Multistage milling and classification for improving both pellet quality and biogas production from hazelnut and olive pruning

Paolo Costa[1], Pier Paolo Dell'Omo[2,*], Sabatino La Froscia[1]

1. *Labo, Université AR_adelec1Scienza per Amore Association – Via Monteleone Sabino, 9 - 00131 Rome, Italy*
2. *AR_adelec1Department of Astronautical, Electrical and Energy Engineering (DIAEE), University La Sapienza, Via Eudossiana 18, 00184, Rome, Italy*

paolo.dellomo@uniroma1.it

ABSTRACT. *The effects of a mechanical process were determined on the solid fuel quality and anaerobic biodegradability of hazelnut and olive pruning. Both the feedstocks did not meet the specification for industrial and residential pellets given in the European Standard EN ISO 17225-2, because of too high ash and nitrogen content. The coarser products from processing were notable for the high reduction in both the ash and nitrogen content. Therefore, as regards hazelnut, they met the requirement of the Standard for both the industrial and residential pellet, whereas those from olive processing met the requirements for the industrial pellet. The finest products showed a high concentration of nitrogenous matter and smaller C/N ratios, far better for anaerobic digestion. The best products from hazelnut and olive achieved methane yields of 118.1 and 176.5* $Nm^3\ t_{VS}^{-1}$*, respectively, corresponding to 70.1 % and 93.5 % gains over the untreated substrates. The process was highly energy efficient, since consumption was low compared with the energy output from the residues of fuel upgrading, intended for anaerobic digestion. The investigated process could be successfully used to improve the fuel quality of pruning and to generate products suitable for anaerobic digestion and the production of advanced biofuels.*

RÉSUMÉ. *Les effets d'un procédé mécanique ont été déterminés sur la qualité du combustible solide et la biodégradabilité anaérobie de la taille des noisettes et des olives. Les deux matières premières ne répondaient pas aux spécifications pour les granulés industriels et résidentiels données dans la norme européenne EN ISO 17225-2, en raison d'une teneur trop élevée en cendres et en azote. Les produits plus grossiers issus de la transformation se caractérisent par une forte réduction de la teneur en cendres et en azote. Par conséquent, en ce qui concerne les noisettes, elles répondaient aux exigences de la norme pour les granulés à la fois industriels et résidentiels, tandis que celles de la transformation des olives répondaient aux exigences applicables aux granulés industriels. Les produits les plus fins présentaient une concentration élevée en matière azotée et des rapports C/N plus faibles, bien meilleurs pour la digestion anaérobie. Les meilleurs produits à base de noisette et d'olive ont respectivement atteint des rendements en méthane de 118,1 et 176,5* $Nm^3\ tVS^{-1}$*, ce qui correspond à des gains* *de 70,1% et 93,5% par rapport aux substrats non traités. Le procédé était très économe en énergie, car sa consommation destinée à la digestion anaérobie était faible par rapport à celle produite par les résidus de valorisation du carburant. Le processus étudié pourrait être utilisé avec succès pour améliorer la qualité du carburant de la taille et pour générer des produits adaptés à la digestion anaérobie et à la production de biocarburants avancés.*

KEYWORDS: *anaerobic digestion, biogas, EN ISO 17225-2, pellet, pruning.*

MOTS-CLÉS: *digestion anaérobie, biogaz, EN ISO 17225-2, granulés, taille.*

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1. Introduction

Concerns about the use of fossil fuels damaging the environment has generated a growing interest over the production of energy using renewable sources. On 10 January 2007, the European Commission draw up a long-term vision for the EU to generate energy from renewable sources (European Commission, 2007); it proposed to establish a target by the year 2020 that 20% of the overall energy consumption in the EU shall be by renewable source. In addition, it proposed to establish, by the same year 2020, that a binding 10% of the overall energy consumption in the transport sector shall be by renewable sources.

In 2012, the total EU27 biomass supply for electricity, heating and cooling amounted to 103.3 Mtoe, accounting for about two-thirds of all renewable energy consumption in the EU, and it is projected to increase to 132 Mtoe by 2020. Therefore, wood for energy and materials will get scarcer and new resources will be necessary to meet the increasing demand through domestic supply (European Commission, 2014; Pedrazzi *et al*., 2018).

In Italy alone, the annual amount of residue derived from the pruning of olive and hazelnut groves, vineyards and other orchards has been estimated to 3.7 million tons of dry matter, and 1.7 million tons are reasonably available for energetic exploitation (Maisano *et al*., 2012).

Until present, pruning residues are either mulched or piled and burned on the fields, accordingly they are not considered an opportunity for additional revenue. Nonetheless, the recent development of dedicated harvesting equipment enhances the technical and economical sustainability of bioenergy production from olive and hazelnut pruning (Spinelli and Picchi, 2010; Fedrizzi *et al.*, 2012). Therefore, orchards by-product biomass may represent a real economic opportunity and a strategic source of fuel in Italy (Monarca *et al.*, 2013) and in other European Countries (Velazquez-Marti *et al*., 2011).

The simplest physical-chemical procedure for exploitation of wood is combustion. Compared to wood, pruning has a higher ash content and a higher concentration of critical inorganic elements, which affect the dust emissions and cause slagging problems during combustion. The dust emissions consist of both carbonaceous particles and vaporised inorganic matter, mainly alkaline metals, sulphur and chlorine (Garcia-Maraver *et al.*, 2014). Inorganic species in biomass fuels, such as silica, alkali oxides and salts, can also intensify agglomeration and intense ash deposition on the heat transfer surfaces of boilers (Vega-Nieva *et al.*, 2016). High concentrations of sulphur (S), chlorine (Cl) and nitrogen (N) increase the emissions of sulphur dioxide (SO_2), hydrogen chloride (HCl) and nitrogen oxides (NO_x), respectively (Carvalho *et al.*, 2013).

The particulate emission generated by pruning combustion can be controlled if medium–large scale boilers are used, which are typically equipped with multiple flue gas control systems. Nevertheless, these medium-large scale plants require a large procurement basin, thus transport distances are excessive for pruning residues, whose economic sustainability rapidly decreases for transport distances greater than 10 km, mostly because of the low bulk density of the resulting biomass (Picchi *et al.*, 2013).

As an alternative, pruning biomass can be processed into pellets, because a dryer, more dense and homogeneous fuel, used in purpose designed boilers, enhances the combustion performance, thus decreasing the polluting emissions.

Commercial pellets from fruit tree pruning have been previously investigated; they showed - as mentioned earlier - the disadvantages of higher ash content and NO_x and SO_2 emissions compared to pine pellets (Arranz *et al.*, 2015).

While wood pellets from forestry residues have successfully established new markets, those from agricultural residues such as pruning have not been developed, mainly because of the said, different characteristics of the feedstock. In fact, the European standard EN ISO 17225-2 (Solid biofuels) establishes quality specifications for wood pellets that those pruning biomass generally do not fulfil.

Accordingly, methods able to reduce the ash, nitrogen and sulphur content in the pruning biomass would be crucial to enhance their quality and commercial value.

Biochemical conversion of pruning into biogas is also attractive, because of its high thermodynamic yield in the conversion of the starting biomass (Samson *et al.*, 2008). The process of anaerobic digestion is also sufficiently consolidated, and the production cost is low even at the small-medium scale.

In October 2014, the Italian Ministry of Economic Development introduced the obligation of placing primarily on the Italian fuel market the so-called "advanced biofuels", that is biofuels, including biomethane, which are produced exclusively from specific feedstock, such as straw, dedicated lignocellulosic crops and residual woody biomass, including pruning. The mandatory share of advanced biofuels for the year 2018 is set at 1.7 % of the energy demand in the transport sector, to grow up to 2 % in 2022 (Italian Ministry of Economic Development).

Bio-methane, resulting from the biogas upgrading, is the biofuel with the best prospects for sustainability, because its use would contribute towards the reduction in greenhouse gas emissions to a much larger amount than the minimum required by European Directive (Bordelannea *et al.*, 2011) and it can be easily distributed through the existing pipeline network (Calabrò *et al.*, 2017).

Woody biomass, and particularly pruning, are currently of very limited use for methane production due to their low biodegradability, given the high content of structural carbohydrates and lignin. Thus, in order to improve the biodegradability, a pretreatment step is needed to weaken the lignocellulosic structure, remove the lignin and increase the surface for enzymatic attack. Several techniques are being studied to improve the anaerobic degradability of such substrates. Among them, pretreatment with organic solvents gave highly effective results (Kabir *et al.*, 2015; Shafiei *et al.*, 2014), but, in the opinion of the authors themselves, it would take enormous sizes - over 200,000 t/year of biomass - to ensure the viability of the installation. Hydrothermal and dilute acid pretreatments of olive and vineyards pruning gave poor results, also using commercial enzyme addiction in the digestion sludge; steam explosion proved to be a more effective pretreatment, especially for olive pruning (Nitsos *et al.*, 2015).

Milling steps following one another are also considered as a pretreatment procedure in the lignocellulose bio-refinery, but, currently, they are not considered cost-effective, because of the high energy demand (Barakat *et al.*, 2013)). Despite this, the great advantages represented by the process speed and the absence of any effluent give a boost to the search for effective and energy-efficient mechanical pretreatment processes or combination of mechanical size reduction with other pretreatments.

Dry milling and fractionation of corn has also been proposed to recover germ and fibers as valuable coproducts prior to fermentation for ethanol production (Murthy *et al.*, 2006). Disc milling and subsequent sieving were used to separate wheat straw into two fractions. Classification was particularly effective with respect to the separation of the non-lignocellulosic components, i.e. protein and extractives (Papatheofanous *et al.*, 1998).

Aim of this study was to assess the performance of an industrial scale device used to mill and classify pruning into four different products, to obtain better quality solid fuels and fractions with improved anaerobic degradability.

Through a detailed analysis of chemical and elemental composition, and a set of anaerobic digestion and calorimetric experiments, both the methane yield and the heat from combustion of the materials were determined. The quality of the solid fuels was also assessed, particularly concerning their ash and nitrogen content. Lastly, an evaluation of the energy balance of the process was carried out.

2. Materials and methods

2.1. Harvesting and processing

Hazelnut and olive pruning were harvested in winter 2016 in the Viterbo province, Central Italy, by a COMBI TR160 pruning harvester. The baled raw materials were then stored in a covered and ventilated area to be naturally dried until summer 2016. The dried raw materials were chopped by a knife mill to an average length of about 30 mm; a portion of these chips was used as a reference.

An industrial scale device, designed for milling and fractionating feedstock with a high dry matter content (>70%), was used to process the chips. It comprises a double stage mill, whose first stage acts predominantly by impact, maximizing the number of shocks in order to obtain a significant breakage due to fatigue, whereas the second exerts strong shear actions on the processed material (Manola, 2016). The milled material is then sieved through a plansichter into three fractions, namely C (Coarse), M (Medium), F (Fines). The fines are further classified according their dimensions, shape and density into two fractions (named F1 and F2) through a centrifugal classifier.

Thirty kilograms of chips of both materials were then processed in the previously described device. A screw conveyer fed the mill at a constant mass flow rate, which was 990 kg h^{-1} and 1,100 kg h^{-1} for hazelnut and olive pruning respectively.

The power drawn by the device was measured using a wattmeter (MTME-485, ABB-SACE, Italy); power, supply voltage, current and time were logged into a PC card at one-second intervals. The specific energy (MJ kg^{-1}) required for milling was determined by integrating the area under the power demand curve for the total time required to grind the 30 kg sample. The tests were performed in duplicates.

2.2. Chemical and elemental composition analysis

The raw materials, hazelnut pruning (HP) and olive pruning (OP), and the processed materials were analyzed for total and volatile solids (TS and VS respectively) according to the APHA standard methods (APHA, 1998). The ultimate elemental composition (C, N, H) of both the raw materials and the products was determined according to EN ISO 16948 (2015). The tests were performed in replicates of three.

2.3. Particle size analysis

The particle size distribution of the products was determined according to the ASABE standard S319.3 (2006). Throughout this test, mass percentages were measured as a function of their particle size by passing through sieves of specified mesh sizes. A sieve analyzer used twelve ISO sieves (3.000, 2.000, 1.400, 1.180, 1.000, 0.700, 0.600, 0.500, 0.425, 0.300, 0.212 and 0.150). Each time we operated the sieve for 15 minutes and the samples mass for the particle size analysis was about 250 g.

2.4. Heating value

The higher heating value (HHV, MJ kg_{TS}^{-1}) of both raw materials and products was evaluated according to the Standard EN 14918 (2009) and, subsequently, the lower heating value (LHV) was calculated according to the following equation given in the Standard

$$LHV = (HHV-2.45\cdot 0.09\cdot H)\cdot(1-U)-2.45\cdot U \quad [\mathrm{MJ\ kg^{-1}}] \tag{1}$$

in which 2.45 MJ kg^{-1} is the heat of evaporation of water at 20°C, H and U the hydrogen and moisture content (%) in biomass, respectively. These tests were performed in replicates of three.

2.5. Anaerobic digestion

Biogas production experiments were carried out on samples of the raw materials and the F1 and F2 fractions from their processing, to assess the biogas and methane yields. The experiments were performed in batch anaerobic reactors with a working volume of 2 litres and equipped with mixing and thermostating systems; the reactors were operated in mesophilic conditions (38°C). Anaerobic sludge from a mesophilic digester, containing 3.67 % total solids (TS) and 2.68 % volatile solids (VS), was used as inoculum to start the biological process. The inoculum/substrate ratio was in the range 0.504 – 0.507 on a VS basis, and the final substrate loading was in the range 69-70 g_{TS}/L. The experiments lasted 28 days and each of them was performed in triplicate, including two controls with inoculum sludge only; the gas produced by controls was subtracted from the actual gas produced through digestion of the media.

Biogas production was measured daily in averaged samples following standard methods. The composition of the biogas, with reference to methane (CH_4) content, was measured using a SG06IOMX6 portable automatic analyzer (B.A.G.G.I. srl, Milan, Italy).

3. Results

Processing of hazelnut pruning gave four products, named HC, HM, HF1 and HF2, whose masses amounted to 8.8 kg, 7.0 kg, 8.4 kg and 5.1 kg, respectively, equivalent to 29.5%, 23.3%, 27.9% and 17.1% of the processed raw material.

Processing of olive pruning gave four products, named OC, OM, OF1 and OF2, whose masses amounted to 8.6 kg, 5.2 kg, 9.5 kg and 5.7 kg, respectively, equivalent to 28.6%, 17.5%, 31.7% and 19.0% of the processed raw material.

The processing loss was 2.3 % and 3.2 %, for hazelnut and olive, respectively. It was mainly due to moisture reduction after milling.

The median particle size of the products from hazelnut processing was 1600, 700, 310 and 180 µm for HC, HM, HF1 and HF2, respectively. Products from olive processing showed a similar distribution of the median particle size, 1800, 710, 280 and 160 µm for OC, OM, OF1 and OF2, respectively.

3.1. Specific energy requirement

The average power drawn by the device, including the need for the feeding conveyer and the pneumatic transport of the processed material, was 72.0 kW for hazelnut, resulting in a specific energy requirement of 72.4 kWh t^{-1} (260.6 ±7.3 kJ kg^{-1}). Whereas, it was 70.1 kW for olive, resulting in a specific energy requirement of 63.0 kWh t^{-1} (226.8 ±6.9 kJ kg^{-1}).

Additional energy was needed for the preliminary chipping of the raw materials. In this case, the knife mill was manually fed and, therefore, the experimental determination of the specific energy consumption would have been unreliable. Therefore, reference is made to data widely available in the literature, which indicate a consumption of about 15 kWh t^{-1} of processed material (Monarca *et al.*, 2011).

3.2. Characterization of the biomasses

The investigated process significantly modified the nitrogen and ash content of the products with respect to the raw materials. In particular, the fractions HC and HM from hazelnut processing showed an ash content of 1.3% and 1.7% respectively, far below that of hazelnut pruning (3.7 %) (Table 1). The ash content of raw olive was 4.1 %, whereas it decreases to 2.1 % and 2.6 % for products OC and OM respectively.

Table 1. Chemical characteristics of raw and processed pruning

	U [%]	Ash [%$_{TS}$]	C [%$_{TS}$]	N [%$_{TS}$]	H [%$_{TS}$]	C/N
HP	10.09 *a*	3.65 *a*	42.8 *a*	0.640 *a*	5.61 *a*	66.9 *a*
HC	7.38 *b*	1.28 *b*	41.3 *b*	0.017 *b*	5.63 *a*	2367 *b*
HM	8.86 *c*	1.73 *b*	43.0 *a*	0.195 *c*	5.72 *a*	220.6 *c*
HF1	9.43 *c*	3.96 *c*	42.5 *a*	0.543 *d*	5.61 *a*	78.2 *d*
HF2	9.18 *c*	9.08 *d*	39.3 *d*	1.109 *e*	5.28 *b*	35.4 *e*
OP	9.20 *a*	4.06 *a*	42.9 *ad*	0.387 *a*	6.54 *a*	110.7 *a*
OC	7.95 *b*	2.09 *b*	41.5 *b*	0.007 *b*	6.52 *a*	592.2 *b*
OM	7.82 *b*	2.56 *b*	41.8 *bc*	0.101 *c*	6.53 *a*	415.5 *c*
OF1	7.14 *bc*	5.28 *c*	42.2 *bc*	0.416 *d*	6.55 *a*	101.5 *d*
OF2	6.83 *c*	6.15 *d*	42.6 *dc*	0.795 *e*	6.59 *a*	53.6 *e*

For each sequence, means followed by the same letter in the same column are not statistically different with a p-value<0.05 (Tukey test)

The highest ash concentrations were found in the F1 and F2 products from both materials: 3.96 % and 9.08 % in HF1 and HF2 respectively, the last being about 2.5 times the value observed in the raw material; whereas they were 5.28 % and 6.15% in OF1 and OF2 respectively, the last being about 1.5 times the value observed in OP.

The C and M fractions from both materials were also notable for the high reduction in the nitrogen content. The product HC from hazelnut processing showed a nitrogen content of 0.017 %, which was only 1/36 of that of the raw material (0.64 %); the product HM showed a nitrogen content of 0.2 %, a third smaller than it was in hazelnut pruning. Particularly concerning olive products, the nitrogen content of OC and OM was 0.07 % and 0.1 % respectively, versus 0.39 % for the raw material.

The nitrogen content significantly increased in both F2 fractions compared to the raw materials, +73.4 % for hazelnut (HF2) and +105.1 % for olive (OF2). For olive pruning the nitrogen in OF1 was 0.41 %, slightly higher than 0.39 % observed for the raw material, whereas in HF1 it was slightly lower than that observed in the raw material (0.54 % vs 0.64 %).

As regards the carbon content, no relevant differences were observed between the raw materials and their respective products, apart from HF2, whose carbon content was 8.2% lower than that measured for HP. As a result, the F2 products from both raw materials showed a C/N ratio far below those of the untreated pruning, which were 66.9 and 110.7 for hazelnut and olive respectively. HF2 and OF2 reached a C/N ratio of 35.4 and 53.6, respectively. A C/N ratio smaller than that of the raw material was also observed for OF1 (101.5 vs 110.7), whereas it was higher for HF1, 78.2 versus 66.9 for HP. The C/N ratio was extremely high for both HC and OC, 2367.0 and 592.2 respectively, due to the tremendous reduction in the nitrogen content of these products.

3.3. Biogas yield and quality

The untreated pruning showed a methane yield of 69.4 $Nm^3\ t_{VS}^{-1}$ (±5.1 $Nm^3\ t_{VS}^{-1}$) for hazelnut (HP) and 91.2 $Nm^3\ t_{VS}^{-1}$ (±5.7 $Nm^3\ t_{VS}^{-1}$) for olive (OP) (Table 2).

Table 2. Methane yield and biogas composition

	CH4 [Nm3 t-1]	CH4 [Nm3 tVS-1]	% CH4 [%]
HP	59.8 a	69.4 a	49.8 a
HF1	67.3 a	78.7 a	50.0 a
HF2	96.8 b	118.1 b	49.8 a
OP	79.4 a	91.2 a	49.3 a
OF1	101.3 b	115.6 b	49.7 a
OF2	153.8 c	176.5 c	50.4 b

Means followed by the same letter in the same column are not statistically different with a p-value<0.05 (Tukey test)

The HF2 product from hazelnut processing reached a yield of 118.1 $Nm^3\ t_{VS}^{-1}$ (±5.5 $Nm^3\ t_{VS}^{-1}$), corresponding to a +70.1 % gain over the untreated substrate, whereas OF2 from olive reached a yield of 176.5 Nm^3 t_{VS}^{-1} (±4.2 Nm^3 t_{VS}^{-1}), corresponding to a +93.5 % gain over the untreated feedstock.

OF1 showed a +26.7 % biomethane yield gain, compared with the raw material. No statistically significant yield gain was observed for HF1. Likewise, no statistically significant differences in the chemical composition of biogas where observed, apart from OF2, whose methane content in biogas was 50.4% versus 49.3% for the raw material.

As expected, the methane production was quite faster in the processed material than in the unprocessed feedstock (Figure 1 and 2).

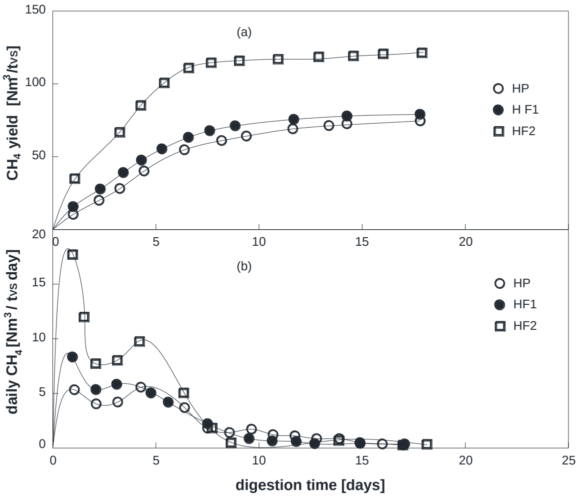

Figure 1. Hazelnut pruning: cumulative (a) and daily (b) methane yield

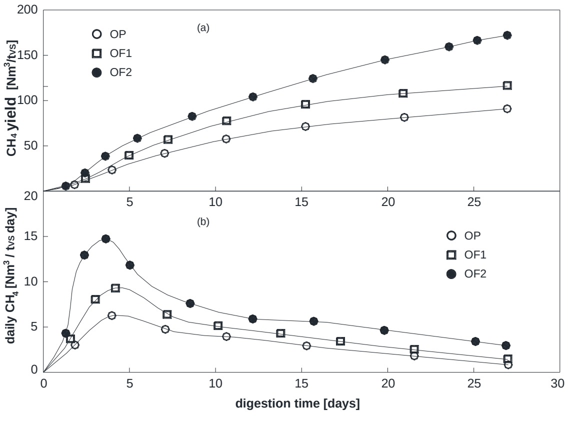

Figure 2. Olive pruning: cumulative (a) and daily (b) methane yield

3.4. Higher and lower heating values

There were no statistically significant differences between the lower heating values of the feedstock and those of their respective products (Table 3).

Table 3. Higher and lower heating values of the processed materials

	HHV [MJ kg_{TS}^{-1}]	LHV [MJ kg^{-1}]
HP	19.31 *bc*	16.00 *a*
HC	19.20 *b*	16.50 *a*
HM	20.08 *c*	16.94 *a*
HF1	19.85 *c*	16.63 *a*
HF2	18.22 *b*	15.27 *a*
OP	19.24 *a*	15.93 *a*
OC	19.21 *a*	16.16 *a*
OM	19.22 *a*	16.20 *a*
OF1	19.25 *a*	16.35 *a*
OF2	20.01 *a*	17.13 *a*

For each sequence, means followed by the same letter in the same column are not statistically different with a p-value<0.05 (Tukey test)

3.5. Energy data analysis

Electrical energy (E_e) produced by the products HF1, HF2, OF1 and OF2, expressed as kWh t^{-1} of raw material (HP and OP, respectively), was calculated using Eq (1):

$$E_e = Y \cdot LHV \cdot \eta_e \cdot (m_P / m_U) \tag{2}$$

where Y was the methane yield of the samples, expressed in Nm^3 t^{-1}, LHV was assumed to be 9.94 kWh Nm^{-3} (35.7 MJ Nm^{-3}), η_e was the co-generator electrical efficiency, estimated as 40 %, m_U and m_P were the masses of the processed sample before and after pretreatment, respectively.

The electric energy gain was obtained by subtracting from the energy produced by the pretreated material the sum of the energy obtained from the raw material and the energy consumption of the process. The results are shown in Table 4.

Table 4. Evaluation of energy efficiency

		ELECTRIC ENERGY		
	CH_4 [Nm^3 t^{-1}]	output [kWh t^{-1}]	consum. [kWh t^{-1}]	net gain [kWh t^{-1}]
HF1	67.3	64.7	87.4	53.0
HF2	212.0	65.8		
OF1	115.6	145.7	78.0	201.0
OF2	176.5	133.3		

Specific energies refer to the mass of the material before processing (HP and OP).

5. Discussion

The investigated process enhanced the pruning quality as solid fuel, allowing their convenient use in pellet supply chains.

Both the raw pruning did not meet the specification for industrial wood pellets given in the European Standard EN ISO 17225-2, because of too high ash content for OP and too high nitrogen and ash content for HP (Table 5).

Table 5. Compliance of raw pruning with the European standard EN ISO 17225-2

EN ISO 17225-2 industrial pellet					
	I1	I2	I3	HP	OP
U [%]	≤10	≤10	≤10	10.1	**9.2**
LHV [MJ/kg]	≥16.5	≥16.5	≥16.5	16.0	15.9
Ash [%$_{TS}$]	≤1	≤1.5	≤3	3.65	4.06
N [%$_{TS}$]	≤0.3	≤0.3	≤0.6	0.64	**0.39**
EN ISO 17225-2 residential pellet					
	A1	A2	B	HP	OP
U [%]	≤10	≤10	≤10	10.1	**9.2**
LHV [MJ/kg]	≥16.5	≥16.5	≥16.5	16.0	15.9
Ash [%$_{TS}$]	≤0.7	≤1.2	≤2	3.65	4.06
N [%$_{TS}$]	≤0.3	≤0.5	≤1	**0.64**	**0.39**

Values in bold numbers comply with the standard.

Likewise, both HP and OP did not meet the specification for residential wood pellets given in the EN ISO 17225-2, because their ash content exceeded the limit of 2% stated for the lower B class (Table 5). Nitrogen content met the requirement of class B for HP and class A2 for OP.

The LHVs of both the feedstock were slightly below the lower limit given in the standard, but this was due to the high amount of moisture in the raw materials, 10.1% and 9.25% for HP and OP respectively. In order to meet the minimum LHV required by the standard, a moisture content between 6% and 7% is needed for hazelnut pruning and no more than 6% for olive pruning.

The products C and M from both the feedstock showed an improved quality as solid fuel (Table 6), since:

a) they had a highly reduced ash content compared to the raw material. Ash reduction is foremost desirable, since it adversely affect the calorific value, the dust emissions and the combustion plant performance, because of the formation of unburnt, accompanied by corrosion, erosion and fouling;

b) they showed a highly reduced nitrogen content compared to the raw pruning. Nitrogen is an undesired element, since in the combustion process generates nitrogen oxides, responsible for irritant effects on humans and the formation of photochemical smog. In small-scale combustion system, in which the flue gas temperatures are above 1300 °C, nitrogen oxides are assumed to be formed mainly from fuel nitrogen during biomass combustion.

Table 6. Compliance of upgraded products with the European standard EN ISO 17225-2

EN ISO 17225-2 industrial pellet							
	A1	A2	B	HC	HM	OC	OM
U [%]		≤10		**7.4**	**8.9**	**7.9**	**7.8**
LHV [MJ/kg]		≥16.5		**16.5**	**16.9**	16.2	16.2
Ash [%$_{TS}$]	≤1	≤1.5	≤3	**1.28**	**1.73**	**2.09**	**2.56**
N [%$_{TS}$]	≤0.3	≤0.3	≤0.6	**0.02**	**0.19**	**0.07**	**0.01**
EN ISO 17225-2 residential pellet							
	A1	A2	B	HC	HM	OC	OM
U [%]	≤10	≤10	≤10	**7.4**	**8.9**	**7.9**	**7.8**
LHV [MJ/kg]	≥16.5	≥16.5	≥16.5	**16.5**	**16.9**	16.2	16.2
Ash [%$_{TS}$]	≤0.7	≤1.2	≤2	**1.28**	**1.73**	2.09	2.56
N [%$_{TS}$]	≤0.3	≤0.5	≤1	**0.02**	**0.19**	**0.07**	**0.01**

Values in bold numbers comply with the standard.

HC met the requirements of class B for residential pellets, and class I2 for industrial use. The limiting factor was the ash content, 1.28%, too high to achieve class A2 for residential use (upper limit 1.2%) and class I1 for industrial use (upper limit 1%). HM from hazelnut processing met the requirements of class B for residential pellets, and class I3 for industrial pellets. In both cases, the limiting factor was the ash content, 1.73%.

For both HC and HM, nitrogen content was far below the upper limit permitted for the top classes A1 and I1.

With the exception of the lower heating values, products OC and OM from olive processing met the requirements for the industrial class I3, but they did not meet the requirements for residential pellet, the limiting factor being the ash content, 2.09 % and 2.56 % respectively, too high for the upper limit permitted by the lower B class (2 %). In this case as well, the nitrogen content was far below the upper limit permitted for the top classes.

The fine fractions F2 from both feedstocks were suitable for biochemical conversion by anaerobic digestion. The high concentration of nitrogenous matter made their C/N ratios smaller than those of the respective raw materials, and far better for anaerobic digestion. As it is well known, a 20–30:1 ratio of C to N is needed for optimal anaerobic digestion, since microorganisms utilize carbon 25–30 times faster than nitrogen (Bardiya and Gaur, 1997). HF2 reached a ratio 35.4, very close to the optimal range, with a reduction of about 49 % compared with HP. Its methane yield reached 118.1 $Nm^3\ t_{VS}^{-1}$, corresponding to a +70.1 % gain over the untreated substrate. The C/N ratio of OF2 was 53.6, with a reduction of about 59.6 % compared with the raw material; as a consequence, the methane production was very high, reaching 176.5 $Nm^3\ t_{VS}^{-1}$, corresponding to a yield gain of about +93.5 % over the untreated raw material.

Biomethane yield of OF1 increased compared with the raw material (+26.7 %), corresponding to a slightly reduction of the C/N ratio, which was 101.5 vs 110.7 of the feedstock. For HF1 the methane production was 78.7 $Nm^3\ t_{VS}^{-1}$ and there was no statistically significant difference compared to the raw material, but the production was quite faster. The C/N ratio of HF1 was slightly higher than that of the raw material (78.2 vs 66.9).

The energy consumption of the investigated process ranged from 78.0 kWh t^{-1} to 87.4 kWh t^{-1} for olive and hazelnut pruning respectively.

In order to assess its energy efficiency, we compared the energy consumption for upgrading the solid fuel with the energy output from residues of upgrading, namely the products F1 and F2, after anaerobic digestion and electric energy production through a standard CHP engine. The electric energy output from the methane was 140.5 kWh for processed ton of hazelnut pruning and 279.0 kWh t^{-1} for olive, which made the energy balance of the process largely positive.

6. Conclusions

Orchard pruning may represent an important source of energy, but they are well known for their poor quality as a fuel, especially in small scale boilers. The hazelnut and olive pruning investigated in the present study did not meet the specification for industrial and residential wood pellets given in the European Standard EN ISO 17225-2 (Graded wood pellets), because of too high ash and nitrogen content.

A mechanical, industrial scale process was investigated for upgrading the fuel quality of pruning through multiple dry milling stages and subsequent classification into four different products. Effective and energy-efficient mechanical processes are of great interest in the field of pretreatment of biomass, because of their speed and the absence of any effluent.

The investigated process greatly reduced both the ash and nitrogen content in the coarser products from processing, named C and M, which accounted for 52.8% and 46.1% of the processed material for hazelnut and olive, respectively.

As regards hazelnut, these products met the requirement of the EN ISO 17225-2 standard for both the industrial and residential pellet. Particularly, both HC and HM met the requirements for residential pellets, class B, whereas they met the requirements for the industrial classes I2 and I3, respectively.

OC and OM from olive processing met the requirements only for the industrial pellet. Their ash content was too high for residential uses, in spite of it was reduced by 48.7% and 36.5% with respect to the raw material, respectively.

The residues from solid fuel upgrading, namely the fine fractions F1 and F2 from both feedstocks, were suitable for the biochemical conversion by anaerobic digestion, due to their micrometric dimensions and to the high concentration of nitrogenous matter, which made their C/N ratios smaller than those of the respective raw materials and far better for anaerobic digestion. Particularly, the methane yield of the finest products from processing were 118.1 Nm^3 t_{VS}^{-1} for hazelnut, corresponding to a +70.1 % gain over the untreated substrate, and 176.5 Nm^3 t_{VS}^{-1} for olive, corresponding to a yield gain of about +93.5 % over the untreated raw material. HF1 showed a methane production similar to the raw material, whereas production from OF1 enhanced by 26.7 % compared with the raw material.

Since the process was undoubtedly effective, its energy efficiency was also assessed, and it resulted largely positive considering only the electric energy output from the residues of solid fuel upgrading, intended for methane production through anaerobic digestion. In fact, the electric energy consumption of the process ranged from 78.0 kWh t-1 to 87.4 kWh t^{-1} of processed raw material, whereas the electric energy output from the residues was 140.5 kWh for processed ton of hazelnut pruning and 279.0 kWh t^{-1} for olive.

These results suggest that the investigated process could be successfully used to improve the quality of pruning and establish new markets for them. Moreover, products suitable for anaerobic digestion and the production of advanced biomethane can be generated.

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Nomenclature

C	carbon content
CH_4	methane
CHP	combined heat and power
d_{50}	median particle size, μm
EU	European Union
kWh	kilowatt hour
H	hydrogen content
HHV	higher heating value, MJ kg^{-1}
LHV	lower heating value, MJ kg^{-1}
N	nitrogen content
Nm^3	cubic meters at normal conditions
TS	total solids
U	water content
VS	volatile solids