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Life Cycle Analysis of Charcoal Production in Masonry Kilns with and without Carbonization Process Generated Gas Combustion

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Abstract: New technologies and emissions controls have been developed for the production of charcoal, but are not widely used in the industry. The present study seeks to evaluate the potential environmental impact of these new technologies as compared to traditional ones. A Life Cycle Assessment (LCA) of Brazilian charcoal produced with different technologies without and with the combustion of the gases in burners or furnaces was carried out. The inclusion of furnaces for the combustion of gases reduces all categories of potential environmental impacts by approximately 90% in both a circular masonry kiln and a rectangular masonry kiln with gas combustion. In the process of producing charcoal (gate-to-gate system boundary), in terms of climate change, the rectangular masonry kiln with gas combustion was approximately 63% less impactful than the circular masonry kiln with gas combustion. In the gate-to-gate analysis, the rectangular masonry kiln with gas combustion presented the best performance when not considering NO_2 and SO_2. Considering these emissions, there were changes in the impact categories of particulate matter emission and terrestrial acidification, and the circular masonry kiln with gas combustion presented better performance (for cradle-to-gate system boundary). The process in a rectangular masonry kiln without gas combustion presented a greater contribution to the categories of terrestrial impact ecotoxicity (98%), due to the emission of acetic acid especially.

Keywords: charcoal production; rectangular kiln; circular kiln; life cycle assessment; environmental performance; pyrolysis gas

1. Introduction

The demand for sustainable growth has increasingly led to use of alternative and renewable energies. Even in some countries where access to other energy sources is widespread, charcoal is an important technological utility, such as in the case of the production of certain iron castings in Brazil that require pig iron as a sulfur-free feedstock, a chemical element that is present in the coal.

The use of charcoal as an energy input is secular and prevalent, and it is the only source of energy for a significant portion of the world's population, such as in many African countries accounting for more than 60% of world production [1]. Of the total charcoal produced in Brazil, more than 90% is consumed in industrial processes, mainly in the metallurgy sector [2]. However, the use of archaic and inefficient kilns for its production prevails, even in Brazil, the world's largest producer of this input (around 6.2 Mt in 2015) [1]. The use of traditional systems such as Kenya's earth mound kiln and unsustainable harvesting of wood, for example, lead to the waste of 85–91% of biomass and cause negative environmental impacts [3].

The quest for sustainability is important for the charcoal value chain to ensure that wood procurement, the carbonization stage, input/product distribution logistics, and charcoal use comply with sustainability criteria. Regarding wood harvesting, the Brazilian charcoal production sector uses wood from eucalypt forest plantations (87% of the total charcoal produced in 2015). This practice lowers the pressure on natural forests [4], but, in sub-Saharan Africa (SSA), the harvesting of wood from planted forests does not reach 5% [5].

The reduction in the volume of carbon emissions varies according to the carbonization technology chosen [6]. However, it is common practice to release the wood pyrolysis gases into the atmosphere [7]. Large Brazilian companies with processes dependent on charcoal have invested in research and development in the search for technologies that are able to obtain higher yield of wood, greater homogeneity of the charcoal, less time in the production process, and scalability, besides the use of the pyrolysis gases for energy cogeneration [8], as these are considered emerging technologies. These technologies, being of industrial nature in most cases, are still in consolidation and are responsible for a small portion of Brazilian charcoal production.

In the same way, Brazilian researchers coupled furnaces to the circular and rectangular masonry kilns for the combustion of carbonization gases, in aim of the following: reducing emissions of greenhouse gases (GHG) [9,10]; identifying the potential of using the heat generated for drying the wood [11]; evaluating the influence of this process on the quality and gravimetric yield of charcoal [12–14]; and obtaining carbon credits [15]. This combined furnace system also has the characteristic of affordable cost [16]. It is also considered to be emerging in the process of consolidation, has the potential to reach most Brazilian producers, and has application anywhere in the world.

The emphasis on the differences between the technologies used in the studies presented in this paper and the emerging technologies reviews the use of the pyrolysis gases in the cycle itself for drying the wood and the cogeneration of electricity on an industrial scale, as shown in Bailis et al. [7] and Vilela et al. [8]. Considering the historical annual volume of 6–9 Mt charcoal production in Brazil, there is a potential of electric generation of 3–5 TWh per year [17].

In a recent and thorough review of the literature on the state of the art of the technologies used to burn the carbonization gases, Pereira et al. [10] mention studies that have presented the reduction in emissions of carbon monoxide (CO) and methane (CH_4) for combined kiln and burner systems. Adoption of these systems is compatible with thousands of producers and can be used in any operation producing charcoal, as a way of mitigating GHG and improving working conditions. However, there is no evidence of LCA research on the charcoal production with burning in combined furnace systems.

One way of verifying whether charcoal production is being conducted sustainably is by assessing its impact on climate change [1]. In this sense, Ugaya and Walter [18] showed that the use of charcoal for steel production in Brazil contributed to the reduction of waste generation in the study of the car's life cycle. In this sense, this article aims to evaluate and compare the environmental impacts on charcoal production with combustion of the pyrolysis gases through LCA of three processes: without combustion of gases in a rectangular masonry kiln (RK1 Process); with combustion of gases in a rectangular masonry kiln coupled to a masonry furnace (RK2 Process); and with the combustion of the gases in a circular masonry kiln coupled to a metal furnace (CK Process). Thus, the technologies and processes involved were identified: three wood carbonization processes; identification of the relevant impact categories; and evaluation and comparison of the potential environmental impacts of these technologies, using LCA.

This study contributes to development of a technology capable of improving the environmental impact of charcoal production for a large portion of the world population, evaluated by the assessment of the life cycle presented. The search for clean and renewable energy sources has retained the attention of researchers, industries, and society. This study results present the advantages and disadvantages of each process and can stimulate the adoption or public policies on charcoal production technologies that have lower environmental impacts.

The article presents the following structure: Section 2 identifies previous studies on LCA in the production of charcoal and the methods used; Section 3 presents the LCA methodology used to analyze the present study; Section 4 compares the results of the analyzed processes, emphasizing those related to the impacts in the wood carbonization phase found in previous studies; and Section 5 evaluates the limitations of the study and presents suggestions for future studies.

2. Charcoal Production LCA

Standard guidelines for LCA were established by the International Organization for Standardization (ISO), currently standardized by ISO 14040: 2006 and ISO 14044: 2006, in a framework that encompasses the following: goal and scope definition (functional unit/reference flow and impact categories); inventory analysis (technology description, period, region, limitations, validation, data quality criteria, and data collection); impact assessment (characterization); and interpretation (process contribution, elementary flow contribution, and sensibility).

The LCA methodology encompasses a cradle-to-grave (C2Gv) analysis, given the environmental impacts subject to a product or process from the collection or extraction of the inputs, transportation, production, and use or final destination. However, the analysis can be performed in certain parts of the life cycle: cradle-to-gate (C2G) includes resource extraction to manufacturing/service operations, excluding subsequent phases; Gate-to-gate (G2G) is restricted to the manufacturing stage; and gate-to-gate (G2G) includes processes of distribution, use and final disposal of the product [19].

Piekarski et al. [20] consider that among the wide range of methodologies used to analyze the environmental profile of products, the most extensive method is the LCA. Khoo et al. [21] also consider the application of LCA to assess the performance of a technology of interest in economic and social terms.

The LCA has been widely used in the evaluation of the impacts caused in the construction industry [22,23]; in the production of biofuels [24]; in the generation of energy [25] from different biomasses; in teaching and research [26]; and in food systems [27], among many applications. Studies demonstrate the joint use of LCA with other methodologies.

As examples, Theodosiou et al. [28] adopted the LCA methodology with the multicriteria analysis model for the development of an optimization model applied in the analysis of energy systems projects; and Koroneos and Stylos [29] used the Exergy Analysis methodology combined with LCA in a sustainability analysis of energy production in photovoltaic systems.

The first study on the amount of air pollution during wood carbonization in an earth mound kiln occurred in 1994 in Côte d'Ivoire [30]. Emission factor calculations in the production of charcoal were done by other authors [31–35]. As an example, other LCA studies have evaluated the use of forest residues in charcoal production as a way to quantify sequestered carbon [36] and to evaluate the environmental performance of different carbonizers in the production of biochar [37].

A LCA of the charcoal value chain encompasses stages from the wood harvest (originated from natural forest and planted forest), carbonization (kiln efficiency and process energy recovery), transport (logistics), and charcoal utilization (domestic and industrial). It also includes the reuse of process products until final disposal (charcoal fine products and cogeneration of electricity) [1]. Stages of the life cycle in the charcoal value chain presented in previous studies are shown in Table 1.

Table 1. Stages of the life cycle in the charcoal value chain considered in previous studies.

Authors	Year	Local	Stages of the Life Cycle in the Charcoal Value Chain Considered
Khoo et al.	2008	Singapore	The first LCA of biomass conversion. The second LCA adopted a unique approach combining social costs of pollution with the economic factors of the two biomass conversion technologies.
Afrane and Ntiamoah	2011	Ghana	Charcoal production, transportation, and utilization of charcoal, excluding emissions arising from machinery use during biomass cultivation and harvesting.
Bailis et al.	2013	Brazil	Charcoal production was divided into five stages: tree nursery, sowing and management (including carbon sequestration), tree harvesting and transport, kiln construction, and the pyrolysis process (including cogeneration in the container kiln system).
Ekeh et al.	2014	Uganda	Charcoal production, transportation, and utilization of charcoal, excluding emissions arising from machinery use during biomass cultivation and harvesting.
Partey et al.	2017	Ghana	Biomass production including plantation development from nurseries; harvesting and processing of biomass; transportation involving transport of biomass to a charcoal production site; and carbonization and packaging of charcoal

The authors' LCA studies presented in Table 1 do not individually consider all stages of the life cycle in the charcoal value chain. The LCA of Bailis et al. [38] is one of the few to contemplate all stages of this value chain and compares emissions from the use of charcoal with emissions from some alternative cooking fuels.

Khoo et al. [21] evaluated the carbonization of wood residues and briquetted tree prunings. They established mass Functional Unit (FU) for 1 t of the charcoal for the G2G border and analyzed the impact on the global warming potential (GWP), acidification, human toxicity, and photochemical oxidant potential categories. The result of the comparison between the technologies evaluated shows a reduction of GHG emissions (54–85%) and a 90% improvement in human health when using the Japanese carbonizer. The other impact categories did not present significant values in any of the technologies.

The study by Afrane and Ntiamoah [39] analyzed the impacts on charcoal production in earth mound kilns. The use of charcoal was compared to the use of biogas and liquefied petroleum gas (LPG) as alternative sources of energy for cooking. The FU used was the production of 1 MJ of useful energy from each fuel system. The cradle-to-grave boundary (C2Gv) was established and analyzed impacts in the following categories: acidification, eutrophication, freshwater aquatic ecotoxicity, GWP, human toxicity, photochemical ozone creation (smog), and terrestrial ecotoxicity potentials. The result shows that, of the total impact of the GWP assigned to the charcoal, 61% occurred during the production and 39% occurred during the use for cooking.

Bailis et al. [7] compared the production of charcoal in hot-tail kiln—the most common in Brazil—with Rima container kilns (RCK) production, using the gases generated in the wood carbonization process in a Brazilian company producing this input. The carbonizations were performed with eucalyptus from planted forests. The adopted FU was 1 kg of charcoal, and C2G was used as the product system. GHG emissions were analyzed by the 100-year GWP. Other empathy categories were analyzed: energy return on investment; ozone depletion potential; photochemical oxidation; acidification potential; eutrophication potential; and water use. The study verified the possibility of reduction of more than 50% in the carbon footprint in the scenario with charcoal production in RCK and use of all the gases for cogeneration of energy.

Ekeh et al. [40], in Kampala, Uganda, used the GHG emission factors found in the study by Pennise et al. [33] to compare with emissions from charcoal production in a CH_4-free process technology ("PYREG methane-free charcoal production equipment"), using wood from a sustainably managed plantation. The established FU was 1 kg of charcoal at the C2G boundary and analyzed the GWP.

This comparison showed that GHG emissions in the carbonization phase decreased by approximately 28% when the PYREG process was used.

Partey et al. [41], in Ghana, compared the impacts related to the carbonization of three biomass species of sustainable origin, using circular masonry kilns. The established FU was 1 MJ energy produced from the three species, at the C2G boundary.

In relation to the LCA methods used in previous studies, GaBi [21,39,40] and SimaPro [7,41] software packages were used in the previous studies using GaBi [21,39,40] and SEMCo [7]. The life cycle impact assessment methods used were EDIP 1997 [21], CML 2001 [39–41], Ecoinvent data v2.1 [7], and Ecoinvent v3 and Idemat 2015 [41]. The analysis of data quality in these studies was not evidenced. The sensitivity analysis was presented by Ekeh et al. [40] and Bailis et al. [7].

3. Research Methodology

This LCA compares three processes of charcoal production in masonry kilns coupled to furnaces for the combustion of carbonization gases: (A) a process without gas combustion in the RK1 Process; (B) a process with combustion of gases in the RK2 Process; and (C) a process with combustion of gases in CK Process coupled to a metal furnace.

3.1. Definition of Scope

In this evaluation, several environmental impacts were analyzed related to small scale artisanal production in circular masonry kilns, which represent about 70% [42] of Brazilian production, considering the combustion of the carbonization gases. In the same way, the environmental impacts caused by the production in rectangular kilns, which represent another 20% of the national production, were analyzed. They considered processes without combustion and with the combustion of the gases during the carbonization of the wood, as listed below.

3.1.1. Functional Unit (FU)

The present study chose to work with calorific value of the charcoal to fulfill the function of the product during the phase of use, in a similar way to the different qualities of the charcoal produced among the technologies evaluated. The FU defined for the study was 41.84 MJ (10,000 kcal) from charcoal. The use of calorific value as FU was chosen because the same mass of charcoal can supply different amounts of energy (which is the function of the charcoal energy). For the charcoal consumer, the amount of energy generated is more important than the mass. The FU was established from the higher calorific value (HCV) of the produced charcoal. Thus, the reference results are shown in Table 2.

Table 2. Reference Flows.

Study's Technology and Reference	Combustion Condition	Higher Calorific Value HCV (MJ/kg)	Reference Flow (kg)
CK Process	With combustion	32.56	1.285
RK2 Process	With combustion	33.18	1.261
RK1 Process	Without combustion	32.06	1.305

Similar criteria were used by Rousset et al. [43], which adopted the delivery 1 kg of briquette energy content to the destination port as the FU.

3.1.2. Product System

The product systems defined in the previous studies were G2G [19], C2Gv [39,40], and C2G [7,41]. The system boundary refers to the analysis of product life cycle stages. The analyzed product system is presented in Figure 1.

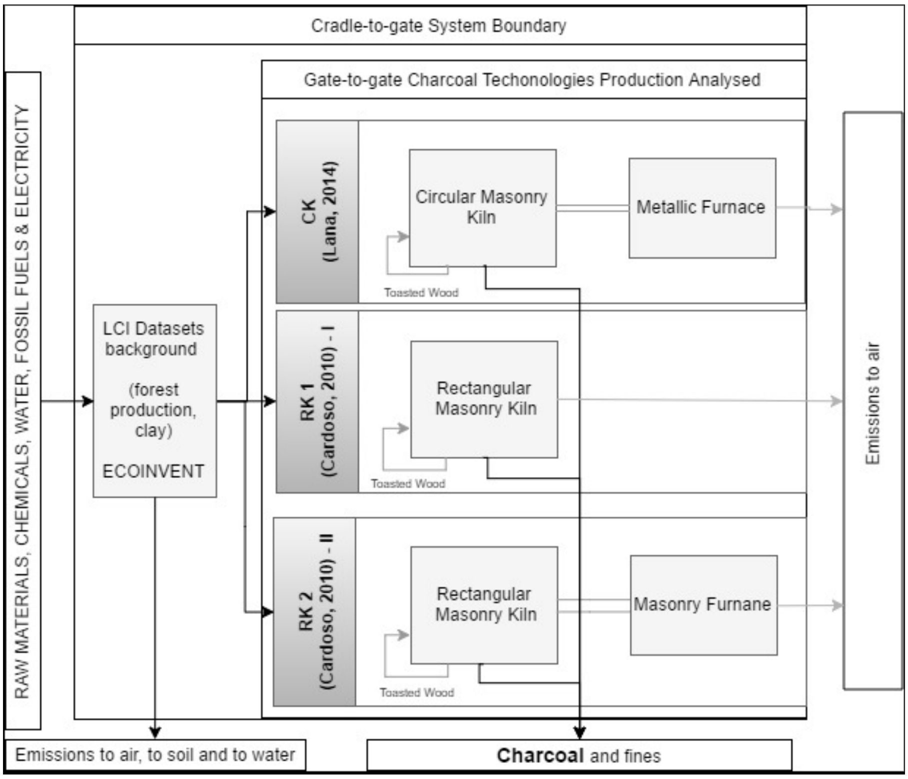

Figure 1. Product system considering the cradle-to-gate system boundary for the three processes analyzed.

The technological processes data used as background in the study were from the ecoinvent database. For the clay input, the dataset used was: "Clay {GLO} | Market for | AllocDef, U". For wood, the dataset used was: "Roundwood {GLO} | Harvest, secondaryforest | AllocDef, U". The kiln data were obtained from literature (see Table 3).

3.1.3. Allocation

Allocation is necessary in the carbonization process because, besides the charcoal, charcoal fines are also produced. Usually, the larger pieces are commercially valued and charcoal fines are disregarded [44]. The charcoal fines have potential for use as an energy input [10,43], and their use contributes to the reduction of GHG emissions [1]. There is no reference to fines in previous studies.

The charcoal was considered as the main stream, that is, the entire impact was allocated to this portion. In this research, the same values of fixed carbon and (HCV) of the produced charcoal were attributed to the charcoal fines. However, there is no denying that HCV of the charcoal fines can decrease due to impurities or contamination when collected at the end of kiln material unloading [45]. In addition, a sensitivity analysis was performed considering three allocation factors: mass, energy, and economics.

3.1.4. Selection of Impact Assessment Method

The selection of the impact assessment method to be applied in the study was performed in the following two steps:

(1) Identification of the impact categories relevant to carbonization processes: A study by Cardoso [12] aimed to evaluate the influence of the gas combustion in the masonry furnace on the yield and quality of the charcoal. The burning of the carbonization pyrolysis gases showed significant reductions in emissions of CO, CH_4, and particulate matter (PM). In turn, Lana [9]

sought to evaluate the reduction of GHG emissions from the burning of the carbonization gases in a metal furnace.

(2) Identification of the impact assessment method contained in these impact categories: For the selection of the life cycle impact assessment method, an analysis was performed on which of the methods covered the previously listed impact categories (i.e., climate change, particulate matter emission, ozone layer depletion, photochemical oxidation, acidification, eutrophication, and water consumption). In addition, fossil energy, abiotic resources, and land use were included, since they are usually evaluated when comparing fossil energy with renewable energy sources. Identification of the impact assessment method was included in these impact categories.

The ReCiPe Endpoint considered all these impact categories, justifying their selection. The version used was (H/A) V.1.12. Calculations were performed using the SimaPro 8.2.3.0 software.

3.2. Life Cycle Inventory (LCI)

The data used were taken from two studies carried out in the Madeira Panels and Energy Laboratory-LAPEM, Federal University of Viçosa-UFV, located in the State of Minas Gerais, the primary producing and consuming state of charcoal in Brazil [4], as described as follows:

(a) A rectangular masonry kiln with capacity of 7.4 m^3 of encased wood was built on a small scale to portray the reality of the industrial kilns used by large producers of charcoal. A connector duct links the kiln to the furnace, both built in the same masonry kiln form, in addition to the cylindrical chimney, as shown in Figure 2.

Figure 2. Kiln-furnace systems of RK1 and RK2 Processes [46].

The process data were collected during 2009 by Cardoso [12] and include carbonization processes using the RK1 Process and the RK2 Process, as presented in Table 3. The Environmental Protection Agency (EPA) standards were used to collect and analyze the gas samples, per Method 18: collection in bags.

(b) A circular masonry kiln with capacity to 8.5 m^3 of wood was connected to a furnace with a metal duct. The furnace, set in a masonry base, was composed of two cylindrical metallic parts, the first one by a combustion chamber and the second by a chimney, as shown in perspective in Figure 3.

Table 3. Technical data of experiments

Sources/Technical Characteristics	Measurement Unit	Lana [9]	Cardoso [12]	
Type of Kiln		Circular masonry	Rectangular Masonry	
Kiln Capacity	m^3	8.5	7.4	
Base Furnace-material	-	Masonry	Masonry	
Furnace-material	-	Metallic	Masonry	
Chimney-material	-	Metallic	Masonry	
Physical-chemical properties of wood per carbonization type		CK Process	RK2 Process	RK1 Process
Wood put in the kiln on a wet basis	Kg	4859.00	3578.50	3428.50
Wood put in the kiln on a dry basis	Kg	3037.00	2680.35	2640.35
Species and Genus	-	*Eucalyptus* spp.		
Age	years	6	8	8
Moisture content	%	37.5	25.04	23.17
Higher calorific value	MJ/kg	Nd	19.77	19.77
Kiln Efficiency: Total Pyrolysis Time	H	78	52	52
Efficiency of the Furnace				
Total Burn Time (TBT)	H	33	28	-
Concentration Reduction				
Methane (CH_4)	%	88.03	96.95	-
Carbon Monoxide (CO)	%	86.24	93.76	-
Particulate Material	%	nd	95	-
Gravimetric Yields:				
Charcoal gravimetric yield	%	33.13	28.73	28.15
Toasted wood gravimetric yield	%	3.27	3.51	12.50
Charcoal fines gravimetric yield	%	2.92	4.32	3.25
Produced Charcoal Properties:				
Higher calorific value	MJ/kg	32.56	33.18	32.06
Fixed Carbon Content	%	79.04	82.56	75.45

Figure 3. Kiln-furnace systems of Process CK [9].

The process data were collected between September and December 2013 by Lana [9] and evaluate carbonizations using the CK process. The process data consider two carbonizations with flaring of the gases and are presented in Table 2. For characterization and quantification of the gases in, and emitted from, this process, *Gasboard 9030* and *3100 Wuhan CUBIC Optoeletronics Co. Ltd (China)* were used.

3.2.1. Data Collection

Eucalyptus spp. wood, originating from commercial forest plantations, was used in all carbonizations, and the physicochemical characteristics are presented in Table 3.

It is observed that the reductions of CH_4 and CO emissions were significant for both technologies. However, it is worth mentioning that the lower percentage in the gas reduction obtained in the CK Process can be explained, at least in part, by the following: furnace design; the quality of the gases generated during the process; and variables dependent on numerous factors related to the wood and to the carbonization process. Thus, the residence time of the gases inside the furnace was not sufficient for the total combustion of these gases to occur.

3.2.2. Estimation of Emissions

Not all emissions were obtained in the laboratory, and it was necessary to perform calculations. For this, the chemical composition of the different substances was estimated according to the percentages presented in Table 4, which identifies the products of the carbonization. The products are separated into fractions, with 33% being coal (solid fraction) and 42% and 25% being the condensable and non-condensable gaseous fractions, respectively. The phenolic compounds of insoluble tar were identified and presented in Alves [47].

Table 4. Carbonization products per ton of wood as a Dry Weight %.

Carbonization Products	% Dry Weight	
Charcoal (80% Fixed Carbon)		33.00
Condensable Gases		42.00
Pyroligneous Acid	35.50	
(Acetic Acid)	(5.00)	
(Methanol)	(2.00)	
(Soluble Tar)	(5.00)	
Water and others	(23.50)	
Insoluble Tar	6.50	
(Phenol—6.00%)	(0.39)	
(Guaiacol—6.00%)	(0.39)	
(2,6-Xylenol—1.00%)	(0.065)	
(Cresol—1.00%)	(0.065)	
(o-Cresol—6.00%)	(0.39)	
(p,m-Cresol—7.00%)	(0.45)	
(4-Ethyl-guaiacol—1.00%)	(0.065)	
(4-Propyl-guaiacol—0.10%)	(0.0065)	
(3,5-Xylenol—5.00%)	(0.325)	
(Syringol—35.00%)	(2.275)	
(4-Methol-syringol—11.00%)	(0.715)	
(4-Ethyll-syringol—5.00%)	(0.325)	
(Water (16~20%)—15.90%)	(1.0335)	
Non-Condensable Gases		25.00
(H_2—0.63%)	(0.16)	
(CO—34%)	(8.50)	
(CO_2—62%)	(15.50)	
(CH_4—2.43%)	(0.61)	
(C_2H_6–0.13%)	(0.03)	
(Others—0.81%)	(0.20)	
Total		100.00

Source: Adapted from Alves [47], Gomes and Oliveira [48], and Ferreira [49].

Note that, from the database, the following were used for identifying products: 3,5-Dimethoxyphenol for syringol, 4-ethyl-syringol, and 4-methyl-syringol; M-cresol for p,m cresol; guaiacol for 4-ethyl-guaiacol; and 4-methyl-guaiacol for 3,5-Xylenol. Although they are not the same compounds because they occupy different positions in the ring, they were used because they have similar chemical functions.

The relative values of NO_2 and SO_2 were obtained considering the transformation of these oxides, due to the difference between the sulfur and nitrogen present in the wood at six years of age as well as in the obtained charcoal. The values of N (0.205%) and S (0.325%) for wood and N (0.170%) and S (0.030%) for charcoal were obtained from Soares et al. [50], calculated by the mean of their values at five and seven years of age.

3.2.3. Validation of Data

The mass balance of the three processes was elaborated based on Section 3.2.2, adapted from Alves [47]; Gomes and Oliveira [48]; Ferreira [49]; and Soares et al. [50]. Data validation was not specified in previous studies except for Ekeh et al. [40].

3.3. LCA Data Quality Analysis

The criteria for data quality analysis used in this study are positioned in Column 1 of Table 5, and data quality requirements are presented in Column 2.

Table 5. Data Quality Analysis via Pedigree Matrix [51].

Criteria	**Desired Information**	**Information Obtained**	
		Cardoso	**Lana**
Technology	In use	Rectangular masonry kiln system, gas combustion chamber, and masonry chimney.	Circular masonry kiln system, gas combustion chamber, and metal chimney.
Temporal	2016	Data from 2010: six years difference compared to the year of this study.	Data for 2014: two years difference compared to the year of this study.
Location	State of Minas Gerais	Data from a similar area, on a prototype scale.	Data of a representative area of the sector, per unit (kiln).
Completeness	100% of cases	If the combustion of the gases is considered, the data represent some locations, and without consideration for gas combustion, the data represent about 10% of the sector.	If the combustion of the gases is considered, data are of unknown representation; and without consideration for gas combustion, the data represent about 70% of the sector.
Consistency	Data collection and method for calculating emissions	Thermocouples for temperature measurements; Pitot tube type "S" for collection of gases in the chimney; Portable barometer for determination of local atmospheric pressure, ABNT [1] standards; EPA [2], Method 18: Collection in bags; and ASTM [3] 1945 for procedures for collection, analysis, storage of samples, and determination of gas concentrations.	Type K thermocouples for temperature measurements; Copper pipes for the collection of gases in the conduit and in the chimney; *Gasboard* 9030 *Wuhan CUBICOptoelectronics Co. Ltd.*, precision filters-FIT1 and FIT2; *Gasboard* 3100 *Wuhan CUBIC Optoelectronics Co. Ltd.*, for procedures for collecting and reading the percentage composition of the gas volume.

Notes: [1] ABNT: Brazilian Association of Technical Standards; [2] EPA: Environmental Protection Agency; [3] ASTM: American Society for Testing and Materials.

In the previous studies, the criteria for data quality analysis were not evidenced. However, Bailis et al. [7] justify the use of emissions data from previous studies and recommend local measurements as a way to increase the degree of accuracy of certain environmental results for the change proposed by the group. Ekeh et al. [40], likewise, use data from previous studies to estimate future emissions in the production and use of charcoal in African countries.

4. Results and Discussion

In this work, the relative environmental impacts of the three technologies mentioned in the scope for the production of charcoal were evaluated. When evaluating only the environmental impacts of the carbonization process and the furnace (G2G), it can be observed that the burning of the gases is significantly better when compared to the direct emissions to the atmosphere. The RK2 Process data are the relatively least impacting G2G boundary, as shown in Figure 4. In general, it is evident that the inclusion of furnaces for the gas combustion reduces all categories of potential environmental impacts by at least 90% for both the CK Process and the RK2 Process.

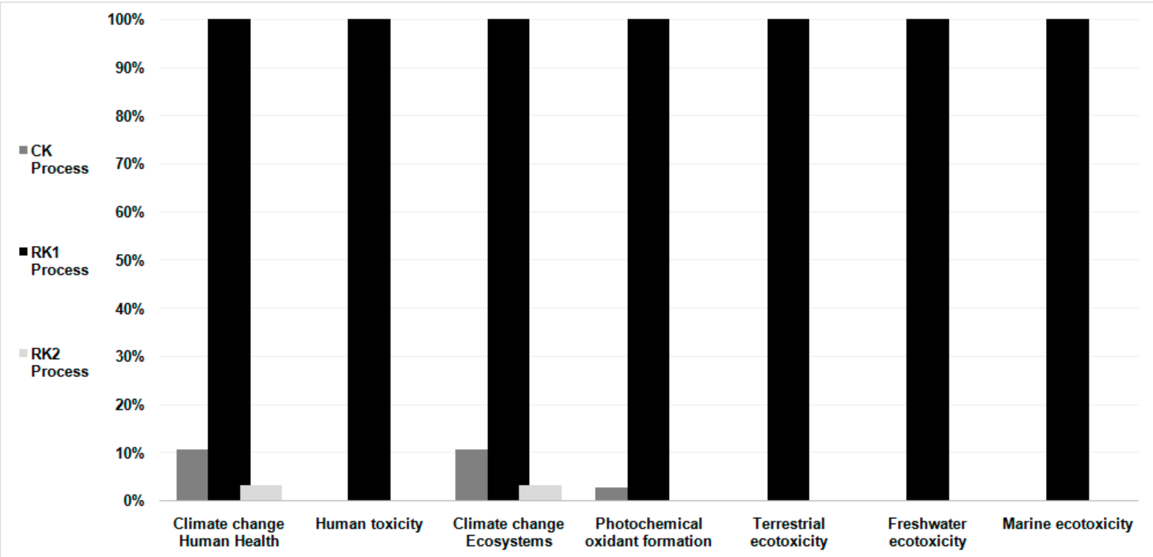

Figure 4. Relative environmental impacts per technological system.

In a second analysis, the calculations were carried out to verify the influence of NO_2 and SO_2 emissions on the G2G analysis for the three technologies analyzed. The emission impacts of particulate matter, photochemical oxidant formation, and terrestrial acidification have been altered due to NO_2 and SO_2 emissions.

For the categories of particulate matter emission, photochemical oxidant formation, and terrestrial acidification, it can be observed that the CK process presents better results, as shown in Figure 5.

Regarding climate change, it is observed that the impact reduction with the combustion of gases is 90% for the CK process in the G2G system. However, in the charcoal production process (G2G), the best technology in terms of climate change was the RK2 Process, having approximately 63% less impact than the CK process.

The search for carbonization alternatives with gas combustion to generate benefits for environmental protection and use of renewable energies is also observed in the study by Khoo et al. [21]. The authors found that, in a Japanese carbonization model, approximately 85% less GHG were produced than in the conventional carbonization system, and the combustion furnaces (RK2 Process and CK Process) presented more than 90% reduction, in terms of impacts on climate change. The study by Khoo et al. [21] also demonstrated reduction in impacts related to acidic gas emissions in the gas combustion process. This corroborates the results of this study, where gas combustion technologies reduce the impacts of acidification, photochemical oxidant potential, and human toxicity.

The study by Ekeh et al. [40], which quantified GHGs from the production, transport, and distribution of charcoal in an earth mound kiln of Uganda, found that approximately 53.3% of the impacts related to climate change (828,316 $tCO_2eq.$) are related to the charcoal production phase. The authors verified the potential of reducing GHG emission through a reduction of "free-methane" in the charcoal production process and the use of non-sustainable wood in the process. In turn, Partey et al. [41] found that the pyrolysis and transport phases did not exceed 10% of the total ecological impact cost in the GWP categories, human health, and ecosystems when analyzing the impacts of production (from nursery of seedlings) and the use of three forest species for charcoal production in C2G boundary.

In this sense, by expanding the boundaries of the study and connecting the technological system of wood production with the ecoinvent database, as presented previously in Figure 1, the results of the C2G impacts can be obtained. The results are presented by category in Figure 6.

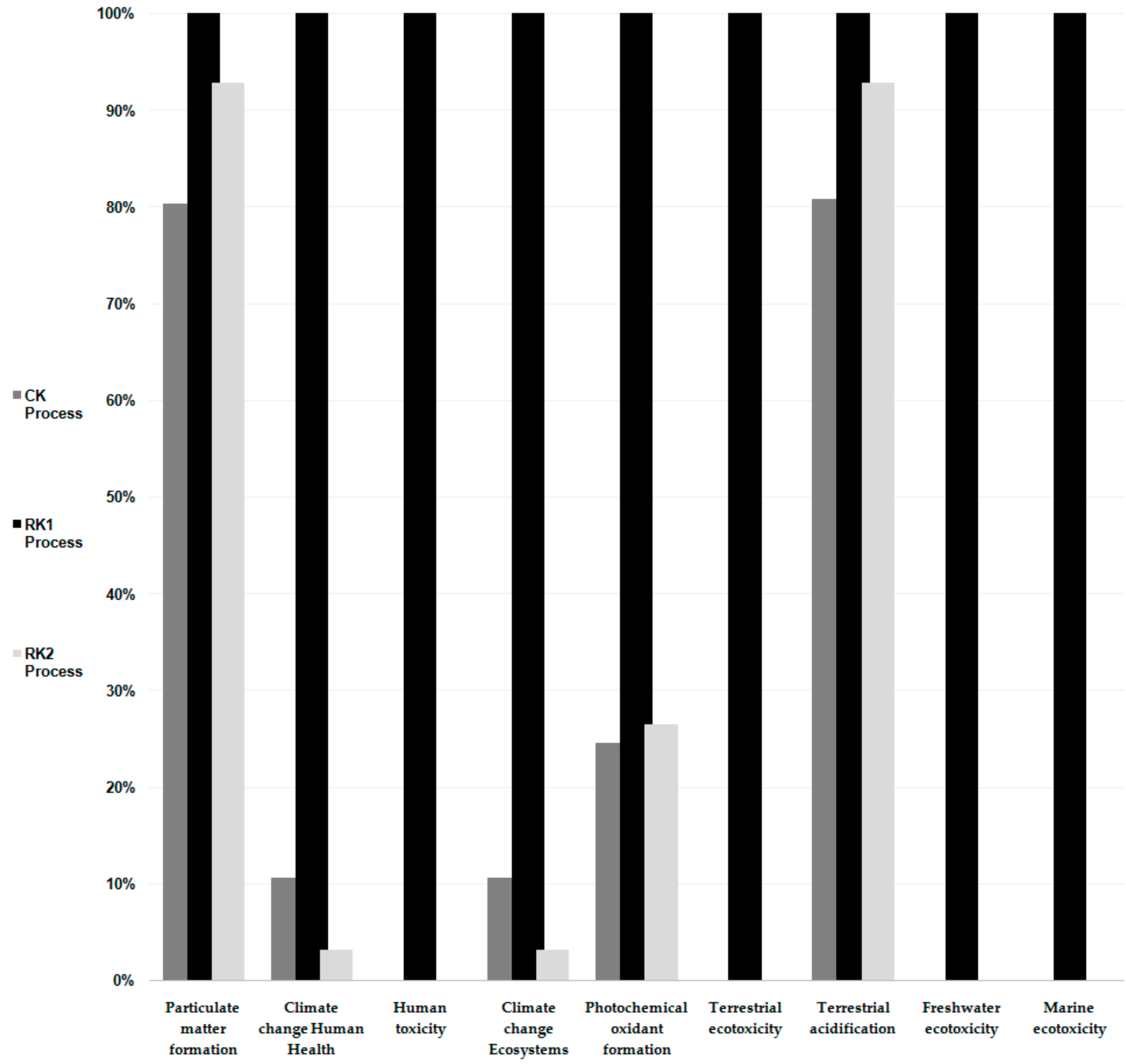

Figure 5. Environmental impacts per technological system considering NO_2 and SO_2.

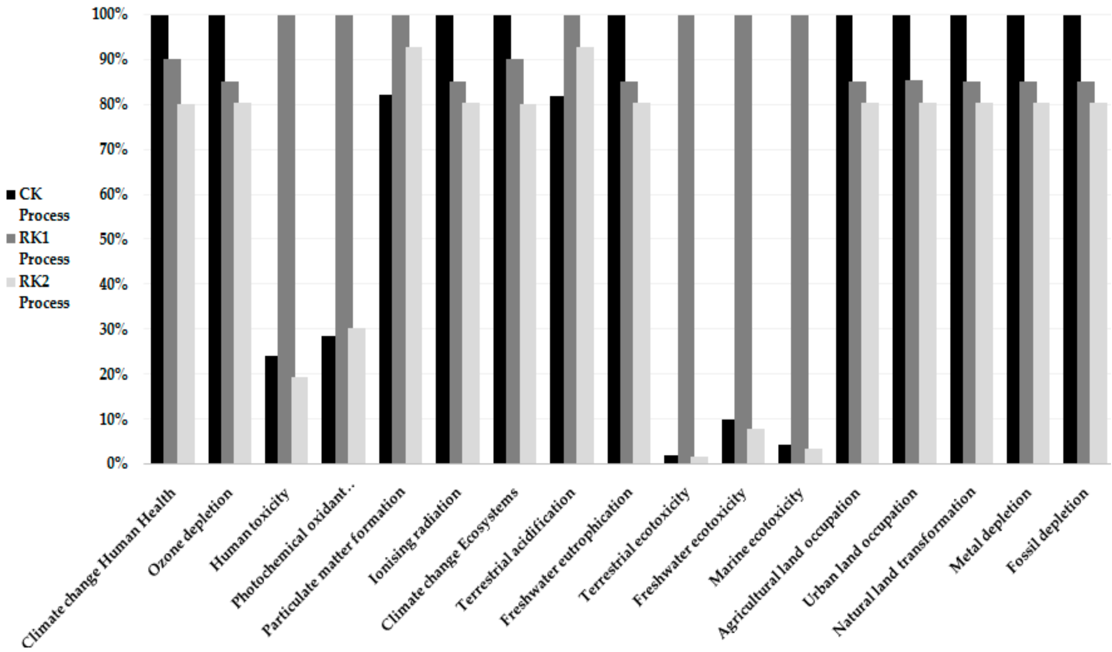

Figure 6. Environmental impacts per technological system, according to Ecoinvent's database.

When considering the life cycle of charcoal production at the C2G boundaries, it can be observed that environmental impacts have a major dependence on the efficiency of each type of technological process in charcoal production. It was observed, for example, that, for the climate change impact category, the gas-fired circular kiln technology (CK Process) had the highest impact (1.38×10^{-5} DALY/41.84 MJ of charcoal), while the best environmental performance for this category was presented by the RK2 Process (1.10×10^{-6} DALY/41.84 MJ of charcoal). In Figure 5, it can be seen that the CK Process has 90% better performance for the impact of climate change compared to the RK1 Process, at the G2G boundary. The fact that the CK process consumes 15% more wood (on a dry basis) when compared to the RK1 Process (see Table 3) results in a higher C2G impact. The G2G boundary represents about 6% of the total impacts of the climate change categories for the RK1 Process. As for the processes with gas combustion, the impact drops to 0.58% and 0.52% (CK Process and RK2 Process, respectively). That is to say, it can be observed that when the impacts of the G2G boundary is low, the behavior of the environmental impacts will be associated with the efficiency of the charcoal production kilns. The efficiency of the analyzed processes is related to the data collected from LCI, and they can be analyzed as points of generation of innovation in the industry, as suggested by Luz et al. [52].

The categories of environmental impacts that presented a low percentage impacts (<10%) for the G2G boundary in the life cycle of charcoal production were: agricultural land occupation, climate change ecosystems, climate change human health, fossil depletion, freshwater eutrophication, ionizing radiation, metal depletion, natural land transformation, ozone depletion, and urban land occupation. For all of these categories, the CK Process had a higher environmental impact (Figure 6) because it had higher wood consumption (on a dry basis) to generate the same calorific value from the charcoal (41.84 MJ) when compared to the rectangular kilns. However, the categories that have a significant impact for the G2G boundary in the charcoal life cycle (C2G) have been shown to be dependent on the impacts generated by the non-combustion of the gases in the processes. In other words, the RK1 Process (which does not burn the combustion gases) had the highest relative environmental impact for the other categories.

In this sense, analyses on the influence of NO_2 and SO_2 emissions during carbonization in C2G analysis were performed. It can be affirmed that the C2G results undergo variations when including NO_2 and SO_2 emissions during carbonization. This is because the carbonization process is one of the most impactful in some categories of the charcoal life cycle. To analyze the relative contribution of the carbonization process in the charcoal production for the three different technologies listed in this study, all impact categories in which the carbonization processes presented contributions above 1% were identified (Table 6).

Table 6. Contributions greater than 1% of the elementary flow of carbonization processes per technological system.

Impact Categories	RK1 Process	RK2 Process	CK Process
Photochemical oxidant formation	96%	87%	83%
Particulate matter formation	95%	95%	93%
Terrestrial acidification	97%	97%	96%
Terrestrial ecotoxicity	98%	-	-
Freshwater ecotoxicity	92%	-	-
Marine ecotoxicity	96%	-	-
Climate change Human Health	6%	-	1%
Climate change Ecosystems	6%	-	1%

Only the categories of photochemical oxidation formation, particulate matter formation, and terrestrial acidification presented contributions above 1% for the three technologies analyzed in this study. Among them, the terrestrial acidification that presents percentage above 95% in all the technologies is highlighted, especially due to the emissions of NO_2 and SO_2.

According to Table 6, the RK1 Process has impact greater than 1% in eight impact categories due to non-combustion of the gases. The greater contributions are from the terrestrial ecotoxicity impact (98%), especially due to the emission of acetic acid. Additionally, to identify the substances that contribute most to the impact categories displayed in Table 6, Table 7 lists the ones whose values surpass 1% of impact contribution in the three technologies.

Table 7. Contribution of more than 1% of the elementary flow of carbonization per process in the life cycle.

Impact Categories	RK1 Process							
	1	2	3	4	5	6	7	8
Reference	3.7960×10^{-9}	3.09655×10^{-6}	2.59241×10^{-10}	1.77076×10^{-9}	1.13728×10^{-12}	4.10×10^{-13}	1.24019×10^{-5}	7.02×10^{-8}
Methane, biogenic	-	-	-	-	-	-	6%	6%
3,5-Dimethoxyphenol	-	-	-	9%	16%	7%	-	-
3,5-Dimethylphenol	-	-	-	-	6%	4%	-	-
Acetic acid	33%	-	-	79%	39%	14%	-	-
Cresol	-	-	-	-	3%	2%	-	-
Guaiacol	-	-	-	4%	7%	6%	-	-
m-Cresol	-	-	-	2%	10%	5%	-	-
o-Cresol	-	-	-	-	3%	2%	-	-
Phenol, 2,6-dimethyl	-	-	-	1%	6%	55%	-	-
Nitrogen dioxide	25%	45%	30%	-	-	-	-	-
Sulfur dioxide	2%	50%	67%	-	-	-	-	-
CO, biogenic	16%	-	-	-	-	-	-	-
Methanol	19%	-	-	-	-	-	-	-
Bromine	-	-	-	-	5%	0%	-	-
Copper	-	-	-	-	-	1%	-	-
Zinc	-	-	-	-	-	1%	-	-

Table 7. *Cont.*

Barium	-	-	-	-	0%	-	-	-
Cypermethrin	-	-	-	-	2%	-	-	-
Phenol	-	-	-	-	1%	-	-	-
Impact Categories	**RK2 Process**							
	1	**2**	**3**	**4**	**5**	**6**	**7**	**8**
Reference	1.15185 $\times 10^{-9}$	2.876 $\times 10^{-6}$	2.40689 $\times 10^{-10}$	2.66101 $\times 10^{-11}$	8.9577 $\times 10^{-14}$	1.41 $\times 10^{-14}$	1.10105 $\times 10^{-5}$	6.2366 $\times 10^{-8}$
Methane, biogenic	-	-	-	-	-	-	0%	0%
Acetic acid	-	-	-	-	-	-	-	-
Nitrogen dioxide	76%	45%	30%	-	-	-	-	-
Sulfur dioxide	8%	50%	67%	-	-	-	-	-
CO, biogenic	-	-	-	-	-	-	-	-
Methanol	-	-	-	-	-	-	-	-
Bromine	-	-	-	-	56%	11%	-	-
Copper	-	-	-	-	-	29%	-	-
Zinc	-	-	-	-	-	18%	-	-
Barium	-	-	-	-	6%	-	-	-
Cypermethrin	-	-	-	-	21%	-	-	-
Phenol	-	-	-	-	0%	-	-	-
Impact Categories	**CK Process**							
	1		**1**		**1**		**1**	
Reference	1.07581 $\times 10^{-9}$	2.54361 $\times 10^{-6}$	2.12153 $\times 10^{-10}$	3.31272 $\times 10^{-11}$	1.11511 $\times 10^{-13}$	1.76 $\times 10^{-14}$	1.37575 $\times 10^{-5}$	7.79245 $\times 10^{-8}$
Methane, biogenic	-	-	-	-	-	-	1%	1%
Acetic acid	-	-	-	-	-	-	-	-
Nitrogen dioxide	69%	43%	29%	-	-	-	-	-
Sulfur dioxide	7%	50%	67%	-	-	-	-	-
CO, biogenic	7%	-	-	-	-	-	-	-
Bromine	-	-	-	-	56%	11%	-	-
Copper	-	-	-	-	-	29%	-	-
Zinc	-	-	-	-	-	18%	-	-
Barium	-	-	-	-	6%	-	-	-
Cypermethrin	-	-	-	-	21%	-	-	-
Phenol	-	-	-	-	-	-	-	-

When comparing the RK1 Process and the RK2 Process, the contribution of NO_2 and SO_2 emissions is approximately 97% for terrestrial acidification. In the formation of photochemical oxidation, NO_2 has a change in contribution, from 76% in RK2 to 25% in RK1. Acetic acid contributes to photochemical oxidation (33%), terrestrial ecotoxicity (79%), fresh water ecotoxicity (39%), and marine ecotoxicity (14%) in the RK1 Process. Phenol, 2,6-dimethyl has a contribution of 55% for marine ecotoxicity when carbonization is performed in the RK1 Process.

A sensitivity analysis of the allocation factors was performed, and the result is presented in Figure 7.

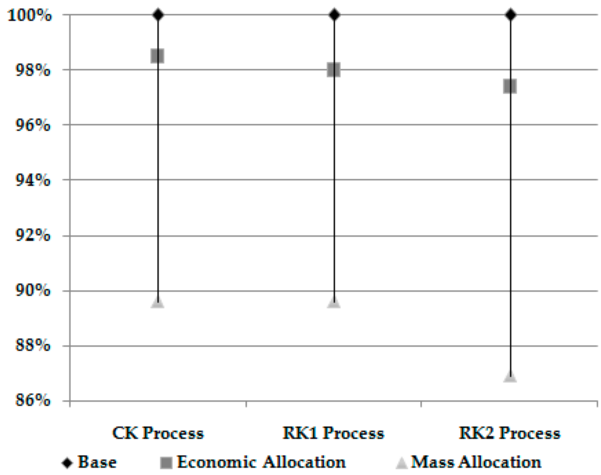

Figure 7. Sensibility analysis in therms of economic and mass allocation for the three processes analyzed.

Figure 7 shows that, in the economic allocation for the CK process, 99% was allocated to the charcoal relative to the base, and 1% was allocated to the fines. For RK1 and RK2, 98% and 97% were allocated to charcoal and the other 2% and 3% to charcoal fines, respectively. Considering the same basis, the mass allocation shows that 90% was allocated to charcoal and 10% to charcoal fines. For RK1 and RK2, 90% and 87% were allocated to the charcoal and the other 10% and 13% to charcoal fines, respectively.

The sensitivity analysis also considered the energy factor. However, the values were the same as those for the mass factor.

5. Conclusions and Limitations

The search for clean and renewable energy sources is of great importance for researchers, society, industries, and public policies. This study allowed for conclusion that different technologies of charcoal production have different environmental performances, which are linked to two major aspects: equipment efficiency and impacts on the G2G boundary.

The substances and impact categories, which do not have high large contribution in the total environmental impact on the cradle-to-gate boundary of the charcoal, suggest that the processes with greater technical efficiency in the conversion of wood to the calorific value for the produced charcoal are the ones that have better environmental performance. In other words, the potential environmental impacts for agricultural land occupation, climate change ecosystems, climate change human health, fossil depletion, freshwater eutrophication, ionizing radiation, metal depletion, natural land transformation, ozone depletion, and urban land occupation tend to be smaller for processes with better efficiency in the charcoal production process. In this study, the best performance for these categories was the RK2 Process. On the other hand, the environmental performance was also better for processes with the combustion of gases in the furnace. There are impact categories that have more impact (>80%) for the G2G boundary in C2G (photochemical oxidant formation, terrestrial acidification, terrestrial ecotoxicity, freshwater ecotoxicity, and marine ecotoxicity). For these categories, the results indicated that the technologies evaluated have a better environmental performance when the furnace gases burn.

In the G2G analysis, the RK2 Process presented the best performance when not considering NO_2 and SO_2. When considering NO_2 and SO_2, there were changes in the particulate matter and terrestrial acidification emission impacts, and, for these two categories, the CK process presented the best performance (C2G). However, it did not present the best performance in terms of consumption of wood vs. calorific value produced from the charcoal. For the other categories (photochemical oxidant formation, terrestrial ecotoxicity, freshwater ecotoxicity, and marine ecotoxicity), the RK2 Process maintained the best environmental performance. The study suggests that the rectangular masonry kiln coupled with the masonry furnace (RK2 Process) presented the best environmental performance in general. Actions that result in reduction in NO_2 and SO_2 emissions can be developed for this process to improve environmental performance in the impact categories of particulate matter and terrestrial acidification, where the circular masonry kiln coupled with the metal furnace (CK Process) presented better performance.

According to Bailis et al. [7], theoretically, the environmental impacts on charcoal production can be reduced with the implementation of pollution controls, as is the case of the gas combustion furnaces analyzed in this work. However, there is little regulatory and fiscal incentive for the deployment, development, and investment of such technologies in rural Brazilian areas. Pereira et al. [10] state that the combustion practice of carbonization gases will reach the entire charcoal production chain in a solid and cohesive way when society's knowledge is disseminated and all technical difficulties are overcome by the productive sector.

Theoretical and Practical Implications

This case study has theoretical and practical value for researchers. It is necessary to advance technological development to increase the efficiency of equipment for the production of charcoal. The most efficient equipment for wood-to-charcoal conversion is one that has the best environmental performance.

Another practical implication highlighted in the study is technological development in the gate-to-gate frontier of charcoal production. Even though the environmental impacts of carbonization are relatively low considering for the entire life cycle, emissions can have significant impacts, both locally and regionally. Although the carbonization gas combustion technologies presented in this study are in development, it is of great importance that new studies evaluate the following: the synchrony between kilns coupled to the same furnace; obtaining the optimum temperature and flow rate of the gases in order to favor their combustion; and the residence time of these gases inside the combustion chamber. These efforts aim to decrease the use of auxiliary fuel and reduce CO and CH_4 emissions. For the gate-to-gate boundaries, gas-fired furnaces showed the best environmental performance.

In addition, process residues that are used for fertilization or other purposes should be evaluated in view of the need to analyze other chemical compounds, such as heavy metals.

Finally, the results presented in this work contribute to the development of life cycle inventories for charcoal production in Brazil. These results can be useful in understanding the advantages and disadvantages of different technologies for the production of charcoal, besides stimulating public policies to encourage the installation of charcoal production technologies with lower environmental impacts and the development of new clean technologies.

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Author Contributions: Miranda Santos wrote items 1–3 and calculated mass and energy balances with the assistance of Piekarski and Ugaya, who supported the development of the LCA study. Donato and Carvalho evaluated and contributed with the information regarding the pyrolysis gas combustion process. Braghini Junior and De Francisco guided authors on the objectives of this study. All authors analyzed the data, and read, and approved the manuscript.

Conflicts of Interest: The authors declare no conflicts of interest.

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