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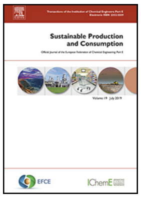

Research article

Substituting wheat with chickpea flour in pasta production delivers more nutrition at a lower environmental cost

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ABSTRACT

The modern food system is characterised by 1) unsustainable agricultural practices, heavily dependent on agrochemical inputs and leaking large amounts of reactive nitrogen (N) whilst degrading soils, and 2) the consumption of energy-rich but nutrient-poor foods, contributing to non-communicable diseases related to malnutrition. Substituting cereals with low-input, protein- and fibre-rich legumes in the production of mainstream foods offers a promising solution to both issues. Chickpea (*Cicer arietinum*) is a leguminous crop that can be grown with little or no synthetic N fertiliser. We performed life cycle assessment (LCA) to compare the environmental footprint of pasta made from chickpeas with conventional pasta made from durum wheat (*Triticum durum*) from cradle to fork. Two functional units were used, an 80g serving of pasta, and a Nutrient Density Unit (NDU). Environmental burdens per serving were smaller for chickpea pasta across at least 10 of the 16 impact categories evaluated. Global warming, resource use minerals and metals, freshwater eutrophication, marine eutrophication, and terrestrial eutrophication burdens were smaller than those of durum wheat pasta by up to 45%, 55%, 50%, 86%, and 76%, respectively. Cooked chickpea pasta contains 1.5 more protein, 3.2 times more fibre and 8 times more essential fatty acids than cooked durum wheat pasta per kcal energy content. Thus, the environmental advantage of chickpea pasta extended to 15 of the 16 impact categories when footprints were compared per unit of nutrition. Global warming, resource use and eutrophication burdens per NDU were 79–95% smaller for chickpea pasta than for durum wheat pasta. The one major trade-off was land use, where chickpea pasta had a burden 200% higher per serving, or 17% higher per NDU, than wheat pasta. We conclude that there is high potential to simultaneously improve the environmental sustainability and nutritional quality of food chains through simple substitution of cereals with legumes in staple foods such as pasta. Breeding and agronomic management improvements for legumes could reduce the yield gap with cereals, mitigating the land use penalty. Meanwhile, the higher protein content of chickpea pasta could contribute towards wider environmental benefits via animal protein substitution in diets, and merits further investigation. Consumers who look for the traditional taste and texture of wheat pasta can achieve these aspects by cooking the chickpea pasta *al dente* and combining it with a typical pasta sauce, which will hide its subtle nutty taste.

1. Introduction

The global food sector faces a major challenge to deliver sustainable nutrition. Intensive agricultural practices adopted to meet growing food demand have driven massive anthropogenic pressures on the Earth's ecosystems, notably via land occupation, fertiliser use and animal-related greenhouse gas (GHG) and ammonia emissions (Steffen and Sorlin, 2015). Synthetic Nitrogen Fertiliser (SNF) use causes significant environmental and economic damage, as its production is resource-intensive, and over-application of SNF causes N leaching and ammonia emissions to air, degrading the

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quality of air, water, and soil (Sutton et al., 2011), changing the climate and promoting biodiversity loss (Mozumder, Berrens, 2007). In terms of cereal production, the use of SNF represents a major source of greenhouse gas emissions to the atmosphere, with typical farm gate values per kg of grain of the order of 0.50 kg CO_2e for oats, wheat and barley (Williams et al., 2020). To put this in perspective, the cultivation of grain legumes (faba bean, chickpea and pea), where nitrogen is provided by biological fixation of atmospheric nitrogen by bacteria present in root nodules, produces typical farm gate emission values of the order of 0.18 kg CO_2e (Williams et al., 2020). Accumulation of this biologically fixed N in plants boosts yields (Peoples et al., 2009), while the use of legumes in agriculture increases biodiversity, reduces weed invasion (Sturludóttir et al., 2014), and can enhance carbon sequestration in the soil (Peoples et al., 2019).

Grain legumes are also more beneficial than cereal grains in term of human health, providing a rich source of nutrients. The risk of type-2 diabetes and cardiovascular diseases decreases with consumption of legumes (Flight, Clifton, 2006; Jayathilake et al., 2018; Kouris-Blazos, Belski, 2016; Viguiliouk et al., 2017), while other positive effects include a reduction in the relative risk of developing myocardial infarction (Miller et al., 2017). The presence of fibre and phytochemicals in legumes lowers cholesterol and helps regulate blood pressure (Bazzano et al., 2011). Moreover, the consumption of legumes improves gut health (Clemente, Olias, 2017) and assists in managing weight (McCrory et al., 2010). Legumes are also a source of anti-cancer peptides (Luna-Vital, González de Mejía, 2018) that can potentially assist in the prevention against prostate (Park et al., 2008) and colorectal cancers (Zhu et al., 2015).

A growing demand for grain legumes for feed and food coupled with supportive policies and yield enhancements in the EU has led to a record production of legume crops in Europe in 2017-2018 (Barel et al., 2018). This demand for pulses, especially chickpeas and lentils is forecast to increase further (Barel et al., 2018). Recently, a growing demand for vegetarian products has been observed across Europe with the rise of flexitarian diets (Derbyshire, 2017; European Commission, 2018a; NatCen, 2016). Launches of vegan and vegetarian food products have increased worldwide by 140% and 21%, respectively (Statista, 2017), and highlight the opportunity to increase legume-based substitutes for staple foods. One such possibility is legume pasta, where substituting durum wheat with pulses in pasta production could dramatically expand legume cultivation and consumption in Europe, with associated environmental and nutritional benefits. Such a substitution could contribute towards crop diversification in the EU and biodiversity restoration (Zander et al., 2016).

Pasta is a popular staple food with high versatility. It is typically made from semolina flour obtained by milling durum wheat, and mixing with water, and sometimes eggs. In Europe alone, 5.4 million tonnes of pasta were produced in 2017 (Eurostat, 2018) and consumption of durum wheat pasta amounted to around 3.4 thousand tonnes per year (Statista, 2019). Despite the high variability in consumption amongst EU countries, with Italians consuming 23.5 kg of pasta per capita annually, Greeks 11.2 kg, British 3.5 kg, and Irish 1 kg (Statista, 2018) for example, pasta production and consumption are growing worldwide, with a compound annual growth rate of 4.4% projected between 2019 and 2023 (Statista, 2019).

Numerous pasta products made from ingredients other than durum wheat are appearing commercially. Major categories include other types of wheat pasta (spelt), other cereals pasta (quinoa, rice), 0-calories pasta (konjac), and legume-based pasta (edamame, adzuki, black bean, chickpea, red or green lentil, and mung bean vermicelli). Legume-based pasta also represents a food opportunity for the 1.4% individuals with celiac disease in the world, due to the legumes pasta being gluten-free (Celiac Disease Foundation, 2018). A sensory analysis of legumes pasta showed consumer acceptability of legumes pasta (Turco et al., 2019). Chickpea pasta can be cooked *al dente*, conserving a similar texture to that of traditional durum wheat pasta, and the difference in taste can be hidden with the sauces typically added to pasta dishes.

In this paper we report results from a ‘farm to fork’ analysis of the environmental burden of both durum wheat and chickpea dry pasta using Life Cycle Assessment (LCA) methodology. LCA is a defined protocol used in assessing the environmental impacts of a product system, by looking at the inputs and outputs of its life cycle (International Organization for Standardization, 2006). It has been widely used in the food sector, and is a powerful tool to support decision making when considering the sustainability of food systems (Sala et al., 2017). Despite the wealth of LCA data on food systems (Heller, Keoleian, Willett, 2013), our literature search on Google Scholar (2000-2010) revealed only two LCA studies that have considered the use of legumes in pasta: Chaudhary et al. (2018) estimated the carbon footprint and nutritional content of a partial substitution of refined wheat flour with Canadian yellow pea flour in pasta (30% pea-70% wheat flours), while Nette et al. (2016) performed a comparative LCA of pasta made with egg or pea protein. Nette et al. (2016), like in numerous other food LCAs, disregarded nutrition by using a weight-based functional unit for comparing different foodstuffs, omitting the key nutritional role of the products involved (Heller et al., 2013), and thus potentially supporting misleading conclusions on the wider sustainability of these foodstuffs. On the other extreme, Chaudhary et al. (2018) compared the Nutrient Balance Score of the food products, requiring the knowledge of 32 macro and micronutrients contents, an expensive process that can dissuade LCA practitioners to use a nutritional functional unit. The study also used nutritional content of raw ingredients, which will ultimately change when cooked (Fabbri, Crosby, 2016). Moreover, both studies found that the legume alternative had a lower global warming potential, but did not assess the foods across other impact categories. Product Environmental Footprint (PEF) guidelines (European Commission, 2018c) recommend a more comprehensive evaluation of 16 environmental impact categories when assessing the environmental sustainability of products. Furthermore, most food LCA studies have used weight-based functional units for comparing

The comparative LCA of chickpea and durum wheat pasta reported here has been assessed over the sixteen impact categories recommended by PEF Guidance (European Commission, 2018c), and has two objectives, 1) to compare the environmental burdens of a serving of cooked pasta using a conventional functional unit of 80g dry weight of pasta, cooked), and 2) to compare the same but use a functional unit based on the nutritional density of the pasta serving, as proposed by Van Dooren (2016).

2. Materials and methods

2.1. Goal, scope, and boundary definition

This LCA study is a comparative assertion of the overall environmental burden from cradle to fork arising from the consumption of chickpea pasta and conventional durum wheat pasta. The open source software OpenLCA 1.10.2 (GreenDelta, 2019) was used to calculate the environmental footprint of the two pasta products, using Agrifootprint 3.0 (Durlinger et al., 2017) and Ecoinvent 3.6 (Wernet et al., 2016) international databases. Inventory data on chickpea pasta were collected specifically for this study from Variva Ltd. (www.variva.bg, 2019), the Bulgarian manufacturer of chickpea pasta Variva®. Data on durum wheat pasta manufacture were adapted from Bevilacqua et al. (2007) and modelled as though the durum pasta was manufactured in Bulgaria to make the geographical origin of the two products identical.

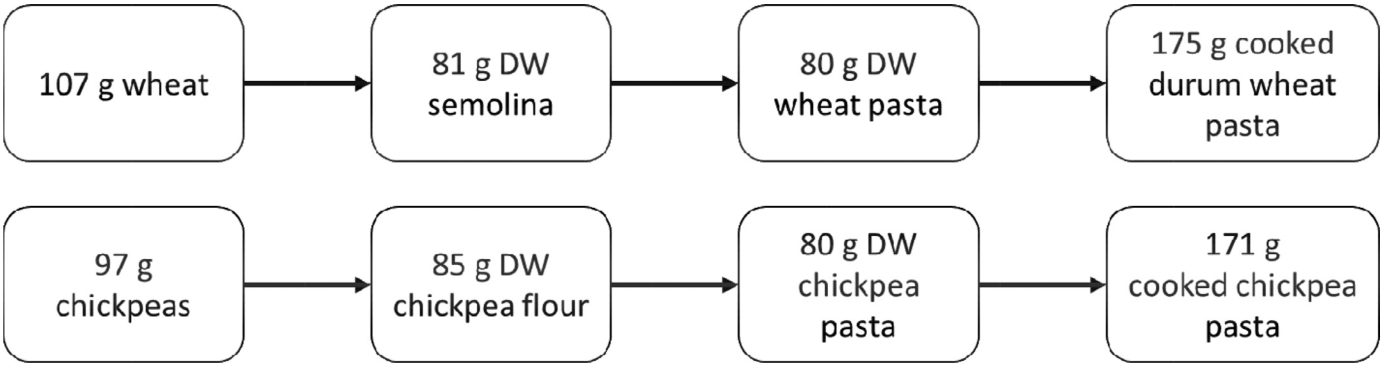

Figure 1. Mass balance flow of chickpea and durum wheat pasta.

In order to represent the differing nutritional profiles of the two types of pasta in the LCA analysis, two functional units were used; 80 grams dry weight pasta, cooked (a mass-based FU) and one NDU. The mass-based functional unit is referred to as a serving in this paper. The NDU represents the integration of nutrient density as a comparative basis between the chickpea and wheat pasta in the LCA, such that the environmental burdens are quantified per integrated content of protein, essential fatty acids, fibre, and calories. The reference flow was defined as a cooked portion of pasta. The mass balance flow for both pastas is represented in figure 1.

An attributional LCA was performed with economic allocation for harvested wheat straw and wheat feed co-products from wheat semolina production. Allocation factors were of 92.5% for wheat grains and 7.5% for wheat straw; 84% for semolina and 16% for wheat feed (EPD International, 2016; European Commission, 2018b). Small amounts of biowaste obtained from chickpea sorting and cleaning were assumed to be discarded in the field, although they could potentially be used as a soil conditioner (which could reduce the environmental burden of chickpea pasta, thus the applied assumption of allocating all burdens to chickpea pasta is conservative). To ensure compatibility between the LCA of durum wheat and chickpea pasta, a second-order approach was used, where the system boundaries included all stages of the life cycle from cradle to fork. Figure 2 illustrates both the system boundaries used and manufacturing steps for the cradle to fork assessment of chickpea and durum wheat pasta value chains. In accordance with the ILCD handbook (JRC, 2010), identical product use was considered, the same life cycle stages were included, and the inventory data had roughly matching degrees of accuracy. The LCA methodology followed PEF guidelines (European Commission, 2018c) as far as possible, excluding end of life, in line with the need to harmonise approaches for improved transparency and comparability. The recycling stage was not modelled, as the focus of the study was to compare the differences between pasta made with different raw materials, wheat or chickpea. Therefore, including recycling for the same packaging would not have contributed any useful differentiation in the study.

Results obtained from running the impact assessment of the LCA procedure were normalised by annual person equivalents, using the factors recommended in the PEF guide (European Commission, 2018c). This was done to facilitate interpretation of impact scores by providing a joint reference impact (Benini et al., 2014).

Four scenarios were assessed in this study to compare different assumptions about cultivation of the wheat and chickpeas used to make the pastas:

1) *Wheat (0% straw)* - pasta made from durum wheat with none of the wheat straw harvested during field operations
2) *Wheat (80% straw)* - pasta made from durum wheat with 80% of the wheat straw harvested during field operations, and cultivation burdens economically allocated between grain (for pasta) and straw, with a 7.5% allocation factor for wheat straw and 92.5% for wheat grain (EPD International, 2016)
3) *Chickpea (Bulgaria)* - pasta made from chickpeas using a Bulgarian case study, with 100% chickpea cultivation residues remaining in the field
4) *Chickpea (Spain)* - pasta made from chickpeas using the Bulgarian case study for all steps but cultivation, which was modelled from a Spanish case study, representing chickpea cultivation with no added fertilisers, chemicals/pesticides based on best practice. As for the other chickpea scenario, 100% chickpea cultivation residues remained in the field

Scenarios 2) *Wheat (80% straw)* and 3) *Chickpea (Bulgaria)* are the baseline scenarios, and scenarios 1) *Wheat (0% straw)* and 4) *Chickpea (Spain)* are alternative scenarios. In scenario 2) *Wheat (80% straw)*, 80% was an estimate of the amount of above ground straw residue that is removed during straw harvesting operations, as in Lienhardt et al. (2019).

2.2. Chickpea pasta inventory

Post-farm gate data were provided by Variva Ltd., and cultivation data provided by the main grower supplying Variva Ltd. with chickpea. Chickpea cultivation was modelled with emission factors of the IPCC 2019 guidelines; N content of ground residues were of 0.008 N for below-ground residues, and the same amount for above-ground residues per hectare (IPCC, 2019). Amount of NPK applied per hectare and yield (1 820 kg.ha^{-1} dry matter) were obtained from the farmer working with Variva Ltd. Distribution of fertiliser types specific to Bulgaria was extracted from the International Fertilizer Association (2020). Field activities included from Ecoinvent 3.6 processes were sowing, tillage and ploughing, fertilising, and harvesting. Two applications of fertiliser were assumed. Due to the high variability in crop protection application, the difficulty of finding trustworthy sources that describe which type and what quantities are needed, and the fact that amounts are usually much smaller than those of fertilisers, crop protection was excluded from the study. Lime application was fixed for all scenarios at 400 kg/ha, as a corrector of acidity. This was a conservative approach, as lime is typically applied to prevent soil acidification as a result of the application of ammonium-based fertilisers (Defra, 2010), which are applied in greater amounts in cereal crops than to legume crops.

Direct field emission of nitrous oxide - from crop residues remaining on field and synthetic N fertilisers (SNFs) – were calculated following equation 11.2 of IPCC (2019). Direct emissions of nitrogen oxides resulting from the application of SNF were modelled according to Nemecek and Kägi (2007), while ammonia emissions from the volatising N fraction of SNF were modelled accord-

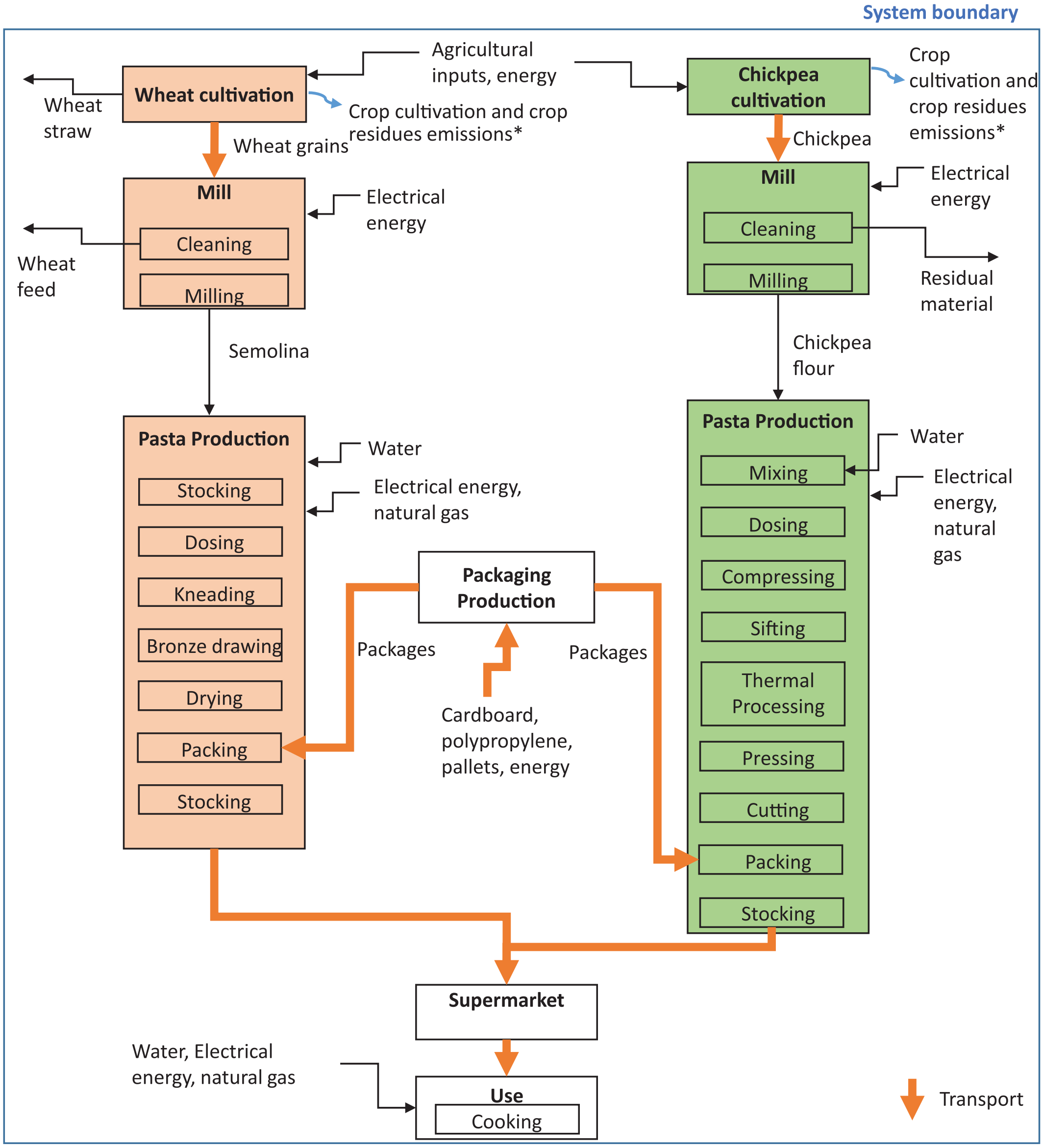

Figure 2. System boundary of chickpea pasta, from cradle to fork. The manufacturing steps of chickpea pasta are indicated in green (right side), those of durum wheat pasta in red (left side), and those shared by the two products are white (middle). For more information on these emissions, see the following section.

ing to IPCC (2019). Indirect emissions of nitrous oxide resulting from a) the volatilisation of SNF and b) from SNF and on field crop residues leaching were calculated with equations 11.9 and 11.10 of IPCC, respectively (IPCC, 2019). Indirect emissions of nitrate losses to water were determined following Reckling and Bachinger (2016), and those of CO_2 emissions from lime and urea applications were calculated according to IPCC (2006). Finally, indirect emissions of phosphorus losses to water from the use of synthetic P fertilisers were determined with the cropping system loss coefficients, as in Styles *et al.* (2015).

Chickpeas were harvested and transported at most 50 kilometres to the cleaning facility. The sorting machine consumed 40Wh per kg, and 5-10% of the material was separated as biowaste. The remaining 90-95% of clean chickpeas were then thermally processed in a 1000 kg capacity oven, transported 150 kilometres to the milling plant where the obtained flour was mixed with water, pressed, cooked in a pasta oven, and finally packaged.

The fertiliser applications in the Bulgarian scenario, *Chickpea (Bulgaria)*, were likely used for boosting yields, though chickpea cultivation guides indicate that high use of nitrogen fertilisers is needless, and may be detrimental to chickpea growth by inhibiting nodulation and atmospheric nitrogen fixation (Corp et al., 2004). Moreover, in some areas, use of phosphorus fertiliser is not required (Corp et al., 2004; GRDC, 2012). Consequently, an additional scenario, *Chickpea (Spain)*, was modelled, using yields from a Spanish field experiment for which no fertiliser, insecticides or pesticides were added. High variability of yields was observed in the Spanish case study, ranging from 0 to 3520 kg.ha^{-1} chickpeas dry matter across the sixteen plots studied. This high variance was due to fungal pathogen ascochyta blight. The average yield, 2014.5 kg.ha^{-1} dry matter was selected for the *Chickpea (Spain)* scenario.

The amount of energy and volume of water used for cooking were determined by adapting data from PEF guidelines for dry pasta (European Commission, 2018b) and information provided by Variva® pasta (variva.bg, 2018). Calculations are listed in Table SI.2 for both durum wheat and chickpea pasta. The energy required for cooking for chickpea pasta is less than wheat pasta, due to the shorter cooking time required (6 minutes versus 11). Adding salt was not mentioned in the chickpea pasta cooking recommendations. However to maintain a consistent comparison, it was assumed that the same amount of salt was added in cooking both products. Electric cooking was modelled with a flow specific to Bulgarian electricity. All of the cooking water was assumed to go to residential wastewater treatment. Information on packaging was provided directly by Variva Ltd.

2.3. Durum wheat pasta inventory

Data for durum wheat pasta cultivation was obtained from the same farm in Bulgaria that supplies the chickpeas, using a combination of Agrifootprint 3.0 and Ecoinvent 3.6 processes. Data for durum wheat pasta manufacturing was adapted from Bevilacqua et al. (2007) using Ecoinvent 3.6 processes. Inputs and outputs for all processes involved in the delivery of an 80 g dry weight serving of *al dente* – a cooking technique in which pasta is still firm - cooked pasta, from cradle to fork, were recorded and are included in full in Table 1. Relevant background processes for chickpea and durum wheat pasta production were extracted from LCA databases Ecoinvent 3.6 and Agrifootprint 3.0.

Agricultural data for both wheat and chickpea production came from the same farm in Bulgaria. Amount of NPK applied per hectare and yield (5 340 kg ha^{-1} dry matter) for wheat were obtained from the farmer working with Variva Ltd. As for chickpea cultivation, wheat cultivation was modelled with emission factors of the IPCC 2019 guidelines based on N contents of above- and below-ground residues, with 0.006 and 0.009 kg N per kg residue, respectively (Liang, Noble, 2019). The breakout of fertiliser types specific to Bulgaria was extracted from the International Fertilizer Association (2020) since the specific fertilisers used in the farm assessed was unknown; one application of fertiliser was assumed along with 400 kg of lime per hectare. Data were collated for the two wheat scenarios, 1) *Wheat (0% straw)* and 2) *Wheat (80% straw)*. Following Product Category Rules (PCR) for arable crops, economic allocation was performed for allocating the burdens between wheat grain and wheat straw during the cultivation phase (EPD International, 2016). Wheat straw and wheat grain co-products were assigned 7.5% and 92.5% of cultivation environmental burdens respectively.

Durum wheat is transported after cultivation to a milling facility. Milling uses 70 Wh of electricity, 10 J of natural gas, and 0.02 L of water for 110 g of durum wheat, producing 80 g of semolina, and 30 g of co-product used as cattle feed. The semolina is transported to another facility where it is dosed, kneaded (using 0.02 L of water per 80 g of semolina), bronze drawn (shaped), and then dried. These manufacturing steps account for 3.02 Wh of heat gas and 13 Wh of electricity, 80 J of natural gas and 60 J of crude oil for the FU. The dried pasta is packaged in cardboard boxes of 250 g and transported to a retail centre and subsequently to the consumer. Transport modes, distances, and weights of products transported within Bulgaria, within Europe, and outside Europe, were assumed to be the same for durum wheat pasta and chickpea pasta so as to avoid any false differentiation between pasta types based on factors independent of the main flour type. Transport modes and distances were modelled following the PEF guidelines (European Commission, 2018c). Based on 2018 data provided by Variva Ltd, 20% of Variva® chickpea pasta produced is sold within Bulgaria, 20% is sold in Europe (excluding Bulgaria), and the remaining 60% is sold in Turkey. The distance between the centroids of Bulgaria and Turkey was determined to be 1162 km with Google Maps. Based on the PEF guidelines, consumer transport to the supermarket included product volume in the equation to determine the share of the product in the shopping environmental burdens. 80 grams of raw pasta are then cooked in 0.8 L of boiling water for 11 minutes with 5 grams of salt (Barilla, 2018; European Commission, 2018b).

Durum wheat and semolina are very similar in terms of energy and protein content (USDA, 2020). Therefore, we assumed that co-products of semolina production were used as wheat-based feed substitutes. Following the product category rules of uncooked pasta, economic allocation was performed for the milling stage, and the co-products semolina and wheat for feed were assigned 84% and 16% of upstream burdens respectively (EPD International, 2010). Because co-products were in small quantities, and because the wheat feed co-products from semolina production are likely to largely replace similar wheat-derived feeds, system expansion was deemed not relevant to perform in this case. For simplification, apart from stated co-products and waste flows, no loss was assumed during any of the life cycle stages of either product. 80 g of dried pasta was used as the reference flow, assuming the same packaging for both pasta products based on packaging information collected from Variva Ltd.

2.4. Nutrient Density Unit (NDU)

A key function of food is to deliver nutrition to the body. LCAs of food products which are based solely on weight FUs do not take this into account, and make inter-food comparisons difficult. LCAs which use a protein FU are still problematic in that nutrition is far more complex than just protein delivery, and that in Europe, daily protein intake is above recommended levels (Westhoek H. et al., 2016). Therefore, in this study, the Nutrient Density Unit (NDU) developed by Van Dooren (2016) was selected a good proxy (see Discussion).

Nutritional composition of both cooked pastas was obtained by nutritional analysis of Variva® chickpea pasta and of durum wheat pasta from the supermarket. Analyses of protein using the Kiedahl method (ISO 1871:2009) (ISO, 2009), energy following the EU regulation 1169/2011 (European Union, 2011), fibre by Enzymatic-Gravimetric Method from the AOAC 991.43 and AOAC 985.29 (Lee, Prosky, Vries, 1992; Prosky et al., 1985), and essential fatty acids through gas chromatography (FID) from ISO 12966-1:2014; 12966-2:2011; 12966-3:2016 (ISO, 2011, 2014, 2016) were performed. Random replicates were performed to 7% of the analysis by a credited laboratory that follows standard, verified, and certificated protocols, and that work mostly with industry. Nutritional characteristics of the two pastas are summarised in Table 2.

The Nutrient Density Unit (NDU) was applied following Van Dooren's 2016) formula ((1).

$$\mathrm{NDU} = \frac{\left(\frac{\mathrm{EFA}}{\mathrm{DV_{EFA}}}\right) + \left(\frac{\mathrm{Protein}}{\mathrm{DV_{prot}}}\right) + \left(\frac{\mathrm{Fibre}}{\mathrm{DV_{fibre}}}\right)}{3 \times \left(\frac{\mathrm{S_i}}{2000\ \mathrm{kcal}}\right)} \tag{1}$$

Where:

EFA is the amount of essential fatty acids in 100g of product, expressed in grams.

Protein is the amount of protein in 100g of product, expressed in grams.

Fibre is the amount of fibre in 100g of product, expressed in grams.

DV_{EFA} is the recommended daily value intake of essential fatty acids, expressed in grams.

DV_{prot} is the recommended daily value intake of protein, expressed in grams.

DV_{fibre} is the recommended daily value intake of fibre, expressed in grams.

S_i is the amount of kilocalories in 100g of product, expressed in kilocalories.

$$NDU_{variva\ pasta(cooked)} = \frac{\frac{1.6}{12.4} + \frac{8.1}{50} + \frac{5.7}{25}}{3 \times \frac{150}{2000}} = 2.3 \tag{2}$$

Table 1
Inventory of inputs and outputs for an 80 g dry pasta serving of *al dente* cooked chickpea or wheat pasta.

Stage	Input/output/process	Units	Wheat		Chickpea (Bulgaria)		Chickpea (Spain)	
			Input	Output	Input	Output	Input	Output
Cultivation	Fertiliser –N	kg	0.0040		0.0016			
	Fertiliser – P_2O_5	kg	0.0025		0.0052			
	Fertiliser – K_2O	kg	0.0024		0.0077			
	Urea	kg	0.0021		0.0009			
	Lime	kg	0.008		0.008		0.008	
	SO_3	kg	0.001					
	Energy, diesel burned in machinery	MJ	0.13		0.04		0.04	
	Seed	kg	0.004		0.004		0.004	
	Land	m_2	0.20		0.53		0.48	
	Chickpeas (dry matter)	g				97		97
	Wheat straw	g		148 / 0*				
	Wheat grain (dry matter)	g		110				
Flour production	Transport 16-32 t lorry (200km)	kg.km	21.27		18		18	
	Electricity	Wh	70		17.4		17.4	
	Organic residual material	kg				0.005		0.005
	Chickpea flour	kg				0.08		0.08
	Water	L	0.0200		0.0200		0.0200	
	Natural gas	J	10					
	Semolina	kg		0.08				
	Wheat grain, feed	kg		0.03				
Pasta production	Machines electricity	Wh	13		16		16	
	Water	L	0.0200		0.0242		0.0242	
	Heat, other than nat. gas	Wh	3.02		19.73		19.73	
	Nat gas- thermal energy	MJ	0.08					
	Heat and power co-generation, oil	MJ	0.06					
Packaging	Film, low density PET	g	1.6		1.6		1.6	
	Kraft paper, unbleached	g	4.5		4.5		4.5	
	Folding boxboard	g	4.5		4.5		4.5	
	Polypropylene, granulates	g	0.0029		0.0029		0.0029	
	Extrusion, plastic film	g	0.0029		0.0029		0.0029	
	Trans PP film >32 t lorry	kg.km	7.40E-4		7.40E-4		7.40E-4	
	Corrugated board box production	g	3.7		3.7		3.7	
	Flat pallet	Unit	8.53E-6		8.53E-6		8.53E-6	
	Transport of boxes >32t lorry	kg.km	2.11		2.11		2.11	
	Packaging electricity	Wh	1.2		1.2		1.2	
	Printing ink, offset	g	0.0149		0.0149		0.0149	
Transport factory-retail-consumer								
	Transport factory-DC (BG) lorry 3.5-7.5 t	kg,km	19.2		19.2		19.2	
Transport factory-retail-consumer	Transport factory-DC (EU) lorry 16-32 t	kg,km	56.0		56.0		56.0	
	Transport factory-DC (outside EU) lorry 16-32 t	kg,km	55.8		55.8		55.8	
	Consumer transport by car	km	1.56E-7		1.56E-7		1.56E-7	
Cooking	Tap water	L	0.80		0.80		0.80	
	Boiling and cooking electricity	kWh	0.694		0.394		0.394	
	Salt	g	5		5		5	

*Depending on whether wheat straw was harvested or not, with 148 g belonging to scenario 2) and 0 to scenario 1).

Table 2
Energy, protein, dietary fibre, and essential fatty acids content of 80 g dry weight durum wheat and chickpea pasta, cooked.

	Durum wheat pasta	Variva® pasta chickpea
Energy (kcal)	144	150
Protein (g)	5.3	8.1
Dietary fibre (g)	1.8	5.7
EFAs (g)	0.2	1.6

$$NDU_{wheat\ pasta(cooked)} = \frac{\frac{0.2}{12.4} + \frac{5.3}{50} + \frac{1.8}{25}}{3 \times \frac{144}{2000}} = 0.90 \qquad (3)$$

The energy content of the two products does not differ greatly, with 144 kilocalories per 80 g dry weight durum wheat pasta, cooked versus 150 kilocalories per 80 g dry weight chickpea pasta, cooked. The protein content of chickpea pasta is 1.5 times higher than that of durum wheat pasta (8.1 g and 5.3 g for chickpea and durum wheat pasta, respectively). The EFA content of chickpea pasta is 8 times higher than that of durum wheat pasta (1.6 g and 0.2 g for chickpea and durum wheat pasta, respectively). The fibre content is 3.2 times higher in chickpea pasta (5.7 g and 1.8 g for chickpea and durum wheat pasta, respectively). Applying these data to NDU Eq. (2) and (3) shows that chickpea pasta is 2.6 times more nutrient dense than durum wheat pasta overall – delivering 2.3 vs 0.90 NDU.

Table 3
Summary of environmental burdens for durum wheat pasta and chickpea pasta, expressed per serving and per NDU.

Impact category	Unit	Impact per serving				Impact per NDU			
		Wheat (0% straw)	Wheat (80% straw)	Chickpea (Bulgaria)	Chickpea (Spain)	Wheat (0% straw)	Wheat (80% straw)	Chickpea (Bulgaria)	Chickpea (Spain)
Acidification ter. & freshwater	mol H+ eq	0.00208	0.00198	0.00132	0.00067	0.00289	0.00275	0.000717	0.000419
Cancer human health	CTUh	1.89E-09	1.86E-09	2.23E-09	1.85E-09	2.62E-09	2.59E-09	1.21E-09	1.00E-09
Climate change	kg CO2 eq	0.207	0.196	0.163	0.114	0.287	0.272	0.088	0.062
Ecotoxicity freshwater	CTUe	0.217	0.214	0.293	0.254	0.301	0.298	0.159	0.138
Eutrophication freshwater	kg P eq	0.00019	0.00019	0.00012	9.551E-05	0.00026	0.00026	0.000065	0.000052
Eutrophication marine	kg N eq	0.00133	0.00111	0.00104	0.00019	0.00185	0.00154	0.00057	0.00010
Eutrophication terrestrial	mol N eq	0.00627	0.00587	0.00388	0.00153	0.00871	0.00815	0.00211	0.00083
Ionising radiation, HH	kBq U-235 eq	0.0424	0.0423	0.0243	0.0230	0.0589	0.0588	0.0132	0.0125
Land use	Pt	24.3	22.5	67.4	60.6	33.7	31.3	36.7	32.9
Non-cancer human health	CTUh	3.50E-08	3.43E-08	7.54E-08	6.43E-08	4.86E-08	4.77E-08	4.10E-08	3.50E-08
Ozone depletion	kg CFC11 eq	1.53E-08	1.51E-08	1.37E-08	1.14E-08	2.12E-08	2.09E-08	7.43E-09	6.17E-09
Photochem. ozone form.	kg NMVOC eq	0.00048	0.00047	0.00052	0.00042	0.00067	0.00065	0.00028	0.00023
Resource use, energy carriers	MJ	2.55	2.53	1.90	1.62	3.54	3.51	1.03	0.883
Resource use mins. & metals	kg Sb eq	1.36E-09	1.26E-09	2.12E-09	6.16E-10	1.89E-09	1.75E-09	1.15E-09	3.35E-10
Respiratory inorganics	disease inc.	1.70E-08	1.63E-08	1.12E-08	6.30E-09	2.36E-08	2.26E-08	6.06E-09	3.43E-09
Water scarcity	m^3 depriv.	0.370	0.368	0.259	0.221	0.514	0.511	0.141	0.120

2.5. Impact Assessment

The environmental burden of the four scenarios was assessed using sixteen impact categories from the PEF recommended methodology (European Commission, 2018c). Impact indicator data were normalised according to PEF guidelines as person equivalents.

3. LCIA Results

Table 3 lists the derived environmental burdens across the sixteen impact categories. Results for each of the two functional units are shown. For both functional units, scenario 4) chickpea pasta with the Spanish agronomic data has a smaller environmental burden across most categories when compared to the other 3 scenarios, and scenario 1) *Wheat (0% straw harvest)* has the highest burdens overall. Per serving, chickpea pasta from the Bulgarian case study (scenario 3) has a smaller environmental burden across 10 of the 16 environmental impact categories. However, chickpea pasta from the Bulgarian case study (scenario 3) requires more than twice the arable land than durum wheat pasta, with 0.53 m^2.yr versus 0.20 m^2.yr, thus has a land use burden that is between 2.8 and 3.0 times the one of durum wheat pasta (Table 3). This is due to significantly lower yields for chickpea than for wheat.

Per serving, chickpea pasta from the Spanish case study (scenario 4) has a smaller environmental burden across 13 of the 16 environmental impact categories when compared to both wheat pasta scenarios (scenarios 1 and 2). For the same weight of pasta, the acidification, climate change, marine eutrophication, terrestrial eutrophication and water scarcity of durum wheat pasta (scenarios 1 and 2) is between 0.7 and 6 times higher than chickpea pasta from the Spanish case study (scenario 4), respectively. However, chickpea pasta from the Spanish case study (scenario 4) still requires more than twice the arable land use than durum wheat pasta (scenarios 1 and 2), with 0.48 m^2 .yr versus 0.20 m^2.yr, respectively (Table 1), and thus has a land use burden that is up to 1.7 higher than the one of durum wheat pasta (Table 3).

When one NDU is used as the FU, the comparative environmental efficiency of chickpea pasta is improved further (Table 3). Chickpea pasta from the Spanish case study (scenario 4) generates smaller environmental burdens than durum wheat pasta per unit of nutrient density across all impact categories, except for the land use one, for which scenario 2) *Wheat (80% straw)* has the lowest burden of all scenarios. For scenario 3) *Chickpea (Bulgaria)*, all environmental burdens are smaller than those of durum wheat pasta per unit of nutrient density except for land use, which is 8-15% smaller for durum wheat pasta. The differences amongst the other impact categories are magnified, with one NDU from wheat pasta generating between 2 and 3 times more climate change, terrestrial eutrophication, and water scarcity burdens than one NDU from chickpea pasta derived from Bulgarian chickpeas, and between 3 and 10 times more in the same categories than one NDU from chickpea pasta derived from Spanish chickpeas. Therefore, to achieve the same nutrition, chickpea pasta from both scenarios (3 and 4) has a considerably lower overall environmental burden than durum wheat pasta (scenarios 1 and 2), with a small land use trade-off.

When comparing the differences between the chickpea pasta scenarios and scenario 1) *Wheat (0% straw)* with the differences between the chickpea pasta scenarios and scenario 2) *Wheat (80% straw)*, no significant change in the pattern of results was observed. Straw harvest not only “dilutes” the environmental burden of grain production, but reduces the quantity of straw residues in the field that give rise to nitrate leaching to water. In the long term, straw incorporation may increase soil organic carbon, though this effect is dependent on factors such as average C:N ration of soil organic matter, climate, soil type, etc. and is beyond the scope of this study.

Figures 3.A. and 3.B. illustrate the environmental burdens normalised per person equivalents as described in Section 2. Figure 3.A. refers to the environmental burdens per serving. Here, we can see the same picture of environmental impact differences between the two products than in the first two result columns of Table 3. Both pasta types contribute relatively more

Figure 3. Environmental burden of cooked chickpea and durum wheat pasta across 13 impact categories, using as a FU: **A.** weight: serving **B**. NDU. Human toxicity-related impact categories were excluded from the graphs, following PEF recommendations (JRC, 2018). Results are normalised per person equivalents.

to the global burdens of freshwater eutrophication, and chickpea pasta to land use. The contribution of one serving of wheat pasta to an average person's annual freshwater eutrophication footprint is nearly times higher than the contribution to an average person's climate change footprint. For a serving of chickpea pasta, it is around 2.5 times higher. Concerning land use, the contribution of one serving of chickpea pasta from Bulgaria to an average person's annual land use footprint is 2 times higher than the contribution to an average person's climate change footprint. For a serving of chickpea pasta from Spain, it is 3 times higher.

Figure 3.B. refers to the environmental burdens per NDU. This graph represents the same broad picture as the last four result columns of Table 3, but in person equivalents. Per NDU, chickpea pasta (scenarios 3 and 4) has a lower environmental burden than durum wheat pasta (scenarios 1 and 2) across all fourteen impact categories displayed in Figure 3.B, excepted for land use, for which the impact is similar. The gap in every other category is greatly widened when comparing with Figure 3.A.

A second aim of attributional LCA is to identify improvement opportunities within a product life cycle. Hotspots in the production chain of chickpea pasta from scenario 3) *Chickpea (Bulgaria)* were identified by examining the burden of each life cycle stage for all impact categories, and were recorded in process contribution graphs (Figure 4). Scenario 2) *Wheat (80% straw)* process contributions are presented as a comparison.

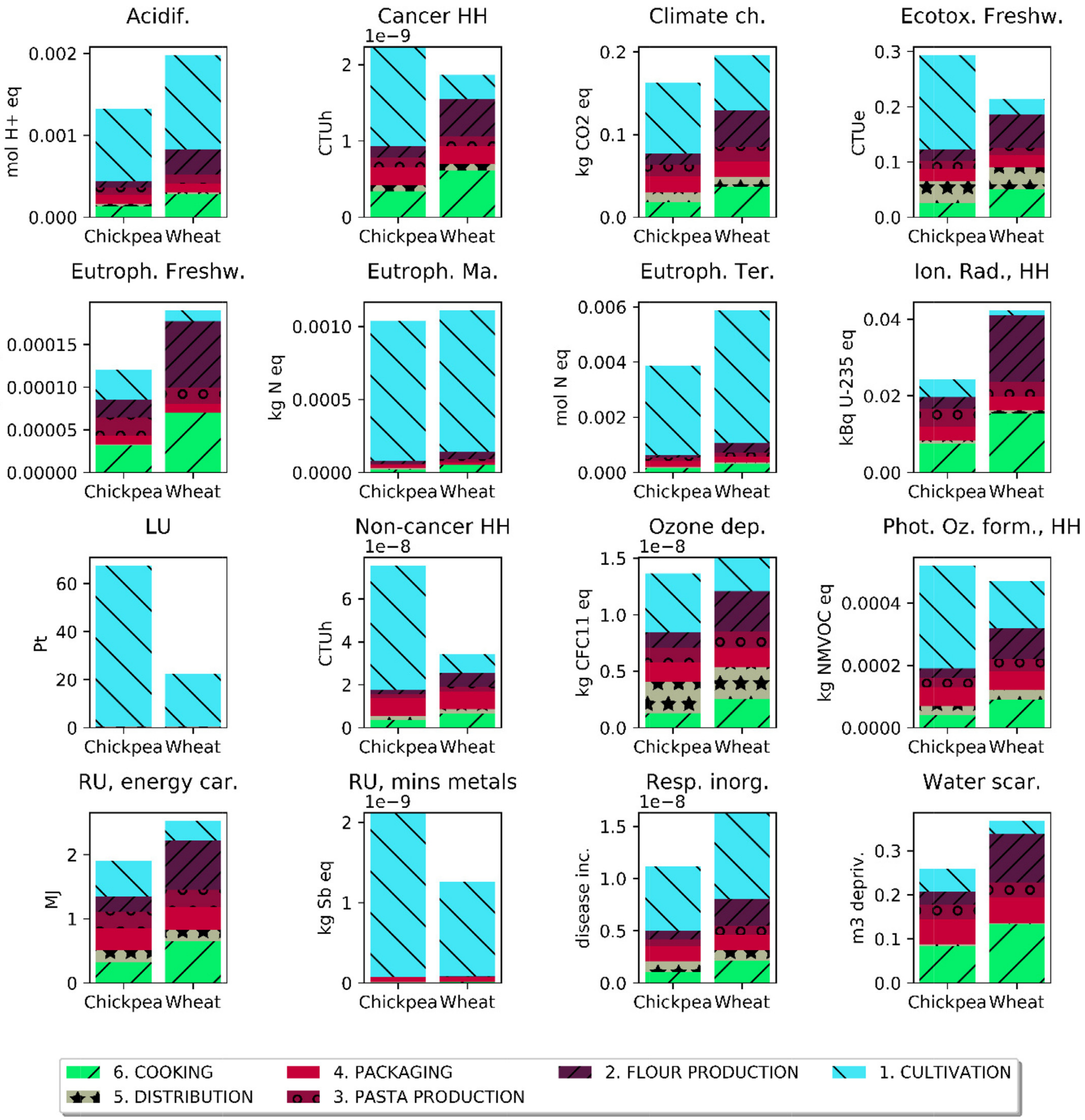

Figure 4. Process contributions of scenario 3) *Chickpea (Bulgaria)* and scenario 2) *Wheat (80% straw)* across 16 impact categories.

3.1. Agricultural stage

The agriculture stage is the major hotspot of the life cycles of both pastas, contributing to at least 20% of total burdens in 15 out of 16 impact categories for scenario 3) *Chickpea (Bulgaria)*, and 10 out of 16 categories for scenario 2) *Wheat (80% straw)*. The agricultural stage in scenario 3) *Chickpea (Bulgaria)* is responsible for more than half of the total GHG, acidification, toxicity, eutrophication marine and terrestrial, land use, photochemical ozone formation, minerals and metals use, and respiratory inorganics emissions of the whole life cycle. A significant proportion of these burdens is due to fertiliser production and application. Acidification was mainly caused by ammonia emission from N fertiliser application. GHG emissions from agriculture are caused mainly by emission of dinitrogen monoxide, mainly from direct N_2O emissions arising from crop residue and fertiliser N inputs. Marine and terrestrial eutrophication is mostly caused by nitrate emission to water and air, from applied N fertilisers and residues.

Chickpea cultivation causes scenario 3) *Chickpea (Bulgaria)* to have a higher environmental burden than durum wheat pasta across the resource use minerals and metals, land use, photochemical ozone formation, and toxicity-related categories. Around a third of the freshwater ecotoxicity burden *of* scenario 3) *Chickpea (Bulgaria)* comes from the production of chickpea seeds. In the land use category, chickpea cultivation has a higher burden because of its much lower yields than durum wheat, with chickpea yields of 1 820 kg DM/ha and 2 015 kg DM/ha in the Bulgarian and Spanish case studies, respectively, compared to wheat yields of 5 340 kg DM/ha. On the other hand, wheat cultivation is responsible for the higher environmental burden of durum wheat pasta compared with chickpea pasta in the terrestrial eutrophication and respiratory inorganics categories. This is mainly due to wheat cultivation releasing ammonia and nitrogen oxides to the air from use of N fertilisers.

3.2. Cooking

Cooking was identified as the second major hotspot behind cultivation, contributing to more than 20% of total burden across 3 impact categories in scenario 3) *Chickpea (Bulgaria)* and across 6 in scenario 2) *Wheat (80% straw)*. In the Climate Change category, cooking is responsible for nearly 20% of the total burden of 2) *Wheat (80% straw)* mainly due to electricity usage, which is higher than that of cooking chickpea pasta, due to the comparatively longer cooking time. Electricity production and the treatment and distribution of tap water in Europe are responsible for the water scarcity burdens from cooking.

3.3. Milling

The high burden of freshwater eutrophication of scenario 2) *Wheat (80% straw)* is due to the high use of electricity in milling. The process “market for electricity, medium voltage- Bulgaria” from Ecoinvent 3.6 was used. This mix relies mainly on lignite, which also remains important in countries such as Germany and Poland. The freshwater eutrophication burden of the Bulgarian electricity mix is around 4.2 times higher than the UK’s, and around 1.5 times lower than Germany’s. Therefore, this hotspot is geographic-dependent, and the burden should significantly decrease once EU countries move away from polluting coal and lignite fuel sources in response to climate change policy obligations.

4. Discussion

4.1. Crop type

Environmental sustainability is affected across multiple dimensions (Steffen et al., 2015), therefore assessing products across multiple impact categories is critical to provide a more precise and holistic indication as to which mitigation options should be adopted to improve the overall sustainability of food systems.

Owing to the importance of the cultivation stage, uncertainty in chickpea yield has a major influence on environmental footprints of chickpea pasta. Comparatively low yields for chickpeas mean that modest per hectare inputs translate into relatively high burdens on a mass basis for the final pasta. Relatively little research has been undertaken on yield improvement in legumes when compared to cereals such as wheat and barley (van Loon et al., 2018). The reason for little improvement in chickpea productivity can be attributed to their subordinate position in consumer likings after cereals, volatile prices due to irregular yields and lack of government incentives as opposed to cereals (Merga, Haji, 2019). There is considerable scope for yield improvement in chickpea cultivation, and therefore potential for the environmental footprint of chickpea pasta to reduce further relative to durum wheat pasta.

Limited available evidence suggests that fertiliser usage is still common in chickpea cultivation, despite research and agronomy guides stating that the practice of N application (and P application in some cases) is not necessary (Corp et al., 2004; Gan et al., 2009; GRDC, 2012). On the European scale, reducing or eliminating fertiliser application will reduce environmental degradation across all impact categories, as shown with 4) *Chickpea (Spain)*. For example, Climate Change will decrease from 0.16 kg CO_2 eq. to 0.11 kg CO_2 eq. per serving. An additional benefit of not applying fertiliser is that costs of production would decrease – the cost of SNF is increasing (Abi-Ghanem et al., 2012; Saghir Khan, Zaidi, Wani, 2007). The alternative scenario, 4) *Chickpea (Spain)*, used data from existing plots in Spain, showing the feasibility of such cultivation methods. However, even under plot trials, yield variability was very high. Therefore, it is crucial to educate farmers on proper chickpea cultivation practices, and on cultivation of legumes in general, to achieve sustainability from environmental, economical, and social perspectives. More breeding is also important to find varieties that produce more reliable yields with improved resilience.

4.2. Impact study (Nutrient density functional unit)

The advantage of the NDU as a FU as opposed to more extensive nutrient indexes lies in its simplicity whilst maintaining much of the nutritional differentiation achieved by the latter indexes (Van Dooren, 2016). It requires only three ’nutrient’ inputs - energy, essential fatty acid, protein and fibre content - and is an excellent proxy for more nutrient data demanding density indices, such as the NRF12:3 nutrient rich food index of Drewnowski, Fulgoni (2008), which requires 15 ’nutrient’ data inputs per food item. To illustrate the convenience of NDU above more conventional NRF indices, Figure 5 adapted with permission from Williams et al. (2020), shows the correlation of NDU with NRF12:3 for a total of 55 separate food items spanning 11 food groups ($r^2>0.64$).

The three macronutrient groups present in the NDU, essential fatty acids, protein, and fibre are essential for the human diet. The two groups of dietary essential fatty acids, linoleic (omega-6) and alpha-linolenic (omega-3) cannot be synthesised by the human body (Di Pasquale, 2009). The main source of omega-3 is in fish and flaxseed oil, while vegetable oils are the main source of omega-6 (Di Pasquale, 2009). These nutrients are crucial for proper growth as constituents of cell membranes (Simopoulos, 1999). Dietary fibre denotes the dietary constituents that mammalian enzymes cannot degrade (Bach Knudsen, 2001). It is a key nutrient, decreasing the risk of obesity and type 2 diabetes (Brennan, 2005; van Dooren et al., 2014), cancer and heart disease (Kendall, Esfahani, Jenkins, 2010). A diet high in fibre also decreases bowel ailments, including constipation through increased faecal substance (Wood, Grusak, 2007). Dietary protein is key to achieve body growth and protein maintenance, and net protein utilisation from animal or plant sources is comparable among adults (WHO, 2007). Consuming chickpea pasta instead of durum wheat pasta has a clear nutritional benefit in terms of fibre, essential fatty acids, and protein, resulting in considerably higher NDU values. Diets in developed countries are too high in omega-6 fatty acids and too low in omega-3 fatty acids (Simopoulos, 2016). Therefore, an additional analysis should be made to see the content of different essential fatty acids in the two products to determine the ratio of omega-3 to omega-6. One could argue that a higher amount of protein in pasta is unnecessary, as the average European consumer already consumes considerably more than the daily recommended protein intake (Westhoek et al., 2011), and that consumers will not reduce the amount of protein consumed from other (higher burden) sources, leading to wasteful protein intake. However, increasing plant protein intake to substitute animal-based protein is a shift that is crucial for environmental and health objectives (Willett et al., 2019). In addition, the presence of viscous fibre in foods is linked to the achievement of satiety (Slavin, Green, 2007), preventing over-eating, suggesting that chickpea pasta could contribute towards reducing excess calorie consumption in industrialised countries. Ideally, chickpea pasta would substitute other sources of pro-

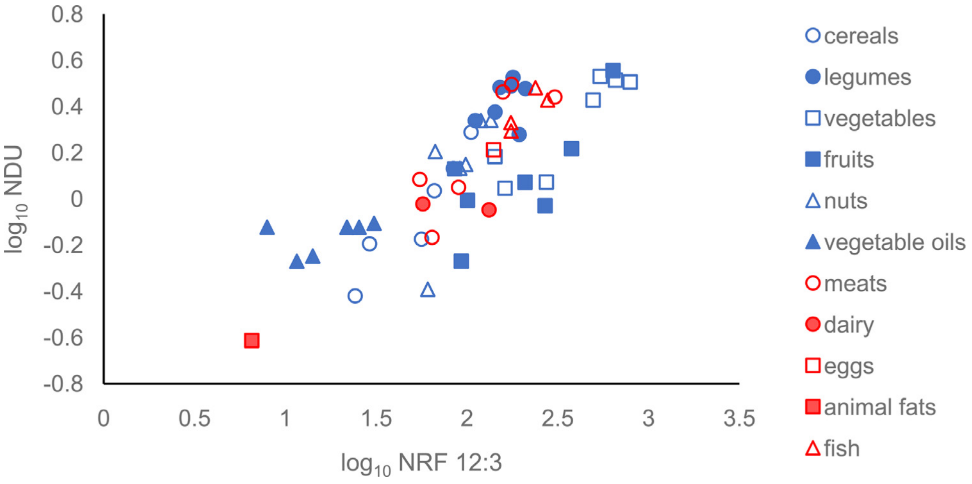

Figure 5. Correlation of NDU with NRF 12:3 for different food categories.

tein, such as meat, dairy and eggs, further reducing the environmental burdens. In their meta-analysis of food products from cradle to retail, Poore & Nemecek (2018) showed that the carbon footprint of pulses versus meat products per kg protein is significantly lower, with the 10th percentile of 100g protein ruminant meat, pork, and poultry being 5, 2.5, and 1.3 times higher than the 90th percentile of pulses other than peas. Therefore, our conclusions here are likely to be conservative, and the aforementioned wider effects of substituting wheat with chickpeas in pasta production require further investigation.

Because the *al dente* texture of legume pasta is possible, and pasta is commonly eaten with sauces, the subtle taste of chickpea pasta can easily be masked if desired. An additional incentive for consumers to opt for legume pasta is the high nutritional value, promoting a healthy diet with weight management due to high fibre content. Finally, a third incentive is the comparatively shorter cooking time of chickpea pasta, which suits a market that takes little time to prepare food.

The PEF guidelines for pasta do not consider any nutritional aspects, recommending a simple weight-based FU as the only FU to be used (European Commission, 2018b), potentially leading to incomplete footprint labelling from a nutritional perspective. We recommend that PEF guidelines incorporate a nutritional FU as an optional alternative to the weight-based FU, in order to support more accurate comparison of different foods.

4.3. Limitations

Processing legumes into “staple” foods such as chickpea pasta increase legume cultivation in EU rotations while encouraging diet change, at least from cereals to legumes and, ideally, from meat to legume protein. This study did not include the benefits of N carryover legumes provide to the following crops in rotations. This is a conservative choice that favours wheat pasta, and enables attributional footprints to be compared. LCAs with legume cropping systems should account for these benefits.

In Bevilacqua et al.’s (2007) study, it was only mentioned that compost and feed were co-products of milling. We adopted a conservative approach in this study, assuming that all co-products of milling (24%) went to cattle feed, as this has a higher value than compost. An economic allocation was performed for the co-products of semolina production and sorting of chickpeas as recommended by the PEF guidelines for dry pasta (European Commission, 2018b). This allocation method is limited by price fluctuation over time due to changes in demand, leading to changes in allocation factors (Nijdam, Rood, Westhoek, 2012).

The application of crop protection agents was excluded in all scenarios. Pesticides are often a hotspot in agricultural LCAs (Zortea, Maciel, Passuello, 2018), therefore the real impact of both types of pastas should be higher, excepted for scenario 4) *Chickpea (Spain)*, in which no pesticides or chemicals were applied in reality. However, chickpea seed production, which contributed significantly to several environmental impact categories including ecotoxicity, is a generic background process for chickpea seed production in the United States taken from Agrifootprint 3.0. These high burdens are in part due to fungicide application, releasing compounds like chlorothalonil to the environment, a highly toxic substance for aquatic animals (IPCS, 1996). A European seed production plant could vary in terms of management, as for example, the use of chlorothalonil has been banned in the European Union (European Commission, 2019).

Limitations of current LCA methodologies relevant to this study include the fact that land use is modelled as part of the technosphere, leading to potentially misleading conclusions around higher yields per unit of land always improving eco-efficiency (Richi et al., 2015). Important impacts and ecosystem services associated with land use are not represented in life cycle impact assessment methods, including, *inter alia*, impacts on pollinators, wider biodiversity (habitats) and soil quality (Ingrao et al., 2019).

5. Conclusions

This study highlights the potential of chickpea pasta to play an important role in the shift from animal protein to plant protein and higher dietary fibre consumption that is critical to achieve more sustainable, healthy diets in industrialised countries. Managed appropriately, chickpea cultivation in Europe could help to diversify crop rotations and decrease the use of synthetic fertilisers through biological nitrogen fixation. However, variable and often low chickpea yields reflect lack of crop breeding and poor agronomic practises, and represent significant potential barriers to more widespread chickpea cultivation. Breeding programmes and targeted advice via extension services could improve chickpea yields, putting this promising sustainable food crop on a level playing field with more-intensively developed wheat.

The use of the Nutrient Density Unit proposed by Van Dooren (2016) as a functional unit to compare different foods, or evaluate modification of foods, is an important advance on food LCA studies that commonly use weight, calories, or protein content as functional units. Foods made with different ingredients have different nutritional qualities that cannot be reliably represented by just one nutritional component. In this case study, the environmental burden per nutrient density unit delivered by chickpea pasta is significantly smaller than for durum wheat pasta across all categories, except for land use, which remains a significant challenge due to the scarcity of the resource. The use of the Nutrient Density Unit as a functional unit is an elegant approach to identify the nutritional (health) and environmental sustainability of different foods. Using the NDU extended the overall environmental efficiency advantage of chickpea pasta.

Further research needs to be carried out to evaluate the indirect burdens associated with a change of production of pasta types. These indirect burdens will be captured in a consequential LCA, taking into account the effects of European diet changes through partial substitution of durum wheat pasta with chickpea pasta, such as indirect land use change, the effect of higher intake of fibre on satiety leading to less food intake, and changing market prices of inputs, substitute products and co-products.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.spc.2020.06.012.

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