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Multiobjective waste management optimization strategy coupling life cycle assessment and genetic algorithms: Application to PET bottles

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ABSTRACT

A mathematical model based on life-cycle assessment (LCA) results is developed to assess the environmental efficiency of the end-of-life management of polyethylene terephthalate (PET) bottles. For this purpose, multiobjective optimization and decision support tools are used to define optimal targets for efficient waste management. The global environmental impacts associated with the treatment of PET bottles from their cradle to their ultimate graves (incineration, landfill, recycling by mechanical, chemical or thermal processes) are computed in function of the flow of bottles in the different valorization paths. They are based on the calculation of the impacts involved in each elementary process with a LCA software tool, using the CML impact assessment method. The model takes into account the fraction λ of PET regenerated into bottles that can be further recycled, the global impacts being the cumulative impacts corresponding to each "end-of-life". A nonlinear model for the bottle waste collection stage is considered, reflecting that the more diffuse the flow of bottles is, the more difficult it is to collect and consequently, the more environmentally impacting. The resulting multiobjective problem is to find the allocation of bottles between valorization paths that minimizes the environmental impacts of bottle end-of-lives. It is solved using a genetic algorithm, and the trade-off between environmental impacts is illustrated through Pareto curves. A decision support tool then determines the best compromise among the set of solutions. The model is applied to the case of France in 2010. The variables that minimize simultaneously abiotic depletion, acidification and global warming potential are determined, in particular the number of recycling loops. The approach can be easily adapted to any specific product like bio-based plastics or organic wastes to find the optimal allocation between valorization paths.

1. Introduction

In a context of petrochemicals resources progressive scarcity, the optimization of fossil materials consumption is a key issue to ensure the sustainable supply of raw materials of future generations. Meanwhile, more than 1.8 billion tons of waste are generated per year in Europe, representing an equivalent pool of potential raw materials. Among those wastes, plastics, with a European demand of 46.4 Mt in 2010 (PlasticsEurope, 2011), are a topical issue. Since they are non-biodegradable, and can be recovered as an advantageous feedstock (Al-Salem et al., 2009), their discarding in landfill is both environmentally and economically detrimental. The European directive 2008/98/EC thus enforces an ambitious re-use or recycling rate of 30% for plastics solid wastes in 2020.

This study focuses on the case of polyethylene terephthalate (PET) bottles. Since 1970 and the development of the blow moulding technique, the volume of PET bottles produced worldwide experienced a rapid growth (Shen et al., 2010). The PET bottles total demand in Europe reached 2.784 Mt in 2010 (PlasticsEurope, 2011). Because of the short life span of bottles, a regular important waste stream is generated. Petcore (PET COntainers Recycling Europe) and EuPR (European Plastics Recyclers) associations published post-sorting European PET collection rate, amounting to 48.3% in 2010, which corresponds to an increase in 6.5% in 2009 (Petcore, 2010). Current applications of resulting r-PET are mostly divided between 39% fibres, 25% bottles, around 20% sheet and 7% strapping. Due to its maturity, the PET recycling industry is a very interesting case study for further understanding the key issues of waste management optimization. The aim of this study is to assess the environmental impacts of the PET waste management system, and to propose appropriate tools to determine how its environmental footprint could be minimized.

Many studies reported on the environmental impacts generated by PET bottles end-of-life by adopting an LCA approach, comparing three or more alternatives for PET waste management, with various objectives: (i) assessing the efficiency of recycling

Nomenclature

BHET	bis-hydroethyl-terephthalate
b-PET	bottle grade PET
B-to-B	bottle-to-bottle
CML	Centrum voor Milieuwetenschappen Leiden (Institute of Environmental Sciences), Leiden University, the Netherlands
CV	coefficient of variation
d_k	$k \in \mathbb{N}$ decision variables
DMT	dimethyl terephthalate
EG	ethylene glycol
GA	genetic algorithm
I_l	total impact l
$I_{l,n}$	impact l, recycling trip n
$I_{l,n,i}$	impact l, recycling trip n, process i
IV	intrinsic viscosity
LCA	life cycle assessment
LCI	life cycle inventory
M	initial amount of PET from fossil material, functional unit of the model
$\bar{m}$	vector of the m_i
m_i	amount treated by the process i expressed in kg
MRF	material recycling facility
NSGA	non-dominated sorting genetic algorithm
PET	polyethylene terephthalate
PREDMA	Plan Régional d'Elimination des Déchets Ménagers et Assimilés
SSP	solid state polycondensation
q	number of recycling trips taken into account in the impact calculation
r-PET	recycled PET
TOPSIS	Technique for order preference by similarity to ideal solution
TPA	terephthalic acid
UP	unsaturated polyester
λ	fraction of initial M kg of PET bottles reprocessed into a bottle

compared to landfill and incineration (Chilton et al., 2010; Arena et al., 2003; Tokai and Furuichi, 2000; Song and Hyun, 1999), (ii) comparing the benefits of thermal recycling versus mechanical recycling (Perugini et al., 2005; Song and Hyun, 1999), (iii) confronting different paths to produce PET fibre from r-PET (Shen et al., 2010), or three chemical recycling routes to recover PET from waste bottles (Sugiyama et al., 2006). The literature indicates that recycling versus either incineration or landfill is generally the best option to reduce environmental impacts (Chilton et al., 2010). However, among recycling routes, closed and open-loop recycling must be differentiated, and end-products characterized, since consequent environmental impacts can be very different. Impacts of mechanical recycling to fibre versus mechanical recycling to bottle are thus compared in Shen et al. (2011). It would be interesting to extend this approach to other products, like PET sheets, or chemicals from recycled PET.

Moreover, recent improvements in recycling technologies now allow bottle-to-bottle (B-to-B) by mechanical recycling (Welle, 2011), and PET initially produced from fossil petrochemicals can be regenerated into a bottle several times (Rieckmann et al., 2011). The implied modifications on PET environmental footprint have yet rarely been considered, apart in Shen et al. (2011), but only (bottle-to-fibre) B-to-F and B-to-B by mechanical recycling are included in the system described.

Finally, if recycling seems like the most desirable solution, a detailed review of the literature shows that conflicting issues arise: (i) above a certain limit, the increasing difficulty to collect bottles aggravates the consequent environmental impacts (Song and Hyun, 1999) and (ii) if incineration is more interesting than landfill in terms of energy use and atmospheric emissions, landfill generates less CO_2 (Chilton et al., 2010). These two points highlight that in such a complex system, there is no ideal solution overcoming all others, but that a right balance has to be found between conflicting objectives. To handle this complexity, single objective optimization tools have already been used (Song and Hyun, 2001, 1999; Tokai and Furuichi, 2000). They assign weighting factors to environmental burdens, which may introduce a bias.

The aim of this work is thus to design a more complete model of the PET waste management system, including both open and closed-loop strategies, and the possibility of multiple recycling trips. The environmental impacts generated by this system will be evaluated as a function of the allocation of bottles between the different valorization and recycling paths. The situation of France in 2010 will serve as an illustration. The allocation of bottles will then be optimized using a multiobjective genetic algorithm in order to minimize simultaneously different environmental impacts.

2. PET waste management system

2.1. Virgin production

Virgin polymer synthesis: Polyethylene terephthalate (PET) is obtained by a reaction between a diacid and a dialcohol, followed by several stages of polycondensation. The bis-hydroxyethyl-terephthalate (BHET) monomer can be produced either by trans-esterification of dimethyl terephthalate (DMT) and ethylene glycol (EG), or by direct esterification of terephthalic acid (TPA) and EG (Karayannidis and Achilias, 2007). Since 1970, the path by direct esterification of TPA and EG has progressively been adopted (Quentin, 2004). Only this process will be further considered in the study.

Injection and blow moulding: Bottle grade PET (b-PET) with a high intrinsic viscosity (IV), around 72–85 mL g^{-1} (Quentin, 2004), is injected into a cold mould to produce an amorphous preform. This preform is then transferred to an air-blowing unit to be blown into a bottle mould (Awaja and Pavel, 2005). The bottle can then be filled, caped and labelled.

2.2. Description of the valorization alternatives

Fig. 1 represents a generic diagram of a waste management system that will be adapted to the case of PET bottles.

2.2.1. Landfill
PET bottle wastes are collected with the usual flow of household waste and disposed of in municipal landfills. Even if it is the worst alternative in terms of resource conservation, more than 21% of PET bottle were still landfilled in France in 2009 (RDC-Environnement, 2007).

2.2.2. Valorization and recycling
Secondary recycling or mechanical recycling: Mechanical recycling includes all processes consisting in physical reprocessing, such as washing, grinding, melting and reforming. PET bottle wastes are selectively collected and transferred to a Material Recovery Facility (MRF). After proper sorting, balled PET bottles are sent to the reprocessing plant according to three main processes:

Conventional mechanical recycling: r-PET flakes production
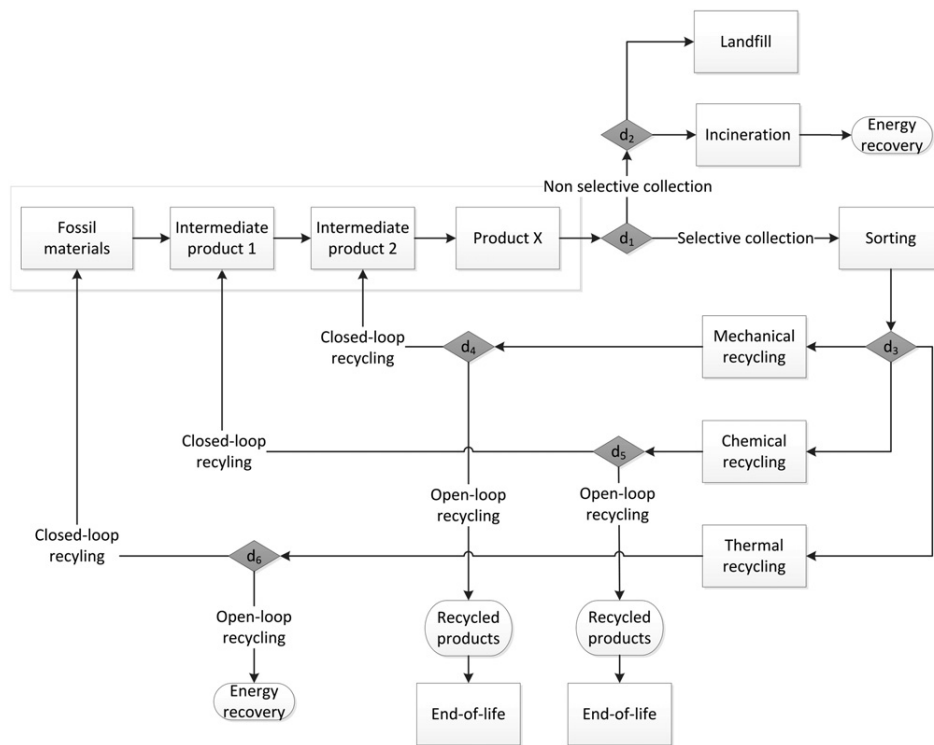Bales of PET bottles are opened to be washed, and then grinded into

Fig. 1. Generic diagram for a waste recycling system, with d_k, decision variables.

flakes. The PET is separated from scraps, caps and labels by flotation and the flakes are then dried. The r–PET flakes thus produced can be used for non-food applications, mostly staple fibres and sheets (Nicklaus, 2011).

r-PET pellets production The r–PET flakes are re-extruded to pellets. The resulting r–PET pellets can be used for food applications (food tray, etc.) but also strapping and non-food PET containers (Nicklaus, 2011).

Super-clean recycling technologies They are new sophisticated decontamination processes that are able to decontaminate PET waste to concentration levels of virgin PET materials (Welle, 2011).

The first stages are common to all processes. After flakes production and extrusion, r–PET pellets are further processed by SSP (solid state polycondensation), to obtain a recycled resin with an IV almost equivalent to that of virgin PET pellets (Nicklaus, 2011). The obtained pellets can be used for the production of new bottles. However, the amount of super-clean recycled PET incorporated in PET bottles is still limited to 35%. Above this limit, there is a risk of colouration, which is unacceptable for commercial use (Shen et al., 2011).

In this work, only the three main end-markets for r–PET in France are considered, namely fibres (52%), films (20%) and bottles (25%) (Valorplast, 2010).

Tertiary or chemical recycling: They lead to either partial depolymerization of PET to oligomers or BHET, or total depolymerization to monomers (TPA, EG and DMT) (Karayannidis and Achilias, 2007). The main methods for chemical recycling of PET are glycolysis, methanolysis, hydrolysis and aminolysis (Awaja and Pavel, 2005). Today, only glycolysis and methanolysis are industrially developed, and in 2006, only 4% PET was chemically recycled worldwide (Karayannidis and Achilias, 2007), mostly in Asia.

Methanolysis involves the degradation of PET chains by methanol, the main products being the monomers DMT and EG.

Besides its high cost, the main disadvantage of the methanolysis comes from the fact that nearly all PET producers are now using the direct esterification path (Karayannidis and Achilias, 2007).

Glycolysis is the reaction of a glycol (ethylene glycol, diethylene glycol or propylene glycol) with PET chains. BHET or oligomers are obtained and can be reused to synthesized new PET or other polymers (Karayannidis and Achilias, 2007). There has been a renewal of interest for PET glycolysis for a few years, as its output can be used as a substrate for the synthesis of high value materials: production of polyurethanes, of acrylate/methacrylate-terminated oligoesters, or production of unsaturated polyesters (UP) (Jankauskaitė et al., 2008).

Given the relative development of each process and the available information in open–literature, only chemical recycling by glycolysis is considered in the model; two alternatives are described, glycolysis and repolymerization into PET or glycolysis followed by synthesis of UP resin.

Quaternary recycling or energy recovery:

Incineration PET bottles are collected with the usual flow of household waste and disposed of in municipal incinerators. Energy in the form of heat or electricity is recovered with varying efficiencies.

Feedstock/thermal recycling This category[1] includes the advanced processes leading to the recovery of primary energy sources like gas or fuel. The main advantage of those processes is the possibility of treating heterogeneous and contaminated polymers. The three main technologies are pyrolysis, hydrogenation and gasification (Al–Salem et al., 2009). Given the availability of data in open–literature, the processes taken into account in this work are pyrolysis and hydrogenation (also called hydrocracking).

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[1] Thermal recycling is not always classified as quaternary recycling but is sometimes assimilated to tertiary or chemical recycling (Al–Salem et al., 2009).

Table 1
The decision variables d_k of the model, $c, i, r, p, m, e, b, f \in [0, 1]$.

d_k	Description
c	Selective collection rate
i	Rate of discarded bottles incinerated (versus landfilled)
r	Rate of collected and then recycled bottles (versus feedstock recycling)
p	Rate of collected and non recycled bottles that are pyrolysed (versus hydrocracking)
m	Rate of recycled bottles mechanically processed (versus chemical recycling)
e	Rate of r-PET flakes that are extruded (versus fiber spinning)
b	Rate of r-PET pellets used for making bottles (versus PET film)
f	Rate of recycled bottles glycolysed and used to synthesize new PET (versus unsaturated polyesters)

Table 2
Yields of the different processes.

Yields	Process	Value	Source
η_S	Sorting	0.84	TERRA (2010)
η_{MR}	Mechanical recycling	0.8	NREL (2008)
η_F	Fiber spinning	0.99	Shen et al. (2010)
η_{GLY}	Glycolysis	0.8	Shen et al. (2010)
η_{UP}	UP synthesis	2.95	Barboza et al. (2009)
η_{RP}	Repolymerization	0.98	Shen et al. (2010)

Pyrolysis is the thermal cracking of polymers in inert atmosphere. The process can treat many different solid hydrocarbon based wastes to produce a clean, high calorific value gas. Hydrogenation or hydrocracking means the addition of hydrogen by chemical reaction through unit operation (Smith et al., 2007). The main outputs are fuel or synthetic oil and gas.

2.3. System diagram and material balance determination

The resulting waste management system diagram of PET bottles is presented in Fig. 2. The decision variables d_k are represented by diamonds and defined in Table 1. The yields of the different processes (see Table 2) are necessary to establish the material flow balance. When no yield is specified, it is considered equal to 1; m_i is introduced as the amount treated by the process i expressed in kg, a function of the decision variables and of the yields of the studied processes. $\vec{m}$ is the resulting vector. The yields are considered as constant values. For example, if M is the initial amount of bottles in kg, the amount of PET bottles incinerated is equal to $m_{inc} = M \times (1 - c) \times i$ kg.

3. Environmental impact assessment

The objective of this section is to develop a model that evaluates the total environmental impact I_l as a function of PET bottle allocation between the different valorization and recycling paths. The LCA software tool Simapro (Pré-Consultants, 2011) used in this study does not give the flexibility to design a module taking into account allocation paths as variables. The calculations are therefore based upon elementary environmental impacts computed with Simapro, and the mass balance $\vec{m}$ between the different recycling pathways. It must be highlighted that a conventional LCA was not implemented *stricto sensu*, but is embedded in a simulation model to perform a analysis based on the influence of valorization paths.

3.1. Goal, functional unit and system boundary

The goal of this LCA is to model the environmental impacts generated by PET bottles from their production to their final disposal in function of the quantity of PET sent to each recycling alternative. The functional unit is defined as *M kg of PET bottles produced from virgin PET*.

An extension of the cradle-to-grave approach is adopted to define the system boundaries. The cradle is considered as the extraction of fossil raw materials. In other words, the first life of the PET bottles is included in the model, which differs from the conventional cut-off approach where waste does not bear any environmental burden from its first life (Shen et al., 2010). The end-of-lives of the bottles according to the quantities sent to each disposal/recycling solution are considered until the final graves, defined as the ultimate end-of-live(s) of the regenerated products,
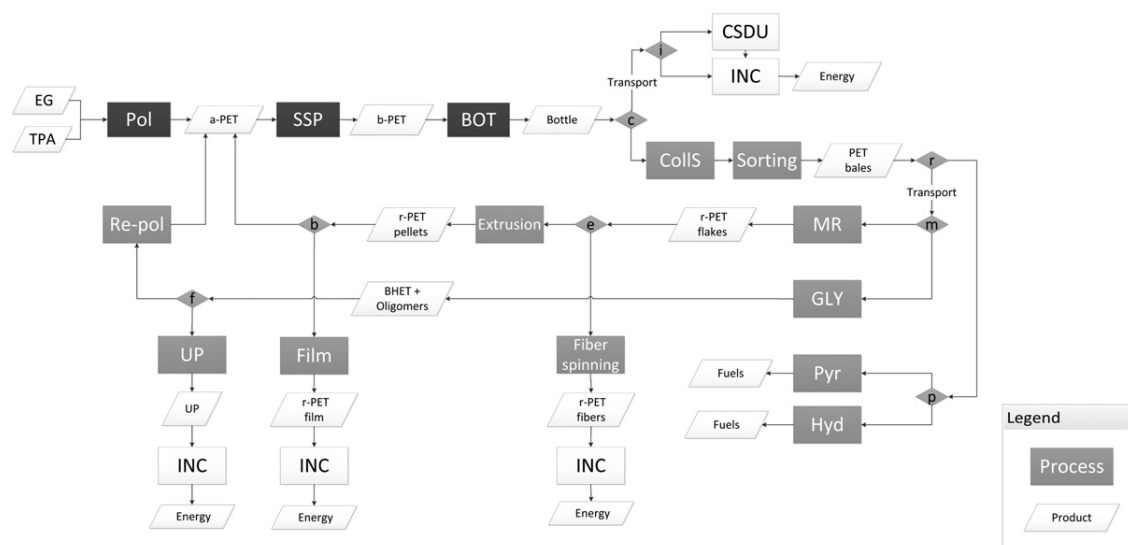
Fig. 2. The PET waste management system. EG: ethylene glycol, TPA: terephthalic acid, Pol: polymerization, a-PET: amorphous PET, SSP: solid state polycondensation, b-PET: bottle grade PET, BOT: bottle production, INC: incineration, CSDU: landfill, CollS: selective collection, Pyr: pyrolysis, Hyd: hydrocracking, MR: mechanical recycling, GLY: glycolysis, UP: unsaturated polyester synthesis, Re-pol: re-polymerization.
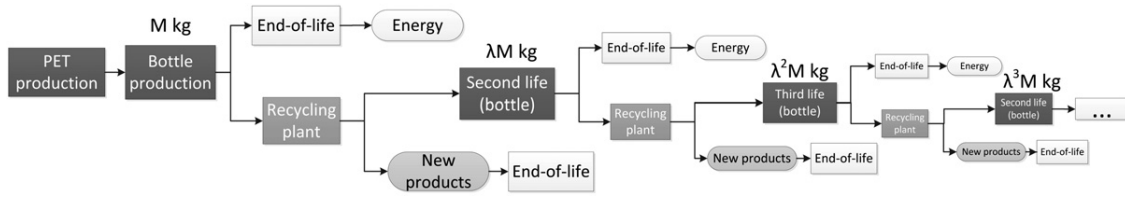
Fig. 3. An exhaustive LCA: the cradle-to-graves approach.

i.e. incineration or landfill. The manufacturing of end products, distribution and use phases are excluded (see Section 3.2).

The end-of-lives of the regenerated products are also part and parcel of modelling. Indeed, according to the quality/ type of product, it can be landfilled, converted thermically or once more recycled, as shown in Fig. 3. This will generally be the case for products following the closed-loop recycling path and regenerated into the original product. According to the type of packaging, there can be an infinity of recycling loops (aluminium cans, glass bottles) or only a few (paper and cardboard). In this study, r-PET regenerated into a bottle can be further recycled, while the other products (fiber, film, unsaturated polyester) can only be incinerated or landfilled. The fraction of the initial M kg of bottle that is recycled into bottle grade PET is called λ. It is then integrated into a flow of virgin PET and reprocessed into λM kg of bottle, which is submitted to the same recycling alternatives as in its first life, since it is not distinguishable from the virgin flow of bottle. The environmental impacts consequent to these successive end-of-lives are taken into account, until the ultimate graves. The life cycle of packaging waste is considered to be short enough to ignore any temporal evolution of the processes and parameters. The allocation of the flow of bottles is consequently the same for each end-of-life cycle. λ is therefore constant in time and given by the material balance of the system, $\lambda = [b \times e\eta_{MR}m + \eta_{RP}f\eta_{GLY}(1-m)]r\eta_T c$. After n recycling trips, the amount of PET recycled to bottles is given by $\lambda^n M$.

3.2. Computation of avoided impacts

The creation of x kg of a new product (fibre, bottle, film, unsaturated polyester) leads to an avoided burden equal to the impact generated by the same amount of the product from virgin raw materials (Nicklaus, 2011). It is therefore considered that 1 kg of PET recycled in x is equivalent to 1 kg of x from virgin production. This justifies not to take into account the distribution and use phases, as they are common to virgin and recycled products. This hypothesis has to be discussed for each case, and a weighting factor could eventually be assigned when the recycled product does not reach the necessary specifications:

- Fibre: as noted in Shen et al. (2011), recycled PET fibers have a narrower application spectrum than virgin fibers and are mostly used as staple fiber. This must be kept in mind when interpreting the results.
- Film: the r-PET film is used for thermoforming like eggs box or food containers, incorporated up to 100% (Nicklaus, 2011), it is thus considered as equivalent to virgin PET film.
- Bottle: bottle grade r-PET is supposed to have the same quality as virgin PET (Rieckmann et al., 2011; Nicklaus, 2011). However, the incorporation of r-PET in bottles is still limited, between 25 and 35% depending on the sources (Nicklaus, 2011) (Shen et al., 2011). This does not modify the proposed model, as we consider that the fraction λ is mixed with a virgin flow of b-PET at each successive "end-of-life". The impact of this virgin flow of bottles is not taken into account.

- Unsaturated polyester: the proprieties of UP produced from r-PET are compared with those of virgin UP and judged as equivalent (Barboza et al., 2009).

3.3. Impact calculation

3.3.1. Elementary impacts

The mid-point impact evaluation method CML 2 baseline 2001 V2.04 is used. The included impacts are Abiotic Depletion (AD), Acidification (Ac), Eutrophication (Eut), Global Warming potential (GWP), Human Toxicity (H.Tox), Fresh water aquatic ecotoxicity (FW.Tox), Terrestrial ecotoxicity (T.Tox) and Photochemical oxidation (PCO). The limited information concerning PET glycolysis (see Section 3.4.1) does not allow the computation of ozone layer depletion and marine aquatic ecotoxicity for this module and therefore for the whole system, they are not further considered in this study. Concerning marine ecotoxicity, the only information reported in the literature gives quite scattered orders of magnitude, suggesting that the indicator is not so reliable. This poor reliability of marine ecotoxicity and the lack of data concerning ozone layer depletion explain why these two indicators were discarded in this analysis. For each process i, an inventory of the elementary streams is carried out, namely reactants, utilities, air, water and waste emissions. The data used come from open literature and scientific publications (see Table 3). The output of the process $i-1$ of the inventory is excluded so that the environmental impacts associated with its production are not counted several times. The impacts $I_{l,n,i}$ associated with the process i per treated kg are then calculated using the software Simapro 7.1. The avoided impacts are computed as negative impacts from virtual processes. Classically from an LCA viewpoint, the $I_{l,n,i}$ are considered constant except from the particular case of selective collection for which a nonlinear model is introduced (see paragraph 3.3.2).

3.3.2. Selective bottle waste collection

The efficiency of collection will rely on the propensity of the inhabitants to sort their garbage, on their location when they discard their bottles, and on the collection system implemented by their municipality. The model must reflect that it would be almost impossible to collect 100% of PET bottles, as the more important the collection rate c, the more diffuse the flow of bottles is. A parallel can be here established with the mining industry, where it is widely demonstrated that as ore grades decline, energy and greenhouse gases costs increase, generally exponentially (Mudd and Ward, 2008). A nonlinear model for the impacts due to the collection process of PET bottles is thus assumed, following (Song and Hyun, 1999):

$$I_{l,n,coll}(c) = P_1(l) + P_2(l) \times \frac{c^{r(l)}}{1-c} \qquad (1)$$

with $P_1(l), P_2(l) \in \mathbb{R}$ and $r(l) \in \mathbb{N}$ parameters to be defined for each impact l.

It is very difficult to find enough empirical data to adjust the parameters, since this requires the values of each impact for a range of c between 0 and 1. The value of c beyond which the

Table 3
Life cycle inventories data sources (EI V2 means EcoInvent V2).

Stage	Sources	Value, commentary	Uncertainty
Ethylene Glycol	EI V2, Frischknecht et al. (2007)	Ethylene glycol, at plant, RER U	Included
Purified terephthalic acid	EI V2, Frischknecht et al. (2007)	Purified terephthalic acid, at plant, RER U	Included
Polymerization a-PET	PlasticsEurope (2010)	Mass balance in kg, $0.333 \times EG + 0.877 \times TPA \rightarrow 1.00 \times PET$	Included
Solid state polycondensation	PlasticsEurope (2010)		Included
Bottle production	PlasticsEurope (2010), RDC-Environnement (2007)	Material balance valid for 1.5 L bottles, 0.908 kg b-PET for 1 kg of PET bottle	From Nicklaus (2011)
Non selective collection	EI V2, Frischknecht et al. (2007) and SINOE (2007)	Transport, municipal waste collection, lorry 21 t/CH U, average distance 12.2 km	Included, uniform ±10%
PET incineration	EI V2, Frischknecht et al. (2007) and Nicklaus (2011)	Disposal, polyethylene terephthalate, 0.2% water, to municipal incineration/CH U	Included, uniform ±10% for recovered energy
PET landfilling	EI V2, Frischknecht et al. (2007)	Disposal, municipal solid waste, 22.9% water, to municipal incineration/CH U	Included
Selective collection	EIV2, Frischknecht et al. (2007)and SINOE (2007)	Transport, municipal waste collection, lorry 21 t/CH U, average distance 86.3 km	Included, uniform ±10%
Sorting at MRF	NREL (2008), Arena et al. (2003)	Mixed recyclables, sorted at MRF	Uniform ±10%
Pyrolysis	Perugini et al. (2005)	Low temperature pyrolysis	Uniform ±10%
Hydrocracking	Perugini et al. (2005)	Veba Combi-cracking process	±10%
Transport to recycling plants	EI V2, Frischknecht et al. (2007) and Nicklaus (2011)	Transport, lorry 3.5–16 t, fleet average/RER U, average distance 400 km	Included, from Nicklaus (2011) for the distance
Mechanical recycling to flakes	NREL (2008)	Recycled postconsumer PET flake	Uniform ±10%
Fiber spinning	Shen et al. (2010)		Uniform ±10%
Extrusion to pellets	NREL (2008)	Recycled postconsumer PET pellet	Uniform ±10%
Film production	EI V2, Frischknecht et al. (2007)	Extrusion, plastic film, RER U	Included
Glycolysis + repolymerization	Shen et al. (2011)	Impacts already calculated, site specific	–
Glycolysis + UP synthesis	Barboza et al. (2009)	Pilot plant	uniform ±10%
UP incineration	EI V2 Frischknecht et al. (2007)	Pilot plant	included
Electricity production	EI V2, Frischknecht et al. (2007)	Electricity, medium voltage FR	Included
Heat production	EI V2, Frischknecht et al. (2007) and Nicklaus (2011)	French heat production 70% natural gas, 20% fuel oil, 10% coal	Included

function $I_{l,n,coll}(c)$ will not be linear is therefore estimated. According to ADEME (2009), 99% of the French population can dispose of their recyclable waste separately, either by kerbside collection or central collection sites. The same source states that an average of 80% (p_1) of people sort their waste when served by kerbside collection, whereas only 67.5% (p_2) does it when they have to take it to central collection sites. According to EcoEmballages (EcoEmballages, 2011), in 2009, 72% (43.3 M) of French people (pop_1) were served by kerbside collection, which means that for 99–72 = 27% of the population (pop_2), the only possibility was to bring their sorted wastes to central collection sites. It is therefore likely that: $c \leq p_1 \times pop_1 + p_2 \times pop_2 = 0.76$. We are of course aware that a lot of PET bottles are used outside the home so a more thorough analysis would be to include in collection efficiency calculations collection facilities and their use at sporting/leisure venues, shopping centres, tourist attractions etc.

In this investigation, a percentage of 76% of PET bottles in domestic waste stream are likely to be sorted. If we consider moreover that no more than 80% of PET bottles enter the domestic waste stream (Wilson, 2002), 60% seems a reasonable value for the limit beyond which the impacts $I_{l,n,coll}(c)$ become nonlinear.

$P_1(l)$ is different for each impact and is calculated by Simapro as the impact resulting from a collection circuit of 86.3 km (the average French value according to SINOE database (SINOE, 2007)). To calculate $P_2(l)$ and $r(l)$, arbitrary assumptions are made:

$$I_{l,n,coll}(c) = P_1(l) + \varepsilon(c, l) \quad \text{with } \varepsilon(c, l) = P_2(l) \times \frac{c^{r(l)}}{1 - c}$$

It is then adopted that $\varepsilon(c, l) \leq 0.05 \times I_{l,n,coll}(c)$ for $c \leq 0.6$, so that $0.6^{r(l)} \times P_{2,l} \leq 0.021 \times P_1(l)$. The growth rate $r(l)$ is taken equal to 7 so that the impacts resulting from the collection double when c is around 0.8.

It can be deduced that $P_2(l) = (0.021 \times P_1(l))/0.6^7 = 0.75 \times P_1(l)$ and $I_{l,n,coll}(c) = P_1(l)(1 + 0.75 \times c^7/(1 - c))$ (see Fig. 4). With this model, impacts become prohibitive when c is superior to 90%.

3.3.3. Assessment of the global impacts

For each impact l, $l \in \{1, \ldots, 8\}$ the total impact I_l is expressed as the sum of impacts caused by each end-of-life, $I_l = \sum_n I_{l,n}$, n being the life cycle number. The $I_{l,0}$ are the impacts resulting from the virgin life of the product, from the extraction of the raw materials to its discarding. $I_{l,1}$ are the impacts resulting from the first life of the used product, from its cradle (bottle waste collection) to its different graves. It is a function of the flow of bottle in each path given by
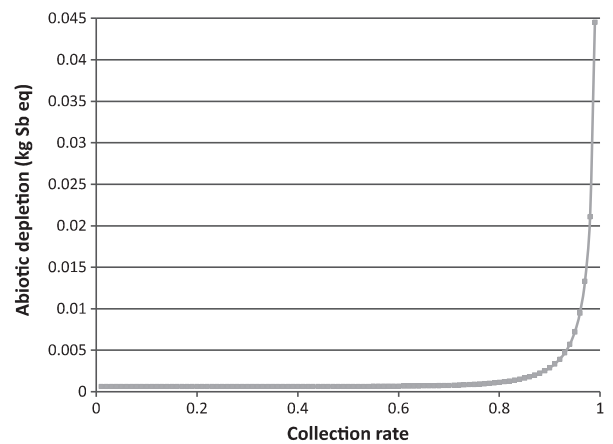

Fig. 4. Impact model for selective collection, the example of abiotic depletion.
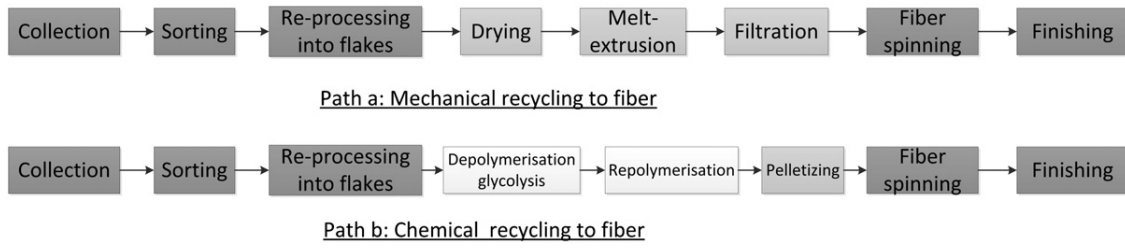
Fig. 5. Chemical recycling and repolymerization path, from Shen et al. (2010) (common steps in dark grey).

m_i and of the elementary impacts $I_{l,1,i}$ per process i. Considering λ as constant in time (see Section 3.1), for each impact l, $I_{l,n}$ is a geometric sequence of initial value $I_{1,n}$ and common ratio λ, $I_{l,n} = I_{l,1} * \lambda^{n-1}$. The $I_{l,n}$ are then given by:

$$
I_{l,n} = \begin{cases} I_{l,0} & \text{for } n = 0 \\ I_{l,1} = \sum_i I_{l,1,i} \times m_i(d_{k,1 \leq k \leq 8}) & \text{for } n = 1 \\ I_{l,n} = \lambda^{n-1} I_{l,1} & \text{for } n \geq 1 \end{cases}
$$

The impact I_l is then easily established, with the number of recycling trips q taken into account:

$$
I_l = \sum_{n=0}^{q} I_{l,n} = I_{l,0} + I_{l,1} \times \sum_{n=1}^{q} \lambda^{n-1} = I_{l,0} + I_{l,1} \times \frac{1 - \lambda^q}{1 - \lambda} \quad (2)
$$

3.4. Specific parameters

3.4.1. Chemical recycling

Chemical recycling of PET bottles is carried out by only a few companies (Karayannidis and Achilias, 2007; Welle, 2011; ADEME, 2002). Two processes are considered in what follows. In both cases, waste bottles are first processed into flakes (Shen et al., 2010), consequent impacts are included in the calculation.

Glycolysis and repolymerization to pellets: The values of the impacts calculated in Shen et al. (2010) obtained from the CML 2 baseline 2001 method are used. The study compares several ways of recycling PET bottles to fibres (Fig. 5). An aggregation was performed for drying, melt-extrusion, filtration and pelletizing, as they both result in PET pellets. If I_a and I_b are respectively the impacts from the first and second path, the impacts for glycolysis and repolymerization only, $I_{Gly+RP} = I_b - I_a$, are easily deduced.

Glycolysis and synthesis of unsaturated polyesters: The inventory is based on the calculations made in Barboza et al. (2009) for mass balance and energy consumption (Fig. 6). It is important to highlight that, unlike other studied processes, those data stem from experimentations in a pilot plant and may not be representative of an industrial practice.

3.4.2. Energy recovery by incineration

Each waste incineration plant has its own efficiency, depending mainly on the kind of energy recovered: electricity, co-generation of electricity and heat, or only heat. It is reported in ADEME (2009) that a total of 3206 GWh of electricity and 6700 GWh of heat were recovered by incinerating 11.04 Mt of waste in 2006. It is deduced that in average, burning 1 kg of waste in France allows to recover 0.29 kWh of electricity and 0.607 kWh (2.18 MJ) of heat. This value will be used in the case of unsaturated polyesters, as no more specific information is available.

To calculate the average energy recovered by PET incineration, a heating value for PET of $HV = 22.95$ MJ/kg (Eco Invent V2 (Frischknecht et al., 2007)), and an efficiency of $\rho_e = 8\%$ for produced electricity and $\rho_h = 19.7\%$ for produced heat (Nicklaus, 2011) are considered. With the amount of energy produced $E_i = HV \times \rho_i$, burning 1 kg of PET therefore produces in average 0.51 kWh of electricity and 4.25 MJ of heat.

The effect on the influence of these values will be performed in Section 5.2.

To assess the impacts induced by the production of 1 kWh of heat, the average mix for heat production in France is taken into account: 70% natural gas, 20% fuel oil and 10% coal (Nicklaus, 2011). Given that France produces a high proportion of its power from nuclear sources, energy recovery from PET in France will have a different CO_2 balance from in countries which rely on fossil fuels for power (particularly those with high coal consumption).

3.5. Uncertainty analysis

The aim of analysis is to evaluate how uncertainty of the input parameters of an LCA affect the final results of impacts (Heijungs and Huijbregts, 2004; Lloyd and Ries, 2007).

Uncertainty evaluation by Monte Carlo analysis is performed for eight valorization routes, as described in Table 4. For example, in case 1, uncertainty of the resulting impacts is calculated when 1 kg of PET bottles is produced, collected and sent to landfill. The nonlinear model for collection stage and avoided impacts are not included here. Uncertainty analysis for B-to-B by chemical recycling is not computed as already calculated impact values are used for glycolysis followed by repolymerization (see Section 3.4.1). The input probability distributions of each parameter (see Table 3)
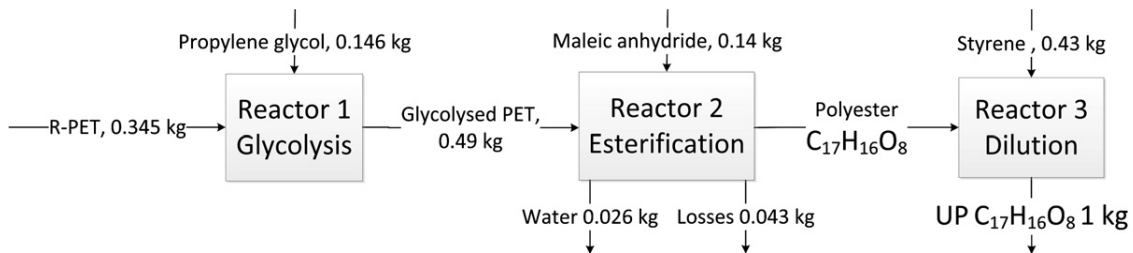

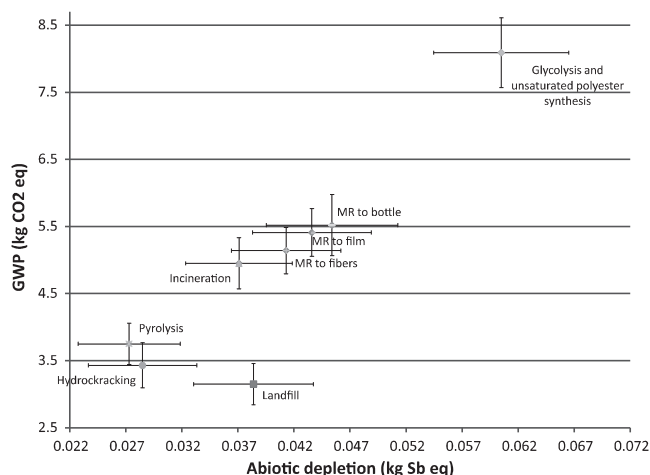**Fig. 6.** Unsaturated polyester synthesis from recycled PET.

Fig. 7. Results from the Monte Carlo analysis.

are either directly specified in EcoInvent modules, or estimated from literature. When information sources are lacking, a restrictive hypothesis of uniform distribution laws of ±10% for all parameters was assumed. The output of this calculation consists in a probability distribution for each impact. They are represented in Fig. 7 for abiotic depletion and GWP, the error bars being equal to the standard deviation. It can be observed that pyrolysis has a higher GWP impact than hydrocracking which can be attributed to the assumptions carried out in EcoInvent for this process. Indeed, hydrocracking is generally based on steam reforming which is identified as a high CO_2 emission process. The hydrogen production module in EcoInvent is based on a mixed process involving water electrolysis which may be surprising at this level.

The coefficient of variation (CV) of each impact is also obtained, it is a normalized measurement of the relative dispersion of the results, defined as the ratio of the standard deviation σ to the mean μ: $c_v = \sigma/\mu$. In average, CV is superior to 80% for fresh water and marine ecotoxicity, and respectively around 52, 37 and 32% for ODP,

human toxicity and terrestrial ecotoxicity. Best results are obtained for GWP, CV being inferior to 10% in all cases and for abiotic depletion (around 13%), acidification, eutrophication and photochemical oxidation (all around 14%).

At this stage, the results of the Monte Carlo analysis show that the level of uncertainty is equivalent for all considered processes for abiotic depletion, acidification, eutrophication, photochemical oxidation and GWP. It also highlights the fact that some impacts are very approximately evaluated, i.e., toxicity and ecotoxicity impacts, and ODP. This will influence the choice of evaluation criteria for the optimization of the system in Section 4.2.

4. Multiobjective optimization and decision support

4.1. Selection of a genetic algorithm

A tool for a parametrizable assessment of the environmental impacts of PET bottles has been proposed. By modifying the decision variables (Table 1) of the model, any recycle alternatives or any combinations can be represented and compared. The aim of this section is to introduce an appropriate tool to find the optimal decision variables in order to minimize the environmental impacts.

The general formulation of a multiobjective optimization problem, according to Fonseca and Fleming (1993), is constituted by a set of n criteria c_k, with $k = 1, \ldots, n$, that are to be maximized or minimized. Each criterion, which can be nonlinear or discontinuous, is a function of m decision variables d in an m-dimensional space.

The first methods developed to solve multiobjective problems consisted in reducing them into a single target by aggregating the different criteria into a single one (Ouattara, 2011). A few publications in the field of waste management have applied this approach ((Song and Hyun, 1999, 2001; Tokai and Furuichi, 2000)). The authors insist on the difficulty of fixing the trade-off coefficients that are the weighting parameters. They have either to be defined by the decision maker, or to be calculated relying on methods such as normalization of the impacts, or transformation of the impacts into equivalent economic values. Both methods can be criticized as they introduce bias/more uncertainty to the results. It is therefore more interesting to resort to *a posteriori* methods leading to a set of non dominated solutions or Pareto Front, the final trade-offs being left to decision makers.

An alternative is to use genetic algorithms (GA), that are easily applicable to multiobjective problems and are therefore widely used in many areas of engineering (Gomez et al., 2010; Ouattara et al., 2012). A variant of the so-called NSGA II algorithm (Non-dominated Sorting Genetic Algorithm) is adopted here (Gomez et al., 2010).

4.2. Choice of the evaluation criteria

If a GA can theoretically be applied to eight or more criteria at a time, the solutions produced would be very difficult to interpret.

Table 4
Valorization routes studied by Monte Carlo analysis. Avoided impacts are not included.

Case	Description
1	From cradle to landfill
2	From cradle to incineration
3	From cradle to pyrolysis
4	From cradle to hydrocracking
5	From cradle to fiber
6	From cradle to film
7	From cradle to bottle, by mechanical recycling
8	From cradle to unsaturated polyester

Table 5
Inventory of the impacts assessed in the literature (y: yes; n: no).

References	AD	Ac	Eut	GWP	H.Tox	FW.Tox	T.Tox	PCO	ODP	M.Tox
Craighill and Powell (1996)	n	y	y	y	n	n	n	n	n	n
Song and Hyun (1999)	y	n	n	y	n	n	n	n	n	n
Perugini et al. (2005)	y	n	n	y	n	n	n	n	n	n
Den Boer et al. (2007)	y	y	y	y	y	n	n	y	n	n
Vellini and Savioli (2009)	y	n	n	y	n	n	n	n	n	n
Shen et al. (2010)	y	y	y	y	y	y	y	y	n	n
Chilton et al. (2010)	y	y	y	y	n	n	n	n	y	n
RDC-Environnement (2007)	y	y	y	y	n	n	n	n	n	n
Napcor (2010)	y	n	n	y	n	n	n	n	n	n
EcoEmballages (2009)	y	y	n	y	n	n	n	n	n	n
Total	9	6	5	10	2	1	1	2	1	0

Table 6
Parameters corresponding to the situation in France in 2010.

Parameters	Value	Source and comments
c	0.5	RDC-Environnement (2007)
i	0.57	RDC-Environnement (2007)
r	1	ADEME (2002)
p	–	ADEME (2002)
m	0.96	Karayannidis and Achilias (2007), worldwide value
e	0.5	Valorplast (2010)
b	0.49	Valorplast (2010)
f	0.8	Valorplast (2010)

It is therefore more interesting to decrease the number of objectives by focusing on the most significant impacts only, as regards to both the relevance of the impact in the field of plastics synthesis and recycling, and the reliability of the impact. From the dedicated literature (see Table 5), ten works were considered: four of them concerning the waste management of several materials (plastics, glass, cardboard, paper), whereas the other six are dealing exclusively with PET. It must yet be emphasized that some criteria are common denominators to all methods, for example GWP and CO_2 emissions, abiotic depletion and energy use. Indeed, abiotic depletion and fossil energy use are correlated in terms of pollutants as high as 98% with representative French values (given in Table 6).

The choice of the evaluated impacts is only justified in RDC-Environnement (2007), where ozone depletion and photochemical oxidation are excluded due to *"the low emissions associated to the processes considered"*. Toxicity and ecotoxicity impacts are also excluded from RDC-Environnement (2007). Indeed, the authors consider that there is too much uncertainty in the calculations for several reasons: the synergies between pollutants are not considered, LCI are often incomplete and uncertain, and characterization factors are lacking for many pollutants. Moreover, those impacts are very dependent of the local ecosystem, the receptors and the period of exposition. It is therefore of limited interest at a global scale as also reported in Sleeswijk et al. (2008), thus confirming the results of Section 3.5.

Based on the results of this inventory, Abiotic depletion and GWP will be included as they are both relevant and very widely used. Indeed, one of the main purpose of recycling PET is to save petrochemical resources, evaluating abiotic depletion is therefore essential to assess the environmental efficiency of the system. Moreover, the material balance and energy consumption of each step being known, it is a rather reliable estimation. As for GWP, it is an unavoidable criterion to be assessed in every environmental evaluation, as shown by the bibliography. It is also an important tool for decision making as it is more and more regulated and discussed by governments. Photochemical oxidation is excluded considering the outcome of the bibliography and the results of Section 3.5. The combination of many arguments against the toxicity factors (uncertainty (see Section 3.5), lack of data, irrelevance, etc.) leads us not to consider these criteria. As for acidification and eutrophication, acidification will be further considered as it gave slightly better results in the uncertainty study, but also because it is traditionally related to industrial emissions, whereas eutrophication is more the consequence of agricultural activities. Finally, the results show that it is not detrimental to excluding marine (not included in any study, and uncertain) and ozone depletion (only once considered, and excluded from RDC-Environnement, 2007).

4.3. Decision support tool

The multiobjective optimization phase presented in Section 4.1 leads to a set of non dominated solutions or Pareto Front. The next step consists in finding tools to guide the decision maker in his final task: finding best compromises within a set of non-dominated solutions, considering the context (regulation, ecosystem, etc.). This is carried out using a modified version of the TOPSIS (technique for order preference by similarity to ideal solution) method, due to the simplicity of its implementation, and the low number of parameters that are to be adjusted.

The basic principle of the method consist in choosing a solution that is simultaneously nearest the ideal solution (best for all criteria) and as far as possible from the worst solution (worst for all criteria) (Ouattara, 2011). The detailed stages of the procedures can be found in Ren et al. (2007). They consist in the construction of the efficiency matrix B, where each column represents a criterion (objective), each line an alternative set of values of the objectives (consequent to a set of decision variables). The matrix is normalized, and weights are assigned to each criteria. Euclidean distances to ideal and non-ideal solutions (respectively D_i^+ and D_i^-) are calculated, and alternative solutions are classified in increasing order according to the value of the ratio $R_i = (D_i^-)/(D_i^- + D_i^+)$.

In the original TOPSIS method, the matrix is normalized so as to convert its elements into values in [0, 1]. The normalized value b_i^j is given by:

$$b_i^j = \frac{m_i^j}{\sqrt{\sum_{i=1}^{n}(m_i^j)^2}}, \quad i = (1, 2, \ldots, n), j = (1, 2, \ldots, m), \text{ and}$$

$$B = [b_i^j]_{n \times m}$$

This normalization step gives good results when all criteria have the same magnitude of variation. However, a criterion varying by only a few percent when the other varies by 100% will be discriminated by the method. As it is the role of the decision maker to decide if 10% variation of impact x is worse than 100% variation of impact y, we choose to introduce an alternative to the TOPSIS method where the normalization step is replaced by:

$$b_i^j = \frac{m_i^j - \min(m_i^j)}{\max(m_i^j) - \min(m_i^j)}, \quad j = (1, 2, \ldots, m).$$

In this case, all impacts have the same magnitude of variation and no bias is introduced.

5. Study of the environmental burdens generated by French PET bottles, considering multi-recycling trips

5.1. Impact values in 2010

The average situation in France in 2010 is simulated, using the values given in Table 6, where **c**, **i**, **r**, **p**, **e** and **b** (refer to Table 1) correspond to actual French values (m is slightly inferior to this value, as chemical recycling is not industrially used in France). As already mentioned, the typical situation of France must be highlighted, since a high proportion of its power comes from nuclear sources (around 75%): this explains why energy recovery from PET in France will have a different CO_2 balance from other countries which more rely on fossil fuels.

If an infinity of recycling loops are considered, from its cradle to its graves, 1 kg of PET bottle generates an abiotic depletion of 2.31×10^{-2} kg Sb equiv., an acidification of 8.16×10^{-3} kg SO_2 equiv. and a GWP of 3.12 kg CO_2 equiv. The results are consistent with previous studies (RDC-Environnement, 2007). The respective contribution of the virgin production and the end-of-lives for the three impacts are given in Fig. 8. For the three impacts, the contribution of the virgin production of the bottle to the final burden is overwhelming. It must be observed that

Table 7
Elasticity of the impacts for a selection of parameters.

	Abiotic depletion	GWP
Mechanical recycling efficiency	−0.83	−0.4
Sorting efficiency	−0.77	−0.31
Transportation distance	0.018	0.02
Efficiency of energy recovery	−0.17	−0.16

currently, the global burden of end-of-life, taking into account the avoided impacts, is negative for abiotic depletion and acidification, whereas it is positive for GWP. This is due to the important amount of PET incinerated (0.285 M), and recycled to fibre and film (around 0.36 M), that must also be incinerated after a single use.
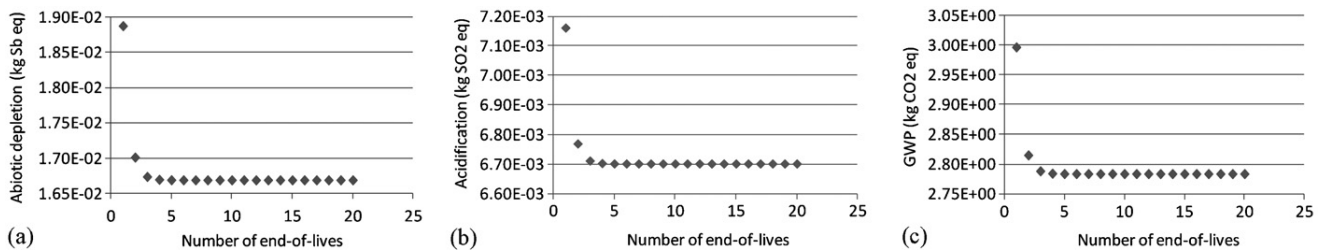
The effect of multiple recycling trips for PET bottles on the final result is analysed (Fig. 9). With the current parameters (Table 6), multiple recycling trips allow a reduction of abiotic depletion (respectively acidification) of 11.1% (respectively 10.9%) more than if there is only a single end-of-life. GWP decreases by only 3.4%. The effect of multiple recycling trips is only significant for the first 2 end-of-lives (see Fig. 9), due to the relatively low value of $\lambda \approx 9\,\%$.
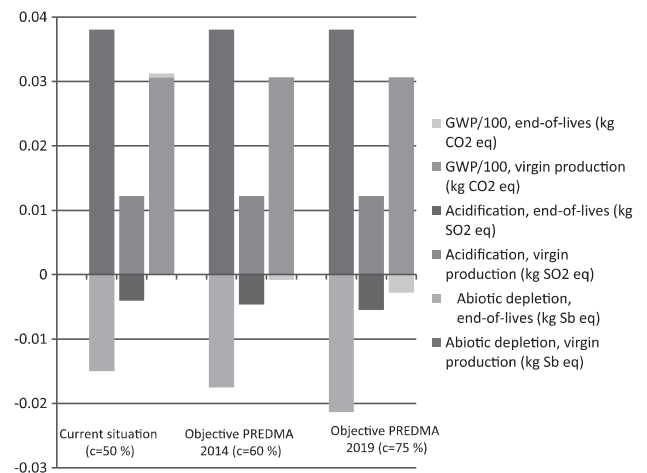
5.2. Sensitivity analysis

A sensitivity analysis was performed for key parameters of the system that are uncorrelated. They are likely to vary according to either the localization in France or the sources of information: yield of mechanical recycling; rate of refusal during sorting at MRF; distance between MRF and recycling plant (a national average distance of 400 km is considered); efficiency of by incineration that can be very variable according to the valorization plant. For the sake of illustration, only the results relative to abiotic depletion and GWP are given (see Table 7) since those for acidification are very similar to abiotic depletion. The elasticity of the impact n for the parameter i is introduced, $\varepsilon = (\Delta I_n/I_n)/(\Delta P_m/P_m)$, with $\Delta I_n/I_n$ relative variation
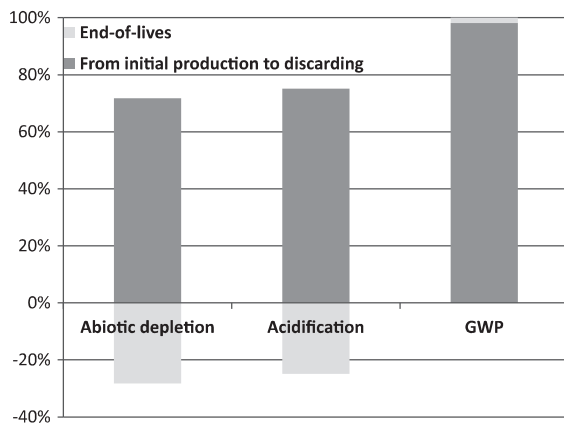
Fig. 10. Effect on the environmental burdens when reaching the PREDMA collection objectives for 1 kg of PET bottles.

of the impact n and $\Delta P_m/P_m$ relative variation of the parameter m. The values of ε are given in Table 7.

Both impacts are rather sensitive to sorting and mechanical recycling efficiency, especially abiotic depletion. Meanwhile, they are moderately sensitive to the efficiency of energy recovery by incineration, and transportation distances have almost no influence. Without increasing the collection and recycling ratio, a significant improvement could be achieved by increasing the efficiency of sorting and mechanical recycling.

5.3. Target PREDMA: measuring the effect of increasing the collection rate

The PREDMA (French acronym for Plan Régional d'Elimination des Déchets Ménagers et Assimilés) is a strategic plan for an eco-efficient waste management in the region Ile-de-France. Concerning packaging waste, the target is to reach a collection rate of 60% by 2014, and 75% by 2019. This target scenario can easily be implemented through simulation while keeping the values of Table 6 for other parameters and under the assumption of multi-recycling trips. With a collection rate increased to 75%, 1 kg of PET bottle would generate from its cradle to its graves an abiotic depletion of 1.67×10^{-2} kg Sb equiv., an acidification of 6.7×10^{-3} kg SO$_2$ equiv. and a GWP of 2.78 kg CO$_2$ equiv. In this situation, the GWP burden implied by the end-of-lives is negative (0.281 kg CO$_2$ equiv. avoided), as the amount of PET bottles directly sent to incineration decreases. The impacts are respectively reduced by 28, 19 and 11% as compared to the current situation Fig. 10.
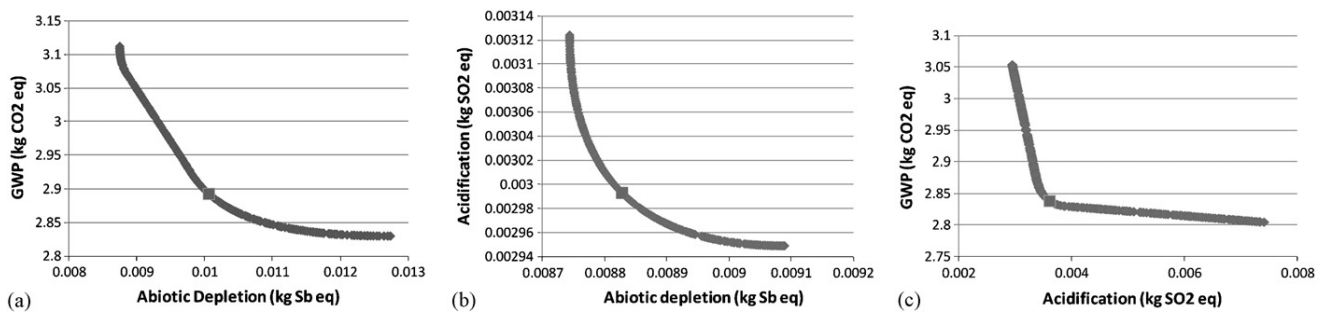

Fig. 8. Contribution of end-of-lives to the impacts from cradle to graves, for 1 kg of PET bottles, France 2010.

Fig. 9. Effect of the number of recycling loops on the final impacts for 1 kg of PET bottles.

Fig. 11. Pareto curves, bi-criteria optimization, Scenario 1 for 1 kg of PET bottles. Best compromises are highlighted by a square.

Table 8
Scenario 1 for 1 kg of PET bottles, decision variables when $q = 1$. Best compromises are in bold letters.

	AD/GWP	AD/Ac	Ac/GWP	AD/GWP/Ac
c	**0.845** (0.74–0.88)	**0.86** (0.83–0.88)	**0.79** (0.75–0.83)	**0.82** (0.75–0.86)
i	**0** (0–1)	1	**0** (0–1)	**0.55** (0–1)
r	1	1	1	1
p	–	–	–	–
m	10^{-6}	10^{-7}	**0** (0–1)	0.8
e	–	–	– (0–1)	10^{-11}
b	–	–	– (0–1)	10^{-5}
f	10^{-7}	10^{-7}	10^{-6}	10^{-11}

Parameters corresponding to the situation in France in 2010.

6. Optimal allocations between valorization routes to reduce impacts

An increase in the collection rate is not the only strategy that can be used to reduce the environmental burdens. Indeed, the selection of the recycling pathway together with the consequent end product for PET influences largely the global impacts. The aim of this part is to optimize the allocation of PET bottles between the different valorization routes so as to minimize the environmental burdens.

Multiobjective problem formulation: For each scenario, the general formulation of the multiobjective nonlinear optimization problem is as follow: determine the decision variables (Table 1) in order to minimize the environmental impacts I_{Ti}, i in 1, ..., m. Eventual additional constraints are specified in each case.

Genetic algorithm parameters: The GA uses the following parameters:

Bi-criteria optimization: population size = 200, number of generations = 300, crossover rate = 0.9, mutation rate = 0.5.
Tri-criteria optimization: population size = 300, number of generations = 1000, crossover rate = 0.9, mutation rate = 0.2.

6.1. Scenario 1: single recycling loop

In this case, a PET bottle can only be recycled once. The fraction λ is regenerated into bottles that are sent to incineration after their use.

Bi-criteria optimization: Three cases of bi-criteria optimization are studied: abiotic depletion versus acidification, abiotic depletion versus GWP, and acidification versus GWP. The resulting Pareto curves are displayed in Fig. 11. Best compromises within the Pareto Front are selected with TOPSIS (see the value of parameters in Table 8). The relative reduction of impacts compared to current situation for the best compromise solution are presented in Table 9.

According to the results in Table 8, some recycling pathways can be directly eliminated. In the three cases, $r \approx 1$ both quaternary recycling by pyrolysis and hydrocraking are therefore less interesting than secondary and tertiary recycling. Similarly, $f \approx 0$, chemical recycling followed by repolymerization into PET is excluded.

Table 9
Scenario 1 for 1 kg of PET bottles, decrease in impacts as compared to current situation for best compromises.

	Bi-criteria			Tri-criteria
	AD/GWP	AD/Ac	Ac/GWP	AD/GWP/Ac
AD	−57.9%	−63%	–	−32.3%
GWP	−10.4%	–	−55.7%	−8.7%
Ac	–	−63.3%	−12.1%	−23.9%

Abiotic depletion/GWP: it is necessary to arbitrate for the determination of the collection rate and the incineration rate. Otherwise, chemical recycling followed by the production of unsaturated polyesters is the chosen pathway. When abiotic depletion is minimal and GWP maximal, $c = 88\%$ and $i = 1\%$. Conversely, $c = 74\%$ and $i = 10^{-6}\%$.

Abiotic depletion/acidification: the only need of arbitration concerns the collection rate. Once again chemical recycling and production of unsaturated polyesters is the chosen pathway.
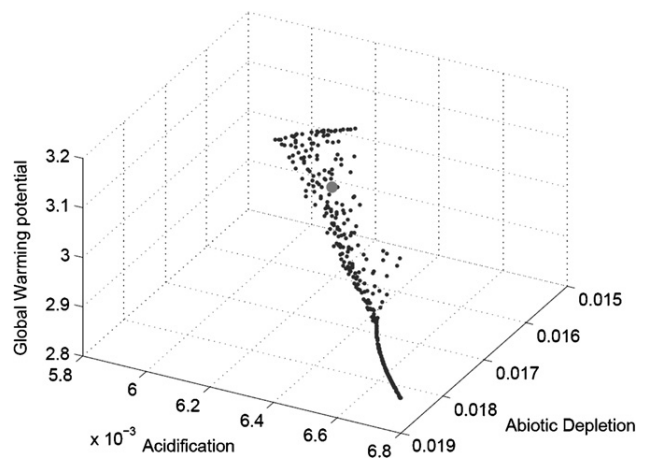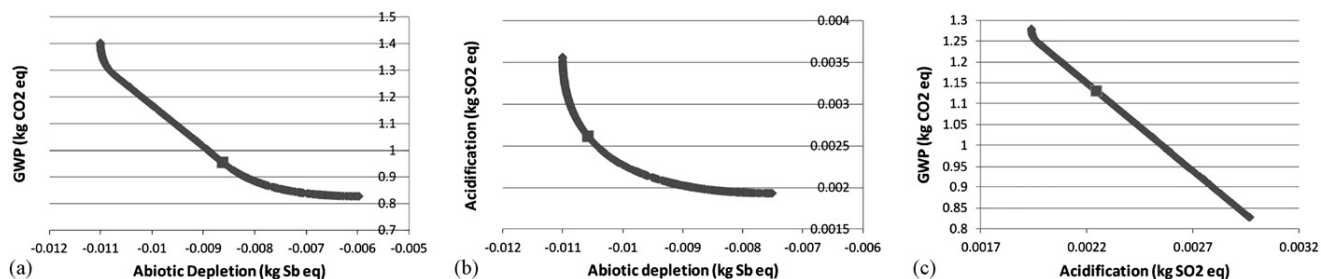

Fig. 12. Pareto curves, tri-criteria optimization, Scenario 1 for 1 kg of PET bottles. Best compromise is highlighted.

Table 10
Scenario 2 for 1 kg of PET bottles, Decision variables when $q \rightarrow +\infty$. Best compromises are in bold letters.

	AD/GWP	AD/Ac	Ac/GWP	AD/GWP/Ac
c	**0.89** (0.86–0.90)	**0.89** (0.84–0.89)	**0.85** (0.84–0.86)	**0.87** (0.84–0.91)
i	**0.04** (0–1)	1	**0.72** (0–1)	**0.94** (0–1)
r	1	1	1	1
p	–	–	–	–
m	1	1	1	–
e	1	1	1	1
b	1	1	1	1
f	–	–	–	–

Fig. 13. Pareto curves, bi-criteria optimization, Scenario 2 for 1 kg of PET bottles. Best compromises are highlighted by a square.

Acidification/GWP: this situation is more complex, as the only consensus being that r clearly tends to 1. GWP is minimized when $c = 75\%$, $i = 0\%$, $m = 1\%$ and $e = 10^{-5}\%$, *i.e.* synthesis of r-PET fibres. Acidification is minimized when $c = 84\%$, $i = 1\%$, $m = 10^{-5}\%$ and $f = 10^{-6}\%$, *i.e.* glycolysis followed by unsaturated polyester synthesis.

In all three cases, the best compromise selected corresponds to glycolysis and unsaturated polyester synthesis of the bottles that are collected. The collection rate is around 80%, being higher when minimizing abiotic depletion is one of the objectives.

Tri-criteria optimization: According to the results of bi-criteria optimization, glycolysis followed by synthesis of unsaturated polyester seems an interesting solution in terms of environmental impacts. Indeed, the use of glycolysed PET for unsaturated synthesis avoids the use of anhydride maleic and other chemicals. However, the data used for the LCA were obtained from a pilot installation (Barboza et al., 2009) and could be different at a larger scale. Moreover, the market of unsaturated polyesters is not large enough to absorb all the flow of PET waste. To be more realistic, the tri-criteria optimization problem is therefore computed with an additional constraint, $m \geq 0.8$, which means that among recycled bottles, at least 80% must be mechanically processed. The resulting Pareto Front is displayed in Fig. 12. The results given in Table 8 show that best solutions are reached when all the collected bottles are recycled, either into fibres by mechanical recycling or into unsaturated polyester by glycolysis. Only the collection and incineration rate have to be adjusted to find the best compromise.

6.2. Scenario 2: infinity of recycling loops

An infinity of closed loop recycling trips is considered, which means that in the model, $q \rightarrow +\infty$. It is now technically possible if the r-PET resin is mixed with enough virgin bottle grade PET (Rieckmann et al., 2011) before being blow moulded, which is the case, proportion of r-PET in bottles being limited to 35%.

The same cases of bi-criteria/tri-criteria optimization than in Section 6.1 are studied. The resulting parameters and best compromises are given in Table 10, the Pareto curves in Figs. 13 and 14. Introducing multiple recycling loops gives an advantage without equivalent to B-to-B by mechanical recycling, whether two or three impacts are simultaneously minimized. In all cases, the only
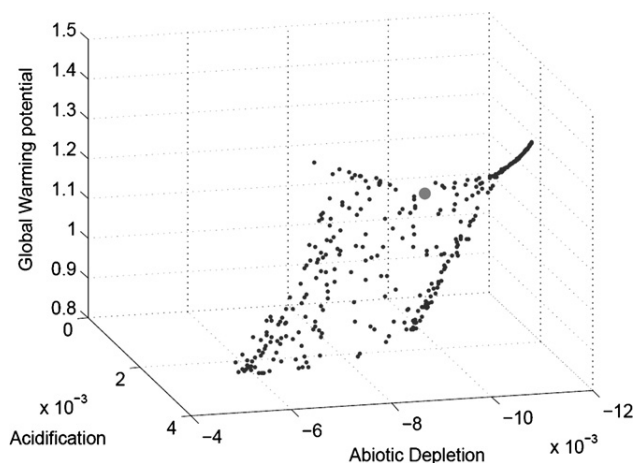
Fig. 14. Pareto curves, tri-criteria optimization, Scenario 2 for 1 kg of PET bottles. Best compromise is highlighted.

variables that have to be adjusted are the collection and incineration rates. The collection rate is higher than in Scenario 1, indeed if we compare best the compromises for tri-criteria optimization, c is 5% higher in Scenario 2. When best compromises are reached, Table 11 shows a drastic decrease in the three environmental burdens as compared to reference situation, especially for abiotic depletion, the decrease being superior to 100%.

The effect of multiple recycling is represented in Fig. 15 where the best compromise for tri-criteria optimization is reached. It is observed that all three impacts decrease significantly up to the

Table 11
Scenario 2, decrease in impacts as compared to current situation for best compromises (for 1 kg of PET bottles).

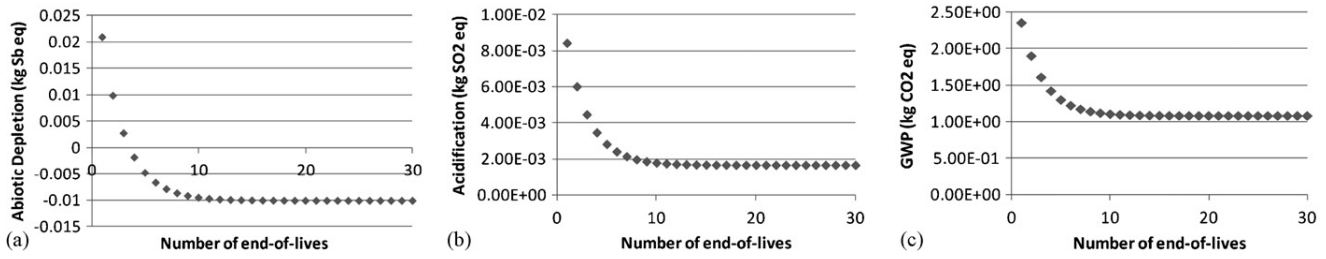|	Bi-criteria			Tri-criteria
	AD/GWP	AD/Ac	Ac/GWP	AD/GWP/Ac
AD	−137.3%	−145.8%	–	−141.4%
GWP	−69.3%	–	−72.5%	−61.4%
Ac	–	−67.9%	−63.7%	−72.5%

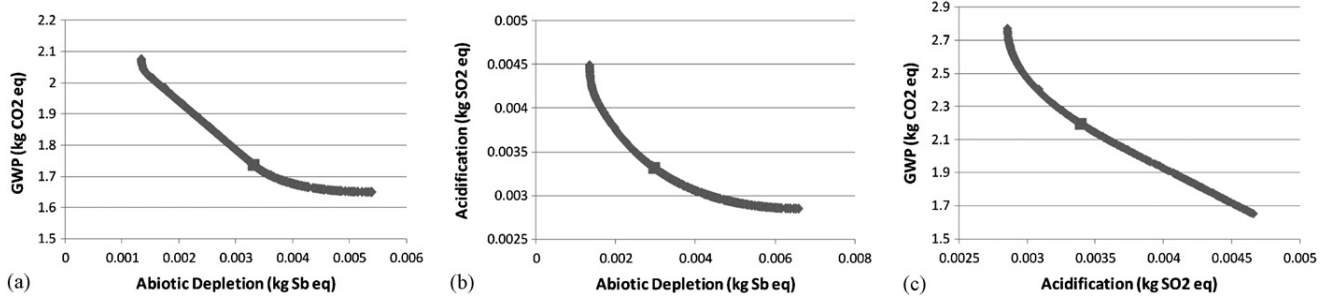Fig. 15. Scenario 2, effect of multiple recycling loops.

Fig. 16. Pareto curves, bi-criteria optimization, Scenario 3 for 1 kg of PET bottles. Best compromises are highlighted by a square.

Table 12
Scenario 3, Decision variables when $q = 3$. Best compromises are in bold letters.

	AD/GWP	AD/Ac	Ac/GWP	AD/GWP/Ac
c	**0.87**(0.82–0.88)	**0.856**(0.83–0.88)	**0.83**(0.82–0.84)	**0.85**(0.82–0.88)
i	**0** (0–1)	1	**1** (0–1)	**0.87**(0–1)
r	1	1	1	1
p	–	–	–	–
m	1	**0.79**(0–1)	**0.86**(0–1)	1
e	1	1	1	1
b	1	1	1	1
f	–	10^{-5}	10^{-5}	1

10th recycling trip, and the threshold is reached after 15 recycling trips. From this value, the decrease in abiotic depletion, acidification and GWP have been respectively multiplied by 2.8, 3.1 and 2.8, as compared to only one recycling loop.

6.3. Scenario 3 for 1 kg of PET bottles: only three recycling loops

A more realistic situation is considered, where PET bottles can only be recycled three times. The resulting parameters and best compromises are given in Table 12, the Pareto curves in Figs. 16 and 17 (Table 13).

Bi-criteria optimization: **Abiotic depletion/GWP**: A compromise is necessary for the collection and incineration rates. Otherwise, B-to-B by mechanical recycling is the chosen pathway.

Abiotic depletion/acidification: There is a need of arbitration concerning the collection rate, and then between B-to-B by mechanical recycling to minimize abiotic depletion, or unsaturated

Table 13
Scenario 3, decrease in impacts as compared to current situation for best compromises.

	Bi-criteria			Tri-criteria
	AD/GWP	AD/Ac	Ac/GWP	AD/GWP/Ac
AD	−85.7%	−87.2%	–	−90.6%
GWP	−44.3%	–	−29.5%	−37.2%
Ac	–	−59.3%	−58.4%	−51%

polyester synthesis to minimize acidification. The best compromise is reached for $m = 0.79$.

Acidification/GWP: The arbitration concerns the collection and incineration rates, and then the allocation between B-to-B by mechanical recycling to minimize GWP, and unsaturated polyester synthesis to minimize acidification. The best compromise is reached for $m = 0.86$.
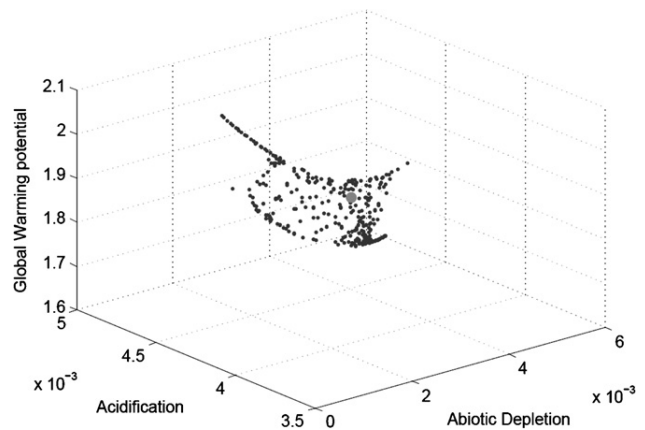

Fig. 17. Pareto curves, tri-criteria optimization, Scenario 3 for 1 kg of PET bottles. Best compromise is highlighted.

For the two bi-objective optimization problems, there is then a compromise to be found between B-to-B and unsaturated polyester synthesis, the former minimizing abiotic depletion and GWP, while the latter minimizes acidification.

Tri-criteria optimization: When the three criteria are simultaneously minimized, only collection and incineration rates are variable within the Pareto front, and B-to-B by mechanical recycling is the chosen path for all recycled bottles. The best compromise is reached for a value of c equal to 85%, which is slightly inferior to the result in Scenario 2.

7. Discussion

Evaluation of the recycling routes: First, it must be said that the three scenarios are in agreement with the literature. With the data used, either secondary or tertiary recycling is always preferable to thermal recycling. This is not surprising since thermal recycling techniques were not created to treat PET (ADEME, 2002), and are more adapted to composite waste flow that can not be sorted. However, the data used for the LCI of pyrolysis and hydrocracking could be outdated as underlined in Perugini et al. (2005). Indeed, this range of recycling techniques suffers from a general lack of data concerning environmental impacts as highlighted in Al-Salem et al. (2009).

Second, for closed loop recycling, mechanical pathway is preferable to glycolysis followed by repolymerization. This difference had already been demonstrated in Shen et al. (2010) in the case of bottle to fibre recycling. Chemical recycling has long been the only possibility to achieve closed loop recycling, but since the beginning of the century mechanical processes have reached the necessary requirements, in terms of mechanical propriety of the polymer and decontamination of the matrix (Welle, 2011). Mechanical processes are also cheaper than chemical depolymerization (Karayannidis and Achilias, 2007; Awaja and Pavel, 2005). This explains the renewal of interest for original applications of glycolysed PET with higher added-value, like unsaturated polyester or polyurethane. Unsaturated polyester synthesis from glycolysed PET seems to be an interesting environmental solution to avoid the used of several chemicals when multi-recycling is not possible. However, the data originating from a pilot plant, additional research is necessary to ensure the viability of the process and confirm the results of this study.

The importance of taking into account the possibility of multi-recycling trips is demonstrated by the difference of results between the three scenarios. Especially, when $q = 1$, as it is the case in many former studies (Chilton et al., 2010; RDC-Environnement, 2007; Nicklaus, 2011) B-to-B by mechanical recycling is not the most interesting solution, the environmental performance being better for unsaturated polyester synthesis and fibres spinning for all three impacts. The comparison between B-to-F and B-to-B by mechanical recycling gives the same results as in RDC-Environnement (2007). However, it must now be included that r-PET mixed with virgin PET and blow-moulded into a bottle has the possibility of being recycled several times, as bottle containing r-PET are not distinguishable from bottles made entirely of fossil PET. With this hypothesis, B-to-B becomes the most interesting path as soon as $q = 3$ when the three impacts are simultaneously minimized. Bottles alone can yet be recycled several times, but PET films from r-PET should soon also be collected to be recycled in France: an experimentation will be conducted by Eco-Emballages in 80 municipalities for two years from January 2012. Of course, energy recovery from PET in France exhibits a typical CO_2 balance due to the fact that nuclear power is the primary source of electric power in France.

Finally, the nonlinear model introduced to assess the impacts of selective collection shows interesting results. Indeed, for all
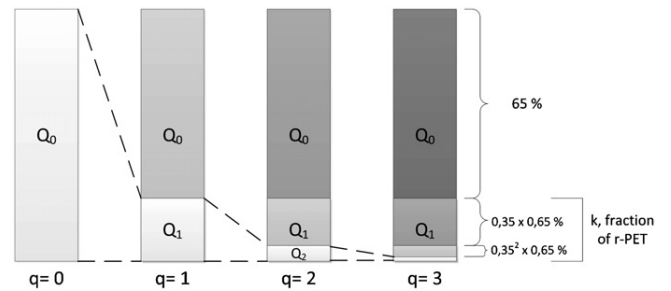
Fig. 18. Mix of resins in a PET bottle. Q_n represents the quality, linked to the number of times this fraction has been recycled, Q_0 being virgin PET and q the number of recycling trips.

scenarios and objectives, best compromises are obtained when c is around 80% or more. This means that even with an increasing difficulty of collection (and consequent environmental impacts) when c is superior to 60%, it is still worth collecting bottles up to 80% rather to discard or incinerate them. It can also be noted that c is higher in the case of multiple closed loop recycling. This illustrates the fact that increasing efforts for bottle collection by fixing a collection rate target superior to 85% is interesting only if closed recycling loop is favoured. This model being partly based on theoretical assumptions, it should be completed by a deeper analysis of consumers propensity to sort their waste and of the collection itineraries. A more regional model including the localization of the facilities (MRF, recycling plant, etc.) with Geographical Information System tools would in this case be interesting,

Multi-recycling loops and quality: In the proposed model, only the impacts generated by M kg of initial fossil PET are assessed. Closed-loop recycling of the fraction λ, either by mechanical or chemical processes, implies that this fraction is mixed with virgin PET before being blow moulded into a bottle. Indeed, even if the final quality of bottle grade r-PET after recycling by mechanical super clean processes is almost similar to the original one (Welle, 2011), there is still a limit of 35% of incorporation of r-PET into bottles (Shen et al., 2011).

A simple model to assess the eventual degradation of the quality of a bottle when a blend of recyled resins are mixed is here developed. A property Q is considered, *e.g.* intrinsic viscosity. After each recycling cycle, the property of the PET resin is degraded, and the relation between cycle $n + 1$ and n is given by $Q_{n+1} = \alpha Q_n$ (Rieckmann et al., 2011), α being the quality retention rate. The propriety after n closed-loop recycling is therefore a geometric sequence and $Q_n = \alpha^n Q_0$. k is the fraction of r-PET introduced in a bottle. This fraction is a mix between PET recycled once, twice, ..., q times, as represented on Fig. 18. The quantity of resin recycled n times introduced in a bottle is equal to $f_n = k^n(1 - k)$ (see Fig. 18 for $q = 1, ..., 4$). Under the hypothesis that the blend of recycled and virgin PET obeys the law of mixes, the final quality Q of a bottle containing r-PET recycled up to q times is given by:

$$Q = \sum_0^q f_n * Q_n = (1 - k)Q_0 \sum_0^q k^n \alpha^n \qquad (3)$$

when $k = 0.35$ and $\alpha = 0.95$ (in the case of intrinsic viscosity, r is near to 0.98 according to the results in Rieckmann et al. (2011)), for $q \rightarrow +\infty$, Q quickly converges towards $\approx 0.973 \times Q_0$. The hypothesis of an infinity a closed-loop recycling therefore does not imply serious degradation of the quality of the bottles considering current practices.

Economical viability: The environmental performance of the different processes is not the only argument that has to be considered to design PET waste management system. An environmentally

friendly solution that is not economically viable will not be selected. The difficulty of modelling the economical flows of the PET waste management system relies on the fact that involved stakeholders generally have conflicting interests, economical profits or costs being different for industrials, municipalities and citizens. A global approach similar to the one adopted for environmental impacts is therefore of limited interest. The model developed in this study is a useful tool to select most interesting processes in terms of environmental impacts. This process being selected, its economical viability must then be tested by classical methods (Net Present Value, annual cost (Ouattara et al., 2012), etc.).

8. Conclusions and perspectives

This study presented a methodology for the optimization of a waste management system, using non-aggregated LCA results for decision support. It was applied to the case of PET bottles in France. A parametrizable assessment of the impacts generated by the end-of-lives of PET bottles was first performed, introducing a nonlinear model for the collection stage. A key assumption of the model is the number of recycling trips taken into account, recent technical improvements now allowing PET bottles to be recycled several times if blended with enough virgin PET. The results show that consequent impacts are very different. Moreover, multi-recycling trips induces a significant decrease in all environmental impacts, as illustrated by the current case of France.

The use of a genetic algorithm is an efficient method to define the optimal allocation of bottles between valorization paths. The output being a set of non-dominated solutions or Pareto front, decision support tools are needed to select best compromises. According to the number of recycling trips possible, the optimal collection rate varies between 80 and 90% which is in average ten percent higher than the target values fixed for 2019 in France. Among recycling paths, for only one recycling trip, synthesis of unsaturated polyester or fibres is less impacting than B-to-B recycling. However, as soon as several recycling trips are possible, as it is now the case in France, r-PET being introduced into bottles, B-to-B becomes by far the best solution for all impacts. This result could be modified if other products made from r-PET (films, etc.), are further recycled instead of being incinerated.

This methodology can be easily adapted to other packaging wastes, e.g. paper, glass or metals, to define optimal applications of recycled products, and to compare different recycling processes or several applications of the recycled materials. Moreover, concerning PET bottles, further insight on their environmental impacts could be gained by applying the method to raw material supply. This study assumes that PET is synthesized from fossil EG and TPA. However, since 2011, different companies launched bio-based bottles, containing EG made from bioethanol. A key issue is the supply of biomass of the biorefinery (wheat, beat, sugar cane, etc.), and the proposed methodology could be a useful tool to determine compromises that simultaneously minimize environmental impacts.

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