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Comparative life cycle assessment of five Greek yogurt production systems: A perspective beyond the plant boundaries

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Abstract: Greek yogurt (GY), a high-protein-low-fat dairy product, particularly prized for its sensory and nutritional benefits, revolutionized the North American yogurt market in less than a decade, bringing with it new sustainability challenges. The standard production of GY generates large volumes of acid whey, a co-product that is a potential source of environmental pollution if not recovered. This study aims to assess the environmental performance of different technologies and identify the main factors for improving GY production. A complete life cycle assessment (LCA) was performed to compare the standard technology (centrifugation) with two new technologies (fortification and ultrafiltration) to reduce acid whey volumes. Three milk protein concentrate alternatives were also assessed. Results show that the technology choice is not a clear discriminant factor. However, minimizing losses and wastage (accounting for 23 to 25% of the environmental impacts for all indicators) beyond the processing plant and selecting milk ingredients (accounting for 63 to 67% of the impacts) with low environmental impacts are key factors in improving the environmental performance of GY systems. From a methodological perspective, the results also highlight a shortcoming in the current International Dairy Federation LCA guidelines (2015) for treating the multifunctionality of GY systems.

Keywords: LCA; Greek-style yogurt processing; environmental impacts; losses and wastage; multifunctionality; allocation

1. Introduction

A global transformation of the food system is urgently needed to support environmental sustainability and nurture a growing population. As stated by the EAT-lancet commission, this transformation must combine a shift towards healthy diets, improving food processes and food production efficiency, and at least halve food losses and waste, to keep the humanity in a safe operating space [1]. Meat and dairy products are widely pointed out as a large source of environmental and/or health burdens [2] but this statement should be nuanced among products. For instance, some would qualify yogurt, as a sustainable food for its health benefits and low environmental impact compared to some other animal source food. Indeed, the production of a regular yogurt emitted 75 to 55% less GHG emissions than cheese and is a good source of calcium and protein but with a lot less [3]. Around the world, yogurts come in various types and compositions [4]. Greek yogurt (GY), also known as Greek-style yogurt, has flooded the North American market. In the past decade, it grew from a market share of 1% to around 40% in the U.S. [5] and 20% in Canada (Nielsen, personal communication, 2017). This spectacular success can likely be attributed to its nutritional and sensory properties. Its high-protein content provides a creamy and thick texture [6] and positioned it as a healthy food product that provides consumers with both pleasure and nutritional benefits [7]. However, in a perspective of sustainable diet, the environmental impacts of GY should also be considered. GY is recognized as a source of incremental environmental impacts in the dairy sector. Indeed, its high protein content is traditionally obtained by concentrating fermented milk through centrifugation to increase the yogurt's protein content from 3.27% over 10% w/w. This operation requires up to three times more milk input than regular yogurt and generates at least twice the weight of GY as acid whey [8,9]. Acid whey may be harmful to ecosystems if not recovered or disposed of correctly and its increasing volumes generate operational challenges for dairy manufacturers [10]. Significant efforts have been made to recover acid whey components. The American Chemical Society registered 3 500 patents in 2017, with most of them focusing on extracting proteins and lactose from GY acid whey using membrane-based filtration processes [11-14] or on enzyme-based approaches [15], but most of these processes are not yet economically viable [5]. Therefore, instead of focusing on whey recovery, recent works have been oriented on processing innovation to reduce or eliminate the production of acid whey. The various technology alternatives explored are extensively described in Jørgensen et al. [7] and summarized in the supplementary materials of this paper (S1). These technologies influence the volume and composition of co-product generated, as well as the GY composition and its sensorial properties [6,16,17]. They also varies in term of production yield, resources, utilities consumption such as energy, water use, chemicals at the manufacturing plant and the capital cost of the processing equipment [7,17,18]. The technical advantages and drawbacks of each technological option are well documented but, to the best of our knowledge, not any study has included a systematic environmental comparison between GY production systems and technologies.

Few life cycle assessment (LCA) studies published in the scientific literature estimated the environmental impacts of regular yogurt manufacturing process [3,19–21] and yogurt packaging and delivery systems [22,23] but non of them focus specifically on Greek yogurt technologies. The first aim of this paper is to fill this gap, comparing the environmental performance of the most common GY production options available in the province of Québec (Canada) in 2018 throughout the entire product life cycle from a cradle-to-grave perspective. This study intends also to identify the main contributors of GY environmental footprint along its life cycle and the key triggers to reduce its environmental burden with a special focus on losses and wastage. The main findings could guide GY manufacturer actions and priorities to improve the environmental performance of this healthy food product. Conjointly, from a LCA methodological perspective, this paper aims to assess the relevance of the mass allocation method based on the dry weight of milk solid content, recommended in the new LCA guidelines of the International Dairy Federation [24], for partitioning the GY environmental impacts from its co-products (cream and whey) and comparing different GY systems consistently.

Section 2 describes the GY technological options and the LCA methodology. Results are presented in Section 3. Section 4 discusses the key findings and determines the limitations of the current study and opportunities for further research. Conclusions follow in Section 5.

2. Materials and Methods

The LCA methodology was based on the International Dairy Federation guideline "A common carbon footprint approach for the dairy sector" [24] and the international ISO 14 040 [25] and 14 044 [26] standards. The attributional LCA compares five scenarios representative of the production options available in the province of Québec (Canada) in 2018, as described after. The scenarios are compared based on a functional unit of 1 kg of GY at 10% protein and 0% fat consumed by an average Québec household in 2018. Despite slight organoleptic and composition differences related to the manufacturing technology, the functional properties of the GY compared in this study were assumed to be equivalent for the five systems.

2.1. Compared technological options

Based on a survey conduct among three major Canadian GY manufacturers in 2017 and 2018, we selected three GY different technologies to manufacture the GY.

- Centrifugation (CE) is the most conventional technology. It concentrates the yogurt proteins
 after the fermentation process stage.
- **Fortification (FO),** consists of adding protein ingredients such as milk protein concentrate (MPC) in the milk before fermentation, which reduces the quantity of acid whey generated

Ultrafiltration (UF) before fermentation, is a new option that is gaining ground. This option
has the benefit of generating neutral pH milk permeate (instead of acid whey), which has a good
potential of valorization on the food ingredients market.

Three ingredient supply alternatives were also considered for FO, resulting in five GY production system scenarios. FO reference scenario was based on solid MPC 80 powder concentrated at 80% proteins (w/w) sourced from the USA (see description of the MPC manufacturing process in S2.1). Two alternatives to liquid diafiltered milk from the USA or Québec were also assessed. The three MPC supply alternative scenarios are named:

- FO-P-US for MPC powder (80% protein) from USA
- FO-L-US for liquid MPC (20% protein) from USA
- FO-L-QC for liquid MPC (20% protein) from Québec

2.2. Product systems description and boundaries

The scope of the study is cradle-to-grave. It considers all the operations, from feed production to milk production at the dairy farm, yogurt factory operations, distribution, consumption and final disposal of the product. The transportation and waste management operations in all stages are also considered. All known exchanges from/to the ecosphere are included in the life cycle inventory. System boundaries are described in Figure 1.

INPUTS FROM ECOSPHERE UF FO CE Subsystem 1 Subsystem 2 Milk Reception & storage T:4°C production Quebec **MPC 80 CREAM** Skimming T:55°C Milk Cleaning Skimming production agents USA Rehydratation Pasteurisation Ultrafiltration Secondary packaging Ultrafiltration Thermal treatment Primary Diafiltration packaging Fermentation Drying Electricity WHEY Centrifugation Packaging Natural gas Cooling Filling, packaging, cooling tunnel, storage ന **PACKAGED** Subsystem 3 Subsystem 4 Subsystem 5 **GREEK YOGURT** Packaging Distribution Household disposal WASTE TREATMENT FU: 1 kg of System boundaries **EMISSIONS TO ECOSPHERE** Greek yogurt Transport consumed

Figure 1. System boundaries including detailed unit process and main parameters of operations for the three technologies: Centrifugation (CE) in blue, Fortification (FO) in orange and Ultrafiltration (UF) in green.

The milk is produced in a typical Québec farm and transported to the GY manufacturing plant where it is transformed in GY (Figure 1).

CE: The raw milk is skimmed, thermally treated, fermented, centrifugated and cooled before reaching the packaging area. The centrifugation process separates the acid whey from the yogurt to concentrate the proteins up to 10%.

FO: Liquid or solid milk protein concentrates (MPC) are introduced between the skimming and thermal treatment operations. Other operations remain similar to CE.

UF: The protein concentration is performed just after the skimming. Consequently, smaller volumes of skimmed milk are treated during subsequent operations. The UF membrane retains the majority of the proteins in the skimmed milk (retentate). Most of the lactose and minerals migrate through the membrane into the permeate. The detailed unit processes of the three technological options are described in S2.1.

Then, regardless of the technological option, 50% of GY is filled into 500 ml polypropylene (PP) containers and the other 50% into 100 ml polystyrene (PS) cups packed by 4 or 8 units. Packaged GY is then boxed, palletized and cooled at 4°C to be stored. The distribution and consumption stages include several transportation and refrigeration operations, as described by [20]. In total, 79% of the cardboard used for PS packs and 15% of PP containers are recycled by consumers [27]. The rest of the materials are landfilled.

2.3. Systems modeling approach

We modeled the three technological options in the prototype of a dairy process simulation software [28] based on the layouts and processing parameters (temperature, pH, pressure) provided by the GY manufacturers. The simulation calculated the mass balance and input and output flows per process unit. Detailed information on processing simulation parameters and input/output flow results are available in S3 (tables S1 to S5). Results from the simulation were subsequently validated with the GY manufacturers to improve the level of confidence of the study.

2.3.1. Mass balance per unit process and technology

The mass balance of main process units reported in Table 1 were based on 1 kg GY output with identical protein concentration (10%). In practice, GY outputs and concentration vary according to the technology. The high selectivity of the UF membrane retains more proteins and less lactose than the traditional CE. FO usually has a higher protein and lower lactose content than CE because of the high protein/lactose ratio contained in MPC 80 [8,29].

Each technology was characterized by a specific protein retention coefficient (Rp) (equation 1).

$$Rp = 1 - [P_w] / [P_{gy}]$$
 (1)

Where $[P_{gy}]$ = protein concentration in GY and $[P_w]$ = protein concentration in the whey.

The simulation was performed with Rp =0.97±0.01 for CE, Rp=0.95±0.01 for FO and Rp=0.98±0.01 for UF based on manufacturer data. Output and inputs were scaled-up to 1 kg of GY produced, resulting in variations of raw milk input and cream and whey outputs according to the technology (Table 1).

Table 1. Simulated mass balance of inputs and outputs for the CE, FO, and UF technologies before L&W

	CONCENTRATION ON WET BASIS							QUANTITY PER KG OF GY PRODUCED						
	Protein	Fat	Lactose	Ash	Dry	Water	Protein	Fat	Lactose	Ash	Water	Total		
					Matter									
	(w/w)	(w/w)	(w/w)	(w/w)	(w/w)	(w/w)	kg	kg	kg	kg	kg	kg		
				(Centrifuga	ation (CE)							
Raw milk	3.27	3.97	4.81	0.75	12.80	87.20	0.113	0.138	0.167	0.026	3.026	3.47		
Skimmed milk	3.40	0.04	5.01	0.78	9.23	90.77	0.106	0.001	0.157	0.024	2.840	3.13		
Cream	2.04	40.00	3.01	0.47	45.52	54.48	0.007	0.137	0.010	0.002	0.186	0.34		
Concentrated yogurt	10.00	0.04	4.66	0.73	15.43	84.57	0.100	0.000	0.047	0.007	0.846	1.00		
Acid whey	0.30	0.04	5.17	0.81	6.32	93.68	0.006	0.001	0.110	0.017	1.994	2.13		
Fortification (FO)														
Raw milk	3.27	3.97	4.81	0.75	12.80	87.20	0.089	0.108	0.131	0.020	2.381	2.73		
Skimmed milk	3.40	0.04	5.01	0.78	9.23	90.77	0.084	0.001	0.123	0.019	2.235	2.46		
Cream	2.04	40.00	3.01	0.47	45.52	54.48	0.005	0.107	0.008	0.001	0.146	0.27		
MPC 80	81.30	1.60	4.60	6.80	94.30	5.70	0.024	0.000	0.001	0.002	0.002	0.03		
Fortified milk	4.20	0.06	4.87	0.83	9.96	90.04	0.107	0.002	0.125	0.021	2.304	2.56		
Concentrated yogurt	10.00	0.06	4.56	0.78	15.40	84.60	0.100	0.001	0.046	0.008	0.846	1.00		
Acid whey	0.48	0.06	5.07	0.86	6.47	93.53	0.007	0.001	0.079	0.013	1.458	1.56		
				1	Ultrafiltra	tion (UF))							
Raw milk	3.27	3.97	4.81	0.75	12.80	87.20	0.111	0.134	0.163	0.025	2.949	3.38		
Skimmed milk	3.40	0.04	5.01	0.78	9.23	90.77	0.104	0.001	0.153	0.024	2.768	3.05		
Cream	2.04	40.00	3.01	0.47	45.52	54.48	0.007	0.133	0.010	0.002	0.181	0.33		
Concentrated yogurt	10.00	0.12	4.65	0.73	15.50	84.50	0.100	0.001	0.047	0.007	0.845	1.00		
Soft whey (permeate)	0.18	0.00	5.18	0.81	6.17	93.83	0.004	0.000	0.106	0.017	1.919	2.04		

2.3.2. Multifunctionality modeling approach

GY systems are multifunctional because they produce jointly GY, cream and whey and thus fulfill several functions. Yogurt is the primary function. The cream is a high-value co-product. The acid whey from CE or FO process is a low-value co-product recovered as an animal feed complement in pig farming but the sweet whey (or permeate) from the UF process could be recovered for higher value application in the food ingredient market. In the Québec context, the transport cost of acid whey to the pig farms is assumed by the GY manufacturer and the pig farmer gets the whey for free. Therefore, the acid whey can be considered as a waste with no economic value whereas the sweet whey from UF has an economic value. This difference in the whey co-product functionality set a challenge on how to allocate the impacts between the product and co-products. As a reference basis, we used IDF guidelines [24] to allocate the impacts between the three products generated by the systems. These guidelines recommend dividing the system into sub-systems when possible and using a mass allocation based on the dry weight of milk solid content of the final product and co-products when sub-division is not possible. The burdens from raw milk and manufacturing processes were then allocated between cream, GY and whey according to equation (2):

$$MAF_i = DM_i \cdot Q_i / \sum_{i=1}^{n} (DM_i \cdot Q_i)$$
 (2)

Where MAF $_i$ is the mass allocation on dry matter factor for the product i; DM $_i$ is the dry matter content of product i (Table 1 column Dry Matter) and Q $_i$ is the quantity of product i for 1 kg GY (Table 1 column Total).

We also tested the robustness of the results by allocating the impacts based on an economic relationship between the co-products. This allocation was calculated on the milk producers' revenues for milk components class 2a price average in Québec for GY and cream and class 7 milk component prices average for the whey in 2017. Whey received an economic allocation factor 0% or 17.5 % depending on its potential value on the market.

- **Scenario 1:** 0% economic allocation is built on the substitution of other feed intakes in a pig farm. In this case, the whey is a cost for the GY manufacturer who pays its transport to the pig farm.
- **Scenario 2:** 17.5 % is built on the valorization of the whey components in the food industry for the UF process only, based on class 7 prices.

The mass and economic (scenario 1) allocation factors are described in Table 2.

MPC manufacturing is also a multifunctional process that uses raw milk to produce cream, MPC and a UF permeate rich in valuable lactose. Economic allocation factors vary depending on the sourcing since the prices of cream and milk ingredients are not the same in the USA and Canada. We used prices according to milk components USA class IV (Proteins: 3.98 USD.kg-1; Fat 5.35 USD.kg-1; Lactose 0.12 USD.kg-1) and Québec class 7 (Proteins: 1.58 CAD.kg-1; Fat 7.24 CAD.kg-1; Lactose 1.58 CAD.kg-1) in 2017. These allocations factors are available in S5 (Table 7).

Table 2. GY production system - mass and economic allocation factors at each point of substitution

SB = subsystem	Mass	allocation	on dry ma	tter	Economic allocation(*)								
		Skimmed			Skimmed								
	Cream	milk	Whey	GY	Cream	milk	Whey	GY					
From farm to plant (SB1) Raw milk production, transportation, losses and wastage													
CE	35%	_	30%	35%	57%	_	0%	43%					
FO	35%	_	29%	36%	57%		0%	43%					
UF	35%	_	29%	36%	57%		0%	43%					
From reception to	From reception to skimming (SB2) Reception and storage, skimming												
CE	35%	65%	_	_	57%	43%	_	_					
FO	35%	65%	_	_	57%	43%	_	_					
UF	35%	65%	_	_	57%	43%	_	_					
Ultrafiltration (SE	32)												
UF	_	_	45%	55%	_	_	0%	100%					
From skimming to centrifugation (SB2) MPC supply, rehydration and mixing (FO only); thermal treatment, (homogenization - optional), fermentation, centrifugation													
CE	_	_	47%	53%	_	_	0%	100%					
FO	_	_	45%	55%	_	_	0%	100%					
UF	_	_	0%	100%	_	_	0%	100%					

^(*) economic allocation is presented only for scenario 1

Table 2. GY production system – mass and economic allocation factors at each point of substitution (count'd and end).

SB = subsystem	Mass	allocation	on dry ma	tter	Economic allocation(*)						
		Skimmed			Skimmed						
	Cream	milk	Whey	GY	Cream	milk	Whey	GY			
General plant operations (SB2) CIP, lighting and conditioned air electricity											
CE	35%	_	30%	35%	57%	_	0%	43%			
FO	35%	_	29%	36%	57%	_	0%	43%			
UF	35%	_	29%	36%	57%	_	0%	43%			
From cooling to final disposal (SB2/SB3/SB4/SB5) SB2: Cooling, filling, packing, storage, packaging, plant wastes: SB3: transport & distribution: SB4: transport and consumption: SB5: final disposal: GV losses and											

wastes; SB3: transport & distribution; SB4: transport and consumption; SB5: final disposal; GY losses and wastage

CE	_	_	_	100%	_	_	_	100%
FO	_	_	_	100%	_	_	_	100%
UF	_	_	_	100%	_	_	_	100%

^(*) economic allocation is presented only for scenario 1

2.3.3. Life cycle inventory (LCI)

Background life cycle inventory data, including milk production, is based on the Québec inventory database [30] as available from the version 3.4 (released in 2017) of the ecoinvent database, cut-off system model (ecoinvent center https://www.ecoinvent.org/; [31]), and targeted USA datasets were extracted from Thoma et al. [32]. Few adjustments to pesticides and water flows were made to ensure the consistency of crop datasets between the USA and Québec databases. Infrastructures and processing equipment were excluded from the analyses.

Foreground data from the process simulation were scaled up to the functional unit and summarized in Table A1 in Appendix A. Process parameters were discussed and validated with industrial partners. We collected the remaining data from the literature and tested the few assumptions with sensitivity analyses. Based on previous studies [20,33,34], we estimated electricity consumption related to packaging, refrigeration and plant ventilation and lighting to be respectively 12%, 18% and 19% for a total of 50% of the plant's total electricity consumption. We also assumed these consumptions to be identical for all options. Water flows resulting from the simulation were corrected with González-García [20] inventory to include plant general water and improve result completeness. Data from distribution and household consumption were based on González-García [20] and adapted to the Canadian context. Refrigerant leakages were excluded, except for chilled trucks. Thoma [35] assessed them at 1% of the GHG impacts at the plant and 19% at distribution but these figures tend to drop as technology evolves. We modeled packaging raw materials recycling based on the cut-off approach using current recycling rates in Québec as recycled content rates. Milk and GY L&W through the value chain, including packaging and all related operations (processing, storage, refrigeration and transportation), except disposal, were assessed at 3.5% during milk production and transportation, 3% at the processing plant and 26% in between distribution and consumption based on a literature overview described in S6 (Table S8). SimaPpro v8.5.2.2 software [36] was used to model the LCI. S4 (Table S6) provides key parameters and the sources used to build each operation.

2.3. Life cycle impact assessment

Impacts are estimated from the LCI using the IMPACT WORLD+ method [37], Endpoint v1.4.1, Midpoint v1.23. Four impact category indicators were selected to compare the GY system options providing a high level of aggregation of environmental concerns relevant for the Québec's dairy sector.

- **Human health** (HH) in DALY, encompassing impact on water availability, human toxicity, particulate matter formation,...
- **Ecosystem quality** (EQ) in pdf·m²-yr encompassing impacts on freshwater eutrophication, land occupation, land transformation,...
- Climate change (CC) in kg CO₂ eq.
- Fossil and nuclear energy use (FEU) in MJ deprived

Climate change are measured at short term, representing a time integrated impact over a time horizon of 100 years. Climate change contribution to HH and EQ endpoint indicators was removed purposely. For the raw milk data from the USA imported from an older version of ecoinvent (v2.2), water supply processes were adapted to ensure the water mass balance. However, the water balance is not consistently ensured for the background processes across ecoinvent v2.2, therefore, the representativity of water availability impact assessment in HH and EQ categories should be taken with care.

3. Results

3.1. Contribution to the environmental impacts

Results presented in Figure 2 are expressed in absolute value. Detailed numerical results are available in S7.1 and 2. The cradle-to-grave perspective reveals the significant contributions of milk production, milk ingredients (MPC) and product L&W to the GY impacts. Packaging materials are also important contributors when it comes to non-renewable energy use.

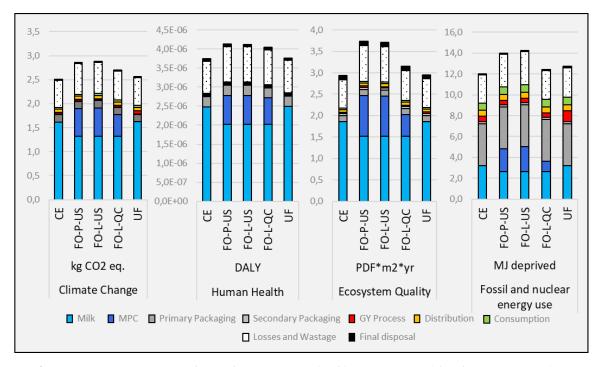


Figure 2: Comparative LCA of centrifugation (CE), ultrafiltration (UF) and fortification (FO) with 3 MPC sourcing alternatives (FO-P-US: fortification with MPC 80 powder from USA; FO-L-US: fortification with liquid MPC from USA; FO-L-Qc: fortification with liquid MPC from Québec) for a functional unit (FU) of 1 kg yogurt consumed including L&W.

The production of milk at the farm (including milk ingredients for FO) contributes to 82 to 88% of the climate change (CC), human health (HH) and ecosystem quality (EQ) impacts and to 33 to 43% of the fossil energy use (FEU) impacts, for all the scenarios before L&W. This is consistent with the IDF report (2009)[38], which states milk production contribution to 80 to 95% of the total LCA GHG emissions based on 60 dairy products studies. When L&W is included, the main contributors remain

milk and dairy ingredients, which contribute to 63 to 67% of CC, HH and EQ impacts and 25 to 35% of the FEU impacts.

Losses and wastage (L&W) contribute to 23 to 25% of the life cycle impacts for all impact indicators. Potential impacts from L&W are notably higher than the cumulative impacts of the processing, distribution, consumption and final disposal stages.

Primary packaging, mainly PP and PS containers, contributes to 6 to 8% of CC impacts and 35% of the FEU category (Figure 2). For half of the GY volumes packaged in PP and half in PS, the latter requires 24% more raw material (Table A1) and has respectively 25% and 50% more impacts on FEU and CC than the former. The contribution of packaging to CC (6%) remains low as compared to the contribution of milk and milk ingredients (> 63%) or to that of L&W (>24%).

Processing operations at plant contribute to less than 3% of CC, HH, EQ and between 3 to 8% of FEU impacts over the entire life cycle for all the scenarios. Operations requiring the use of natural gas (heating treatment and CIP) stand out in the FEU category. They contribute to 94 to 98% of the processing stage impacts. More details on the contribution of each processing operation are available in S7.1 (Table S10). These results are specific to Québec, where renewable energy sources (mostly hydropower) represent 99.5% of the electricity mix [39]. The impacts related to processing operations would increase in regions where fossil thermal power plants are the primary source of electricity. For instance, with the energy mix average of USA– 35% natural gas; 27% coal; 19% nuclear; 17% renewable [40] – the impacts related to CC would only rise up to 2%. Therefore, the contribution of the processing stage to the entire GY life cycle impacts remains low in any case.

Distribution and consumption: These two stages include mainly electricity consumption for refrigeration and fuel consumption for transportation. The sum of all these operations accounts for 4% of CC impacts and 10% of FEU impacts over the entire life cycle (Figure 2) in this study but may vary significantly according to transportation distances. For instance, contribution to CC impacts vary from 1 to 18% over the entire life cycle (1% for transport from the grocery to home on foot and distribution distances < 50 km; 18% being for 20 km from the grocery by car and distribution distance equal to 600 km).

Final disposal counts for less than 1% of CC, 2% of HH, 3.5% of EQ and 1% of FEU impacts over the entire GY life cycle.

3.2. LCA scenarios comparison

The relative discrepancy between the highest and lowest environmental impact scores among scenarios varies from 0 to 20% across indicators. The two fortification scenarios sourced in the USA have the highest impacts for all four indicators. CE has the lowest impact. The discrepancies for CE, UF and FO-L-QC are less than 8% for each indicator.

3.2.1. Protein yield and raw milk input

Seeing as the major contributor to the environmental impacts is milk production (Figure 2), the technologies that consume less milk to achieve 10% protein concentration in GY were expected to perform better. So, the higher the protein yield performance, the more GY is produced with the same quantity of milk and the more the impacts correlated to GY production might decrease. As reported in Table 1, FO is the most intensive technology in terms of milk input (3.51 per kg of GY produced including 2.73 kg of QC raw milk and 0.78 kg of milk for MPC), followed by CE (3.47 kg) and UF (3.38 kg). FO has a low protein retention rate in GY during the centrifugation process (Rp=0.95). The performance of UF is attributed to the good selectivity of the membrane, which retains more protein in the GY (Rp=0.98) than CE separator (Rp=0.97). Nevertheless, LCA results are not only correlated with protein yield. Several other factors are involved, such as the influence of the milk producing region and the type of MPC (liquid or powder) used. These key factors are discussed below, other less influential factors are also detailed in S7.3.

3.2.2. Influence of the MPC milk producing region and type

Influence of the milk producing region (Figure 3): The discrepancies between the three MPC sourcing alternatives (*FO-S-US, FO-L-US and FO-L-QC*) for the FO process (Figure 3) are due to a combination of three main factors: milk producing region, MPC drying process and transportation distances.

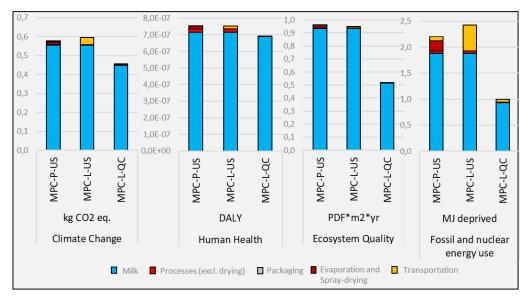


Figure 3: Impact profile for the production and transportation of three MPC sourcing alternatives with quantities scaled up to fulfill the functional unit (1 kg of GY). MPC-P-US: 0.029 kg of MPC 80 powder from the USA transported over 1500 km; MPC-L-US: 0.12 kg of liquid MPC from the USA transported over 1500 km; MPC-L-Qc: 0.12 kg of liquid MPC from Québec transported over 180 km.

Milk production contributes to 77 to 99% of the MPC impacts for all the FO scenarios and impact categories. Milk produced in the USA (national average) has a greater impact than milk produced in Québec by 19% for CC, 4% for HH, 45% for EQ and 56% for FEU. Differences in farming system parameters such as feed intakes, crop production practices, irrigation requirement, manure management and regional climate conditions, cause most of these variations [41]. On average, the USA's agricultural practices are more intensive than Canada's. The USA uses more fertilizer, requires more fossil energy and relies on more maize for cow feeding (a crop with a high environmental footprint) than Canada [42]. Furthermore, a sensitivity analysis based on data collected by Thoma et al. [32] on American farms reveals significant variations between USA regions (S7.1 Table S11).

Influence of the MPC drying process and transportation distances (Figure 4): For CC and FEU impact categories, the supply of 1 kg of MPC powder (MPC-P-US) transported over 1500 km has respectively only 4 and 9% less impacts than the supply of 4 kg of liquid MPC (MPC-L-US). By reducing the distance of transportation to 750 km or less, the supply of 1 kg of MPC powder (MPC-P-US) has more impacts than the supply of 4 kg of liquid MPC across all categories. As already demonstrated by Depping et al. (2017), transporting large volumes of liquid proteins over long distances often has fewer impacts than drying operations. As shown in Figure 4 and detailed in S8.2, the region of milk sourcing and the type of MPC (powder versus liquid) are much more sensitive parameters than the distances of transportation.

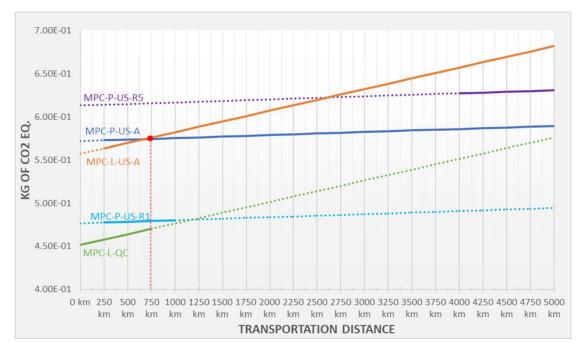


Figure 4: CC impacts variation as a function of transportation distance from MPC plant to GY plant for the three MPC sourcing alternatives scaled-up to the FU (1 kg of GY): MPC-P-US-A: 0.03 kg MPC 80 powder from USA with USA raw milk average; 0.12 kg MPC-L-US-A: liquid MPC from USA with USA raw milk average; 0.03 kg MPC-P-US-R1: MPC 80 powder from north east USA; 0.03 kg MPC-P-US-R5 MPC 80 powder from west coast USA; ; 0.12 kg MPC-L-Qc: liquid MPC from Québec.

3.3. Results sensitivity

The different sensitivity analyses performed on the modeling parameters or the modeling and methodological choices (detailed in S8.1 to S8.4) show that the results are mainly influenced by the quantity and the origin of supplied raw milk or MPC, the allocations rules and factors, and the L&W rates.

Economic versus mass allocation on dry matter: As summarized in Table 3, conclusions change with respect to the allocation rule (mass versus economic) and the allocation factor (value attributed to the whey). With economic allocation (scenario 1), the whey is considered a waste and is attributed 0% of the impacts. With mass allocation on dry matter, the whey (or permeate) is attributed 29% to 30% of the raw milk impacts for UF and CE, respectively. This difference changes the impacts allocated to GY and modifies the conclusions between scenarios. Indeed, with economic allocation, the higher protein retention coefficient of UF gives UF an advantage as compared to CE by slightly reducing the mass of raw milk required at the input (3.47 kg/kg GY for CE and 3.38 kg/kg GY for UF) to produce 1 kg of GY. However, with mass allocation on dry matter, this advantage is offset by the higher dry matter retention rate of UF versus CE (Table 1), which increases the impacts allocated to GY processed by UF versus CE (Table 2).

If the milk components from the UF permeate is recovered (scenario 2 of the economic allocation) instead of being treated as waste like acid whey (scenario 1 of the economic allocation), UF performs even better than CE and FO. Indeed, in this situation, only 82.5% of the UF system impacts are allocated to the GY whereas GY made from CE and FO systems still received 100% of the impacts.

Region of milk sourcing: Conclusions are also sensitive to the MPC milk sourcing region (Table 3 and S7.4). For instance, if MPCs are produced with US milk sourced from the state of New York (R1), FO-L-US becomes more interesting than FO-P-US in all impact categories. In contrast, if MPCs are produced with US milk sourced from California (R5), FO-P-US becomes more interesting than FO-L-US. FO-L-QC performs better than the FO-US scenarios but CC impact results show low gaps (0.2%) with milk production regions such as R1 and high gaps (14%) with milk production regions such as R5 (Figure 4 and S8.4 Table S15). These findings reinforce the fact that the region from which the milk and milk ingredients are sourced has a major influence on GY environmental performance.

Table 3. Change in scenarios classification according to sensitivity analyses (more details in S8.4); R1: north east; R5: west coast

OBJECT	MODIFICATI ON	IMPACT CATEGOR Y	CONCLUSI ON VS. REFERENCE	LCA RESULTS	GENERAL CLASSIFICA TION	
		CC		CE <uf<fo-l-qc <fo-p-us<fo-l-us< td=""><td></td></fo-p-us<fo-l-us<></uf<fo-l-qc 		
D (27.4	НН	N .	CE <uf<fo-l-qc <fo-l-us<fo-p-us< td=""><td>CE<uf<fo-l- QC</uf<fo-l- </td></fo-l-us<fo-p-us<></uf<fo-l-qc 	CE <uf<fo-l- QC</uf<fo-l- 	
Reference	NA	EO	CE <uf<fo-l-qc <fo-l-us <fo-p-us<="" td=""><td>except for FEU FO alternatives</td></fo-l-us></uf<fo-l-qc 	except for FEU FO alternatives		
		FEU		CE <fo-l-qc <uf<br=""><fo-p-us <fo-l-us<="" td=""><td>variable</td></fo-p-us></fo-l-qc>	variable	
		CC	Changed	FO-L-QC< UF < CE <fo-p-us<fo-l-us< td=""><td></td></fo-p-us<fo-l-us<>		
	Economic instead of	НН	Changed	FO-L-QC <uf< ce<fo-l-us="FO-P-US</td"><td>Lowest: FO-L- QC<uf< ce<="" td=""></uf<></td></uf<>	Lowest: FO-L- QC <uf< ce<="" td=""></uf<>	
	mass allocation on dry matter	EQ	Changed	FO-L-QC< UF <ce <fo-l-us <fo-p-us<="" td=""><td>Except for FEU others vary</td></fo-l-us></ce 	Except for FEU others vary	
		FEU	Changed	FO-L-QC <ce<uf< <fo-l-us="" <fo-p-us<="" td=""><td>•</td></ce<uf<>	•	
Allocation		CC	Changed	UF< FO-L-QC < FO-L- US <fo-p-us< ce<="" td=""><td></td></fo-p-us<>		
	Economic allocation	НН	Changed	UF< FO-L-QC <ce <fo-l-us<fo-p-us< td=""><td>Lowest: UF<fo-l- QC</fo-l- </td></fo-l-us<fo-p-us<></ce 	Lowest: UF <fo-l- QC</fo-l- 	
	with whey UF at 17.5% instead of 0%	EQ	Changed	UF <fo-l-qc<ce< td=""><td>Except for FEU others vary</td></fo-l-qc<ce<>	Except for FEU others vary	
		FEU	Changed	FO-L-QC <uf<ce <fo-<br="">L-US <fo-p-us< td=""><td></td></fo-p-us<></uf<ce>		
		CC	Changed	CE <uf<fo-l-qc< td=""><td></td></uf<fo-l-qc<>		
	R1 350 km vs	НН	Unchanged	CE <uf<fo-l-qc <fo-l-us<fo-p-us<="" td=""><td></td></uf<fo-l-qc>		
	national average 1500 km	EQ	Unchanged	CE <uf<fo-l-qc <FO-L-US<fo-p-us< b=""></fo-p-us<></uf<fo-l-qc 	FO-L-QC <fo-l- US<fo-p-us< td=""></fo-p-us<></fo-l- 	
US region		FEU	Changed	CE <fo-l-qc<uf <fo-l-us <fo-p-us<="" td=""><td></td></fo-l-us></fo-l-qc<uf 		
of milk sourcing		CC	Unchanged	CE <uf<fo-l-qc <FO-P-US<fo-l-us< b=""></fo-l-us<></uf<fo-l-qc 		
	R5 5000 km vs	НН	Changed	CE <uf<fo-l-qc <FO-P-US<fo-l-us< b=""></fo-l-us<></uf<fo-l-qc 	FO-L-QC <fo-p-< b=""></fo-p-<>	
	national average 1500 km	EQ	Changed	CE <uf<fo-l-qc <fo-p-us <fo-l-us<="" td=""><td>US <fo-l-us< td=""></fo-l-us<></td></fo-p-us></uf<fo-l-qc 	US <fo-l-us< td=""></fo-l-us<>	
		FEU	Unchanged	CE <fo-l-qc <uf<="" td=""><td></td></fo-l-qc>		

L&W rate reduction potential: A sensitivity analysis (detailed in S8.4 shows that the potential of CC impacts reduction is 45 times higher for L&W than for energy consumption (electricity and natural gas) at the plant. Indeed, a 1% L&W reduction at the GY plant is more effective than a 10% reduction in energy consumption to reduce CC impacts. The L&W from distribution to consumer (20% of the life cycle impacts) present even higher impact mitigation potential.

4. Discussion

4.1. key findings for GY manufacturers

This study demonstrates that milk production is the most important contributor to the environmental profile of GY. Any initiatives from manufacturers to select milk from regions with a low environmental footprint and reduce milk inputs and product L&W could significantly improve the environmental performance of GY. Manufacturers could also promote and support efforts to mitigate the impacts of dairy farms.

Procurement decisions about milk and milk protein sourcing significantly impact the performances of GY systems. Selecting liquid protein ingredients or protein powders depending on transportation distances can improve the environmental performances of GY. However, the milk production region remains the most important factor. The large variations observed among regions are mainly due to differences in farming practices such as crop feeding and manure management and, to a lesser extent, to regional climate. Therefore, any efforts by manufacturers to improve the traceability and impact assessment of their supply chains (MPC and milk sourcing) could have a positive influence on the environmental profile of GY.

Waste minimization has already been identified as a priority in some LCA studies in dairy literature [19,44–47]. Our results reinforce the idea that reducing L&W along the life cycle is an effective lever to improve the environmental impact of GY and should be investigated further. We demonstrated that reducing L&W by 1% at the plant level is more effective than reducing energy consumption by 10% and could even be much more effective if the initiative goes beyond the plant boundaries. For instance, cooperating with food security legislators to improve consumer communication on product shelf life labeling and participating in public awareness campaigns on the environmental impact of dairy food wastage could influence the reduction in losses at the distribution and consumption stages. Instead of being viewed as a potential factor that lowers sales volumes, it could become a factor of competitive differentiation and a commitment in terms of corporate social responsibility.

However, it is difficult to discriminate the environmental performance of each technology (CE, FO, UF) based on these results. Indeed, scenario results are sensitive to the allocation rules used between GY and its co-products and the milk sourcing regions. Although, CE tends to be the preferred scenario with mass allocation on dry matter, UF performs best with economic allocation. Furthermore, the low contribution of the processing operations stage (limited to 3% of CC, HH, EQ impacts for all the scenarios) demonstrates that the energy consumption at the manufacturing plant is not a critical factor over the entire product life cycle and the differences of energy consumption between each technology are not discriminating.

4.2. Limitations and opportunities

This study paves the way for a range of opportunities for further research. The major limitation pertains to the multifunctionality approach, which is a highly sensitive choice in determining the environmental burdens of various co-products. The accuracy of the L&W data is another area of improvement.

4.2.1. Multifunctionality and allocation method

Comparative LCA results between the five studied systems are sensitive to the allocation rules. While the sweet whey from the UF technology as a good potential to be recovered in the food industry, the acid whey from the CE and FO technologies is still considered as a by-product managed as a waste. In this respect, the IDF allocation based on dry-matter content is somewhat misleading in this comparative LCA. This rule attributes more impacts to the product that retains more dry matter in GY. Consequently, 1% more impacts from the raw milk (Table 2) are allocated to GY produced with the UF technology despite generating a co-product that is more valuable than the acid whey from CE or FO. Furthermore, fewer impacts are allocated to GY with the technologies that reject more valuable milk components in the acid whey. Reducing the impacts on GY by allocating the

environmental burdens to the acid whey is hardly justifiable choice should the results be used in an environmental declaration initiative. Therefore, despite ISO recommendation to prioritize causal physical relationship (such as dry-matter content) in allocation method (ISO 14044), in the current situation, in which GY manufacturers are still struggling to recover the milk solid content that ends up in the acid whey, we would recommend an economic allocation zeroing the allocation to acid whey. However, economic allocations depend upon price volatility and market demand. With this rule, results and conclusions may vary over time. For instance, the sensitivity analysis on the economic allocation value of whey (Table 3) showed that the fluctuations in the prices of UF whey components change the conclusions. UF may score better or worse as compared to the other technologies depending on the market value of the permeate.

Dealing with multifunctionality has always been an issue in LCA [48]. Many different solutions to the allocation problems have been suggested but, such as in this study, it has been demonstrated that choosing one among the other might have a decisive impact on the LCA results [46,49,50]. Recent LCA dairy studies [33,51,52] using the allocation approach recommended by the IDF, also conclude that the allocation section of the guideline should be improved. Our work demonstrated that mass allocation on dry matter is not the most appropriate for GY and that the economic allocation is not a panacea as well. Further work, evaluating other approaches addressing the multifunctionality of GY and, more generally dairy production, is therefore recommended but was out of the scope of this paper.

4.2.2. L&W data improvement

We used available data in the literature to assess the environmental burdens (S6) of L&W but sources are limited and often based on assumptions. The amount of milk and GY L&W along the supply chain were accounted for in the reference flows calculation. However, the treatment of these wastes along the supply chain was not considered due to the lack of information on the different waste treatment pathways and technologies involved.

Work to improve the accuracy of L&W data is not trivial. Since it is a major contributor to the entire product life cycle, any reduction in this area could lead to a significant improvement in the environmental performances of dairies.

5. Conclusion

This first comprehensive LCA comparing three GY producing technologies (CE, FO, UF) and three different MPC sourcing alternatives concluded that it is not possible to clearly discriminate the environmental performance between the assessed scenarios. The conclusions remain sensitive to the allocation modeling choices between GY and its co-products. Nevertheless, our findings serve to identify the environmental hotspots across the GY value chain, which has never been done before. It comes as no surprise that the major contributor is raw milk production and shows that the GY manufacturing stage is a low contributor to the environmental burdens of GY systems. Minimizing product L&W across the entire value chain (estimated at 32.5% overall) and sourcing milk from regions with the lowest environmental impacts could significantly improve the performance of GY. Therefore, L&W minimization should remain a key priority at the manufacturing plant. In addition, GY manufacturers can also significantly influence the life cycle performance of GY systems beyond their plant operations through targeted strategic decision-making related to ingredient procurement, product shelf-life and returned product management. Such a broader life cycle vision, that goes beyond the plant boundaries, is a step forward to guide the dairy industry toward a sustainable path.

Beyond these findings, this study also contributes to the debate on defining the most appropriate modeling to solve multifunctionality of dairy systems. We demonstrated the shortcomings of applying the mass allocation method based on the dry matter weight recommended by the IDF to compare GY systems producing yogurt, cream and whey with different characteristics. Further research is required to gain a better understanding of whey valorization pathways and find more holistic approaches to model such multifunctional systems.

Supplementary Materials: The following are available online at www.mdpi.com/xxx/s1

Author Contributions: Conceptualization, C.H.; methodology, C.H., S.B. and D.M.; software, S.B.; validation, M.M., D.M. and Y.P.; formal analysis, C.H.; investigation, C.H.,S.B.; resources, Y.P; writing – original preparation, C.H., writing – review and editing, M.M., S.B., D.M., Y.P.; supervision, M.M. and Y.P.; project administration, M.M.; funding acquisition, Y.P.

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Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

Appendix A

Table A1. Global inventory scaled up to the functional unit (FU): 1 kg of GY at 10% protein and 0% fat consumed by an average Québec household in 2018 before L&W. CE: centrifugation; FO-P-US: fortification with MPC 80 powder from the USA; FO-L-US: fortification with liquid MPC from the USA; FO-L-QC: fortification with liquid MPC from Canada; UF: ultrafiltration.

			FO-P-	FO-L-	FO-L-					
	Unit	CE	US	US	QC	UF	Source			
Inputs from technosphere										
Raw material procurement (SB1)										
Raw milk	kg	3.47	2.73	2.73	2.73	3.38	Simulation Benoit &			
	8	0.17	0	2 0	2., 0	5.5 6	Houssard (2017)			
MPC powder	kg	_	0.030	_	_	_	Houssard (2018) S4			
MPC liquid	kg	-	_	0.119	0.119	_	110 400414 (2010) 01			
Culture of lactic ferments (not included)		NI	NI	NI	NI	NI				
Raw milk transportation to Qc plant	t·km	0.654	0.516	0.516	0.516	0.637	PLQ (2016)			
MPC transportation to GY plant										
	t·km	-	0.044	0.178	0.018	-	Houssard (2018) S4			
Primary packaging (SB2)										
PP containers (50% of FU) -	œ	15.025	15.025	15.025	15.025	15.025				
Polyethylene (virgin content)	g	13.023	15.025	15.025	13.023	13.023				
PP containers recycled content	g	2.640	2.640	2.640	2.640	2.640	C 2010			
PS containers (50% of FU)	~	23.041	23.041	23.041	23.041	23.041	Survey 2018			
Polystyrene	g	25.041	23.041	25.041	23.041	25.041	S4			
PET seal for PP containers	g	0.512	0.512	0.512	0.512	0.512	<i>0</i> 1			
Laminated paper seal for PS containers	g	1.200	1.200	1.200	1.200	1.200				

Table A1. Global inventory scaled up to the functional unit (FU): 1 kg of GY at 10% protein and 0% fat consumed by an average Québec household in 2018 before L&W (cont'd)

			FO-P-	FO-L-	FO-L-		
	Unit	CE	US	US	QC	UF	Source
		Inputs f	rom tecl	nnosphe	re		
Primary packaging (SB2)							
HDPE lid for PP containers	g	7.172	7.172	7.172	7.172	7.172	Survey 2018
Bleached cardboard for PS	~	3.230	3.230	3.230	3.230	3.230	
containers (virgin content)	g	3.230	3.230	3.230	3.230	3.230	S4
Bleached cardboard for PS	α.	12.320	12.320	12.320	12.320	12.320	
containers (recycled content)	g	12.520	12.520	12.320	12.520	12.320	
Secondary packaging (SB2)							
Corrugated board	g	48.860	48.860	48.860	48.860	48.860	Survey 2018
LLDPE stretch wrap film	g	0.788	0.788	0.788	0.788	0.788	
Wood pallet	g	0.141	0.141	0.141	0.141	0.141	S4
GY Processing at plant (SB2)							
Electricity							
Milk filling and storage at 4° C	Wh	0.136	0.107	0.107	0.107	0.133	
Heating at 55°C	Wh	0.156	0.127	0.127	0.127	0.247	
Skimming at 55°C	Wh	3.611	2.842	2.842	2.842	3.520	
Fortification at 55°C	Wh	-	0.830	0.077	0.077	-	
Ultrafiltration at 55°C	Wh	-	-	-	_	1.011	
Thermal treatment at 88°C for 6 min.	Wh	1.440	1.350	1.350	1.350	1.041	
Homogenisation at 65 °C & 170-200	Wh	17.400					Simulation Benoit &
bars (optional)			_	_	_	_	Houssard (2017)
Cooling at 42°C	Wh	0.247	0.208	0.208	0.208	0.041	
Fermentation at 42°C during 5 to 8	Wh	0.289	0.245	0.245	0.245	0.044	
hours							
Centrifugation at 35 - 40°C	Wh	15.053	11.847	11.847	11.847	-	
Cooling at 15°C in 20 to 30 sec.	Wh	3.102	3.538	3.538	3.538	3.059	
CIP	Wh	0.167	0.164	0.164	0.164	0.162	
Packaging and storage at 4°C	Wh	14.236	14.236	14.236	14.236	14.236	Prasad (2003, 2004)
Plant ventilation and lighting	Wh	9.491	9.491	9.491	9.491	9.491	Prasad (2003, 2004)
Natural gas							
Heating treatments regeneration	MJ	0.618	0.490	0.490	0.490	0.738	Simulation Benoit &
system		0.00=	0.007	0.004	0.007	0.00=	Houssard (2017
CIP	MJ	0.087	0.086	0.086	0.086	0.085	
Chemicals and water							
Sodium hydroxide in 50% solution	g	0.361	0.356	0.356	0.356	0.351	Simulation Benoit &
state		0.400	0.127	0.40=	0.40=	0.125	Houssard (2017)
Nitric acid in 50% solution state	g	0.139	0.137	0.137	0.137	0.135	

Table A1. Global inventory scaled up to the functional unit (FU): 1 kg of GY at 10% protein and 0% fat consumed by an average Québec household in 2018 before L&W (cont'd and end)

			FO-P-	FO-L-	FO-L-		
	Unit	CE	US	US	QC	UF	Source
		Inpu	ts from to	echnospł	nere		
Chemicals and water							
Deionized water for MPC powder hydration	kg	_	0.090	_	_	_	Simulation Benoit &
Deionised water for CIP	kg	0.255	0.252	0.252	0.252	0.249	Houssard (2017)
Other plant tap water usage	kg	2.941	2.315	2.315	2.315	2.867	Gonzalez-Garcia (2013)
Distribution (SB3)							
Electricity	Wh	186.100	186.100	186.100	186.100	186.100	Gonzalez-Garcia (2013)
Transportation							Calculation
	t·km	0.145	0.145	0.145	0.145	0.145	
							S4
Consumption (SB4)							
Plastic bag	g	2.000	2.000	2.000	2.000	2.000	Hospido A (2006)
							Calculation
	km	0.146	0.146	0.146	0.146	0.146	
Transportation							S4
Electricity (refrigeration)	Wh	54.700	54.700	54.700	54.700	54.700	Gonzalez-Garcia (2013)
Tap water	kg	0.8045	0.8045	0.8045	0.8045	0.8045	Gonzalez-Garcia (2013)
		Out	put to te	chnosph	ere		
Wastes to treatment (SB2, SB3,	SB4, SB	5)					
White water from plant	m3	3.20E-	2.57E-	2.57E-	2.57E-03	3.12E-	Simulation Benoit &
White water from plant	nio	03	03	03	2.57 £ 65	03	Houssard (2017)
Other waste water treatment	m3	8.05E-	8.05E-	8.05E-	8.05E-04	8.05E-	
Offici waste water treatment	1113	04	04	04	0.03L-04	04	Calculation
Cardboard and corrugated	~	71.160	71.160	71.160	71.160	71.160	
board	g	71.100	71.100	71.100	71.100	71.100	S4
Plastic mixture landfill	g	49.178	49.178	49.178	49.178	49.178	
Municipal waste collection	41	1.19E-	1.13E-	1.13E-	1 12E 02	1.13E-	
(transportation)	t∙km	02	02	02	1.13E-02	02	
Product and co-products (SB2)							
Cream	kg	0.341	0.268	0.268	0.268	0.332	
Greek Yogurt (GY)	kg	1.000	1.000	1.000	1.000	1.000	
Whey	kg	2.129	1.559	1.559	1.559	2.044	

S4: See extensive details in section 4 of the supplementary materials file

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