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ABSTRACT

Many industrial food processes are multi-steps, multi-products systems, and sharing the environmental impacts produced by each step to each product is critical when implementing life cycle assessment (LCA) method. The objective of the study was therefore to investigate subdivision and different allocation rules as means of accounting for branched process itineraries such as the fractionation of milk into cream, casein, whey proteins and lactose as a case study. Depending on the mass, dry matter, protein or economic allocation, single products may or may not bear significant environmental impacts, thereby stressing the need for equally detailed inventories for all co-products. Aggregating the results by step or by input further helps identifying hotspots. Methodological choices in LCA of multifunctional systems are therefore strategic decisions that ultimately affect the eco-design of products, processes and food chains.

1. Introduction

In most food sectors, processing involves partitioning the agricultural commodity into various co-products and wastes. Even in the case of fresh items like fruit or fish, operations like cleaning, sorting and waste valorization generates different food co-products, while extraction and separation technologies are frequent, in particular in the mill, oil, meat, egg and dairy industries, in the purpose of offering diversified products to consumers or of easing reassembly in industrial food recipes. Food production is currently one of the human activities that contribute most to many environmental impacts. It is responsible for about 30% of greenhouse gas (GHG) emissions, 70% of freshwater withdrawals and 40% of land occupation (Poore and Nemecek, 2018; Willett et al., 2019; Campbell et al., 2017). In addition, it is associated with biodiversity losses (Foley et al., 2005; Rigal et al., 2023). Off-farm activities, among which food processing demonstrates small contributions compared to on-farm activities. For instance, processing alone represents less than 5% of GHG emissions, terrestrial acidification or freshwater and marine eutrophication due to food production globally (Poore and Nemecek, 2018). Nevertheless, it has significant responsibility in reaching the United Nations sustainable development goals for food systems. First, processing prevents food spoilage and food waste by stabilization of products and by management of co-products, therefore notably acting for resource sparing and for sustaining a growing population (De Marco and Iannone, 2017; Sasaki et al., 2022a, 2022b; Bacenetti et al., 2018). Second, being downstream to agricultural production implies that food processing is the link where the farms' environmental burden is managed and, eventually, shared between the co-products, with important consequences on the way public policies and consumers will consider single products as environmentally friendly or not (Biswas and Naude, 2016). Third, food processing consumes large amounts of energy and water, which are both critical resources in the context of climate change and unstable geopolitical context.

All these responsibilities apply to dairy manufacturers, especially as milk is a nutritious, culturally accepted and sanitary fragile animal product. The dairy sector alone accounts for 3–4% of total GHG

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emissions worldwide (Milani et al., 2011) and 6–7% in France (CITEPA and Floréal, 2020; CNIEL, 2021) including meat co-production and dairy processes and storage. In Western countries, processing accounts for 5–35% of the dairy products' impacts on GHG emissions, land use, water scarcity, acidification or eutrophication of natural compartments (CITEPA and Floréal, 2020; Lovarelli et al., 2022; Cortesi et al., 2022; Djekic et al., 2014; Hayek et al., 2021; Jungbluth et al., 2018). Taking eco-design and mitigation actions requires reliable and consensual assessment of the products' potential impacts on the environment, such as life cycle assessment (LCA – (ISO, 2006; European Commission, 2010)). However, LCA's results can be strongly influenced by the practitioner's goal and methodological choices, regarding e.g. the functional unit, system detail and boundaries (Jolliet et al., 2017). Many agri-food processes being multi-step and multi-product systems, another key issue is that of the respective share of impacts to be distributed between products. Three main approaches exist for attributional LCAs.

- Subdivision, consisting in dividing the unit process into sub-processes for which specific inputs and outputs of each product can be identified and the related impacts calculated and attributed to them;
- System expansion, consisting in including the additional functions related to the co-products into the functional unit;
- Allocation, consisting in attributing a share of the total impacts to each co-product, according to a chosen allocation rule.

The ISO standards establish a hierarchy between these options (ISO, 2006). If possible, allocation should be avoided by applying subdivision or system expansion that maintain integrity and realism of the input data. In the latter, the impacts of a given product are deduced by subtracting the impacts of equivalent products (substitution) from the total impacts of the expanded system (Jolliet et al., 2017). However, it can be impaired by a lack of knowledge on the substitutable systems or imperfect functional equivalence (Houssard et al., 2020; Ekvall and Finnveden, 2001). Subdivision alone attributes all the impacts of common processes onto the main product, even though co-products are generated (Aguirre-Villegas et al., 2012). When allocation cannot be avoided, impacts should be partitioned in a way that reflects the "underlying physical relationship" between the inputs/outputs flows and the products, or by default in a way that reflects another relationship between co-products, such as respective economical values (ISO, 2006; Jolliet et al., 2017). Applying allocation matrices directly to multi-product plants as "black boxes" to alleviate LCA practitioners' workload in the industry helps for comparative evaluation (Jungbluth et al., 2018; Feitz et al., 2007) but fail to support eco-design of industrial processes. Notably, they can only refer to constant technologies and products (Feitz et al., 2007) and lack sufficient detail to spot virtuous in-situ heat or mass exchanges. They also take the risk to allocate downstream burdens to upstream products, e.g. refrigeration of yoghurts to UHT skim milk. In a pioneer paper, a gate-to-gate life cycle of Wisconsin's cheese production has been assessed using subdivision, allocation or a combination of both in order to take whey valorization into account after cheese manufacture has reached an industrial scale sufficient to avoid whey spreading (Aguirre-Villegas et al., 2012). The system involved 2 separation operations and 3 co-products, and the functional unit was defined as 1 kg of cheddar cheese, thereby stating a hierarchy between cheese, whey cream and dry whey. However, dairy plants have continued to gain in complexity to best valorize all milk components. Depending on market trends, a co-product like butter or sweet whey may be in a position to replace cheese as a driver for milk processing. The objective of this paper is therefore to investigate the sensitivity of LCA's results to practitioner's choices when it comes to partition the environmental burden between 5 equally treated co-products of a French milk processing plant. Using a 4-step fractionation cascade as a case study, the present report compares a simplified black box approach with a combination of subdivision and allocation (Aguirre-Villegas et al., 2012). When allocation is used, LCA's results are sensitive to the chosen rule, raising the issue of potential "impact transfers" between co-products depending on stakeholders' diverging interests (Wilfart et al., 2021; Kyttä et al., 2022). Although the least recommended choice according to ISO, economic allocation is widely used in food LCAs (Wilfart et al., 2021; Kyttä et al., 2022; van der Werf and Nguyen, 2015). The International Dairy Federation (IDF) recommends dry matter allocation for the processing phase (IDF, 2022). Composition-based allocation rules (or functional units) are also in debate to account for technology-based drivers in dairy processing (Flysjö et al., 2014). For these reasons, dry matter (as the IDF reference), mass, protein and economic allocations were compared and discussed in this report. The detailed life cycle inventory further allowed to analyze the results either by processing step or by type of input, for a cross-identification of hotspots.

2. Materials and Methods

2.1. Goal and scope

The goal of this study is to investigate the influence of methodological choices related to multifunctionality on LCA results when evaluating the respective environmental impacts of 5 co-products, generated by a cascade of separation processes. The sensitivity of the results to the allocation rule is also considered. The scope is that of typical dairy plants in France during the 2015–2020 period, processing over 100 000 tons of milk yearly each and collecting over 80% of dairy farms altogether (CNIEL, 2022).

2.1.1. System description

To achieve this goal, a complete food processing system was required, producing multiple products through multiple separation operations (fractionation). Regarded as a "black box", the system is a dairy plant that processes raw milk into 5 co-products: raw cream, micellar casein, concentrated lactose, β-lactoglobulin and α-lactalbumin enriched ingredients (Fig. 1A). Complete system description and life cycle inventory can be found in Gésan-Guiziou et al. (2019) and in Guyomarc'h et al. (Guyomarc'h et al., 2023). Briefly, raw milk is first skimmed to produce raw cream as a first product; then skim milk is subjected to microfiltration to produce micellar casein that is the second product of the system (Fig. 1B). At this stage, whey is generated as a co-product of cream and casein. Long regarded as a waste, whey has motivated efforts to retrieve value out of its nutrients and water contents. In particular, the major whey proteins α-lactalbumin and β-lactoglobulin have good texturing properties, and α-lactalbumin is sought for its nutritional value in infant formula. In the considered system, lactose (and minerals) is separated from proteins using ultrafiltration, then reverse osmosis is used to concentrate lactose as the third product. In the protein flow, α-lactabumin is selectively precipitated then separated from β-lactoglobulin by microfiltration. Once resolubilized, each protein is concentrated by ultrafiltration and evaporation, then spray-dried to yield the fourth and fifth products (Fig. 1B). While membrane separations are typically run at 50–55 °C, intermediate products are kept safe by cooling and cool storage at 4 °C up to final stabilization by drying. Beyond pasteurization, the system therefore involves numerous heat exchanges. It also involves chemicals, water and heat for cleaning (cleaning in place + rinsing water) at each operation (not shown in Fig. 1). The life cycle inventory also includes farming (embedded in the raw milk input), equipment materials (machines and consumables, as for instance tanks, pipes or membranes) and transport of the whey from one factory to another, but excludes other infrastructures (e.g. buildings) or labour-related activities (e.g. support and administration). It also excludes downstream processing of cream, micellar casein and lactose concentrate into butter, cheese and powder, respectively, as these processes were not required for the study's goal, centered on handling a protein fractionation system (Gésan-Guiziou

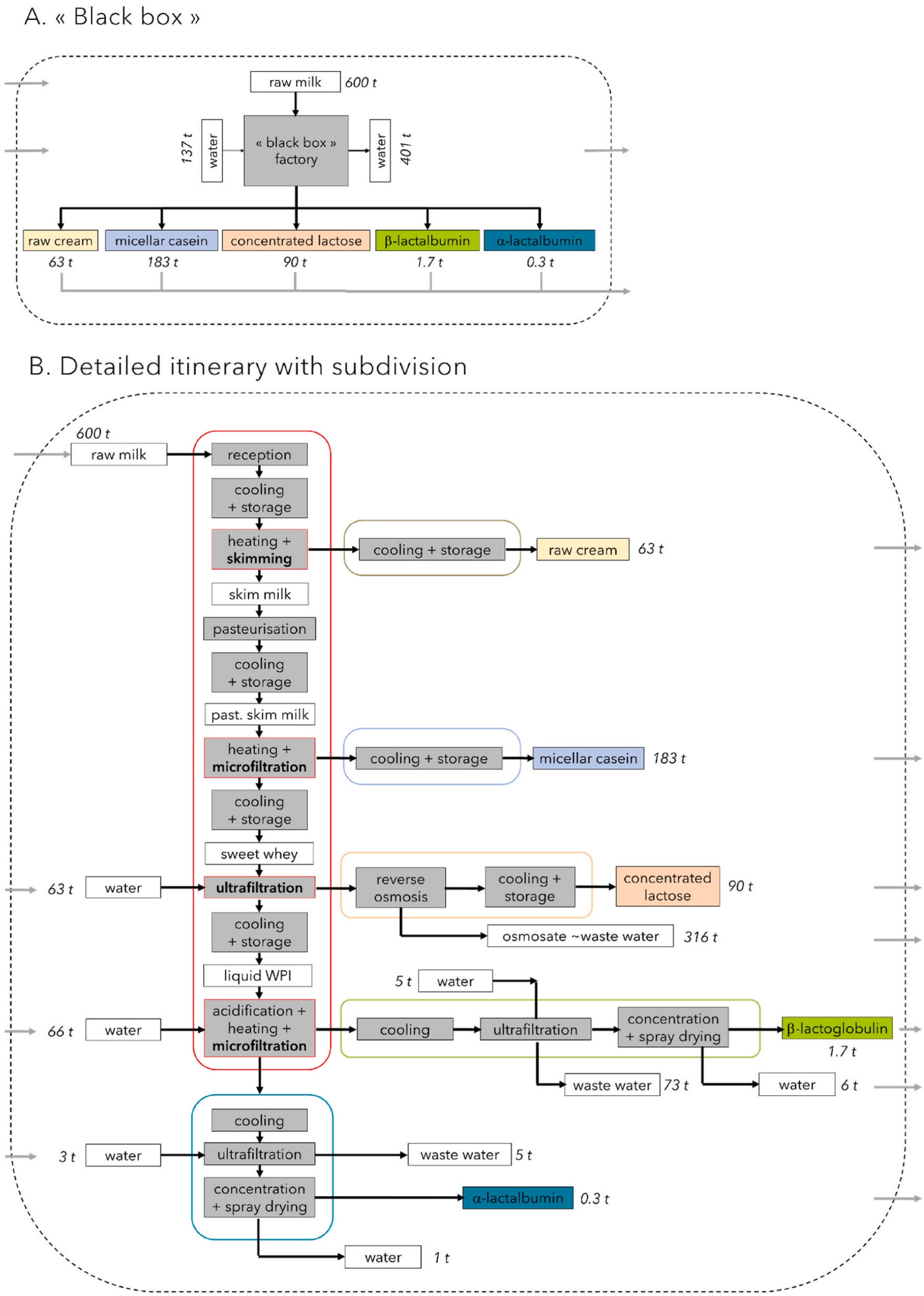

Fig. 1. (A) Black box and (B) detailed itinerary with subdivision diagrams of the same farm-to-gate milk fractionation system. The dashed box represents the system boundary. Input raw milk and the intermediate products are in white boxes. The 5 co-products, i.e. raw cream, micellar casein, concentrated lactose and the two enriched β-lactoglobulin and α-lactalbumin ingredients are in colored boxes. In (B), the colored lines show the subdivisions' boundaries. Environmental impacts of the processes enclosed in the yellow, sky blue, orange, green and green blue boundaries are 100% attributed to raw cream, micellar casein, lactose, β-lactoglobulin and α-lactalbumin, respectively. Environmental impacts of the processes enclosed in the red boundary are shared between the intermediate products at each separation process (namely: skimming, microfiltrations and ultrafiltrations). The operation units that required allocation are in bold. All the allocation factors can be found in (Guyomarc'h et al., 2023). Less than 1% error in mass balance is due to rounding and uncertainties. WPI: whey protein isolate.

et al., 2019).

2.1.2. Functional unit, system boundary and approach

The chosen approach is that of attributional LCA with farm-to-gate system boundary. The system boundary includes milk production, cooling and storage at the farm stage and all thermal, separation and cleaning processes at the plant stage, as well as storage, transport and wastewater treatment (Fig. 1). The fate of the 5 co-products inside or outside the plant is not included. The functional unit is taken as 1 day of plant activity, resulting in the transformation of 600 tons of raw milk into 63 tons of raw cream, 183 tons of micellar casein, 90 tons of concentrated lactose, 1.7 ton of β-lactoglobulin enriched ingredient and 0.3 ton of α-lactalbumin enriched ingredient. While the production of α-lactalbumin is not a driver for cream or casein production, it is a driver for processing whey into enriched protein ingredients. To illustrate the relevance of subdivision combined with sensitivity on allocation in the purpose of eco-design, the potential environmental impacts of the most downstream product, the α-lactalbumin enriched dried ingredient, were analyzed along its farm-to-gate life cycle. In that case, the functional unit was 1 kg of the final product.

2.2. Densities, compositions and prices of products and intermediate products

Foreground data is that of Gésan-Guiziou et al. (2019), where operational data is in volume and no allocation is applied, as impacts were assessed for the whole plant and not related to any specific output (i.e. the functional unit was to treat a daily volume of milk). Table 1 shows the compositions, density and price of products and intermediate products that are necessary to apply allocation at every separation step.

The density of skim milk, micellar casein retentate, sweet whey permeate, liquid whey protein isolate and the aqueous milk phase was measured at 50 °C with a DMA48 densitometer (Anton Paar, Courtaboeuf, France). That of the lactose concentrate was calculated from the mass balance of the reverse osmosis operation, regarding the osmosate as water. The density of the liquid whey protein isolate was taken as a proxy for all single-protein intermediate products. The dry matter content, fat content and total protein content of skim milk, micellar casein retentate, sweet whey permeate and the liquid whey protein isolate were determined experimentally using the standard desiccation method, gravimetric method and Kjeldahl method, respectively (ISO, 2010; ISO, 2022; ISO, 2009; ISO, 2016). From the separation of sweet whey onwards, all downstream products are regarded as fat-free. Products' prices were established from expert opinion (see Acknowledgements). A fictive price was attributed to intermediate products resulting from separation steps, namely the aqueous milk phase, the liquid whey protein isolate, and wet fractions of α-lactalbumin and β-lactoglobulin after microfiltration. Fictive prices were calculated as to keep the final revenue constant (i.e. mass × price of the final product – (Jolliet et al., 2017)) through the lactose and whey streams up to the sweet whey ultrafiltration. The cost of intermediate operations was neglected. All other required information came from the literature.

2.3. Partitioning of the environmental impacts

In the present study, subdivision was applied to attribute the environmental impacts of downstream operational units only to the co-product that is generated by them. Allocation was applied to the rest

Table 1
Producing step, dry matter, fat and protein content, density and price of the products and intermediate products of the detailed system. Expert opinion for protein fraction's composition comes from confidential industrial experiments at UMR STLO. Expert opinion for prices comes from personal communications by professionals within the dairy industry. Sources: Jolliet et al. (2017); Gésan-Guiziou et al. (2019); IDF (IDF, 2004); Jeantet et al. (2001); ANSES (Agribalyse, 2024); FranceAgriMer (FranceAgriMer and Prix des produits laitiers, 2022).

Product	From	Dry matter (% g.g^{-1})	Fat content (% g.g^{-1})	Protein content (% g.g^{-1})	Density (kg.m^{-3})	Sources	Price (€.kg-1)	Source
Raw milk	Farm + transport	12.7 (1)	4.0 (1)	3.2 (1)	1030 (at 20 °C) (2)	(1) IDF (2004) (2) Jeantet et al. (2001)		
Raw cream	Skimming	37.1 (1)	30.7 (1)	2.4 (1)	981 (at 50 °C) (2)	(1) ANSES (2020) (2) Jeantet et al. (2001)	4.50	FranceAgriMer (2022)
Skim milk	Skimming	9.2 (1)	<0.1 (1)	3.3 (1)	1023 (at 50 °C) (1)	(1) Measured	0.50	FranceAgriMer (2022)
Micellar casein	Milk MF	13.9 (1)	0.2 (2)	8.0 (1)	1045 (at 50 °C) (1)	(1) Measured (2) Calculated	10.50	Expert opinion
Sweet whey	Milk MF	6.3 (1)	0.0 (2)	0.6 (1)	1014 (at 50 °C) (1)	(1) Measured (2) Hypothesized	1.05	Expert opinion
Liquid whey protein isolate (WPI)	Whey UF	11.9 (1)	0.0 (2)	11.4 (1)	1026 (at 50 °C) (1)	(1) Measured (2) Hypothesized	2.26	Fictive price (Jolliet et al., 2017)
Aqueous milk phase	Whey UF	5.1 (1)	0.0 (2)	0.0 (2)	1012 (at 50 °C) (3)	(1) Calculated (2) Hypothesized (3) Measured	~0	Fictive price (Jolliet et al., 2017)
Concentrated lactose solution	RO	24.0 (1)	0.0 (2)	0.0 (2)	1056 (–) (1)	(1) Calculated (2) Hypothesized	1.50	Expert opinion
Osmosate	RO	<0.1 (1)	0.0 (1)	0.0 (1)	1000 (–) (1)	(1) Hypothesized	0	
a-lactalbumin enriched precipitate	Acidification + MF	7.1 (1)	0.0 (2)	7.1 (1)	1026 (–) (2)	(1) Expert opinion (2) Hypothesized	1.40	Fictive price (Jolliet et al., 2017)
b-lactoglobulin enriched solution	Acidification + MF	1.5 (1)	0.0 (2)	1.5 (1)	1026 (–) (2)	(1) Expert opinion (2) Hypothesized	0.29	Fictive price (Jolliet et al., 2017)
Concentrated b-lactoglobulin	UF	15.3 (1)	0.0 (2)	14.7 (1)	1026 (–) (2)	(1)Calculated (2) Hypothesized	–	
Dried b-lactoglobulin enriched ingredient	evaporation. + spray-drying	96.0 (1)	0.0 (2)	91.8 (1)	–	(1) Gésan-Guiziou et al. (Gésan-Guiziou et al., 2019) (2) Hypothesized	12.5	Expert opinion
Concentrated a-lactalbumin	UF	17.7 (1)	0.0 (2)	17.6 (1)	1026 (–) (2)	(1)Calculated (2) Hypothesized	–	
Dried a-lactalbumin enriched ingredient	evaporation + spray-drying	96.0 (1)	0.0 (2)	95.1 (1)	–	(1) Gésan-Guiziou et al. (Gésan-Guiziou et al., 2019) (2) Hypothesized	15	Expert opinion

of the system, i.e. whenever subdivision was not possible ((Aguirre--Villegas et al., 2012; IDF, 2022) – Fig. 1B). Whole-system allocation, corresponding to a black box situation was also calculated for comparison (Fig. 1A). In compliance with the International Dairy Federation guidelines, dry matter (i.e. milk solids) allocation was applied as reference at the processing stage for all food grade products (IDF, 2022).

2.4. Sensitivity to the allocation rule

The same allocation rule was applied throughout the system for the assessment's consistency, but was varied from one assessment to another to evaluate the consequence of this choice on the results. Mass, protein and economic allocations were considered. Each product received a proportion of the total environmental impacts of the input flows respective of its share of the total mass, total protein mass or total economic revenue of the products, respectively.

2.5. Impact assessment

LCA assessment was performed for the final quantity (in kg) of each product or for 1 kg of α-lactabumin using the SimaPro Analyst software (release 9.5.0.1, PRé Sustainability, Amersfoort, The Netherlands) loaded with the Agribalyse 3.0.1 and EcoInvent 3.8 databases. The Environmental Footprint characterization method (EF 3.0 release 1.03) was chosen and the 16 midpoint impact categories were calculated and complete results are provided in the associated dataset (reference (Guyomarc'h et al., 2023)). Excepting for general results (Table 2), the present study focuses on the following impact categories: climate change as a marker of carbon footprint, land use as a marker of agriculture, ionizing radiations as a marker of electricity consumption (for the French electricity mix), water use as a marker of water footprint and consumption of fossil energy resources. Fossil energies (especially gas) and electricity consumptions are markers of industrial heating and cooling processes, respectively, as illustrated by the thermal history of the products along the process itinerary (Fig. 1B). For the detailed impact assessment of 1 kg α-lactalbumin in the perspective of eco-design, the results were aggregated according to 2 different rules.

- aggregation by processing stage: milk production, skimming, pasteurization, microfiltration, cooling of whey, whey protein ultrafiltration and concentration, cooling of whey protein retentate, transports, whey protein microfiltration and separation, cooling of α-lactalbumin retentate, α-lactalbumin ultrafiltration and concentration and spray drying (results in Fig. 4)
- aggregation by input type: milk, machines, energy for processing, energy for drying, water, cleaning, transport and chemicals (results in Fig. 5)

3. Results

3.1. Comparison between black box and subdivision with a single allocation rule

The black box and the subdivision systems were compared, using dry matter (DM) as the allocation rule. In the case of a black box system, the impacts are allocated to each of the 5 co-products proportionally to their dry matter balance, i.e. the shares are the same for any impact category (Fig. 2, first left column). Impacts are 30–35% on lactose, casein and cream, which reflects the milk composition where lactose, proteins and fat are the 3 major milk components (Table 1).

With subdivision and dry matter allocation at every separation step, the share of impacts attributed to the α-lactabumin and β-lactoglobulin enriched ingredients increased from 2 to 3% in the black box system to 15–23% in the detailed system, mostly to the benefit of lactose (decrease from 30% in black box to 14–20% with subdivision) or cream (decrease from 32 to 21%). This increase was largest (+20%) for the "ionizing radiations" impact category, and 19% for the "fossil resource use" impact category, which are respectively related to electricity and gas consumptions. With the subdivision approach, the environmental impacts of the industrial cascade are shared between the co-products with the closest possible reference to the actual process itinerary. While the black box system shares the impact of the whey fractionation, cooling and spray-drying between all co-products according to their share in total DM, the combination of subdivision and allocation only attributed these processes to the α-lactalbumin and β-lactoglobulin ingredients (Fig. 1). This result evidences subdivision is as a preferred approach for the LCA of industrial cascades with products released at different stages along the itinerary.

3.2. Comparison of the results using four different allocation rules

As an illustration, the impacts on climate change are presented in Fig. 3 to illustrate the consequences of the allocation rules on LCA's results, while similar results were observed for other impact categories. Regardless of the allocation rule, the subdivision approach increased

Table 2
Environmental impacts, in absolute values, of the manufacture of 1 kg α-lactalbumin enriched ingredient, using the EF 3.0 characterization method and a subdivision + allocation approach. Abbreviations: kg CO_2 eq = kg CO_2 equivalent; kg CFC-11 eq = kg trichlorofluoromethane equivalent; kBq U-235 eq = kBq uranium 235 equivalent; kg NMVOC eq = kg non-methane volatile organic compounds equivalent; inc. = increase; CTUh = comparative toxic unit for human; CTUe = comparative toxic unit for ecosystems; Pt and mPt = point and millipoint; depriv. = deprivation.

impact categories	unit	mass allocation	dry matter allocation	protein allocation	economic allocation
Climate change	kg CO_2 eq	5.0×10^{0}	$6.2 \times 10^{+1}$	$5.3 \times 10^{+1}$	$3.7 \times 10^{+1}$
Ozone depletion	kg CFC-11 eq	7.2×10^{-7}	2.7×10^{-6}	2.5×10^{-6}	2.1×10^{-6}
Ionizing radiation	kBq U-235 eq	1.2×10^{0}	5.3×10^{0}	5.1×10^{0}	4.6×10^{0}
Photochemical ozone formation	kg NMVOC eq	9.5×10^{-3}	9.9×10^{-2}	8.5×10^{-2}	6.1×10^{-2}
Particulate matter	disease inc.	2.6×10^{-7}	4.7×10^{-6}	4.0×10^{-6}	2.8×10^{-6}
Human toxicity, non-cancer	CTUh	7.0×10^{-8}	1.2×10^{-6}	1.0×10^{-6}	7.1×10^{-7}
Human toxicity, cancer	CTUh	3.1×10^{-9}	2.7×10^{-8}	2.3×10^{-8}	1.7×10^{-8}
Acidification	mol H+ eq	3.3×10^{-2}	6.9×10^{-1}	5.8×10^{-1}	4.0×10^{-1}
Eutrophication, freshwater	kg P eq	8.6×10^{-4}	6.8×10^{-3}	6.0×10^{-3}	4.5×10^{-3}
Eutrophication, marine	kg N eq	8.6×10^{-3}	2.1×10^{-1}	1.7×10^{-1}	1.2×10^{-1}
Eutrophication, terrestrial	mol N eq	1.1×10^{-1}	2.9×10^{0}	2.5×10^{0}	1.7×10^{0}
Ecotoxicity, freshwater	CTUe	$8.7 \times 10^{+1}$	$1.0 \times 10^{+3}$	$8.9 \times 10^{+2}$	$6.4 \times 10^{+2}$
Land use	Pt	$9.4 \times 10^{+1}$	$3.1 \times 10^{+3}$	$2.6 \times 10^{+3}$	$1.7 \times 10^{+3}$
Water use	m3 depriv.	5.3×10^{0}	$2.7 \times 10^{+1}$	$2.6 \times 10^{+1}$	$2.1 \times 10^{+1}$
Resource use, fossils	MJ	$6.5 \times 10^{+1}$	$2.6 \times 10^{+2}$	$2.5 \times 10^{+2}$	$2.1 \times 10^{+2}$
Resource use, minerals and metals	kg Sb eq	2.3×10^{-5}	8.9×10^{-5}	8.5×10^{-5}	7.7×10^{-5}
Product Environmental Footprint (PEF) score	mPt	0.49	6.01	5.09	3.61

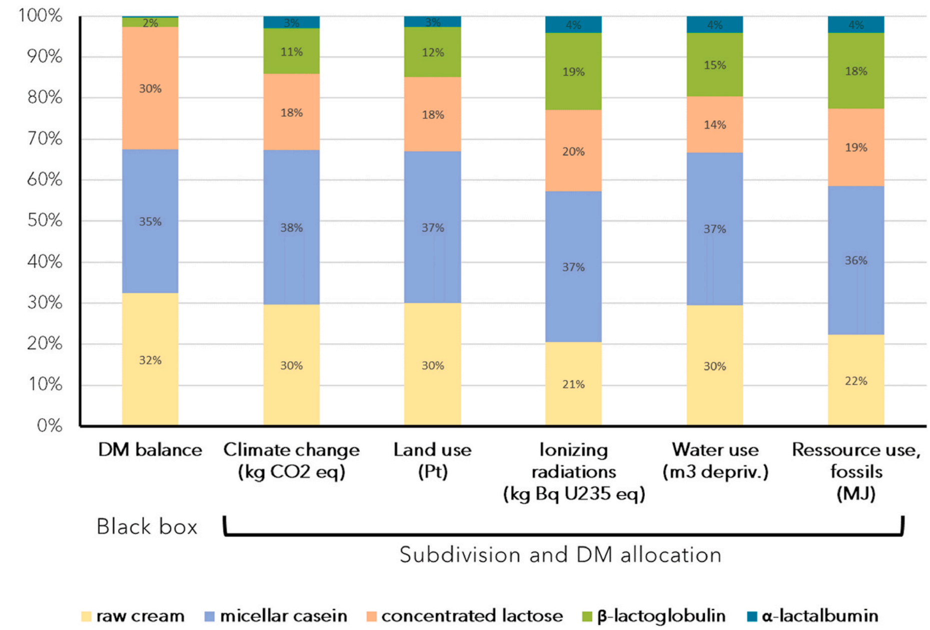

Fig. 2. Respective shares of environmental impacts attributed to raw cream, micellar casein, concentrated lactose, β-lactoglobulin and α-lactalbumin enriched ingredients using dry matter (DM) allocation in a black box system (first left column, representing any impact category) or in a detailed system with subdivision (second to sixth columns, representing in that order: climate change, land use, ionizing radiations, water use and use of fossils resources). Abbreviations: kg CO_2 eq = kg CO_2 equivalent; Pt = point; kBq U-235 eq = kBq uranium 235 equivalent; m3 depriv. = m^3 deprivation.

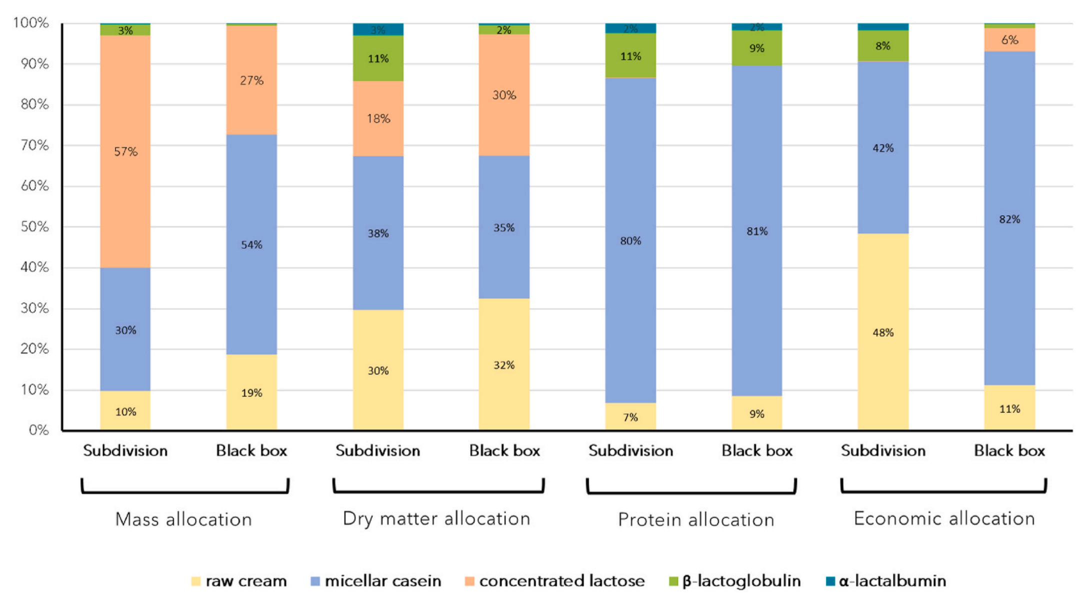

Fig. 3. Respective shares of the impacts on climate change attributed to raw cream, micellar casein, concentrated lactose, β-lactoglobulin and α-lactalbumin enriched ingredients, depending on the allocation rule. For each allocation rule, the results were calculated using black box system or the detailed system with explicit subdivision (respectively left and right columns for each allocation rule).

again the share of impacts attributed to the α-lactalbumin and β-lactoglobulin ingredients, by preventing the transfer of impacts that occurs in the black box approach from downstream processes towards upstream products. Compared to the dry matter allocation, mass allocation increased the share of the impacts attributed to lactose in a subdivision system from 18 to 57% (Fig. 3). Water accounts for the difference between mass and dry matter and this illustrated the issue of water content (genuine + input) in allocating environmental impacts and eventually in comparing environmental footprints of different food products. In agreement with their compositions (Table 1), no environmental burden is allocated to lactose and less than 10% is allocated to raw cream when using a protein allocation. Micellar casein was attributed 30–82% of the environmental impact on climate change across all allocation rules. As a main product of dairy industry, it is ponderous, protein-rich and valuable altogether.

Economic is the allocation that was most sensitive to subdivision in this system (Fig. 3). Based on the revenue, it is affected by both price and the weight produced. At the first separation step (skimming), the environmental impacts were shared in 50% halves, because of the high price of raw cream with respect to skim milk (Guyomarc'h et al., 2023). Consequently, at the skimming stage only 50% of the impacts went to the protein/lactose streams with the subdivision approach, whereas they receive 89% of the burden in the black box approach. The fact that casein, lactose and whey proteins were further loaded with more impacts of downstream processes than cream only decreased the cream's share to 48% in the subdivision approach (Guyomarc'h et al., 2023). At the microfiltration stage, 84% of the carbon footprint went to the casein using economic allocation, leaving only 16% for whey, from which only 0.5% went to lactose (Guyomarc'h et al., 2023). This explains that lactose was virtually burden-free using subdivision prior to economic allocation, while its large mass accounted for 6% of the revenue – and hence of the impacts – when using only a black box approach. Altogether, the results showed that allocation introduced significant variations in the respective environmental footprint of the 5 co-products.

3.3. Hotspots identification: example of the α-lactalbumin enriched ingredient

The potential of α-lactalbumin enriched ingredient for incorporation in infant formulas is the driver for applying industrial membrane separation processes to whey. The environmental impacts of 1 kg ingredient were therefore calculated using a combined subdivision and allocation approach. The impact on climate change was 5–62 kg CO_2 eq/kg α-lactalbumin enriched ingredient, depending on the allocation rule (Table 2). Values are ranging from 1.5 to 17 kg CO_2 eq/kg of dried whey in the Agribalyse or Agri-Footprint databases, or from 1 to 12 CO_2 eq/kg of soy or pea protein isolate, depending on allocation (mass or economic) or geographic scope. Considering the nutritional value attached to α-lactalbumin, comparison at equal nutritional function would require amino acid correction (3–15 kg CO_2 eq/kg). Table 2 and Figs. 4 and 5 further illustrate the sensitivity of the results to the allocation rule. Figs. 4 and 5 depict the contributions of each subparts of the system to total environmental impacts, using respectively the aggregation by processing stage and the aggregation by input type, introduced in the Materials and Methods section 2.5.

In agreement with Fig. 3 for climate change, the environmental impact of 1 kg α-lactalbumin enriched ingredient was about 10-fold less when applying mass allocation (left column; PEF score 0.49 mPt) than dry matter, protein or economic allocation (3 right columns; PEF scores 6.01, 5.09 and 3.61 respectively). The maximum ratio observed was 100-fold for land use, from 94 Pt kg^{-1} for mass allocation to 1700–3100 Pt kg^{-1} for the other rules (Table 2). As seen in Fig. 3 for climate change, mass allocation attributed a larger share of the total impacts to watery (weighty) co-products like lactose, which resulted in smaller absolute values for α-lactalbumin across all impact categories using this rule. Dry matter and protein allocations were the rules that calculated the highest impacts to the product. Finally, in spite of its role as an economic driver, the price of α-lactalbumin is not so higher than that of β-lactoglobulin that it could compensate for the respective masses.

With dry matter, protein or economic allocation rules, milk production and collection contributed from 25 to over 95% of the environmental impacts across the selected impact categories (Fig. 4 or 5). Due to its agricultural origin and large requirements for crops and grasslands, milk production was almost the sole contribution to land use. For this impact category, milk production's contribution was yet 89.5% even using the mass allocation rule compared to other impact categories (<30%) or other allocations rules (>98% - Fig. 4). In other words, the contribution of gate-to-gate processing to land use was less than 1% using dry matter, protein or economic allocations, and reached about 10% with mass allocation only because the milk's impacts (~land use) were mostly allocated to cream, casein and lactose (Table 2). Membrane separations, involving skim milk microfiltration, lactose removal from whey using ultrafiltration, and the separation and washing of α-lactabumin using micro- and ultrafiltration (Fig. 1) contributed for 46–57% to the ionizing radiation impact category (i.e. electricity consumption) and for 33–45% to fossil resources use (Fig. 4). When aggregating the impacts by type of input, processing energies (even without drying) and cleaning (containing the impacts of cleaning water, heating and cleaning agents) were confirmed as the largest contributors to ionizing radiation (54–66%) and consumption of fossil resource (37–46%) during the production of the α-lactalbumin rich ingredient, besides milk itself (Fig. 5 – (Bacenetti et al., 2018; Gésan-Guiziou et al., 2019)). Water involved in whey protein diafiltrations and resolubilization of α-lactalbumin contributed for 29–62% to water use, prior to cleaning (5–10% – Figs. 4 and 5). The chemicals not otherwise involved in cleaning were mostly refrigerants, with significant impacts relative to their mass input

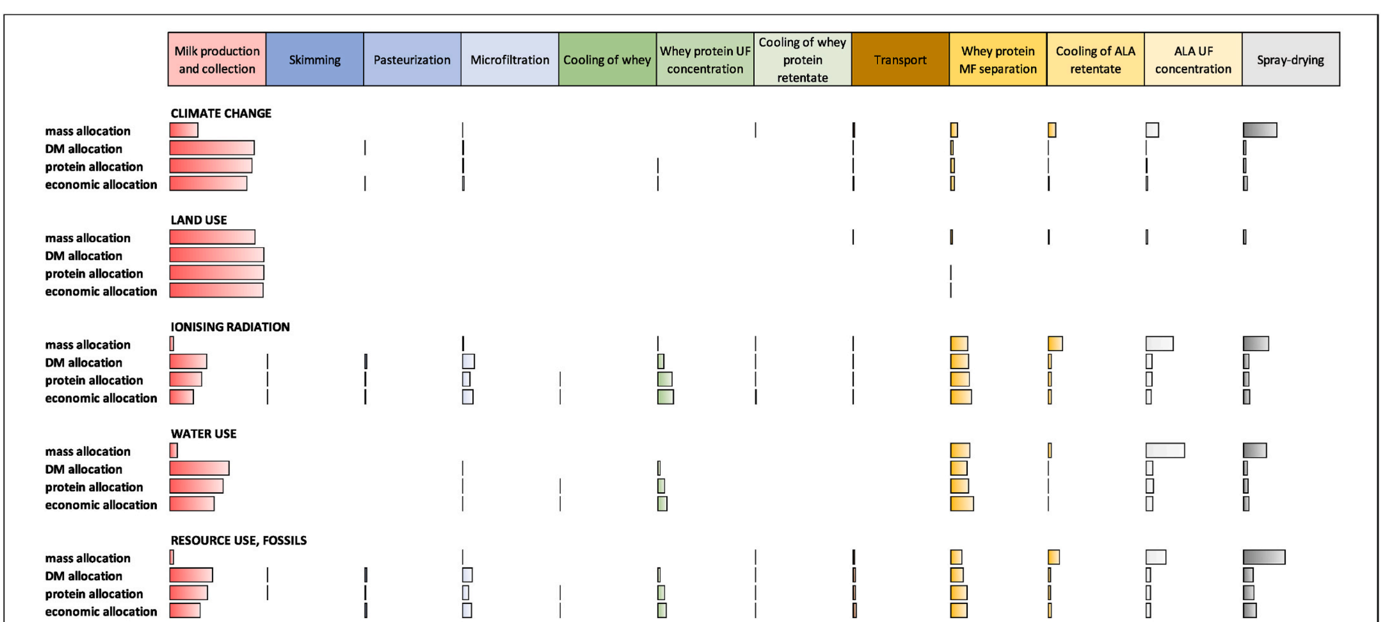

Fig. 4. Contributions of the processing steps to the environmental impacts of 1 kg α-lactalbumin enriched ingredient, for each allocation rule and for the following impact categories: climate change (greenhouse gases emissions, in kg of CO_2 equivalent), land use (marker of agricultural activity, in points), ionizing radiation (marker of electricity consumption, in kBq of uranium 235 equivalent), water deprivation (in m^3) and consumption of fossil resources (in MJ). The impact assessment method was EF 3.0. Abbreviations: ALA = α-lactalbumin; MF = microfiltration; UF = ultrafiltration.

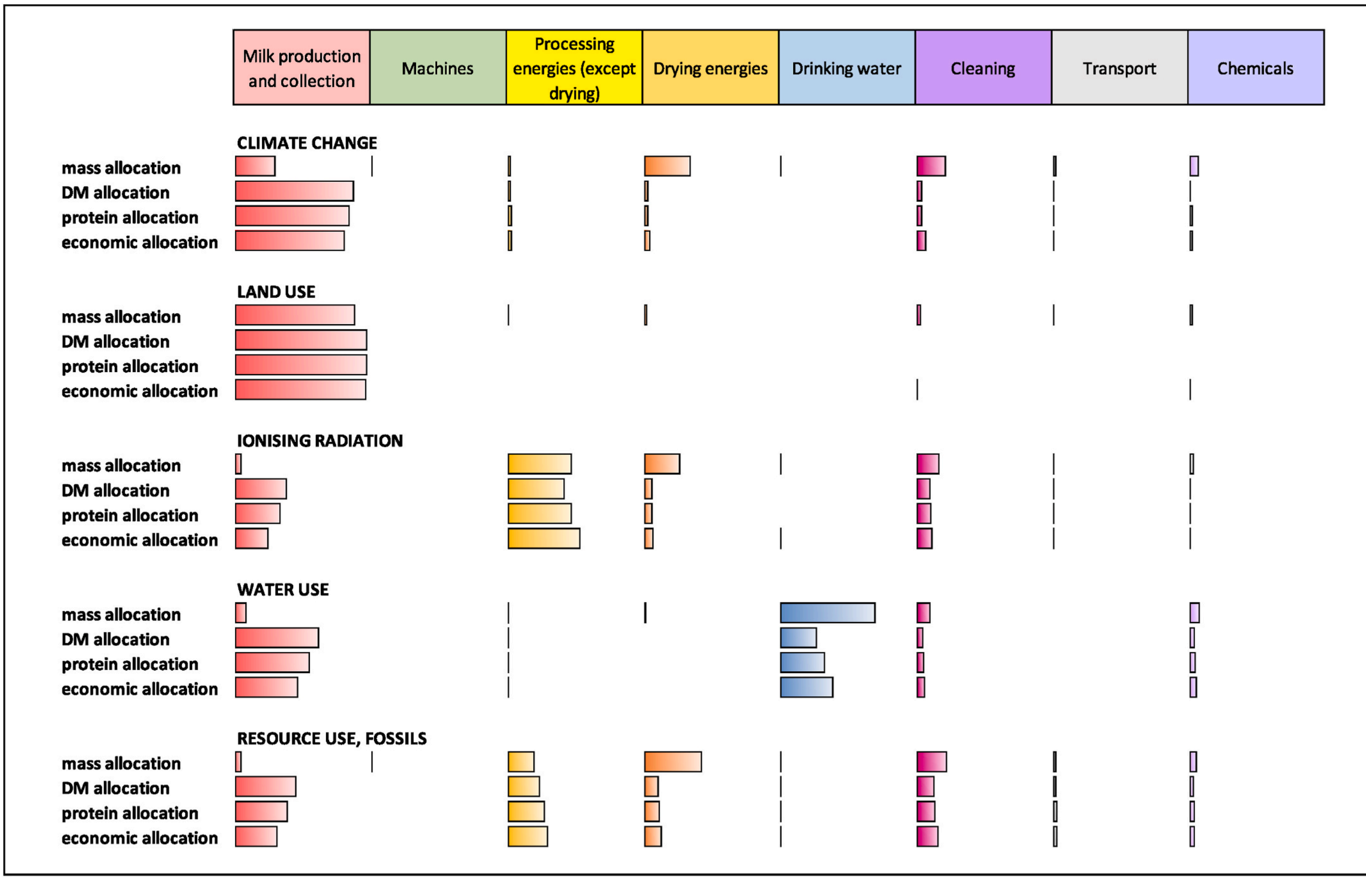

Fig. 5. Contributions of the type of inputs of the environmental impacts of 1 kg α-lactalbumin enriched ingredient, for each allocation rule and for the following impact categories: climate change (greenhouse gases emissions, in kg of CO_2 equivalent), land use (marker of agricultural activity, in points), ionizing radiation (marker of electricity consumption, in kBq of uranium 235 equivalent), water deprivation (in m^3) and consumption of fossil resources (in MJ). The impact assessment method was EF 3.0.

(up to 7% in contribution – Fig. 5).

When using mass as the allocation rule and processing steps as an aggregation rule, attention was called on spray-drying as a hot spot, while both membrane separations and spray-drying were both flagged when using dry matter, protein or economic allocation rules (Fig. 4). Furthermore, it is only when using input as an aggregation rule that cleaning appeared as another hot spot of the system (Fig. 5). These results illustrate that practitioner's choices affect LCA's results and hence, decisions for reducing the system's environmental impacts.

4. Discussion

Attributional LCA is currently used with two major objectives: to assess the environmental footprint of products in order to compare them and to identify hot spots in the system in order to improve the environmental performance or to eco-design processes.

In the first objective, the main issue of LCA is to assess the impacts of individual products without ignoring the fate of co-products in multi-functional systems. In the present report, the large variation in the shares that the 5 co-products received depending on the allocation rule (Fig. 3) illustrated the issue of consistent, and if not, transparent methods to share the environmental burden in such systems (Ekvall and Finnveden, 2001; Dominguez Aldama et al., 2023). Typically, economic allocation is often used to assess agrifood products although physical allocation rules are recommended by ISO (Kyttä et al., 2022; van der Werf and Nguyen, 2015). The presented example shows that even with subdivision, economic allocation attributes only 1.7% of the burden to α-lactalbumin, against 2.4 and 2.8% in protein and dry matter allocations respectively, essentially because the price of whey was low (Fig. 3 and Table 1). As discussed by IDF for the milk sector, economic allocation alleviates a tension between ISO and socio-economic operators, for whom exists a hierarchy between products and "by-products", the latter being neither a product nor a waste (IDF, 2022; Dominguez Aldama et al., 2023). In the case of a multi-functional system, the IDF therefore recommends to first apply allocation "on the economic value of the main products for human consumption and the economic value of the by-products going to animal feed" prior to applying dry matter allocation (IDF, 2022). The issue with this recommendation is that valuable products come virtually burden-free on the market, often at low prices. This could support the emergence of new profitable outlets that question the initial choice calls for actualization of the allocation rule in the entire sector – something that would of course go against the newcomers' interests. Infant formulas, for instance, have considerably increased the economic value of skim milk or whey proteins generated by butter- and cheese-making. Another example is that of lactose, which is used as excipient in pharmaceuticals and could come burden-free when using protein instead of dry matter allocation (Fig. 3). Other examples exist outside the dairy industry with the example of plant proteins, co-products of the starch or oil industries used in animal feed, which are currently drawn towards plant-based food alternatives. This means that they now compete more with micellar casein than with whey. In the same order of thought, choosing or not to process the whey stream would have opened even more options of substitutable systems, from waste management (e.g. biogas production) to fertilization or feed. Comparing the overall impacts of the 5 co-products system with those of a 3-co-product system containing only cream, casein and liquid whey production showed that processing the whey into lactose, α-lactalbumin and β-lactoglobulin enriched ingredients generated less than +2% impacts on climate change, land use and water use, no matter the allocation rule (dataset in (Guyomarc'h et al., 2023)). However, it generated +12–13.6% of impacts on the use of fossil resources and +16.4–17.2% of impacts on ionizing radiations, in coherence with extra uses of gas and electricity. This illustrates how important are system expansion(s), parallel evaluations of the system's co-products and wastes to prevent burden transfer and at least consistent and updated rules not only within but also between food sectors (Wiedemann et al., 2015).

In the second objective, the main issue of LCA is that the life cycle inventory provides as many details as possible to prevent unwanted cut-offs (e.g. on refrigerants – Fig. 5 (De Marco and Iannone, 2017);) and to allow as many subdivisions as possible. In agreement with Aguirre--Villegas et al. (2012) who investigated a shorter processing cascade, subdivision appeared critical for the attribution of the processes' environmental impacts to the sole products that required them. Sensitivities on the allocation and on the aggregation rules are also important to have a clear view of all the possible hot spots. For instance, levers for reduction of the membrane separations' environmental impacts are to be sought on improving cleaning procedures, using renewable electricity and/or questioning purity requirements.

The present comparison of 4 allocation rules furthermore raised the issue of water, which accounts for the difference between mass and dry matter allocations (Fig. 3). IDF's rationale is that dry matter allocation alleviates water from environmental burden, as the total energy required to heat, cool or dry milk (and cleaning water) is affected to the nutrient-rich product (IDF, 2022). On the other hand, water can also be added to the reference flow (e.g. during diafiltration – Fig. 1) and/or issued during the processing cascade (e.g. during reverse osmosis, evaporation and drying). Comparing the results of mass and dry matter allocations draws attention to the fate of water, which is important now that most countries have authorized or are about to authorize food and drink uses of "co-produced" water. It also helps evaluate the nutritional interest of the product, along with discussion on the functional unit, in conditions where a product "diluted" with water or other burden-discounted ingredients (oil, sugar) is likely to present a lower environmental footprint than a product concentrated in milk nutrients.

With increasing end-of-life options of the co-produced water (reuse in the food chain) and of nitrogen and sugar contained in whey (e.g. towards food-grade valorization or towards energy and fertilization), system expansion appears as an increasingly attractive strategy for LCA of multi-functional systems in a context where circular economy and sobriety are priorities. However, it requires even more data as a greater number of substitutable products needs to be identified and documented. In agri-food systems, such products in may themselves be co-products of multi-functional systems, creating competition for outlets (e.g. between feed sources) and a risk of oversupply. Assessment and eco-design of multi-functional systems therefore raise attention on the integrative aspect of processing. LCA should help designing industrial processes with virtuous compromises between in-door or local recycling or exchanges of resources (sobriety) and extra expenses for upgrading co-products (valorization). Both strategies require substantial effort in food technology, process engineering and industrial ecology.

5. Conclusion

Consistent and transparent methodological choices in LCA of multi-product processes are critical both for comparison between food products' environmental footprints and improvement of the system's environmental performance. Subdivision was found necessary to avoid irrelevant allocation of impacts to upstream products, and sensitivities on the allocation and the aggregation rules help to identify and confirm hot spots. The method through which environmental impacts – and possible environmental credits – are distributed between co-products reveals possible strategies for substitution and/or valorization. This study contributes to the dissemination of LCA in complex food system for eco-design purposes, by drawing attention to the fate of co-products as part of the environmental impact assessment of any of the system's product, by making the fight against food waste as a priority knowing the contribution of farming and by questioning the balance between the impacts and value of downstream recovery processes.

CRediT authorship contribution statement

Fanny Guyomarc'h: Writing – original draft, Methodology, Investigation, Data curation, Conceptualization. **Félicie Héquet:** Methodology, Investigation, Formal analysis, Conceptualization. **Samuel Le Féon:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Nadine Leconte:** Writing – review & editing, Resources, Investigation, Conceptualization. **Fabienne Garnier-Lambrouin:** Writing – review & editing, Resources, Investigation, Conceptualization. **Julie Auberger:** Writing – review & editing, Software, Conceptualization. **Caroline Malnoë:** Writing – review & editing, Software, Conceptualization. **Caroline Pénicaud:** Writing – review & editing, Funding acquisition, Conceptualization. **Geneviève Gésan-Guiziou:** Writing – review & editing, Supervision, Resources, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Gesan-Guiziou reports financial support was provided by French National Research Agency. Penicaud reports financial support was provided by French National Research Agency. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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