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Dry matter losses during biomass storage

Measures to minimize feedstock degradation

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Measures to minimize feedstock degradation

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Abstract

The degradation of biomass during storage leads to several unfavourable outcomes including Greenhouse gas (GHG) emissions, feedstock/energy losses, and economic losses. Optimization of biomass storage along the supply chain for the reduction of these negative effects is essential in order to improve bioenergy as a renewable and profitable energy source. The overall effect of the biological, chemical and physical reactions, which occur in biomass piles, leads to a succession of microorganisms as pile temperatures increase, which in turn releases GHG emissions, such as CO2, CH4 and CO, into the atmosphere. Furthermore, valuable extractive compounds from tree biomass begin to diminish directly after harvesting occurs. Management that facilitates drying can reduce dry matter losses caused by microbial activity. Pre-storage in small heaps during favourable storage conditions can increase the drying rate at the harvesting site. However, forest residues stored at the harvest site will retain moisture more rapidly than windrows at landing during precipitation. Favourable exposure to sun and wind during storage is essential for the result. Coverage of forest residues stored in windrows can protect the biomass from rewetting leading to lower moisture content and higher net calorific value. The effect of coverage may be large on some landings and negligible in others and therefore location and season must be considered. Comminution to chips increase the surface area exposed to potential microbial degradation, increase pile compaction decrease the permeability leading to increased activity and temperature within the pile. It is highly recommended to strive to comminute to the largest possible particle size accepted from the end-user. Minimize compaction during storage both by avoid using heavy machinery on the piles and limit the height of piles below 7 m. Coverage with a semipermeable material can protect wood chips from rewetting leading to lower moisture content, which will reduce dry matter losses and energy losses. If possible, limit storage time to 3-4 months if possible and expect a major temperature increase during the first weeks of storage. Methods for monitoring degradation include calculating dry matter losses using sampling net bags in storage piles. However, other methods are used and there is no general standard. Predictive models of temperature development, fuel quality parameters and dry matter losses allow for the simulation of pile dynamics based on input parameters. As these models develop, they will allow useful information to be obtained for improved storage management without the need for excessive sampling. Thereby, adverse effects on storage can be reduced.

Keywords:

Bark, Biomass, Degradation, Dry matter loss, Gas regime, Gaseous emissions, Forest residues, Pellets, Storage, Stumps, Mass loss, Moisture content, Woody biomass

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Introduction

In the face of climate change, stabilizing atmospheric concentrations of greenhouse gases (GHG) remains a major global environmental and political challenge. Alternative renewable energy sources can contribute to phasing out technologies based on fossil fuels to reduce emissions. Biomass can be considered as a renewable energy source since theoretically the carbon released into the atmosphere through combustion can be re-sequestered during the next generation of biomass growth. Carbon neutrality has been questioned however and extensive biomass harvesting can have a multitude of effects on biogenic carbon stocks, depending on the characteristics of the bioenergy system and land-use history. Bioenergy is currently the largest renewable source used in EU and several member states have increased the use of forest biomass for energy to reach their 2020 renewable targets. Common practice (at least for the Nordic countries) is to manage forests initially for timber production and secondly for pulp production. Less valuable parts of trees, classified as primary forest residues (e.g. logging residues, tree parts, wood from early thinning and stumps) and secondary forest residues (which are the residues from the industrial processing of wood) are attractive for bioenergy production. Benders et al. (2016) concludes that emissions derived from biomass supply chain operations are minor when forest biomass is transported within a relatively short distance. Furthermore, GHG emissions from the bioenergy supply chain can be highly reduced by utilizing more effective handling methods and efficient transport strategies over longer distances (Berndes et al., 2016).

Harvesting operations and residue production occurs all year round. Forest residues are still mainly utilised for heat and power generation in temperate developed countries where the highest demand for such biomass occurs during the winter season. Immediate utilization of forest feedstock is often unfeasible however due to high moisture content and varying demand. As a consequence, it is often necessary to store wood fuel (at least temporarily) close to harvesting sites or at industrial terminals. Biological, chemical and thermo-oxidative reactions lead to the degradation of biomass during storage (Jirjis, 2001) where degradation rates are dependent on microbial populations and species, available carbon, oxygen concentration, temperature and moisture levels. These reactions result in dry matter losses and gaseous emissions (particularly CO and CO2). Numerous studies state the occurrence of dry matter losses, ranging between 0.3 to 4.2 % per month. These losses reduce the available energy content in feedstock thus leading to increased costs and negatively influencing emissions per unit of energy delivered.

Since some form of biomass storage is essentially inevitable in all forest bioenergy supply systems, it is critical to consider the issues relating to feedstock quality changes and processes leading to risks during storage. Moisture management is a key element to improve net calorific value and the cost-efficiency of energy wood supply, through the whole supply chain. The choice of storage location and method is usually influenced by biological, economic and logistic considerations (Richardson, Björheden, Hakkila, Lowe, & Smith, 2006). Dry matter losses can affect both the profitability of using forest biomass as well as any GHG reduction benefits from replacing fossil fuels. A higher understanding of the complex processes, which occur during storage combined with the implementation of dry-matter loss mitigation strategies, can significantly reduce these negative effects and risks during storage. The following report describes the processes responsible for mass loss and moisture changes in woody biomass during industrial storage. The woody biomasses considered in this report are forest residues and sawmill residues in the form of chips or ground material, including fresh wood, branch wood, bark and some foliage. This report outlines several management strategies aiming to preserve/improve fuel quality and minimize net energy and GHG losses during storage of unprocessed forest biomass in both small and large-scale storage trials.

OBJECTIVES OF STUDY

The overall objective was to review recent results related to dry matter losses during biomass storage as well as preventative strategies for the purpose of improving management efforts and minimizing the associated consequences of feedstock degradation (such as GHG emissions and economic losses in the supply chain). This work is limited to only consider woody biomass.

TARGET GROUP OF REPORT

The following report is primarily targeting producers, traders and users of solid biomass for energy production. The information contained herein is also pertinent to the scientific community for alternative energy and climate change research (e.g. when comparing the climate mitigation potential of alternate supply chains).

Theory of biological degradation of woody biomass under industrial storage conditions

1. BIOLOGICAL DEGRADATION

In fresh, un-dried woody biomass, microorganisms are by far the most predominant mechanism for the deterioration of the material under storage. Microorganisms can enter the biomass pile via several routes. First, certain types of decay, soft rot, or staining fungi, may inhabit the wood prior to harvesting and hence will continue to thrive under chipped/ground conditions once stored in a pile. Secondly, harvesting activity, cutting, hauling and piling, will incorporate soil, which harbours a variety of wood attacking microorganisms (decay fungi, soft rots, moulds and bacteria), into the bark/branches (Scheffer, 1973). Thirdly, standing water, either around the pile or under it, may introduce bacteria that is dispersed in the water. Finally, liberated fungal spores may start new infections in the storage pile if they find suitable microclimate and substrate conditions where they land or when conditions within the pile dictate.

There are a number of studies that have identified the varied composition of microbial communities in stored woody biomass (Kropacz & Fojutowski, 2014; Noll & Jirjis, 2012) . In the study by Kropacz and Fojutowski the species of microorganisms identified in the stored biomass showed a succession over the 120 day study period. They found the initial material had evidence of staining fungi and bacteria, which later shifted to moulds, and after 120 days, the first wood decaying basidiomycetes were identified. Noll & Jirjis (2012) provide a summary of 10 studies and included the types of ascomycetes, basidiomycetes, bacteria and zygomycetes, which were identified in each of the studies. They concluded from the diversity of the communities revealed in their study that there was no universally common composition of microbes in stored woody biomass piles. Instead, the populations were dependent on the origin of the material and the ability of the microorganisms to survive on the specific substrate under prevailing conditions.

The process of biological degradation of wood by bacteria and fungi is extremely complex. While both are able to use wood as a food source, bacteria are propagated only by cell division and thus can only move in wood where there is liquid water available. They prefer to establish their colonies in parenchyma cells where they can utilize proteins for energy metabolism (Fengel & Wegener, 1983). Wood decaying microorganisms on the other hand, are far more damaging to wood. They have the ability to consume all components of the wood and can rapidly expand into wood by secreting a diverse array of enzymes from their hyphae. The system of enzymes act synergistically to convert the polysaccharides (cellulose and hemicelluloses) in wood to simple sugars, such as glucose or xylose that can then be utilized by the organism for energy metabolism. One class of fungi, known as brown rot fungi and belonging to the subdivision of Basidiomycetes, predominately attack holocellulose and commonly thrive in softwoods. Another class of fungi also belonging to subdivision Basidiomycetes are commonly known as white rot fungi. These organisms have the additional capacity to produce oxidative enzymes that can degrade lignin as well. The white rot fungi are known to prefer hardwood species (Fengel & Wegener, 1983). The microflora populations available within the biomass, the environmental conditions for growth and the substrate will determine the overall level of decay as well as the composition of the material remaining.

Biochemical reactions are the key mechanism responsible for biomass changes, both moisture (addition or loss) and mass loss within a woody biomass pile (Krigstin & Wetzel, 2016). The biological degradation is influenced by three physical, albeit changing conditions within the pile. These are moisture, temperature, and oxygen (gas regime) and to a minor extent nutrients and species of wood. These conditions determine both the type of microorganisms present as well the activity of the microorganisms.

1.1 GAS REGIME

The amount of void space within a pile is governed by an array of factors but dominated by the physical size of the pile, how the pile is constructed and the comminuted size and shape of the biomass particles. The greater the pile height, the greater will be the compaction of the biomass, resulting in lower volumetric air space towards the bottom layers of the pile as compared to the top layer. The compaction is estimated to be 0.6% for every 0.3 m of height (Janzé, 2014). In large piles of over 5 metres in height, there is compaction of the material, which decreases the air spaces between particles and restricts movement of fresh air into the pile and reactant gases out. Pile construction using front-end loaders that repeatedly drive over the pile will also increase pile compaction. Finally, the size of the particles will largely affect the permeability of the pile. It has been suggested that particle size in excess of 100 mm might provide and retain adequate voids for natural convection through a pile (Jirjis, 2005).

Within a biomass pile, the void spaces are occupied with varying concentrations of gases. The concentration of gases within these voids are inclined to change as the biomass degradation proceeds. In addition, the air composition is an important consideration for biological metabolism and organism growth and is central to determining which types or microorganisms can survive. With respect to wood inhabiting bacteria, there are three main categories; aerobic, anaerobic and facultative. Aerobic bacteria require oxygen to survive and use oxygen for the process of energy metabolism. Anaerobic bacteria, on the other hand, generate energy by fermentation and can survive with only 3-5% oxygen concentration in the air. Facultative anaerobes can grow in either the presence of oxygen or in its absence, producing energy both through respiration and fermentation. Bacterial species such as Pseudomonas (aerobic), Bacillus (facultatively anaerobic) or Clostridium (anaerobic) have been identified in wood (Schmidt & Liese, 1994). Cellulose degrading bacteria have been identified in both oxic and anoxic environments and in woody biomass storage situations (Noll & Jirjis, 2012).

Microorganisms growing within a biomass pile are responsible for changing the atmosphere within the void spaces as they metabolize organic matter. Woody biomass contains food for bacteria and fungi in a number of different forms. The preferred form is as simple sugars like glucose and xylose that are readily available in freshly harvested biomass. The general biochemical reaction for aerobic conversion of simple sugars found in parenchyma cells or from hydrolysis of the holocellulose polymers can be summarized by the simplified reaction shown in equation [1]. Thus, for every mole of glucose consumed by the organism, 6 moles of O2 are consumed and 6 moles each of CO2 and water are produced. Hence it is evident that as aerobic decomposition proceeds the temperature of the pile increases, moisture increases, oxygen is depleted, and anaerobic conditions are gradually created where methanogenic microbial communities can flourish (Ferrero, Malow, & Noll, 2011;

Whittaker et al., 2017). It should be noted that in practice not all glucose will covert to CO2 and water but a large proportion (40%) of consumed cellulose is processed to intermediate metabolites for the production of fungal biomass (Schmidt, 2006). Significant amounts of CO2 are produced by fungi. Polyporus vaporaria grown on pine produced 1.98 g per kg wood per day (S. G. Cartwright & Findlay, 1934). Anaerobic conversion of woody material consists of a number of steps depending on the microorganism and environment. Biochemical reactions include acetogenesis and methanogenesis (equations [2] & [3]) with approximately 70% of methanogens using acetate as a substrate, while others use carbon dioxide and hydrogen (Ritchie, Edwards, McDonald, & Murrell, 1997). In natural environments, methanogens are found deep in the soil profile but have also been found in the heartwood of decomposing trees (Wang et al., 2016).

Cellular respiration:	$C_6H_{12}O_6 + 6O_2 \rightarrow 6CO2 + 6H_2O$	$\Delta G0' = -2805$ kJ/mol	[1]
Acetogens:	$C_6H_{12}O_6 \rightarrow 3CH_3COO^- + 3H^+$	$\Delta G0' = -310.9$ kJ/mol	[2]
Methanogens:	$CH_3COO- + H+ \rightleftharpoons CH_4 + CO_2$	$\Delta Go' = -36$ kJ/mol CH_4	[3]

The dynamic nature of the microorganism communities is illustrated in Whittaker et al. (2015) where monitored concentrations of CO2, CH4 and N2O at depths of 1m and 3m within a short rotation coppice willow chip pile saw increase of CO2 concentration early in the storage period followed by a peak in CH4 concentration some time later (Whittaker, Yates, Powers, Misselbrook, & Shield, 2016) . This illustrates that the change in the microbial community over the course of the storage period from aerobic to predominately anaerobic, due to the changing ambient gas concentrations.

Other studies have shown that wood degrading fungi can produce methane under aerobic conditions (Guenet et al., 2012; White & Boddy, 1992). In work by White and Boddy (1992), wood degrading fungi Phlebia rufa and Phlebia radiate (cultured in reduced oxygen environment (5% v/v) with 20% CO2) showed slightly lower extension for the colony than observed under a normal atmospheric regime, however certain levels of CO2 at fixed oxygen content (5%) did show improved growth. For Coriolus versicolor the opposite was observed, with decreased extension rate as CO2 concentration was increased at 5% O2 level. Other fungi, such as Merulius lacrymans, was entirely inhibited in an atmosphere of 25% CO2. Certain reactions, such as the degradation of lignin by white rot fungi require oxygen so these fungi are not expected to function in an oxygen depleted atmosphere. Not only can wood rotting fungi function over a range of toxic concentrations, there have also been a large number of organisms identified that are capable of remaining viable under anaerobic conditions for at least one month (Kurakov, Lavrent'Ev, Nechitailo, Golyshin, & Zvyagintsev, 2008). It is of interest to note that fungi which commonly cause heart rot are more tolerant of a reduced oxygen environment (K. G. Cartwright & Findlay, 1958).

The survival of microorganisms within a biomass pile may be limited by one or a combination of factors. By monitoring CO2 and CH4 emissions from a biomass pile, Whittaker et al. (2016) showed that the emissions reached a moderate baseline at around 60 days and did not increase even once temperature had fallen back to mesophilic levels (Whittaker, Yates, et al., 2016). This may suggest that anaerobic organisms, which metabolize much slower and produce less energy, are the only ones surviving as the availability of easily degradable sugars, amenable oxygen levels, and possibly other factors such as acidity or nitrogen availability are not encouraging rapid growth (and increasing temperature).

Gaseous emissions

Gaseous emissions from stored biomass is related to losses in dry matter (He et al., 2012). Typically the gases produced from stored woody biomasses are CO, CO2, CH4 and other volatile hydrocarbons e.g., aldehydes and terpenes (Alakoski, Jämsén, Agar, Tampio, & Wihersaari, 2016). Volatile Organic Compounds (VOC) includes all organic compounds with a boiling point between 50-240°C, or 100-260°C for polar compounds (Rupar-Gadd, 2006). This property limits the size of these compounds to a maximum of 12 carbon atoms (Alakoski et al., 2016). The mechanisms of formation of CO, CO2 and CH4 from woody biomass storage is at present not entirely clear (K. M. Granström, 2014; Whittaker, Yates, et al., 2016).

The respiration of living wood cells and microorganisms means consumption of atmospheric oxygen and production of carbon dioxide and water (Assarsson, Croon, & Frisk, 1970). Carbon dioxide (CO2) is also formed e.g., in thermal oxidation, aerobic biodegradation or anaerobic biodegradation of organic material (Alakoski et al., 2016). For instance, the breakdown of carbohydrates with oxygen consumption may be represented by the reaction C6H12O6 + 6O2 → 6CO2 + 6H2O (He et al., 2012). It is also possible that wood stacks undergo composting, as they contain some readily available carbohydrates that can be fermented to lactic acid, volatile fatty acids and alcohols with the release of carbon dioxide and heat (Whittaker, Yates, et al., 2016). CO and CO2 emissions and oxygen depletion have also been observed during transportation of wood chips and logs (Whittaker, Yates, et al., 2016).

Carbon dioxide has been found to accumulate in piles of sawdust to form as much as 20% of the atmosphere of the pile, which is 700 times more than in the fresh air (Kubler, 1987). Whittaker et al. (2016) studied the dry matter losses and the GHG emissions within two short rotation coppice (SRC) willow wood chip storage heaps. In the study by Whittaker et al. (2016), carbon dioxide was the only GHG present in appreciable quantities, as also indicated in the pine chip storage study by Ferrero et al. (2011) (Ferrero et al., 2011). This suggests that aerobic processes dominated. The freshly cut pine wood (approx. 400 tons of fresh weight) in the study of Ferrero et al. also contained bark but no needles and branches.

Jylhä et al. (2017) studied CO2 release from stockpiled whole-tree chips and chips made from delimbed, unbarked stemwood. Both chips were made of small-diameter Scots pine. They found that stockpiled whole-tree chips emitted more CO2 and lost their dry matter more rapidly than chips made of delimbed material. This happened in spite of the initially lower moisture content and greater median particle size of whole-tree chips. The first few weeks were crucial in terms of CO2 emission and dry matter loss. In addition to the type of raw material used for the chips, the magnitude of CO2 emissions also depended on storage duration and the temperature within the stockpile. Any lowering of temperature within the stockpile was followed by a decrease in CO2 emissions. During the six-month storage period, the estimated CO2 emissions amounted to 9.3–10.8 kg per solid cubic meter of wood. In the case of whole-tree chips, over three-quarters (77%) of the total emission was emitted during the first storage month. Reaching the same proportion of pile-specific CO2 emission took twice the time for the stemwood chips. According to Jylhä et al. (2017) storing of Scots pine forest chips for only one month would cause a CO2 release of about 4.8–8.3 kg per a solid cubic meter.

Carbon monoxide (CO) is formed from incomplete combustion processes, from sluggish decomposition of organic material, and from the autooxidative degradation of wood lipids and fatty acids (Alakoski et al., 2016; He et al., 2012). Temperature is one of the critical factors in autooxidative degradation. It is also suggested that CO generation is independent of microbial activity in the feedstock, but is promoted by increased temperatures and available oxygen. Ferrero et al. (2011) detected carbon monoxide concentrations of approx. 100 fold the atmospheric concentration within the heap. As for CO_2 and CO, the higher values were achieved in the center of the heap and were smaller on the boundaries. This indicated occurrence of the incomplete oxidations which are typical of stored bulk material. However, the concentrations of CO were still in the parts per million by volume (ppmv) range meaning that the oxidations were negligible in comparison with the aerobic biological processes. Thus it seems reasonable that CO concentrations have been measured to be significantly lower from wood chip and timber transportation than from the pellet transportation (Alakoski et al., 2016).

CH_4 generation is usually due to anaerobic decomposition of biomass due to the action of microorganisms (Alakoski et al., 2016; He et al., 2012). Methane production usually occurs after O_2 has been depleted by aerobic processes, and when CO_2 production and temperatures are high (Whittaker, Yates, et al., 2016). Compaction is also known to contribute to CH_4 production. Whittaker et al. (2016a,b) detected a peak in methane concentration (around 400 ppm) in willow wood chip heaps after around 55 days. In both cases, the peak CH_4 concentration occurred as CO_2 concentration dropped. This suggested that after an active period of aerobic decomposition in the first 2 months of storage the conditions in heaps became anaerobic. In a study by Ferrero at al. (2011) methane was detectable only in the ppmv-range which is a sign of the almost total absence of anaerobic processes. They monitored the pile for a period of 150 days. In biological systems containing CO_2, O_2 and CH_4, it is possible that CH_4 is oxidized by methanotrophic bacteria to H_2O and CO_2 (Whittaker, Yates, et al., 2016).

Volatile organic compounds

Monoterpenes are the dominant volatile organic compounds emitted from fresh spruce and pine wood (Alakoski et al., 2016). All machining, tooling and shaping of biomass is likely to cause monoterpene emissions (K. Granström, 2005). Activities that cause anthropogenic terpene emissions include e.g., logging, chipping, debarking, sawing and storage. During roundwood storage, terpenes are slowly released into the air (Strömvall & Petersson, 2000). Chipping and long storage of wood chips increases terpene losses remarkably. Most of the terpenes are emitted in the first few weeks of storage. More than half of the terpene content may be lost during chip pile storage for a few weeks. These losses occur mainly by evaporation to air. However, microbial activity increases losses primarily by increasing the chip pile temperature. Especially in summer terpenes are released fast from chip piles.

Sesquiterpenes are also naturally emitted into the air (Strömvall & Petersson, 2000). Their boiling points are approximately 100°C higher than the boiling points of monoterpenes. The low vapour pressure at ambient temperature may be enough for evaporation from the exudate e.g., when distributed over a wound. However, monoterpenes are more easily released to air than sesquiterpenes, but the sesquiterpenes are more reactive in the atmosphere and are more rapidly converted from the gas phase to liquid phase in the form of polar aerosols, due to their higher molecular weight.

Terpenoids include a wide range of terpene derivatives, such as ethers, alcohols, aldehydes, esters, and carboxylic acids. They contain oxygen and have a lower vapour pressure and more soluble in water than the terpene hydrocarbons. This means that they are less volatile, and since they are usually present in small amounts, they cause low emissions to air from the industrial use of wood.

More terpenes have been found to be released if wood chip piles stored outdoors were mixed with bark during storage, especially when the amount of precipitation increased (Rupar & Sanati, 2005).

Jirjis and Andersson (2005) conducted a field trial lasting 7 weeks in April-May 2003(Jirjis, Andersson, & Aronsson, 2005). The pile consisted of a mixture of 50% pine bark and 50% spruce bark which was delivered directly from the sawmills after debarking. About 250 m3 of each bark variety was used for pile construction and the materials were well mixed with an excavator before construction. Bark piles give rise to a certain emission of VOC's and especially monoterpenes. With the repeated measurements of total volatile organic compounds (TVOC) in the air vented from the pile via the flow chamber, a rapid and sharp decrease of TVOC over time appeared. After 2 weeks, the level had fallen by 95% from the initial concentration, and in the following 4 weeks, the level further decreased to 0.3% of the original. The concentration of the identified monoterpenes, increased between day 2 and day 6, and then decreased sharply. In addition to the first measurement, the monoterpenes generally represented a very high proportion of measured TVOC. At the first sampling, the measured TVOC content was dominated by xylene and toluene. The rapidly declining concentrations indicates that the gassing stops after two weeks.

Based on the measured values and calculations, 5 g of monoterpenes and 12 g of TVOC were emitted per square meter during the first 6 weeks. Assuming that the bark pile with the bottom surface 150 m2 had a volume of 500 m3 and that the basic density was 160 kg m-3, this corresponds to 10 mg kg-1 bark (dry weight) monoterpenes and 20 mg kg-1 bark TVOC. Emissions from spruce bark were dominated by the α- and β-pinenes while the α-pinene and 3-carene dominates emissions from pine. This is also often the case with emission from the wood of these species. Results pointed out that the main emissions of terpenes occurs during the first 2-3 weeks.

It is also suggested that there are emissions of nitrous oxide from wood chip stacks (Whittaker, Macalpine, Yates, & Shield, 2016). These emissions would result from the activity of nitrifying of denitrifying bacteria that utilize nitrogen derived from bark, cambium and foliage. There are estimations that the emissions of N2O-N can be between 0.5 to 0.7% of the total initial nitrogen present in biomass. As nitrifying bacteria is sensitive to high temperatures (> 40 °C), no high emissions of N2O can be expected (Whittaker, Macalpine, et al., 2016; Wihersaari, 2005). Nitrous oxide is formed either initially, before temperature increase, or after the thermophilic phase when temperature is low again.

He et al. (2012) studied dry matter losses and gaseous emissions from stored fresh logging residues at laboratory scale. The released NMVOCs (Non-methanous VOC's) they identified included alcohols, terpenes, aldehydes, acids, acetone, benzene, ethers, esters, sulphur and nitrogen compounds. They also observed that the gas concentrations increased significantly at higher temperature of 35 °C compared to lower temperature of 15 °C. This effect was especially notable in case of CO. The oxygen level decreased to 0% at the end of storage period 35 days. Hexanal from fatty acid oxidation has been found to be an important volatile compound during the storage of solid wood fuels (Laitinen et al., 2016). Wihersaari (2005) found that the GHG emissions were almost three times higher for fresh versus dried forest residues.

The dominating compounds released from softwood pellets during storage are terpenes, aldehydes, CO, CO2 and CH4 (K. M. Granström, 2014). Also emissions of e.g., alkanes, alkenes, ketones, alcohols and organic acids have been detected (Alakoski et al., 2016). Aldehydes are formed from unsaturated lipid compounds such as resin and fatty acids through autooxidative chemical reactions (K. M. Granström, 2014). The fats such as triglyceridesin wood can hydrolyze (so called lipolysis) to form free unsaturated fatty acids. Free fatty acids are more susceptible to oxidation than the fatty acids esterified to glycerol. Oxidation of unsaturated fatty acids is a complex self-catalysing free radical chain reaction started by free radicals, which can be produced by light photons or by metal ions or by the spontaneous reaction of oxygen with a material with a readily abstractable hydrogen. The radicals cause free fatty acids to become hydroperoxides which are unstable and start to decompose soon after they are formed. Hydroperoxides break down to one alkoxy radical and one hydroxyl radical, which enter into numerous complex radical reactions ending with a myriad of different hydrocarbons. When the oxidation has started, it self-catalyzes and continues until all the radicals have been neutralized. One of the most notable compounds emitted from the oxidation of fat is hexanal, and thus it is a reliable indicator of lipid oxidation. Acetone and aldehydes such as butanal, pentanal, hexanal, heptanal, octanal and nonanal are also emitted. The problems with fatty acid oxidation products are more prevalent for pellets produced from Scots pine as compared to the pellets produced e.g., from Norway spruce. This is due to the remarkably higher concentration of fatty acids in pine.

Another key conclusion in Granström's study was that the sawdust age should be taken into consideration when assessing the risk of aldehyde emissions from pellets (K. M. Granström, 2014). She found out that the pellets made from the fresh Scots pine sawdust were low-emitting after 80 days, whereas the pellets made from the aged sawdust did not reach the same level until after 190 days of storage. The aged sawdust pellets had maximum emissions at the same time as the emissions ceased from the fresh sawdust pellets. All pellets had a peak emission of about 30 mg/kg dry substance (DS) (except fresh sawdust pellets stored at elevated temperature). The majority of the studies examining the GHG emissions from wood pellets during silo storage have found that CO2 emissions are the greatest and CH4 emissions the least (Whittaker et al. 2016). Storage temperature has been found to be the key factor affecting the gaseous emissions from wood pellets (Alakoski et al., 2016). Increase in temperature enhances gaseous emissions. It seems that off-gassing from pellet storages is mainly the result of chemical processes.

1.2 TEMPERATURE

Temperature is an important environmental factor which influences both the growth rate of fungi as well as the predominance of certain species. Fungi, as with most green plants, favour a moderate temperature for their growth. Generally, the optimum growth for most wood decaying fungi is between 20-32°C with the minimum generally believed to be about 10°C and maximum just below 40°C, thus falling into the category of mesophilic organisms. Research has shown that there appears to be differences in optimal temperature ranges for white and brown rot fungi, with brown rot fungi's between 30-35°C and white rot somewhat lower, at 20-30°C (Fukasawa, 2018). The temperature range and optimum growth rate of many individual wood destroying fungi are available in literature. A study published in 1934 measured the growth rate (on agar) of a number of important Basidiomycetes known to grow on wood (S. G. Cartwright & Findlay, 1934). A chart prepared from this work illustrates the range of temperatures for the optimum growth rates of three fungi, and also shows the range over which they will survive (Fig. 1). It is evident that temperature plays a large part in determining which species will dominate and how much decay will take place.

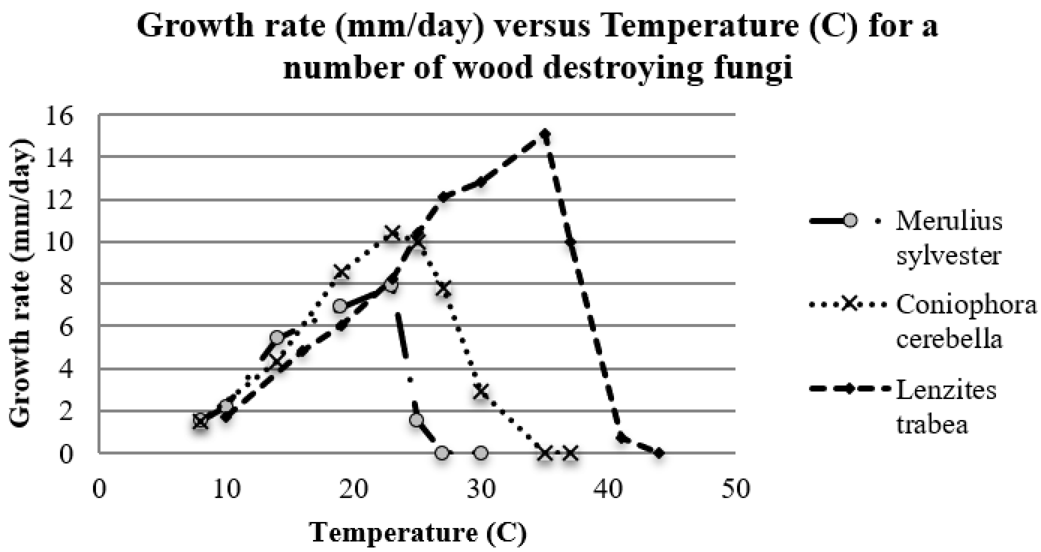

Fig. 1. Growth rate (mm/day) versus Temperature (oC) for three wood destroying Fungi. Chart derived from Cartwright and Findlay, (1934).

Important to woody biomass storage are thermophilic fungi. This is a group of fungi that require a minimum of about 20˚C, and continue to grow well above 50˚C. In Thambirajah & Kuthubutheen (1988) it has been reported that thermophilic bacteria will tolerate even higher temperatures than thermophilic fungi (Thambirajah & Kuthubutheen, 1989). Maheshwari et al., (2000) provide a table identifying 29 unique fungal species with optimal growth rates varying between 35-55oC and maximum temperatures of up to 61oC. It has long been theorized (Chang & Hudson, 1967; Maheshwari, Bharadwaj, & Bhat, 2000; Thambirajah & Kuthubutheen, 1989) that in self-heating situations, such as in wet hay or compost, an initial microbial community establishes itself and through its exothermic reactions causes the temperature to rise over 40˚C. This warm environment favours germination and growth of a thermophilic microflora, reduced activity and the demise of less thermally tolerant organisms. Due to the relatively low thermal conductivity of compacted biomass, the heat build-up inside biomass piles is relatively fast and hence after an initial heat up period it can be conjectured that the thermophilic microflora dominate. As the maximum survival temperature for the thermophiles is surpassed, they too will cease to grow and heat generation within the pile will diminish. Fungi can survive excessively low temperatures however high temperatures, even for a short time, can be lethal. It has been shown that fungi in wood heated at 65˚C for a minimum of 75 minutes will be killed (Scheffer, 1973). Eventually thermophilic and even mesophilic organisms surviving in the outer, cooler regions of the pile might recolonize inwards as the temperature falls below each fungus' upper temperature range. An excellent study of the change in fungal populations over decomposition of wheat straw in relation to temperature can be found in Chang & Hudson (1967), Fig. 2. The figure shows the predominance of mesophilic over thermophilic fungus early in the composting process. Then, once the temperature reaches its maximum (70˚C) all fungi die off completely. The thermophiles begin to recolonize once temperature falls below about 65˚C and continue to expand as the temperature in the compost declines. The mesophilic do not begin to recolonize until the temperature in the compost falls below 50˚C. Both types of fungi appear to reach a steady state level about 8 days after recolonization begins.

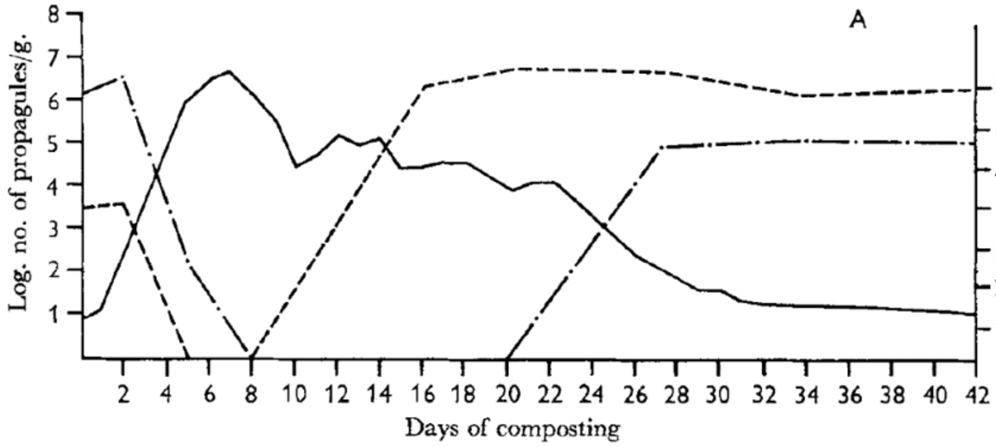

Fig. 2. Fungal populations in wheat straw compost. The solid line represents temperature; dashed line is thermophilic fungi, and dash-dot line represents mesophilic fungi. (Chang & Hudson, 1967).

Thermophilic bacteria growth rates are also influenced by temperature. In fact, the thermophiles show a very high rate of multiplication at the higher temperature range of the compost (Figure 3). The growth of the mesophilic is somewhat reduced at the highest temperature but the thermophilic bacteria, with their higher maximum growth temperature continue to thrive (Chang & Hudson, 1967). The bacteria show a much higher population growth compared to fungi during the initial heating stages of the compost, which suggests that it may be the bacteria that contribute more to the initial rise in temperature than the fungi community does.

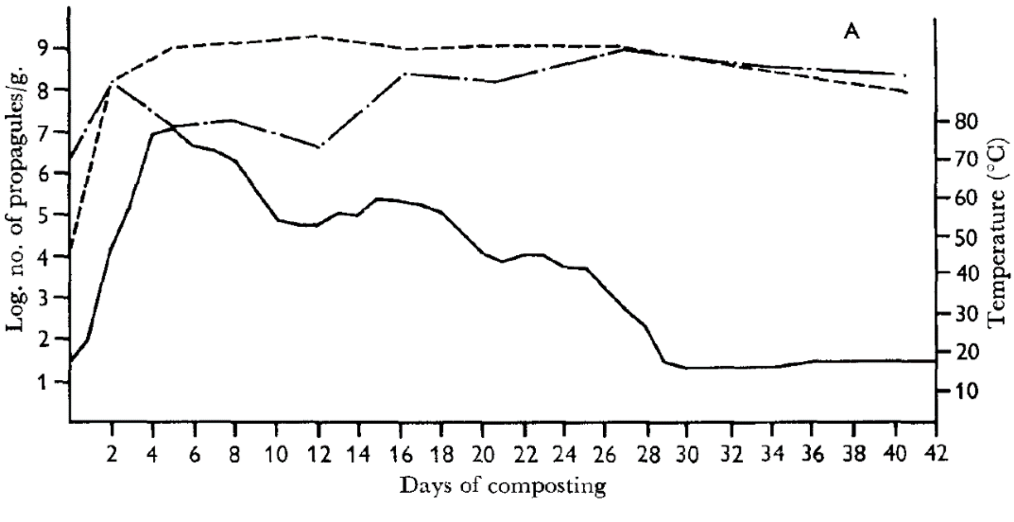

Fig. 3. Bacterial populations in wheat straw compost. The solid line represents temperature; dashed line is thermophiles, and dash-dot line represents mesophilics. (Chang & Hudson, 1967)

1.3 MOISTURE

A certain level of moisture in woody biomass is essential for fungal activity. Freshly chipped biomass has very high moisture content, especially if harvested in late spring or summer. During this time, the tree is actively transporting water from the roots to the leaves for photosynthesis. There is much variability in moisture within a tree however. In some species, the moisture content of green sapwood is higher than the heartwood; however, the reverse can also be true. Softwoods in general have a very large difference in moisture content between sapwood and heartwood (for Eastern spruce: sapwood ~56%, heartwood ~25%) as compared to hardwoods (for Aspen: sapwood ~53%, heartwood ~49%) (Simpson & TenWolde, 1999). Recent studies involving storage of freshly chipped biomass materials reported 56.4% for SRC willow chips (Whittaker, Yates, Powers, Misselbrook, & Shield, 2018), 48%-57% for spruce wood chips (Hofmann et al., 2018) as an example.

All wood decay fungi and bacteria require moisture for degradation to occur. The optimal condition for fungal degradation is when the cell walls are saturated with water (bound water) and there is a layer of free water in the cell lumen, also known as the fibre saturation point (FSP) (Kollmann & Côté, 1968). The void space in the lumen allows for gas exchange and the layer of water allows for movement of the enzymes into the cell wall. The moisture content at FSP is approximately 23-30%, measured as mass of water / total mass. At moisture levels above FSP the lumen will partially fill with liquid water through capillary action. If the entire lumen is filled with water, then there will be insufficient space for gas exchange to occur and the fungi cannot function. Some fungi can survive in wood when moisture content is less than FSP however, and some can even remain viable yet inactive for up to 3 years (Scheffer, 1973). The fungi will resume their activity once favourable moisture conditions return. Different fungal organisms have various moisture requirements for optimal growth. It has been suggested that white rot fungi may require more moisture than brown-rot fungi (Scheffer, 1973). The optimal environment for fungal growth depends on a balance between moisture and air requirements, which is likely species dependent. Optimum moisture ratio (%u) for some common species are as follows: Phlebiopsis gigantea, 100-130%, Coniophora puteana, 30-70% (Schmidt, 2006). This balance varies by the density and cell structure of the material.

Water serves a number of important functions in the wood degradation process. As mentioned earlier, it is the medium by which the hyphae secreted enzymes are transported to the cell wall surface and the solubilized molecules are transported back to the microorganism. It is also one of the reactants used in the enzyme-catalysed hydrolysis reaction that breaks down the glycosidic bonds between adjoining 5 or 6-carbon monosaccharides in the cellulose or hemicellulose polymers. And finally, it serves to swell the micro capillaries within the cell wall, which enables more rapid penetration of the fungal digestive enzymes into the substrate (Zabel & Morrell, 2012).

Fungi and bacteria produce moisture during their metabolic processes, and therefore as biomass degrades under storage, it will actually become wetter. It has been demonstrated that from 1 m3 of wood degraded to 50% of its original mass, the cellulose portion alone produces 139 L of water (K. G. Cartwright & Findlay, 1958). It is expected that the water vapour, being a condensable gas, will condense on the biomass and be available for capillary absorption back into the biomass. This allows the fungus to continue to grow without requiring a surplus of water.

1.4 NUTRIENTS

The nutritional requirements of wood decaying microorganisms do vary but woody biomass seem to provide the minimum requirement for most. Nitrogen is a fundamental requirement of microorganisms for production of enzymes and cell material but the sparse amount naturally occurring in wood seems to be adequate. Nitrogen in woody biomass may be augmented by the fungi's ability to solubilize nitrogen in the protoplast of older hyphae and translocate (Scheffer, 1973), or by diastrophic bacteria, such as Pseudomonas , which live in wood chip piles (Noll & Jirjis, 2012). Nitrogen content in forest biomass (0.7% daf) is higher than in wood (poplar, 0.6% daf; pine chips, 0.5%) (Vassilev, Baxter, Andersen, & Vassileva, 2010) alone since most forest residues contain a certain amount of foliage, which generally contain a high nitrogen content.

FINAL THOUGHTS

Biological degradation occurring within stored woody biomass piles is extremely complex and ever shifting. There is a complicated co-dependence of temperature, atmospheric composition, substrate chemistry and moisture in determining the microflora composition, which in turn will affect all of the factors mentioned. Therefore, within any given biomass pile, there may be anaerobic, aerobic, mesophilic, thermophiles, bacteria and fungi all existing within their suitable environment. Beyond influencing the type of microorganisms present, the environmental conditions will influence the rate of decay (dry matter loss) and the moisture content of the biomass. Knowledge of the fundamental aspects of biological degradation within a biomass pile structure is key to obtain the quality of biomass required by the end user.

Microbiological decay of wood in storage causes potential for several negative consequences including self-heating leading to ignition, loss of dry matter and excessive moisture. However, with an understanding of the biological decay mechanisms and conditions for growth in storage, there is potential to design the decay process for beneficial changes to heterogeneous forest residue biomass, making it more homogeneous from a chemical perspective. It is well documented that wood decay will increase the porosity of the cell wall which can lead to improved penetration of chemicals (for pulping) and reduce the mechanical energy needed for refining wood, (López, Silva, & Santos, 2017; Reinprecht, Solar, Geffert, & Kacik, 2007; Scott, Akhtar, Swaney, & Houtman, 2002). A semi-commercial scale process developed in conjunction with the US Forest Services has shown that through control of the conditions and inoculation with specific delignifying fungi, TMP pulp manufacturing cost savings can be achieved (i.e. US$258/ton versus US$278/ton) (Scott et al., 2002). Similarly, long-term storage of woody biomass could potentially be the first step in the bio refining process where designer decayed material gives a cost advantage at the next step of the refining process.

Dry matter losses in the biomass supply chain

2.1 UNPROCESSED FOREST BIOMASS

Storing forest biomass outdoors can cause substantial dry-matter losses where degradation rates can vary depending on climatic conditions and weather regimes. For logging residues, average dry matter losses of 1-3% per month have been reported (Filbakk, Høibø, Dibdiakova, & Nurmi, 2011; Golser, Pichler, & Hader, 2005; Jirjis, 1995; Jirjis & Norden, 2005; Nurmi, 1999; Pettersson & Nordfjell, 2007; Routa, Kolström, Ruotsalainen, & Sikanen, 2015a). Pile heating and decomposition alike in chip piles are reported for logging residues piles (Golser et al., 2005).

Pre-dried logging residues show lower dry matter losses (Filbakk et al. 2011, Routa et al. 2015a) (Fig. 4.). Flinkman et al. (1986) reported that loss of forest residues stored in heaps on the harvest site could be attributed to needle loss (Flinkman, 1986). Nurmi (1999) calculated the dry matter loss of needles stored at the harvest site versus those transported to landing for storage. Needle composition fell from 27.7% to 6.9% for the biomass left at the harvest site compared to 18.9% for biomass stored on the landing. Since needles are nutrient-rich, leaving a high proportion of needles at the harvest site is always advisable to sustain tree regeneration. Defoliation at the harvesting site is therefore beneficial both in terms of drying performance and for the forest nutrient cycle (Nurmi, 1999; Routa et al., 2018; Suadicani & Gamborg, 1999). In addition, compared to other tree parts, needles contain higher amounts of chloride, which is known to increase corrosion inside power plant boilers. Lastly, the retention of green needles and increased moisture can stimulate fungi and bacteria growth during storage further supporting defoliation at the harvesting site.

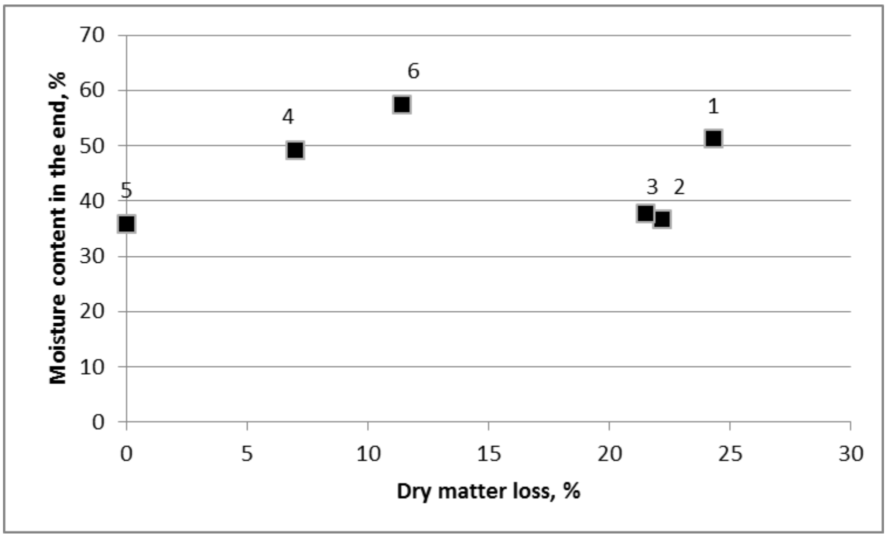

Fig. 4. Moisture content at the end of the experiment and dry matter loss during storage (8 months) in the different study piles of logging residues (Routa et al. 2015a). The lowest moisture content and no dry-matter losses at all was observed from the pile, which was pre-dried to 20% before piling to bigger storage pile (pile number 5), even the moisture increased to 36% during 8 months storing.

Covering piles (e.g. with a paper based cover) is recommended, especially when precipitation levels are high or prolonged (e.g. In Scandinavia or Scotland) for both logging residues and stem wood (Filbakk, Høibø, & Nurmi, 2011; Jirjis, 1995; Nurmi & Hillebrand, 2007; Pettersson & Nordfjell, 2007; Röser et al., 2010). No cover is necessary for stem wood where conditions are dry (such as in Italy) (Röser et al., 2010), but it probably could be beneficial in the alpine regions (Elber, 2007; Erber et al., 2012; Golser et al., 2005). The overall effect of covering piles is considered to be an additional 3–6% reduction in moisture content compared to uncovered piles (Jirjis, 1995; Nurmi & Hillebrand, 2007).

2.2 WOOD CHIPS

During the storage of coniferous wood chips, monthly dry matter losses between 0.3 and 5.5% are reported (Anerud, Jirjis, Larsson, & Eliasson, 2018; Bergman & Nilsson, 1971; Heding, 1989; Heinek et al., 2013; Hofmann et al., 2018; Juntunen, Hirvonen, & Paukkunen, 2013; Jylhä, Hytönen, & Alm, 2017; Mitchell, Hudson, Gardner, & Storry, 1988; T. Thörnqvist, 1985). Generally, half of the losses occurring during the first months consist of low molecular carbohydrates, resins, acetic acid etc. (Assarsson et al., 1970). The decomposition rate in whole-tree or forest residue chip piles has been found to be higher than in stemwood chip piles (Hofmann et al., 2018; Jylhä et al., 2017; Thörnqvist, 1985). A high green and fine content in forest residues offers a large surface area for microbial attack and many easily available nutrients for microorganisms.

In piles of poplar, alder and willow chips, dry matter losses of up to 4.4% per month have been reported (Barontini et al., 2014; Scholz, Idler, Daries, & Egert, 2005; Whittaker et al., 2018). In a Canadian trial with woodchips from birch, monthly dry matter losses ranged from 0.7 to 2.3% (Afzal, Bedane, Sokhansanj, & Mahmood, 2009). To date, the storage behaviour of other deciduous wood chips has not been investigated in depth.

2.3 BARK

The physical structure and chemical composition of bark differs from wood considerably. In general, bark has higher proportion of parenchyma cells, implying higher store of easily accessible sugars, which gives rise to higher and longer respiration period and causes greater heat generation. Bark may also be more susceptible to fungal invasion, as studies have found that parenchyma facilitate the spread of fungi (Schwarze, 2007). According to Thörnqvist (1985), piles containing bark compared to those that were bark-free, contained more fungal spores and exhibited increased degradation. In addition to structural differences, there are significant chemical differences between wood and bark where bark typically contains higher concentrations of lignin, extractives and inorganic metals (P. Lehtikangas, 2001). The bark of certain tree species (e.g. Norway spruce) can also contain antifungal agents which help to resist fungal invasion during storage; Hammerbacher et al, 2013). In a Swedish study of coniferous bark, dry-matter losses around 5-10 % were reported during a 2-5 months storage time (Fredholm & Jirjis, 1988; Lehtikangas & Jirjis, 1998) (Fredholm & Jirjis, 1988; Lehtikangas & Jirjis, 1998).

2.4 PROCESSED FOREST BIOMASS

Pellets

Sawdust supply fluctuates according to sawmill activity, which makes it a common practice to store sawdust up to several months at pellets plants, generally in large heaps outdoors (K. M. Granström, 2014). Produced pellets are also stored due to seasonal demand patterns.

Energy content losses/changes and chemical form of matter losses

Heating value is the amount of heat produced by a complete combustion of fuel. The heat of combustion of fuels is expressed by the higher and lower heating values (HHV and LHV). The higher heating value is also known as the gross calorific value. The HHV is an indicator of the value of a material as a direct energy resource; however, the moisture content of biomass has a marked influence on its usable energy. The actual usable energy in a fuel is often referred to as the net heating value (NHV). Moisture gain or loss in the biomass affects its NHV, and therefore storage can have a significant impact on this value. A change in calorific value occurs when the different wood components do not decompose according to their specific calorific values. For example, lignin has a distinctly higher calorific value than cellulose (Kaltschmidt, Hartmann, & Hofbauer, 2009), but usually decomposes at a lower rate than hemicellulose and cellulose. Therefore, in most studies concerning the storage of wood chips, no major changes in the net calorific value are measured (Afzal et al., 2009; Jirjis, 2005; Lenz, Idler, Hartung, & Pecenka, 2015; T. Thörnqvist & Jirjis, 1990a).

In the study by Reisinger and Kluender (1982), energy content of the whole tree chips decreased and continued to decline for approximately 4 months (25% loss of heating value) after which losses were negligible (Reisinger & Kluender, 1982). Mitchell et al. (1988) studied percentage changes in net calorific value (NCV), including both dry matter loss and moisture change, reporting that covered chips and chunk wood had minimal losses in NCV (1.2% and 0.8%) while uncovered chips had 5.0% and 2.3% losses over the 6 month trial period (Mitchell et al., 1988). Juntunen et al., (2013) reported an energy loss of 28% in forest chips following 6 months of storage (Juntunen et al., 2013). According to Thörnqvist, an energy loss of 6-23% occurs during comminuted forest residue storage over 3–9 months, depending upon particle size, initial moisture content, proportions of foliage, bark, and wood (Thörnqvist, 1984, 1985). A study by Routa et al (2018) found that the heating value of uncomminuted stem woodpiles increased in 64% of the piles depending on storage period, species and moisture content. Furthermore, according to Acquah et al. (2015), the change in NHV during storage was strongly influenced by the position of the material in the pile (Acquah, Via, Fasina, & Eckhardt, 2015). The material on the outer layer showed an increase in NHV of 30% and 48% whereas the inner layers showed a decrease of 59% and 68% over 1 and 2 years of outdoor storage.

Overall, the energy quality of biomass is not uniform and can be positively or negatively affected by storage depending on the duration and conditions of the storage trial. Reduction of moisture is obviously a positive change which increases the material's NHV, increase its energy density, and hence reduce the unit energy cost for transportation. Depending on the nature of the changes to the chemical characteristics of the biomass during storage, changes may increase the material's HHV.

Factors affecting the loss of valuable extractives

The extractives contained within woody biomass begin to degrade immediately after tree felling and this degradation continues throughout storage (Alén, 2000; Assarsson & Cronn, 1963; Ekman, 2000; Jirjis & Theander, 1990). The chemical composition of the extractives-based fraction changes gradually, however a few months of outdoor chip storage can decrease the extractives content by 25-75%. Other phases of the wood handling process can have an effect on the extractives content and composition of the extractives fraction as well. The nature and rate of change in the properties of wood resin are determined by several factors (Ekman, 2000; Rupar & Sanati, 2005) including harvesting method, transportation and the inventory-control systems used at the mill site. They also depend on the tree species, type of material, age of the material, time in storage, physical form of the wood, weather and other environmental conditions in all phases of the wood-handling process.

The major chemical changes in the resin during wood storage can be divided into three types: (1) rapid hydrolysis of triglycerides accompanied by slower hydrolysis of waxes, especially steryl esters, (2) oxidation/degradation/polymerization of resin acids, unsaturated fatty acids and to some extent other unsaturated compounds, and (3) evaporation of volatile terpenoids, mainly monoterpenes. Esters (fats and waxes) are enzymically hydrolysed to fatty acids and alcohols (Assarsson, Croon & Frisk 1970). Fatty acids and, to some extent, higher alcohols are metabolized by respiration to carbon dioxide and water. These reactions might take place either in living wood cells or be caused by moulding fungi. Unsaturated extractives easily react with atmospheric oxygen in autoxidation reactions. These proceed via radical reactions. The rate of all these reactions increases with increasing temperature (Ekman, 2000). Transition metal ions and light generally accelerate auto-oxidation reactions (Alén, 2000). For example, according to Zahri et al. (2007), UV light induces the degradation of phenolic compounds present in oak extract (Zahri et al., 2007). It is also known that stilbenes are sensitive to daylight. Increasing ventilation, and thereby increasing the access of air and oxygen in the chip pile, further speeds up evaporation and oxidation reactions (Ekman, 2000). Some extractives are water-soluble (hydrophilic), which means that both rainfall and water added before debarking at the mill will leach some compounds of extractives from the biomass. Different tree species contain varying concentrations and types of water-soluble compounds in their wood and their leaching rates vary as well (Hedmark & Scholz, 2008). The compounds of extractives that are generally found in woodyard runoff include phenolic compounds, resin acids and short chain fatty acids. According to Rupar and Sanati (2005), there seems to be a correlation between the amount of precipitation and the emission levels of terpenes into the air (Rupar & Sanati, 2005). They also concluded that this phenomenon is more obvious for bark and wood chips than for forest residues since bark and wood chips are more affected by precipitation owing to the smaller particle size of the material, and for bark, owing to the porosity of the material. Evaporation of volatile terpenes is faster for more porous material (Rupar & Sanati, 2005) .

Outdoor/indoor storage and seasoning in water are all utilized when supplying wood to the mills. Environmental storage conditions (e.g. temperature and precipitation) as well as the duration of storage post-harvest are all important factors, which determine the remaining amount and composition of wood resins after the storage period. It is well known that the hydrolysis of glycerides leading to free fatty acids and glycerol proceeds faster when the conditions for wood storage are wet as opposed to dry (Alén, 2000). Conversely, water and a high moisture content of wood protect it from damage caused by fungi or insects (Ateş, Pütün, & Pütün, 2006). This is particularly important during the storage of wood logs in water in the summer (Alén, 2000). During seasoning, there is an increase in the amount of fatty acids and a reduction in neutral resin components (i.e. hydrocarbons, waxes, glycerides and higher alcohols) which is ascribed mostly to the saponifiable substances (Assarsson & Åkerlund, 1967). Unsaponifiables have shown only a small decrease. The only chemical reaction-taking place when wood is seasoned under water is the hydrolysis of glycerides to fatty acids. No changes indicative of metabolism or autoxidation have been observed (Assarsson, 1966). Chemical reactions are markedly faster when the wood is stored in the form of chips instead of logs (Alén, 2000). Promberger et al. (2004) concluded that the faster deterioration of compounds in wood chips is due to the larger surface area that makes substances more easily accessible (Promberger, Weber, Stockinger, & Sixta, 2004). As an example, it has been reported that the degree of hydrolysis of triglycerides after eight weeks of outdoor chip storage was about the same as round-wood storage for one year (Ekman, 2000).

Different tree species vary with regards to their extractive content as well as the composition of this fraction which in turn affects the chemical decomposition reactions occurring during storage (Routa et al., 2017). Furthermore, different parts of wood have different extractive contents and compositions of extractive fractions (Routa et al., 2017).

Fatty acids are oxidized much faster than resin acids during chip storage and at a faster rate than the amount of resin acids. Tall oil from fresh pine yields approximately equal amounts of fatty and resin acids. According to Fuller (1985), within 4-8 weeks of storage 60-80% of the yield of by-products, tall oil and turpentine, are lost from coniferous species chip piles (Fuller, 1985). Turpentine is lost more rapidly than tall oil. According to Ekman (2000), after four months storage, only 25% of the fatty acids remain while 56% of the resin acids are retained. A few weeks of storing pine wood chips can lower the turpentine yield by about 50%. The turpentine losses are due to evaporation of volatile terpenes that are carried away in the convection of moist air currents that normally occur in the pile.

According to Assarsson et al. (1963), the total resin content in chipped spruce wood was almost constant for two weeks. Thereafter the resin content decreased and the rate of hydrolysis reaction slowed down after four weeks. In spruce wood chip seasoning, the amounts of triglycerides decreased by 90% and the amounts of waxes decreased by 70% in 3 months (Assarsson & Åkerlund, 1967). The remaining esters might have been sterol esters largely, as these are more difficult to hydrolyse than glycerol esters. The amounts of free fatty acids increased strongly at the beginning and reached a maximum level after one week in a spruce chip pile (Assarsson & Cronn, 1963). After that period, the rate of free fatty acid oxidation became greater than that of ester hydrolysis, hence, the amount of free fatty acids started to decrease. They also found that the resin acids remained unaffected until after 2-4 weeks of seasoning. In a later study, the amounts of resin acids had decreased by 60% after 3 months of seasoning (Assarsson & Åkerlund, 1967). The unsaponifiable compounds seemed to have remained constant during the first eight weeks in the chip pile, whereafter a decrease was discernible. With regards to the log storage of spruce wood, the unsaponifiable fraction was found to be almost unaffected (Assarsson & Cronn, 1963).

Halmemies et al. (2018) studied the effects of 24 week storage on spruce (Picea abies) bark extractives fraction. To study the way the different storage conditions affect the preservation of spruce bark extractives, two experimental setups were established: a single stem setup where spruce logs were stored with bark intact, and a bark pile setup of spruce saw mill bark (Fig. 6.). Special emphasis was on valuable phenolic compounds, like stilbenes. Lipophilic hexane extracted fractions remained quite stable within the whole storage period in both setups, whereas there was a big difference between the different storage setups in the rate of degradation of the hydrophilic fractions. While the hydrophilic fraction of the single stem samples remained stable for 12 weeks the hydrophilic fraction of the bark pile samples showed significant deterioration just after 4 weeks. The different sampling locations in the pile also depicted clear trends: the rate of decrease in extractives content was the fastest in the top of the pile, the slowest in the middle of the pile, and somewhere in between them in the side of the pile.

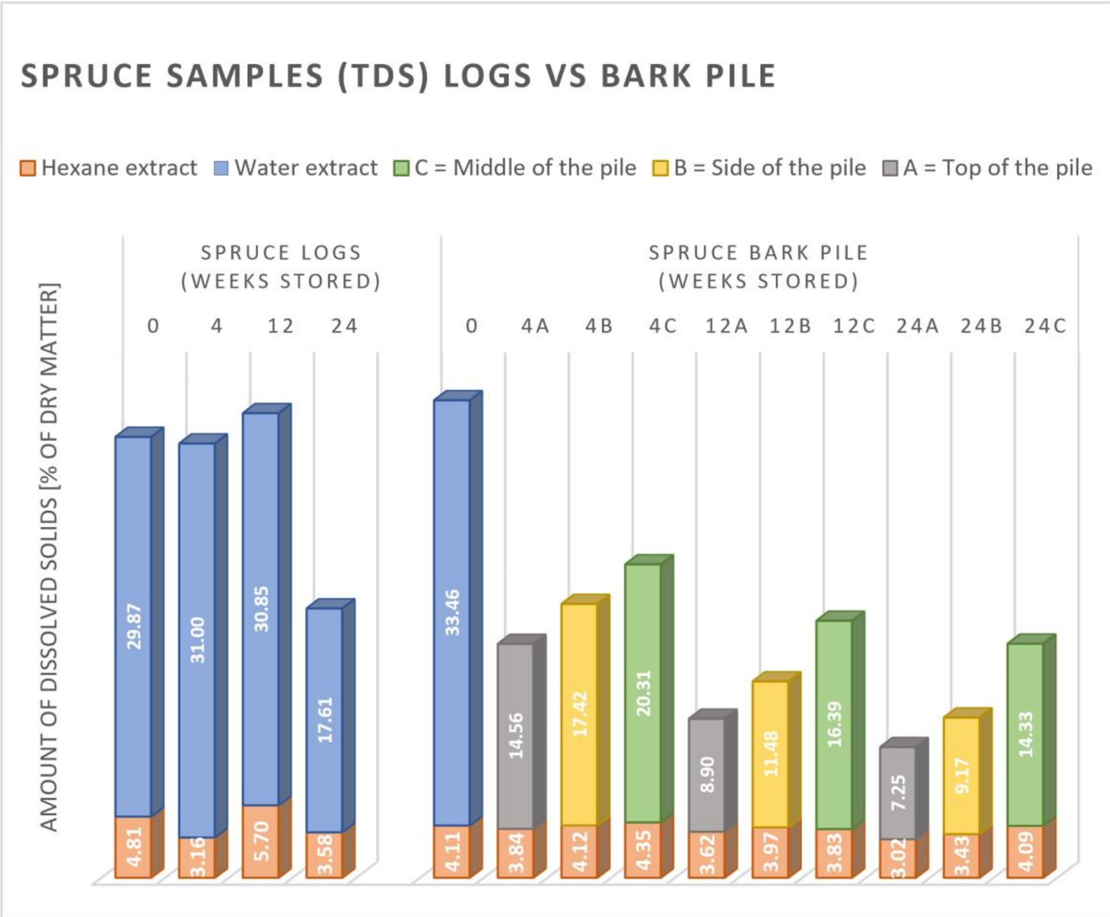

Fig. 5. Total dissolved solids of the single stem and bark pile experimental setups. Experimental setup for Norway spruce saw log (Picea abies) bark storage studies was constructed February 7 2017 in Kälviä, and saw mill bark pile was built up February 20 2017 in Pietarsaari. Both storage studies were located in Western Finland.

The concentration of the major stilbene glucosides (piceid, astringin, and isorhapontin) in the spruce bark samples from the single stem and bark pile experimental settings are illustrated in Fig.6. The freshly debarked spruce bark from saw mill seemed to correspond to the bark that was stored intact on saw logs for 4 weeks in terms of stilbene concentration. Most significantly, at four weeks the stilbene concentration in the bark pile reached undetectable levels, while the stilbenes in log bark could still be found after 24 weeks.

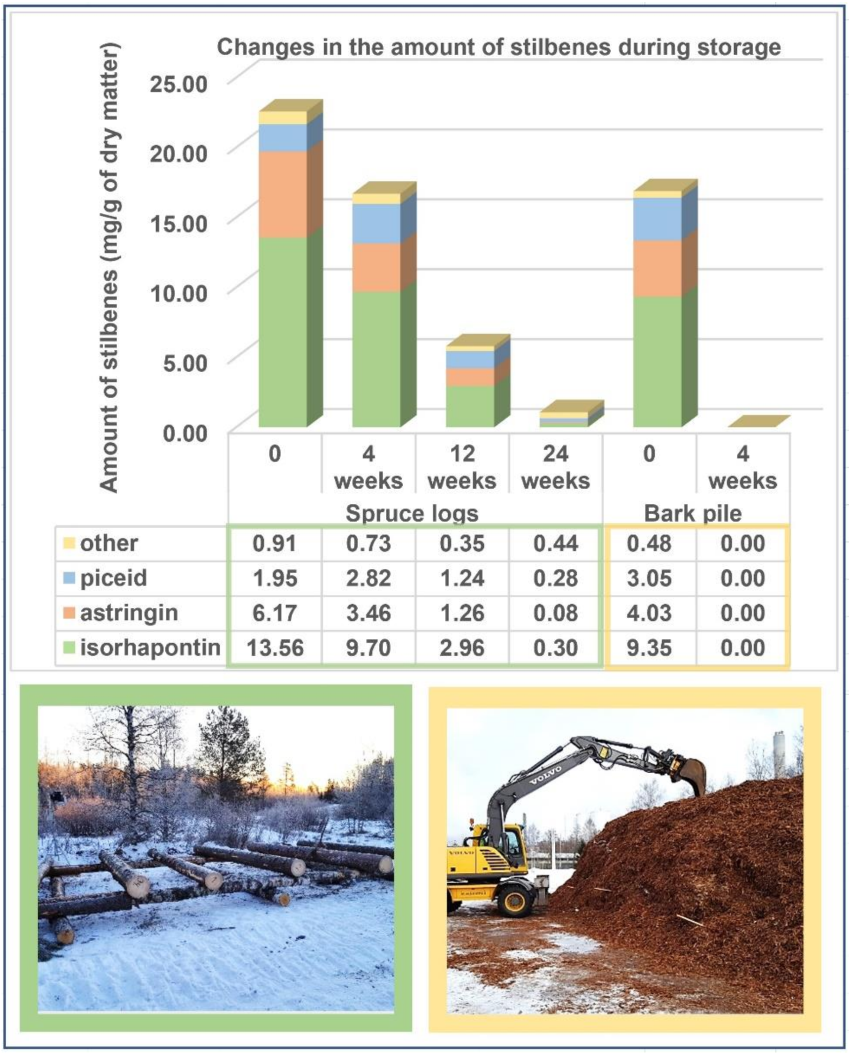

Fig. 6. The changes in the concentration of stilbenes in spruce bark during the storage of bark in logs and bark pile (Halmemies et al. 2018).

Different methods of storing raw materials like spruce bark can have huge impact on the content and composition of its extractives-fractions. For example, the valuable stilbenes in spruce bark are quickly lost if the bark is stored in a pile, whereas storing the bark in logs shelters the stilbenes from oxidation and UV light induced reactions.

How can degradation be measured and monitored?

3.1 METHODS FOR MEASURING DRY MATTER LOSSES

Dry matter losses are expressed as change in total dry weight before and after storage, thus reflecting the amount of degradation, which occurred in stored biomass over time. There are, at present, no established standard method for determining dry matter losses, thus the development of such would simplify comparison between studies. One commonly used method of calculating dry matter loss involves placing net bags (filled with the feedstock material) within storage piles and windrows (Fig. 7 a). The initial moisture content of the piled feedstock is then compared to the moisture content of the feedstock in the sample bags after the storage period to determine the amount of material loss. This method requires a large amount of sampling and evenly distributed sampling points in order to obtain accurate representative values since moisture content can vary within different layers and sections of biomass (Fig. 7 b).

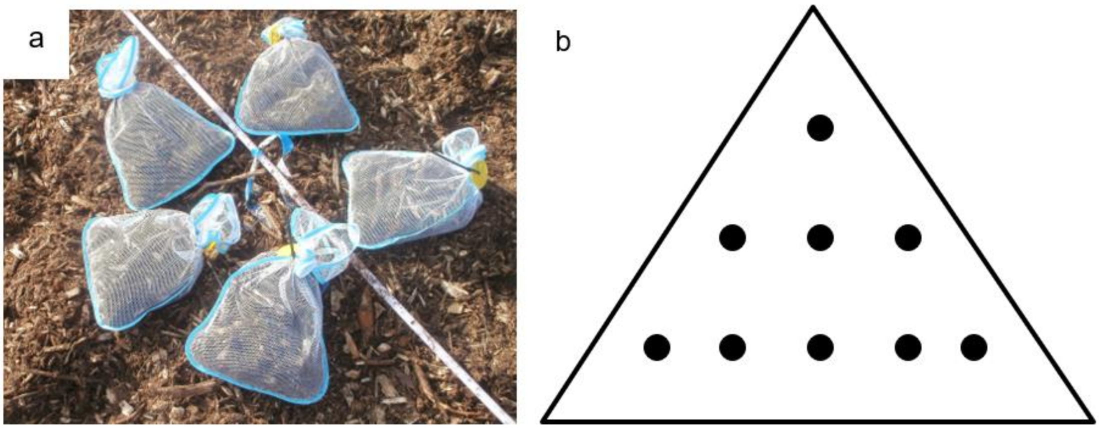

Fig. 7. Net bags for determination of dry matter losses during storage (a) and sampling points within a pile (b)

An alternative method is to weigh all biomass before and after storage and then calculate the substance losses based on difference in dry matter calculated from mean moisture content before and after storage. Negative dry matter losses (an increase in the dry matter during storage) are reported in several studies, implying that the initial dry weight of the sample bags were likely calculated from an initial average moisture content. A comparison of mean dry matter loss obtained are still complicated due to major differences between studies. Thus, proper sampling methodology and data handling are critical when determining dry matter losses over time since misleading information can easily be produced otherwise. Lenz, et al., (2017), stated that it is possible to determine degradation by comparing the difference in ash content before and after storage (Lenz et al., 2017). Ash content however derives from both natural inherent minerals and inorganic contamination where some parts are water-soluble while others are not. In addition, contamination of the pile surface is common during storage at a terminal. Currently, it is not feasible to quantify or monitor degradation of neither residues nor chips and bark during storage under practical storage conditions. It is possible to measure pile temperature on the surface and thereby reflect the activity in a pile. Continuous measurement of temperature and off gassing can provide important information about the activity within a pile. A rapid increase in surface temperature may indicate a strong self-ignition and fire hazard. However, the temperature on the surface does not directly correspond to the temperature fluctuations taking place within a pile, and no temperature curves for estimating critical temperatures have been developed.

3.2 MODELLING AND MONITORING CHANGES DURING STORAGE

Moisture content is a key factor and one of the most important fuel quality parameters affecting both net heating value and dry matter losses during storage. Finding methods to monitor moisture content and dry matter losses without the need for frequent sampling and measurement remains a major challenge. Drying models, where changes in moisture content during storage of different wood assortments (e.g. logwood, stumps, and various types of forest residues) have been reported in numerous studies (Erber et al., 2012; Erber, Kanzian, & Stampfer, 2016; Erber, Routa, Kolström, et al., 2014; Erber, Routa, Wilhelmsson, et al., 2014; Filbakk, Høibø, Dibdiakova, et al., 2011; Raitila, Heiskanen, Routa, Kolström, & Sikanen, 2015;Routa, Kolström, Ruotsalainen & Sikanen, 2015b, 2016; Lindblad, Routa, Ruotsalainen et al. 2018). With the ability to predict changes in feedstock moisture content in relation to local meteorological parameters such as relative air humidity, air temperature, precipitation and evaporation along with specific factors such as tree species, material type (e.g. logs or forest residues), storage duration and initial moisture content, quality assessments can be estimated without the need for extensive sampling trials. Such models can be used to improve the wood supply chain and enable the optimization of transport and fuel deliveries as well to better meet the demands of the customer (Erber, Routa, Wilhelmsson, et al., 2014). Erber et al., (2012) developed a model for estimating drying of Scots pine logs and found that the best way to simulate the effect of moisture content was to flatten the model curves using a moving average (Erber et al., 2012). A mean deviation of – 0.51% ± 0.71% and an R2adj = 0.62 was reported for the produced model. Drying models for small Pine logs in Finland and Austria considered the cumulated sum of metrological data (Erber, Routa, Kolstrom, et al., 2014). Erber et al., (2014) produced a linear regression model of drying under Austrian conditions of the cumulative alteration in moisture content based on cumulative sums of daily means for air temperature, wind speed, relative humidity and precipitation. In contrast, the Finnish drying model only considered the daily sum of net evaporation with an aim to achieve a linear regression model with a deviation of the moisture content (up to ± 5%). Erber et al. (2014) reported that the mean deviations of moisture content calculated from produced linear models were –1.04 % ± 1.43 % for the model produced using the Austrian data, and 0.52 % ± 0.87 % using the Finnish data (Erber, Routa, Kolstrom, et al., 2014). A drying model for piled European beech (Fagus sylvatica L.) logs based on meteorological data, employing the continuous weighing approach including two drying cycles and three piles altogether were conducted in Austria (Erber et al., 2016). This model was validated by applying the model developed from the second cycle's data on the first cycle's data resulting in a mean deviation of − 0.41% ± 0.72% from the observed curve. Routa et al. (2016) has validated spruce logging residues models against data from field studies, over 200 reference piles were studied, and the difference between measured and modelled moisture was on average 0.35%. With small diameter stem wood the validation was done against data from forest companies (22piles), and the difference between measured and modelled moisture was on average 0.3% with covered piles and 2.5% with uncovered piles(Routa et al 2015b). Overall, the development of models, which can accurately estimate moisture content and dry matter losses in bark and woodchip piles, remains a work in progress. Further advancements in predictive modelling for moisture, temperature, gas emissions, and dry matter loss within piles are therefore currently highly desirable.

Measures to minimize dry matter losses

4.1 NATURAL DRYING AND STORAGE OF UNCOVERED FOREST RESIDUES AND WOOD LOGS

The moisture content of biomass is the most important parameter during storage since it affects fuel quality, in particular, the net calorific value, biological activity leading to dry matter losses, transport costs and revenue of the fuel (Erber et al., 2016). Moisture management during storage is a key factor for improving the whole revenue of the supply chain of wood fuel and storage has to be considered in each supply chain system. In general, moisture content declines during storage of uncomminuted forest residues (Afzal et al., 2009). However, biomass, which is a hygroscopic material, is exposed to varying weather conditions and can easily be rewetted during storage (Fig. 8). Therefore, the rate of drying depends on many factors including ambient temperature, relative humidity, wind speed, season, precipitation pattern, tree species, and size of the stored biomass. The storage conditions are essential in maintaining fuel quality (Wetzel, Volpe, Damianopoulos, &

Krigstin, 2017). The best season for drying is during periods of high ambient temperature and low humidity, which usually occur during late spring/early summer in temperate climates and dry season in the tropics (Richardson et al., 2006).

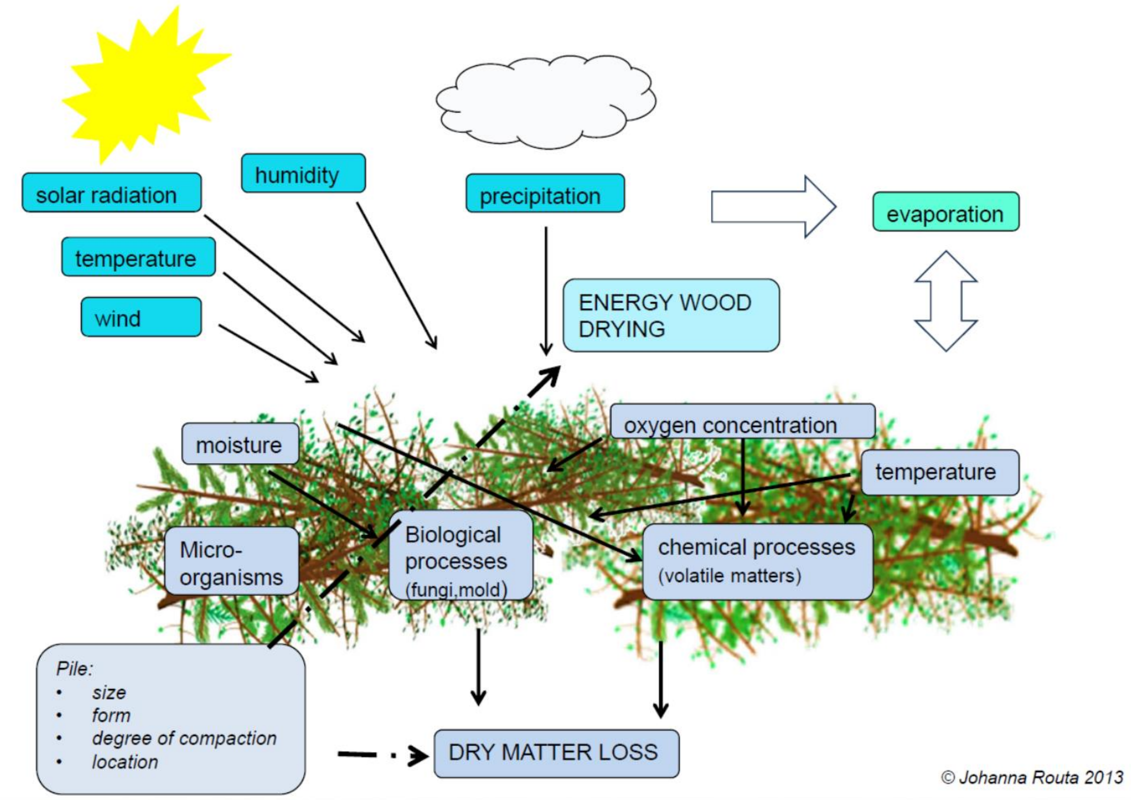

Fig. 8. Energy wood drying and dry matter loss processes in natural conditions.

Storage conditions facilitating the natural drying rate, which in turn affect energy content and the total amount of assessable energy, is essential from an energy perspective. Therefore, the focus is to reduce moisture content and biological activity, which will improve fuel quality and actually could increase the assessable amount of energy (since less energy would be used up for the evaporation of water). Nurmi & Hillebrand (2007) reported that moisture content in pine and birch stem wood can drop below 30 % during the summer (Nurmi & Hillebrand, 2007). Similarly, a decrease from 53 % to between 33 and 21 %, was reported for radiata pine logs after 24 weeks of summer storage in New Zealand (Visser, Berkett, & Spinelli, 2014). Erber et al. (2014) showed that moisture content in small logs dropped from 50.1 % to 32.2 % when stored in Austria and from 62.2 % to 38.6 % when stored in Finland (Erber, Routa, Kolstrom, et al., 2014). Civitarese et al. (2015) reported that moisture content declined with 10 to 20 percent points during storage of poplar stem wood logs stored in windrows during December to June in central Italy. According to Civitarese et al., no dry matter losses due to microbial activity could be recorded during the same period. Assarson et al. (1970) on the other hand have reported dry matter losses of about 1.1 % during storage of roundwood after one summer, while more recently dry matter losses of 0.07 -1.52 % per month for small sized delimbed roundwood have been reported (Routa et al., 2018). Manzone (2015) reported dry matter losses ranging between 1.2 to 6.6% after storage of poplar and black lotus logs from March to September (Manzone, 2015). However, the differences in the dry matter between initial and final values were not statistically significant. At the same period, the moisture content dropped to 19% from an initial value of 60% in poplar and 45% for black lotus. The highest loss was observed for the smallest diameters, while the lowest moisture was observed for the largest diameters

(Manzone, 2015). However, logs could be rewetted during autumn, which could increase the moisture content to the initial value of fresh biomass (Röser et al., 2011). Considering the overall assessment of drying rate and low dry matter losses reported during log storage, this implies that open-air storage of wood logs in windrows could be a viable solution for fuelwood management during spring and summer.

Storage conditions, which facilitates drying is an important prerequisite for maintaining as much energy as possible. Nurmi concluded, after showing that the average moisture content dropped to 28.5% at the harvesting site compared to 42.2% at the landing, that storage in small heaps at the harvesting site was a better drying site for forest residues than storage in windrows at landing (Nurmi, 1999). Pettersson & Nordfjell showed that the moisture content of freshly harvested forest residues could drop to 28.6% after 3 weeks when stored in small heaps at the harvesting site in May (Pettersson & Nordfjell, 2007). Nurmi and Hildebrand reported that fresh logging residues lose moisture very quickly during early summer and storage for only 2 to 3 weeks at the harvesting site may lower the moisture content up to 30% (Juha Nurmi & Kari Hillebrand, 2001). It is clear that pre-storage in small heaps and during favourable storage conditions increases the drying rate at the harvesting site; however, the fuel will retain moisture more rapidly during periods of precipitation. Guatam et al. reported that windrows displayed lower moisture content than small heaps after storage and that smaller piles regained moisture more rapidly when the temperature and relative humidity dropped (Gautam, Pulkki, Shahi, & Leitch, 2012). Nilsson et al. showed that both handling methods provided adequate drying and their results indicated that weather conditions have a greater impact on the moisture content than the handling method (Nilsson, Blom, & Thörnqvist, 2013). Storage at the harvesting site allows for the mechanical loss of needles, small twigs and leaves, which reduce the impact of nutrient removal. Less biomass, mainly needles and twigs could be gathered when stored in heaps at the harvesting site than when stored in windrows. However, needles contain high levels of ash forming minerals, which negatively could affect the ash melting behaviour. If properly planned, gathering of pre-stored heaps into windrows at landing during summer months can lead to lower moisture content, better storage properties during the autumn and winter seasons and facilitates management when the fuel is required. The proportion of biomass lost through handling compared to degradation during storage in small heaps is uncertain. Dry matter losses ranging between 1-3% per month are reported during storage of forest residues in uncovered windrows at the harvesting site and at the landing (Jirjis, 1995; Nurmi, 1999; Pettersson & Nordfjell, 2007; Routa et al., 2015a). It is recommended to leave forest residues in small heaps at the harvesting site to the end of the summer and then forwarding the material to a windrow at a landing to ensure the loss of nutrient-rich components as well as a fuel with lower moisture content and better storage characteristics.

4.2 COVERAGE OF FOREST RESIDUES AND LOGS

Numerous studies have shown that coverage of forest residues and wood logs can protect against precipitation penetration into the stored material and thus rewetting of the fuel (Jirjis, Gärdenäs, & Hedman, 1989; Jirjis & Lehtikangas, 1993; P Lehtikangas & Jirjis, 1993; J Nurmi & K Hillebrand, 2001; Röser et al., 2011). According to Nurmi and Hillebrand, coverage will result in up to 6% units lower moisture content in comparison with non-covered piles (Nurmi & Hillebrand, 2007). Jirjis reported that the achievable effect of covering range from an additional 3 to 6% reduction in moisture content compared to uncovered piles (Jirjis, 1995). During storage of forest residues, the average moisture content of a covered pile decreased from 36.1% to 32.2% under a covered part between August to February, while the forest residues in the uncovered part were clearly rewetted to 44.0% (Jirjis et al., 1989). Coverage of forest residues during November until September showed that moisture content decreased from 42% to 21% in a covered windrow and to 35% in an uncovered part (Jirjis & Lehtikangas, 1993). The total dry matter loss in this windrow was 2.3% in the covered part and about 10% in the uncovered part. Lehitkangas & Jirjis (1993) showed that moisture content decreased from 28.6% to 20.9% when forest residues were stored in covered windrows during a period from September to March, while the moisture content increased to 29.7% in the uncovered part (P Lehtikangas & Jirjis, 1993). Coverage of windrows (e.g. with a paper-based cover) is recommended, especially under wet seasons like autumn in Scandinavia, for both logging residues and stem wood (Jirjis, 1995). In addition, it could probably be beneficial in alpine regions (Elber, 2007; Erber et al., 2012; Golser et al., 2005) as well as in Scotland (Röser et al., 2011).

Röser et al. evaluated the effect of debarking and coverage of stem wood in Finland, Scotland and Italy (Röser et al., 2011). Röser et al. (2011), reported that the moisture content of un-debarked and debarked Sitka spruce logs stored in covered piles, during June- February in Scotland, decreased from 55% to 30% and that moisture content in lodgepole pine decreased from approximately 51% to 36% during the same period. The drying rate within the uncovered piles was slower and the moisture content was only slightly lower compared to the moisture content of fresh lodgepole pine (Röser et al., 2011). In Finland, moisture content decreased from 53% to between 30 – 40% during the summer months. Coverage of piles in September had a clear significant effect where moisture content decrease with approximately 2% units, whereas moisture contents increased with approximately 5% units in the uncovered piles (Röser et al., 2011).

Lehtikangas and Jirjis concluded that proper coverage of the entire surface is important to avoid accumulation of water from rain and melting snow (P Lehtikangas & Jirjis, 1993). A breathable cover material like a paperboard (Fig. 9 a) can protect the material from precipitation, but not from rewetting caused by increased humidity or unfavourable placement (Fig. 9 b). The use of plastic tarps, which could be easier to work with, do not allow ventilation. Wetzel et al., (2017) reported that moisture content increased from 49% to 65% when forest residues were stored in a windrow covered with a plastic tarp during one year (Wetzel et al., 2017). To conclude, management that facilitates drying can reduce dry matter losses caused by microbial activity. Coverage of forest residues and logs with a breathable covering material during seasons with high humidity and precipitation is recommended. The effect of cover will decrease with the decomposition of the covering material. In addition, coverage is not necessary under dry conditions. Mechanical losses of dry matter, such as loss of needles, even though needles are energy-rich, increase fuel quality, increase ash-melting temperature and decrease ash content, while nutrients remain on the harvesting site.

Fig. 9. Storage of forest residues in windrows covered with a paperboard that protect the biomass from rewetting caused by precipitation (a) and unfavourable placement of a windrow which increase the moisture content due to remoistening from the ground (b).

4.3 STORAGE OF WOOD CHIPS

Proper storage before comminution refer to storage methods that facilitate moisture content reduction, increase wood fuel quality and the amount of assessable energy. The role of storage and drying process in the chain between harvesting and utilisation is critical since moisture content is the most important physical parameter that determines fuel quality. Storage in uncomminuted form maintains the quality of the fuel. However, certain volumes of wood fuel have to be stored in the form of chips at least as a buffer. Temperature development in piles of chunk wood is also lower and more dependent on ambient conditions. Despite these positive results, comminution to chip size is still the most commonly used method (Jirjis, 1995). Storage before comminution helps in minimizing decay by reducing the surface area exposed to potential microbial degradation (Barontini et al., 2014). However, handling cost for uncomminuted forest residues is higher than for woodchips (Barontini et al., 2014). This could partly be related to increased payload utilisation during transport of chips, which reduces the total costs (L. O. Eriksson & Björheden, 1989). Various approaches, aiming to increase the efficiency in the supply chain of forest fuels by improving management efficiency have been suggested (e.g. (Asikainen et al., 2015; Carl-Henrik Palmér, 2016; A. Eriksson, Eliasson, Sikanen, Hansson, & Jirjis, 2017; Flisberg, Frisk, & Ronnqvist, 2012; Windisch et al., 2015; Å. Thorsén, 2011)). An increase of the annual utilisation of chippers and chip trucks is one obvious way to reduce the supply cost (Anerud et al., 2018). This requires storage of comminuted wood and wood chips are extremely vulnerable to microbial degradation during storage. Extensive dry matter losses and fires, caused by spontaneous heating and self-ignition, constitute a high risk during large-scale storage of wood chips.

Average dry matter losses of 0.7 to 5% per month are reported in several studies (Table 1). Storage of poplar wood chips leads to greater losses than during storage of Norway spruce chips, but since the studies differ in so many ways, a direct comparison is not possible. In general, the rate of decomposition is faster at the beginning of the storage period and then decreases with storage time (Anerud et al., 2018; Gislerud, 1974; T. Thörnqvist & Jirjis, 1990b). Several factors affect the commutative dry matter loss, for instance, the particle size of the comminuted wood affects the outcome of storage (Jirjis, 1995).

4.3.1 Fraction size and compaction of comminuted wood chips

A number of studies have compared storage of chunk wood and chips (Table 2). Baadgaard-Jensen, (1988) reported that dry matter loss was much greater for chips than chunks, 2.9% vs. 0.3% per month, during small-scale storage (Baadsgaard-Jensen, 1988). Pari et al, (2015) reported accumulated dry matter losses of 12.9% in fine poplar wood chips during March to June, while coarse wood chips reached 7.2% during the same period (Pari et al., 2015). The temperature is a measure of the total biological and chemical activity in a pile and the cumulative heat is a measure of the degradation that occurs during storage. A rapid rise in temperature to above 60˚C, often followed by a long period of slow temperature decrease is reported in numerous studies (Table. 1). Barentoni et al., (2014) reported a rapid peak near 70˚C within in the first week of storage of poplar crown wood chips (Barontini et al., 2014). Increased temperature indicates and reflects biological activity and chemical reactions. The growth of microorganisms, at least for most species, ceases at about 55°C (Jirjis & Lehtikangas, 1992). A further increase is attributed to chemical processes such as hydrolysis and oxidation. The internal temperature is strongly affected by ambient air temperature within small piles (Richardson et al., 2006). The impact on large-scale piles is low, but weather conditions such as ambient temperature as well as precipitation still affect storage conditions.

Table 1. Dry matter losses during storage of comminuted forest residues and wood logs.

Fuel	Chip size (mm)	Pile volume (m3)	Method	Coverage	Moisture content (%, w.b)	Storage period	Dry matter loss (% per moth)	Dry matter loss (% total)	Max temp ˚C	Reference
Birch	2-25		Net bag	Tarp	51.0	Jul-Jul	0.7	8.0		(Afzal et al., 2009)
	2-25		Net bag		59.0	Jul-Jul	2.6	27.0		(Afzal et al., 2009)
	6	400	Net bag		40.0	May-Dec	1.1	7.5		(Jirjis, 1995)
	19	400	Net bag		40.0	May-Dec	1.3	8.7		(Jirjis, 1995)
Norway spruce	P45	725	Net bag		50.6	Jun-Jan	1.1	7.3	67	(Anerud et al., 2018)
	P45	725	Net bag	Toptex	50.6	Jun-Jan	0.9	5.8	64	(Anerud et al., 2018)
	Fine	200	Net bag		56.6	Nov-Apr	0.6	3.4	60	(Hofmann et al., 2018)
	Fine	200	Net bag		51.1	Nov-Apr	1.3	7.8	60	(Hofmann et al., 2018)
	Fine	200	Net bag	Toptex	55.1	Nov-Apr	0.7	4.2	60	(Hofmann et al., 2018)
	Fine	200	Net bag	Toptex	50.7	Nov-Apr	1.4	8.0	60	(Hofmann et al., 2018)
	Fine	200	Net bag		47.9	May-Oct	1.4	6.8	60	(Hofmann et al., 2018)
	Fine	200	Net bag		50.4	May-Oct	1.4	7.0	60	(Hofmann et al., 2018)
	Fine	200	Net bag	Toptex	53.2	May-Oct	1.4	6.9	60	(Hofmann et al., 2018)
	Fine	200	Net bag	Toptex	56.8	May-Oct	2.3	11.1	60	(Hofmann et al., 2018)
	P45	1067	Weighted		41.5	Jan-May	1.4	5.6		(Nilsson et al., 2013)
	P45	997	Weighted		32.2	Jun-Sep	3.5	10.1		(Nilsson et al., 2013)
	P45	524	Net bag	Toptex	50.0	Jun-Oct	1.5	5.8	68	(Wästerlund, Nilsson, & Gref, 2017)
	P45	498	Net bag		48.0	Jun-Oct	1.9	7.2	68	(Wästerlund et al., 2017)
Poplar	P31	1000	Net bag	Toptex	60.0	Feb-Nov	2.5	22.0	62	(Lenz et al., 2015)
	P45	600	Net bag	Toptex	59.0	Jan-Nov	2.1	21.0	57	(Lenz et al., 2015)
	P31	100	Net bag	Toptex	62.0	Feb-Oct	3.3	23.3	62	(Lenz et al., 2017)
	P31	100	Net bag	Toptex	62.1	Feb-Oct	3.2	22.7	62	(Lenz et al., 2017)
	P31	100	Net bag	Toptex	62.2	Feb-Oct	1.8	13.9	62	(Lenz et al., 2017)
	P31	100	Net bag	Toptex	62.3	Feb-Oct	2.2	16.6	62	(Lenz et al., 2017)
	P45	35	Weighted		61.0	Jun-Sep	3.2	9.4	60	(Manzone & Balsari, 2016)
	P45	35	Weighted		61.0	Jun-Sep	3.7	10.7	64	(Manzone & Balsari, 2016)
	P45	70	Weighted		61.0	Jun-Sep	3.5	10.1	60	(Manzone & Balsari, 2016)
	P45	70	Weighted		61.0	Jun-Sep	3.3	9.5	58	(Manzone & Balsari, 2016)
	P63	20	Weighted		45.0	Mars-Sep	1.7	9.8	61	(Manzone, Balsari, & Spinelli, 2013)
	P63	20	Weighted	Roof	40.0	Mars-Sep	0.9	5.1	41	(Manzone et al., 2013)
	P63	20	Weighted	Plastic	45.0	Mars-Sep	1.2	7.1	41	(Manzone et al., 2013)
	P63	20	Weighted	Toptex	55.0	Mars-Sep	1.6	9.3	62	(Manzone et al., 2013)
	Fine	130	Plastic bag		62.0	Mar-Jun	3.4	12.9	65	(Pari et al., 2015)
	Fine	130	Plastic bag	Toptex	62.0	Mar-Jun	2.9	11.1	65	(Pari et al., 2015)
	Coarse	130	Plastic bag	Toptex	62.0	Mar-Jun	1.9	7.2	65	(Pari et al., 2015)
	Coarse	500	Plastic bag		60.0	Jan-Aug	4.4	27.0	14	(Pecenka, Lenz, Idler, Daries, & Ehlert, 2014)
	Fine	500	Net bag	Toptex	62.0	Jan-Aug	3.9	24.0	18	(Pecenka et al., 2014)
	Fine	500	Net bag	Toptex	54.0	Jan-Jul	2.7	15.0	60	(Pecenka et al., 2014)
	P31	117	Net bag		54.0	Mar-Sep	1.1	6.6	78	(Barontini et al., 2014)
	P31	117	Weighted		54.0	Mar-Sep	1.7	10.0	78	(Barontini et al., 2014)
	P31	117	Net bag		48.0	Mar-Sep	4.0	21.6	50	(Barontini et al., 2014)
	P31	117	Weighted		48.0	Mar-Sep	5.0	26.6	50	(Barontini et al., 2014)

Table 2. Effect of average particle size on dry matter losses during storage of comminuted forest residues

	Effect of average particle size	
Baadgaard-Jensen, 1988	▬	
Jirjis, 1995	▬	▬
Barontini et al., 2014	▬	
Pari et al, 2015	▬	
Lenz et al,, 2015	▬	▬
	Positive	Negative

Comminution to a large fraction is cheaper and requires less energy (Eliasson, von Hofsten, Johannesson, Spinelli, & Thierfelder, 2015). In addition, comminution to a coarse fraction results in a small proportion of fine fraction, which is preferred from a storage and combustion perspective, since high content of fines leads to higher cumulative heat during storage and to an uneven combustion at an undesired location. Manzone & Balsari, (2016) concluded that the influence of form or density for small sized piles (35-70 m3) of uncovered wood chips did not affect the final poplar woodchip quality or dry matter losses (Manzone & Balsari, 2016). Löwegren and Jonsson, (1987) reported dry matter losses between 12-24% when Norway spruce and Scots Pine chips were stored in large compact piles (5000 m3) for 7 months (Löwegren, 1987). During the same period, cumulative dry matter losses reached 8.1% in uncompact piles. Storage under a roof resulted in average dry matter losses of 9.5% and 4.8% in a compacted and uncompact pile respectively. More recently, Wästerlund et al. (2017) reported cumulative dry matter losses of 11% when Norway spruce was stored in compact cakes for 7 months (Wästerlund et al., 2017). To conclude, comminution to a coarse fraction decreases the rate of compaction, which is preferable when biomass has to be stored as chips.

4.3.2 Coverage of wood chips

Coverage of wood fuel piles can, if the cover material protects the biomass from rain and snow, both increase the drying performance and reduce the risk of dry matter losses (Table 3.). It is necessary to achieve a good airflow within a pile during storage to disperse accumulated water vapour and thus decrease moisture content. The effect of covering depends on several factors including assortment, season, storage duration, amount of precipitation, rain protection properties of the cover material and the ability to permeate water vapour and heat. Nurmi, (1990), reported increased moisture content when wood chips were covered with a plastic tarp (Nurmi, 1990) and Spinelli et al, (2007) showed that sealing wood chips in a trench led to anaerobic fermentation (Spinelli, Kofman, & Magagnotti, 2007). Moisture content in a chip pile increased from 49% to 65% during one year of storage when the pile was covered with a plastic tarp (Wetzel et al., 2017). Afzal, et al., (2009), reported a moisture content reduction from 51% to 26% during the storage period July-November when a 3 m high cone-shaped wood chip pile was covered with a tarp (Afzal et al., 2009). Future storage for 8 months showed continued decline to 16.5% and a uniform moisture content distribution within the pile. The moisture content of an uncovered wood chip pile continuously increased through the same storage period, resulting in more than double in magnitude from 59% to 160% (d.b) (Afzal et al., 2009). Wästerlund et al. (2017), reported similar results where moisture content in a covered pile dropped from 50% to 34.4% after 4 months of storage, while the moisture content in an uncovered pile increased from 48% 63% during the same period (Wästerlund et al, 2017). Anerud, et al, (2018) reported that coverage of a large-scale pile with a semi permeable cover could reduce moisture content from 52% to 45% when stored during June to January, while an uncovered part reached 48% during the same period (Anerud et al, 2018). Afzal et al., (2009) reported that the cumulative dry matter losses after 12 months of storage reached 8% when the wood chip pile was covered and 27% when stored in uncovered piles (Afzal et al, 2009). The use of breathable cover materials such as paper-based tarps and Toptex result in less degradation of biomass (Anerud, 2018, Wetzel et al., 2017, Wästerlund, 2016). It could be concluded that coverage facilitates drying and lead to comparatively low dry matter losses compared to uncovered wood chips However, the magnitude depends on numerous factors such as biomass assortment, particle size distribution, season, initial moisture content, pile size and shape, local weather conditions and tarp material.

Table 3. Effect of coverage on dry matter losses during storage of comminuted forest residues

	Effect of coverage	
Afzal et al, 2017	▬	
Anerud et al, 2018	▬	
Hofmann et al, 2018	▬	▬
Manzone et al, 2013	▬	
Pari et al, 2013	▬	
Wetzel et al., 2017	▬	
Wästerlund et al, 2017	▬	
	Positive	Negative

4.4 STORAGE OF BARK

Only a few storage trials with bark have been performed. Fredholm & Jirjis (1988) reported dry matter losses of 26% during 5.5 months of bark storage, with an initial moisture content of 65-68%, in a 4 m high pile (Fredholm & Jirjis, 1988). It was assumed that the storage ability may be improved during the storage of bark if water was removed by a mechanical press so that the moisture content drop below 50 % (Fredholm & Jirjis, 1988). Jirjis & Lehtikangas (1992) studied the incorporation of dry biomass to reduce moisture content (e.g. mixing bark and dry shavings) (Jirjis & Lehtikangas, 1992). Dry matter losses of such mixes resulted in 2.4% to 5.0% after one month of storage, while at the end of the storage period the cumulative losses reached 8 – 9% (Jirjis & Lehtikangas, 1992). Lehtikangas and Jirjis examined coverage of 4 m high bark piles with a tarpaulin and the effect on moisture content and dry matter losses (Lehtikangas & Jirjis, 1998). Changes in moisture content and amount of dry matter were measured after 1 to 2 months and the temperature within the piles was continuously monitored. The temperature within both piles increased to 60°C during the first month of storage. The moisture content after 2 months of storage was decreased from 47% to 38% in the covered and from 49% to 43% in the uncovered. Dry matter losses reached above 10% irrespective of coverage. One main conclusion drawn from the bark studies was that the cover material has to be efficient in ventilating water vapour and heat.

4.5 STORAGE OF STUMPS

Harvested stumps contaminated with soil results in higher ash content. Current practice during handling of stumps is based on the assumption that storage in heaps on the harvesting site for a couple of months can contribute to reducing contaminants by precipitation. Contaminants as well as stump wood dry during storage in small heaps, but roughly, handling during forwarding is still required to achieve an acceptable fuel quality. In the Nordic countries, contaminants are usually frozen onto the stumps when the fuel is required during late autumn and winter. Direct storage in windrows is an available option, which can be considered if windrows are exposed to favourable storage conditions such as wind and sun exposure (Fig. 10). The moisture content in spring-harvested stumps decreases rapidly during storage. Anerud and Jirjis reported a reduction in moisture content from 40-50% to around 25% when stumps were stored in heaps and windrows for three months with only marginal re-wetting during autumn (Anerud & Jirjis, 2011). Similar drying and rewetting were reported during small-scale storage (Nylinder, 1981) and in large-scale storage in Finland (Laurila & Lauhanen, 2010). Storage of stumps in heaps compared to direct storage in windrows had no significant effect on moisture content and ash content according to Anerud and Jirjis (Anerud & Jirjis, 2011). Cumulative dry matter losses in the range of 1.5-2.0 % were reported when stumps were stored for three months (Anerud & Jirjis, 2011). Storage for 9 months showed cumulative dry matter losses in the range of 2.5-5.0%. Gathering split stumps directly into windrows at the roadside instead of storing them in small heaps at the harvesting site provides two advantages: the logistics for forwarder use for collecting forest residues can be more efficiently coordinated with stump removal from the clear-cut area and such coordination of forwarding, when possible, can reduce transport costs. Windrow storage at the roadside facilitates regeneration operations, which can result in faster tree establishment. Referred to moisture content, it is possible to shorten the storage duration. However, shortening the storage duration can affect the possibility of contaminant removal. To solve such a problem, faster and more efficient methods to clean the stumps are highly desirable in those countries were stump harvesting still is acceptable.

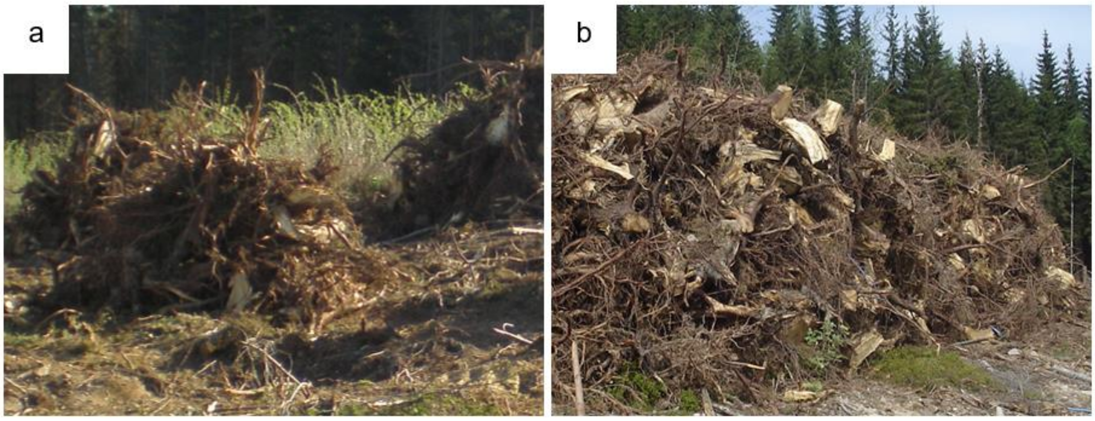

Fig. 10. Storage of stumps in small heaps at the clear cut (a) and storage of stumps in windrows (b)

4.6 STORAGE OF PELLETS

Storage of wood pellets is substantially different from the storage of unprocessed raw materials, one of the most obvious divergences being the low moisture content of pellets (Päivi Lehtikangas, 2000). During wood pellet storage, there is the added risk of catastrophic dust explosions as wood pellets are hydrophilic and need to be stored in dry ambient conditions. The pre-treatment stages during pellet production, especially dehydration at high temperatures reduces the bacterial activity in pellets. This in turn reduces the risk of spontaneous heating and storage losses compared to wood chips (Alakoski et al., 2016).

Off gassing is another concern where wood pellets during storage and transportation (under certain conditions) may emit condensable gases such as volatile organic compounds (VOC's) (e.g. aldehydes and ketones), as well as non-condensable gases, primarily CO, CO2, and CH4. The emissions from condensable gases are often combined with a pungent smell. In a previous study, (Arshadi & Gref, 2005) the emissions from wood pellets made from fresh and stored Norway spruce and Scots pine were investigated. Measurements were done by headspace (HS) analyses, using gas chromatography and mass spectrometry (GC-MS).

The amount of fatty and resin acids varies in different raw materials. For example, spruce sawdust normally contains considerably lower amounts of fatty and resin acids than pine sawdust. A systematic investigation by Near Infrared (NIR) spectroscopy and GC-MS showed that during large scale storage (16 weeks) of Scots pine and Norway spruce sawdust, the amount of fatty and resin acids decreased (Arshadi, Nilsson, & Geladi, 2007). The studies have shown that auto oxidation of fatty and resin acids may occur during storage of sawdust as well as wood pellets. Auto oxidation starts with the formation of free radicals that, in presence of oxygen, form hydroperoxide radicals. These can then interact with an unsaturated fatty acid producing two hydroperoxides and a new free radical (Baduí-Dergal, 1996).

There are several hypothesized causes for these phenomena, but the most important seems to suggest auto oxidative processes in the raw woody material are responsible in combination with moisture absorption from ambient air (Arshadi, et al., 2018, Arshadi, et al., 2009, Arshadi et al., 2007, Arshadi, Gref 2005, Nilsson et al. 2018). Recently, major laboratory trials (Blomqvist, et al., 2007 and Persson et al. 2006) have shown that in the case of pellets, the self-heating process is linked to emissions of toxic substances. In conjunction with heating, volatile substances are released, as well as those found naturally in the wood, and partly as formed by oxidation of stored fats, such as aldehydes (VOC), carbon monoxide (CO), carbon dioxide (CO2), methane (CH4) and low molecular weight carboxylic acids. A number of the substances (such as several aldehydes) were found to have a low odor threshold and can be corrosive to the eyes and upper respiratory tracts. CO and CO2 are harmful in high doses due to asphyxiation and can in the worst-case result in death. Lastly, methane is a flammable gas and therefore it’s accumulation may pose an ignition hazard. By removing extractives, the amount of off-gassing is reduced drastically (Attard et al 1016). Various methods for off-gassing measurements were compared (Sedlmayer al 2018).

During the auto-oxidation processes, the temperature in the pellet piles may often increase up to 60°C (Arshadi et al., 2007; (Arshadi, Geladi, Gref, & Fjallstrom, 2009). In these studies, chemical characterizations of fatty and resin acids in sawdust and pellets were performed. It was found that stored pellets contained a lower amount of fatty and resin acids compared to the corresponding fresh pellets. In a previous study (Arshadi et al., 2009); the correlation between the auto oxidation of fatty and resin acids and the emissions of volatile organic compounds (VOC) was investigated. In pellets with a pungent smell, high levels of hexanal and pentanal together with minor amounts of other aldehydes were detected by Arshadi et al (2005). A comparison between newly produced (fresh) pellets and pellets stored for a short time after production showed that there were more emissions from the stored pellets. The three-week-old pellet sample emitted about 28 times more pentanal and 8 times more hexanal than the reference pellets. The emissions of terpenes were low in wood pellets since most monoterpenes leave the sawdust during high temperature drying in the pelletizing process. A comparison of monoterpenes in fresh pine sawdust and the corresponding pellets showed that more than 95% of the monoterpenes were gone after pellets production (Arshadi, et al 2005). Only a small amount of α-pinene, β-pinene and 3-carene could still be detected in the pellets. This was not surprising since the boiling point of most terpenes is below 160°C and during the drying process of sawdust; the temperature is often higher than 195°C. Headspace analyses of pellets confirmed that several aldehydes and low molecular carboxylic acids were emitted from pellets. Some of these volatile organic compounds may have a negative impact on human health (e.g. irritation in eyes and upper airways) (Hagström, 2008).

4.7 CHEMICAL ADDITIVES

Alakoski et al, (2016) stated that eliminating microbial action inside the pile would also eliminate the risk of fires due to spontaneous ignition (Alakoski et al., 2016). Wood chips undergo no pre-treatments prior to chipping, except the natural drying of the raw material in outdoor conditions. Additives have been tested during the 1970's. Large-scale trial with chips treated with sodium hydroxide and then stored in a 10000 m3 pile showed that the temperature rose more rapidly than in untreated piles but also decreased more rapidly (Bergman, Nilsson, & Jerkeman, 1970). According to Bergman et al. (1970), the cumulative dry matter losses only reached 1.7% when chips were treated with Sodium hydroxide, while untreated reached 2.9% during the same period (Bergman et al., 1970). However, sodium hydroxide as an additive for wood fuel is undesirable due to its agglomeration properties and can adversely affect the working environment. Thirty chemicals or chemical mixtures were evaluated for their effectiveness in controlling fungal degradation in wood chips during storage (Eslyn, 1973). The effective concentration of each chemical was determined by laboratory-controlled incubation of red pine inoculated with five mesophilic and three thermophilic wood-degrading fungi. The additives were considered effective if the weight losses caused by each of the test fungi was less than 0.5 % during a storage period of 12 weeks. Eslyn (1973) showed that 23 of the tested chemicals were effective at various concentrations. Though effective, they generally caused unwanted reactions during combustion.

Recommendations

5.1 UNPROCESSED FOREST BIOMASS

1. Pre-storage in small heaps during favourable storage conditions can increase the drying rate at the harvesting site. However, weather conditions must be considered since forest residues stored at the harvest site will retain moisture more rapidly than windrows at landing during periods of precipitation. Consider sun and wind exposure during storage of windrows at landing.

2. Coverage of forest residues stored in windrows with a paper-based cover will protect the biomass from rewetting leading to lower moisture content and higher net calorific value. However, effect of coverage may be large on some landings and negligible in others. Consider location and season.

3. Biomass can easily be rewetted from the ground, consider effect of capillary rise of moisture from the ground.

4. Limit storage time of logging residues to as short as possible.

5. Avoid storing of forest residues over the winter time.

5.2 WOODCHIPS AND BARK

1. Strive to comminute to the largest possible particle size. Consider the boiler size requirements and remove fines (< 3 mm) before storage.

2. Avoid mixing various feedstock’s, species, material, biomass type, different moisture content, within the same windrow.

3. Minimize compaction during construction of windrows (e.g. avoid using heavy machinery on the piles).

4. Store wood chips and bark in a windrow shape. The height should not exceed 7 m, when the biomass consists of forest residue chips or bark.

5. Coverage with a semipermeable material or a paper-based material will protect the biomass from rewetting leading to lower moisture content and higher net calorific value. In addition, coverage will reduce dry matter losses and energy losses.

6. Limit storage time to 3-4 months if possible and expect a major temperature increase during the first weeks of storage.

Conclusions

Dry matter loss during storage is extremely important to consider since it affects both the revenue of the whole supply chain system as well as the environmental impact of forest biomass. The processes leading to dry matter losses are thoroughly reported and based on numerous studies aiming to improve fuel quality and reduce the risks associated with biomass storage. It is stated that different types of biomass and storage alternatives lead to different dry matter loss outcomes. Storage method, biomass origin, size and shape of the fuel, size and degree of compaction and storage time as well as ambient factors as e.g. temperature and humidity are all factors that simultaneously affect the processes leading to dry matter losses, and must all be taken into account. Several strategies can be used to reduce dry matter loss and should be considered during pile management. Minimizing variation in moisture content and control moisture content during storage is a key factor to improving fuel quality and reducing losses. Coverage of biomass, both loose logging residues stored in windrows and coverage of comminuted biomass have a clear advantage since it simultaneously protect the biomass form rewetting and reduce dry matter losses during storage. The effect of cover and its profitability is related to local weather conditions and storage time. Postponing comminution or comminution to a coarse fraction may also be considered since less surface area is known to decrease degradation rates. Transport and handling of biomass needs also to be considered to increase cost-efficiency in the supply chain. The development of models for predicting changes in fuel quality, temperature development within piles and dry matter losses during large-scale storage is important when tailoring optimized pile management and fuel quality during storage. The simultaneous development of storage guidelines and management models/tools will contribute towards uniform utilization of forest resources, a reduction in dry matter losses, less environmental impact and cost throughout the production chain. These efforts will undoubtedly elevate energy derived from biomass as a sustainable alternative to fossil fuels.

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