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Life cycle assessment of supercritical CO_2 extraction of caffeine from coffee beans

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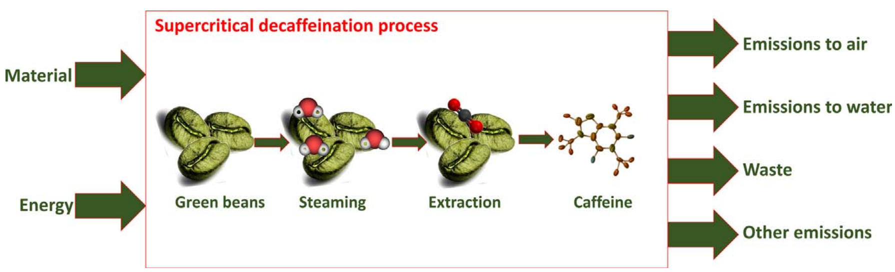
GRAPHICAL ABSTRACT



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ABSTRACT

The environmental impacts of caffeine extraction from coffee beans using supercritical carbon dioxide ($scCO_2$) were evaluated, through a Life Cycle Assessment (LCA) approach. Using this process, two products of interest were obtained: caffeine and decaffeinated coffee. All the emissions to air, water and soil were reported to 1 kg of decaf coffee constituted by a 60/40 Arabica/Robusta blend. The performed analysis showed that agricultural stages, transportation and caffeine extraction are the steps mostly affecting the environmental categories under study. Therefore, the process was optimised, considering the fertilisers' amount reduction and the substitution of a portion of electricity at grid with electricity produced by photovoltaic panels. Using this improved scenario, a reduction of the environmental impact equal to 176% in terms of human health, 10.3% in terms of ecosystem diversity and 16.1% in terms of resource availability can be obtained.

1. Introduction

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

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

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Table 1
Process details and assumptions.

Process	Characteristics and details	
	Arabica	*Robusta*
Agricultural stages	Energy and water supply Nitrogen and fertilisers supply Herbicides and pesticides supply Diesel and fuel supply	Energy and water supply Nitrogen and fertilisers supply Herbicides and pesticides 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 = 5 h; energy, water and fuel supply	T = 90 °C; t = 5 h; 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

beans is about 1.6–2.5%.

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

Four main methods are used worldwide for the decaffeination: in the solvent based methods, organic solvents (mainly methylene chloride and ethyl acetate) are employed, whereas in the non-solvent based methods, water or supercritical carbon dioxide ($scCO_2$) are used for the caffeine extraction [11]. In all the cases, coffee is decaffeinated in its green state; i.e., before the roasting operation.

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

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

Since supercritical fluids' based technologies are considered as "eco-friendly", it is important to study the environmental emissions due to a specific production. One of the most common ways to determine, in a quantitative way, the environmental impact of a process or a product is the use of life cycle assessment (LCA) analysis.

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

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

Therefore, the aim of this work is to carry out a cradle-to-gate LCA analysis of the production of caffeine and of $scCO_2$ decaf coffee constituted by a 60/40 Arabica/Robusta blend. In the analysis, the considered steps are coffee cultivation (in Brazil for Arabica and in Vietnam for Robusta), its transportation to Italy, its processing to obtain decaf coffee and caffeine. Data regarding agricultural stages in Brazil and Vietnam were obtained from literature [36,42], whereas data regarding the industrial stages were collected from an Italian processor.

2. Process description

In Table 1, the main activities and the details of the process under