

1 **Life cycle assessment of supercritical CO₂ extraction of caffeine from coffee beans**

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8 The environmental impacts of caffeine extraction from coffee beans using supercritical carbon dioxide
9 (scCO₂) were evaluated, through a Life Cycle Assessment (LCA) approach. Using this process, two products of
10 interest were obtained: caffeine and decaffeinated coffee. All the emissions to air, water and soil were
11 reported to 1 kg of decaf coffee constituted by a 60/40 Arabica/Robusta blend. The performed analysis
12 showed that agricultural stages, transportation and caffeine extraction are the steps mostly affecting the
13 environmental categories under study. Therefore, the process was optimised, considering the fertilisers'
14 amount reduction and the substitution of a portion of electricity at grid with electricity produced by
15 photovoltaic panels. Using this improved scenario, a reduction of the environmental impact equal to 176 %
16 in terms of human health, 10.3 % in terms of ecosystem diversity and 16.1 % in terms of resource availability
17 can be obtained.

18

19 Keywords: LCA; supercritical decaffeination; minimized emissions; process optimization; sustainability.

20 1. Introduction

21 Coffee is one of the most consumed beverages in the world and, after petroleum, is the second traded
22 product worldwide [1]. In Western countries, a significant portion of the daily beverage is constituted by the
23 different varieties of coffee [2]. Coffee grows mainly in Africa and South America and nowadays, among a
24 large number of known species, only two varieties are successfully used in commercial cultivation: *Coffea*
25 *arabica* var. Arabica and *Coffea canephora* var. Robusta. In particular, Arabica is mainly cultivated in Brazil,
26 Colombia, Costa Rica, Guatemala and India, whereas Robusta is mainly cultivated in Vietnam, Ivory Coast,
27 Guatemala and India.

28 Coffee is second only to water as the most widely consumed beverage in the US and Europe and it is the main
29 source of caffeine in daily consumption in adults, even if caffeine is contained also in tea, chocolate, and soft
30 drinks [3, 4]. The two coffee bean varieties of worldwide importance differ considerably in price, quality and
31 consumers' acceptance. Indeed, Arabica is preferable for the aroma effect and Robusta for taste and body
32 [5]; for these reasons, a good flavour is commonly obtained by blending the two varieties. Moreover, caffeine
33 content of green coffee beans varies according to the species: Arabica beans contain about 1.0–1.2 %,
34 whereas the caffeine content in Robusta beans is about 1.6–2.5 %.

35 Even if a moderate consumption of caffeine can have beneficial effects on adults' behaviour, numerous
36 studies, in recent years, reported the effect of caffeine consumption on cardiovascular diseases [6] and on
37 central nervous system [7], leading to an increasing consumption of decaffeinated coffee [8]. Moreover,
38 caffeine recovery is important, because it can be used in cola-type drinks or in combination with other active
39 principles in the pharmaceutical field (in the treatment of headache and neuralgia) [9], or as an ingredient in
40 the cosmetic field (in the treatment of cellulitis and localised excess fat) [10].

41 Four main methods are used worldwide for the decaffeination: in the solvent based methods, organic
42 solvents (mainly methylene chloride and ethyl acetate) are employed, whereas in the non-solvent based
43 methods, water or supercritical carbon dioxide (scCO₂) are used for the caffeine extraction [11]. In all the
44 cases, coffee is decaffeinated in its green state; i.e., before the roasting operation.

45 Until the mid-1970s, methylene chloride was considered the best solvent for extraction of caffeine with
46 satisfactory results. However, subsequently, doubts arose about its risk to humans, due to the solvent high
47 toxicity. Although the residual amount of methylene chloride in decaffeinated coffee was well below the limit
48 of 10 ppm, established by the Food and Drug Administration, the suspected carcinogenicity of this solvent
49 led to the choice of a less toxic solvent, such as ethyl acetate, a natural component detected in coffee aroma
50 and found to occur naturally in different fruits. The use of ethyl acetate has two considerable drawbacks: it
51 is highly flammable and has a fruity aroma. It has to be handled carefully, increasing production costs, and it
52 tends to pass on its characteristic aroma to the coffee, slightly altering the flavour. The decaffeination using
53 water was developed in Switzerland, and constitutes a green process with respect to the product.
54 Unfortunately, water is not a particularly selective solvent and, therefore, not only caffeine but also various
55 flavours were removed from coffee beans using this method. As a result, a less flavourful brew with respect
56 to other methods was obtained. The most selective process for removing just caffeine and not the other
57 flavour precursors from coffee is based on the use of $scCO_2$. This process was successfully developed on an
58 industrial scale in the 1970s, based on two patents developed by Zosel: in the first one, the process was
59 presented for the recovery of caffeine [12], whereas, in the second one, a detailed description aimed at
60 obtaining decaffeinated coffee was proposed [13].

61 Supercritical fluids (SCFs) based techniques were proposed as an alternative to conventional processes,
62 thanks to their specific characteristics, mainly, solvent power and liquid-like densities with gas-like transport
63 properties that can be tuned by varying pressure and temperature. They were successfully applied in several
64 fields, such as, for example, micronization [14], porous structures formation [15], adsorption [16]. Among
65 the different $scCO_2$ based processes, one of the most studied one was supercritical fluids extraction (SFE), for
66 the possibility of continuously modulating the solvent power/selectivity; this process was frequently used for
67 the extraction of essential oils [17-19]. SFE was used also for the extraction of caffeine from natural sources,
68 such as coffee husks [20], coffee beans [21] and tea leaves [22].

69 Since supercritical fluids' based technologies are considered as "eco-friendly", it is important to study the
70 environmental emissions due to a specific production. One of the most common ways to determine, in a

71 quantitative way, the environmental impact of a process or a product is the use of life cycle assessment (LCA)
72 analysis.

73 Many papers based on LCA analyses were published in different research fields, such as energy [23],
74 beverages [24-26], wines [27, 28], fruits [29, 30], pharmaceutical delivery systems [31, 32] and wastewater
75 treatments [33]. In particular, food sector is among the most impactful ones for the environment, due to
76 production, preservation and distribution steps [34], which consume a considerable amount of energy [35].
77 LCA studies regarding the food sector were addressed on agricultural stages [36], production steps [28] or
78 packaging systems [37]. Some LCA studies regarding coffee were performed, considering agricultural stages
79 [13], processing steps [38, 39] and packaging [40].

80 Concerning papers on LCA of decaffeination processes, a preliminary gate-to-gate study on the
81 decaffeination of Arabica seeds, considering only the industrial stages of the process was attempted [41], but
82 a complete LCA study on decaffeination of commercial decaf coffee (constituted by Arabica and Robusta
83 blends) was not endeavoured until now.

84 Therefore, the aim of this work is to carry out a cradle-to-gate LCA analysis of the production of caffeine and
85 of scCO₂ decaf coffee constituted by a 60/40 Arabica/Robusta blend. In the analysis, the considered steps are
86 coffee cultivation (in Brazil for Arabica and in Vietnam for Robusta), its transportation to Italy, its processing
87 to obtain decaf coffee and caffeine. Data regarding agricultural stages in Brazil and Vietnam were obtained
88 from literature [36, 42], whereas data regarding the industrial stages were collected from an Italian
89 processor.

90 **2. Process description**

91 In Table 1, the main activities and the details of the process under investigation are reported.

92 *Table 1: Process details and assumptions*

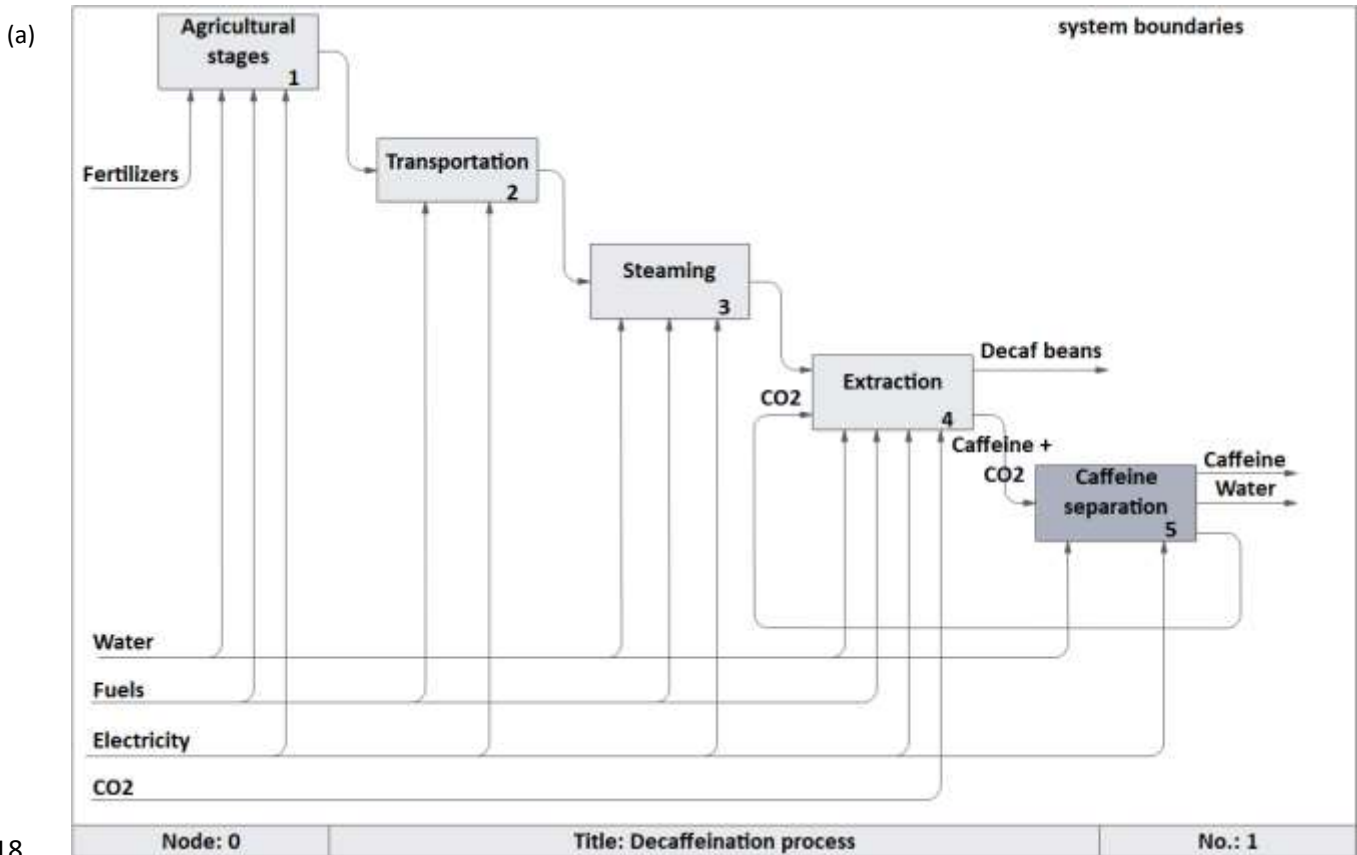
Process	Characteristics and details	
	<i>Arabica</i>	<i>Robusta</i>
Agricultural stages	Energy and water supply	Energy and water supply

	Nitrogen and fertilisers supply	Nitrogen and fertilisers supply
	Herbicides and pesticides supply	Herbicides and pesticides supply
	Diesel and fuel supply	Diesel and fuel supply
Energy supply for agricultural stages	Brazilian energy mix low voltage	Vietnamese energy mix low voltage
Coffee beans supply to facility	Transport by truck, 40 t from Genoa (distance = 650 km) + by large tanker from Brazil (distance = 9100 km)	Transport by truck, 40 t from Trieste (distance = 800 km) + by large tanker from Vietnam (distance = 13600 km)
Energy supply for processing stages	Italian energy mix medium voltage	Italian energy mix medium voltage
Coffee beans steaming	T = 90 °C; t = 5h; energy, water and fuel supply	T = 90 °C; t = 5h; energy, water and fuel supply
Pressurization	t=0.25 h; carbon dioxide, energy, water and fuel supply	t=0.25 h; carbon dioxide, energy, water and fuel supply
Operating conditions' stabilization	T=90 °C; P=25 MPa; t=0.25 h; carbon dioxide, energy, water and fuel supply	T=90 °C; P=25 MPa; t=0.25 h; carbon dioxide, energy, water and fuel supply
Caffeine extraction	T=90 °C; P=25 MPa; t=11.5 h; carbon dioxide, energy, water and fuel supply	T=90 °C; P=25 MPa; t=22 h; carbon dioxide, energy, water and fuel supply
Depressurization	T=25 °C; P=0.1 MPa; t=1 h	T=25 °C; P=0.1 MPa; t=1 h
Caffeine recovery	Water, energy and fuel supply	Water, energy and fuel supply

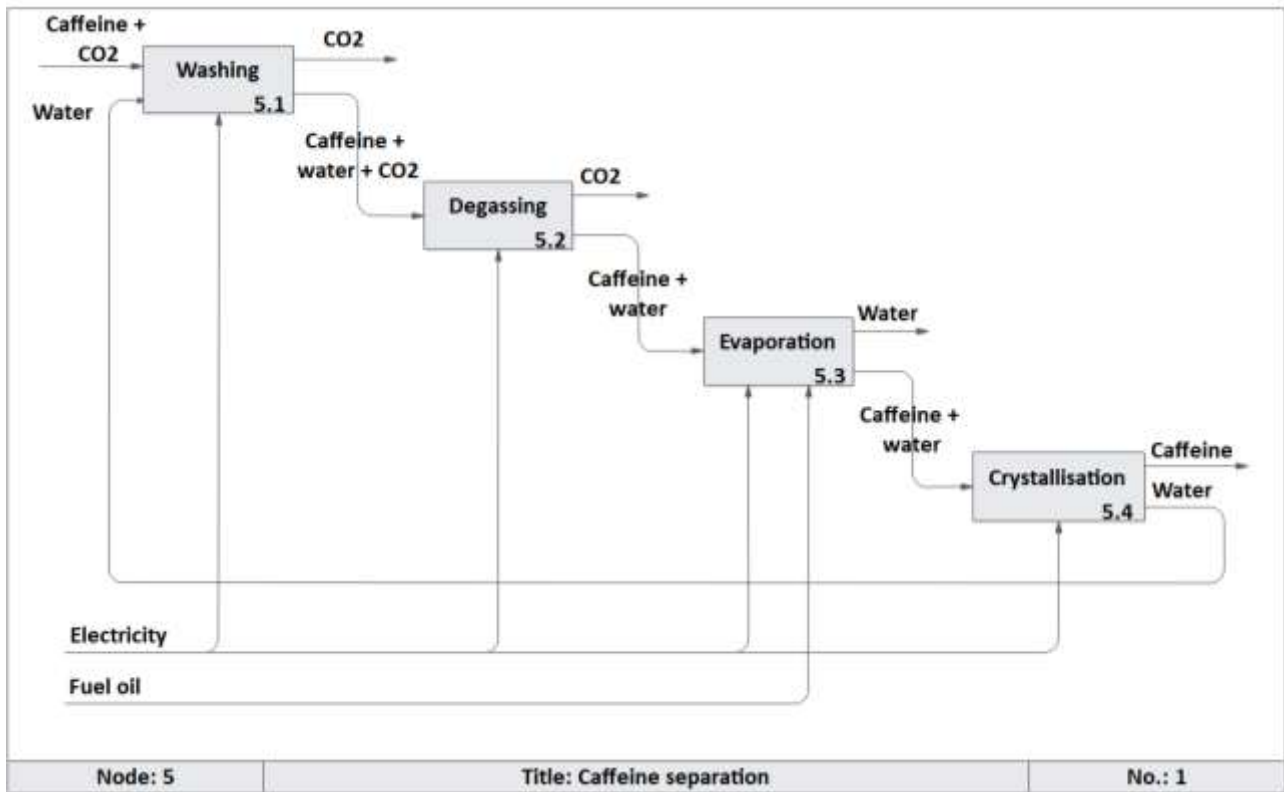
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94 Coffee beans are cultivated in Brazil in the case of Arabica and in Vietnam in the case of Robusta. After
95 harvesting, they are transported to the Italian processor by large tanker and by truck. Caffeine extraction
96 from coffee beans using scCO₂ can be distinguished in three main steps: steaming, supercritical extraction
97 and caffeine recovery, as represented in Figure 1a. After cultivation (stage 1) and transportation to the
98 processor (stage 2), coffee beans are put in contact with superheated steam at elevated temperature (stage
99 3) until their moisture content reaches 30 % by weight; as a result, the beans swell considerably. Then, a 9
100 tons batch of water-moistened coffee beans is charged in a vessel with a volume equal to 22 m³ (stage 4) and
101 an extraction cycle begins. Indeed, the extraction vessel is pressurised by pumping CO₂ until the desired
102 conditions of pressure and temperature are reached (25 MPa and 90 °C). The scCO₂ is passed through the
103 beans' bed for approximately 11.5 hours in the case of Arabica coffee beans and 22 hours in the case of
104 Robusta coffee beans, with the aim of extracting 97 % of the caffeine. The caffeine+CO₂ mixture leaves the

105 extractor and accumulates in a holding tank, until the extraction is completed. Then, the extractor begins a
 106 down time, in order to be emptied from decaffeinated beans (which can be stored in silos) and charged with
 107 fresh beans. The last step is the caffeine recovery, shown in Figure 1b. The stream of caffeine rich CO₂ begins
 108 to flow at a steady state through a water wash packed column (stage 5.1), at conditions that are the same as
 109 those of the extractor to keep the carbon dioxide in its supercritical state. Here, the supercritical stream is
 110 counter-currently contacted with a stream of water, which removes 99.5 % of the caffeine from the CO₂,
 111 considering that caffeine has a higher affinity for water compared to scCO₂. The regenerated carbon dioxide
 112 leaves the water wash column and is pumped into its storage tank. The caffeine rich water stream exiting the
 113 water wash column is throttled to approximately atmospheric pressure, during a degassing step (stage 5.2).
 114 During this stage, a small amount of carbon dioxide dissolved in the mixture is vented as a gas. The caffeine
 115 + water mixture is sent to two evaporators in series (stage 5.3), where it is concentrated from 1 to 15 % in
 116 weight. In the last step (stage 5.4), the mixture is sent to a crystalliser where solid caffeine precipitates
 117 because of a sudden temperature decrease.



(b)



119

120 Figure 1: Main stages and system boundaries for the extraction of caffeine from coffee beans: (a) complete process
121 scheme; (b) details of caffeine separation.

122 3. LCA methodology

123 LCA analysis allows to correlate a broad set of data regarding the life-cycle of a product or a process in order
124 to identify the phases of the process that are critical from an environmental point of view. The main steps of
125 an LCA analysis are presented in the following sub-sections.

126 3.1 Goal definition, functional unit and system boundaries

127 Goal definition is one of the most important phases of the LCA methodology, because the choices made at
128 this stage influence the entire study. The goal of this study is the in-depth evaluation of the environmental
129 impacts of coffee decaffeination using supercritical carbon dioxide (scCO₂). The functional unit (FU) is the
130 reference to which all the inputs and outputs have to be related. Considering that:

131 1) the work has a dual objective, consisting in the LCA of both decaf coffee and caffeine products;

132 2) different data are obtained for Arabica and Robusta seeds processing,

133 the FU was defined as 1 kg of decaf blend coffee beans (constituted by 600 g of Arabica and 400 g of Robusta)
134 corresponding to 11.4 g of dry caffeine recovered from the blend.

135 Through mass and energy balances of each operation, a cradle-to-gate analysis was performed; therefore,
136 the system boundaries (reported in Figure 1a) are set from coffee cultivation to decaffeinated coffee and
137 caffeine attainment.

138 *3.2 Data collection and life cycle inventory*

139 The life cycle inventory (LCI) is one of the most time consuming steps, because it consists of the activities
140 related to the search, collection, and interpretation of the data necessary for the environmental assessment
141 of the observed system. During this step, all inputs and outputs such as usage of resources and materials,
142 consumption of electricity and fuels, and determination of transportation have to be quantified for the stages
143 included in the system boundaries.

144 Data regarding the stages of Arabica beans cultivation in Brazil [36] and Robusta beans cultivation in Vietnam
145 [42] were recovered from literature. Primary data regarding the industrial stages of caffeine extraction from
146 the beans were collected from an Italian processor, which uses supercritical carbon dioxide to extract caffeine
147 from coffee beans (the process details are listed in Table 1) through questionnaires and personal interviews.
148 Background data regarding, for example, inputs and outputs associated with the production of 1 kWh of
149 electricity were recovered from [43] for Brazilian and Italian energy mix. The Brazilian mix process available
150 in Ecoinvent was also adapted to Vietnam, by replacing the proportions of energy sources for Brazil with the
151 ones gathered for this country from [44]. Through mass and energy balances made on each of the single unit
152 operations, data were organised in tables that constitute the inventory. The numerical results obtained after
153 mass and energy balances are listed in Table 2.

154 *Table 2. Life cycle inventory of the main inputs and outputs for decaf coffee production and caffeine extraction; data are*
155 *referred to 1 kg of decaf coffee beans (constituted by 600 g of Arabica and 400 g of Robusta) and 11.4 g of caffeine*

156

Phase	Input/Output	Unit
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Agricultural	Pesticides	kg	5.85E-03
	Nitrogen	kg	1.04E-01
	Phosphorous (P ₂ O ₅)	kg	6.29E-02
	Potassium (K ₂ O)	kg	6.57E-02
	Herbicides	kg	6.36E-04
	Diesel	kg	1.06E-01
Transportation	Green coffee beans	kg	8.77E-01
	Transport by truck	tkm	6.24E-01
	Transport by large tanker	tkm	9.58E+00
Steaming	Coffee beans	kg	8.69E-01
	Water	kg	1.65E-01
	Methane	m ³	2.07E-03
Supercritical extraction	Coffee beans	kg	1.03E+00
	Carbon dioxide	kg	1.24E-01
	Methane	m ³	2.79E-01
	Electricity	MJ	5.24E+00
	<i>Output</i>		
	Decaffeinated beans	kg	1.00E+00
Washing	Caffeine	kg	1.28E-02
	Carbon dioxide	kg	7.43E-01
	Water	kg	1.12E+00
	Electricity	MJ	7.59E-05
	<i>Output</i>		
	Carbon dioxide	kg	6.86E-01
Degassing	Caffeine	kg	1.27E-02
	Carbon dioxide	kg	5.79E-02
	Water	kg	1.12E+00
	Electricity	MJ	8.07E-06
	<i>Output</i>		
	Carbon dioxide	kg	5.79E-02
Evaporation	Caffeine	kg	1.27E-02
	Water	kg	1.19E+00

	Fuel oil	kg	2.11E-02
	<i>Output</i>		
	Water	kg	1.12E+00
Crystallisation	Caffeine	kg	1.27E-02
	Water	kg	7.2E-02
	Electricity	MJ	1.18E-02
	<i>Output</i>		
	Caffeine	kg	1.14E-02

157 3.3 Impact assessment

158 The elaboration of the inventory data was performed through the LCA software SimaPro 8.0.5 (PRé
159 Consultants, 2014) in agreement with the reference standard for LCA (i.e. ISO 14040-14044). ReCiPe method
160 [45] was used to aggregate the inventory results first in terms of 18 midpoint categories and, then, in terms
161 of damages to human health, ecosystem diversity and resource availability (endpoint). The list of the impact
162 categories at midpoint and endpoint level assessed in the present study is shown in the first column of Table
163 3. The chosen time perspective, among the three proposed by the ReCiPe method, is the hierarchist (H),
164 which is based on the most common policy principles concerning time-frame and is the most balanced one.

165 *Table 3: Environmental impact categories with their respective acronyms and units and impact assessment at midpoint*
166 *level*

Impact category	Acronym	Unit	Impact assessment
<i>Midpoint level</i>			
Climate change	CC	kg CO ₂ eq	3.29E+00
Ozone depletion	OD	kg CFC-11 eq ¹	6.82E-07
Terrestrial acidification	TA	kg SO ₂ eq	1.82E-02
Freshwater eutrophication	FE	kg P eq	7.27E-04
Marine eutrophication	ME	kg N eq	7.68E-02
Human toxicity	HT	kg 1,4DCB eq ¹	8.69E-01
Photochemical oxidant formation	POF	kg NMVOC ¹	1.22E-02
Particulate matter formation	PMF	kg PM ₁₀ eq	5.95E-03
Terrestrial ecotoxicity	TET	kg 1,4DCB eq ¹	1.54E-03

Freshwater ecotoxicity	FET	kg 1,4DCB eq ¹	5.45E-02
Marine ecotoxicity	MET	kg 1,4DCB eq ¹	4.92E-02
Ionising radiation	IR	kBq U235 eq ¹	3.92E-01
Agricultural land occupation	ALO	m ² x yr	2.33E+00
Urban land occupation	ULO	m ² x yr	4.00E-02
Natural land transformation	NLT	m ²	6.47E-04
Water depletion	WD	m ³	3.67E-02
Mineral resource depletion	MRD	kg Fe eq	1.87E-01
Fossil fuel depletion	FD	kg oil eq	8.86E-01

Endpoint level

Human health	HH	DALY ¹
Ecosystem diversity	ED	species.yr
Resource availability	RA	\$

167 ¹CFC-11: Chlorofluorocarbon; 1,4DCB: 1,4 dichlorobenzene; NMVOC: Non Methane Volatile Organic Carbon compound; U235:
168 Uranium 235; DALY: disability-adjusted life years; species.yr: loss of species during a year; \$: increased cost.

169 **4. Results and discussion**

170 *4.1 Environmental analysis of coffee decaffeination*

171 The environmental analysis of the production of decaffeinated coffee was performed in terms of ReCiPe
172 midpoint categories. The analysis was performed considering the production of 1 kg of decaf coffee and 11.4
173 g of pure caffeine; the results of the impact assessment are reported in the last column of Table 3.

174 In order to identify the processing steps that generate the higher impact, an in-depth analysis was performed.

175 The impact assessments at midpoint level related to each stage of the process are reported in Table 4.

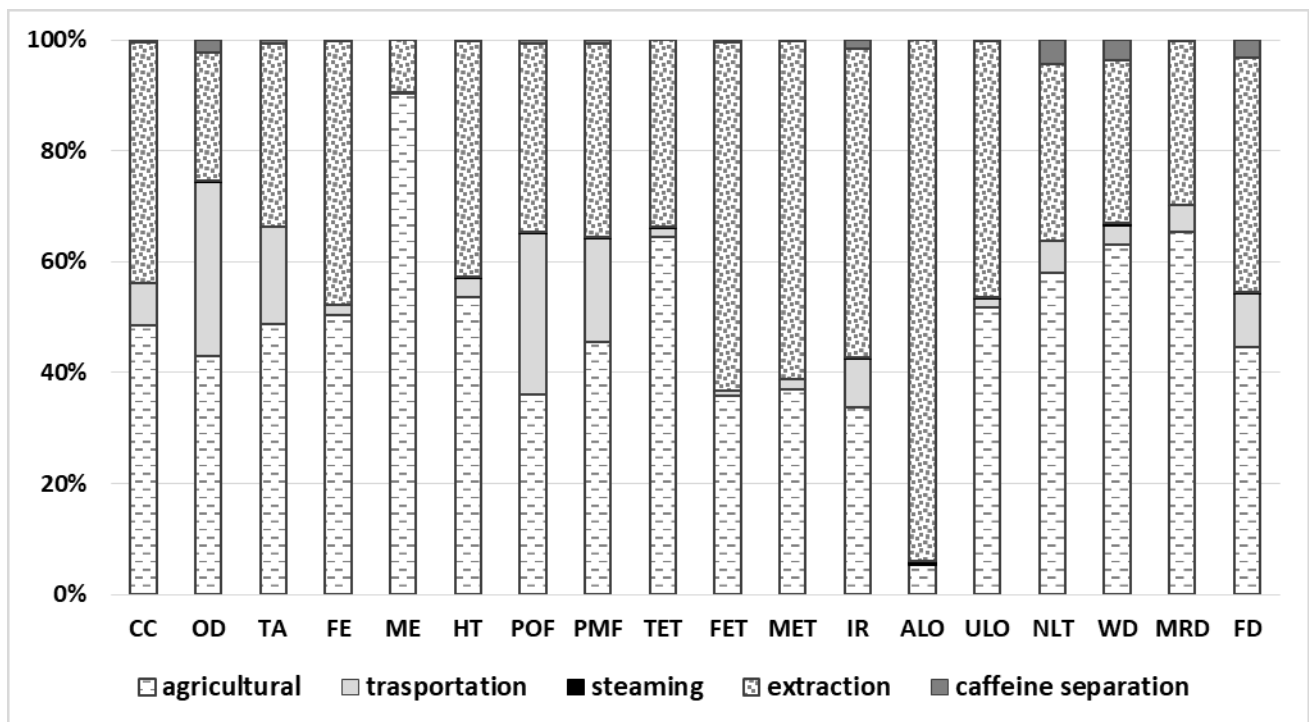
176 *Table 4: Impact assessment at midpoint level with respect to 1 kg of decaf coffee and 11.4 g of caffeine*

Impact category	agricultural	transportation	steaming	extraction	washing	degassing	evaporation	crystallisation
CC	1.59E+00	2.54E-01	2.25E-03	1.43E+00	7.94E-04	1.41E-06	1.23E-02	2.07E-03
OD	2.94E-07	2.14E-07	2.26E-10	1.59E-07	3.25E-10	2.03E-13	1.46E-08	2.97E-10
TA	8.88E-03	3.20E-03	1.60E-05	6.01E-03	4.80E-06	5.61E-09	1.08E-04	8.23E-06

FE	3.67E-04	1.21E-05	1.22E-06	3.45E-04	2.64E-07	2.59E-10	1.28E-06	3.80E-07
ME	6.93E-02	1.76E-04	1.03E-05	7.32E-03	7.28E-07	9.18E-09	9.68E-06	1.35E-05
HT	4.66E-01	3.02E-02	1.27E-03	3.69E-01	3.72E-04	2.78E-07	1.90E-03	4.08E-04
POF	4.40E-03	3.56E-03	1.45E-05	4.16E-03	2.77E-06	3.30E-09	7.74E-05	4.85E-06
PMF	2.71E-03	1.11E-03	6.44E-06	2.09E-03	2.02E-06	1.78E-09	3.14E-05	2.61E-06
TET	9.91E-04	2.29E-05	3.38E-06	5.18E-04	6.78E-08	9.85E-11	1.31E-06	1.45E-07
FET	1.95E-02	4.75E-04	4.32E-05	3.43E-02	5.08E-05	4.35E-08	8.20E-05	6.38E-05
MET	1.82E-02	8.11E-04	3.87E-05	3.00E-02	4.45E-05	3.77E-08	6.80E-05	5.54E-05
IR	1.32E-01	3.44E-02	4.08E-04	2.18E-01	1.25E-04	2.46E-07	5.64E-03	3.60E-04
ALO	1.22E-01	1.19E-03	1.71E-02	2.19E+00	2.94E-05	5.77E-08	1.64E-04	8.46E-05
ULO	2.07E-02	5.93E-04	1.17E-04	1.84E-02	8.34E-06	4.97E-09	1.20E-04	7.30E-06
NLT	3.75E-04	3.74E-05	3.89E-07	2.05E-04	1.04E-07	2.28E-10	2.84E-05	3.35E-07
WD	2.31E-02	1.31E-03	1.93E-04	1.07E-02	1.12E-03	9.82E-09	2.05E-04	1.44E-05
MRD	1.22E-01	9.06E-03	8.44E-05	5.52E-02	7.67E-05	5.80E-08	3.73E-04	8.51E-05
FD	3.96E-01	8.57E-02	5.92E-04	3.76E-01	2.04E-04	4.36E-07	2.74E-02	6.40E-04

177

178 The relative contributions of the different stages of the complete process are reported in Figure 2.



179

180 Figure 2. Relative contributions of the different stages with respect to the overall impact

181 It is clear that the steaming process contribution is negligible on all the midpoint categories. Indeed, its
182 contribution is equal at most to 0.7 %, in the case of agricultural land occupation. The caffeine separation
183 process is appreciable only in terms of ozone depletion (2.2 %), natural land transformation (4.5 %), water
184 depletion (3.6 %) and fossil fuel depletion (3.2 %). The remaining steps; i.e., agricultural stages, transportation
185 and caffeine extraction contribute to different extents on the midpoint categories.

186 Transportation contribution is higher than 15 % with respect to the overall impact in terms of ozone
187 depletion, terrestrial acidification, photochemical oxidant formation and particulate matter formation.

188 The relative contribution of the agricultural step is higher than the contribution of supercritical extraction in
189 terms of ozone depletion, terrestrial acidification, marine eutrophication, terrestrial ecotoxicity, natural land
190 transformation, water depletion and mineral resource depletion. In particular:

- 191 • the emissions in terms of ozone depletion and natural land transformation are mainly due to diesel
192 consumption for planting/harvesting operations and to the use of nitrogen fertiliser;
- 193 • the emissions in terms of terrestrial acidification, water depletion and mineral resource depletion are
194 mainly due to the use of nitrogen fertiliser;
- 195 • eventually, the emissions in terms of marine eutrophication and terrestrial ecotoxicity are due to both
196 nitrogen and potassium fertilisers.

197 On the contrary, the relative contribution of the extraction step is higher than the contribution of the
198 agricultural step in terms of freshwater ecotoxicity, marine ecotoxicity, ionising radiation and agricultural
199 land occupation. The emissions in terms of agricultural land occupation are mainly due to the production of
200 the methane used in the hot utility, whereas the emissions in terms of the other categories are mainly due
201 to the electrical energy generation.

202 Agricultural and extraction steps' contributions are comparable in terms of the remaining impact categories.

203 *4.2 Scenario analysis and improved solution*

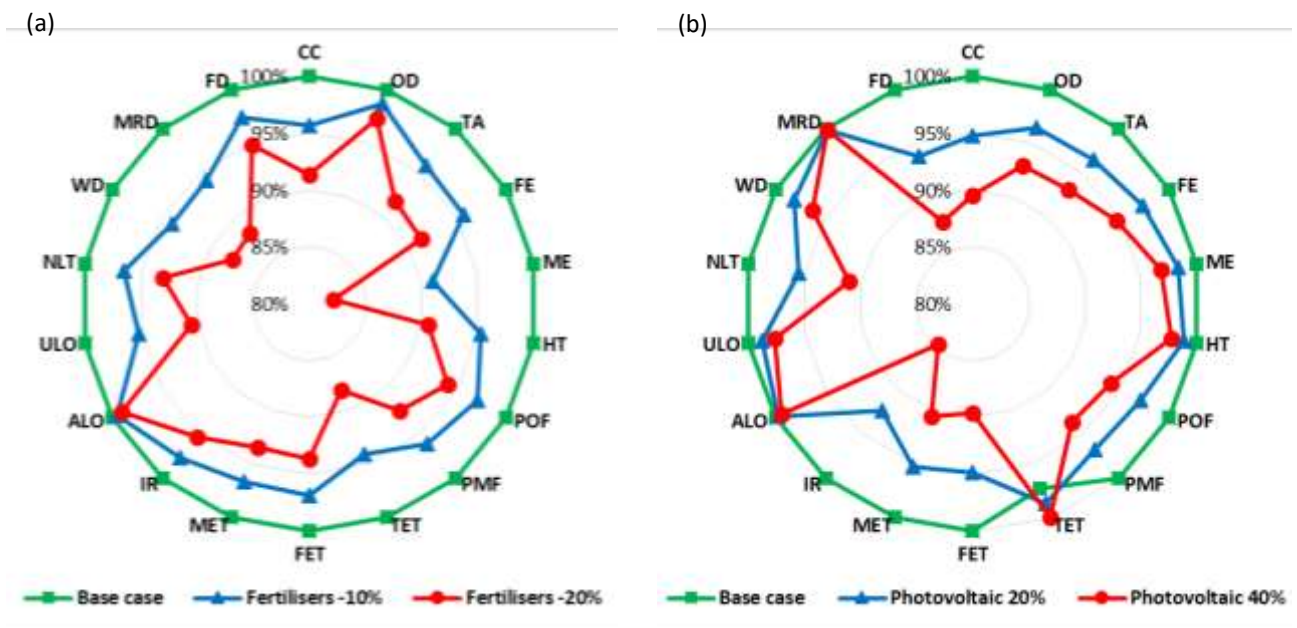
204 In order to estimate the possible reduction of the emissions due to the modification of some input variables,
205 a scenario analysis is proposed. The performed analysis considers the possibility of reducing the amount of

206 fertilisers used in the agricultural stage and modifying the electricity source. Indeed, in the last years,
 207 renewable energy sources were frequently used with the aim of improving processes' sustainability [46-48].

208 Therefore, the scenario analysis considers:

- 209 • different fertilisation strategies leading to a reduction of the amount of fertilisers (10 or 20 %) for the
 210 same coffee beans yield; indeed, the fertilisation can be carried out in different ways depending, for
 211 example, on the agricultural practice (conventional or biological), on the type of plantation (in shade
 212 or sun, monoculture or not), on the method of harvesting (manual or mechanised);
- 213 • for the electricity source, the installation and usage of photovoltaic panels and the substitution of a
 214 portion (20 or 40 %) of the electricity at grid referred to the Italian energy mix with electricity
 215 produced by the installed photovoltaic panels.

216 The results of the scenario analysis are shown in the radar charts in Figure 3.



217
 218 *Figure 3: Scenario analysis at midpoint level: (a) different amount of fertilisers; (b) introduction of photovoltaic panels*

219 It is possible to observe that:

- 220 1) the reduction of the amount of fertilisers used in the agricultural stages implies a lowering of the emissions
 221 in terms of all the midpoint categories;

222 2) the substitution of a portion of the electricity source using photovoltaic panels lowered the emissions in
 223 terms of all the ReCiPe midpoint categories, with an exception of TET. This result is in agreement with life
 224 cycle assessment studies performed on multicrystalline silicon photovoltaic panels, asserting that TET was
 225 the category most significantly affected by photovoltaic modules usage [49, 50].

226 On the basis of the performed analysis, an improved scenario is proposed, considering a reduction of the
 227 amount of fertilisers equal to 20 % and the substitution of the 40 % of the electricity grid with electricity
 228 produced by photovoltaic panels. In Table 5, the emissions at midpoint level of this improved scenario and
 229 its comparison with the base case are reported.

230 *Table 5: Impact assessment at midpoint level of the improved scenario ant its comparision with the base case. Data are*
 231 *referred to the production of 1 kg of decaf coffee and 11.4 g of caffeine*

Impact category	Base case	Improved scenario	Emissions' reduction with respect to the base case
	(a)	(b)	$\frac{(b - a)}{a} \times 100$
CC	3.29E+00	2.66E+00	-19.0%
OD	6.82E-07	6.19E-07	-9.3%
TA	1.82E-02	1.55E-02	-15.1%
FE	7.27E-04	6.25E-04	-14.1%
ME	7.68E-02	6.07E-02	-21.0%
HT	8.69E-01	7.67E-01	-11.8%
POF	1.22E-02	1.08E-02	-11.9%
PMF	5.95E-03	5.11E-03	-14.2%
TET	1.54E-03	1.39E-03	-9.5%
FET	5.45E-02	4.54E-02	-16.6%
MET	4.92E-02	4.13E-02	-16.0%
IR	3.92E-01	3.15E-01	-19.7%
ALO	2.33E+00	2.29E+00	-1.5%
ULO	4.00E-02	3.52E-02	-11.9%
NLT	6.47E-04	5.48E-04	-15.3%
WD	3.67E-02	3.11E-02	-15.3%
MRD	1.87E-01	1.65E-01	-12.2%

FD

8.86E-01

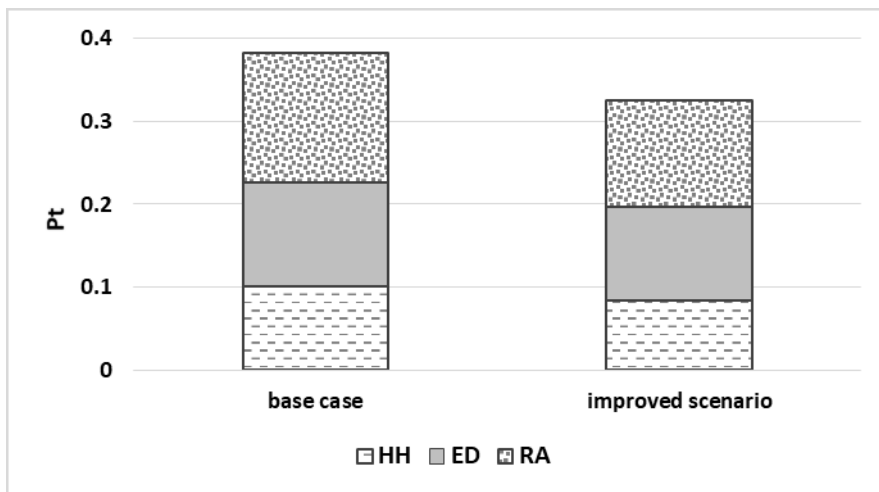
7.35E-01

-17.0%

232

233 It is clear that the improved scenario implies an appreciable reduction of the impacts on all the considered
234 midpoint categories.

235 Finally, the environmental impacts were grouped and normalised, according to the ReCiPe method,
236 considering the damage at the endpoint level; i.e., in terms of damage to human health (HH), to ecosystem
237 diversity (ED) and to resource availability (RA). The comparison between the base case and the proposed
238 improved scenario is reported in Figure 4.



239

240 *Figure 4: Environmental impact according to the normalised ReCiPe damage categories (point, Pt)*

241 It is possible to observe that the improved solution generated a reduction of the environmental impact equal
242 to 17.6 % in terms of human health, 10.3 % in terms of ecosystem diversity and 16.1 % in terms of resource
243 availability with respect to the base case.

244 5. Conclusions and perspectives

245 The supercritical carbon dioxide caffeine extraction from coffee beans is a suitable process to obtain, in one
246 single process, decaf coffee and pure caffeine. Besides, the cradle-to-gate LCA analysis provided quantitative
247 information of the environmental performance of the process, showing that the major contributors to the
248 environmental impact are agricultural stages, transportation and caffeine extraction steps. The scenario
249 analysis demonstrated the importance of fertilisers' amount and electricity sources. An improved solution

250 was proposed, obtaining a global reduction of the impact with respect to the base case equal to 14.6 %. A
251 step forward of this work would be to analyse the correlation between the obtainable emissions' percentage
252 reduction and the investment and management costs needed to obtain it.

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