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Solid biofuels: Environmental concern and integrated analysis of energy sustainability

Cristian Bozzato, Md. Badiuzzaman Khan, and Bruno Pavoni

Dipartimento di Scienze Ambientali, Informatica e Statistica,
Università Ca' Foscari Venezia,
Dorsoduro 2137, Venice 30123, Italy

ABSTRACT: A co-generative plant situated in Northern Italy (CPN) was used to test an approach for evaluating the environmental pros and contras of using wood chips of different origin as biofuel. The EROI (Energy Return On Energy Investment) of the plant was also assessed. Used woodchips were of the two main categories and were obtained from i) natural ligneous trees from energy crops and ii) uncontaminated ligneous biomasses from agriculture and forestry wastes. Woodchips were characterized by determining the relevant physical and chemical properties for the plant functioning. Biofuels used in CPN have different physical properties and this may be the reason explaining a lower performance of some features (LHV and bulk density) of the wood chips from energy crops compared to agriculture and forestry recovery materials. Although recovered material contained higher Cl, S and heavy metal levels than wood chips from energy crops, but LHV (Low Heating Value) and lower price of recovery materials suggested that this one could be a good alternative source of energy especially in developing countries. This observation demands frequent integrity checks of the pollutants in order to guarantee a low level of environmental risks. This results obtained enable a comparison between the different kinds of materials used and an energy analysis to assess sustainability in the studied territory.

KEYWORDS: Biofuels, wood-chips, Chemical characterization, EROI.

1 INTRODUCTION

Energy is a key factor for the generation of wealth and an important prerequisite for economic development. Globally, 80% of global energy production is based on fossil fuels like coal or oil [1]. However, adverse impact on human health and environment such as global warming and acid rain due to emissions from fossil fuels have stimulated the use of renewable energy resources (RESs) [2]. Moreover, the European Commission EU-20 strategy [20% reduction of greenhouse gases, 20% increase in the national energy production based on renewable sources and another 20% of improvement in energy efficiency] has forced the European countries to become more responsible for using renewable energy resources. Among the main RESs, including hydroelectric, solar (thermosolar and photovoltaic), wind and aeolian sources, biomasses appear to be the most versatile. In addition, according to the Kyoto Protocol, biomasses appear a dependable and environmentally safe energy supplier. In Italy, it was shown that biomasses may have a great role in achieving the objectives set out in the framework of the EU 2020 strategy [3]. However, there are several issues (especially long-term availability, technology, price changes, resource issues and logistics) that need to be improved to enable a partial or total replacement of fossil fuels. Currently, research is going on all over the world to identify more sustainable conversion technologies. Of course, like all technologies, the improvement of the process can only be achieved by knowing the full properties of the material. Moreover, energy production is also influenced by several factors, firstly, the physical and chemical properties of biomasses; secondly, biomass exploitation depends on logistic needs [4]. Physical properties have been described by a number of authors ([5], [6]) and the most important ones are bulk density, calorific value, particle size distribution, moisture and ash content. More details are added as *Supplementary material 1.1.*

On the other hand, various harmful pollutants such as Polycyclic Aromatic Hydrocarbons (PAHs), dioxins (PCDD/F), nitrogen oxides (NO_x), sulfur oxides (SO_x), and heavy metals (HMs) are emitted to the atmosphere from the direct combustion which is the most commonly used and suitable technology for a cellulose biofuel like wood chips.

Therefore, the characterization of physical and chemical properties of woodchips is essential to assess the environmental impact through woodchips burning emissions and to establish proper controls for natural biofuels in order to guarantee a certain security in bioenergetics production. Although RESs have a lower environmental impact [2], it is questionable whether to encourage RES adoption without a coherent analysis of environmental and energy advantages. So, an objective based evaluation by Life Cycle Assessment (LCA) [7] is necessary, because it considers environmental aspects and the potential impact of a product or a service system throughout its entire life cycle [8]. In the same way, NEA (Net Energy Analysis), which underpins Energy Return on Investment (EROI), permits to evaluate the amount of net energy obtained from a Primary Energy Source (PES) after the outputs (extraction, processing, delivering) to make it exploitable have been subtracted [9]. Moreover, society's economic growth and industrial processes depend on high EROI resources [10].

This research examines a co-generative plant situated in Northern Italy (CPN). The plant is supplied solely with mechanically treated wood chips obtained from two sources: (1) natural ligneous trees from energy crops, and (2) uncontaminated ligneous biomasses from agriculture and forestry wastes. Italian legislative Decree number 152/2006 lists allowed biofuels in the Italian territory. More Details of the co-generative plant are added as *Supplementary material 1.2*.

In this paper the tested approach was based on the following steps: (1) wood chips were characterized by determining the relevant physical and chemical properties for plant functioning; (2) HMs concentrations in wood chips were analyzed to assess a possible environmental pollution effect through emissions; and, (3) an approach to calculate EROI of CPN was suggested.

2 MATERIALS AND METHODS

2.1 BIOFUEL SAMPLES

Biofuel samples were of two categories. A first group included three kinds of natural wood chips from energy crops (A1, A2 and A3) and a second group was a mixture of wood chips obtained from agriculture and forestry recovery material (B1). The biofuel samples were collected directly from the stock area of CPN, producing a composite sample for every wood chip type. Samples were collected daily for five days and stored in laboratory conditions (15/22 [°C]), in sealed plastic bags to avoid moisture loss. Analyses were carried out using UNI, APAT (Agency for Environmental and Territory Protection) and EPA (Environmental Protection Agency) methods.

2.2 PHYSICAL PROPERTIES OF BIOFUELS

Particle size distribution was determined by sieving, as per UNI 15149-1:2011. Bulk density is based on the overall space occupied by an amount or a group of biomass particles. Calculations were made applying the UNI 15103:2010 Method. Bulk density was determined on dry weight basis with a digital balance (Kern 572-27) and a graduated container.

Calorific value was determined using a Mahler bomb calorimeter (UNI 14918:2010 Method). Benzoic acid was used for the calibration and pure oxygen 99.5 [%] to load the bomb.

Ashes, or the inorganic fraction, were determined by gravimetric analysis after desiccation at 550 °C, (UNI 14775:2010). A Muffle Gefran 1200 was used. Moisture content was determined on the same date of sampling by following the UNI 14774-2:2010 Method.

2.3 CHEMICAL ANALYSES OF BIOFUELS

Chemical components of the woodchips were also analyzed concurrently with the physical property determinations. Cl and S concentrations were determined by analyzing the moisture condensed on the wall of the Mahler bomb calorimeter by HPLC (UNI 15408:2006 and APAT 4020). The Portlab HPLC and Portlab Conductivity detector ID 510 were used. Concentrations of HMs (As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Sb, V, Zn, Se and Sn) were determined according to UNI 15297:2011 and 6020A US-EPA methods. After acid digestion of the sample with HNO_3 (67 [%]) and H_2O_2 (30 [%]), the solution obtained was analyzed by ICP-MS (7500 Series Agilent Technologies).

2.4 DETERMINATION OF CPN'S EROI

EROI (Energy-Return-On-Energy-Investment) is the ratio of how much energy is obtained from an energy production process to how much of that energy is needed to extract it ([11], [12]). It is defined by the following equation:

$$EROI = E_g/(E_c + E_{op} + E_d)$$

Where E_g stands for the constant gross flow of energy at rate over the whole lifetime, while E_c, E_{op} and E_d stands for total energy input to construct the system, the energy flow required to operate and maintain the system and the energy required for decommissioning, respectively.

All energy gains are included in the numerator, whereas all costs in the denominator. In theory, when the ratio is greater than one, energy output gives an effective yield or it could be regarded at least viable. Nonetheless, some researchers support the thesis that the social sustainability is achieved reaching the ratio of 3 at least ([13], [14]). Weißbach, who prefers economic threshold given by the society's GDP (Gross Domestic Product), establish this limit to 7 [15].

The EROI, in this article is substantially an energy balance calculation based on direct factors associated with wood chip use in CPN. Therefore, net electric energy produced and net thermal energy produced are two types of energy production and both work towards creating a positive EROI determination. Conversely, plant construction, equipment working, biofuel production, biofuel logistic charges and plant demolition, for instance, determine a negative EROI. We included the direct energy demand for processing the recovered biofuels, as indicated by Weißbach [15]. Moreover, an extended survey has been conduced to account the totality of "the upstream factors" [16] implicated in the direct energy and material inputs.

The information used in the research was based on data from the CPN control plan, reference values from the EPD (Environmental Product Declaration) of the materials used to build the energy plant and finally, interviews with staff directly involved in CPN operations. The reference method applied was drawn from attachment A of the Veneto Regional Decree n° 1713 dated 16 June 2006.

Based on the recent literature, the method adopted for this article can be assimilated to the Extended EROI ($EROI_{EXT}$) category [14], but only the amount of direct inputs relevant to make the energy usable to the final user was considered. It consists of a process analysis, as defined by Murphy [17] and the method could be classified as $EROI_{3.d}$, following his classification. In addition, where it was possible, the nomenclature has been harmonized with the one used by the same author and the energetic contributions have been explicated as indicated by Zhang [13].

The applied methodology doesn't comply with some important recommendations: (1) the energetic analysis doesn't consider indirect energy and material inputs, and (2) no energy quality adjustment has been done, although it was highly recommended by Murphy [17] and by Weißbach et al. [15] - the concept of exergy. These issues are matters for next investigations.

3 RESULTS AND DISCUSSION

3.1 CHARACTERISTICS OF BIOFUELS

The results are organized in the same order used to describe the analytical methods. Results from dimensional size distributions of samples (Fig. 1) are quite similar to those found in another article by Hartmann [18]. Jiris [18] states that a homogeneous distribution of dimensional classes favors the stability of the combustion process. In fact, particle dimensions, in addition to storage and desiccation processes, influence the regularity of the supply system in a combustion plant [19].

Bulk density was particularly high for B1 (Table 1), whereas significantly lower values were observed for A2 and A3. The percentage of ash (Fig. 2a) in the samples was higher, on average, than the values found in literature [20]. The peculiarity of B1 can be due to a larger amount of inorganic matter from soil attached to roots and to bark which is richer in inorganic fractions than wood [21]. It has been reported that ashes cause adverse effects toward plant equipment like fouling and corrosion. Considering that a portion of PM is generated by the inorganic fraction, a selection of cleaner biomasses would be advisable.

The calorific values of biofuels determined via the Mahler bomb calorimeter are shown in Fig. 2b. The values obtained are generally lower than those reported by other researchers ([6], [18]). LHV (Low heating value) is regarded as a more useful parameter in evaluating an energy yield. A1 shows the highest value, followed by B1, A2 and A3. Natural biofuels from energy crops, (material that is purchased) have a lower yield than B1 (not paid for).

Results from moisture analyses (not displayed) showed that the used on-field storage system of biomasses was unsatisfactory [4].

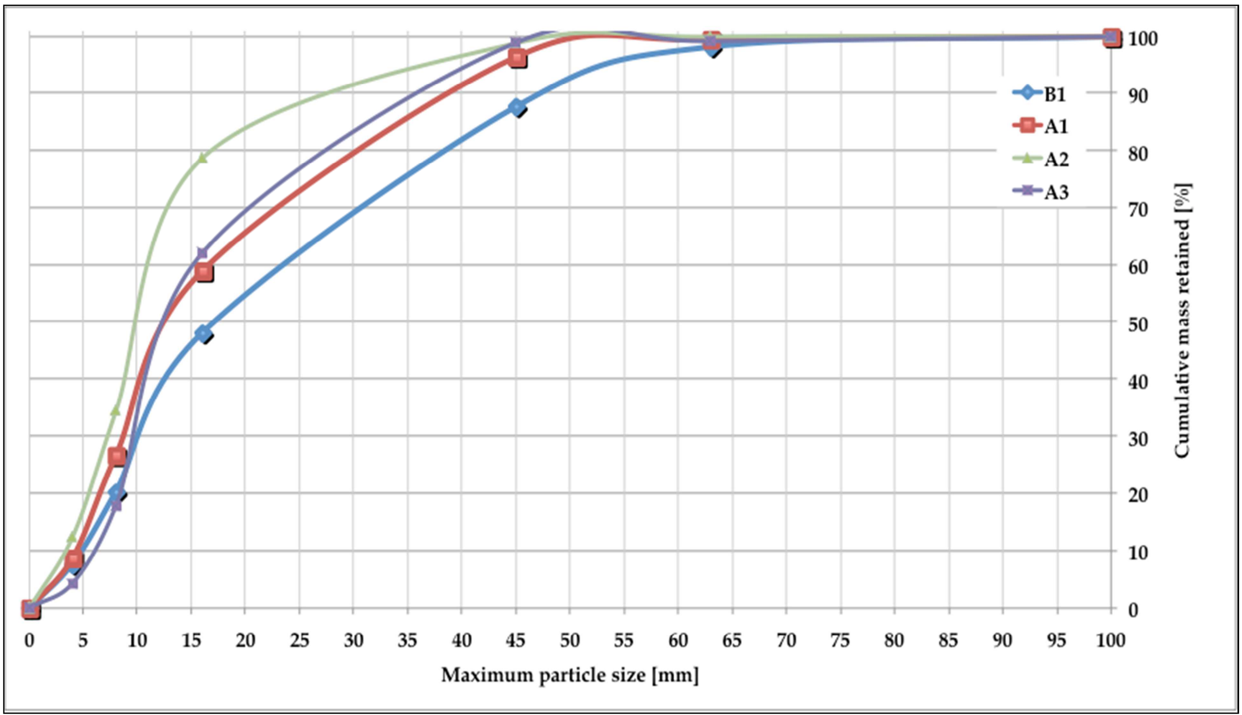

Fig. 1. Cumulative particles size distribution of test materials

Table 1. Bulk density of the tested materials

Dimensional class	Biofuel samples [Kg m^{-3}]							
	A1		A2		A3		B1	
	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.	Mean	St. Dev.
0-20	249.2	64	232.8	27.8	225.7	23.2	257.8	89.3
0-40	270.8	65.6	251.4	38.6	249.8	33.8	281.8	57.5
0-100	270.8	65.6	251.4	38.6	249.8	33.8	305.2	52.3

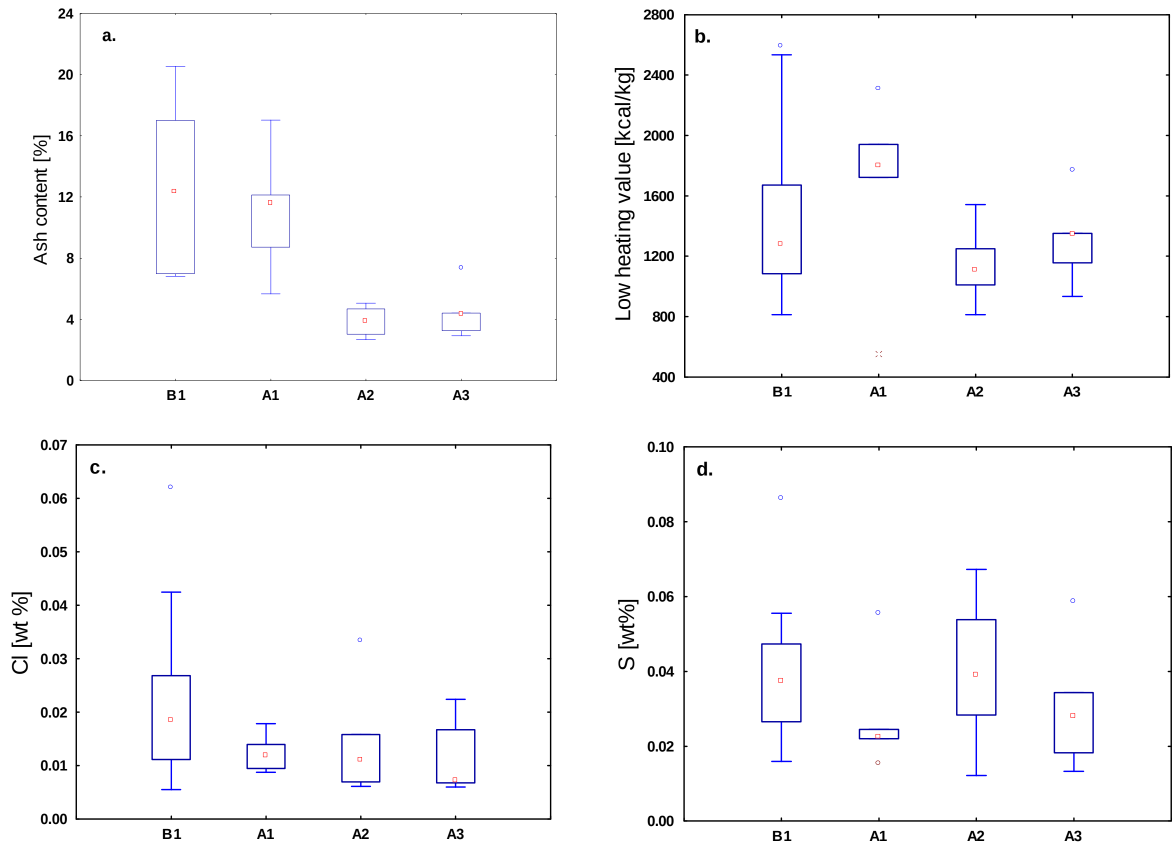

Fig. 2. Ash content (a), Low heating value (b), Chlorine (c) and Sulphur (d) concentration of the tested material

Moisture content is negatively correlated to the energy potential of the biofuel and values above 50 [%] make the process counterproductive [5]. Moreover, a high moisture content favors microbial proliferation and, consequently, natural biomass deterioration [22].

Chemical composition analysis focused on Cl, S and HMs concentrations, all calculated on dry matter basis. Cl and S were determined in mass percentage, whereas HMs are expressed in [mg/kg]. Cl (Fig. 2c) and S (Fig. 2d) concentrations are slightly higher than values reported by [23].

Higher values of Cl in B1 samples can be explained on the basis of their origin. In fact, a large fraction of B1 was collected from areas bordering the Venetian lagoon and high trafficked roads. Wind transported marine spray and sea salt used to prevent icing can reasonably explain a higher Cl content. Cl and S (Fig. 2a-b) do not exceed the limits suggested [6].

The presence of HMs in biofuels depends upon their source [24]. The composition of B1 clearly reveals its origin from high traffic zones (Fig. 3). HMs can be transported by the flow of gas produced during combustion and ultimately escape together with fly ash and vapors. HMs concentrations are similar to those previously reported [23]. Some technical features deserve consideration. During combustion HMs' behavior is different. Cd, Zn and Pb are mainly adsorbed onto fly ash [25], rather than onto bottom ash [6]. As and Cu are mainly in gas phase. Towards HMs, Cl operates similarly to alkaline metals [27] and, finally, HMs are accumulated mainly in the bark of plants [23].

High concentrations of HMs in dust emissions are conventionally related to biofuels containing wood from demolition activity and chemical treatments and not of natural origin [27]. A slight contamination could be derived from the mechanical treatment of wood (e.g.: cutting and crushing) during wood chip production. It is important to highlight that storage and transport act on biomass quality and, consequently, on financial costs. Moreover, environmental matters are directly related to the logistic phase, particularly, transportation.

Considering physical and chemical properties, it could be concluded that recovered waste (B1) could be a good alternative energy sources in developing countries as its physical and chemical properties are in compliance with stuffs of wood chips and economically profitable. Production of energy from recovering waste material in developing countries may be an interesting opportunity and a crucial factor in reducing poverty [28]. In developing countries (especially in rural, remote and coastal areas), approximately 80% of the total energy consumption comes from biomass fuel [29]. However, agriculture and forest recovery materials are more suitable source of renewable energy instead of wood chips since most of the developing countries have shortage of wood fuels. For instance, Bangladesh has lack of fuel wood since forest land is decreasing due to urbanization, industrialization and cultivation [30]. Therefore, it would be wise to generate power from recovery materials as Bangladesh is very vulnerable to climate change and cutting of forest in a large scale will disrupt the eco-system balance. However, suitable measures should be taken during collection and selection of recovery materials, selection of energy conversion methods, energy utilization efficiency and economy of the biological system to make the biomass resources more efficient and useful to meet the energy requirement of the developing countries.

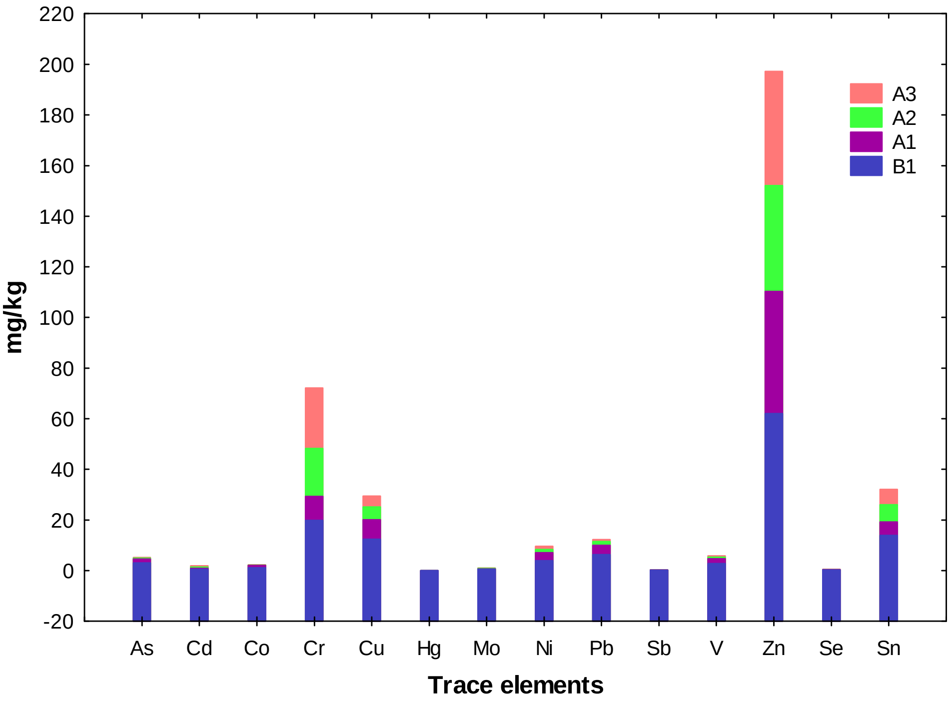

Fig.3. Trace elements concentrations of tested materials on a dry basis

3.2 EROI Result

An objective assessment of energy sustainability rises from a complete evaluation of the effects of RES exploitation including logistical issues, energy plant setting, environmental pollution, energy production and other secondary effects [17]. Energy charges and investment are both listed (Table 2). The lifespan of the plant was estimated to be 15 years.

During the life of CPN, working equipment and biofuel cultivation are weighed considerably on energy charges. The remaining energy charges are not so significant. Energy charges for the main building materials (a fraction of EC_{me}) used for CPN realization are extracted from the EPD of similar materials. The energy charge for the biomass production (EC_{bio}) is then calculated on the assumption that the biomass obtained from an energy crop is conserved for the whole period of the CPN lifespan. Transportation (a fraction of EC_{tmc} and the whole of $EC_{traspbio}$) is based on the effective equipment assigned to this service. Therefore, the approximations concerning maintenance and fuel provisioning suggested by Weißbach [15] could not be applied in this case.

The calculated EROI of CPN is 9.18 which correspond to a yield of nine times the investment. However, this EROI remains an optimistic value, because accidents and extraordinary maintenance have not been factored in, due to the fact that contingency prevision goes beyond the objectives of this research. A hypothetical scenario where the functioning of the CPN is totally based on recovered material will permit to add up to 4 additional points (a value of 13.24 instead of 9.18) to the EROI. This increase could be obtained by maintaining minimal distance between the site of supply and the CPN and the savings deriving from energy consumption of synthetic fertilizer production [10].

In accord with Murphy's classification an $EROI_{3,d}$ analysis has been conducted. Therefore, from a methodological perspective, the "state of the art" of EROI has not been reached in this paper, but a reasonable result has been achieved with the available amount of data.

The possibility of comparing the EROI determined in this paper with other results is quite difficult due to (1) the absence of an official standardized method and (2) the poor divulgation of other publications concerning the same specific issue. Although, the implementation of a standard method is "work in progress" since Murphy [14] has proposed a standard protocol. Method standardization could enable direct comparisons between bio-energy technologies and would avoid misleading results as suggested by several authors. Instead, it is advisable to increase the researches in biofuels exploitation, which likely could represent a deterrent during the transitional period from fossil fuels to more sustainable technologies like photovoltaic and wind turbines as presumed [14].

Table 2. Budget of EROEI

Entry			Result [MJ]
EC_{me}			6229230
EC_{tmc}			88
EC_{ma}			652032
EC_{imp}			2496690
EC_{att}			69258646
EC_{bio} =	EC_{ccu}	13526	34796701
	EC_{irr}	19543	
	$EC_{conctot}$	68083	
$EC_{traspbio}$			1651
EC_{mo}			163008
EC_{tm}			88
EI			113598134
ER			1043280000

Notes: energy charges for: building materials (EC_{me}); building materials transportation (EC_{tmc}); energy plant construction (EC_{ma}); plant equipment construction (EC_{imp}); plant working (EC_{att}); biofuels production (EC_{bio}); crop management per year per hectare (EC_{ccu}); crop irrigation per year per hectare (EC_{irr}); crop fertilization and defense ($EC_{conctot}$); biofuels transportation ($EC_{traspbio}$); energy plant demolition (EC_{mo}); demolition wastes transportation (EC_{tm}). Energy investment (EI). Energy gained (ER).

The final aim is to create a common platform to stimulate the discussion on energy technologies. Further, the method adopted requires detailed data collection in order to reach plausible results. On the other hand, the energy required for environmental reclamation, the energy to compensate pollutants emissions (i.e. the installation of the best available treatment technology) and other externalities are absent in the method adopted.

This last feature will increase the complexity to create a comprehensive EROI calculation [14], but it represents the milestone to reduce the EROI's gap between the RES and the fossil fuels. For instance, an additional consideration could be done with regards to the allocation of co-products obtained by the process. The fly ashes and bottom ashes produced during the combustion phase, represent an interesting binder for the technology known as stabilization/solidification (S/S) of solid wastes [a,b,c]. This could be a benefit: (1) in terms of energy saved from the production of additional clinker, reserved to waste management purposes; (2) it could be introduced in the energetic budget as highlighted by other authors ([13], [17]).

4 CONCLUSION

Biomasses are one of the most promising RES, even though there remains an evident yield efficiency difference with respect to fossil fuels. The biofuels selected for this study derived from energy crops and the recovery of natural materials from agriculture and forestry maintenance. Experimental activities showed that biofuels adopted in CPN have different properties and this is may be the reason explaining a lower performance of some features (LHV and bulk density) of the wood chips from energy crops than those from recovered materials (B1). LHV results demonstrate that economical and environmental benefits can be obtained by reusing residual biomasses, containing e.g. agricultural waste wood and braches from pruned plants. In fact, LHV performances are not far from the results obtained with virgin stocks. This valorization could intercept a large amount of vegetal material preventing its landfill disposal. From the perspective developed in this case-study, recovery material represents the most preferable way to obtain benefits in several environmental spheres like CO_2 reduction, sustainable waste management and preservation of soil quality. However, this observation demands for more frequent integrity checks of pollutants (especially ash content) in order to guarantee a low level of environmental risks.

EROI acts as an objective indicator to assess the energy sustainability of a plant in a specific scenario and the results of this study can be taken as an indication for a more comprehensive study. The EROI of CPN - an $EROI_{3,d}$ according to Murphy's classification - was found to be significantly positive (9.18), and this result can be a reference for other energy technologies in the same context. However the result should be refined by the use of extended analysis boundaries in order to include the direct and indirect energetic charges associated to CPN. If the inclusion of these further specifications could decrease the EROI performance of CPN, the allocation of the ashes for S/S purposes could partially compensate this reduction. A hypothetic scenario demonstrates that providing uniquely waste material an optimistic, but indicative, increment of EROI up to 13.24 could be reached. In addition, the exclusive exploitation of waste material may be a meaningful strategy where biofuels with low LHV are available. In conclusion, biofuels and in general RES deserve more attention and research as they are a promising energy source becoming more and more competitive or maybe essential to conserve the social well-being or to extend it to developing countries, where large amount of waste biological material may be available.

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SUPPLEMENTARY MATERIALS

1.1 .BIOMASSES PROPERTIES AND THEIR EFFECTS ON THE COMBUSTION PROCESS

1. Physical properties influence energy conversion yield and are determined by two factors: (1) the types of biomass (the respective calorific value, e.g.) and (2) the treatment undergone during the logistic and processing phase. Physical properties have been described by a number of authors [1, 2, 3] and the most important ones are bulk density, calorific value, particle size distribution, moisture and ash content.
2. Bulk density has considerable value for technical and economic reasons in all the processing phases. Coal bulk density is higher compared to biofuel, consequently the latter requires more space to produce the same amount of energy.
3. Calorific value represents the potential energy releasable by a determined kind of fuel and it is identified in two types: high heating value (HHV) and low heating value (LHV). HHV corresponds to the total energy released from the combustion and, therefore, the maximal energy recoverable from an energy source. LHV, on the other hand, is determined by subtracting the heat of water vaporization from the HHV produced by the combustion [3].
4. Moisture content, or the water content, of a biomass can inhibit combustion [4], repressing the full oxidation of the reagents. Mathematical models have been developed to estimate this factor [5].
5. Particle size distribution is defined as the dimensional distribution of the particles of the biofuel. Wood chips are conventionally classified in five classes: 4-7, 8-15, 16-44, 45-62, 63-100 [mm].
6. Ash content is the inorganic fraction of biomasses or, in other terms, the residual substance produced at the end of combustion. Ashes are of two kinds: (1) the intrinsic fraction that relies on ash composition; and (2) the assimilated fraction that depends on both the site of plant development [6] and on the processing operations employed. Both classes are mainly composed of the following elements: Si, Al, Fe, Ca, Mg, Na, S, K , and P (Table 1).
7. Ash content ranges from 1 to 30-40 [%] of mass weight. This subject has been widely explored for its important implications in technical and environmental issues. Ash is the main factor in the mechanism of corrosion that consists in interphase reactions deteriorating the mechanical devices of plant components. The main elements involved in this process are: S, K an Cl where chlorine acts as the vector for the other corrosive elements. Corrosion reactions have been described and modeled in specific bodies of research [7]. The inorganic fraction is also associated with the other negative processes of fouling and slagging. Fouling consists in the accumulation of ashes on the surfaces of mechanical devices, whereas slagging is defined as the formation of waste matter with stony features due to heat radiation on the walls of the combustion chamber [8]. Fouling and slagging are favored by
8. The presence of alkaline (e.g. K and Na) and earth alkaline (e.g. Mg) metals in biomasses. These could potentially reduce the life cycle of a combustion plant.

1.2. CO-GENERATIVE PLANT

The supply chain of CPN is coherent with a multi-biomass approach [9]. Besides, cogeneration enables a simultaneous production of thermal and electric energy starting from the same source, so that considerable fuel saving [10] is obtained. CPN produces 900 [kW] of electric energy, which is sold to the national manager of the energy grid (GSE) and 1910 [kWt] of thermal energy which is used for soil reclamation purposes. The plant project estimates 13.500 tonnes of biofuel to be supplied per year [t/y]. The purification system of CPN includes a multicyclon and an ESP system. At present, energy production from biomasses is more expensive than that from fossil fuels. Despite this, improvements can be obtained by biofuel characterization [8, 11, 12]. In the same way, this operation permits to the check the integrity of natural biofuels from HMs pollution which could be potentially dispersed during the combustion process.

Table 1. Typical mean values for the chemical composition of wood chips ashes (softwood, Wt [%] d.b.: dry weight basis per cent). Re-edited from: Obernberger et al., 2006[6]; Kalac et al., 2004[13].

Parameter	Unit	Values
Si	wt [%] d.b.	4 - 11
Ca	wt [%] d.b.	26 - 38
K	wt [%] d.b.	4.9 - 6.3
Mg	wt [%] d.b.	2.2 - 3.6
Na	wt [%] d.b.	0.3 - 0.5
SiO_2	wt [%] d.b.	49.3
Al_2O_2	wt [%] d.b.	9.4
Fe_2O_3	wt [%] d.b.	8.3
MgO	wt [%] d.b.	1.1
Cl	wt [%] d.b.	0.8
Na_2O	wt [%] d.b.	0.5
K_2O	wt [%] d.b.	9.6
CaO	wt [%] d.b.	17.2
SO_3	wt [%] d.b.	2.6
Cd	wt [%] d.b.	3 - 6.6
Zn	mg/kg d.b.	260 - 500