# Emissions from wood pellets during storage referring to the extractive content

Irene Schmutzer-Roseneder[1]* tel.:+43 7416 52238 51, email: Irene.schmutzer-roseneder@bioenergy2020.eu
Waltraud Emhofer[1] tel.:+43 7416 52238 49, email: waltraud.emhofer@bioenergy2020.eu
Walter Haslinger[1] tel.:+43 7416 52238 20, email: walter.haslinger@bioenergy2020.eu
* corresponding author
[1] BIOENERGY 2020+ GmbH, Gewerbepark Haag 3, 3250 Wieselburg-Land, Austria, fax:+43 7416 52238 99

**Abstract:** Wood pellets and wood raw materials such as chips or sawdust emit hazardous gases such as carbon monoxide (CO) and volatile organic compounds (VOCs) during processing and storage. Due to the high toxicity of CO it is necessary to identify the release mechanisms for CO and VOCs. Several studies show that organic extractives decrease during storage as well as the emissions. Therefore, the purpose of this study was to investigate a possible correlation between the organic extractive content and the release of CO and VOCs. Sawdust and pellets from Norway spruce (*Picea abies*), European larch (*Larix decidua*) and loblolly pine (*Pinus taeda*) were examined. Additionally, five different pellet samples from Austrian pellet producers were investigated. Soxhlet extraction with acetone was used to extract the organic content. The concentration of CO and VOCs was measured from stored wood materials and pellets in sealed glass flasks. The highest (3,41 mg CO/kg sample dm/d) and the lowest (0,02 mg CO/kg sample dm/d) release of CO were reported with freshly produced pine pellets and a spruce pellets sample from an Austrian do-it-yourself store, respectively. The results showed that the pelletizing process reduced the content of organic extractives. The emissions of pine samples concerning CO and VOCs were higher than of the spruce and larch samples. Moreover, the organic extractive content also decreased in that order. However, a direct correlation between organic extractive content and released quantities of emissions could not be established.

**Introduction:** Wood pellets are gaining more and more importance because of high energy density, consistent quality and increasing demand. Within the European Standard ÖNORM EN 14961-1 2010 various quality characteristics such as density, net calorific value, water content, ash content and durability are defined. In Austria 425 pellet heating systems (< 100 kW) were installed in 1997, however in 2011 about 10.400 heating systems were installed (Furtner and Haneder, 2011; Biermayr *et al.,* 2012). Furthermore the pellet production increased from about 450.000 t in the year 2005 to about 940.000 t in 2011 (ProPellets Austria, 2012; Biermayr *et al.,* 2012). This means that both the production and the consumption of pellets have risen.

In Gauthier *et al.* (2012) twelve fatalities referring to CO connected with woodworking, paper production and pellets are mentioned. According to Svedberg *et al.* (2004 and 2008) stored pellets emit apart from CO volatile organic compounds such as hexanal, pentanal and methanol. The oxidative degradation of organic extractives of wood is suggested as the most likely mechanism behind the formation of CO. Therefore, the chemical decomposition has an important influence on the release of CO, $CO_2$ and $CH_4$ (Kuang *et al.* 2008). The release rate of CO and VOCs as well as the content of fatty acids and resins decline during increasing time of storage (Emhofer and Pointner, 2009; Arshadi *et al.*, 2009). The emissions of pure spruce and pine pellets are investigated. The pine pellets emit more CO and VOCs than pellets produced from spruce. The different release rates of emissions are probably caused by the varying amounts of organic extractives and lipids (Arshadi and Gref, 2005).

**Methodology:** The release of carbon monoxide and volatile organic compounds as well as the content of organic extractives of sawdust and wood pellets were investigated. The analysed pellets were produced by a laboratory press at Bioenergy2020+. Various species of wood like European larch (*Larix decidua*), Norway spruce (*Picea abies*) and Loblolly pine (*Pinus taeda*) are examined. In additional experiments another five spruce pellet samples obtained in Austrian do-it-yourself stores were analysed. In the course of the experiment sawdust and wood pellets were filled in glass flasks. The sealed flasks were stored in a dark room at about 21 ±1° C temperature. After about 72 hours of storage the formed emissions were measured with a gas analyser (type Emerson NGA 2000) and a flame ionisation detector (type Horiba). Furthermore the organic extractive content of these wood samples were determined by Soxhlet extraction. For this experiment milled samples of pellets and sawdust were extracted for 6 hours in a Soxhlet apparatus using a mixture of acetone and water (96 % v/v). The inorganic extractives were separated by liquid-liquid extraction and the content of organic extractives was ascertained. The determination of the release of emissions as well as the content of organic extractives were carried out using at least three samples.

**Results:** The results of the determinations of the release of emissions in sealed flasks and the organic extractive content of various wood raw materials and pellets are shown in Table 1 and Table 2.

Table 1: Content of organic extractives and release of CO and VOCs of different wood raw materials (* dm/d = dry matter /day)

| Sample | Sample in kg | Storage time in d | Organic extractives in m % (dm) | CO release in ppm | CO in mg/kg sample dm /d* | VOCs release in ppm | VOCs in mg/kg sample dm /d |
|---|---|---|---|---|---|---|---|
| Larch Sawdust | 0,8 | 2,63 | 2,46 | 23 | 0,07 | 1 | 0,004 |
| Spruce Sawdust | 0,8 | 3,13 | 2,55 | 167 | 0,32 | 15 | 0,040 |
| Pine Sawdust | 0,8 | 3,04 | 3,49 | 257 | 0,62 | 55 | 0,142 |

The measured emissions were converted from ppm in mg/kg sample dry matter / day (dm/d) to enable comparability of the results. The highest (6089 ppm) and the lowest (23 ppm) formation of CO were reported with fresh pine pellets and larch sawdust, respectively. In this experiments 3 kg of fresh pine pellets released in sealed 5 l flask in average 6089 ppm CO. The formed concentration was high and would be toxic to humans (Hollemann *et al*., 1995).

All examined sawdust and pellet samples formed fewer VOCs than CO. The storage experiments of sawdust showed that larch emitted the least amount of CO and VOCs. The pine sawdust indicated the highest release of emissions, and contained a higher amount of organic extractives than spruce. Moreover, the spruce sawdust had a higher content of organic extractives than larch sawdust. For the sawdust samples it could be established that, regarding the formation of CO and VOCs arranged from lowest to highest, the samples also showed an increasing amount of extractive content.

Table 2: Content of organic extractives and release of CO and VOCs of various wood pellets

| Sample | Sample in kg | Storage time in d | Organic extractives in m % | CO release in ppm | CO in mg/kg sample dm /d | VOCs release in ppm | VOCs in mg/kg sample dm /d |
|---|---|---|---|---|---|---|---|
| Larch pellets | 3.0 | 2,63 | 1,76 | 70 | 0,03 | 8 | 0,004 |
| Spruce pellets | 2.5 | 2,75 | 1,99 | 1318 | 0,80 | 88 | 0,057 |
| Pine pellets | 2.5 | 2,75 | 3,43 | 6089 | 3,70 | 1865 | 1,216 |

A first comparison of the extractive content of the sawdust samples with the pellet samples produced from these sawdust samples showed, that the content of organic extractives was decreasing as an effect of the pelletizing process.

The larch sawdust and also the larch pellets had the lowest content of organic extractives and both samples released the least amounts of CO and VOC. The pine samples, in comparison, contained the highest amounts of organic extractives and also release the highest levels of emissions. The comparison of the amounts of released emissions for the spruce and the pine samples showed that the sawdust samples emitted fewer VOCs and CO than the produced pellet samples. This trend was also true for the larch sawdust sample which contained a higher amount of organic extractives than the larch pellet sample. However, contrary to the results from the spruce and pine samples, the larch pellets proofed to emit lower amounts of VOCs and CO than the larch sawdust sample.

In a subsequent experiment additional five spruce pellet samples obtained in Austrian do-if-yourself stores were analysed, to investigate the possibility of a correlation between extractive content and release of emissions from pellets more closely. In Table 3 the determined characteristics of pellets samples number 1 to 5 are listed.

Table 3: Content of organic extractives and release of CO and VOCs of pellets from Austrian market

| Pellet sample number | Organic extractives in m % | CO in mg/kg sample dm /d | VOCs in mg/kg sample dm /d |
|---|---|---|---|
| 1 | 1.73 | 0.06 | 0.013 |
| 2 | 1.92 | 0.10 | 0.012 |
| 3 | 1.98 | 0.02 | 0.003 |
| 4 | 2.15 | 0.24 | 0.064 |
| 5 | 2.85 | 0.02 | 0.004 |

The pellet samples were numbered according to their content of organic extractives starting from lowest to highest. Contrary to the initial hypothesis the results also showed, that the release rates of CO and VOCs did not follow the same sequence. For example, the pellet sample number 5 contained the highest amount of organic extractives, however pellet number 5 released the least amount of CO. Pellet sample number 4, on the other hand, emitted the highest amount of CO and contained the second highest amount of organic extractives.

**Discussion and Conclusion:**

The aim of this work was to find out if there are correlations between sawdust and pellets of different wood species, regarding the release of CO and VOC and the content of organic extractives. For the sawdust samples a correlation between the organic extractive content and the amount of emitted VOCs and CO could be established. The pine sawdust contained the most organic extractives, the spruce sawdust had the second highest content and the larch sawdust covered the fewest extractives. The pine sawdust also emitted the highest concentration of CO and VOC, and the lowest emissions of CO and VOC were reported with larch sawdust. For the pellet samples made from the sawdust samples a comparison of the contents of organic showed that the pellet samples contained fewer extractives than the respective sawdust samples, indicating that the pelletizing process reduced the content of organic extractives. Comparing only the pellet samples once more the pine pellets contained the highest amount of organic extractives and the larch pellets the lowest. Furthermore, the release rates of CO and VOC of the pellets samples were also arranged in the same order, from highest to lowest pine, spruce and larch pellets. According to these results there was a correlation between the release of CO and VOC and the content of organic extractives.

However, even though all produced pellets contained fewer extractives than the sawdust, the spruce and pine pellets showed a higher release of emissions than the spruce and pine sawdust. The same trend could not be seen in the release rate of CO for the two larch samples, the larch pellets emitted less CO than the larch sawdust. However, due to the overall low measured CO values during the measurements of the larch samples, the calculated emission rates for these samples were also afflicted with a higher margin of error. Therefore, we concluded that the pelletizing process stimulated and enhanced the release of CO and VOC in the pellets in comparison to the raw material.

In order to look at a possible correlation between the organic extractives and the release of emissions more closely, the CO and VOC release rates of five different pellets available on the Austrian market were investigated. Hence the order of the content of extractives differed from the ranking of the release rates no direct correlation between the formed emissions and the content of extractives was observed. The influence or a possible correlation between the individual fatty acids and resins to the release of CO and VOC of wood sawdust and pellets were not analyzed. Moreover the investigation of the composition of the extractive content and the change of composition during the pelletizing process could provide additional information on the cause of the release of emissions. Additionally, the pelletizing parameters temperature and pressure as well as the storage duration and type of the used sawdust and pellets may have impacts on the later formed emissions.

Furthermore it could be shown, that even small amounts of fresh pellets could produce a high value of CO (see pine pellets). Therefore, it is really essential to learn what is responsible for the high formation of CO. For that the pellet production process and the release of emissions must be further investigated. In addition, safety instructions for trading, using and storing pellets are required to protect the user along the entire pellet supply chain.

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