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Ciclo: XXVII

Study on the optimization of the pellet production  
and on the development of a forest-wood-pellet chain

*Studio per l'ottimizzazione della produzione di pellet  
e lo sviluppo di una filiera foresta-legno-pellet dedicata*

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## 1 Summary

The biomass and biofuels markets have increased substantially in the last two decades, mainly driven by the national and European policy targets on green energy. The same positive trend is registered in particular for wood pellet markets.

In comparison with other wooden biofuels (firewood or wood-chips) pellets have a number of positive aspects such as:

- regular and optimized shape;
- wide range of energetic applications (to produce thermal energy, syngas, electricity);
- high solid density resulting in the concentration of a huge amount of energy in small volume;
- high calorific value on volume bases (i.e.: energy density);
- high energetic conversion factors due to the efficiency and regularity of their burning;
- possibility to enhance the qualitative parameters of some biomasses or bio-wastes in a recycling perspective.

The supply chains of wood pellets were traditionally based on residues and by-products from the lumber production industry which could be purchased at low cost locally. Even if these raw materials represent the best option, the potential of residual biomasses has now already reached its availability limit and new forms of supply are needed.

The research program aims to verify the technical and economical feasibility to develop and improve alternative raw materials supply chains for wood pellet production verifying their qualitative output and environmental performances compared with traditional ones.

The research will consider the forestry sector as main source of raw material, as a large amount of low quality timber is annually harvested and even bigger amounts of allowable annual cut (AAC) are not actually harvested both for technical or economic issues. The possibility to build a forest-wood-pellet chain is going to be studied and tested.

The project is mainly focus on four pillars:

- 1) The study of the actual quality of commercial pellets in the market. Through a consistent sampling activity and the statistical elaboration of the acquired data via SIMCA modeling it is possible to build a model able to classify each sample and

predict its certifiability. The model defines a critical metric distance below which a sample can be considered of certifiable quality.

- 2) The study of the achievable quality that can be technically obtained from forestry assortments. An experimental design is established for the sampling of different raw material retrievable from forest operation in the alpine region. The analysis of wood pellets, produced by the mean of a pilot pelletizing plant, give answers about the quality of the final product.
- 3) The study of the color of pellets and its relationship with quality parameters. The color of each sample from point 1) is measured through a spectrometer and image scanner and the results elaborated and statistically analyzed to find correlation between color and quality parameter of pellets.
- 4) The impact assessment of different supply and production pellet chains with the LCA methodology. Five supply and production scenarios are studied and discussed from their environmental performance point of view. Each of them is separated into elementary processes involved in the whole chain and their energetic input and emission output studied and analyzed to detect the best option possible compared with reference one.

The results of this study show that there are still chances of improvement in the production potential of wood pellets by an optimal organization of the supply, not only of traditional raw materials, but also of innovative ones. Their exploitation, moreover, can find a proper position in the market of commercial pellets since the quality performance of such products is comparable with traditional ones.

## 2 Riassunto

I mercati delle biomasse e biocombustibili sono aumentati notevolmente negli ultimi decenni, principalmente sotto la spinta degli obiettivi di politica nazionale ed europea in materia di energia rinnovabile. Lo stesso trend positivo si è registrato in particolar modo per i mercati del pellet di legno.

In confronto con altri biocombustibili legnosi (legna o cippato) il pellet ha un certo numero di aspetti positivi, tra i quali:

- forma regolare e ottimizzata;
- vasta gamma di applicazioni energetiche (per produrre energia termica, syngas, energia elettrica);
- alta densità risultante della concentrazione di una grande quantità di energia in un ridotto volume;
- elevato potere calorifico su base volumetrica (ad esempio: densità energetica);
- elevati fattori di conversione energetici grazie all'efficienza e la uniformità di combustione;
- possibilità di migliorare i parametri qualitativi di alcune biomasse e bio-rifiuti in una prospettiva di riciclaggio.

Le catene di approvvigionamento di pellet di legno sono tradizionalmente basate sui residui e sottoprodotti dell'industria di produzione di legname che potevano essere acquistati a basso costo a livello locale. Anche se queste materie prime rappresentano l'opzione tecnicamente migliore, il potenziale di biomasse residuali ha ormai già raggiunto il limite di disponibilità massimo e sono necessarie nuove forme di approvvigionamento.

Il programma di ricerca si propone di verificare la fattibilità tecnica ed economica dello sviluppo e miglioramento delle catene di approvvigionamento di materie prime alternative per la produzione di pellet di legno e verificare la loro qualità e prestazioni ambientali in confronto con quelli tradizionali.

La ricerca prenderà in considerazione il settore forestale come fonte principale di materia prima, dal momento che una grande quantità di legname di bassa qualità viene annualmente raccolto e una ancora più grande quantità di provvigione tagliabile e disponibile non è effettivamente utilizzata, sia per questioni tecniche che economiche. La possibilità di implementare una catena foresta-legno-pellet è qui studiata e testata.

Il progetto si basa principalmente su quattro pilastri:

- 1) Lo studio della qualità effettiva di pellet commerciali nel mercato. Attraverso una consistente attività di campionamento e di elaborazione statistica dei dati acquisiti tramite modellazione SIMCA è possibile costruire un modello in grado di classificare ciascun campione e prevederne la sua possibile certificabilità. Il modello definisce una distanza metrica critica sotto la quale un campione può essere considerato di qualità certificabile.
- 2) Lo studio della qualità tecnicamente ottenibile dalla produzione di pellet da assortimenti forestali. Si è definito uno schema di campionamento delle materie prime recuperabili da operazioni di utilizzazione forestale nella regione alpina. L'analisi dei pellet prodotti con un impianto di pelletizzazione pilota industriale dà risposte sulla qualità del prodotto finale.
- 3) Lo studio del colore del pellet e il suo rapporto con i parametri di qualità. Il colore di ciascun campione del punto 1) viene misurato attraverso uno spettrometro e uno scanner di immagine e i risultati elaborati ed analizzati statisticamente per trovare eventuali correlazioni tra il colore e i parametri qualitativi del pellet.
- 4) La valutazione d'impatto delle diverse filiere di approvvigionamento e produzione di pellet con la metodologia LCA. Cinque scenari di diverse catene di approvvigionamento e produzione sono studiati e discussi dal punto di vista delle loro prestazioni ambientali. Ciascuno di essi è stato scomposto in processi elementari coinvolti nell'intera catena e i loro input energetici e output di emissioni sono studiati e analizzati per rilevare la migliore opzione possibile rispetto allo scenario di riferimento.

I risultati di questo studio mostrano che c'è ancora possibilità di miglioramento del potenziale di produzione di pellet da un'organizzazione ottimale della fornitura non solo di materie prime tradizionali, ma anche di altre innovative. Il loro sfruttamento può inoltre trovare un posizionamento sul mercato di pellet commerciali poiché le caratteristiche di qualità di tali prodotti sembrano essere paragonabili a quelli commerciali.

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## 3 Introduction

### 3.1 Pellet production process

Wood pellets, are obtained from a multi-steps process that transforms biomass feedstock into final product. The cores of this process are the fine grinding and the compaction that create a product with high physical proprieties able to be used in a huge variety of heating devices with high conversion efficiency. The standard shape of pellet is cylindrical, with a diameter of 6 to 8 mm and a length of no more than 45 mm. Larger pellets could also be produced, and this is of particular interest in Central and Northern Europe. Compacted biomasses in the shape of pellets and larger than 25 millimeters in diameter are usually called briquettes. Some researches are now studying the benefits related to the production of pellets with different shape, especially spherical.

The production process (Figure 3-1 - Figure 3-2) comprises the following steps (Magelli et al., 2009):

#### Raw material purchasing and selection

The final quality of wood pellets is strongly influenced by the raw material, pure or blends, involved in the production. A fine selection of feedstock and proper handling and storing contribute to maintain high quality standards.

#### Raw material milling

Pellet presses generally require to be fed with sawdust of 5-8 mm in nominal top size particles distribution. If the feedstock is in form of logs (large and solid), the material is first run through a chipper that reduces the material into chips by cutting the wood with sharp knives. A second step involves the final reduction of chips into sawdust through a hammer mill with blunt knives to reduce the particles to the required size.

#### Moisture control

Controlling and maintaining an appropriate moisture level of the raw material is crucial to obtain high quality levels of the final pellets. When pelletizing wood, the required moisture level of the feedstock is about 8 - 13%. Moister materials are not able to remain compact after pressing due the spring-back effect caused by the evaporation of water during pelletization. Drier materials result in less durable pellets and in increased power

consumption for extrusion. Raw materials are dried in industrial ovens or by blowing hot gases (flue gases – steam – hot air) over or through the particles depending on the type of dryer. To regulate the moisture level when the feedstock is too dry (for example sawdust from the wood processing industry), the moisture content can be increased by injecting water or steam into the feedstock in the conditioner. Steam is usually preferred because, even if it involves more energy for its production, it also increases the temperature of sawdust to better prepare wood fiber to be pelletized.

### Additives

In some cases it is preferable to add some natural binders to the feedstock materials before pelletization to i) increase the mechanical properties of final pellets since additives act as inter-particles binder, ii) reduce the power consumption of the pellet press due to the lubricating effect produced by the additive at high temperatures, iii) increase the life of the die reducing the die and rollers wear. The most used additive is corn starch, that is added in percentages ranging from 0.1 to 2%.

### Extrusion

Two rollers compress the fine biomass against a metal die that can be flat or circular. The die is equipped with several holes of the required diameter drilled through it with a conical shape, which allow the biomass to be pressed under high temperature and pressure conditions. In this environment the lignin melts and acts as natural binder for the particles constituting the final pellet. The compression factor of the die is represented by the ratio between the diameter of the hole and the depth of the active channel. Specific compression ratios have to be chosen for different types of woods.

A fix blade cut the pellet to a predefined length as it exits the die.

### Cooling

As pellets exit the die they are hot (90-150°C), soft and releasing a high amount of steam. They must be cooled and dried before being packed to avoid the condensed water to affect final quality. This is usually achieved by blowing air through the mass of bulk pellets as they stay in a so-called counter-flow cooler. The final moisture content of the pellets should be no higher than 10%.

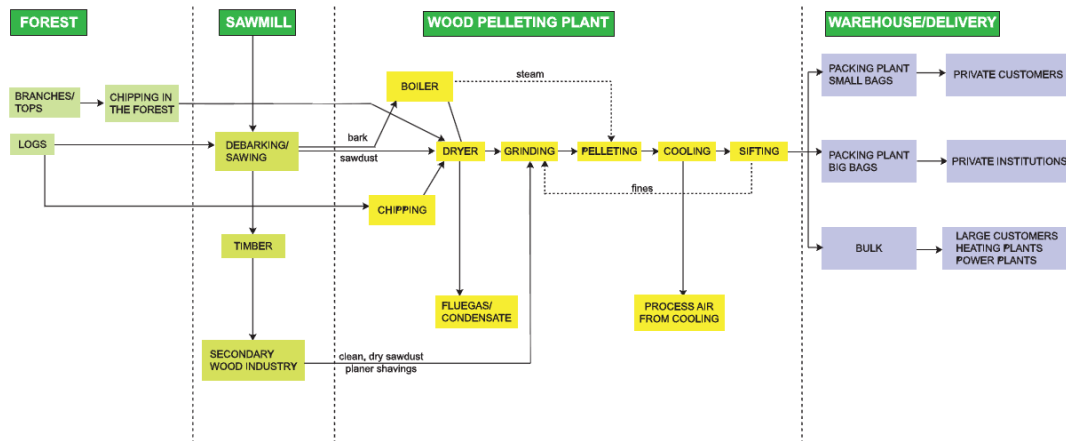


Figure 3-1: the whole pellet supply chain (Kofman, 2010)

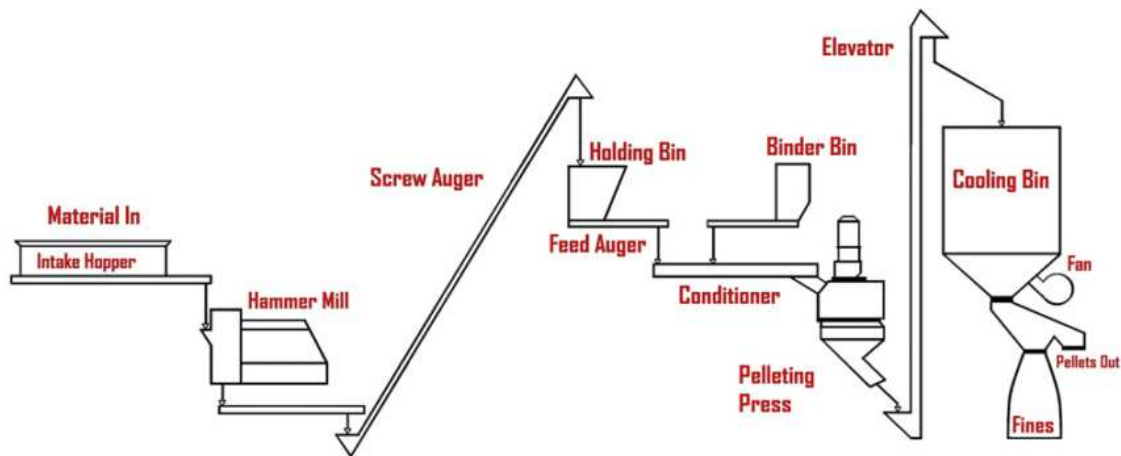


Figure 3-2: the pellet production process (Carroll and Finnan, 2012)

### 3.2 Current status of the pellet market

The increased global demand of energy is forcing the renewable sector to find new strategies and supply solutions. The demand for renewable energy sources has continued to rise, and data available for 2011 and 2012, published in the EU reports, give an estimated supply of final renewable energy of about 19% share among others (McGinn et al., 2013). The development of alternative energy and fuel supply chains has been chosen as a key action for the European Union to produce cleaner and less pollutant economy. Biomass feedstock is though one of the most important actors of this action since its share is growing among all other renewable energy sources.

This concern has affected the biomass trade with a specific reference to wood based products as substitutes of fossil fuels. The production and use of traditional or

technologically improved woody biofuels can contribute to the overall greenhouse gases (GHG) reduction (Katers et al., 2012) due to their close-to-neutral carbon emission (Sjølie and Solberg, 2011) up to the point that many European Countries have strongly encouraged and subsidized the substitution of oil, coal and gas residential and industrial heating systems and power plants with others run on biomaterials.

Wood pellets are one of the wider internationally spread biofuel commodities for energy production and their market is booming in Europe (Sikkema et al., 2011). This is due mainly by the fairly low cost per energy unit, the European and National subsidy systems to achieve the 2020 targets, the high prices of other energy sources and the level of technology required to exploit them. The pellet market is expected to grow more than 10 times by the year 2020 (Sikkema et al., 2011). Nowadays the continuous increasing demand in wood pellets is not to be totally met by the domestic production, not for technology limitation, but for shortage in primary resources and due to the competition with large power plants (Monteiro et al., 2012a) and wood industry. Even if specific and accurate statistics for pellets sector are missing, the European market is dependent on importation (Uasuf and Becker, 2011) and the ENplus annual report underlines that the Italian import of wood pellets from abroad (within EU-27, Canada, United States, South America and New Zealand) is actually filling the gap between the domestic consumption and production (Audigane et al., 2012).

In the period 2009-2012 the production of pellets in EU has increased by 30%, reaching 9.2 million tons in 2010, and 10.6 million tons in 2012 (AEBIOM, 2013). More than half of this production is concentrated in few core productive areas mainly located in the North and Central Europe (Mola-Yudego et al., 2013).

EU wood pellets consumption increased to 11 million tons in 2010 (AEBIOM, 2012). This gap has caused a lack of pellets in the European market, that has resulted in increasing pellets prices and the dependence of Europe on importations (Fantozzi et al., 2005) from USA, Canada, South America, New Zealand and Russia (Uasuf and Becker, 2011).

### **3.3 Pellet quality**

Physical and mechanical properties of pellets play a key role in their wide spreading, since more energy can be compacted and stored in the same amount of volume and, when compared with other traditional wood fuels, it keeps transportation and storage costs fairly

low (Monteiro et al., 2012b; Oveisi et al., 2013). The regular geometrics and steady energetic characteristics (Oveisi et al., 2013) allow to handle, store and use pellets in automatic appliances (García-Maraver et al., 2011; Obernberger and Thek, 2010) both equipped with mechanical or pneumatic conveyors.

Wood pellets quality is then affected by a number of different parameters, that are linked with the: i) feedstock properties following the general rules *what goes in, comes out*, ii) feedstock pre-treatment such as comminution before storage, seasoning, condition and drying technology, iii) production settings, iv) post-treatment. Raw materials properties affect pellets quality, as their constituents are found more or less unchanged in the final product especially regarding the minerals content. By-products from the wood processing industry are traditionally the most used raw materials, but their exploitation has already reached its maximum limit (Selkimäki et al., 2010). Alternative raw materials are then needed and their utilization has to be tested to allow economic and quality productions (Dibdiakova et al., 2014). Storage conditions, that can modify the chemical constitution of wood, affect the mechanical compaction behavior of the sawdust. In particular the natural seasoning and low temperature drying results in more durable pellets compared with ones made from artificially and fast dried feedstock at high temperatures (Filbakk et al., 2011).

Tree species and wood provenance has been found to influence the pelletizing process (Holm et al., 2007), and the final pellet quality (Gil et al., 2010; Nielsen et al., 2009), especially if related with the sawdust particle size distribution and its content in fatty acids (Arshadi et al., 2008; Samuelsson et al., 2009). The pelletizing pressure, the hold time of pellets in the die channel and the extruding temperature are directly related with feedstock characteristics (Holm et al., 2007).

The total amount of ash in the biofuel is also related with tree species introduced in the blend of feedstock as different woods are characterized by different amount of unburnt residues (Lestander et al., 2012b).

The final quality of pellets are related to the optimal moisture content of feedstock and its precise control and adjustment (Larsson et al., 2013). The moisture contained in the raw material as bound water is a key player in the formation of inter-particles linkages as its values are maintained between 8% and 13% (Filbakk et al., 2011; Nielsen et al., 2009). Delivery and handling chains have even the same importance to maintain the initial quality

level and allow the proper and wide use of pellets for energy production able to compete with other bioenergy sources (Selkimäki et al., 2010).

The fast European growth and evolution of the pellet sector has encouraged the development of specific standards and regulations in the attempt to set some baselines for production, storage and distribution of wood pellets. The standardization process has resulted, by the end of 2012, to the publication of some harmonized European Norms (EN) regulating pellets quality requirements. The EN 14961, in its part 2, sets the parameters limits to fulfill different quality classes (A1, A2 and B), while EN 15234-2 refers specifically to the pellet fuel quality assurance. The general objective of the quality assurance is to guarantee the overall pellet quality level through the entire supply chain giving evidence of the maintenance of a control system with reliable records at every critical control point (CCP) of the chain. The European standard, officially adopted by the Italian Standardization Body (UNI), perfectly applies to the national market which is mainly based on residential appliances, as stoves and small boilers (Olsson et al., 2011) which are pretty sensitive to the variability in quality of the used fuel.

### 3.4 Aims of the research

One of the main objectives of the research program is the study of the whole pellet production chain considering its technical feasibility, economic sustainability and environmental impacts.

Almost every step of the production within the pelletizing plants have been studied and analyzed to optimize the quality of the final product. Lots of information are available, for example, about the optimal wood blends and mixture (Lehtikangas, 2001) or about calorific values of different wood species (Gil et al., 2010; Telmo and Lousada, 2011). On the other hand, less information are available about the different possibilities offered to optimize the supply stage, or either, the utilization and energetic valorization stages. Most of the raw materials used for pellet production come from wood sawdust and other secondary products that obviously have lower price in comparison to round wood or other raw materials. This particular sector of the chain will be improved and studied to enhance its efficiency. But the rising demand of biofuels, especially wood based ones, could not be fulfilled only by reusing waste and by-product materials. Besides, there is a strong dependence of Italy and other European countries from foreign markets for the supply of raw material and pellet as well (Hillring, 2006; Pa, 2009). These are the reasons why the project aims to study and focus on the possibility to set up a pellet production chain directly from forest raw materials. Moreover the study aims to define the quality of the pellet produced from this chain. A valid example is represented by branches and treetops, actually left on the felling area or harvested to produce low quality wood-chips (Spinelli et al., 2011). To do this the study was divided into four main blocks, each one focusing on a specific aspect of the pellet quality evaluation:

- the study of the actual quality of commercial pellets
- the study pellet quality from forest residues
- the LCA study of these supply chains
- the study of the relationships between color and quality in wood pellets.



## 4 Study of actual quality of commercial wood pellets

### 4.1 Introduction

The first step of the project aims to define the so-called *zero line* which is defined by the average values of some parameters found on the actual wood pellets market. This gives figures about the actual quality and the gap that is still present between the measured quality parameters values and the declared ones. These minimum requirements will be then compared with the ones from pellet produced out of forestry logging residues. This comparison gives an idea about the overall reachable quality of pellets produced with by-products as specified in the experimental design. A huge amount of pellets from different commercial brands and types of final products are actually imported, produced and sold in our Country and not always the declared quality is found to be the real one, able to meet the basic requirements as requested from boilers and stoves producers. In particular the big difference between the domestic demand and offer of pellets makes the risk of frauds even higher.

To overpass this problem and to have a clear snapshot of the actual situation an experimental study has been set up based on the collection of a number of commercial pellets samples and a multivariate class modeling approach which could extract an index from the measured quality parameters of a wood pellets population randomly purchased from the commercial products available on the market. The index calculated can be set to be a baseline for the real quality of the analyzed pellet samples on the market and acts as a threshold in the evaluation of newly analyzed samples. The soft independent modeling of class analogy (SIMCA) is built on the actual properties of certified pellet samples (reference population), and it is able to predict the position of a test sample (SIMCA distance) on a scale, defining its potential certificability referred to the critical distance previously calculated via SIMCA. The model was built on a limited number of pellet quality parameters. The quality analysis has been related to the declared parameters stated in the package or in the product declaration (EN 15234-2) by the producer or trader. The presence of any kind of certification seal has been checked and the compliance of measured parameters with reference ones has been verified. Moreover we tested five reference raw materials that were chosen considering the

economic potential of small trees from thinning operations and residues from final felling. The market demand for energy biomass can create a good opportunity to develop supply chains also for these base materials considering that residual or non-commercial timber is actually mainly left on the felling site.

## 4.2 Materials and methods

### 4.2.1 Sampling

The sampling campaign lasted for 6 months when a total number of 51 pellet bags (15 kg each) were randomly collected from 25 producers and retailers active in the Italian market regardless to their provenience, cost or commercial names. The sample supplier was not told about the scientific scope of the work to avoid any kind of positive selection of the material prior the delivery. This procedure has ensured that no influence of the production parameter affecting the final quality of the pellet could occur due to the consciousness of the producer about the study going on. When possible the sampling was done directly by the staff of the University specifically trained for this operation. Standard EN 14778, the one regulating the sampling procedure, was adopted to ensure that the representativeness of the final sample was maintained.

All the possible information about the origin, source and raw material used, certification and measured parameters were recorded. Some information were retrieved from the producer or the importer when reachable. When this was not possible only the information reported on the packaging were considered.

Some samples were certified by national standards certification schemes (ÖNORM, DINplus, PelletGold), some were certified by the European schemes (ENplus), while some other were not certified at all. For some of them the quality parameters were stated printed on the bag while some others quality characteristics were completely undeclared. Those with stated values were divided into two groups: ones with precise data records and ones with only ranges of values declared. This second group made the analysis little more complicated due to the uncertainty of the properties stated. The validation of the declared data by the company was impossible for all the samples taken because:

- for some samples it was impossible to retrieve the producer name due to the lack in the traceability chain. In the international market there are no obligation for companies to state the producer's facility location and contact;
- some companies did not answer back after our specification request;
- some companies just declared on the bag some average values of the average production, regardless the presence of detailed and precise analysis;
- some bags were totally blank with no reference about producer, trader or even quality specification.

The same issue was recorded for the origin of the product, as for some pellets the production plant and the source of raw material were declared, while for some others no information about the producer, but only about the dealers, were given.

In addition five reference feedstock materials were collected, sorted and pre-dried. After grinding in a hammer mill (sieve apertures 6 mm), the moisture content was normalized (12%) and each material was pelletized using an industrial pilot plant (ring die aperture 6 mm), that reproduced the average production condition available. The pellets produced were sampled and analyzed for the scope of this work as they were processed under the same conditions of feedstock pre-treatment and seasoning, production settings and post-treatment. Two species were used: Norway spruce (*Picea abies* Karst) and beech (*Fagus sylvatica* L). For the former, whole tree (AbTr), whole stem (AbSt) and debarked stem (AbDe) were collected. For the latter whole stem (FaSt) and branches (FaBr) were collected. Finally a virtual certified sample (VCS) was added to the population used to build the model. Its properties were set on the minimum-maximum requirements level for a pellet sample to be certified according with the ENplus scheme (EPC, 2013) according to EN 14961-2. The specific moisture state at which each quality parameter was considered refers to the value measured directly on the sample (*as received*), that includes the moisture level within the wood except for the ash (A) which determination implies the complete evaporation (Table 4-1).

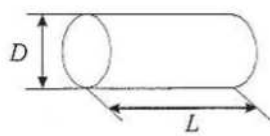
The expected outcome is an overview on the actual level of pellets quality within the Italian market and the verification of their compliance with the national regulations on labeling and certification (Table 4-2). The samples were processed at the Biofuel Laboratory of the Padova University for further analysis. At the arrival in the laboratory all the samples were

managed following the EN 14778 (sampling) and EN 14780 (sample preparation) on sampling reduction and test sample preparation. To do so all samples were transferred to some anonymous plastic containers and marked with a unique ID number to make them recognizable afterward in the study.

Table 4-1: pools of variables selected for the models construction

Variable	Abbreviation	Unit	Limit value for ENplus A1 class
Moisture content as received	$M_{ar}$	%	$\leq 10$
Mechanical Durability as received	$DU_{ar}$	%	$\geq 97.5$
Bulk Density as received	$BD_{ar}$	$\text{kg m}^{-3}_b$	$\geq 600$
Solid Density as received	$DE_{ar}$	$\text{g cm}^{-3}$	$\geq 1.2$
Ash content on dry basis	$A_d$	%	$\leq 0.7$
Low Heating Value as received	$LHV_{ar}$	$\text{MJ kg}^{-1}$	$\geq 16.5$

Table 4-2: reference values for the definition of pellet quality as define by the EN 14961-1

<b>Origin and source UNI EN 14961-1</b>	Woody biomass Herbaceous biomass Fruit residues biomass Blends and mixture	
<b>Traded form</b>	Pellets	
<b>Dimensions (mm)</b>		
Diameter (D) and length (L)		
D 06	D = 6 ± 1.0 and 3.15 ≤ L ≤ 40	
D 08	D = 8 ± 1.0 and 3.15 ≤ L ≤ 40	
D 10	D = 10 ± 1.0 and 3.15 ≤ L ≤ 40	
D 12	D = 12 ± 1.0 and 3.15 ≤ L ≤ 50	
D 25	D = 25 ± 1.0 and 10.0 ≤ L ≤ 50	
		
<b>Moisture content, M (% on fresh bases ) UNI EN 14774-1, UNI EN 14774-2</b>		
M 10	≤ 10	
M 15	≤ 15	
<b>Ash content, A (% on dry bases) UNI EN 14775</b>		
A 0.5	≤ 0.5	
A 0.7	≤ 0.7	
A 1.0	≤ 1.0	
A 1.5	≤ 1.5	
A 2.0	≤ 2.0	
A 3.0	≤ 3.0	
A 5.0	≤ 5.0	
A 7.0	≤ 7.0	
A 10.0	≤ 10.0	
A 10.0+	> 10.0 (max value to be stated)	
<b>Mechanical durability, DU (%) UNI EN 15210-1</b>		
DU 97.5	≥ 97.5	
DU 96.5	≥ 96.5	
DU 95.0	≥ 95.0	
DU 95.0-	< 95.0 (max value to be stated)	
<b>Fines, F (% , &lt; 3.15 mm), UNI EN 15149-1</b>		
F 1.0	≤ 1.0	
F 2.0	≤ 2.0	
F 3.0	≤ 3.0	
F 5.0	≤ 5.0	
F 5.0+	> 5.0 (max value to be stated)	
<b>Bulk density, BD (kg·m<sup>-3</sup>) UNI EN 15103</b>		
BD 550	≥ 550	
BD 600	≥ 600	
BD 650	≥ 650	
BD 700	≥ 700	
BD 700+	> 700 (max value to be stated)	
<b>Additives (%)</b>		
<b>Net calorific value, Q (MJ·kg<sup>-1</sup> or kWh·kg<sup>-1</sup>) UNI EN 14918</b>	Min value to be stated	
Chemical composition (macro- and micro-elements)		

#### 4.2.2 Physical and mechanical properties

The quality analyses were based on the classes proposed by the standard EN 14961-1. It delegates to other satellite norms the role to specify the methodology and procedures to determine the quality parameters. More deeply into detail the standard EN 14961-2 was applied whenever possible.

The quality analyses were done based on the classes proposed by the standard EN 14961-1 (Fuel specifications and classes. Part 1: General requirements). It delegates to a number of other satellite norms the role to specify the methodology and procedures to determine the quality parameters. More deeply into detail we applied, wherever possible, the standard EN 14961-2 (Fuel specifications and classes. Part 2: Wood pellets for non-industrial use). Pellets produced according to this European Standard can be used in pellet stoves that are tested according to EN 14785, pellet burners tested according to EN 15270 and pellet boilers or integrated-pellet burner systems tested according to EN 303-5 (< 500 kW<sub>th</sub>).

The fines content (F), mechanical durability (DU), moisture content (M), bulk density (BD), diameter (D), length (L), particle density ( $\rho$ ), ash content (A), calorific value (Q), and metals content were determined (Table 4-2).

The amount of dust and **fines** was determined by weighting and sieving the sample as stated in the EN 15210 (Solid biofuels. Determination of mechanical durability of pellets and briquettes. Part 1: Pellets). The measurement was carried out on the total amount of pellets for each bag to be able to measure to complete amount of fines produced during the packaging, handling and transporting phases. A 3.15 mm round sieve ISO 3310 certified was used to hand sieving the sample. The sieving was done avoiding creation of new fine particles but in a way that the fine particles were completely separated. This was done shaking a sample portion of 1.5 kg in some circular movements on a sieve of 40 cm diameter as suggested by the standard.

Then the weight of the amount of pellets retained on the sieve allowed the calculation of the initial amount of particles passing through the 3.15 mm sieve in the sample.

The **mechanical durability** of pellet is correlated with the mechanical properties of the pellet and with its tendency to disintegrate under stress. Once the pellets have been sieved the mechanical durability test was performed on 500 g subsamples on 3 repetitions. A prototype box tumbler machine was involved (Figure 4-1). One baffle is placed inside the container to

enhance the tumbling effect. The percentage of pellets remained integral, compared to the initial subsample weight gives the DU value (EN 15210 - Solid biofuels Determination of mechanical durability of pellets and briquettes Part 1: Pellets). The sieving after tumbling was done following the same procedure as for fines.

Calculations were done according to:

$$DU = \frac{m_A}{m_E} * 100$$

where:

DU mechanical durability, in%;

$m_A$  mass of sieved pellets after the tumbling treatment, in g;

$m_E$  mass of pre-sieved pellets before the tumbling treatment, in g.



Figure 4-1: pellet durability tester



The **moisture content** on wet basis was determined following the EN 14774-1 (Solid biofuels. Determination of moisture content - Oven dry method Part 1: Total moisture - Reference method) by placing 3 subsamples of each pellets type in a ventilated oven at 105°C until constancy in weight of two determination after one hour. The different buoyancy of the empty cold and hot steel containers was considered.

Calculations were done according to:

$$M = \frac{(m_2 - m_3) - (m_4 - m_5)}{(m_2 - m_1) + (m_6)} * 100$$

where:

$m_1$  mass in g of the empty tray;

- $m_2$  is the mass in g of the tray and sample before drying;
- $m_3$  is the mass in g of the tray and sample after drying;
- $m_4$  is the mass in g of the reference empty tray before drying (weight at room temperature);
- $m_5$  is the mass in g of the reference empty tray after drying (weight when still hot);
- $m_6$  is the mass in g of moisture associated with the packing.

The **bulk density** (EN 15103 Solid biofuels. Determination of bulk density) was determined by the mean of a 5 l steel container (Figure 4-2). The measuring container has a filling volume of 5 l ( $\pm 2\%$ ). It has an precise diameter of 167 mm and an precise height of 228 mm measured in the inner volume. The calculation is determined by the relationship between the mass of some pellets and their bulk volume. The container was firstly overfilled, then the material compacted by dropping the container from 15 cm on a hard surface and finally refilled and leveled to the top. The final weight referred to the cubic meter (1000 l) gives the bulk density value (i.e.: multiplying the net weight 200 times).



Figure 4-2: apparatus to measure the bulk density

The **dimensions** were measured by the mean of a caliper recording the length in mm of a 80 g subsample, selected from the whole pellet batch as specified in the standard EN 16127 (Solid Biofuels. Determination of length and diameter of pellets). In order to determine the diameter class, a minimum of 10 pellets were selected and their diameter measured twice in perpendicular positions and averaged. Pellets with a length greater than the requirement were separated from the test portion by hand and measured separately.

The **density of each single pellet** was determined as stated in the EN 15150 (Solid biofuels Determination of particle density).

The standard describes the method for the determination of the density of the single average particle of a pellet sample. The measuring of article density does not give an absolute precise value, and to achieve reasonable level of repeatability the operating conditions have to be standardized to determine comparative repetitions to be made since the methodology involves a number of critical points that can lead to measuring errors.

The calculation is determined by the relationship between the mass of a particle and its volume. For solid materials with irregular shape the first source of error for the density estimation is the volume, since the mass is easily measurable by precision weighting.

Both mass and volume of an individual particle or a group of particles are determined. The volume is measured by the buoyancy in a liquid. This method is based on the physical principle that the buoyancy of a body equals the weight of its volume filled with a specified fluid (Archimedes principle). The different weight between the pellet measured in air and the following weight measured in liquid (distilled water) marks its buoyancy. The volume of the sample pellet is retrieved considering the specific density of the liquid used at room temperature (20°C).

The particle density could also be estimated by stereometric means by cutting the pellets in sharp surfaces to determine their volume by measuring their dimensions. Due to the higher variability between the replications when applying the stereometric measuring principle this methodology was discard (Rabier et al., 2006).

The density determination kit (Figure 4-3) consists of a metal arch that is placed on a analytical scale with  $10^{-4}$  g accuracy. It is composed of a bridge that keeps the glass of water high and separated from the weighing plate. In this way the glass and its content never loaded the balance. The glass is filled with a solution of water and a detergent ( $1,5 \text{ g} \cdot \text{l}^{-1}$ ). A supporting U frame keeps the weighing dish out in air and lets a second dish to hung freely into the beaker glass. The dish is large enough to hold at least four pellets a time (dish diameter: 3.5 cm). The supporting frame holding the dish is loaded on the balance plate. The bottom of the dish is perforated by small openings, which allow the liquid to fill the dish from underneath when it is submerged and avoid its complete floating. No other additional weights are to be put on the dish to make it sinking, since pellets density is generally higher

than water one. In some cases a metal wire was used to fasten the pellets to the dish and help the sinking phase.

Calculations were made according to:

$$\rho = \frac{m_a}{(m_a - m_l)} * \rho_l$$

where:

$\rho$  is the density in  $\text{g cm}^{-3}$  of the pellets

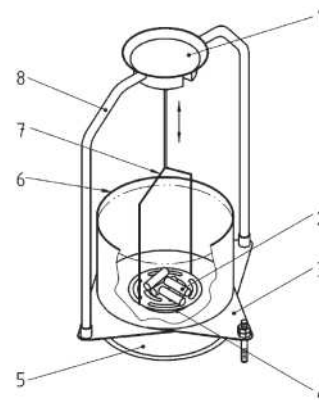
$m_a$  is the mass of the sample in air (including sample moisture)

$m_l$  is the mass of the sample in liquid (including sample moisture)

$\rho_l$  is the density of the applied liquid in  $\text{g cm}^{-3}$ . The density of the liquid (water and detergent) was considered to be  $0,9958 \text{ g cm}^{-3}$  at  $20^\circ\text{C}$ .



Figure 4-3: instrument for determination of the particle density of pellets



- 1 weighing dish (weighing in air)
- 2 pellets
- 3 bridge
- 4 perforated submergence dish (weighing in water)
- 5 weighing plate (balance)
- 6 beaker glass
- 7 dish suspension
- 8 supporting frame

A subsample was prepared for further analysis using a laboratory cutting mill to reduce the particle size distribution to a nominal top size of 1 mm (general analysis sample - GAS) following the standard EN 14780. This European Standard describes methods for reducing combined samples (or increments) to laboratory samples and laboratory samples to subsamples and general analysis samples (GAS). The method described in this European Standard may be used for sample preparation, for example, when they are to be tested for

calorific value, moisture content, ash content, ash melting behavior, chemical composition, and impurities.

The **ash content** was determined on the GAS following the EN 14775 (Solid biofuels. Determination of ash content). A minimum of two determinations was carried out on the test sample. The procedure comprises four phases: complete cleaning and drying of the crucibles done in the muffle at 550°C. Crucibles are then let to cool down in a desiccator to prevent any moisture adsorption from the air. After that each crucible was weighted to the nearest 10<sup>-4</sup> g, filled with minimum 1 g of GAS and re-weighted for the fresh weight. The filled crucible was put in a drying cabinet at 105°C for at least 4 hours (until constancy in weight), cooled into the desiccator and weighted for the dry weight. Finally they were put into the muffle and heated up to 550°C following a specific ramp procedure. After few hours, when the complete combustion of all the organic matter has occurred, the crucibles were cooled down and weighted to determine the ash amount.

Calculations were done according to:

$$A_d = \frac{(m_3 - m_1)}{(m_2 - m_1)} * 100 * \frac{100}{100 - M}$$

where:

A<sub>d</sub> ash content;

m<sub>1</sub> is the mass, in g, of the empty dish;

m<sub>2</sub> is the mass, in g, of the dish plus the test sample;

m<sub>3</sub> is the mass, in g, of the dish plus ash;

M is the % moisture content of the test sample at the moment of the determination.

The **calorific value** was determined according to the EN 14918 using the IKA C-200 calorimeter which gives the high heating value as received (HHV<sub>ar</sub>).

To determine the high heating value as received (also called gross calorific value as received GCV<sub>ar</sub>) a weighed portion of the general analysis sample of the pellet is burnt in high-pressure oxygen bomb under specified conditions. The heat capacity of the calorimeter is determined in calibration experiments. This is done burning some certified benzoic acid pellets under the same conditions. 1.5·10<sup>-3</sup> l of water is added in bomb during the test preparation to create saturated vapor atmosphere before the running of the test. This

allows all the water (the one newly created during combustion and the one of the sample moisture) to be considered as remained liquid (energetically passive).

The low heating value (also called net calorific value GCV) of the biofuel is then obtained by calculation from the gross calorific value determined on the analysis sample discounting from the HHV the energy invested to make all the water to evaporate.

The calculation of the net calorific value requires information about the moisture and hydrogen contents of the analysis sample. Further conversions allow to determine the following values (Table 4-3).

Table 4-3: description of different heating values measurements

High heating value as received	HHV <sub>ar</sub>	Calculated at the moisture content of the original sample
High heating value as analyzed	HHV <sub>aa</sub>	Calculated at the moisture content of the sample at the moment of the test
High heating value on dry basis	HHV <sub>db</sub>	Calculate on anhydrous sample
High heating value on dry basis ash free	HHV <sub>dbaf</sub>	Calculate on anhydrous sample not considering the weight of ashes
Low heating value as received	LHV <sub>ar</sub>	Calculated at the moisture content of the original sample
Low heating value as analyzed	LHV <sub>aa</sub>	Calculated at the moisture content of the sample at the moment of the test
Low heating value on dry basis	LHV <sub>db</sub>	Calculate on anhydrous sample
Low heating value on dry basis ash free	LHV <sub>dbah</sub>	Calculate on anhydrous sample not considering the weight of ashes

#### 4.2.3 Chemical analysis (in collaboration with DiSC of UNIPD)

The general procedure comprises the following steps:

- finding the optimum digestion method (optimization);
- mineralization (digestion) of the samples in the microwave reactor;
- analysis of the ICP reacted samples;
- results evaluation.

The first results from the digestion were used to assess the amount of sample to be mineralized: it is needed a compromise between costs and results of the analysis (it is necessary to ensure sufficient sample consistency to ensure that the concentration of metals in the digested sample exceeds the detection limit of the instrument).

The first step was the **optimization of digestion method**.

Per each sample 0.3 g were taken, digested in 3 g of ultra-pure nitric acid. If 0.6 g would be used, some samples could carbonize during digestion in microwave, while using less pure acid could lead to the contamination of the sample with external metals.

The optimal method for the digestion of wood in the microwave reactor was chosen as follows: 4 minutes ramp up to 200°C at 400 psi for 2 minutes.

To verify and cross-check the repeatability of the method a pellet sample was selected (number 46) and 7 repetitions of digestion by the reference method were made. The results obtained in ICP then had to give the same values, proving that the method of sample preparation (microwave digestion) was repeatable.

Figure 4-4 shows the summary graphs of the mineralization of the reference (3 g ultra-pure nitric acid 67%) and one of the samples. This is also representative of all the others (all have very similar trends).

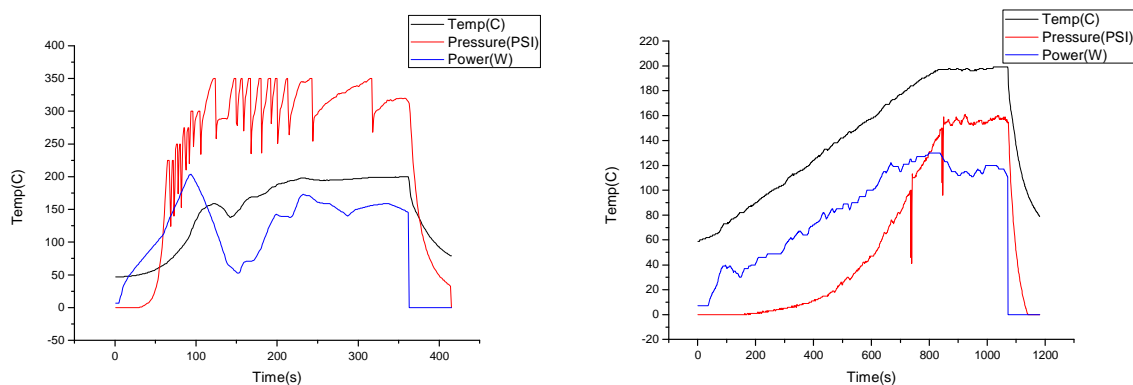


Figure 4-4: trends in temperature, pressure and power of samples in the digester: left pellet sample 46, right reference sample

### Mineralization of samples

Once the methodology was tested and the reproducibility verified, all 51 pellet samples were milled with a laboratory cutting mill down to a nominal top size of 1 mm. Sawdust samples were then digested in one repetition each. The solutions resulting from the mineralization were mostly clear and their color ranges between yellow and green. Only a few samples have submitted turbidity or gray precipitate on the bottom of the tube.

The European standard UNI EN 15297 (Solid biofuels - Determination of minor elements - As, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, V and Zn) suggests for digestion not only nitric acid. For this reason a mixture 3 : 1 = nitric acid : hydrogen peroxide 30% was prepared and 5

repetitions of the same sample with the addition of 1 g of hydrogen peroxide done to evaluate the difference in the results. All the samples digested with hydrogen peroxide were comparable to the samples tested before (only nitric acid), either for the solution quality and for reaction trend. This led to the decision of using only nitric acid for digestion.

All the solutions of the mineralized samples were then diluted coming to be 6% acid solutions, ready for analysis in ICP-MS.

### **ICP test**

Inductively coupled plasma mass spectrometry (ICP-MS) is capable of detecting metals and several non-metals at really low concentrations in digested liquid samples. This is achieved by ionizing the sample and then using a mass spectrometer to separate and quantify those charged ions.

The instrument was calibrated using an external calibration method and a 12 multi-element standard solutions at concentrations of 0.2 - 0.5 - 1 - 5 - 10 - 25 - 50 - 100 - 250 - 500 - 750 - 1000 ppm for each element.

#### **4.2.4 Statistical analysis**

As stated by (Forina et al., 2008) multivariate class-modeling techniques answer to the general question if an object O, stated of class A, really belong to class A. This is a typical question in multivariate quality control. On the contrary, the classification techniques assign objects to one of the classes in the problem. Class-modeling techniques calculate the “prediction probability” with a classification threshold for each modeled class. By using a class-modeling approach, it is possible to attribute objects not only into one or more classes but also to none (i.e., in this case, the object is an outlier).

In order to find an index able to assess the general quality of a population of wood pellets samples randomly taken among commercial products on the Italian market and to define a baseline for quality parameters of the general pellet market, a SIMCA (Casale et al., 2007; Hall and Kenny, 2007; Menesatti et al., 2013) was applied to the six qualitative variables dataset. The SIMCA model, computed with the software V-Parvus 2010, is used for supervised pattern recognition. It is a collection of PCA (Principal Component Analysis) models (computed by the NIPALS algorithm), one for each class of data set (one in this case), after separate category autoscaling. SIMCA cross-validates (CV) the PCA model of each class (training set), splitting the data (evaluation set) into 4 contiguous groups (CV groups). In our

cases, the modified model with expanded range was used in respect to the technique firstly introduced (Wold and Sjöström, 1977). Also, in building the models the unweighted augmented SIMCA distance was considered. For each class, the number of significant components of the inner space was estimated considering five PC (lowest noise found). For each modeled class a critical square distance based on the F-distribution, was calculated using 95% confidence interval. The class boundary was also determined according the confidence limit. In CV, new observations are projected into each PC model and the residual distances calculated. An observation is assigned to the model class when its residual distance from the model is below the statistical limit for the class. SIMCA allows both the classification and modeling analysis. For classification, all the observations have to be attributed into one of the pre-defined classes. The efficiency was indicated by a classification (training set) and a prediction (evaluation sets) matrix, which reported the percentage of correct classification for each considered class and total percentage ability. The observations for each class, classified outside the model, were also reported. SIMCA expressed the statistical parameters indicating the modeling efficiency. In fact, the observation may be found to belong to multiple classes or to fit not to any of them (outlier). Also unknown objects could be either classified into one of the classes or recognized as outlier. The modeling efficiency was indicated by sensitivity and specificity parameters. The sensitivity is the measure of how well the classification test correctly identifies the cases really belonging to the class. The modeling power of each variable, that represent the influence of that variable in the definition of the model, was expressed.

In order to express an index for each pellet sample, square SIMCA distances were linearized converting the values into logarithmic scale and then translating them adding a value of 2.5 (in order to have all positive values).

The SIMCA model was built on a training data set consisting of 18 out of the 23 certified samples (included the VCS) with a measured set of attributes. This partitioning was optimally chosen with the Euclidean distances based on the algorithm of (Kennard and Stone, 1969) that selects objects without the *a priori* knowledge of a regression model (i.e., the hypothesis is that a flat distribution of the data is preferable for a regression model). The rest of the samples (34) were used as external test.

### 4.3 Results and discussion

The total explained variance of the five SIMCA model principal components is 95.77%.

The adimensional logarithmic translated square critical distance, based on the F-distribution, is 2.88. This identifies the class-boundary of the model, below which the distance value of an observation is included in the model, while a distance value higher is rejected. All (100%) the 18 certified samples (included the VCS) used to build the model were included in the SIMCA model (internal sensitivity = 100%). The certified samples used as external test were also included in the SIMCA model (external sensitivity = 100%).

Among the 34 non-certified samples used as external test, 19 were included in the model. These results are summarized in Figure 4-5. On the other hand none of the certified samples were excluded from the model.

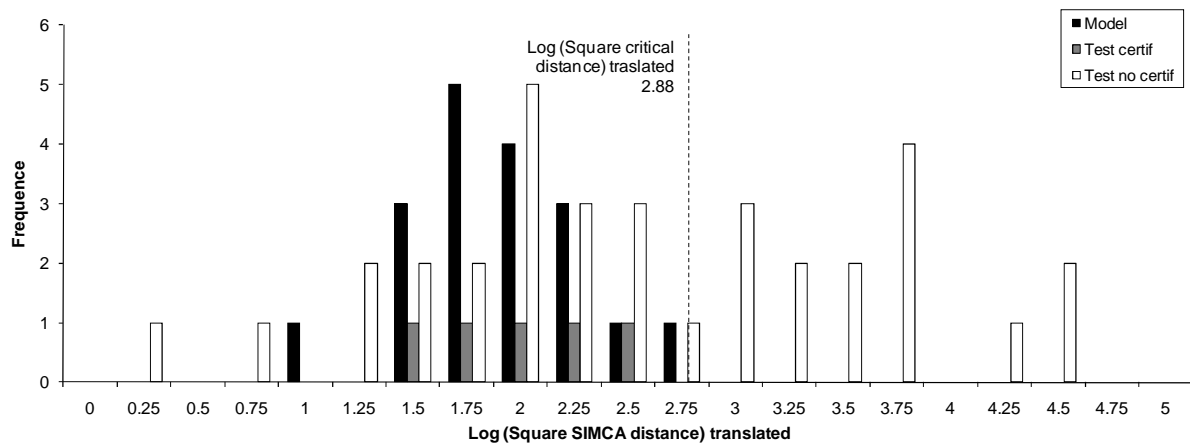


Figure 4-5: Number of pellet samples having different logarithmic translated square SIMCA distances from the 18 certified samples used to build the model (*Model*; black), the 5 certified samples used as external test (*Test certif*; Gray) and the 34 non certified samples used as external test (*Test no certif*; White). The logarithmic translated square critical distance is reported (dashed line).

The adimensional logarithmic translated square SIMCA distance represents a real metric scale where each sample could be positioned. It expresses the inclusion (values below the critical distance) in the model or the distance from the model. Higher distance values could be considered a proxy of lower quality, and this lower quality is obtained in a multivariate approach. Up to now in literature just one paper in Italy treated the aim of the pellet market characterization based on quality parameters and adopting a multiple correlation approach (Toscano et al., 2013).

The modeling power values of the six variables are reported in Figure 4-6. Ad, LHV<sub>ar</sub> and BD<sub>ar</sub> are the most important variables and this is practically reflected in the importance that

these three variables have in the grading of commercial pellets and the consequence related to their use. No power values are below 0.59 (adimensional scale).

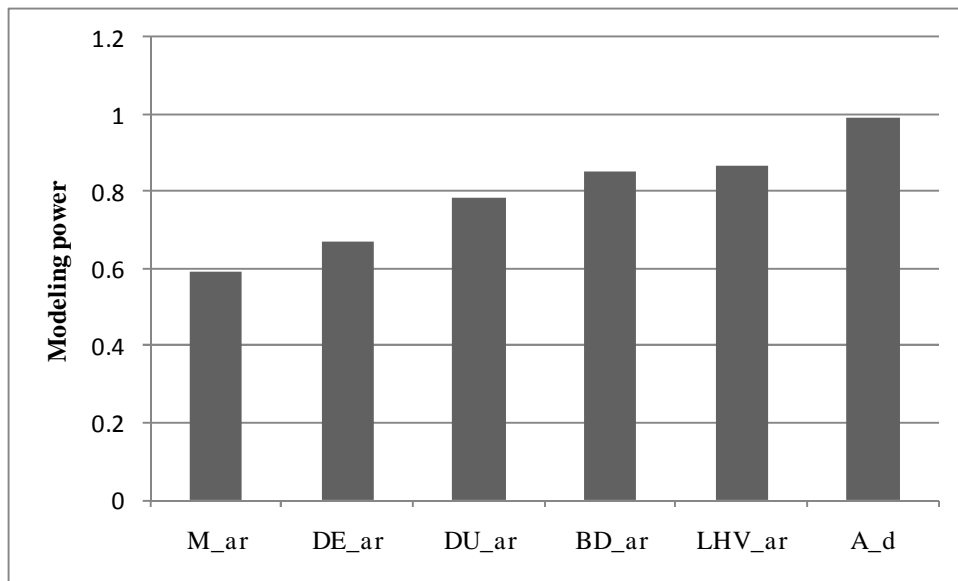


Figure 4-6: Variables included in the model ordered by their corresponding contribution to the model definition (modeling power)

These variables were the main responsible for the exclusion of the samples from the SIMCA model. The position of samples which raw materials were agricultural residues or a mixture of non-debarked wood species is negatively affected by  $A_d$ . At the same time the modeling power of this variable is the highest among the others, giving reason of its weight in the model classification.

The relationship between the  $A_d$  and the raw material used for the pellet production is confirmed considering the five samples specifically produced for the aim of the study. In fact for all of them the ash content was exceeding the maximum limit value. Only the ash content for  $AbDe$ , which lacks the bark fraction due to the debarking process, meets the maximum ash threshold. On the other side the absence of bark, that act as binding agent during pelletization (Lestander et al., 2012a), lowered the  $DU_{ar}$  below the minimum.

The  $LHV_{ar}$  was the second major contributor in the exclusion of these samples due to low values. It has to be specified that the high heating value of wood is more or less constant with some slight differences related to the content of resins and oils in some species. On the other hand the low heating values is mainly affected by the moisture content of the sample, when some of the energy content has to be invested in the evaporation of such water. This is the case of the excluded samples, which moisture content was slightly over the population average.

The  $BD_{ar}$  of excluded samples is in general within the minimum specified requirements except for one of them.

On the other hand it can be noticed that some samples, which quality parameters exceed the minimum value for a specified variable, are still accepted from the model. This is explained taking into consideration that the model is built on the real quality of pellet samples taken from the actual market that have no direct connection with the threshold of the certification schemes. While the limits imposed by the schemes are fixed and based on some preliminary studies, the model is adapted to the representativeness of the population been sampled for this work. The acceptance of all the certified pellets within the model is, on the other hand, a remark that the model itself is fully adapted and applicable.

In Table 4-4 are reported the concentrations of each element within the material under analysis (values expressed in ppm). The data in **bold** and underlined are the concentrations that exceed the standard limit for the pellet (with the exception of iron, whose limit is not provided by UNI EN). It is possible to notice that only few samples are actually exceeding some of the limits imposed by the EN standard on high quality wood pellets. In particular only pellet number 6 has more than one element over the acceptable limits, but from a deeper analysis of the provenance and the declared value no relationship or clear causes of this could be found.

Samples number 22 – 31 – 33 – 38 have pretty high copper contents values and this is found to be in strict relationship with the raw material involved in their production: vineyard pruning. It is well known that the quality of this kind of material is strongly affected by the chemical treatments done during the vegetative season with copper-based solutions. The utilization of such biofuel is therefore possible with no safety hazards only in specific designed plants.

Table 4-4: metals concentration in tested samples. In **bold** underlined values that exceed the upper limit for A quality class

	<b>Cr</b>	<b>Ni</b>	<b>Cu</b>	<b>Zn</b>	<b>As</b>	<b>Cd</b>	<b>Pb</b>
<b>Limit A1 class UNI EN 1496</b>							
<b>Sample ID</b>	<b>10</b>	<b>10</b>	<b>10</b>	<b>100</b>	<b>1</b>	<b>0.5</b>	<b>10</b>
1	0.36	1.03	1.49	8.59	0.03	0.05	0.62
2	0.72	0.86	1.61	23.24	0.03	0.20	0.46
3	0.10	0.11	0.78	30.70	0.03	0.16	0.07
4	1.65	0.87	2.58	8.24	0.03	0.06	0.42
5	1.86	2.39	1.98	12.69	0.45	0.11	1.04

6	6.57	2.03	<b><u>12.31</u></b>	<b><u>107.96</u></b>	0.98	0.40	<b><u>34.73</u></b>
7	0.23	1.22	1.58	8.07	0.03	0.06	0.28
8	1.43	0.27	0.79	10.77	0.03	0.10	0.18
9	0.43	0.24	1.15	16.64	0.03	0.17	0.67
10	0.46	0.20	0.98	15.11	0.03	0.03	0.04
11	1.48	1.39	1.62	19.36	0.03	0.11	0.68
12	0.13	0.12	0.77	12.15	0.03	0.15	0.34
13	0.13	0.13	0.82	23.98	0.03	0.09	0.03
14	0.18	0.18	0.85	14.23	0.03	0.10	0.17
15	1.35	0.56	2.07	42.71	0.03	0.15	7.92
16	0.16	0.10	0.83	32.38	0.03	0.13	0.29
17	0.43	0.24	1.07	9.05	0.03	0.11	0.49
18	0.65	0.90	1.56	10.53	0.03	0.07	0.38
19	0.16	0.59	0.97	11.48	0.03	0.08	0.16
20	0.09	0.46	1.76	9.07	0.03	0.08	0.17
21	0.15	0.06	3.68	8.40	0.03	0.03	0.03
22	2.45	1.42	<b><u>15.02</u></b>	39.56	0.03	0.04	0.59
23	0.39	0.31	0.77	11.17	0.03	0.21	0.46
24	0.46	0.35	1.24	8.40	0.03	0.13	0.30
25	1.83	0.94	1.45	13.26	0.03	0.04	1.26
26	0.30	0.33	1.07	3.21	0.03	0.07	0.11
27	0.53	0.80	1.49	16.32	0.03	0.14	0.31
28	0.85	0.52	0.96	42.73	0.03	0.03	0.33
29	0.32	0.35	1.22	26.94	0.03	0.17	0.35
30	0.41	0.25	0.90	11.52	0.03	0.08	0.24
31	0.59	0.42	<b><u>16.13</u></b>	47.34	0.03	0.03	0.42
32	0.27	0.15	2.85	38.64	0.03	0.03	0.51
33	8.63	4.56	<b><u>18.17</u></b>	45.84	0.03	0.03	0.52
34	0.36	0.30	1.27	13.24	0.03	0.15	0.32
35	0.28	0.18	0.83	54.76	0.03	0.11	0.19
36	1.41	0.97	5.74	11.97	0.03	0.06	0.83
37	1.16	2.25	2.01	14.42	0.83	0.15	2.04
38	7.63	4.83	<b><u>12.81</u></b>	54.16	0.29	0.12	1.42
39	0.25	0.32	0.87	9.72	0.03	0.03	0.75
40	0.16	0.51	1.28	6.43	0.03	0.03	0.42
41	0.12	0.11	0.68	16.67	0.03	0.12	0.20
42	0.10	0.10	0.73	43.40	0.03	0.10	0.22
43	0.26	0.21	0.74	15.94	0.03	0.09	0.52
44	0.36	0.34	1.37	27.85	0.03	0.03	0.35
45	0.34	0.30	1.20	48.59	0.03	0.05	0.82
46	0.17	0.40	0.85	23.37	0.03	0.10	0.29
47	1.65	0.85	1.11	10.81	0.03	0.05	0.24
48	1.41	0.72	7.58	17.63	0.03	0.03	0.58
49	0.24	0.15	2.08	12.14	0.03	0.03	0.10

50	0.09	0.09	0.70	21.63	0.03	0.07	0.10
51	0.43	0.38	1.12	3.61	0.03	0.03	0.16

#### 4.4 Conclusions

The innovative multivariate concept adopted in this study is applied for the construction of a model based on the real quality parameters of wood pellets. The SIMCA approach allows to define a critical distance for each sample that defines its possible certificability and locates it in a specific quality level or range. While the actual certification schemes are based on a single sample analysis over a period of one year the model can be used and updated continuously giving more narrow limits to the certification bodies. The innovation is represented by considering the entire dataset, that can be set at national or international level, in a multivariate fashion with respect to traditional approaches which generally only consider each single parameter referred to some definite limits (minimum or maximum acceptable values for that specific parameter). Such methodology can also be spread and applied directly by the market operators to monitor the quality of the measured data and their reaction to improvement action taken by producers and traders.

Moreover the model, designed to be dynamically updated, can be further improved by adding new samples during its revision to periodically adapt it to the actual quality changes within the market due to raw material variations, seasonal quality fluctuations, import from other countries. The proposed multivariate SIMCA model could be easily implemented on a web site where web users such as other Italian or European laboratories could test their samples and verify the certificability of their pellet.

The low number of variables necessary to make the model stable and reliable together with the low investment necessary to determine them is the key factor for the application of the model in the practice.

## 5 Pellets from forest residues

### 5.1 Introduction

A preliminary study was done to detect and identify the most frequent harvesting techniques adopted in Italy for the utilization of timber. This was done considering that the study should test not a new raw material supply chain but a integrated raw material supply chain, that generates from the optimization of already existing methodologies from retrievable products which properties have already been studied (Dibdiakova et al., 2014). That's the case of forestry residues, mainly represented by top and branches of tree that are accumulate at the roadside when the logging methods is the Full Tree extracted with a cable yarder. Another example is represented by the pre-commercial and commercial thinning. Often these operations are non-economic since the quality of the harvested timber is very low for industrial use. As soon as timber quality is not an issue for its energetic exploitation, the pellet production can represent an alternative.

Nevertheless the quality of the final pellet produced is a direct function of the incoming feedstock following the rule *what goes in, comes out*. Some studies have tested the influence of the raw material characteristics on the quality of the final pellet. It was demonstrated (Arshadi et al., 2008; Lestander et al., 2012a) that it is possible to produce high quality pellets from whole-stem logs that are not suitable for commercial purposes. To do so it is necessary to maintain certain moisture content in the raw material immediately prior to pelletizing. Optimum pellet quality can be obtained within a range of sawdust moisture content of 11–13%. The water determining the moisture content of the sawdust is, moreover, the responsible for lubricating of the die as it lowers the friction in the pelletizing process (Samuelsson et al., 2012).

Also the controlling of the blend composition of the feedstock is a key factor to predict the final quality. Some tests were carried out to produce pellets from biomasses differentiated by tree species. Their mechanical properties and combustion behavior were studied in order to select the best raw materials available in the area of study for pellet production. While the influences of blending raw material seem to be crucial for the main quality parameters

of pellets, it does not affect combustion, which remains similar to those of the individual raw materials (Gil et al., 2010).

The influence of bark content in the raw material was also demonstrated to strongly influence some pellet characteristic such as bulk density, calorific value and ash content. All these values increase with higher percentages of bark in the mixture, but such effect on ashes makes the overall quality lower especially for pellet used in small appliances. Other studies have shown that pellets made from pure bark had the best mechanical properties compared with traditional wood pellets containing various concentrations of bark (Filbakk et al., 2011). No problems with sintering of ashes when the bark content was low (5 - 10%) have been registered. Fuel quality (chemical composition) and burner type (design of combustion chamber and setting of gases exchange) affect the amounts of ash deposit produced. As the same, the fuel composition is the main responsible of the degree of sintering and the strength of the residual deposits. The sintering of ashes on the bottom part of the furnace negatively affects the combustion quality mainly due by the obstruction of the air channels on the bottom grid of the combustion chamber. It is therefore recommended that fuels rich in ashes, like bark and logging residues, should not be used in the domestic and residential pellet burners if not specifically design to burn such biofuels. Some controlled sintering tests of the produced ashes showed that critical slagging temperatures are reached already at 900 - 1000°C for bark and logging residues (Öhman et al., 2004). But such temperature values are normal for a high efficiency domestic pellet burners as well as for industrial heating plants (Eskilsson et al., 2004; Nussbaumer, 2003).

The main point, as said before, is that the Italian biofuel sector, and in particular the wood pellets one, is now unable to fulfill the total domestic demand of this product. On the other hand the forestry sector needs new answers for the management of big amount of low quality timber that have to be harvested during the pre-commercial thinning as well as huge quantities of low quality forestry residues that often are accumulated at the roadside during wood processing phases. The effectiveness of such operations depends on the proper organization of forest operations involved (Mirabella et al., 2014).

The feasibility study for the production of pellets from forest residues consists of checking the quality characteristics, defined by European standards UNI EN 14961, from pellets

obtained by processing selected plants parts from spruce and beech, which are considered to be the two main species used in the pellet production sector.

## 5.2 Materials and methods

To evaluate the possibility to do this the following experimental design was set up.

The overall process comprises the following phases:

1. the selection, felling and processing of representative trees in the forests. The species used for this study are spruce (*Picea abies* Karst) and beech (*Fagus sylvatica* L), selected as they represent the most widespread material available for energy purposes in our region. The collection of the material was done with the technical collaboration of the Belluno Regional Forestry Service in October 2012. Nine spruce trees and six beech trees were selected according with the prescribed dimensions (spruce 15-30 cm DBH, beech 10-20 cm DBH). Such diameters are considered to be representative of the average dimensions of the non-economic timber harvesting (especially in juvenile and pre-mature stands). This choice was taken considering that in this study a correct allocation of timber products and a responsible management of forestry woody resources has to be done (Notarangelo et al., 2013), also considering that the technology used to exploit such resources is mainly immature (Lindholm et al., 2010). For the experimental study though we do not consider saw logs as a profitable base material for energy production. Spruces with diameters bigger than 15-30 cm are considered to be delivered to the sawmill industry for low quality lumber production. Beeches with diameters bigger than 10-20 cm are considered to be designated to the production of firewood, when no other richer supply chains can be caught;
2. the selected trees were reduced into smaller parts and processed following the scheme in Table 5-1 to represent all the possible quality ranges of feedstock. Those materials were then comminuted with a disc chipper powered from the PTO of a tractor. The chipper was feed in such a way that the resulted hip was homogenous in terms of material composition and size distribution enough to be representative of the material being collected. The chipping operation was done separately for each

base material to avoid any contamination. The samples were then collected into sealed big-bags, labeled and transported;

Table 5-1: the experimental design

	Norway spruce	Beech
High quality	Debarked stem*	Whole stem
Medium quality	Whole stem	Branches $\varnothing < 5$ cm
Low quality	Whole tree	

\* with debarked it is intended a stem partially debarked as a result of harvesting with harvester or processor. For the beech, cutting with the harvester or otherwise debarking in forest is not practiced in common operations

3. the natural seasoning of the chipped materials was done in the facilities of the Azienda Agraria Sperimentale Lucio Toniolo in Legnaro. The wood-chips were put inside plastic bins provided with apertures on the side and bottom to allow the maximum circulation of air (Figure 5-1). Each bin contained approximately 250 kg of fresh woodchips;
4. in the first day, and then periodically for all the seasoning period, samples of wood chips were collected and brought to LabABC for the moisture content analysis. Moreover each bin was weighted every two weeks (Figure 5-2) to monitor the loss in moisture content during the seasoning (Figure 5-3). Each bin content was poured, during the overall period at least once, onto a clean surface and evenly spread to fasten the drying process and avoid organic matter losses due to the internal fermentation. After 80 days all the base materials were dried down to a moisture content of 10-15%;
5. the milling of the seasoned wood chips to reduce them to the nominal top size necessary for being pelletized. This was found to be equal to the diameter of the die channel through which the raw material has to pass during the pelletizing process. Since the pellet press die we used in this study was made for 6 mm pellets, this was also the diameter of the apertures in the hammer mill screen. The equipment used was a power hammer mill;

Spruce: debarked stem



Spruce: whole stem



Spruce: whole tree



Beech: whole stem



Beech: branches < 5 cm



Figure 5-1: wood chips collected and stored in plastic bins



Figure 5-2: weighting the bins

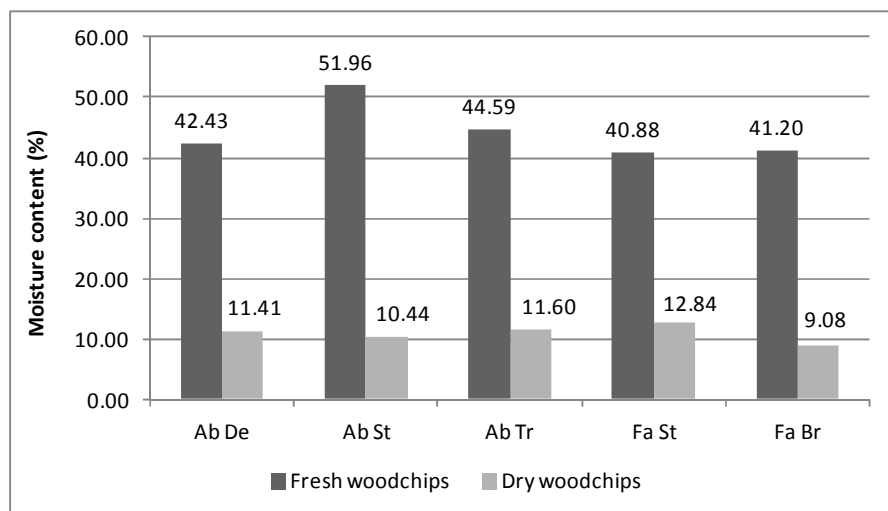
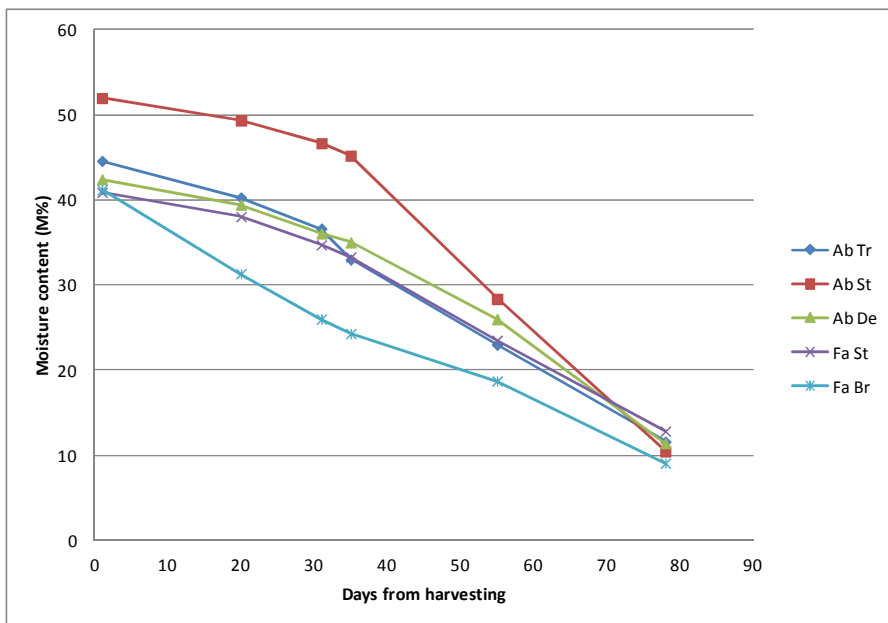


Figure 5-3: the seasoning and drying trends for the five base materials collected. Ab: Spruce, Fa: beech; Tr: whole tree, St: whole stem; De: debarked stem; Br: branches

6. once all the samples have been milled we proceeded with the balancing of the moisture content of the sawdust used as base material, since the moisture contents values of the wood chips seasoned were not uniform and adequate for the pelletizing. The conditioning was done by adding a certain mass of water, specific for each sample of sawdust, on the basis of its current water content, to bring it to the value of optimal moisture content before pelletizing of 12%. This value was selected considering the specific requirement of the pellet press used: the heating of the material during the pelletizing process allow the vaporization of part of the bound water and its subsequent release during the cooling procedure;
7. the production of wood pellets into a small scale pellet mill capable to reproduce the production conditions at industrial scale (pressure, temperature, dimensions).

The pelletizing of the material has been done by means of a small scale pellet mill for experimental use CLM200 (Figure 5-4) produced and made available by *La Meccanica* company, manufacturer of machines and equipment for pelletizing. The machine is equipped with a 20 kW electric motor. The die has a diameter of 200 mm, the holes of 6 mm and the die channel has a length of 30 mm. The speed of the feed screw and the length of the pellet in output were adjusted by the operator of the press in relation to the quality of pellet produced. Parameters such as the feeding screw speed have to be regulated to avoid the stop of the machine due to overloading of the press chamber with sawdust. The different speed of the feeding system also influences the time necessary for pellet to be extruded (i.e. the productivity of the machines) on the basis of the self-lubrication of sawdust when it undergoes high temperatures and pressures (Table 5-2). It was observed that the chosen die was suitable for both species (spruce and beech) and for all five different base materials, since it led to a uniform final pellet quality.

The absorption of the motor during the production was 18 A for the three types of spruce and 20 A for the two types beech. This difference in the power consumption during the pelletizing process is due to the different mechanical and physical properties of the two species. The hardness and higher density of beech produces a higher friction between the pellet and the wall of the die channel;

Table 5-2: production data from the experimental pelletizing process

		Ab De	Ab St	Ab Tr	Fa St	Fa Br
<b>Die holes diameter</b>	mm	5	5	5	5	5
<b>Die channel length</b>	mm	30	30	30	30	30
<b>Engine absorption</b>	A	18	18	19	20	20
<b>Screw feeder</b>	rpm	90	125	115	70	87
<b>Productivity</b>	kg h <sup>-1</sup>	33	41	73	54	56
<b>Moisture content of sawdust</b>	% approx.	12	12	12	12	12
<b>Pellet temperature</b>	°C	80	80	85	85	85
<b>Test duration</b>	min	39	29	22	26	20



Figure 5-4: the pellet press CLM200 open (left) and in operation (right)

- the cooling of pellet produced. The experimental pellet press was not equipped with any automatic cooling device. This could be a dangerous source of variability for the final mechanical properties of pellets produces because the high amount of steam produced by the evaporation of bound water in the sawdust at high temperature, if not properly removed, could condensate on the pellets walls and destroy them again.

To avoid this, pellets exiting the press were collected into a container and constantly moved to a sieve to remove dust and fines. Then, to cool them in the shortest time possible, they were introduced in a counter flow cooler specifically designed and built for the purposes of this study. The device consists of a box, open at the top, equipped with a grate placed horizontally at half height of the walls. In the lower part a electric fan blew fresh air from the outside into the cooler chamber and through the grate, cooling the overlaying grate (Figure 5-5). The pellets were let to cool down to room temperature and then placed in 5 sealed plastic bags and transported to the laboratory.



Figure 5-5: the cooling system in operation



9. the analysis of the pellet produced. This comprises the mechanical and physical qualification at Biofuel Laboratory of the Padua University, as well as the chemical characterization. This was done in accordance with all the European Standards (UNI EN) already available for the biomass characterization and in relation to the final destination of the biofuels. The analysis on pellets was done using the same methodologies explained in Chapter 4.2.2 for the commercial pellets;
10. the statistical evaluation of results. The objective of the statistical evaluation was to test if there are significant differences among the pellet samples produced with five different base materials. The test carried out is one-way ANOVA. The first step was to check for the basic assumption for the statistics applied. Levene's test was used to assess the similarity of the variances for the 5 datasets involved in the analysis

(origin). Usually the common statistical methodologies are based on the assumption that variances of the sample populations are equal within a certain confidential level and the test of Levene is used to verify this assumption. The null hypothesis that this test verifies is the similarity of the variances of the population. This is generally called homogeneity of variance or homoscedasticity. If the calculated P-value of the test is lower than the fixed threshold ( $P < 0.05$ ), the obtained differences among variances are unlikely to have occurred based on random sampling from a population with equal variances. Thus, the null hypothesis of homoscedasticity is rejected because there is a significant difference of the variances in the population. When Levene's test results in significant differences, it is necessary to adopt generalized tests, that are not dependent or influenced by the homogeneity of variance assumptions.

In our study only the test between Ash and Origin meet the basic requirement of homoscedasticity.

ANOVA tells that at least one of the groups tested differs from the other groups. However, ANOVA says nothing about which couple of groups differs. The method used to discriminate the differences is the LSD (Least Significant Difference) of Fisher. This allows to make comparisons between two means from two pairs of groups. Any difference larger than the LSD is considered a significant result. With this method, there is a risk of 5% to declare each pair of means significantly different when the actual difference is 0.

For all the other variables except the ash content, the homogeneity of variance was not demonstrated so some other *post hoc* tests have to be applied to determine such pairs. There are many *post-hoc* tests. The choice depends on the characteristics of the data but since in our case the observed variances are unequal the Tamhane test was found to be suitable.

### 5.3 Other materials studied

There are several studies focused on finding the ideal blend of biomass for the production of good quality pellets, mixing both different parts of the same species (wood, bark and needles) and different species (Arshadi et al., 2008; Filbakk et al., 2011; Gil et al., 2010). The mixtures of different raw materials have good results on the mechanical characteristics of

the pellets, whose durability is improved and the calorific value of the pellet thus obtained, however, remains similar to that of the starting materials.

Therefore by varying the content of bark and needles in the mixtures, the characteristics that are affected are primarily mechanical durability, calorific value and ash content. These values increase with increasing percentage of bark in the mixture, but the aspect that most influences the quality of the pellet, in a negative way, especially for small scale appliances, is the high content of ash, which excessive sedimentation can cause problems in the air circulation within the combustion chamber in the heating plant.

Pellets with high content of bark, on the other hand, can be used without this problem in industrial plants boilers which combustion chamber is equipped with mechanical and automatic ash removal.

In order to verify these observations, we studied the influence of bark, twigs and needles on the calorific value and ash content of mixtures of spruce, beech and larch.

## 5.4 Results

In the graphic presentation of results all the comparisons are referred to the average market value (horizontal line in graphs) from commercial samples (Chapter 4).

The Figure 5-6 shows the comparison of bulk density of dry woodchips and pellets produced from the same material. It can be noticed that the pellet bulk density is almost three times higher than the woodchips one, proving its effectiveness in compacting high amount of material and energy in the same volume. Looking to the differenced on BD within the five reference pellets it is possible to notice that: i) the highest value of bulk density is registered for AbDe, ii) statistical similarities can be detected for AbSt, FaSt and FaBr, iii) AbTr has the lowest values, statistical similar only with FaBr, iv) all the values are in line if compared with the market average except for AbDe exceptionally high. To find the reason why AbDe has this abnormal BD we compared it with the particle density values  $t$ . Figure 5-8 shows that: i) AbDe has the highest D value, ii) that differs statistically with all the others. On the other hand the particle density itself does not seem to be the reason of the BD supremacy of AbDe. The total length, given the same diameter of all pellets (6 mm), is in fact the lowest for that material (17 mm). This means that these pellets have a better capacity to occupy

efficiently the space, resulting in higher BD. The length is then a result of the low mechanical durability. Pellet of AbDe were prone to break soon after being pushed out of the die.

The mechanical durability of pellets has been determined and is shown in Figure 5-9. The low value of pellets produced only with wood is statistically different from all the other materials. This is mainly caused by the absence of bark in the feeding mixture, that has been proved to be a binding agent in the pellet manufacturing (Oberberger and Thek, 2010). That is also the reason why producers who use only clean sawdust in their facility have, most of the time, to improve the mechanical properties of pellets by adding some binders. All the other feedstock materials resulted in pretty durable pellets, comparable with the average values found in the market. Since the DU is tested at the actual moisture content and the results are referred to the sample *as received*, the figure shows also the actual moisture content of tested samples. It can be noticed that the final M differs significantly among all types of pellet. If we consider that the same moisture level of the five types of saw dust involved in the pellet production was ensured by initial conditioning, this suggests a different hygroscopicity behavior of base materials. Since the water present within wood fibers as bound water is demonstrated to play a key role in binding capacity of particles, the low value for SpDe (M 5.2%) contributes to the failure of DU result (Kaliyan and Vance, 2009; Lee et al., 2013).

The ash content of the five fresh samples of wood chips is shown in Figure 5-10. Wood chips of debarked spruce have a rather low content of ash (class A 0.5) because in this case only pure wood is sampled. The whole stems of spruce and beech both belong to the class A 1.0, due to the presence of bark in the sample. The increasing in ash content compared to an increase in the percentage of bark has already been observed by Filbakk et al. (2011) in Scots pine and can be explained primarily by mineral impurities naturally present in the bark. The increased ash content can also be caused by some organic matter losses during storage due to fermentation and degradation of wood by fungi and bacteria.

In beech branches the ratio between pure wood and bark is lower due to the small diameter of each branch and the consequent low volume of wood. Moreover this material is rich in buds and vegetative parts that lead to the high ash content value of 2.16%. The whole plant of spruce has also a high ash content (2.1%). The cause of this is two-fold: i) in AbTr there is a significant presence of bark and needles, that are typically richer in minerals and ashes than

pure wood, ii) some degree of sampling error. This error was due to the sampling of fresh woodchips from the whole tree material. The fresh twigs of spruce have maintained the needles attached until their complete drying, affecting the real percentage of needles, branches, wood and bark being collected. The same error was not repeated when collection and analyzing samples of dry and milled woodchips, when the grinding process has homogenized the whole material. Other authors recorded the same sampling limit when material stored in a windrow showed lower ash content than fresh logging residues, which was mainly due to partial needle fall (Lehtikangas, 2001).

The comparison of ash contents from fresh woodchips and pellets shows (Figure 5-10) higher values for the latter product. This is mainly caused by the fermentation that has occurred during the seasoning of wet woodchips that caused a loss in organic matter. Other sources of ashes could be due to the processing stages that can bring some metals into the final product (milling - pelletizing) incrementing the unburnt residues.

Finally the statistical evaluation of ash contents in the wood pellets produced is shown in Figure 5-11. Increasing values of ash contents are recorded in this sequence: AbDe – AbSt – FaSt – AbTr – FaBr and all the differences are statistical significant. This is in line with the consideration done before, given that increasing amounts of bark and branches occur in higher deposits after the incineration process. From this perspective only AbDe, AbSt and FaSt have ash contents below the maximum limit for the A quality classification. Any other addition of contaminants (bark, branches and leaves) resulted in higher ash contents that fail the high quality classification.

The comparison of calorific values is presented in Figure 5-12. To better appreciate the difference among calorific values of the different types of wood it is necessary to transform the value given by the calorimeter into high calorific value ash-free. In other words this allow to deny the effect of ashes in the weight of the tested sample referring all the energy produced to the real organic matter being burnt. In fact, the high ash content lowers the calorific value calculated by the bomb calorimeter, because, in the same weight of sample it is collected a lower burnable substance amount. HHV<sub>af</sub> is then a theoretical value of energy content that can be retrieved from calculation but never directly from the calorimeter or in a burning appliance. To maintain this differentiation in Figure 5-12 a dot identifies the actual ash content of each material already explained. The statistical evaluation of HHV<sub>af</sub> data

reveals that: i) AbDe and AbSt have similar calorific value, but the internal distribution of values is wider for AbSt due to the higher heterogeneity of the milled product containing a certain amount of bark, ii) conifers have higher HHV<sub>af</sub> values than broadleaves due to higher content in resins and oils, iii) both in conifers and broadleaves HHV<sub>af</sub> is higher for materials containing higher percentages of bark and needles since these pure materials are more energetic than pure wood, iv) the highest HHV<sub>af</sub> is produced by AbTr due to the high energetic mixture of materials contained. It should be remarked that these values do not consider the ash content. In fact giving an eye to the ash content in the graph it is evident that HHV<sub>af</sub> and A<sub>d</sub> have the same trend, resulting in pretty similar values of HHV<sub>0</sub>.

In Figure 5-13 the energy density values for all the produced pellets are reported. Given the average value of 13.4 GJ m<sup>-3</sup> for commercial pellets, the values for debarked spruce pellets are much higher, thanks to the high calorific value and high bulk density. Follow the pellets produced with whole stem and spruce whole tree, both above the market average. Pellets from beech have lower calorific values and bulk densities.

In Figure 5-14 and Figure 5-15 data about ash content and calorific value of other tested materials are presented. It can be noticed that there is a constant trend in increasing ash values following the order: *wood – branches – needles – bark*, while for heating values it is *wood – bark – branches – needles*. This is caused by the different mineral content, especially in bark and needles that affects directly the ash content while the oil and resin content, especially in twigs and needles, affects directly the heating value.

For all the three species bark is the material providing the greater contribution of ash compared to other parts of the plant: respectively 7.08% spruce, 1.60% larch, 8.89% beech. When considering the whole stem, however, this value is significantly lower since the percentage of bark is low and its effect mitigated by wood. Also the needles of spruce contain a significant amount of ash: 3.90%.

The high ash content leads to a decrease, in some cases significant, (for example in bark of spruce and beech) of the calorific value compared to what potentially achievable without considering the contribution of ash (gross calorific value, ash-free).

A study pointed out that the HHV<sub>af</sub> of spruce is, in order, *wood < needles < bark* (Ivask, 1999); while in the case of this study the sequence is *wood < bark < needles*. This diversity can be produced by different causes, whose main finding is uncertain; there may be

different content of lignin in the various species, or more generally by the different site conditions of growth, or also by the particular circumstances of storage and seasoning.

In any case, not only for spruce, but also for beech and larch, pure wood has the lower ash content than the other organs of the plant. In this sense, larch is in first place, followed by spruce. The wood of the latter has the highest energy content between the three species considered (HHV<sub>af</sub> 20.15 MJ · kg<sup>-1</sup>), then beech that also in this case has the least favorable value (18.77 MJ · kg<sup>-1</sup>) and larch in an intermediate position (19.56 MJ · kg<sup>-1</sup>).

These results underline the necessity to investigate and prepare the feedstock considering the contribution of each material. Already Lehtikangas (2001) has come to a similar conclusion with respect to pellets made from bark and waste products of two typical mixed forest trees in Sweden, spruce and scot pine.

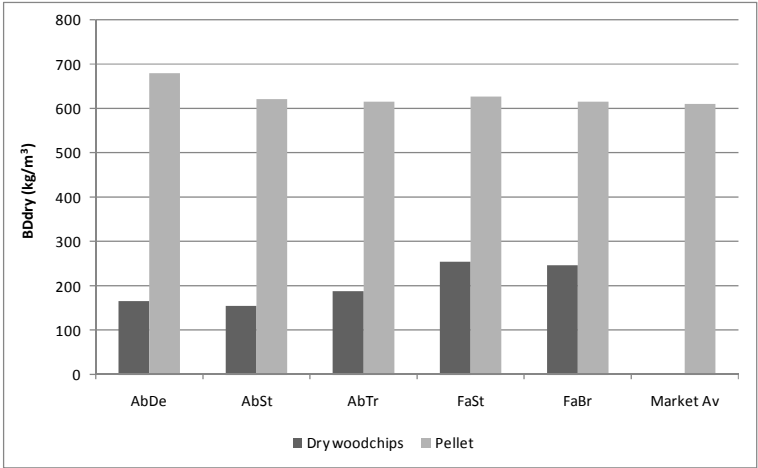


Figure 5-6: comparison between bulk densities

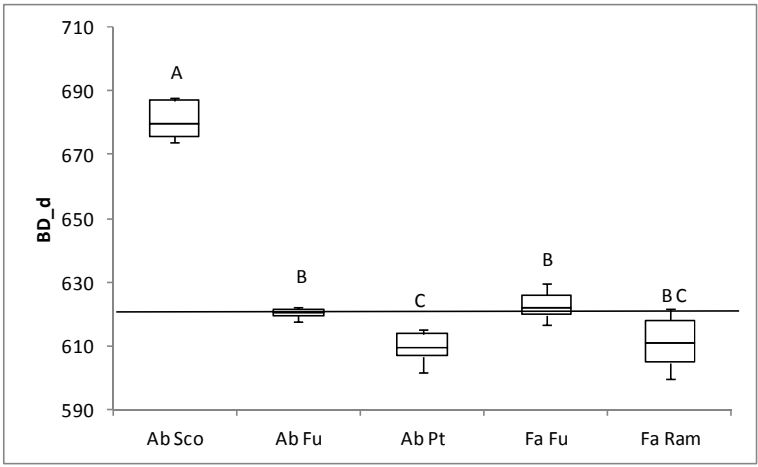


Figure 5-7: bulk density of produced pellets. The horizontal line represents the market average

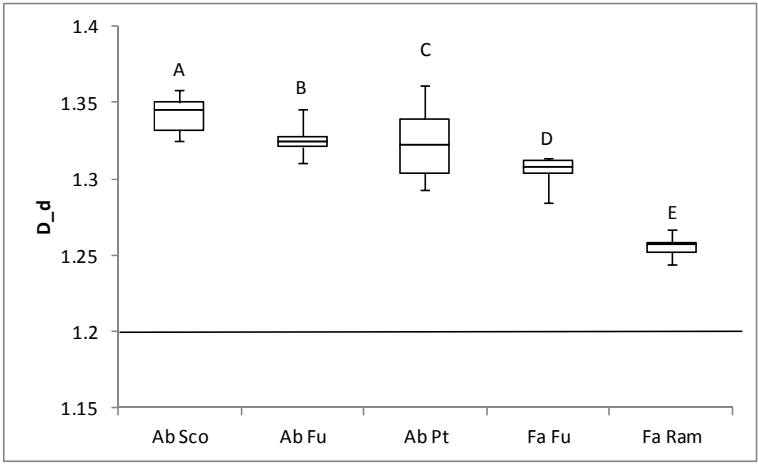


Figure 5-8: particle density of produced pellets. The horizontal line represents the market average

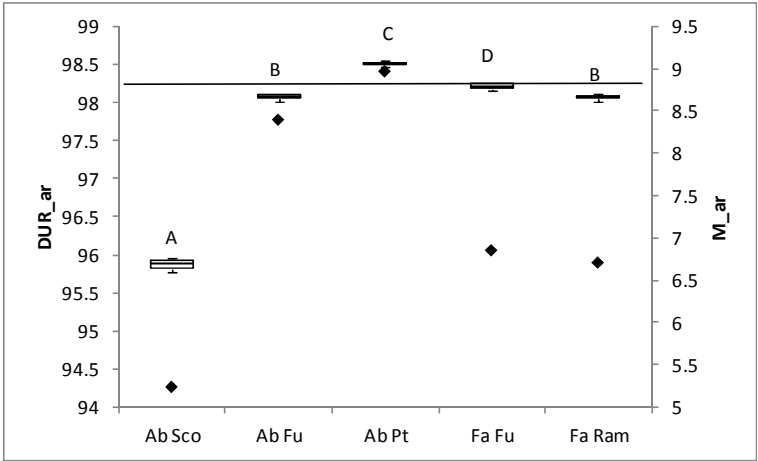


Figure 5-9: mechanical durability of produced pellets. The horizontal line represents the market average. Black dots represent the moisture content

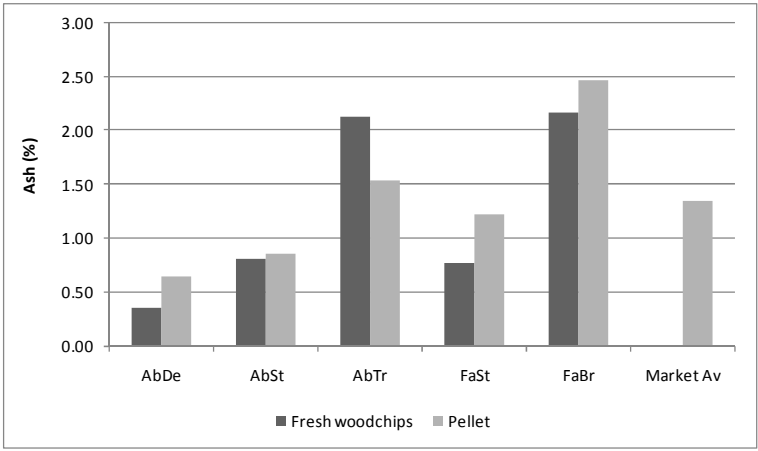


Figure 5-10: comparison between ash contents

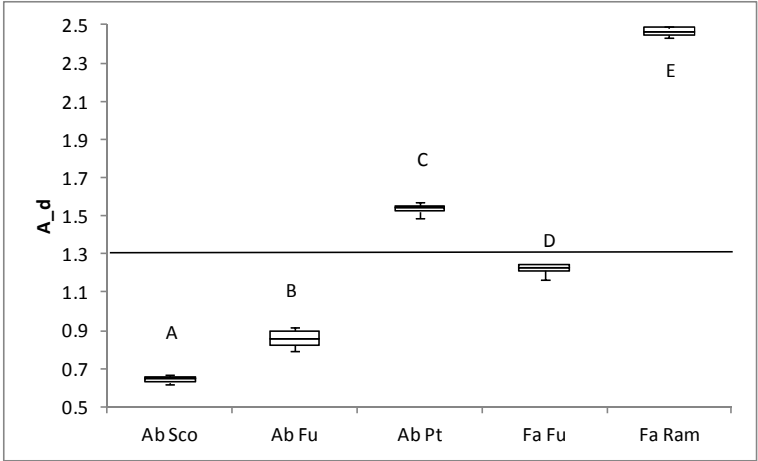


Figure 5-11: ash content of produced pellets. The horizontal line represents the market average

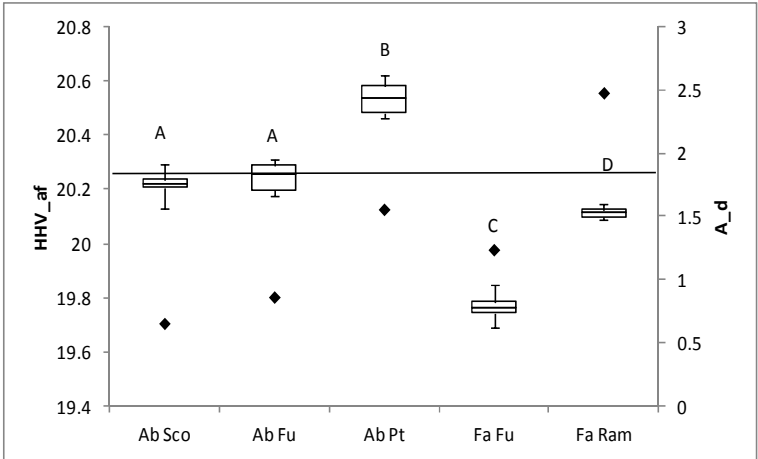


Figure 5-12: high calorific value ash free of produced pellets. The horizontal line represents the market average. Black dots represent the ash content

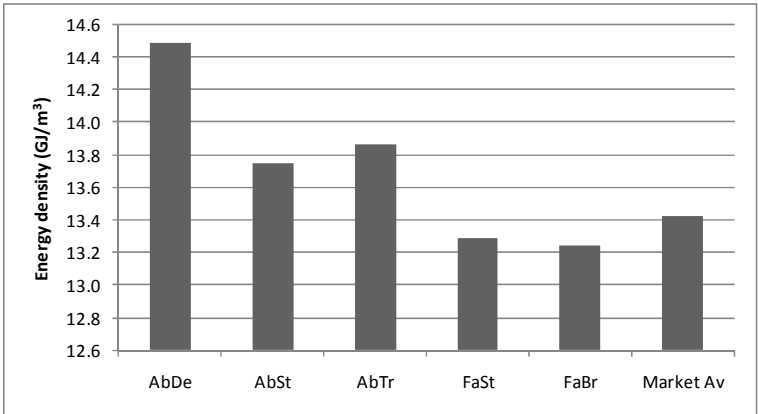


Figure 5-13: energy densities of produced pellets

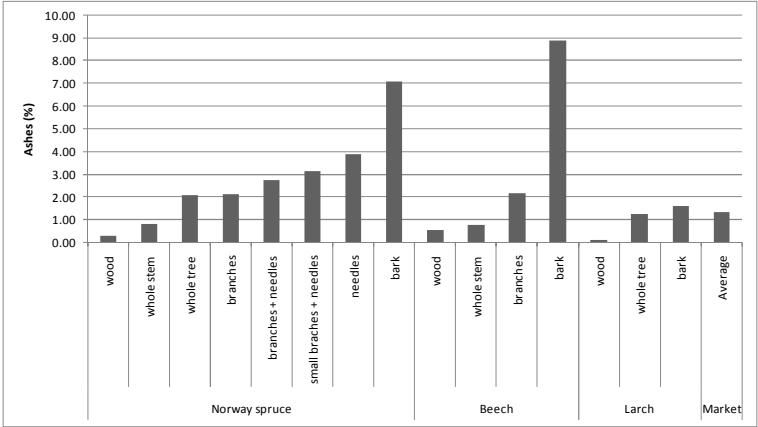


Figure 5-14: ash contents in different materials

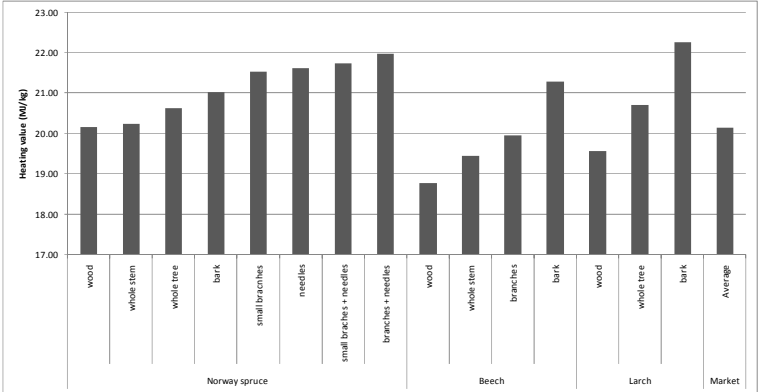


Figure 5-15: high heating value ash free in different materials

The chemical analysis of produced pellets is reported in the table below. It was conducted for all the metals and elements requested by the EN 14961-2 (wood pellets for non-industrial use). Results showed that almost all the parameters respected the maximum values, confirming that chemicals are not a limit in the production of high quality pellets. A specific remark should be done for the Nitrogen content. The **bold** and underlined values in the table reveal that this element is fairly present in AbTr and FaBr, while it does not represent a problem for all the other material. This is caused by the presence in needles, leaves and buds of significant amount of protein used by the tree for its physiological functions. Since the N limit for A1 pellets is 0.3% and for A2 pellets is 0.5% no issues are related with the combustion of this AbTr and FaBr in domestic appliances.

	<b>As</b>	<b>Cd</b>	<b>Cr</b>	<b>Cu</b>	<b>Hg</b>	<b>Ni</b>	<b>Pb</b>	<b>Zn</b>	<b>S</b>	<b>Cl</b>	<b>N</b>
	ppm ss	ppm ss	ppm ss	ppm ss	ppm ss	ppm ss	ppm ss	ppm ss	%	%	%
Ab De	0.23	0.15	5.47	1.15	<0.001	0.23	0.31	16.32	0.005	0.020	0.223
Ab St	0.16	0.24	0.32	1.21	<0.001	0.24	0.24	20.80	0.007	0.009	0.227
Ab Tr	0.39	0.23	0.31	1.87	<0.001	0.47	0.55	28.72	0.013	0.017	<b><u>0.328</u></b>
Fa St	0.24	0.08	0.64	1.53	<0.001	0.56	0.64	7.56	0.010	0.008	0.272
Fa Br	0.24	0.08	0.40	5.87	<0.001	0.64	0.64	10.70	0.019	0.012	<b><u>0.398</u></b>

## 5.5 Conclusions

All five types of pellet produced are good or acceptable: the best is the one made from debarked spruce stem wood, classified A1 according to the UNI EN 14961-2:2011 except for the value of durability. Nevertheless this parameter can be easily improved with the addition of some natural binders. In the class A2 falls the pellet made of whole stem, which is the second in terms of ash content and calorific value, and the pellet obtained from the whole spruce tree and beech stem. Finally, the pellets produced with branch wood beech falls into quality class B.

It can be concluded that, according to the analysis carried out, all the produced pellets can be destined for the domestic market. Some suitable blends and mixtures can be prepared to improve the quality of the pellets in the lower categories through the control of the ash content.

If the qualitative requirements laid down by the EN are fully respected, there may be a real opportunity to provide a higher added value and a renewed interest to this residual feedstock, enhancing them as a source of raw materials in the production chain of the pellets. Positive results may be found in further studies on the economic sustainability of the production chain of pellets from residues and forestry assortments of poor and very poor quality. This would give an added value, and thus it would return a productive function to marginal forests and abandoned stands, also considering the increasing need to exploit new renewable resources in sustainable way.

## 6 Color of pellets

### 6.1 Introduction

The color of pellets made from pure wood sawdust, processing residues or debarked stems and shavings have generally lighter colors. The absence of bark results in lower ash contents compared with wood (Filbakk et al., 2011; Lehtikangas, 2001) but, on the other hand, in low mechanical properties, especially durability (Lestander et al., 2012a), that detects the proneness of pellet to disintegrate when exposed to mechanical stresses. This can result in higher fine particles content that affects pellets quality during transportation, storing, stove feeding and final burning (Filbakk et al., 2011). Pellets produced from whole stems, including bark or mineral contaminants, are instead darker in color and usually their ash content is higher (Lam et al., 2012), lowering the content of energetic matter in the biofuel and the quality of the combustion behavior, to the point that pure bark pellets can be burnt only in large heating plants (Filbakk et al., 2011).

The exposure of raw material to high temperatures during drying changes both its chemical properties and external appearance affecting also the resulting pellet color (Lam et al., 2011; Nuopponen et al., 2004). Heat exposure, in fact, modifies the wood structure and color in dependence of temperature and presence or absence of oxygen (Shen et al., 2012).

The moisture in the feedstock as bound water plays a key role in the formation of interparticles linkages. Moist feedstock results in weaker pellets, to the point that sawdust is no longer able to remain compacted after exiting the die channel. On the other hand, the moisture content, acting as lubricant, can affect positively the resulting pelletizing pressure and temperature (Larsson et al., 2013), again influencing the final color result (Lam et al., 2012). The optimal ranges for water content values are found to be 8-13% (Filbakk et al., 2011; Nielsen et al., 2009).

This variability in pellets quality parameters has stimulated many countries to develop specific standards and regulations in the attempt to set some baselines for production, storage and distribution of wood pellets (García-Maraver et al., 2011). The national standardization process has resulted, at the end of the year 2012, to the publication of some harmonized EN (European Norms) regulating pellets quality parameters and classes.

The experience developed in the study of the wood pellet sector for residential heating has shown that a strong emphasis is put on the color of pellets, starting from the consideration that color could be a direct indicator of product quality. The market is very reactive to this parameter, and this is verified considering that dark color pellets have a lower commercial value than light wood pellets (Lam et al., 2012) (the so called *white pellets*), especially for residential applications. This is mainly associated with the contamination of raw materials with impurities such as bark, which high ash content is reflected in the final pellet quality.

Even if color is not a parameter for grading pellets quality as defined by the European and International Norms (EN - ISO), the market trend and the consumer choice are strongly affected by this parameter that is the only one directly verifiable at first sight (Hellrigl, 2004; Kofman, 2007). On the other side, this consideration is not supported by scientific studies and a lack of knowledge regarding color and quality applied to pellet properties is registered. Very little literature is available (mainly on change in color of feedstock that underwent thermal drying) and no clear relationships among pellet characteristics and its color have been proved.

Each color depends on the type of emission source that irradiates an object, as well as on the physical properties of the object itself (which reflects the radiation consequently detected by the sensor), and finally on the in-between medium (e.g., air or water) (Duntley, 1952). Generally, the color spaces applied in product classification are the standard RGB (sRGB; red, green, blue) and  $L^*a^*b^*$ . sRGB can be obtained rapidly using computer vision systems. Outputs signals are generated by the camera sensors (e.g., CCD or CMOS), but the rendering is device dependent, since the range of colors varies according to the display device specifications (Cubero et al., 2011). In order to overcome this problem, sRGB values are often transformed to other color spaces such  $L^*a^*b^*$ . Moreover, even the result of such transformation is device dependent (Ford and Roberts, 1998). Another technique is the Near-infrared (NIR) spectroscopy that has been already tested to study the species composition in wood pellets (Arshadi et al., 2008) or to detect the presence of specific chemicals (Lestander and Samuelsson, 2010). The application of this technology is already well known and broadly applied to online estimation of moisture content from woody materials (Lestander et al., 2009; Nyström and Dahlquist, 2004).

The current work investigated the color characteristics of some commercial and control pellets with the aim to detect if quality parameters are effectively related with colorimetric values and image patterns and finally clarify the possibility to base the consumer biofuel choice on visual evaluation.

The study was set up in collaboration with CRA (Consiglio per la Ricerca e la Sperimentazione in Agricoltura) to verify and prove if any relationship between color and pellet quality parameters can be found.

## 6.2 Materials and methods

### 6.2.1 Color analysis

All the sampling and sample preparation are the same as exposed in Chapter 4.2.

Color measurement was performed with two different kinds of approaches. The first method, applied on whole pellets samples and GAS, is based on CIE (*Commission Internationale de l'Eclairage*) L\* (lightness) a\* [along the X axis red (+) to green (-)] b\* [along the Y axis yellow (+) to blue (-)] color space (Zanetti et al., 2003), the second is based on RGB and HSV color spaces.

CIE L\*a\*b\* colorimetric information were acquired using an imaging spectrometer (Spectral Scanner, v. 1.4.1, DV Optics, Padova, Italy) which digitized images (Figure 6-1) ranging from 400 to 790 nm (step 5 nm) following details and settings exposed by (Menesatti et al., 2010). Each frame contained the line pixels in one dimension (spatial axis) and the spectral pixels in the other dimension (spectral axis), providing full spectral information for each pixel line. The reconstruction of the entire hyperspectral image of the sample was performed by scanning the sample line by line as the transportation plate moved it through the field of view of the sensor. The resolution of the line image was 700 pixels per line for 500 lines. The system was operated in a dark laboratory to minimize interference from ambient light, and the system was calibrated using black and white references. The spectral scanner acquired images of single pellets and GAS. Mean CIE L\*a\*b\* values (3 values) of 5 region of interest (ROI) were used for each pellet sample.

RGB and HSV color information of single pellets were acquired through a professional high resolution scanner (600 d.p.i.) (Figure 6-2). Images of more than 100 pellets for each run were acquired together with a GretagMachbeth ColorChecker 24 color patches standard. In

order to standardize the RGB color, images were calibrated using the Thin Plate Spline interpolation algorithm (TPS 3D) (Menesatti et al., 2012). Inside the calibrated images each pellet were outlined from the dark background automatically. Image binarization was performed by means of fixed threshold value applied on each RGB channel respectively (R=25, G=25, B=25). Mean RGB and HSV values of all the pellets belonging to the same sample were used in the further statistical analysis.

Differences in terms of CIE L\*a\*b\* values between pellets and GAS were estimated using the  $\Delta E$  calculation respectively to observe whether same color results can be obtained from different sample form (values lower than 3). The  $\Delta E$ , and further on the  $\Delta RGB$ , represent the Euclidean distance between two colorimetric patches in the color space, CIE L\*a\*b or RGB respectively.

$$\Delta E = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2} \quad (1)$$

$$\Delta RGB = [(\Delta R^*)^2 + (\Delta G^*)^2 + (\Delta B^*)^2]^{1/2} \quad (2)$$

$\Delta E$  and  $\Delta RGB$  were calculated for samples of whole pellets and GAS having respectively high and low ash content values, to define the possibility for the human eye to detect some differences among different pellets types and qualities based on colorimetric characteristics (Buchelt and Wagenführ, 2012; Pallottino et al., 2013).

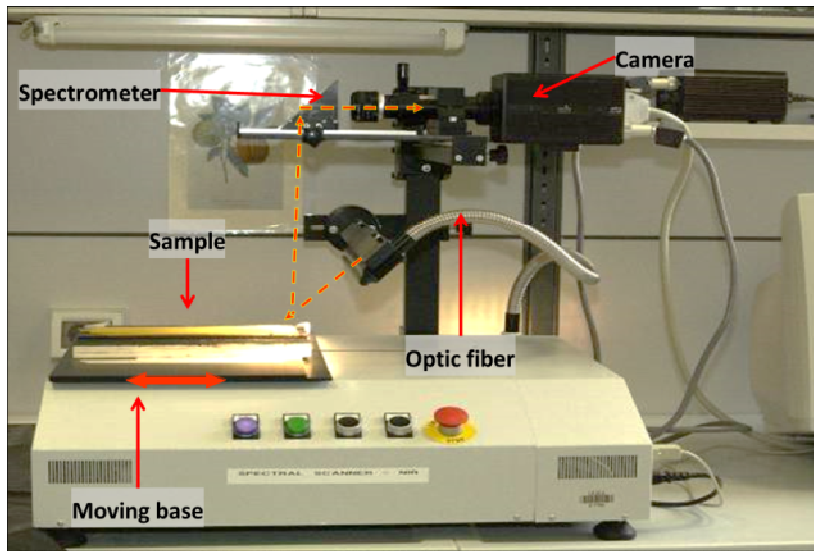


Figure 6-1: VIS-NIR spectral scanner (left) and the taken image (right)



Figure 6-2: RGB scanner (left) and the taken image (right)

## 6.2.2 Statistical evaluation

Canonical correlation analysis (CCA) was used to observe the degree of correlation of ash with CIE L\*a\*b, RGB and HSV separately. CCA is a way of measuring the linear relationship between two multidimensional variables. It finds two bases, one for each variable, that are optimal with respect to correlations and, at the same time, it finds the corresponding correlations. Canonical correlations are invariant with respect to affine transformations of the variables. This is the most important difference between CCA and ordinary correlation analysis which highly depends on the basis in which the variables are described (Sherry and Henson, 2005). Canonical correlation coefficients representing the degree of correlation between ash and the color coordinates were reported together with their significance ( $\chi^2$  test). To investigate the single relationship between each colorimetric variable and ash content, the Pearson linear correlation coefficient was reported.

In this study the Principal Component Analysis (PCA) was performed to verify the influence of quality parameters on the overall sample classification and to relate those with sample color (RGB). The PCA is a projection method which allows the display of information in a data matrix considering the influence of each one in a limited number of components expressed as linear function of the original ones. The data matrix, consisting of the quality pellet parameters for all the variables, contains a large amount of information partially hidden because the differences among samples are complex and not of easy representation. Two principal components (PC) were selected. The analysis was done considering the loading of six parameters: D, A, M, DU, HHV and BD on each PC. The results allow to classify the samples according with their scores. Similar scores along a component detect similarities for variables with a high loading for that component.

### 6.3 Results

To observe if differences in terms of CIE L\*a\*b\* between whole pellets and GAS were perceptible by human eye, mean  $\Delta E$  were calculated. Human eye perceives clearly color differences for  $\Delta E$  values greater than 3 (Dirckx et al., 1992). They resulted to be  $8.0 \pm 3.9$ . This result proved that these slight differences are perceptible by human eye as they resulted to be significantly higher than 3 (one sample t test,  $p < 0.0001$ ).

In Table 6-1 the summary of the CCA results is reported. It is possible to observe good correlations between ash and CIE L\*a\*b\* (pellet and GAS), RGB and HSV values. In particular GAS L\*a\*b\* values are highly correlated with ash (0.78). The single colorimetric components of the color space are all negatively and highly correlated with ASH a part Hue (highly positively correlated) and Saturation (low negatively correlated) of the HSV space and a\* (low positively correlated) of the CIE L\*a\*b\* space.

Table 6-1: summary of the results of the Canonical Correlation Analysis (CCA)

	Canonical correlation coefficient	CCA coefficient significance	Pearson L*/R/H vs ASH	Pearson a*/G/S vs ASH	Pearson b*/B/V vs ASH
Pellet - CIE L*a*b	0.70	<0.0001	-0.67	-0.19	-0.61
GAS - CIE L*a*b	0.78	<0.0001	-0.73	0.09	-0.64
Pellet - RGB	0.70	<0.0001	-0.69	-0.67	-0.63
Pellet - HSV	0.72	<0.0001	0.53	-0.20	-0.69

Figure 6-3 reports the colorimetric representations of CIE L\*a\*b\* on GAS and on pellets and RGB mean values (only on pellets) for the 4 samples having respectively highest (> 3.85%) and lowest (< 0.36%) ash content percentages values.  $\Delta E$  and  $\Delta RGB$  values between these two group mean values are respectively 30.3, 36.0 and 104.6. These high delta values are perceptible by human eye since they are higher than the reference values 3 ( $\Delta E$ ) and 6 ( $\Delta RGB$ ) respectively. It demonstrates that high ash concentrations are likely to be related with darker colors regardless it is measured on GAS or whole pellets sample.







	CIE L*a*b* GAS	CIE L*a*b* pellet	RGB pellet
High ash content			
Low ash content			
	$\Delta E$	$\Delta E$	$\Delta RGB$
	30.3	36.0	104.6

Figure 6-3: colorimetric patches representing the CIE L\*a\*b\* (GAS and pellet) and RGB mean values of the 4 pellet samples having respectively higher (> 3.85) and lower (< 0.36) ash values

Figure 6-4 represents the output of the PCA. The position of each variable defines its score along each component. The total variance explained by the model is 64,2% where 43,8% is explained by the first PC, while only 20,4 is explained by the second PC.

For each sample a dot locates its position in the graph, while the color of the dot itself represents the average RGB value for that sample at its initial moisture condition (referred as received) measured during the image analysis.

Along the first PC axis the variables A, D and BD give a negative contribution (*i.e.* the variables decrease their values when the PC1 scores increase). On the contrary DU and HHV variables give a higher and positive contribution to the samples ordination (and also along the second one), while, M is less strong. Along the second PC axis the major contributors are M (negative), BD and D (positive). Visually it can be noticed that the majority of the darkest samples are located in the third quadrant, where A values have higher load, while the

lightest ones are mainly aggregated in the first quadrant where HHV and DU have higher loads. In the second and fourth quadrants the samples with intermediate colors are located. In these regions the loadings and significance of the representative variables (M, D and BD) are lower and no clear relationships can be detected.

The presence of some light brown samples in the second quadrant is mainly driven by the influence of the moisture content that makes colors darker. This factor is averted for the other parameters as they are referred on dry basis.

It can be noticed in Figure 6-4 that a dense cluster of certified samples (squares) is aggregated in the first quadrant, with few cases of samples trespassing to the second and fourth. Only one sample is located further from the cluster on the fourth quadrant even if it was declared as certified. This is due to its abnormal values of M (very low) and A (very high). A second group of samples, mainly clustered around the origin of the axis, is represented by the five reference pellets types (AbDe, AbSt, AbTr, FaSt, FaBr). Both the beech samples are located in the negative sector of the first PC due to their relatively high ash content and low values of HHV, while, on the contrary, AbSt and AbTr are located in the opposite side. The outer position of AbDe, close to the load value of D and BD along the first PC, is due to its high D and BD, while it has the lowest value of HHV among spruce samples and the lowest DU value among all, confirming that pure wood pellets often results in low mechanical and energetic properties.

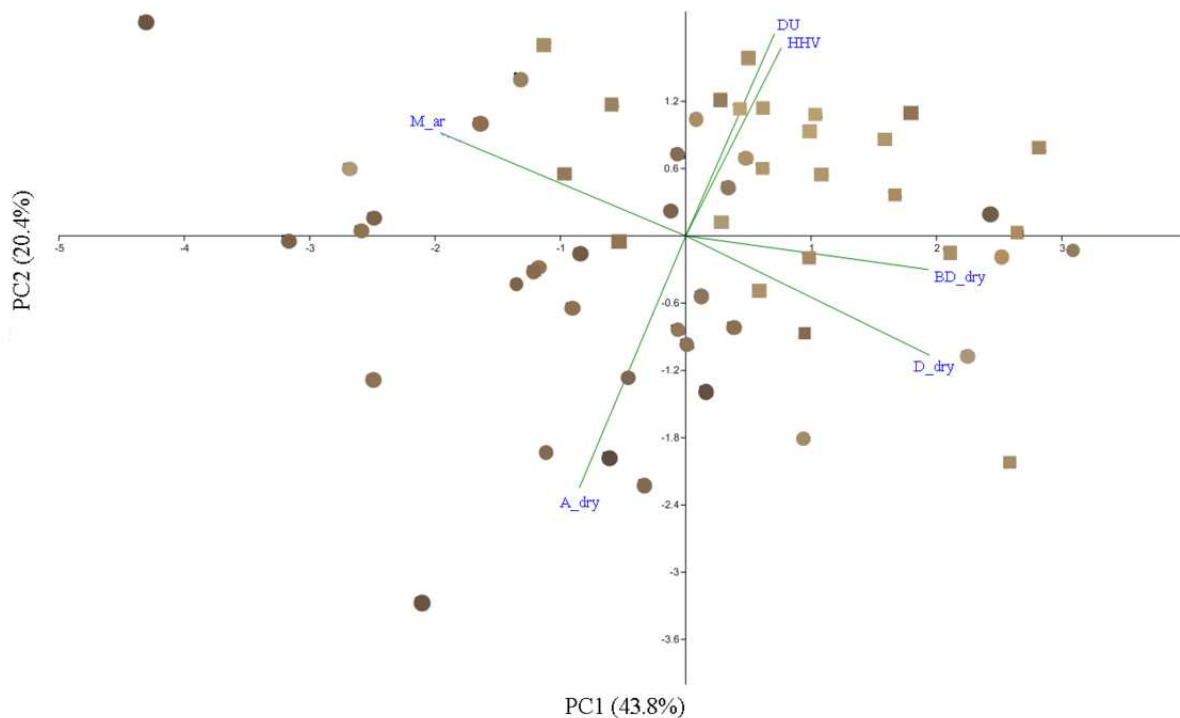


Figure 6-4: graphical layout of the PCA results. Dot color represents the average RGB for that sample. The position of each variable detects its loading on each PC. Squares represent certified samples

## 6.4 Discussion

The explanation why the final appearance of some pellets is darker than some others could not be found only in their constituent formula. It has been said how the outer color of pellets is affected by a number of factors such as moisture content, feedstock treatment and production parameters.

The color analysis revealed a perceptible difference between the CIE  $L^*a^*b^*$  of pellets (the outer surface) and GAS (ground pellets) that is proven to be greater than 3. The reasons for that are twofold. The cylindrical form of pellets caused the presence in the image taken with VIS-NIR of areas of reflection and areas of shadow because their convex surfaces did not reflect the light evenly when illuminated at  $45^\circ$  (device dependent). This effect is emphasized by the lucid pattern of the outer pellet layer due to the glass transition (softening point) of lignin and hemicelluloses at the pelletizing temperatures that enhances the bonding effect of wood particles (Lehtikangas, 2001). Secondly thermal shock to which wood is subjected during the production makes it generally darker in color with lower brightening and values of  $L^*$  (Chen et al., 2012) and different hygroscopicity (Lam et al., 2011). The contact with the hot die at high friction forces during extrusion causes the

external layer darkening to be enhanced than the inside. This is confirmed by (Lam et al., 2012) who have found that the  $L^*$  values of the ground raw material compared with the pellets made out from the same biomass are higher and result in lighter colors. In the same work, to avoid the affection of the color response by the moisture content, all the samples were oven dried. As the aim of this study is to prove the relation between visual appearances of pellets with their quality, the water content of each sample was determined and included (not discarded) from the analysis.

Significant correlations (CCA) were observed between color and ash content for CIE  $L^*a^*b^*$  (GAS and pellet) and RGB-HSV (pellet). The direction of the correlation has showed that higher values of ash are mostly associated with darker colors. These differences, detected by the device, are visually perceptible ( $\Delta E - \Delta RGB$ ) when considering the color of samples at the extremes of ash contents distribution (highest and lowest) while comparisons between samples with similar colors is less appropriate. For this reason the capability of the consumer to define the quality of pellets only referring to their external color still has a wide degree of uncertainty.

The PCA underlined the influence of the measured parameters, in addition to the ash content, in the quality grading of pellets. Good pellets are those able to undergo mechanical stresses and are converted in high energy when burnt. To achieve this, low moisture contents, high densities and high heating values are required. Complementarily the content in minerals, resulting in ashes, is to be avoided as it reduces the energy content of pellets and can affect negatively the combustion (Casal et al., 2010; Öhman et al., 2004; Toscano et al., 2013). The distribution pattern of all the samples has confirmed a strong relation of RGB mean values with their overall quality and has also outlined that pellets declared as certified by the producers have generally good quality and lighter colors. This is in good agreement with what it was expected from the previous analysis.

Systems based on the spectral measure of the color ( $L^*a^*b^*$ ) are expensive, complex and require high utilization standard, as their sensitivity to the environment conditions (mainly lighting) is high. Furthermore they do not give any appreciable advantage to the estimation of the quality parameters if compared with the other systems studied. The device used for the study, which is an imaging spectrometer, provides a better appreciation of the color of

individual pellet net of shadows and steric geometric characteristics of pellets compared to the standard colorimeters with integration of area.

On the other hand the RGB imaging system, once properly calibrated (Menesatti et al., 2012) is proven to be a convenient alternative when applied for color analysis. These systems are not only cheaper and more efficient than the previous ones, but they also can be easily implemented directly on production processing lines.

Spectroscopy and color measurement has been used in a large variety of applied sciences and their utilization is well developed and robust. On the other hand the biomass sector is seeking for new application for detecting quality parameters online and continuously. The advantages connected with the fast estimation of moisture content, ash content and calorific value are mostly not compensated by the high initial investment of purchasing all the devices for internal analysis set up. Instruments for the qualification of product quality directly online are nowadays easily available and implementable on the production process (Lam et al., 2012).

## 6.5 Conclusions

The relationship between colorimetric characteristics of wood pellets for energy purposes and their quality parameters was investigated. High degrees of correlation were found between CIE L\*a\*b, RGB and HSV separately and values of ash contents, which has been considered to be the most appropriate indicator as it is the biggest concern among final consumers in domestic appliances. The direction of the correlation was proved to be constant and darker pellets are more prone to have higher ash content. The same results were obtained analyzing the pellets as they are or grinding them to a nominal top size of 1mm (GAS). Moreover the mean  $\Delta E$  between pellets and the respective GAS were detected to be appreciable by human eyes as they were all higher than 3. This is due to the modification that wood particles and fiber constituent (lining and cellulose) undergo during the pelletizing phase and the different moisture condition of the inside of the pellet compared with the outer layer.  $\Delta E$  and  $\Delta RGB$  calculated between the 4 samples with highest and lowest ash content showed results fairly high, confirming that the direction of change of ash and colors is the same, and visually appreciable. This is not confirmed for sample with color similar to each other, where the predictability of their quality is less certain. Samples

have shown a clear distribution along the two PC considering mechanical (DU), physical (A, M, BD, D) and energetic (HHV) variables. The same general pattern was verified for pellets sold as certified products that are supposed to have higher quality properties. Sample lighter in color mainly clustered in the space where HHV, DU and densities have higher scores. On the other hand darker samples were dispersing in the opposite space dominated by higher ash and moisture contents.

The application of such methodology directly to the industry seems to be possible considering some developing factors:

- Vis NIR spectroscopy, which is fairly expensive, need specific work environment and can be highly affected by the lighting conditions and the stereometrics of the objects analyzed;
- RGB imaging systems are cheaper and lead to reliable results similar to the previous ones. Appropriate color calibration has to be applied to avoid errors.

## 7 LCA on pellet supply chain

### 7.1 Introduction

The development of alternative energy and fuel supply chains has been chosen as a key action for the European Union to produce and enhance a cleaner and less pollutant economy.

The global demand for energy from renewable sources has continued to rise, and the attest data available for 2011 and 2012 published by the EU report gives a estimated supplying of global final energy consumption of about 19% (Mcginn et al., 2013).

Biomass feedstock is though one of the most important actors of this actions since their share is growing among the overall renewable energy production sector.

Half of this energy share is from traditional biomasses (Figure 7-1), which utilization is now under discussion if to be considered renewable or not, since burning biomasses in low technology appliance (i.e. domestic stoves or open burners) can result in high pollutants emission. A recent study has applied the LCA methodology in the comparison of traditional wood burners and innovative pellet ones to highlight if some differences in efficiency and pollution potential can be detected. Results showed that the LCA is a good approach to investigate such field and gives consistent results helping development choices. Moreover it is evident how the installation of the latest generation pellet stoves, integrated with continuous combustion regulation through gas emissions control units, can results in a improvements of the air quality. A different scenario is resulted when considering obsolete wood stoves (Cespi et al., 2014).

On the other hand in some regions the available feedstock are not sufficient to fulfill growing demand for bioenergy, whereas other regions can produce supplies in excess. The international pattern of the energy market has led to a substantial need in global transportation of energy feedstock, increasing questions on the sustainability of such markets. The wood pellet production and transportation from British Columbia in Canada to Northern Europe showed that up to 40% of the energy content of pellets are invested in the production and transportation processes, leaving only 60% of the energy content available for energy production (Magelli et al., 2009).

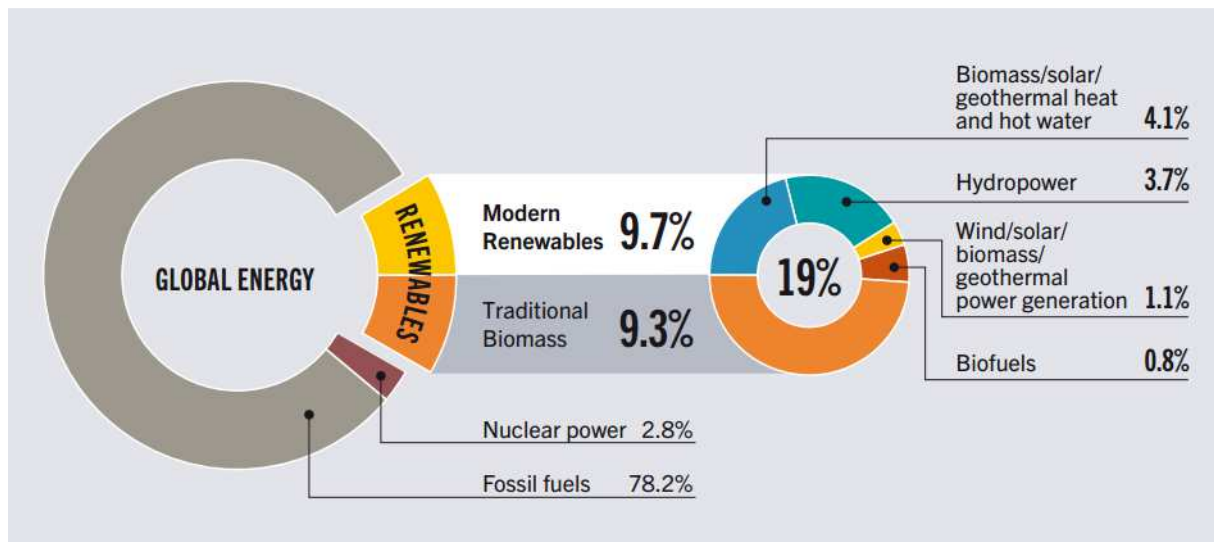


Figure 7-1: total energy share in Europe (McGinn et al., 2013)

The overall emissions of greenhouse gases for the production of one unit of electricity in a big power plant run on wood pellets imported from the USA to the UK has been calculated to be at least 50% lower than the same unit derived from fossil fuels power production (Ghilardi et al., 2014). These results give evidences of how intensive the investment of energy could be in moving materials with low energy density worldwide, but on the other hand it is clear that the energetic balance could justify a so wide market.

Wood pellets have experienced more rapid growth and account for a large share of solid biomass trade: in 2012, global production of pellets exceeded 22 million tons. Wood pellets account for only 1–2% of total global solid biomass demand and about two-thirds of pellet production is used in small heat plants and one-third in larger power plants (McGinn et al., 2013).

The reasons why the interest in pellets is so high have to be found in their physical and mechanical properties, since more energy is compacted and stored in the same amount of volume, if compared with other traditional woody fuels (Dhamodaran and Afzal, 2012). Moreover the regular shape and the small dimension allow to handle, store and use them in automatic appliances.

The European Union is still the most important market for wood pellets due to the high prices of other energy sources and the level of technology required to exploit them. In the period 2009-2012 the production of pellets in EU has increased by 30%, reaching 9.2 million tons in 2010, equal to 61% of the global production. In the same period, EU wood pellet consumption increased by 43.5% up to 11.4 million tons consumed in 2010 (AEBIOM, 2012).

Moreover biomaterial for energy proposes could be produced or exploited almost worldwide by planning and adapting forest operations and agricultural management properly. The availability of specific importation data is often poor but some estimations say that the total income of wood pellets from the USA will increase from 1.5 to 5.2 Mt between 2012 and 2015 (Ghilardi et al., 2014).

Finally the carbon footprint of biomass resources can be considered almost neutral (Cherubini et al., 2009; Helin et al., 2013; Lippke et al., 2011; McKechnie et al., 2010; McManus, 2010; Routa et al., 2012; Whittaker et al., 2011). In fact the total amount of carbon dioxide that has been adsorbed during the growing life of a plant is then re-emitted with combustion. However this consideration does not take into account the emissions correlated with fossil sources burnt during the whole life cycle of the product. To the neutral share of carbon dioxide it is necessary to add all the black energy inputs, mainly from fossils and non-renewable resources, involved for the supplying, producing and upgrading operations.

Although bioenergy is almost CO<sub>2</sub>-neutral, the time scale to be considered to fix the same amount of carbon over a long period of time and then released instantaneously when burnt has to be take at proper dimension (Raymer and Kristin, 2006). Another aspect is the dynamics of a managed forest. Harvesting wood for energy or products releases land for regeneration and further carbon fixation. These aspects can be studied by including substitution effects into a forest management model. Moreover many processes, from combustion to degradation, emits non only CO<sub>2</sub>, but also other gases that could not be offset by plant photosynthesis.

It has been proved that a consistent share of the energy content in wood pellets could have been used for its production and transportation (Mani et al., 2006). In this regard, the distance covered from the harvesting site of the raw material to the manufacturing site gives an important contribution in the overall carbon footprint per km travelled (Heinimann, 2012; Karjalainen and Asikanene, 1996). It is globally accepted and considered that transportation is usually the critical emitting phase of many supply chains representing a consistent source of gaseous carbon dioxide, the reference climate altering gas (IPCC, 2014). The need to find alternative solutions for raw materials supply gives reasons to invest on research in this field (Loo and Koppejan, 2008).

To better understand the sustainability of a wood pellet project it is necessary to assess the energy consumption and the related emissions and refer them to different framework conditions. For example it should be considered that there are still wide ranges of improvement to make the pellet supply more efficient for example reducing the energy consumption of the pelletizing process (Cespi et al., 2014).

Others studies on the LCA of wood pellets have been conducted in Canada (Magelli et al., 2009; Pa et al., 2012), Europe (Fantozzi and Buratti, 2010; Sjølie and Solberg, 2011; Valente et al., 2011), and the United States (Katers et al., 2012). Some other works have focused their attention on the economic aspects of producing pellets in different framework conditions (Mani et al., 2006; Thek and Obernberger, 2004; Wang et al., 2013).

The focus of each of these studies was different from the one we want to study here.

The main objective of this work is the definition and technical evaluation of alternative supply chain models based on the evaluation of the wood pellets sector energy consumption under different framework conditions based on a survey campaign of primary data in Sweden and Italy, and secondary data from international literature review.

On one hand it will focus on the traditional by-products supply chains based on residues and waste form the lumber processing industry. On the other hand new forms of supply chains directly from forests products will be assessed and the most sensitive elements influencing them will be highlighted. From previous studies it resulted that the GHG emissions from pellet production are generally similar if different types of feedstock are used for manufacturing (Ghilardi et al., 2014). This implies that the best option to reduce the environmental impact of pellet production and utilization could be the use of all the by-products coming from forest operation, when not referring these advantages to the final pellet quality.

The overall system will be finally compared with a reference scenario, where the wood resource is directly used, in form of woodchips, in a district heating plant (DHP). This last scenario is thought to give a comparison term and quantify the energy investment, the added value and the environmental impact of wood upgraded into pellets. Since these solutions could have different environmental impact their evaluation should be considered as a starting point for the study of a project.

## 7.2 Methods

Life cycle assessment is a methodology to assess environmental impacts related with all the stages and steps of goods' production or services provision (i.e., from raw material extraction, product processing, transportation and distribution, intermediate and final use, disposal or recycling). The objective of LCA is to compare all the measured effects on environment that derive from the origin and production of products and services by quantifying all inputs from and outputs to the environment. It considers not only material flows but also energy transformation and conversion to finally assess how these procedures impact on the environment. If properly done the information retrievable from LCA can be used as valid support to improve industrial processes, help policy makers' activity and provide basis for decisions.

The LCA methodology could be used to evaluate also the environmental and energetic sustainability of biomass energy chains, and in particular their greenhouse gas emission factors. LCA methodology, as defined by the ISO 14040 (Environmental management - Life cycle assessment - Principles and framework) and ISO 14044 (Environmental management - Life cycle assessment - Requirements and guidelines) standards, proved to be a valuable tool for documenting and analyzing environmental considerations of product and service systems, that have to be part of decision-making process.

### 7.2.1 Study structure

The LCA on pellet production chains study comprises the following phases:

- the definition of goal and scope;
- the inventory analysis;
- the impact assessment;
- the interpretation of results.

The scope of the study is the environmental evaluation of alternative supply chain models for pellets production, which structures is summarized in Figure 7-2. The system boundaries (*solid* and *dotted* lines) determine which unit process shall be included within the LCA. The selection of the system boundary, which is consistent with the goal of the study, starts from the forest operations or harvesting of wood and ends at the energy production plant in a *cradle-to-grave* perspective. This means that the study originates from the resource

collecting and ends with the product consumption (i.e.: energy exploitation). The *solid* boundary defines the limits of processes regarding with pellet production and burning from different base materials, while *dotted* boundary refers to the supply of thermal energy from woodchips in a big district heating plant.

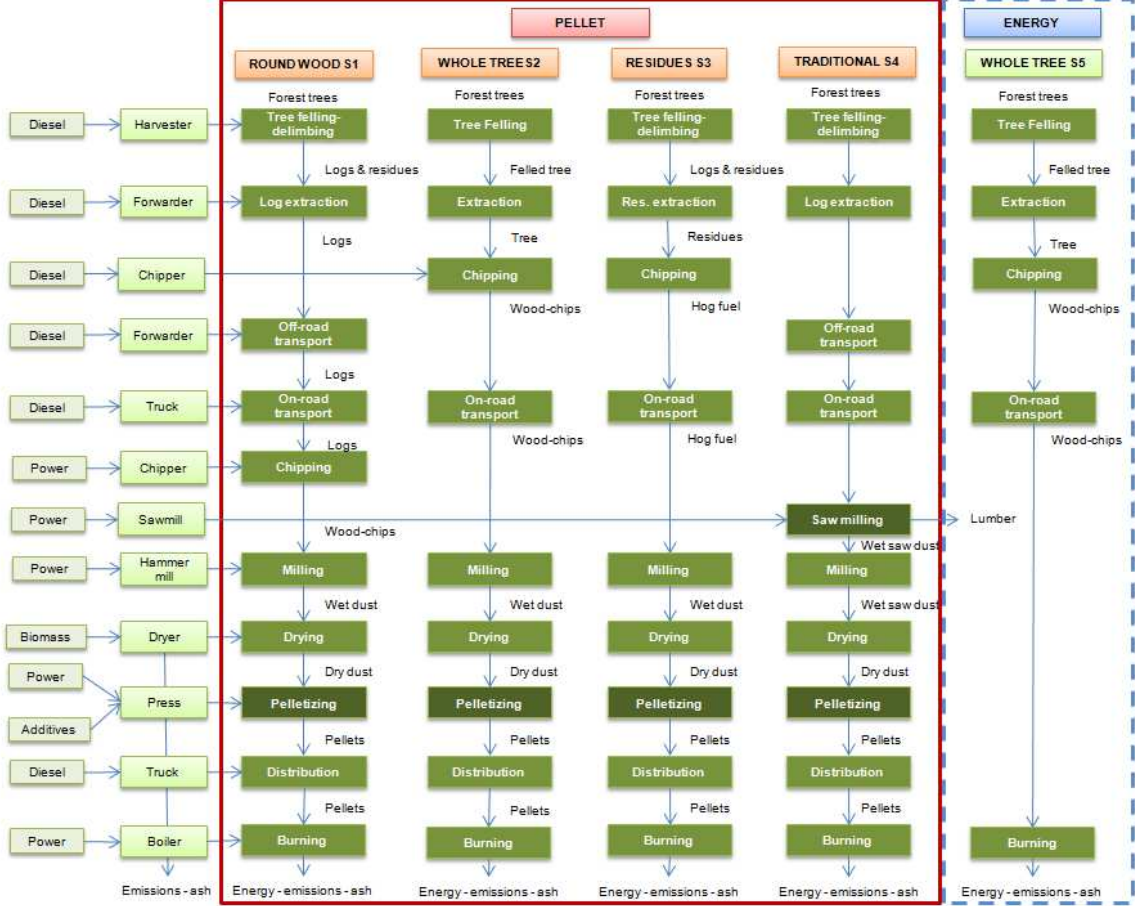


Figure 7-2: flowchart and logical framework of the LCA study

The depth and the level of detail of the LCA can differ considerably depending on the goal of a particular study. In our work the relatively wide system boundary chosen will be balanced with specific consideration about each single processing phase. To do this the attention on the quality of the data will be stressed on those phases that differ among the five scenarios. The life cycle inventory phase (LCI phase) results in list of input/output data, recorded in a matrix, with regard to the system being studied. It involves the collection of the data referred to each and every process considered in the study. This part of the work, which was mainly done during the study period abroad at SLU in Sweden, is built on two types of data: primary data that are collected directly from sectors operators or in the field by sensors and recorders, while secondary data are retrieved form literature review and public LCI databases. Secondary data were also utilized for the emission factors provided by Ecoinvent

database, that is widely recognized by the scientific community to be one of the most complete databases to perform LCA studies (Briggs, 2010).

A good LCA study should be characterized by a major share of primary data, which quality has to be tested and verified. Secondary data, whenever possible, have to be used for validation of primary ones instead of using them directly in the LCI.

The life cycle impact assessment phase (LCIA) has the purpose to provide additional information to help the evaluation of a product system and understand its environmental significance. This phase of LCA aims to evaluate the significance and amount of the potential environmental impacts that can derive based on the LCI flow results.

To perform the life cycle analysis and generate the emissions factors and to analyze the relative contribution of the various pellet chain processes to emissions we use the GaBi 6 software from PE International. The time frame used by the Kyoto Protocol (1997) to assess the global warming impact is 100 years. The environmental impacts of the pellet chain processes have been calculated using the CML 2001 – Apr. 2013 method developed by the Institute of Environmental Sciences (CML) of Leiden University (The Netherlands).

The environmental impacts are taken into account and the functional unit is 1 MJ of energy produced by pellet or wood-chips. Four impact categories have been considered: two global - Global Warming Potential (GWP) and Ozone Depletion Potential (ODP); and two local - Photochemical Ozone Creation Potential (POCP) and Human Toxicity Potential (HTP).

Global Warming is expressed in terms of carbon dioxide equivalents ( $\text{CO}_{2\text{-eq}}$ ). All the effects of the greenhouse gas emissions on global warming are referred to the  $\text{CO}_2$  multiplying the concentrations of each gas by its Global Warming Potential (GWP). The greenhouse effect is expressed in terms of increased temperature in the lower atmosphere due to an increased level of light reflection. This effect is the driving force of the climate changes at global scale. The Ozone depletion (expressed in terms of trichlorofluoromethane equivalents ( $\text{R11}_{\text{eq}}$ )) causes the reduction of the ozone layer protection from UV-radiation. Since there is no correlation between GWP and ODP emissions with the specific place of their emission, they are considered to contribute to the same impact and effect categories at a global level.

On the other hand we considered the photochemical smog creation (expressed in terms of ethylene equivalents ( $\text{Ethene}_{\text{eq}}$ )) and the human toxicity potential (expressed in terms of dichlorobenzene equivalents ( $\text{DCB}_{\text{eq}}$ )), to be at local scale, since their direct effects are

strongly related with the place where they are emitted. Particularly, all the considered impact categories have an effect on human health but the HTP measuring the potential effects of emissions on human health, takes into account also the heavy metals and particles (dust).

Life cycle interpretation is then the final phase of the LCA procedure, in which the results of the LCIA are summarized and discussed to provide a base for conclusions and decision-making.

### **7.2.2 Data collection and characterization**

The survey campaign to collect primary data was done with the collaboration of some companies involved in the chain being studied. A survey sheet was prepared and submitted to all those companies, which agree to participate in the study. The selection of the companies was done considering their specific environment and structure to select those that could be considered as a model for our specific work.

The forest operations sector is represented by enterprises that use harvesters and forwarders in fully mechanized forest operations to avoid the extreme variability in productivity of chainsaw based working sites. The three base materials selected (round wood, whole tree and forest residues) are then transferred to the road side in off-road transportation and, according to their destination, chipped there (full tree system S2 and residues S3) or transported in solid form to the factory for further processing (whole logs S1 and S4).

The pellet factory sector is represented by companies with a annual production rate between 38.000 and 90.000 t. This value has been chosen as a average of the EU pellet production companies equipped with 1 or more presses with nominal output of  $10 \text{ t h}^{-1}$ .

S5 is based on the thermal energy produced directly in a big district heating plant run on woodchips. The upgrading processes that involve the pellet production, in this last scenario, are discarded. S5 will provide a reference scenario to which refer all the results calculated for S1-S4.

S1 represents the production of pellets using whole logs as base material. This process involves the comminution of logs into woodchips and their further reduction into sawdust ready for the pelletization step.

S2 represents the production of pellets using whole trees as base material. For this process we consider the harvested material resulting from thinning operation in softwood stands. This operation results in huge volume of trees which stem has not the minimum quality and dimension requirements to be designated to lumber production. All the parts of the standing tree (stem, branches and needles) are comminuted and used in the pellet production.

S3 represents the production of pellets using only logging residues as base material. For this scenario we considered the residues produced during harvesting in softwood stand with fully mechanized system. The harvester cuts the trees and provides their delimiting. The extraction of such biomass is then operated with a forwarder.

S4 represents the classic wood pellet supply chain, based on sawdust and by-product from the lumber processing industry, typically sawmills. The forest operations of this scenario are the same as S1. Sawmill residues are represented by sawdust, shavings, trims and bark. They can be used as raw materials or fuel for pellet production. Since they are considered as co-products of the sawmill, they should share some of the emissions associated with the lumber production. That's the reason why we adopted the *allocation* to divide all the emissions that occur in the life cycle until the co-product is generated (at sawmill) proportionally by their mass between the lumber and the co-product itself. In this way all emissions at the sawmill are allocated simultaneously to the main product (sawn wood) and to the residues themselves.

S1, S2, S3, S4 share together the nominal productivity of the pellet press fixed in  $10 \text{ t h}^{-1}$ . The real productivity considered in the study derives then from primary data retrieved in trial tests carried out during the research (Chapter 5.2).

The energy production phase will be represented by pellet boilers in domestic appliances with a nominal power of 100 kW. The emission data and efficiency rates were retrieved from the reports of some emission test published by *BLT Josephinum* in Austria. Only for S5 the burning appliance is represented by a DHP plant of 18 MW power which data were directly measured at the plant in Sweden.

Based on the flow chart and the flows among processes, the calculation resulted in all system input and output data being referred to the functional unit. The functional unit has to provide a quantity reference to which the input and output data are normalized and referred. In this work the functional unit is the MJ of thermal energy produced burning wood

pellets (or woodchips in S5) delivered to the final user's place. All the material flows are referred to the same unit, as the energy densities, energy production and energy conversion. In Table 7-1 are reported the general characteristic of the pieces of equipment used in the five scenarios.

Table 7-1: characterization of the machines and tools involved in the study for each scenario

	Unit	S1 ROUND WOOD	S2 WHOLE TREE	S3 RESIDUES	S4 TRADITIONAL	S5 WHOLE TREE
Harvester	kW	190	150	190	190	150
Forwarder	kW	180	180	180	180	180
Chipper	kW	132	335	335	-	335
Truck	km	50	50	50	50	50
Sawmill	kWh t <sub>0</sub> <sup>-1</sup>	-	-	-	90.5	-
Hammer mill	kW	350	350	350	350	-
Dryer heat	kWh t <sub>0</sub> <sup>-1</sup>	896	727	1097	896	-
Conditioner	kW	30	30	30	30	-
Press	kW	750	750	750	750	-
Nom. press prod.	t h <sup>-1</sup>	10	10	10	10	-
Cooler	kW	50	50	50	50	-
Sieve	kW	81	81	81	81	-
Internal transportation	kW	220 x 3	220 x 2	220 x 2	220 x 2	220 x 1
Ventilation and conveying system	kW	100	100	100	100	-
Packaging	kW	90	90	90	90	-
Real plant productivity	t h <sup>-1</sup>	4.5	10	8	8	-
Truck	km	300	300	300	300	50
Boiler	kW	100	100	100	100	18000 + DHP *

\* DHP: District Heating Plant. It comprises the heat distribution through a network of approximately 18 km and 250 users connected

### 7.3 Results and discussion

Figure 7-3 shows the relative emission contribution of the 5 scenarios for the four impact categories analyzed. Data are provided in relative percentages given S4 being the reference scenario which contribution is fixed to be 100%. All the calculations are referred to S4 which represents the production of pellet from by-product of the sawmilling industry. From the graphic it is possible to note that S2 is the scenario with the best emission performances for all the four impact categories chosen, and that for none of them are recorded values greater

than for S4. In particular the widest differential is recorded for GWP that is 54% of S4. S1 is the second best scenario, since only for ODP a value greater than 100% is recorded. For all the other impact categories the values of relative contribution are lower than this value but never less than 92%, proving a little deviation from the reference S4. S3, on the contrary, has pretty good performance for GWP (65%) while results get worse for other impact categories exceeding for all of them the 100% threshold. S4 itself represent an average situation that is located in median position among others. Finally results for S5 give evidences of the relative low emission characteristics of this last scenario. The dotted line indicates that the comparison of S5 with the others has to be done considering that all the production phase of pellet is missing from S5 (basic assumption). The percentages can be used to investigate the net contribution of GHG emission exploiting the raw materials directly as wood chips in a big DH plant instead of bringing them to further processing in the pellet plant. The highest value is recorded for the HTP since for this category the contribution of pollutants emission from burning is determinant, while the non-biotic emissions for GWP are very low as few energy input are involved in the process.

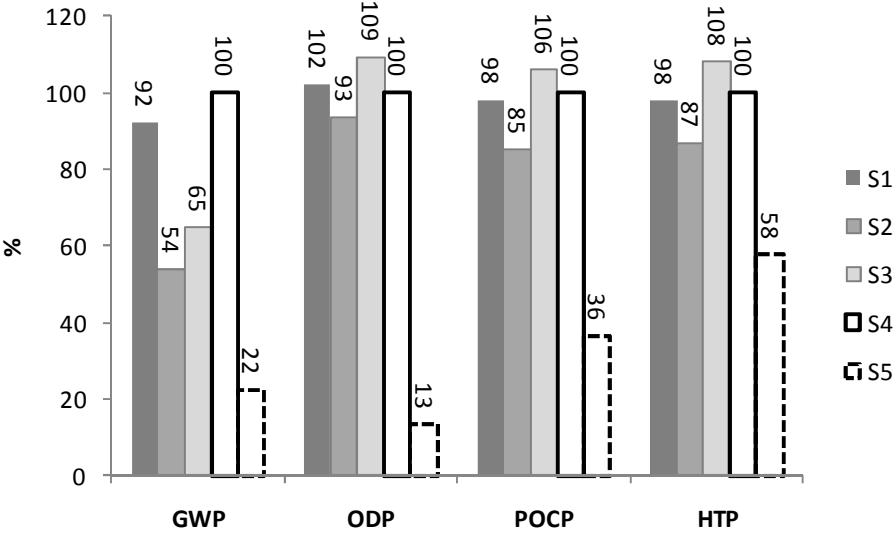


Figure 7-3: relative emission contribution for each impact category grouped per scenario

To better understand the total emission calculated in this study and represented in Figure 7-3 a further subdivision of the result was operated to underline the specific contribution given by each step in the supply chain. To do this every process of each scenario was grouped into six phases specifically designed for the scope of the study. In this paragraph, in order to describe the final results, we refer to *processes* and *phases*. A *process* is considered

to be the smallest analytical unit to which the elementary flows of energy, materials and emissions refer. A *phase* is considered to be a group of subsequent processes as mentioned in Table 7-2. The phases are *forest operations*, *transportation of resource*, *preparation*, *production*, *transportation* of final product and final *burning*. Table 7-2 shows which process contributes to the definition of the specific phase.

*Forest operations* groups all the processes from the felling of the tree to the loading on the truck at the road side. For S2 and S3 this comprises also the *chipping* operation. *Transport of the resource* is simply the carrying step of feedstock from the forest road to the factory gate. At this point the *preparation* phase starts from *debarking* of logs to the feeding hopper of the pellet press. The *production* phase starts with *pelletization* and ends with the *packaging* of the final product. *Internal transportation* and *ventilation and conveying* were added to this phase to comprise the emissions related with these procedures that are common to all the operation within the factory. The *transport* represents the distribution of pellets within a distance of 300 km or woodchips within a distance of 50 km. Finally the *burning* phase is considered separately to discriminate the emission related with burning.

Table 7-2: description of all the *processes* involved in each *phase* in the supply and production chain for each scenario

	S1	S2	S3	S4	S5
	Logs	Whole tree	Residues	Traditional	Heating plant
Burning	Burning	Burning	Burning	Burning	Burning and distribution
Transport	- Transport pellet	- Transport pellet	- Transport pellet	- Transport pellet	Transport woodchips -
Production	Internal transportation	Internal transportation	Internal transportation	Internal transportation	Internal transportation
	Ventilation and conveying	Ventilation and conveying	Ventilation and conveying	Ventilation and conveying	-
	Packaging	Packaging	Packaging	Packaging	-
	Sieving	Sieving	Sieving	Sieving	-
	Cooling	Cooling	Cooling	Cooling	-
	Pelletizing	Pelletizing	Pelletizing	Pelletizing	-
Preparation	Conditioning	Conditioning	Conditioning	Conditioning	-
	Drying	Drying	Drying	Drying	-
	Milling	Milling	Milling	Milling	-
	Chipping	-	-	-	-
	-	-	-	Sawing	-
	Debarking	-	-	Debarking	-
Transport resource	Road transportation	Road transportation	Road transportation	Road transportation	Road transportation
Forest operations	-	Chipping	Chipping	-	Chipping
	Forwarding	Forwarding	Forwarding	Forwarding	Forwarding
	Felling and disbranching	-	Felling and disbranching	Felling and disbranching	-
	-	Felling	-	-	Felling

## GWP

As far as the GWP is concerned, the biggest share of the emissions is represented by biogenic CO<sub>2</sub> produced during the production of energy through the final combustion. The way to evaluate the emissions of biogenic source is still under discussion because they are

often internationally assumed to be the same as the carbon adsorbed in forest through the photosynthesis phase. For this reason in many studies this contribution is neglected. The carbon neutrality assumption based on international standards and guidelines, allow not accounting for the biogenic carbon dioxide in LCA studies. This is also the choice in this work in which we kept the biogenic contribution of CO<sub>2</sub> separate from the fossil one. The same was not operated for methane and other GWP gases that are commonly measured in CO<sub>2eq</sub> since the same benefit from tree activity cannot be assumed for CH<sub>4</sub>.

Including the biogenic CO<sub>2</sub> emissions, the impact on global warming for the production of 1 MJ of energy from pellet in domestic heating (or woodchips for S5) through the whole life cycle for S1 is 186 gCO<sub>2eq</sub>, for S2 is 160 gCO<sub>2eq</sub>, for S3 is 196 gCO<sub>2eq</sub>, for S4 is 188 gCO<sub>2eq</sub>, for S5 is 119 gCO<sub>2eq</sub>. Excluding the biogenic CO<sub>2</sub> emissions, the GWP for S1 is 25.4 gCO<sub>2eq</sub>, for S2 is 14.9 gCO<sub>2eq</sub>, for S3 is 17.9 gCO<sub>2eq</sub>, for S4 is 27.6 gCO<sub>2eq</sub>, for S5 is 6.2 gCO<sub>2eq</sub>. This results in very steady levels of carbon dioxide emissions from biotic sources which differences among the five scenarios are basically caused by: i) the different calorific value of the three type of materials involved in *burning* phase; ii) the different moisture content of the feedstock before the *drying* phase that influences the quantity of biomass needed for drying; iii) the different processes added to *preparation* phase in S4.

From Figure 7-4 the share of this contribution among all phases is evident. S1 and S4 has both a huge emission level in *forest operation*, that is not recorded for the others scenarios. The main contributor for this is the biotic methane released in the forest during the decomposition of the residues left on the felling site, since for these scenarios forest residues are not exploited. On the other hand all the fertilizing effect of releasing minerals into the forest could not be accounted in the study. The second major contributor in GWP is given by the preparation stage. This is dramatically high for S4, follow by S1, S3 and S2. The reason for that have to be found in the very high emission of CO<sub>2</sub> and N<sub>2</sub>O related to the drying process, that involves biomass combustion. Moreover S4 comprises in the *preparation* phase the entire sawmilling procedures, that even if they involve only electric and diesel consumption, the energetic input for them is fairly high. The decreasing values for the other scenarios follows the decreasing productivity of the plant run on different feedstock materials. The same is registered for the *production* phase which trend follows the net pellet output from the plant that is different for each feedstock considered.

As far as *burning* phase is concerned for S1 – S2 – S3 – S4 the emission level of GWP, not considering the biogenic CO<sub>2</sub>, is more or less equal as the same amount of wood pellet has to be burnt to produce 1 MJ (approx. 66.6 g). Some slight differences have to be accredited to the different calorific value of the feedstock used affecting the net calorific value of pellets (moisture content is assumed to be constant at 8%). For S5, even not considering the biogenic share, the total CO<sub>2</sub> is about 99% because of the relative big investment of electricity for the heat distribution. The transport distance of chips is set at 50 km. The transportation of pellets for their distribution, based on 300 km trip, represents an important GWP emission source that is kept constant for all scenarios. Its level varies linearly with respect of the distance covered.

### ODP

With regard to the ozone depletion potential, its value is not influenced by the combustion for all the scenarios. In fact during this phase none of the chemical substances responsible for ODP are produced and emitted in the life cycle. For this study the only ODP emission is related to organic emission to air (group VOC – volatile organic compound).

The production of 1 MJ of energy from pellet in domestic heating (or woodchips for S5) through the whole life cycle for S1 is 2.02 µgR11<sub>eq</sub>, for S2 is 1.85 µgR11<sub>eq</sub>, for S3 is 2.16 µgR11<sub>eq</sub>, for S4 is 1.98 µgR11<sub>eq</sub>, for S5 is 0.261 µgR11<sub>eq</sub>.

It is important to notice that, even if ODP is a global index, the total quantity of chemicals with impact on the ozone depletion are produced in quantity of few micrograms (10<sup>-6</sup> g) that are very small in terms of absolute values.

The specific emission potential of each phase of the supply chain in the life cycle analysis varies considering the energetic input due to fossil fuel consumption. For this reason to the high level of emission related with the *forest operation* of S2 and S3, together with pretty low productivity of harvesting and forwarding operation, are associated the chipping operation at road side. The comminution at this place is done by the mean of diesel engine powered chippers. The *preparation* phase of S4 again is influenced by the sawmilling process that contributes substantially for the overall ODP factor while *drying* is the major contributor of this phase common for all the scenarios. The *production* phase is influenced by the specific press productivity and secondarily by the internal transportation stage. In S1 there are three front loaders involved, while in the other scenarios only two. The fairly high level

of ODP for S5, that is related for more than 90% to the *burning* phase, is dependent on the distribution phase of the heat in the DH network, from the centralized boiler to the final user's radiators.

### **POPC**

The production of 1 MJ of energy from pellet for domestic heating (or woodchips for S5) through the whole life cycle for S1 is 19.4 mg Ethene<sub>eq</sub>, for S2 is 16.9 mg Ethene<sub>eq</sub>, for S3 is 21.0 mg Ethene<sub>eq</sub>, for S4 is 19.8 mg Ethene<sub>eq</sub>, for S5 is 7.2 mg Ethene<sub>eq</sub>.

Combustion is widely considered to be a crucial step in terms of pollutants formation (Cespi et al., 2014; Solli et al., 2009). The main variables that affect the combustion behavior and the emissions though have to be found in the combustion efficiency and the pollutant removal efficiency. All these variables are then affected by the fuel type and its composition (Cespi et al., 2014).

In addition to carbon dioxide, in a real combustion, some other pollutants are produced: carbon monoxide, sulfur oxides and nitrogen oxides, volatile organic compounds, dust and ammonia. The oxides of sulfur and nitrogen, not only have toxic effects on living creatures and on the environment, but they also cause acid rains and represent precursors of secondary pollutants. Moreover nitrogen oxides, volatile organic compounds (VOC) and ozone are some of the causes of photochemical pollution (or smog). Photochemical smog is a specific type of pollution that is created in days characterized by weather conditions of stability and strong sunlight. Nitrogen oxides and VOC together, emitted in many natural processes in the long term, undergo a complex system of photochemical reactions induced by UV light from the sun that lead to the formation of ozone, peroxiacetyl nitrate (PAN), peroxibenzoil nitrate (PBN) and hundreds of other substances. These secondary pollutants are indicated with the collective name of photochemical smog because they are generated by chemical reactions catalyzed by light. Among the many volatile organic compounds emitted during combustion, many are toxic by themselves (benzene, toluene, xylenes, aldehydes etc.).

From the results it is evident how the potential impact on photochemical ozone creation is considerably higher in the phases where some sort of active combustions are present (i.e.: *preparation* and *burning*). Not only this assumption is generally speaking valid, but this also results in higher values of POCP for the *preparation* phase of S3 ( $8.44 \cdot 10^{-6}$  conditioning,

*drying and milling*) than S4 ( $7.05 \cdot 10^{-6}$  *debarking, sawing, conditioning, drying and milling*) even if more processes are involved in the latter.

For this indicator the most efficient scenario is S2, especially for what it concerns the emissions related to the *forest operations* that basically miss the *disbranching* process since for S2 the *forwarding* is done with the whole tree that substantially increases the productivity.

Taking a look to the singular pollutant that contributes to the POCP definition, among others N<sub>2</sub>O is the first contributor of this indicator for the non organic emissions, followed by biotic CO and SO<sub>2</sub>. For the organic emission the first contributors are NMVOC that cover the biggest share among others, lately followed by biotic CH<sub>4</sub>.

### **HTP**

The production of 1 MJ of energy from pellet for domestic heating (or woodchips for S5) through the whole life cycle for S1 is 19.1 g DCB<sub>eq</sub>, for S2 is 16.9 DCB<sub>eq</sub>, for S3 is 21.1 DCB<sub>eq</sub>, for S4 is 19.5 DCB<sub>eq</sub>, for S5 is 11.2 DCB<sub>eq</sub>.

The human toxicity potential is defined in LCA by four main groups of pollutants: heavy metals, inorganic emissions, VOC and dust to air. As it was for the POCP, also HTP is mainly influenced by the contribution of biomass combustion emissions, so the detection of higher values of this index, in terms of equivalents, is expected in the phases that comprise combustion: *drying* and *burning*. For all the scenarios the share of HTP is higher than 24% for *drying* (max. value 40.8% for S3) and 40% for *burning* (max value 59.6% for S2). In all cases only these two processes cover at least two third of the total HTP.

The decreasing order of sorted contribution for the four groups of pollutants is the same found in other studies on firewood: organic emissions, heavy metals, inorganic emissions and particles/dust.

Organic emissions produced by combustion include polycyclic aromatic hydrocarbons (PAH), halogenated organic emissions, polychlorinated dibenzo-p-dioxins (2,3,7,8-TCDD), benzene and formaldehyde that could create a great hazard for human health even in very small concentrations. Heavy metals are naturally contained into the wood and they are then released after *burning*. Among others the heavy metals that represent the biggest hazard for humans are: Arsenic (+V), Nickel (+II), Chromium (+VI), Cadmium (+II), Lead (+II) (Table 7-3).

The inorganic emissions, already present for other impact categories accounting such as GWP and POPC, have also a direct impact on human health and their potential effect on living beings is mainly affected by nitrogen oxides, hydrogen fluoride, hydrogen chloride and ammonia. This last is released in combustion processes.

The combustion of solids and liquids is always responsible of the production of some particles which size distribution allows determining their dangerousness for health. The smaller they are, the worse is their effect, especially for fines and ultrafines (PM<sub>2.5</sub>). Regarding the dusts emitted, their degree of penetration into the human respiratory system and their consequent toxicity depends on the particle size distribution: they may be the vehicle of chemical agents toxic to humans, such as heavy metals, polycyclic aromatic hydrocarbons, polychlorinated biphenyls and dioxins generated by burning.

Considering the whole HTP, as it was for all the other indexes, S5 reported the lowest value but in this case the differences with other scenarios is less than 50%.

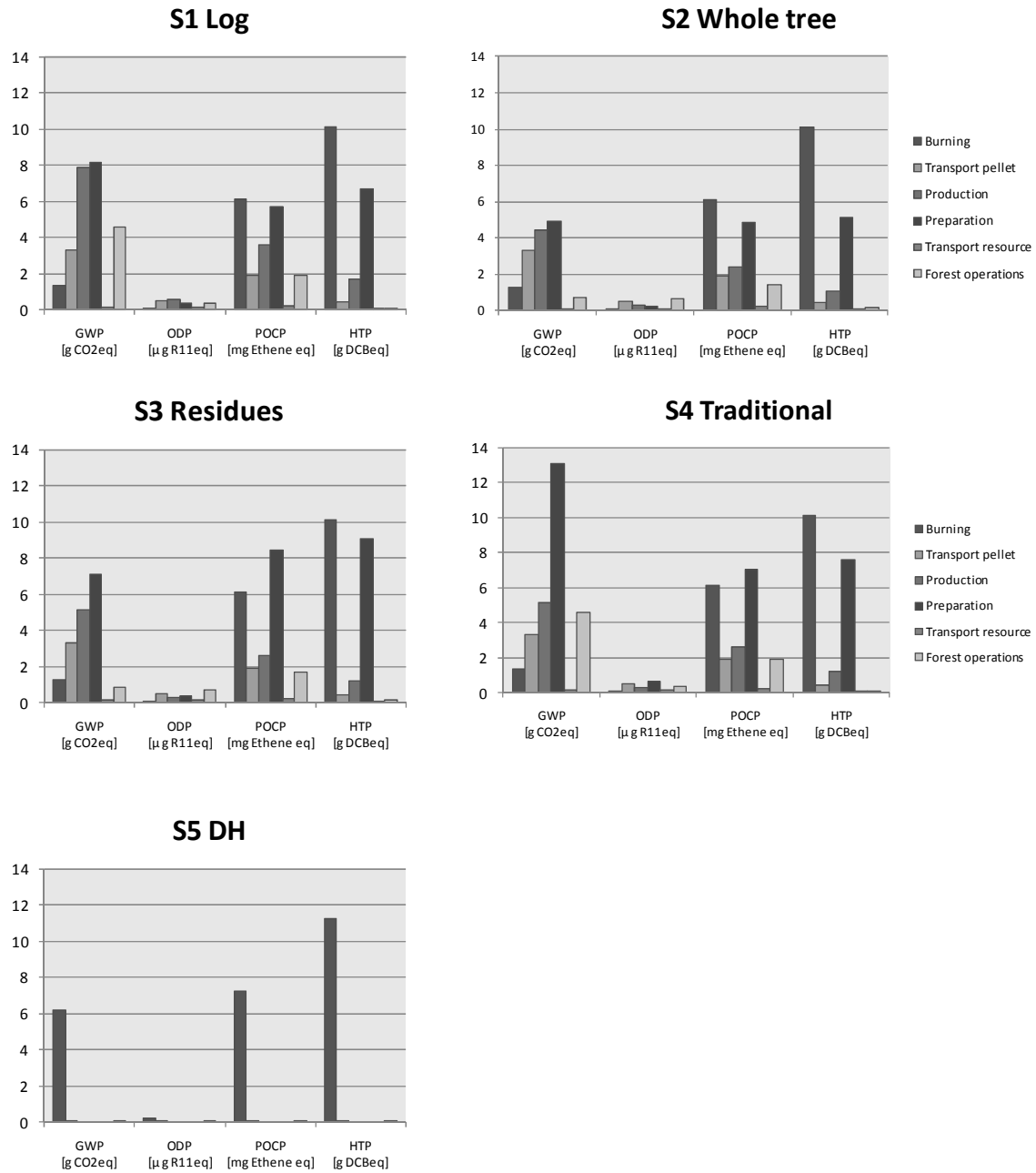


Figure 7-4: specific emission factors for all the scenarios grouped per factor. GWP is given without the contribution of biotic CO<sub>2</sub>

Table 7-3: specific emission quantities grouped per impact category and scenario

	S1	S2	S3	S4	S5
<b>GWP</b>	g CO <sub>2eq</sub>				
<b>Emissions to air (total)</b>	185.70	159.84	195.79	187.97	118.92
Carbon dioxide (biotic)	160.25	144.94	177.89	160.36	112.72
Carbon dioxide	18.92	12.97	15.58	20.97	5.15
Nitrous oxide (laughing gas)	1.31	1.18	1.42	1.33	0.79
Methane (biotic)	4.24	0.04	0.05	4.24	0.03
Methane	0.95	0.68	0.80	1.03	0.24
Group NMVOC to air	0.03	0.03	0.05	0.03	1.19E-03
Sulphur hexafluoride	2.51E-03	2.54E-03	3.01E-03	2.49E-03	1.68E-04
<b>ODP</b>	µg R11 <sub>eq</sub>				
<b>Halogenated organic emissions to air*</b>	2.02	1.85	2.16	1.98	0.26
*Halon (1301), Halon (1211), R 22 (chlorodifluoromethane), R 114 (dichlorotetrafluoroethane), Carbon tetrachloride (tetrachloromethane), R 12 (dichlorodifluoromethane), R 113 (trichlorotrifluoroethane), Chloromethane (methyl chloride), R 11 (trichlorofluoromethane), 1,1,1-Trichloroethane					
<b>POPC</b>	mg Ethene <sub>eq</sub>				
<b>Emissions to air (total)</b>	19.45	16.88	20.98	19.82	7.22
Carbon monoxide (biotic)	2.49	2.34	2.70	2.49	1.22
Group NMVOC to air*	7.44	7.53	9.44	7.41	0.06
Nitrogen oxides	4.89	4.29	5.40	4.98	3.25
Carbon monoxide	0.49	0.56	0.80	0.50	0.04
Sulphur dioxide	2.89	1.99	2.45	3.16	0.92
Methane (biotic)	1.02	0.01	0.01	1.02	3.42E-04
Methane	0.23	0.16	0.19	0.25	0.01
*1,1,1-Trichloroethane, Chloromethane (methyl chloride), Dichloromethane (methylene chloride), Tetrachloroethene (perchloroethylene), Trichloromethane (chloroform), Vinyl chloride (VCM; chloroethene), 1-Butanol, 1-Pentene, 2-Methyl-2-butene, Acetaldehyde (Ethanal), Acetic acid, Acetone (dimethylcetone), Alkane (unspecified), Benzaldehyde, Benzene, Butadiene, Butane, Butanone (methyl ethyl ketone), Cumene (isopropylbenzene), Ethane, Ethanol, Ethene (ethylene), Ethine (acetylene), Ethyl benzene, Ethylene acetate (ethyl acetate), Formaldehyde (methanal), Formic acid (methane acid), Heptane (isomers), Hexane (isomers), iso-Butanol, Isoprene, Isopropanol, Methanol, Methyl acetate, Methyl formate, Methyl tert-butylether, NMVOC (unspecified), Pentane (n-pentane), Propane, Propene (propylene), Propionaldehyde, Propionic acid (propane acid), Styrene, Toluene (methyl benzene), Xylene					
<b>HTP</b>	g DCB <sub>eq</sub>				
<b>Heavy metals to air*</b>	3.72	2.97	3.71	3.96	1.63
*Arsenic (+V), Nickel (+II), Chromium (+VI), Cadmium (+II), Copper (+II), Zinc (+II), Vanadium (+III), Lead (+II), Chromium (unspecified), Selenium, Mercury (+II), Cobalt, Antimony, Molybdenum, Thallium, Tin (+IV), Hydrogen arsenic (arsine)					
<b>Inorganic emissions to air*</b>	0.83	0.53	0.67	0.92	0.35
*Nitrogen oxides, Hydrogen fluoride, Sulphur dioxide, Ammonia, Beryllium, Barium, Carbon disulphide, Hydrogen chloride, Hydrogen sulphide					
<b>Organic emissions to air (group VOC)</b>	14.46	13.36	16.57	14.54	9.20
Polycyclic aromatic hydrocarbons (PAH)	11.30	10.50	13.10	11.30	7.18
Other organic emissions to air*	3.05	2.76	3.35	3.13	1.95
Halogenated organic emissions to air**	0.11	0.10	0.13	0.11	0.07
*Benzene, NMVOC (unspecified), Formaldehyde (methanal), Toluene (methyl benzene), Propylene oxide, Ethyl benzene, Ethylene oxide, Xylene (meta-Xylene; 1,3-Dimethylbenzene), Ethene (ethylene), Xylene (dimethyl benzene), Butadiene, Acrolein, Phenol (hydroxy benzene), Styrene					
**Polychlorinated dibenzo-p-dioxins (2,3,7,8 - TCDD), Hexachlorobenzene (Perchlorobenzene), Vinyl chloride (VCM; chloroethene), Dichloroethane (ethylene dichloride), Carbon tetrachloride (tetrachloromethane), Pentachlorophenol (PCP), Trichloromethane (chloroform), Dichlorobenzene (o-DCB; 1,2-dichlorobenzene), 2,4-Dichlorophenol, Pentachlorobenzene, Dichloromethane (methylene chloride), 1,1,1-Trichloroethane, Tetrachloroethene (perchloroethylene), Methyl bromide					
<b>Particles</b>	9.9E-02	8.4E-02	1.1E-01	1.0E-01	8.7E-02
Dust (PM2.5)	4.8E-02	4.0E-02	5.7E-02	4.8E-02	5.6E-02
Dust (> PM10)	5.0E-02	4.3E-02	5.2E-02	5.1E-02	3.1E-02
Dust (PM2.5 - PM10)	1.5E-03	1.3E-03	1.6E-03	1.6E-03	2.5E-04
Silicon dust	4.1E-05	2.6E-05	3.4E-05	4.6E-05	1.4E-05

## 7.4 Conclusions

The study has been based on the comparison of the specific emissions of five different supply chains for energy production from forest products and residues. The aims of the study were to evaluate the environmental impact of pellet production and energy production comparing different supply chains, and to refer them to a reference scenario. Four of these were based on pellet production while one, kept as reference, refers to a DHP run on fresh woodchips from whole tree.

Primary and secondary data were collected, normalized and processed by the mean of GaBi LCA software. Results were expressed in four emission and impact categories, chosen among all for their significance in discriminating alternative scenarios. The environmental impacts were calculated based on the method CML 2001 – Apr. 2013 from the Leiden University.

Two of them were considered as local indicators (POCP, HTP), while other two ad global indicators (GWP, ODP).

Results generally show that the best option, when comparing different productive alternatives, is always the one where less processes and energy inputs are required. It was found that some specific processes (*burning, drying, pelletizing*) are the biggest contributors for all the four impact categories, but nevertheless, scenarios with more processes are not able to compete with simplest ones.

This is the reason why S5, even if it is not included in the direct comparison object of this study, has the lowest absolute emission since more than half of the processes involved in other scenarios, are discarded for this last one. Same reasons can be given for S2 that has the best environmental performance among all the others. The whole tree system extraction allows to avoid one of the most energy-demanding phase and to chip all the unsorted material at the road side. For that reason the *preparation* phase misses *debarking* and any sorting processes. This is then coupled with pretty high press productivity that is facilitated by the optimal mixture of wood and bark into the feedstock, able to produce very durable pellets.

These comparisons could be set as starting point in the definition of the best environmental and energetic performance of different supply options, and should always be considered before accurate judgments can be formulated. Nevertheless it is crucial to consider that the chains analyzed in this study are very long and complex. A number of assumption and

simplification were made and sometime finding reliable and consistent data was not easy. The straight application of the international standards and guidelines should always be considered to maintain the comparability of data and results high and reliable worldwide.



## 8 Conclusions

The study, structured in four subsequent functional pillars, aims to verify the possibility to set up a wood pellet supply chain that originates directly from the output material of forest operations. The first step was to investigate the real quality of commercial pellet samples that could be purchased in the market and set a baseline for further comparisons. Through a consistent sampling randomly done over several months and the SIMCA elaboration, the results showed a total explained variance of 95.77%. This proved that the metric distance set by the model is able to define the quality of that sample and predict the certifiability of a pellet sample based on few core quality parameters measured (M, DE, DU, BD, LHV, A). The SIMCA distance is a real scale for positioning each sample on a line. Values below the critical distance are included in the model while higher distance values are a symptom of lower quality.

This is then proven considering that the internal sensitivity of the model is 100% (all the certified samples used to build the model were included in the SIMCA model itself) and that external sensitivity is also 100% (all the certified samples used as external test were also included in the SIMCA model). The final overview of the commercial pellet market based on the model showed that, over a population of 57 samples, 42 resulted to be certifiable while only 15 were not.

The following step in the concept map of the thesis was to verify the possible quality retrievable producing pellets from different forest assortments, and in particular from by-products or residues of forest operations based on two species: spruce and beech.

The five types of pellet were analyzed and results showed that the best option is the one made from spruce debarked stem, even if some issues related to the mechanical properties were recorded. Lower properties were measured for pellet made of whole stem (mainly because the ash content and the calorific value) and pellet from the whole spruce tree and beech stem. The worst properties were for pellets produced with branch wood from beech that still felled in class B. These results express the possibility to exploit such resources in the production of pellets destined for the domestic consumption or to improve the final quality with some blends of raw materials able to upgrade the same residues and allow their utilization in small appliances. This could improve the added value of some materials which

exploitation is now non economic, and increase the productive function of marginal forests or abandoned stands.

The production of pellets with different raw materials or through different plant set up could lead to differences in color of the final product. Color of pellets has been recorded as a key element in the commercial choice of the final consumer. Since no scientific evidences of relationships between color and quality of pellet were studied before, this aspect was investigated in this study.

High correlation were found between colorimetrics and quality parameters especially for ash contents, the first indicator of big concern for final costumers. Moreover the direction of this relation is constant and darker pellets have higher ash contents. The capacity of the human eye to detect such differences was investigated and the parameter  $\Delta E$  between high and low ash content samples confirmed that extreme differences are appreciable at first sight. On the other hand the same predictability is not confirmed for samples showing near colors. Moreover the statistical analysis resulted in a clear cloud of certified sample clustered together around high quality parameters values, while pellets from crop and agriculture residues were clustered at the opposite.

The study has then shown that the application of such methodology in industry sector can help producers to achieve high quality properties of pellets coupled with reasonable level of color to satisfy the need of the market that requests pretty light materials.

The last step of the study regarded the environmental impact evaluation of five alternative supply chains for energy production from forest products and residues with the objective to evaluate their environmental performance with reference to a base scenario.

Results showed that the best option, with regard to the energy final output, is obtained when few processes and energy inputs are involved along the chain following the general rule that the simpler is the chain the best is its performance. Moreover some specific processes give wider emission contribution (*burning, drying, pelletizing*) and since all of them are required to achieve the final product (pellet) it is necessary to study the chain to reduce their impact and make them more efficient (natural drying, chain).

The application of these results on real cases should then include a more detailed data analysis considering some key aspects of the chain being studied and adapting them to the measured one.

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