this zone for the three objectives.

A TOPSIS and a FUCA analysis are carried on the global set of objectives (annual cost, EP, AP, GWP, HTP and POCP). The two best solutions obtained from TOPSIS (respectively FUCA) are called TT1 and TT2 (respectively TF1 and TF2) on the 3D curve. The two procedures give results that are in agreement with the simple graphical analysis.

The gains provided by solutions TT1, TT2, TF1, TF2 versus a non optimized solution, called Douglas_{300}, where the decision variables are those used by Douglas [9] for a benzene production updated at 300 kmol/h, are given in Table 3. According to the mean gain, the solutions provided by FUCA are much better than those obtained by TOPSIS, and the solution TF2 is slightly better than TF1. As it was observed on numerous numerical examples treated in the research group, the FUCA method gives always better results than the TOPSIS procedure. This explains why only the FUCA method is implemented for determining "good" solutions in the following section devoted to the UPS of the HDA process fed by natural gas.

The decision variables for solutions TT1, TT2, TF1, TF2 and Douglas_{300} are reported in Table 4. The main differences between solutions TT1, TT2, TF1 and TF2 and the reference case Douglas_{300} concern the purged hydrogen, the column 1 pressure and the ratio fuel/gas in the furnace.

The design parameters for the main equipment items of the HDA process are presented in detail in Table 5. The main differences between the two groups of solutions are related to the furnace power, the reactor volume, the HP flash pressure, the height and diameter of column 1, the heat exchanger area and the compressor power.

An additional comparison is carried out by reporting solutions TT1, TT2, TF1, TF2 and Douglas_{300} on a normalized radar graph as shown in Fig. 3. The solution Douglas_{300} is deficient for objectives AC, AP, GWP and POCP, while TF1 is worst for EP and HTP. TF1 and TF2 give the lowest values for all the objectives.

The solution TF2 being slightly better than TF1 for the mean gain is adopted for the following studies. This solution corresponds to annual cost = 209 M$/y, EP = 9770.4 t PO_4^{3-} equivalent/y, AP = 4782.5 t SO_2 equivalent/y, GWP = 1,432,472 t CO_2 equivalent/y, HTP = 19,370.4 t C_6H_6 equivalent/y and POCP = 1928.7 t C_2H_4-equivalent/y.

6.3. Environmental impacts of the HDA process and the UPS for the solution TF2

The solution TF2 is now analyzed in terms of contribution of the HDA process and the UPS on environmental impacts. The results are displayed in Fig. 4.

The HDA process only contributes to HTP and EP impacts, while the UPS is involved alone in AP impact while HDA and UPS both contribute to GWP and POCP. The contribution of the HDA process

Table 2
Multilinear regression (fuel oil – steam turbine).

Objective	AC (M$/y)	EP (t PO_4^3 eq$^-$/y)	AP (t C_2H_4 eq/y)	*y* (constant term)	Coef. corr.	Max error (%)
GWP (t CO_2 eq/y)	5445.29	−0.64	43.95	90,529.67	0.9988	0.51
HTP (t C_6H_6 eq/y)	-6.2910^{-5}	1.92	-3.7010^{-5}	9.2510^{-3}	1.000	10^{-6}
POCP (t C_2H_4 eq/y)	0.44	4.0710^{-3}	8.6710^{-2}	1377.95	0.9999	0.03

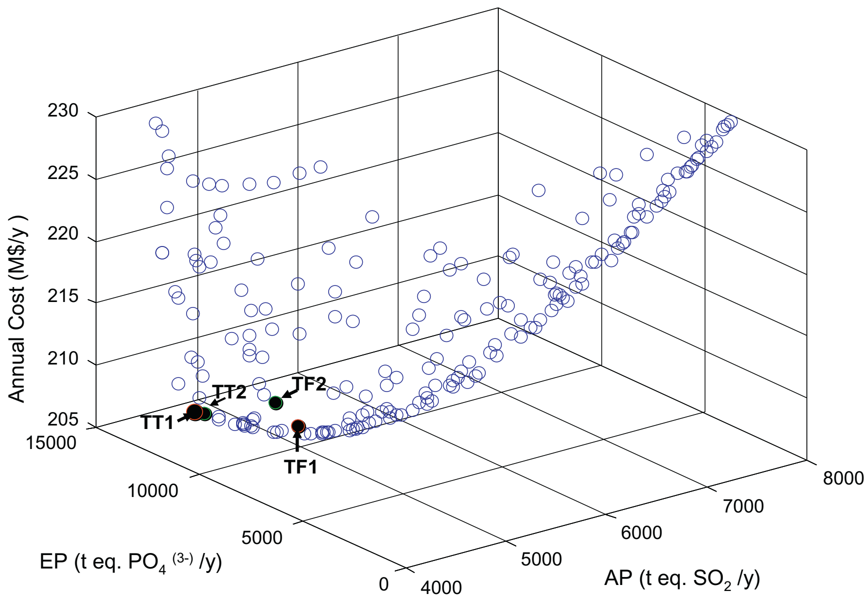

Fig. 2. Tri-objective optimization (annual cost, EP, AP) – fuel oil turbine.

Table 3
Comparison of solutions TT1, TT2, TF1 and TF2 vs. Douglas_{300}.

Solutions	Annual cost (M$/y)	EP (t PO_4^3 eq/y)	AP (t SO_2 eq/y)	GWP (t CO_2 eq /y)	HTP (t C_6H_6 eq/y)	POCP (t C_2H_4 eq/y)	Mean gain
Douglas_{300}	327	9759.1	14,777.6	2,175,735.0	18,699.6	2794.3	
TT1	205.3	13,954.3	4759.1	1,408,841.5	26,737.7	1940.8	
Gain (%)	37.2	−43.0	67.8	35.2	−43.0	30.6	14.1
TT2	205.32	13,671.8	4792.3	1,410,408.7	26,196.4	1942.5	
Gain (%)	37.2	−40.1	67.6	35.2	−40.1	30.5	15.1
TF1	207	10,109.4	4930.2	1,428,118.4	18,720.8	1939.3	
Gain (%)	36.7	−3.6	66.6	34.4	−0.12	30.6	27.4
TF2	209.0	9770.4	4782.5	1,432,472.4	19,370.4	1928.7	
Gain (%)	36.1	−0.12	67.6	34.2	−3.6	31.0	27.5

Table 4
Values of decision variables.

Decision variables	TT1	TT2	TF1	TF2	Douglas_{300}
Toluene conversion rate	0.80	0.79	0.75	0.76	0.75
Purged hydrogen flow rate (kmol/h)	300	300	300	300	198
HP flash pressure (bar)	34.0	33.9	33.9	33.9	34.4
LP flash pressure (bar)	10.0	10.0	9.7	10.0	10.3
Pressure column 1 (bar)	3	3	3	3	1
Pressure column 2 (bar)	1.2	1.7	1.1	1.0	1.0
Ratio fuel flowrate/gas flowrate at the furnace	0.44	0.45	0.40	0.34	0.90

Table 5
Design parameters for equipment items of the HDA process – fuel oil steam-turbine.

Equipment items	TT1	TT2	TF1	TF2	Douglas_{300}
Furnace power (GJ/h)	93.1	93.4	98.4	97.8	121.5
Reactor volume (m^3)	173.3	173.0	170.3	170.4	251.6
HP flash volume (m^3)	22.9	23.0	24.8	24.6	38.7
LP flash volume (m^3)	2.3	2.3	2.5	2.4	2.9
Column 1: height (m)	42.4	42.4	43.0	43.0	36.9
Diameter (m)	2.9	2.9	2.9	2.9	3.7
Column 2: height (m)	18.0	18.6	18.0	18.0	18.0
Diameter (m)	1.7	1.6	1.9	1.9	1.9
Heat exchanger area (m^2)	525.9	527.1	554.4	551.0	668.7
Compressor power (kW)	100.0	100.3	107.1	106.2	145.1
Feed pump power (kW)	2.5	2.5	2.6	2.6	2.6
Recycle pump power (kW)	11.1	11.0	14.5	14.2	14.6

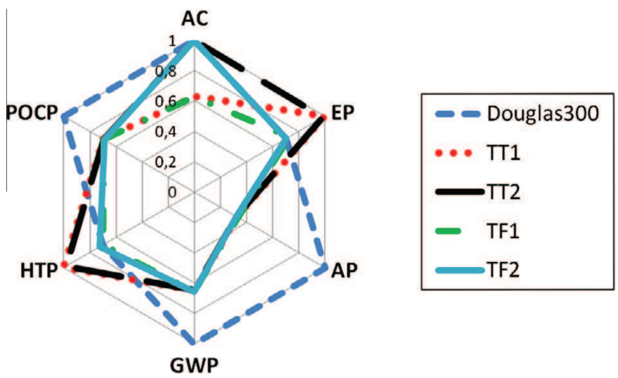

Fig. 3. Radar graph for solutions TT1, TT2, TF1, TF2 and Douglas_{300}.

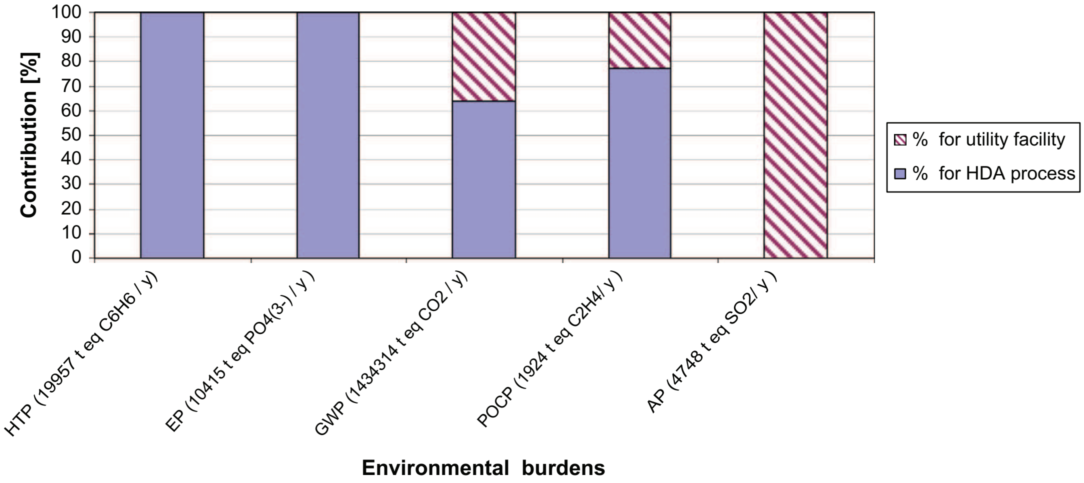

Fig. 4. Environmental impacts of the HDA process and the UPS for solution TF2.

in GWP and POCP is quite significant (65% and 78% respectively), mainly due to the purged methane, since the impact factor of methane related to GWP is 21 times higher than the one of carbon dioxide.

6.4. Environmental impacts of unit operations of the HDA process for the solution TF2

In this section, the environmental impacts of main equipment items of the HDA process for the solution TF2 are studied. The results are reported in Fig. 5. The impacts HTP and HP are only generated by column 2, due to the biphenyl emissions. The GWP high value is attributed to the purged methane.

6.5. Life Cycle Assessment (LCA) of the TF2 solution

The "gate-to-gate" environmental study carried out in the previous section is now enlarged by performing a "cradle-to-gate" LCA [35], for accounting of emission inventory and extraction. The package SimaPro 7.1 [36] with the database EcoInvent [37] is implemented for this purpose; the life cycle impact assessment methodology IMPACT 2002 + based on a combined midpoint/damage-oriented approach was selected [38]. The data for LCA are given in Table 6. In this LCA, only intermediate categories are considered.

Table 6
Data for LCA of TF2 solution.

	TF2
Benzene production (kmol/h)	300
Effluents of HDA process	
Purged hydrogen (kmol/h)	300.0
Purged methane (kmol/h)	341.7
Biphenyl flow rate (kmol/h)	4.9
Energy consumption	
Fuel oil flow rate in furnace (t/h)	4.1
Natural gas flow rate in furnace (Nm^3/h)	8375.7
Fuel oil flow rate in boiler (t/h)	10.5
Raw materials	
Toluene flow rate (kmol/h)	309.8
Hydrogen flow rate (kmol/h)	604.9
Methane flow rate (kmol/h)	31.8
Water consumption	
Water flow rate (kg/s)	1117.9
Pollutant emissions	
CO_2 emission (kg/h)	61,722.9
SO_2 emission (kg/h)	597.8
NO_x emission (kg/h)	0.09
CO emission (kg/h)	972.7
Dust emission (kg/h)	0.02

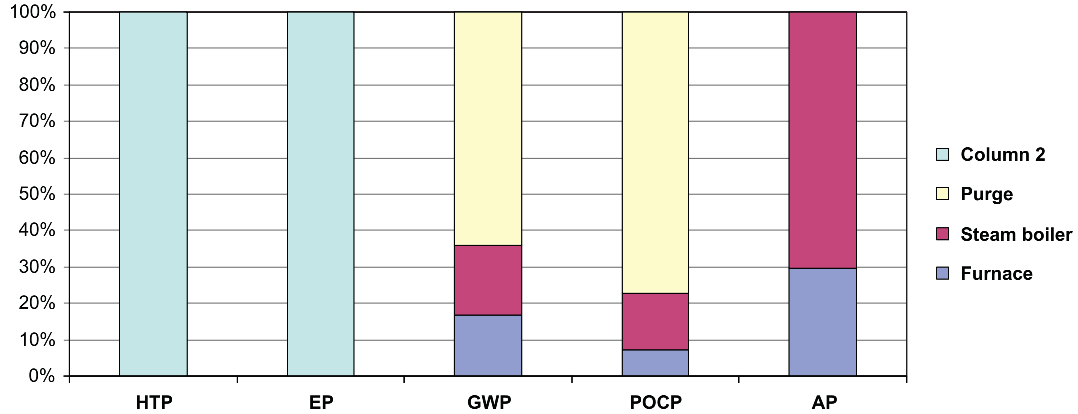

Fig. 5. Environmental impacts of unit operations for solution TF2.

Table 7
Multilinear regression (natural gas turbine).

Objective	AC (M$/y)	EP (t PO_4^3 eq^-/y)	AP (t C_2H_4 eq/y)	*y* (constant term)	Coef. corr.	Max error (%)
GWP (t CO_2 eq/y)	5630.29	−0.68	45.00	901,850.16	0.9977	0.45
HTP (t C_6H_6 eq/y)	-7.3210^{-5}	1.98	-2.9110^{-5}	7.7510^{-3}	0.9988	10^{-6}
POCP (t C_2H_4 eq/y)	0.48	4.3810^{-3}	7.7810^{-2}	1477.65	0.9999	0.06

This LCA study shows that the predominant midpoint environmental categories are Global Warming Potential, followed by non-renewable energy, inorganic respiratory impact, and carcinogens, the other items being quasi-null or null (see Table 12).

7. Multi-objective optimization of the HDA process – natural gas turbine

7.1. Problem formulation

In this section, the influence of the utility production system is studied by replacing the system boiler – steam turbine (which used fuel oil in the previous section) by a gas turbine, also simulated by use of Ariane™ [13]. For a benzene production always fixed at 300 kmol/h, the problem formulation, the parameters of the NSGA IIb algorithm and the bounds on decision variables are the same as in the previous section.

The multilinear regressions for expressing GWP, HTP and POCP in terms of AC, EP and AP were carried out again according to the same procedure as the one described above. The results reported in Table 7 are close to the ones obtained in the case of a fuel oil turbine. From the values of the correlation coefficient and the max error between the experimental points and those predicted by the model, it can be said that the regression is very good.

Table 9
Values of decision variables.

Decision variables	TF3	TF4	TF5	TF2
Toluene conversion rate	0.74	0.74	0.75	0.76
Purged hydrogen flow rate (kmol/h)	299.9	299.8	299.8	300
HP flash pressure (bar)	31.0	31.0	30.0	33.9
LP flash pressure (bar)	8.1	9.1	8.8	10.0
Pressure column 1 (bar)	2.2	2.8	2.3	3
Pressure column 2 (bar)	1.4	1.4	1.8	1.0
Ratio fuel flowrate/gas flowrate at the furnace	0.39	0.37	0.37	0.34
Turbine chamber pressure (bar)	15.27	13.70	15.22	–

7.2. Problem solution

The Pareto front provided by NSGA IIb is displayed in Fig. 6. The solutions ranked 1, 2 and 3 by the FUCA method correspond to points named TF3, TF4 and TF5 in the 3D curve of Fig. 6.

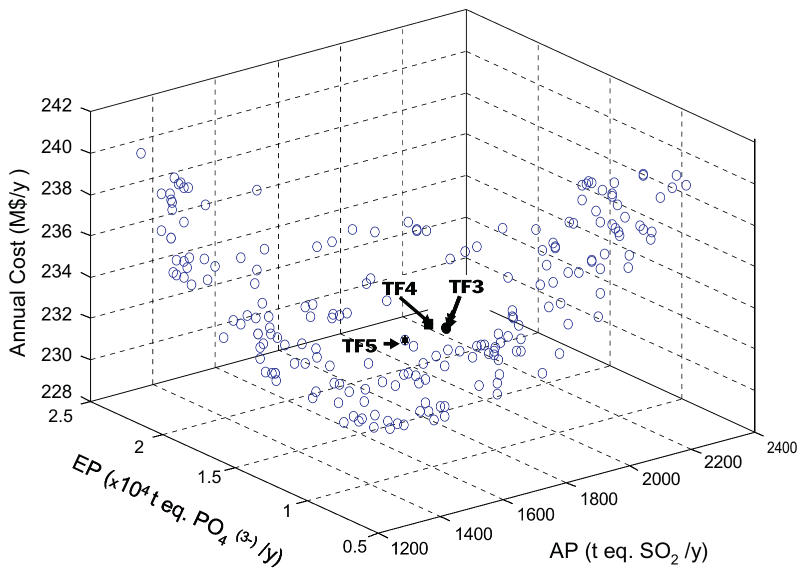

Fig. 6. Tri-objective optimization (annual cost, EP, AP) – natural gas turbine.

Table 8
Comparison of solutions TF3, TF4, TF5 with TF2.

Solution	AC (M$/y)	EP (t PO_4^3 eq^-/y	AP (t SO_2 eq/y)	GWP (t CO_2 eq/y)	HTP (t C_6H_6 eq/y)	POCP (t C_2H_4 eq/y)	Mean gain
TF2	209.0	9770.4	4782.5	1,432,472.4	19,370.4	1928.7	
TF3	234.9	9252.5	1609.6	1,250,006.7	17,729.3	1644.1	
Gain (%)	−12.4	5.3	66.3	12.7	8.5	14.8	16.4
TF4	235.3	9252.5	1554.7	1,251,164.1	17,729.3	1639.63	
Gain (%)	−12.6	8.5	67.5	12.7	8.5	15.0	16.6
TF5	234.3	10,413.8	1535.0	1,247,782.2	19,954.5	1642.5	
Gain (%)	−12.1	−3.0	67.9	12.9	−3.0	14.8	12.9

Table 10
Design parameters for equipments of the HDA process.

Equipment	TF3	TF4	TF5	TF2
Furnace power (GJ/h)	99.71	99.60	98.47	97.8
Reactor volume (m^3)	170.26	170.30	170.39	170.4
HP flash volume (m^3)	27.11	27.03	27.23	24.6
LP flash volume (m^3)	0.02	0.02	0.02	2.4
Column 1 height (m)	41.76	42.98	41.76	43.0
Diameter (m)	3.15	2.99	3.11	2.9
Column 2 height (m)	17.98	17.98	18.59	18.0
Diameter (m)	1.84	1.83	1.71	1.9
Heat exchanger area (m^2)	561.17	560.54	554.34	551.0
Compressor power (kW)	108.80	108.67	107.16	106.2
Feed pump power (kW)	2.66	2.65	2.63	2.6
Recycled pump power (kW)	15.19	15.11	14.27	14.2

The gains provided by solutions TF3, TF4 and TF5 versus the best solution TF2 found for the fuel oil turbine are given in Table 8. According to the mean gain, the three solutions provide a significant gain versus TF2, the solutions TF3 and TF4 are better than TF5, TF4 being slightly better than TF3.

In Table 9, the decision variables for solutions TF3, TF4, TF5 and TF2 are reported. The main differences between the two groups of solutions concern the HP and LP pressure of the flash and the pressure of the columns 1 and 2.

The design parameters for the main equipment items of the HDA process are displayed in Table 10. The power generated by the turbine for the solutions TF2 and TF4 is respectively 9.7 kW and 22.3 kW.

As in the previous case, the results can be visualized by reporting a normalized radar graph for solutions TF3, TF4, TF5 and TF2 as shown in Fig. 7.

Finally, for the natural gas turbine, the choice has to be carried out between solutions TF3 and TF4. The solution TF4 being slightly better than TF3 according to the mean gain will be adopted for the following studies. This solution corresponds to annual cost = 235.3 M$/y, EP = 9252.5 t PO_4^{3-} equivalent/y, AP = 1554.7 t SO_2 equivalent/y, GWP = 1,251,164.1 t CO_2 equivalent/y, HTP = 17,729.3 t C_6H_6 - equivalent/y and POCP = 1639.6 t C_2H_4 equivalent/y.

At this point, the final choice has to be performed between the solution TF2, which exhibits a better economic performance, and TF4, which is better from environmental aspects. As in the previous case, the environmental impacts of the HDA and UPS processes and the LCA of the TF4 solution are detailed in the following subsections.

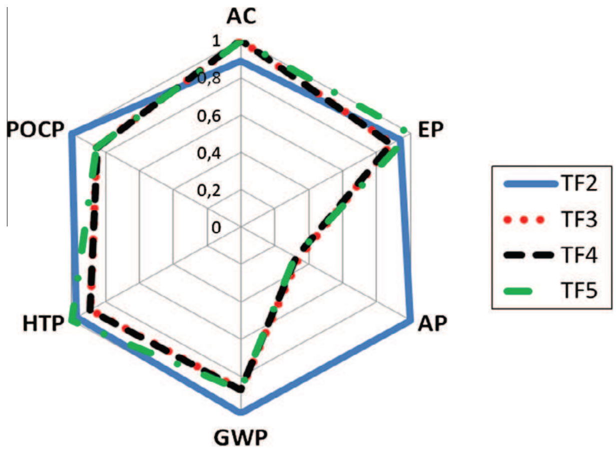

Fig. 7. Radar graph for solutions TF2, TF3, TF4 and TF5.

7.3. Environmental impacts of the HDA process and the UPS for solution TF4

The solution TF4 is now analyzed in terms of contribution of both the HDA process and the UPS on environmental impacts. The results are displayed in Fig. 8. Compared with the UPS using fuel oil (Fig. 4), the contribution of the UPS of the GWP decreases from 35% to 31%, and the one of POCP decreases from 22% to 10%.

7.4. Environmental impacts of unit operations of the HDA process for solution TF4

In this section, the environmental impacts of some equipment items of the HDA process for the solution TF4 are studied. The results are reported in Fig. 9. The contributions of the UPS in environmental burdens GWP, POCP and AP lead to significant reductions (from 20% to 10% for GWP, 14–1% for POCP and 69–5% for AP) versus the fuel oil turbine case (Fig. 5).

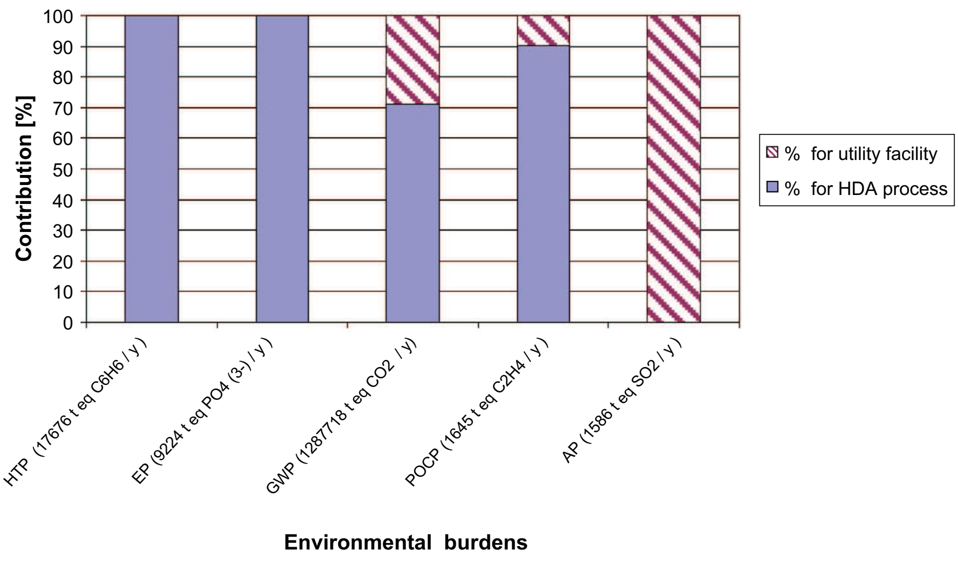

Fig. 8. Environmental impacts of the HDA process and the UPS for solution TF4.

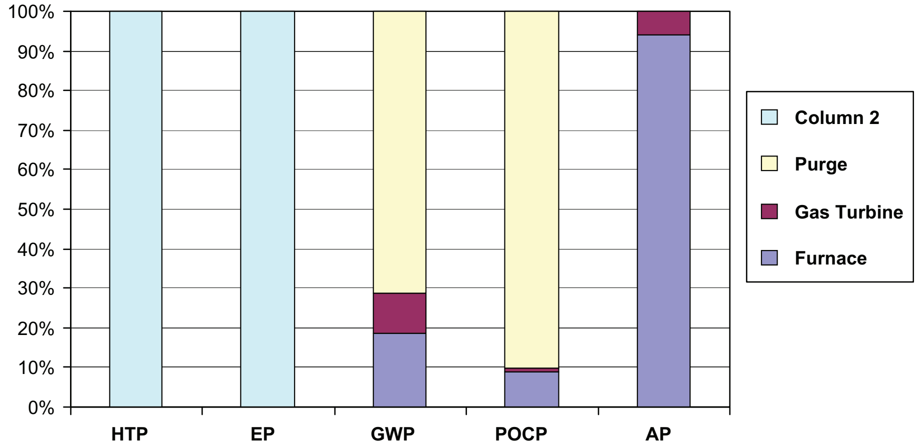

Fig. 9. Environmental impacts of unit operations for the solution TF4.

Table 11
Data for LCA of TF4 solution.

	TF4
Benzene production (kmol/h)	300
Effluents of HDA process	
Purged hydrogen (kmol/h)	300
Purged methane (kmol/h)	340.8
Biphenyl flow rate (kmol/h)	4.48
Energy consumption	
Fuel oil flow rate in furnace (t/h)	2.76
Natural gas flow rate in furnace (Nm^3/h)	7933.42
Natural gas flow rate in gas turbine (t/h)	6010.30
Raw materials	
Toluene flow rate (kmol/h)	308.96
Hydrogen flow rate (kmol/h)	604.28
Methane flow rate (kmol/h)	31.8
Water consumption	
Water flow rate (kg/s)	1135.10
Pollutant emissions	
CO_2 emission (kg/h)	40,255.65
SO_2 emission (kg/h)	133.48
NO_x emission (kg/h)	0.02
CO emission (kg/h)	297.60

7.5. LCA of the TF4 solution

A cradle-to-gate LCA is now performed on the solution TF4; the data are given in Table 11. As in the previous case, only intermediate categories are considered. This LCA study shows that, as in the previous case, the predominant impact categories are nonrenewable energy, followed by Global Warming Potential, inorganic respiratory impact, and carcinogens, the other items being quasi-null or null. As it can be highlighted in Table 12, the solution TF4 is much better than the solution TF2 from a LCA point of view.

8. Discussion

The problem remains to choose between solution TF2 corresponding to a fuel oil steam turbine used for the UPS, and TF4 related to a natural gas turbine for the UPS. Solution TF2 is better than TF4 on an economic point of view (12.6%, see Table 8) while TF4 is better than TF2 regarding environmental impacts AC, EP, GWP, HTP and POCP (mean gain in the five objectives of 21.5%).

A more thorough study concerning the environmental impacts of the HDA process and the UPS shows that the UPS using a natural gas turbine leads to a decrease in 4% in the GWP and 12% in the POCP (Figs. 4 and 8). This trend was further confirmed by a more detailed analysis of environmental impacts of unit operations of the HDA process (Figs. 5 and 9); the contributions of the UPS towards environmental burdens GWP, POCP and AP decrease significantly (10% for GWP, 13% for POCP and 64% for AP) as compared to the fuel oil steam turbine case (Fig. 5).

This gate-to-gate framework was then extended to a cradle-to-gate approach by implementing an LCA on the two solutions that were identified as potential candidates. As it is shown in Table 12, from a LCA point of view, the results of the midpoint category impacts show that solution TF4 turns out to be more environmental-friendly than solution TF2, even if it is more capital intensive.

9. Conclusion and perspectives

In this paper, multi-objective optimization based on a genetic algorithm, coupled with Multiple Choice Decision Making procedures was implemented to study the classical benchmark HDA process, where the utility production system was modelled either by use of a fuel oil steam turbine or a natural gas one. A key point is to capture in the modelling phase both process and utility production system, since the environmental impact of a chemical process is not only embedded in the products involved in the process, but

Table 12
Impacts for solutions TF2 and TF4.

	Unit		HDA	Steam	Toluene	Heat boiler	Heat furnace	Hydrogen	Fuel	Total
Carcinogens	Daily	TF2			0.7		0.1			0.8
		TF4			0.5		0.2			0.7
Respiratory inorganics	Daily	TF2	4.8	0.1	1.5	0.4	2.9	0.7	0.4	10.3
		TF4	1.6		1.5		0.5	0.7		4.8
GWP	kg eq. CO_2 in air	TF2	10.2	0.4	3.8	1.3	6.2	0.6		22.1
		TF4	8.5		3.7	1.5	3.5	0.7		18.3
Nonrenewable energy	MJ	TF2		0.5	12.1	1.4	6.7	0.6		20.8
		TF4			12	1.3	4.4	0.8		19

is also related among others to the energy consumption, the effect of flow recycle and conversion rate.

For a fixed benzene production, the multi-objective optimizations were carried out by considering six objectives, the total annual cost of the process and five environmental burdens. In each case, some good solutions were identified, and the two best ones corresponding, on the one hand, to a fuel oil steam turbine and, on the other hand, to a natural gas turbine were compared according to several items: economic, environmental burdens and Life Cycle Assessment. Even if the methodology is supported by the HDA process that was intensively studied, mainly from an academic viewpoint, it can be extended to industrial case studies. The genericity of the approach and the development of an integrated framework combining the simulation of the process and energy production unit coupled with Life Cycle Assessment, multi-objective optimization and multiple criteria decision making tools are currently under investigation in our research group.

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