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Less refined ingredients have lower environmental impact – A life cycle assessment of protein-rich ingredients from oil- and starch-bearing crops

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A B S T R A C T

In the coming decades, meat-based protein foods will increasingly be replaced by plant-based protein foods. These are typically prepared from highly refined protein isolates or concentrates, which require a lot of energy and auxiliary chemicals to be produced. Milder techniques such as dry fractionation or mild aqueous fractionation deliver alternative ingredients that have a multicomponent character but do not need the same amount of chemicals and energy. This study aims to assess the effect of reducing the degree of refining on the environmental impact using a life cycle assessment. The functional unit was 1000 kg of the processed crop. As protein is considered key in these ingredients, the functional unit of 1 kg of protein in the produced fractions was also assessed. The contribution of processing to the overall impact was found to be significant and, in some cases, larger than the contribution by the crop cultivation. Therefore, any analysis of the environmental impact should include both. Reducing the degree of refining substantially reduces global warming potential, human carcinogenic toxicity, fossil resource scarcity, and water consumption. However, for all impact categories, drying remains the largest contributor. The global warming potential of less refined ingredients was still lower compared to the conventionally refined ingredients when expressed per kg of protein, despite the significantly lower amount of protein. The fractions obtained through mild aqueous fractionation have a higher protein yield and a lower global warming potential compared to conventional full refining. Both dry fractionation and the combination of dry and mild aqueous fractionation substantially lower the environmental impact, but the protein yield and purity are also considerably lower. Overall, linking environmental impact to protein purity and yield allows for a comprehensive selection of sustainable food ingredients.

1. Introduction

Ready-to-eat soups, vegan ice-cream, and meat replacers are processed food products require texturisers that are often protein-based. These ingredients can be produced from animal products, such as whey protein isolate, or plants, such as pea protein isolate. Especially plant proteins are of increasing importance due to pressing climate issues, which require food producers to become more sustainable (Aiking, 2011). Currently, proteins that originate from oil-, starch-, or protein-bearing crops are isolated by isoelectric precipitation, requiring chemicals such as caustic (NaOH) and hydrochloric acid (HCL) for extraction and precipitation (Passe et al., 2008), and hexane in case of an oil-bearing seed (Wäsche et al., 2002). Since every step in such a multi-step process involves intrinsic losses, the total yield of the desired components decreases with the number of steps. The side streams that are generated are often too dilute to use or may have such reduced quality that they are not suitable for human consumption anymore (Berghout et al., 2015). Therefore, we assert that to have adequate sustainability in our food production, it is important to utilize the complete crop and hence, the least amount of resources.

Processing, or fractionation, methods have been developed that result in a lower degree of refining. These methods omit chemicals and aim at valorising the whole crop. Perhaps the most radical example is dry fractionation. The use of water is avoided, which

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normally dilutes the raw materials and inevitably takes some of the raw materials into the wastewater. Dry fractionation typically relies on differences in the size, density, and tribocharging properties of particles. This method includes milling and air classification or uses electrostatics to separate flours of pulses such as yellow pea, chickpea (Schutyser and van der Goot, 2011), or wheat bran (Hemery et al., 2011) into protein- and starch-rich fractions. Another method is mild aqueous fractionation, which is a simplification of the conventional wet fractionation method – largely omitting the use of chemicals – and has been demonstrated for the oil-bearing lupine seeds (Pelgrom et al., 2014) and soybean (Peng et al., 2020). The same method was applied to the starch-bearing crop yellow pea (Geerts et al., 2017a,b).

A lower degree of processing results in fractions that have a multicomponent character rather than being pure in one component. Nevertheless, promising functional properties have been found; such as the thickening capacity of mildly refined yellow pea or (Kornet et al., 2020a) soy fractions (Peng et al., 2020) and emulsifying properties of pea flour (Sridharan et al., 2020). These milder fractionation methods for yellow pea (Geerts et al., 2018) and lupine (Berghout et al., 2015) have a better exergy efficiency compared to the conventional fractionation. Combining dry and mild aqueous fractionation can further reduce water and energy consumption. However, these exergy analyses only considered processing and did not include crop cultivation. Besides, they only considered exergy (useful energy) efficiency and therefore did not consider other modes of environmental impact. Lastly, these methods for lower levels of refinement create fractions with a lower protein content, while it is considered the most important component for which more sustainable alternatives must be found. To evaluate these three factors, we here report on a full attributional life cycle assessment comparing different degrees of refining; based on the whole process and protein content, from cultivation to processing.

A life cycle assessment (LCA) allows for a multidimensional assessment of the environmental impact of products at all stages in the food production chain. This includes the extraction of resources and emission of hazardous substances (Guinée, 2002). Most current LCA studies focus on calculating the footprint of the production of only conventional protein isolates (Berardy et al., 2015) while less have focused on the milder fractionation methods (Vogelsang-o'Dwyer et al., 2020). Therefore, this study aims to directly compare the environmental impact of protein-rich fractions from conventional and milder fractionation methods from starch- and oil-bearing crops. It is hypothesized that a lower degree of refining decreases all sustainability indicators due to the lower use of resources. Further, expressing the environmental impact in protein will negatively influence the footprint of a lower degree of refining as their products contain less protein than the fully refined isolates. To exemplify the effect of processing on the impact of protein-rich ingredients, a starch- and an oil-bearing crop were selected: lupine and yellow pea. These crops were selected due to the significant protein content of approximately 40 (Rodríguez-Ambriz et al. (2005)) and 23% (USDA, 2017) respectively. In addition, these crops have established functional properties such as foaming abilities (Adebiyi and Aluko, 2011), emulsifying capabilities (Geerts et al., 2017a), and gelation (Kornet et al., 2020b). At first, the most significant environmental impact categories of processing these crops will be presented. After this, the relative impacts of processing and cultivation will be compared. Finally, the impact of the individual process steps and the fractions relative to the purity and yield of protein is discussed. Evidently, the selection of sustainable ingredients is not merely based on the purity of the fraction, as other factors such as functional properties are also important. However, these are not within the scope of this study.

2. Goal and scope

The goal of this study was to quantify the impact of decreasing the degree of refining on the environmental impact of food ingredients using an attributional LCA. To achieve this, four different refining processes were compared for yellow pea and lupine, namely: conventional protein extraction, mild aqueous fractionation, dry fractionation, and combined dry and mild aqueous fractionation, or hybrid fractionation.

2.1. Ingredient selection

In this study, the production of ingredients from lupine legume seeds (*Lupinus angustifolius* L.) and yellow pea (*Pisum sativum*) in the Netherlands was considered. The yields of the ingredients from the starting material and the purities were obtained or based on information of the processes in literature (further explained in 3.1.2) (Table 1). In the case of hybrid fractionation of yellow pea, the protein content was determined experimentally, for which the dry fractionation by air classification (Pelgrom, Boom. et al., 2015) was combined with mild aqueous fractionation (Geerts et al., 2017a). The protein content of the ingredients was determined using Dumas analysis (Nitrogen analyser, FlashEA 1112 series, Thermo Scientific, Interscience, Breda, The Netherlands). A conversion factor of 5.52 was used for the calculation of the protein content (Holt and Sosulski, 1979). Generally, the milder fractionation methods lead to ingredients that contain less protein but could have a higher protein yield. The latter is defined as the protein in the ingredients as a percentage of the original protein present in the crop and can be calculated using Equation (1).

$$Protein\ yield(\%) = Ingredient\ yield(\%) \cdot \frac{Protein\ content\ of\ ingredient}{Protein\ content\ of\ crop\ flour} \tag{1}$$

Ingredient yield is the mass of the obtained ingredients as a percentage of the initial mass of the starting material and the protein content is the amount of protein in the specific ingredient or crop flour.

2.2. System boundaries

The system boundaries were set from cultivation to the end of ingredient processing, in other words: cradle-to-processing-gate (Fig. 1). These boundaries were picked because the food assembly, distribution, use of the product, and disposal after the production were considered not relevant for the current comparison between high and low degrees of refining. The analyses in this study were performed both including and excluding the impact of the cultivation and transportation, to highlight the effect of decreasing the degree of refining in processing only. Energy, chemicals, and water that go into the system were considered for this analysis, as well as

Table 1
Total yield of the ingredient from starting material and protein content in the ingredients based on dry matter (db%) of the protein-rich fractions obtained through conventional, mild aqueous, dry, and hybrid fractionation.

Crop	Method	Fraction	Ingredient yield (db%)	Protein content (db%)
Yellow Pea	Raw material	Yellow pea flour[a]		21.4
	Conventional fractionation	Protein isolate[a,b]	22.3	78.8
	Mild aqueous fractionation	Soluble protein[c]	23.9	55.9
		Insoluble protein[c]	10.2	53.3
	Dry fractionation	Fine fraction[a]	22.8	42.9
	Hybrid fractionation	Soluble protein	5.1[e]	62.2[d]
		Insoluble protein	3.8[e]	37.4[d]
Lupine	Raw material	Lupine flour[f]		39.5
	Conventional fractionation	Protein isolate[f]	27.0	87.0
	Mild aqueous fractionation	Enriched protein[f]	29.0	86.5
	Dry fractionation	Fine fraction[f]	33.0	57.6
	Hybrid fractionation	Enriched protein[f]	14.0	87.0

[a] Pelgrom et al. (2015).
[b] Passe et al. (2008)
[c] Geerts et al. (2017a) (supplementary information).
[d] Obtained experimentally by this study.
[e] Schutyser et al. (2015) (supplementary information).
[f] Berghout et al. (2015) (supplementary information).

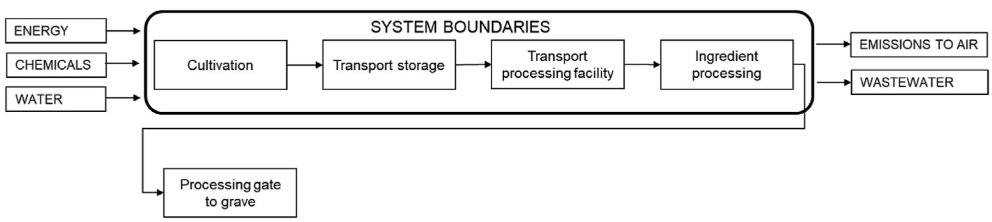

Fig. 1. System boundaries for the life cycle assessment of the production of food ingredients.

the emissions to air and wastewater that come out of the system, which is further explained in the life cycle inventory in section 3.

2.3. Functional unit & allocation

The functional unit was defined as 1000 kg of the processed crop to compare the impact among the different processes. This functional unit was picked since the goal of this study was to assess the overall impact of reducing the degree of refining on the complete process. Since protein is the key component of the ingredients, 1 kg of protein as a functional unit was also investigated. The allocation was done based on mass (dry matter) allocation. Fractions that were considered as a loss (i.e. soluble solids after precipitation), were treated as waste; hence no impact was allocated. This means that all useful co-streams produced during the development of the protein-rich ingredients (i.e. starch or fibre-rich fractions) received an allocation according to mass in all analyses. In this study, when discussing the functional unit per 1000 kg crop, the footprint of all fractions was considered. For the discussion of the environmental impact per kg protein, only the allocated impact of the protein-rich ingredients was considered.

3. Life cycle inventory

The data collected for the agricultural production and transportation of yellow pea and lupine were retrieved from the Agri-footprint 5.0 database (van Paassen et al., 2019). For more details on how this data is derived one can consult the database description, which is publicly available. The impact for all individual fractionation processes was calculated using data for electricity, process steam, water, and chemicals retrieved from the Agri-footprint 5.0 database (Table S1 in Supplementary information).

Table 2
Energy requirements for the process parameters used in the LCA impact assessment, a detailed description of all processes can be found in the supplementary information.

Process parameters	Unit	Energy consumption
Electricity use		
Mill	MJ/kg feed	0.5[a]
Air classifier	MJ/kg feed	0.023[a]
Dispersion mixing	MJ/kg protein	1.5[a]
Hydrocyclones	MJ/kg feed	0.0018[b]
Centrifugal decanter	MJ/kg feed	0.0024[b]
Vacuum drum filter	MJ/m^3 feed	99[b]
Ultrafiltration	MJ/m^3 feed	10[b]
Air pump	MJ/kg feed	0.019[b]
Electricity oil extractor	MJ/kg flour	0.07[c]
Fuel energy use drying		
Pneumatic ring dryer	MJ/kg water removed	4.3[b]
Evaporator	MJ/kg water removed	0.8[b]
Spray dryer	MJ/kg water removed	4.8[a]
Fluidized bed	MJ/kg water removed	6.75[b]

[a] Schutyser and van der Goot (2011).
[b] Geerts, van Veghel et al., 2018.
[c] Berghout et al. (2015).

3.1. Cultivation & transportation

Both the agricultural production and the transportation to storage or feed plants were based on the impact of the consumption mix of the specific crop in the Netherlands and extracted from the Agri-footprint 5.0 database, which is available in the supplementary data (Table S2) (van Paassen et al., 2019). The impact of yellow pea was determined from pea in general, as only this was available in the database. The consumption mix for pea originated from Belgium (0.1%), Canada (4.6%), Czech Republic (1.3%), Estonia (0.2%), Finland (23.4%), France (16.9%), Germany (1.7%), Hungary (8.6%), Lithuania (5.1%), the Netherlands (1.7%), Poland (18.4%), Russia (12.0%), Ukraine (4.3%), the United Kingdom (0.7%), and the United States (1.1%). The lupine seeds originated from Germany (20%) and Australia (80%). The distance between the location of cultivation versus the location of processing will influence the overall environmental impact, which is case study dependent.

3.2. Ingredient processing

In this study, only the resources related to the production of the ingredients were considered relevant for the comparison between processes with a different degree of refining. Additional impact by for example construction, maintenance, and cleaning of equipment was not included. The information for the conventional and mild aqueous fractionation of yellow was mainly based on literature (Geerts et al., 2018). A patent was also used for additional information for the conventional process (Passe et al., 2008). The conventional fractionation of lupine was also based on literature (Berghout et al., 2015) and a patent (Wäsche et al., 2002). The mild aqueous fractionation of lupine was based on the same study as the conventional variant and a patent (Snowden et al., 2007). As the exergy study by Berghout et al. (2015) mainly focussed on the drying and oil extraction steps, the processes were completed using information from the fractionation of yellow pea in this study. The dry fractionation of both crops was based on literature (Schutyser et al., 2015). The different degrees of processing were defined as follows and presented in more detail including flow diagrams in the supplementary information.

In general, all wet processes start with a milling and steeping step. The initial separation of starch and fibre fractions is done with either hydrocyclones or decanters. These fractions are subsequently dried by evaporation or a vacuum drum filter and a pneumatic ring drier. In the conventional way of fractionation, the oil (in the case of oil-bearing crops) is initially removed, after which the proteins are precipitated isoelectrically and finally neutralized. The precipitation and neutralization steps are omitted in the mild aqueous fractionation for yellow pea. The protein-rich fractions are concentrated using ultrafiltration and/or dried through spray drying and a fluidized bed. In the milder process for oil-bearing crops the precipitation step is still included, yet, the oil extraction is omitted. The mildest fractionation method for both crops is dry fractionation; using a milling step and air classifier. In hybrid fractionation, the fine fraction obtained through dry fractionation is further processed using the mild aqueous fractionation. A detailed description including the assumptions for these fractionation methods can be found in the supplementary information. The energy requirements were based on the parameters presented in Table 2.

4. Life cycle impact assessment

The life cycle impact (LCI) assessment was performed by combining the existing impact of the agricultural production and transportation and the modelled fractionation processes using Simapro LCA software version 9.0 and the ReCiPe 2016 Midpoint (H) V1.03 method (Huijbregts et al., 2016). First, the LCI data points and the connected background processes (Table S1) are combined and compiled into an inventory list of the substance flows (e.g. “methane, biogenic” to Air in kg CH_4/kg product or “dinitrogen monoxide” to Air in kg N_2O/kg product). Next, the ReCiPe method will translate the substance flows to the various impact categories, based on characterization factors as indicated in the ReCiPe method. A substance flow can influence more impact categories, such as “dinitrogen monoxide” to Air has a characterization factor of 298 kg CO_2 eq/kg N_2O for global warming and 0.011 kg CFC-11/kg N_2O for Stratospheric Ozone Depletion.

5. Results & discussion

Environmental impacts of the different fractionation methods for processing 1000 kg of yellow pea and lupine with a lower degree of refining are compared excluding cultivation and transportation (Table 3). In general, all impact categories decrease with a lower degree of refining in the case of yellow pea and lupine. Both the conventional and mild aqueous fractionation of lupine have a higher environmental impact compared to yellow pea, which can be attributed to the differences between processing starch- or oil-bearing crops. Dry fractionation of lupine seeds is similar to yellow pea as the process is the same. In the following paragraphs, the main differences between the environmental impacts will be discussed and related to the differences among the fractionation processes. The overall impact to process 1000 kg of crop including cultivation and transportation is presented in the supplementary information.

5.1. Comparison impacts from a high to low degree of processing

Four impact categories were selected to compare the impacts of only the different fractionation processes; hence, without cultivation and transportation. The categories represent the impact on 1) ecosystems using global warming potential, 2) human health with human carcinogenic toxicity, and 3) resources using water consumption expressed in blue water use and 4) fossil resource scarcity. For processing 1000 kg yellow pea or lupine, all environmental impact factors decrease drastically compared to the conventional way of isolation, which is set as 100% in Fig. 2. The results for human carcinogenic toxicity introduce a degree of uncertainty since it is difficult to measure, calculate, and translate into a single impact indicator (Fantke et al., 2018). Therefore, the numbers are used for a comparison of health damage, rather than absolute values.

Mild aqueous fractionation reduces all environmental indicators with approximately 30–40% for yellow pea and 20–35% for lupine compared to conventional fractionation. The impact of yellow pea can be decreased more compared to lupine due to several differences between processing oil- or starch-bearing crops. The fractionation of lupine requires an oil extraction step and involves larger quantities of water compared to the fractionation of yellow pea. This water eventually needs to be removed again to produce dried ingredients. Moreover, in the mild aqueous fractionation of lupine, protein is still extracted by isoelectric precipitation. This step is not used in the milder fractionation of yellow pea. Therefore, the extraction of protein from lupine seeds could be rendered even more sustainable by omitting oil extraction *and* protein precipitation. Using dry instead of conventional fractionation decreases the impacts by up to 99% for both crops. The impact of hybrid fractionation lays in between the other two methods. Geerts et al. (2018), also reported that mild aqueous fractionation has a higher exergy efficiency of 54%, compared to 35% in conventional fractionation, mainly due to the loss of immaterial exergy. Similarly, an exergy efficiency for dry fractionation of 99–100% was found, since all materials and limited amounts of electricity were used. Furthermore, Berghout et al. (2015) presented that oil extraction leads to significant exergy losses and destruction. The removal of this step is the main cause of the decrease in environmental indicators in mild aqueous and hybrid fractionation found in this research. The results show that a life cycle assessment can be very valuable in sustainability analyses next to exergy analyses. They provide a detailed insight into the translation of higher efficiencies to environmental impacts.

Table 3
All environmental impacts for the fractionation of yellow pea and lupine to process 1000 kg crop, excluding cultivation. Green-yellow-orange-red highlighted boxes indicate the fractions with the lowest to highest impact among all fractions from both lupine and yellow pea. In bold are the impact categories that are further discussed.

		Yellow pea				*Lupine*			
Impact category	***Unit***	***Conventional fractionation***	***Mild Aqueous fractionation***	***Dry fractionation***	***Hybrid fractionation***	***Conventional fractionation***	***Mild Aqueous fractionation***	***Dry fractionation***	***Hybrid fractionation***
Global warming	**kg CO eq**	**940**	**541**	**107**	**251**	**1464**	**1156**	**107**	**564**
Stratospheric ozone depletion	kg CFC11 eq	0.00020	0.00010	0.00003	0.00006	0.00034	0.00030	0.00003	0.00014
Ionizing radiation	kBq Co-60 eq	16.8	9.0	3.8	6.0	24.5	24.6	3.8	12.1
Ozone formation, Human health	kg NOx eq	0.012	0.0042	0.0008	0.0019	0.0979	0.0613	0.0008	0.0563
Fine particulate matter formation	kg PM2.5 eq	0.26	0.13	0.03	0.07	0.48	0.41	0.03	0.19
Ozone formation, Terrestrial ecosystems	kg NOx eq	0.0168	0.0068	0.0013	0.0031	0.1277	0.0688	0.0013	0.0607
Terrestrial acidification	kg SO^2 eq	0.83	0.41	0.10	0.21	1.57	1.33	0.10	0.61
Freshwater eutrophication	kg P eq	0.00006	0.00003	0.00001	0.00001	0.00013	0.00012	0.00001	0.00006
Marine eutrophication	kg N eq	0.0040	0.0016	0.0002	0.0006	0.0091	0.0067	0.0002	0.0027
Terrestrial ecotoxicity	kg 1,4-DCB	39.1	18.3	6.8	11.5	81.1	77.7	6.8	37.2
Freshwater ecotoxicity	kg 1,4-DCB	0.017	0.009	0.003	0.005	0.028	0.026	0.003	0.013
Marine ecotoxicity	kg 1,4-DCB	0.084	0.045	0.013	0.025	0.157	0.143	0.013	0.082
Human carcinogenic toxicity	**kg 1,4-DCB**	**0.28**	**0.17**	**0.02**	**0.07**	**0.43**	**0.31**	**0.02**	**0.15**
Human non-carcinogenic toxicity	kg 1,4-DCB	2.04	1.07	0.35	0.62	4.11	3.32	0.35	1.69
Land use	m^2a crop eq	0.11	0.07	0.00	0.02	0.20	0.14	0.00	0.08
Mineral resource scarcity	kg Cu eq	0.030	0.017	0.004	0.009	0.048	0.041	0.004	0.021
Fossil resource scarcity	**kg oil eq**	**248+**	**153**	**25**	**67**	**370**	**283**	**25**	**146**
Water consumption	**m^3**	**15.2**	**10.4**	**0.2**	**3.1**	**27.0**	**18.3**	**0.2**	**9.6**

5.2. Environmental impacts broken down per process step

The impact to process 1000 kg of crops without cultivation and transportation is divided into the main processing steps: milling, oil removal, steeping, separation (hydrocylone or decanter), precipitation and neutralization, and drying or ultrafiltration ((Fig. 3 and Fig. 4). This division aimed at determining the origin of the impact of the fractionation of yellow pea and lupine.

5.2.1. Yellow pea

For yellow pea, the global warming potential, human carcinogenicity, fossil resource scarcity, and water consumption all decrease with a lower degree of refining. A closer look into the process shows that the omission of the acidic precipitation step, with the associated heating step, is mostly responsible for the decrease in all impact categories (Fig. 3). With the removal of this step, less or even no water, no chemicals, and less electricity or process steam were required. In addition, treating the wastewater of the fraction containing soluble solids after precipitation leads to negative water usage, as water is returned to the environment. As the precipitation step is removed, the subsequent decanter step could also be left out. The latter was mainly responsible for the impact in the separation category and created a large waste stream. The impact of the hydrocyclones/decanters category is also lower for mild aqueous fractionation since starch is separated in decanters. In contrast, conventional fractionation uses hydrocyclones, which require more energy compared to decanters. However, the decanters in mild aqueous fractionation produce a larger starch fraction compared to the conventional method. This requires more energy to be dried by the subsequent pneumatic drying step. Therefore, replacing decanters with hydrocyclones in this case study will have a low effect on the overall environmental impact. The environmental impact is further reduced with dry fractionation, which results in a decrease ranging from 87 to 99% in impact. Overall, drying is responsible for a large part of the impact and does not decrease very dramatically in the mild aqueous fractionation

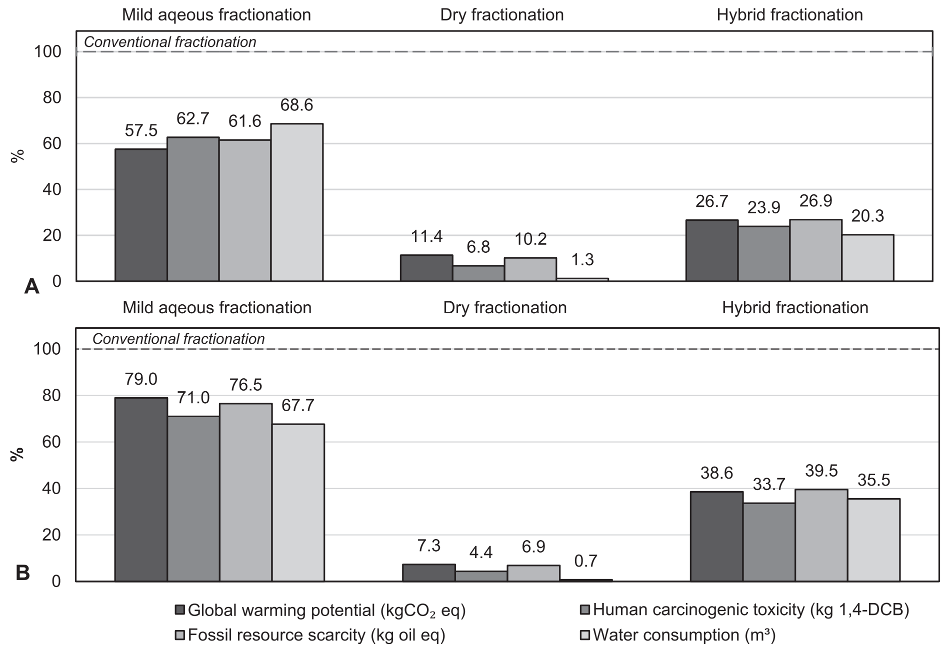

Fig. 2. Global warming potential, fossil resource scarcity, human carcinogenic toxicity, and water consumption of mild, dry, and hybrid fractionation of 1000 kg crop without cultivation. The environmental impacts are expressed as a percentage from the conventional fractionation method for (A) yellow pea and (B) lupine.

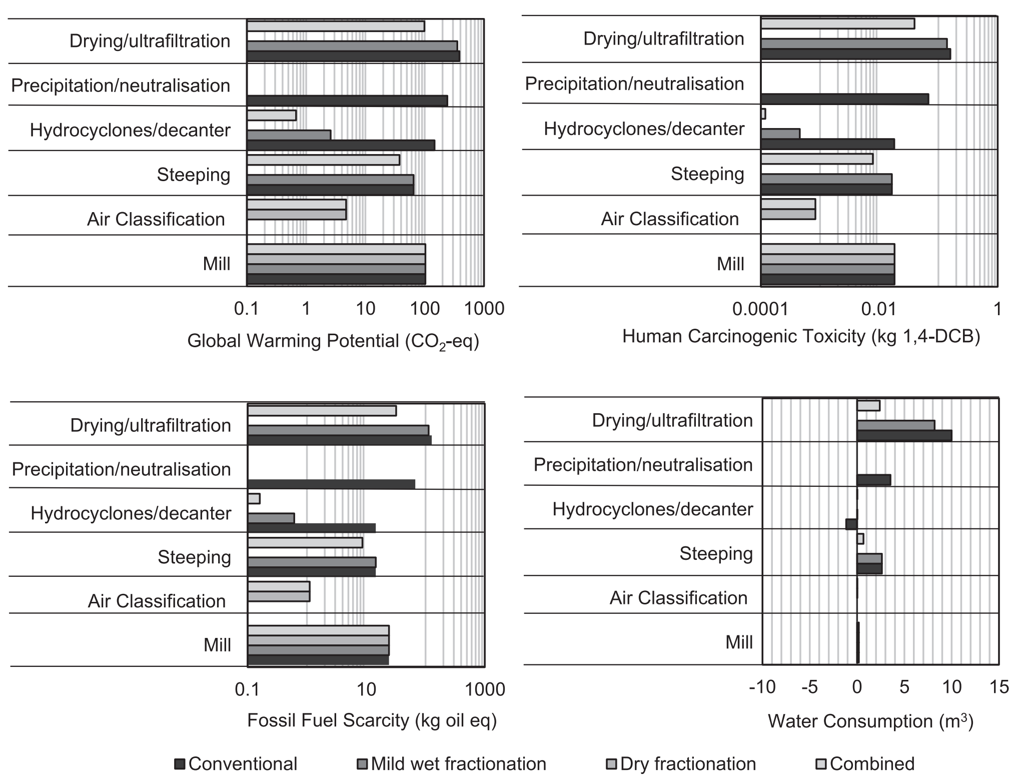

Fig. 3. The global warming potential, human carcinogenic toxicity, fossil fuel scarcity, and water consumption for each process step in conventional, mild aqueous, dry, and hybrid fractionation of yellow pea. Please note the log scale.

methods. Geerts et al. (2018) showed that drying was the main driver for exergy losses in both conventional and mild wet fractionation, whereas the precipitation step was responsible for only 10% of the exergy losses. This is most likely a slight underestimation because the material and immaterial costs of the materials (acids and bases) were not considered in that study. This can now also be translated into the environmental impact, which shows that the omission of the extraction-precipitation process can drastically reduce the environmental indicators.

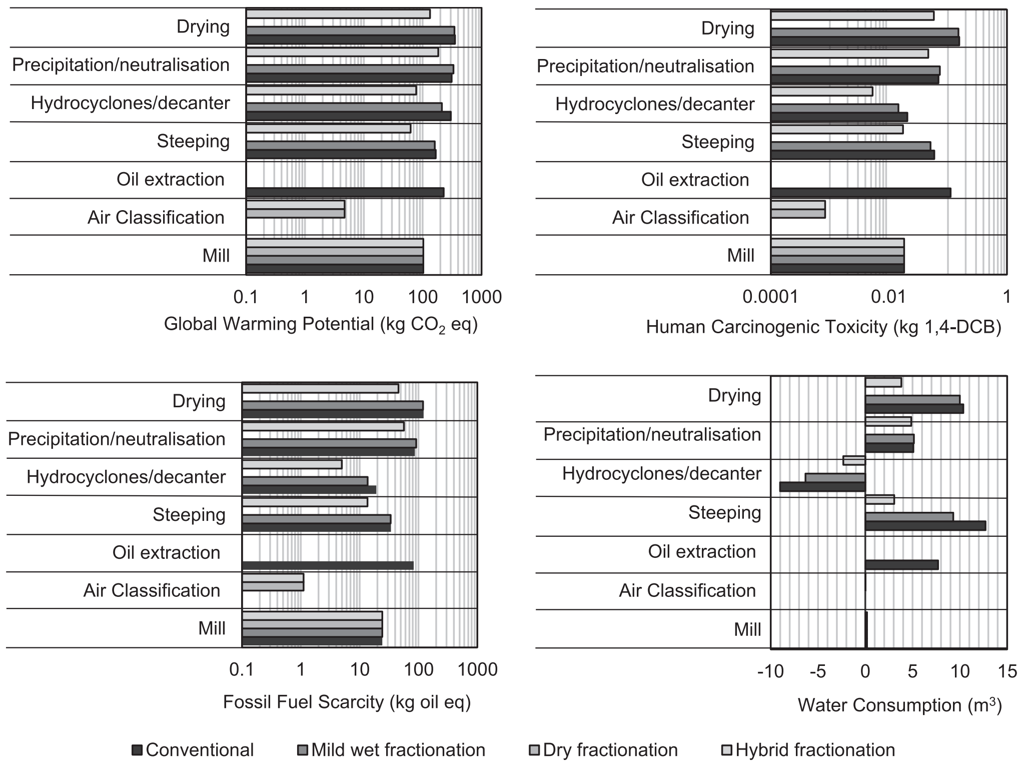

Fig. 4. The global warming potential, human carcinogenic toxicity, fossil fuel scarcity, and water consumption for each process step in conventional, mild aqueous, dry, and hybrid fractionation of lupine. Please note the log scale.

5.2.2. Lupine

Fractionating lupine without the removal of oil reduces all impact categories, as the use of hexane and the distillation of hexane is omitted. Through mild aqueous fractionation of lupine, the oil fraction (approximately 10% in lupine flour based on dry matter) ends up in very small amounts in the protein isolate (0.02–0.07 g oil/g protein isolate) and the rest in the fibre-rich pellet. The functional properties of the protein-rich fraction are not significnatly altered by the presence of oil compared to the conventional protein isolate (Berghout et al., 2014). The same was found for the dry fractionated fine fraction of lupine, which showed increased foam stability compared to the conventional lupine protein isolates (Pelgrom et al., 2014). For both crops, dry fractionation leads to the largest decrease in footprint, as less electricity, no process steam, and no chemicals are used; of course, at the cost of product purity. The combined method is situated between the dry and mild aqueous fractionation as the fine fraction was further processed (Fig. 4). The drying steps are responsible for a large part of the footprint. This is corroborated by Berghout et al. who also considered alternatives for processes that require a lot of energy for drying, such as dry fractionation (Berghout et al., 2015). The water use was reduced in the milder fractionation method, mainly due to the different ratio of water to material that was used. As for fractionation yellow pea, wastewater was also returned to the environment after receiving treatments. Especially the human carcinogenic toxicity is reduced drastically with the removal of the oil step. This is mostly attributed to heating hexane during the oil distillation rather than the use of hexane, since the hexane is reused almost completely. More specifically, only 3 kg hexane per ton lupine protein isolate is typically lost into the atmosphere during production (European Commission, 2008) and hence, affects the human carcinogenic toxicity, whereas the remainder is not considered.

In general, these results confirm the importance of reducing the degree of processing for food ingredients from the viewpoint of the environmental impact. Mainly the removal of the alkaline-acidic extraction-precipitation process and the oil extraction are responsible for this. Moreover, drying remains a dominant process step, which indicates that this remains an important issue to focus on. One should bear in mind that the milder fractionation pathways are still based on lab-scale processes that could be better optimized when developed for industrial scale. Therefore, the results may still change due to upcoming developments. We do believe however that the conclusions will not change, and in fact, will only become more distinct due to better efficiencies on larger scales. Furthermore, the different processes deliver products with very different qualities and properties. The conventional protein isolates are quite pure, while the concentrates from the other processes contain significant amounts of other components, but may still have good, though different functionality. Therefore, the direct comparison of these processes is not without complexity.

5.3. Comparison between cultivation and processing

Until now the environmental impact of different ways of refining 1000 kg yellow pea and lupine was discussed without including cultivation and transportation. Fig. 5 illustrates the effect of including the global warming potential, human carcinogenic toxicity, fossil resource scarcity, and water consumption of cultivation, transportation, and processing of 1000 kg of lupine or yellow pea. The combined environmental impact to produce 1 kg of ingredient and protein can be found in the supplementary information. The global warming potential and fossil resource scarcity of the cultivation of lupine or yellow pea are similar. The cultivation of lupine has more effect on human carcinogenic toxicity compared to yellow pea. The latter is explained by the higher toxicity emission

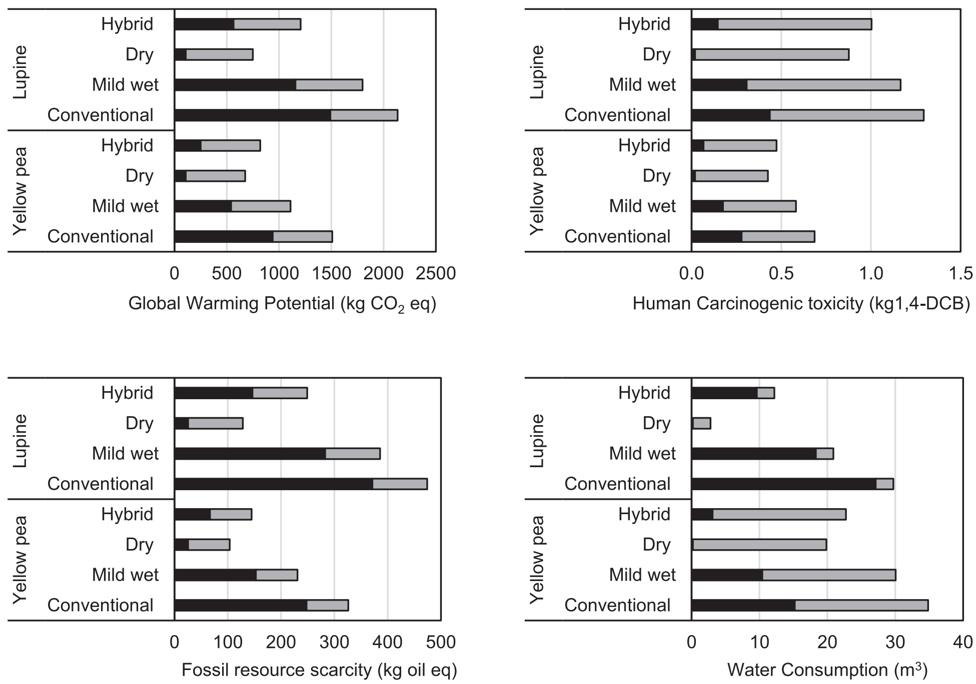

Fig. 5. The global warming potential, human carcinogenic toxicity, fossil resource scarcity, and water consumption of cultivation (grey) and processing (black) for yellow pea and lupine for processing 1000 kg crop.

related to the use of insecticides and herbicides outside of the EU (lupine originates partly from Australia) and longer transportation distances. In contrast, yellow pea requires more water during cultivation compared to lupine, as it is cultivated using irrigation (van Paassen et al., 2019).

For the global warming potential and fossil resource scarcity of both conventional and mild wet fractionation, the processing is dominant over the cultivation due to the relatively low energy requirements of the cultivation. The human carcinogenic toxicity of cultivation is dominant for all processes, mainly caused by the use of pesticides. With regards to water consumption, the processing of lupine contributes substantially more than its cultivation. This is different for yellow pea due to the difference in irrigation and water use in the steeping step during fractionation. The ratio between cultivation and fractionation is different for each environmental indicator (full data set in the supplementary information). As both have a significant contribution, it is important to include both the environmental impact of cultivation and fractionation when selecting the most sustainable ingredients. Evidently, the place and method of cultivation highly influence the environmental indicators as different countries have different climates and distances to the processing location. This could result in a different outcome in a case study with different cultivation locations. Nevertheless, the comparisons made in this case study are to emphasize the impact of cultivation compared to processing.

5.4. Environmental impact of the production of 1 kg of protein

Earlier, it was shown that a lower degree of refining decreases the environmental impact of the processes and hence, also of the embedded environmental impacts in the food ingredients. Moreover, we showed in the previous section that both the impact of cultivation, transportation, and processing of the crops should be included in any sustainability assessment. One should however bear in mind that a lower degree of processing results in fractions with a lower protein purity, which is considered a key component in food ingredients. Therefore, instead of using the amount of processed raw material as a functional unit, the global warming potential of the fractionation processes can also be expressed per kg of protein within that ingredient, now including the impact of cultivation and transportation (Fig. 6). The other environmental impact indicators expressed per kg of protein are presented in the supplementary information and follow the same reasoning as the global warming potential.

The conversion from total mass to protein as a functional unit is inversely related to the environmental indicators due to the low purity of the milder fractionated ingredients. Nevertheless, a lower degree of refining can still deliver the same amount of protein with a lower footprint (Fig. 6). The carbon footprint to produce conventional yellow pea protein isolate is 5.3 kg CO_2-equivalents/kg protein and 4.9 kg CO_2-equivalents/kg protein for mild aqueous fractions. The fine fraction has the lowest climate change potential with 1.6 kg CO_2-equivalents/kg protein. The global warming potential of hybrid protein-rich fractions is 5.1 kg CO_2-equivalents/kg protein. The impact of the hybrid protein fractions surpasses the impact of the mild aqueous protein fractions. This is due to a matter of impact allocation to the protein/fibre-rich stream and the starch stream in the first decanter. As the starch-rich fraction produced from the fine fraction is smaller than the starch-rich fraction that originates from yellow pea flour, relatively more impact is allocated to the hybrid protein-rich fractions. Moreover, the global warming potential of the protein-rich fractions from lupine is slightly higher compared to yellow pea. The conventional fractionation of lupine requires oil removal and more water compared to the fractionation of yellow pea. The excess water also needs to be evaporated, which requires additional energy. The climate change potential for the conventional protein isolate of lupine is 5.8 kg CO_2-equivalents/kg protein, while the milder aqueous variant is

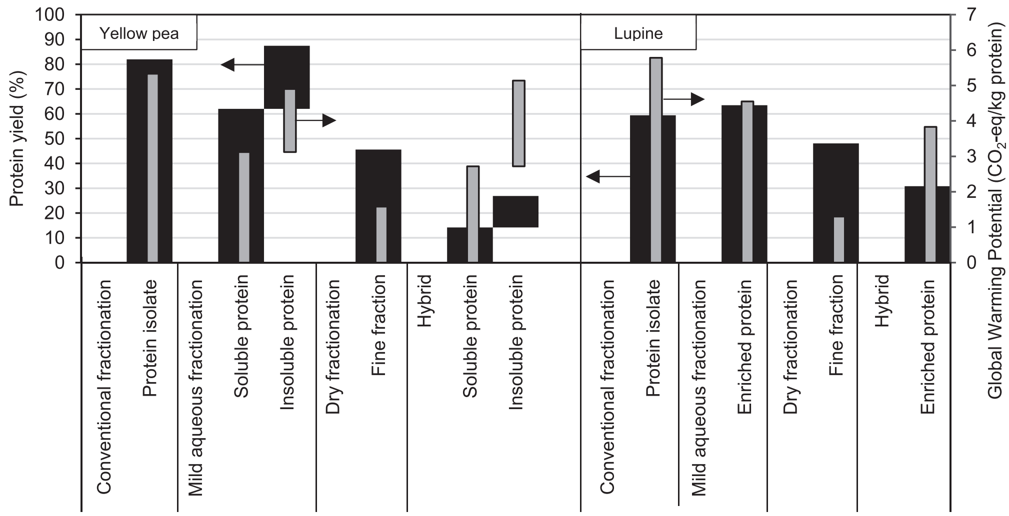

Fig. 6. Protein yield (protein in the ingredient as a percentage of the protein in the crop flour) (left axis in black) and the global warming potential (right axis in grey) to produce 1 kg of protein in each fraction.

4.6 kg CO_2-equivalents/kg protein. The dry fractionation again has the lowest carbon footprint, of 1.3 kg CO_2-equivalents while the hybrid method is 3.8 kg CO_2-equivalents per kg protein. This proves that the mildly fractionated ingredients have the potential to deliver as much protein as the conventional way, but with a reduced carbon footprint. The milder fractions also have a lower carbon footprint than for example soy protein isolate, which was estimated by Thrane et al. (2017) 6.1 kg CO_2-equivalents/kg protein.

Comparing the impact between different processes and fractions, one should realize that the impact per fraction (e.g. starch isolate and protein isolate) is dependent on the allocation of the process steps and the impact of cultivation. More specifically, for conventional fractionation, the impact of cultivation is allocated according to the yield of the three fractions; the protein-, fibre-, and oil/starch-rich fractions. For dry fractionation the impact is allocated to only the fine and coarse fractions, meaning a relative higher cultivation impact per fraction. It is however still debatable whether the fibre fraction from conventional fractionation is considered to be food grade; hence, a useful fraction. The removal of such a fraction from the allocation will increase the final impact of each fraction. Therefore, the allocation to valuable fractions should always be considered carefully, either based on mass or economical value.

Next to the reduction of the global warming potential during the production of food ingredients, it is evident that for a sustainable food chain a minimal loss of valuable components is essential. Even though conventional fractionation of yellow pea delivers a high protein yield of 82% (Fig. 6), all other protein originally present is lost. Even if they could be recovered, they are not food grade anymore. In contrast, the protein-rich ingredients from mild aqueous fractionation of yellow pea combined have a higher protein yield of 87%, assuming that both the insoluble and the soluble proteins are recovered. The rest of the proteins end up in the starch fraction and could still be used for human consumption. A similar relation is found for the fractionation of lupine. The protein yield of mild aqueous fractionation of lupine is 64%, compared to conventional fractionation of 59%, with a lower impact. These findings indicate that besides the lower global warming potential of the milder alternatives to conventional fractionation, they also deliver a higher protein yield.

Dry fractionation features a protein yield of 100% for both fractions. It is assumed that no fractions are degraded because the milled flour is just separated into two fractions through an air classifier. On top of that, the global warming potential is very low for this method. However, the downside of this method is the low purity that is obtained; with a significant amount of protein ending up in the protein lean coarse fraction. A method to increase the purity was proposed by Berghout et al. (2015), by further purifying the fine fraction using mild aqueous fractionation. Interestingly, combining dry fractionation and mild aqueous fractionation for yellow pea only slightly changes the purity of the fine fraction from 43% to 62 and 37% for the soluble and insoluble protein fraction respectively. As the fine fraction is the starting material of the second step of the hybrid fractionation, the protein yield compared to the initial crop is approximately 15 and 13% for the soluble and insoluble protein fraction, respectively. Next to the rather slight increase in purity with a low total protein yield of the hybrid fractions, the hybrid fractionation comes at a cost of 3.5 CO_2-equivalents/kg of protein extra. One should however realize that no protein is lost during both mild fractionation techniques, since the proteins in the coarse fraction can be utilized completely in food, as opposed to conventional fractionation. In addition, the combined method is still lab-based and can be optimized. The hybrid fractionation of lupine increases the purity of the fine fraction from 58 to 87%, with a protein yield of 30% (Berghout et al., 2015, *supplementary information*). While this looks very promising, the values are uncertain since it is calculated theoretically by the author. The same distribution of mild aqueous fractionation was used, starting with lupine flour rather than the fine fraction. This might be an unjustified assumption as the fine fraction has a different composition; hence, the hybrid protein-rich fraction will have a different composition. This could also be an explanation for why the carbon footprint of the protein-rich hybrid fraction does not surpass the mild aqueous protein-rich fraction as is the case with yellow pea. Therefore, we conclude for now that for yellow pea, hybrid fractionation can slightly increase the purity of the fine fraction after dry fractionation, yet, this comes at a high cost in terms of global warming potential. For lupine, the situation may be the opposite, but more research is required. Overall, the use of protein as a functional unit in this sustainability assessment indicates that a lower degree of refining can deliver as much protein as the conventional fractionation, with a lower footprint. On top of that, milder methods have a higher protein yield, in which close to no protein is lost.

6. Conclusion

This study quantified the environmental impact of several degrees of refining for food ingredients. The environmental impact of the production of plant protein-rich ingredients can be reduced substantially by decreasing the degree of refining, which we assessed for yellow pea and lupine. Overall, processing is a very important element in the total environmental impact and in some cases larger than that of the crop cultivation. Therefore, any assessment should include both. The omission of the protein precipitation step in mild aqueous fractionation significantly decreases the global warming potential, human carcinogenic toxicity, water consumption, and fossil resource scarcity by 30–40%. Moreover, the removal of the oil extraction step in the fractionation process of oil-rich seeds such as lupine decreases these impact categories by 20–30%. The final drying steps remain mostly responsible for the impact in all categories. The largest decrease in global warming potential was obtained with only dry processing with a decrease of up to 93% compared to the conventional way of fractionation. With the amount of recovered protein as the functional unit, the processes that refine less still have a lower impact compared to the conventional isolation method. From all ingredients, the conventional method gives the highest protein yield per protein-rich fraction, although at the costs of a higher environmental impact. In general, the consideration of purity, yield, and environmental indicators offer more insight into the process of choosing the right ingredients, which will benefit the sustainability of the food chain.

CRediT authorship contribution statement

Anouk Lie-Piang: Conceptualization, Methodology, Formal analysis, Validation, Investigation, Visualization, Writing - original draft. **Nicoló Braconi:** Methodology, Formal analysis, Resources, Writing - review & editing. **Remko M. Boom:** Supervision, Conceptualization, Visualization, Writing - review & editing, Project administration. **Albert van der Padt:** Supervision, Conceptualization, Visualization, Writing - review & editing, Project administration.

Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2021.126046.

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