

Emission of Volatile Aldehydes and Ketones from Wood Pellets under Controlled Conditions

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Different qualities of biofuel pellets were made from pine and spruce sawdust according to an industrial experimental design. The fatty/resin acid compositions were determined by gas chromatography-mass spectrometry for both newly produced pellets and those after 2 and 4 weeks of storage. The aldehydes/ketones compositions were determined by high performance liquid chromatography at 0, 2, and 4 weeks. The designs were analyzed for the response variables: total fatty/resin acids and total aldehydes/ketones. The design showed a strong correlation between the pine fraction in the pellets and the fatty/resin acid content but the influence decreased over storage time. The amount of fatty/resin acids decreased ~40% during 4 weeks. The influence of drying temperature on the aldehyde/ketone emission of fresh pellets was also shown. The amounts of emitted aldehydes/ketones generally decreased by 45% during storage as a consequence of fatty/resin acid oxidation. The matrices of individual concentrations were subjected to multivariate data analysis. This showed clustering of the different experimental runs and demonstrated the important mechanism of fatty/resin acid conversion.

Keywords: aldehydes; fatty acids; industrial experimental design; multivariate data analysis; softwood pellets

INTRODUCTION

Fuel pellets made of sawdust represent a renewable energy source for heat production and the use of fuel pellets will increase in the future. About 1 million tons of wood pellets were manufactured in Sweden in 2004, making this country the biggest fuel pellet producer in Europe (Larsson, 2004). The principal raw materials for pellets in Sweden are fresh and stored sawdust of Norway spruce and Scots pine together with cutter shavings of these species (Lehtikangas, 2001).

In the pelletizing process, sawdust is dried at an elevated temperature, ground, and pressed through the holes of a steel die to form cylindrical pellets. Finally, the warm pellets are cooled and stored.

At high temperatures, in lumber drying and hot pressing of boards, the content and composition of lipophilic wood extractives are modified by auto-oxidative reactions (Hemingway *et al.*, 1971; Back and Allen, 2000; Manninen *et al.*, 2002). Liberated

unsaturated fatty acids may undergo cleavage and oxidation of double bonds and form volatile aldehydes (Bengtsson and Sanati, 2004; Makowski *et al.*, 2005). Such reactions could also be expected to occur during the pelletizing process and in stored pellets.

It has been reported that under certain conditions, stored pellets emit high levels of volatile organic compounds (VOCs) (Svedberg *et al.*, 2004). Previous studies (Arshadi and Gref, 2005) have suggested that the amount and composition of VOC emitted from stored pellets were strongly correlated with the drying temperature and quality of the raw material and with self-heating in the stored pellets.

The major constituents of VOC emitted from wood pellets are aldehydes some of which are known to be upper airway irritants (Hagström, 2008). Aldehydes are also known irritants to eyes and mucous membranes and methanal and ethanal are suspected carcinogens. Methanal is also known to be sensitizing (Fjällström, 2003). Pentanal and hexanal are known to cause unpleasant and irritant odors in closed spaces (Jensen *et al.*, 1995; Risholm-Sundman *et al.*, 1998; Back and Allen, 2000). For hexanal, which is formed through

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auto-oxidation of linoleic acids (Back and Allen, 2000), there is no threshold limit value for occupational exposure and scientific information about the potential health risk is insufficient (Svedberg *et al.*, 2004).

The aim of the present study was to investigate the influence of the raw material and the process parameters on fatty/resin acids composition in pellets and the emission of aldehydes/ketones from fresh and stored pellets. The study was carried out in an industrial production plant using an experimental design and multivariate data analysis.

MATERIAL AND METHODS

Wood pellets

Pellets were made from fresh sawdust of Norway spruce (*Picea abies* Karst) and Scots pine (*Pinus sylvestris* L), in a large-scale pellet production plant in Sweden. Eleven different piles of pellets, each pile containing ~12 tons, were stored in the plant for 30 days. Temperatures were logged in all piles every hour during the whole experiment. Newly produced (fresh) pellets from the piles are referred to as P1 to P11 (P standing for pile). Two-week-old pellets from the piles are referred to as P1-2 to P11-2 (2 indicating the number of storage weeks) and 4-week-old pellets P1-4 to P11-4 (4 indicating the number of storage weeks). The sampling time was between 5 April 2005 and 4 May 2005. Samples were collected from 10 different positions in each pile and mixed to prepare a general sample (~10 kg). After collection, all samples were kept in sealed gastight plastic bags at +2°C and analyzed within 1–4 days. The moisture content of the fresh pellet piles varied between 2.7 and 8.1% of the dry weight.

Chemicals

Dinitrophenylhydrazone derivatives of 2-ethylhexanal, octanal, and nonanal were used as reference substances and synthesized according to previous work (Shriner *et al.*, 2004). Reference solutions of other aldehydes (DNPH Mix 2, Supelco) were also used for identification. Eight aldehydes—methanal, ethanal, propanal, butanal, hexanal, 2-ethylhexanal, heptanal, and octanal—were purchased from Aldrich. Pentanal and nonanal were obtained from Fluka. Dinitrophenylhydrazine (DNPH) cartridges (LpDNPH H10 and LpDNPH S10) were obtained from Supelco. Acetonitrile [high performance liquid chromatography (HPLC) grade S] was used for the elution of samplers and for the liquid chromatography mobile phase (Rathburn, Peeblesshire, UK). Tetrahydrofuran (THF) (Merck) was used as the second mobile phase.

Extraction and analysis of fatty acids and resin acids

Fatty acids and resin acids in wood pellets were extracted in a Soxhlet apparatus with a mixture of

petroleum ether (bp 40–60°C) and acetone (90 to 10 v/v) as the solvent for 6 h and analyzed by GC-MS according to a previously reported method (Arshadi and Gref, 2005; Samuelsson *et al.*, 2006). A complete list of all fatty acids and resin acids and their maximum concentrations in the pellets is available in Table 1.

Aldehyde and ketone sampling

The pellet samples were analyzed by using a laboratory setup designed specifically for the purpose (Fig. 1). The air was taken from the surrounding room and passed through an airflow meter (1) into the first water trap (2) and into an absorbing carbon tube that acted like a contamination trap (3). The airflow was introduced into an oven (4) (kept at +60 ± 2°C) via a Teflon tube to make sure that it was heated

Table 1. Maximum concentration of fatty/resin acids extracted from fuel pellets

No.	Compound	Concentration ($\mu\text{g g}^{-1}$)
1	Octanoic acid	8.1
2	Nonanoic acid	17.2
3	Decanoic acid	5.2
4	Dodecanoic acid	5.2
5	Tridecanoic acid	2.3
6	2-Undecenoic acid	0.9
7	Tetradecanoic acid	16.8
8	Pentadecanoic acid	16.4
9	Pentadecanoic acid, anteiso	12.3
10	Hexadecanoic acid, anteiso	18.5
11	9-Hexadecenoic acid	27.0
12	Hexadecanoic acid	150.7
13	Heptadecanoic acid, anteiso	102.8
14	9-Octadecenoic acid	193.1
15	9,12,15-Octadecatrienoic acid	39.0
16	9,12-Octadecadienoic acid	131.7
17	11-Octadecenoic acid	696.5
18	Octadecanoic acid	86.6
19	Pimaric acid	239.2
20	Pimaric acid, isomer	45.0
21	Isopimaric acid	77.9
22	Abietic acid	154.8
23	Dehydroabietic acid	1164.5
24	Abietic acid, isomer	70.7
25	7-Oxo-dehydroabietic acid	888.3
26	11-Eicosonic acid	24.2
27	Docosanoic acid	33.5
28	Dehydroabietic acid, isomer	39.0
29	Arachidonic acid	13.5
30	Undecanoic acid	21.0
31	Tricosonic acid	20.9
32	9-Octadecenoic acid	18.6

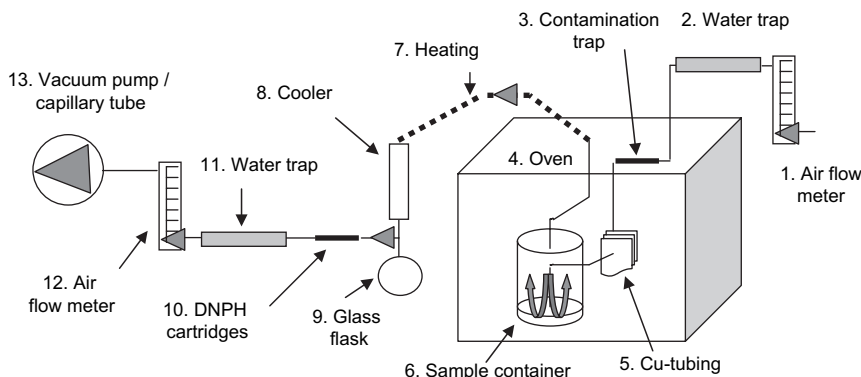


Fig. 1. Laboratory setup designed for sampling of aldehyde/ketone emission.

to 60°C and then passed through a 2 m long coil of copper tubing (5). The airflow was split after the copper tubing using one to three parallel lines depending on how many samples were to be run at the same time. The heated air was led into sample containers (6), which contained the experimental material. The evaporated gases (containing all VOCs) were then passed through a Teflon tube surrounded by a heating mantle (7), which extended all the way to coolers (8) to prevent early condensation. The water vapor was then condensed in a carefully monitored glass flask (9). This was done so as not to flood the samplers since the heated pellets initially gave off considerable amounts of moisture. The collected water evaporated slowly before the sampling was terminated and the flask was washed to make sure no compounds of interest were lost. The volatile components were subsequently trapped in the DNP cartridges (10). After that, the gas passed a second water trap (11) filled with magnesium perchlorate, to enable monitoring of the amount of water released from the pellets during the experiment. Airflow regulators (12) were placed on each line to set the flow through the system (200 ml min⁻¹) and the three lines were reduced to one and connected to a vacuum pump (SIDEPAK™ Personal Sampling Pump SP 730 from TSI Inc.) (13).

Approximately, 10 g of pellets were placed into the sample container and pumping was started as soon as possible and maintained for 49 h. In total, ~600 l of air passed through each sample and collected all aldehydes and ketones emitted from the samples in seven DNP cartridges during this time. Samples were collected at $t = 0, 40 \text{ min}, 2 \text{ h}, 6 \text{ h}, 12 \text{ h}, 24 \text{ h},$ and 48 h and each sampling was maintained for 20–60 min. The aldehydes and ketones were converted to corresponding hydrazones by reacting them with DNP inside the cartridges. The hydrazones were released by elution with acetonitrile. 2-Ethylhexanal (112 µg), as an internal standard, was added to each sample. Thereafter, each sample was analyzed by HPLC.

Gas chromatography–mass spectrometry

A Hewlett-Packard (HP6890-5973) gas chromatography–mass spectrometry (GC-MS) instrument with an auto-sampler operating in the electron impact mode (EI 70 eV) was used. The instrument was equipped with a 30 m × 0.25 mm i.d., 0.25-µm film HP-5 MS capillary column coated with cross-linked 5% phenyl methyl siloxane. The column was temperature programmed as follows: 100°C isothermal for 0 min, to 220°C at 10°C min⁻¹, then to 235°C at 1°C min⁻¹, then to 260°C at 10°C min⁻¹ and held for 1.5 min. Aliquots of 1 µl silylated samples, prepared according to previous work (Ånäs *et al.*, 1983; Ekman and Holmbom, 1989; Örså and Holmbom, 1994), were injected. For quantitative analysis of fatty acids and resin acids, an internal standard, heptadecanoic acid, was used.

HPLC

HPLC was used for the measurement of DNP derivatives of aldehydes and ketones. The separation was carried out on a Crom-sil 120 Butyl-1 SF 5 µm 150 × 4 mm-column thermostated to 20°C, flow rate 1 ml min⁻¹. Before use, the column was conditioned with 65% water/THF (90/10 v/v%) referred to as (A) and 35% acetonitrile referred to as (B). The following binary mobile phase was used: isocratic elution 65% (A) and 35% (B) 0–7 min, linear gradient from 35% B to 60% B, 7–18 min, to 65% B, 18–25 min, there it was maintained 25–27 min. Thereafter, the concentration was returned to the initial value for 27–32 min. The ultraviolet detector recorded a signal at 360 nm. In total, 20 µl of each sample was injected and 2-ethylhexanal was added as an internal standard to each sample. Known reference substances were used to identify the aldehydes.

Experimental design

An experimental design is a systematic way of carrying out experiments in order to obtain optimally interpretable results that can be tested for statistical

significance and incur minimal effort or cost (Box *et al.*, 1978; Myers and Montgomery, 1995). When used industrially, a number of factors (x_1 , x_2 , x_3 , ...) which can influence the result are identified and set at fixed levels. For each combination of levels chosen (also called a run), the result of the experiment, called the response (y), is recorded.

This allows the construction of a model equation:

$$y = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_{12}x_1x_2 + b_{13}x_1x_3 + b_{23}x_2x_3 + e,$$

where

y : the response

b_0 : the constant term

b_1 – b_3 : the regression coefficients

b_{12} , b_{13} , and b_{23} : the interaction regression coefficients

x_1 – x_3 : the factors

e : the residual, often assumed to be normally or t -distributed with a zero mean.

The most popular setting of the factor levels is a full factorial design, sometimes with extra center points, see Table 2. The center points are added to estimate replicate error and they are placed at the beginning, middle, and end of the sequence of runs.

The final result will then be that: (i) unimportant (insignificant) factors are ignored in the future; (ii) important (significant) factors are used at the correct settings that give an optimal response; and (iii) new tests are carried out close to the optimal settings. The three factors chosen were drying gas temperature, ingoing material moisture content, and mixture composition of spruce/pine. The levels for these factors are given in Table 2.

Table 2. Settings of the design with three center points

Run	x_1	x_2	x_3	y	x_1 real	x_2 real
P1	400	9	55	y_1	396	9.5
P2	350	11	10	y_2	363	11.6
P3	450	7	10	y_3	457	7.4
P4	350	7	10	y_4	355	7.5
P5	450	11	10	y_5	461	11.7
P6	400	9	55	y_6	401	10.2
P7	350	11	100	y_7	332	11.6
P8	350	7	100	y_8	329	8.0
P9	450	11	100	y_9	375	10.6
P10	450	7	100	y_{10}	380	7.8
P11	400	9	55	y_{11}	400	9.8

x_1 is planned drying gas temperature ($^{\circ}\text{C}$), x_2 is planned moisture content of sawdust (%), and x_3 is the percentage of spruce and the rest is pine in percent. The responses y_1 – y_{11} are filled in after each run. The responses are fatty/resin acids and aldehydes/ketones. x_1 real and x_2 real are the accurate process mean values measured during each experiment. P = pile number.

Table 2 contains not only the ideal factor levels for a full factorial design but it also contains obtainable levels used in practice. In this case, it was not possible, practically, to reach the factor levels of drying gas temperature and moisture content of the sawdust. After the experiments, it was possible to collect process data and to use the exact values in the calculations. The other difficulty was that the order of the runs could not be fully randomized. From a practical point of view, it was also not possible to randomize mixtures of spruce and pine sawdust. Therefore, the mixtures of 10% spruce and 90% pine were run in a sequence (Run 2–5) and 100% spruce were done in another sequence (Run 7–10). This is called blocking.

Two responses were used: total emitted aldehyde/ketone concentration from the pellets and total fatty/resin acid concentration in the pellets. These were measured fresh and after 2 and 4 weeks of storage. For the aldehyde/ketone response, temperature profile results after 2 weeks showed stabilization and only a few responses after 4 weeks were measured.

In realistic industrial situations, large piles of pellets are stored and expected to self-generate heat. This self-heating is dependent on, for example, heat transfer and thus on pile size. Therefore, it was necessary to make pellet piles large enough to be able to compare the results from the storage experiments with realistic storage conditions in industrial pellet plants.

Multivariate data analysis

The response in Table 2 may be a single value, e.g. a product quality index, but sometimes there is more than one response. The use of spectroscopy or chromatography can lead to tens of responses for each run. In this study, the response comprises several data from GC-MS and HPLC. In such a case, the responses form a data matrix. In this work, there were 11 samples, each giving a maximum of 32 GC peaks for each fatty/resin acids. The result then is an 11×32 data matrix. A simple statistical analysis is not possible if the responses in the matrix are correlated, so in this case, multivariate data analysis is needed. In the example of an 11×32 data matrix, the 11 rows are called the objects and the 32 columns are called the variables.

Principal component analysis (PCA) is a technique used for data reduction (Jackson, 1991). It also allows the detection of outliers, clustering in the data and the visual estimation of variability in the clusters. What PCA does is construct new axes in the data that are a better representation of the important phenomena, the principal component axes. These new axes can then be used to make plots in which the runs can be seen as points (score plots) and plots in which the variables can be seen as points (loading plots). This is illustrated in the 'Results and discussion' section.

An important diagnostic is how much (as %) of the original sum of squares of the data in the data matrix is explained by the new axes (Beebe *et al.*, 1998; Brereton, 2003).

If two principal components explain 97.1% of the sum of squares of an 11×32 matrix, then this is a substantial data reduction.

RESULTS AND DISCUSSION

The moisture content of the pellet piles changed during storage. The driest pellets were the most hygroscopic. In one case, the moisture content increased from 2.65 to 4.74% over 4 weeks of storage. For pellets with an initial moisture content $\sim 8\%$, almost no change was detected.

Fatty/resin acids identified in pellets by GC-MS are presented in Table 3. In this study, the amounts of di- and trienoic fatty acids in pellets were lower compared to fresh wood (Holmbom and Ekman, 1978), which confirms that di- and trienoic unsaturated fatty acids are more sensitive to oxidation than monoenoic fatty acids (Back and Allen, 2000). The main resin acids in the pellets were dehydroabietic acid and 7-oxo-dehydroabietic acid which were in greater amounts compared to typical fresh pine and spruce wood (Holmbom and Ekman, 1978).

The total concentration of aldehydes/ketones emitted from newly produced and stored pellets is shown in Table 4. The following aldehydes and ketones were detected: methanal, ethanal, acrolein, 2-propanone, propanal, butanal, benzaldehyde, pentanal, toloulaldehyde, hexanal, octanal, and nonanal.

The drying gas temperature is the only significant factor for aldehyde/ketone emission, but only for fresh material. The pine fraction, which is also the blocking variable, gave no significant coefficient. From this it can be concluded that the blocking was not detrimental to the experiment. This is due to

differences in chemical composition of pine and spruce extractives. This temperature effect on aldehyde/ketone emissions was also mentioned in the paper by Arshadi and Gref (2005). The average result (b_0) for total aldehyde/ketone emissions decreased by 45% after 2 weeks. This is in good agreement with the report on auto-oxidation of lipids in pellets during storage (Arshadi and Gref, 2005).

The *t*-tests for the fatty/resin acids showed the following results: no significant effect for drying gas temperature and moisture content and no significant effect for the interactions. The fraction of spruce content is extremely significant and negatively correlated, but this significance diminishes with storage. The blocking is of course confounded with the spruce fraction, but it is unlikely that a highly significant factor should come from the blocking. The average fatty/resin acid content decreased steadily over time, by $\sim 20\%$ after 2 weeks and by $\sim 30\%$ after 4 weeks referred to initial values. These findings are supported by studies on wood chips (Quinde and Paszner, 1991; Back and Allen, 2000).

The design results are for the total and individual amount of aldehyde/ketone emissions and fatty/resin acid concentrations. There are more responses available, but then as individual ketone/aldehyde emissions or fatty/resin acid concentrations. Experimental designs with many individual responses of fatty/resin acids and aldehydes/ketones give tedious and confusing interpretations (Arshadi *et al.*, 2008). Therefore, it was considered more appropriate to use the response data for exploratory multivariate data analysis (Geladi and Forsström, 2002).

The data matrix of the fatty/resin acids contained 33 objects (11 runs at 0, 2, and 4 weeks) and 32 variables after removal of those acids that are always below the detection limit. The data were pretreated by mean centering over the variables. The resulting data matrix gave a very good PCA model with two

Table 3. Total concentration of fatty/resin acids ($\mu\text{g g}^{-1}$) in newly produced pellets, after 2 and 4 weeks of storage in 11 piles (P1 to P11)

Experiment no.	Fresh	2 Weeks storage	4 Weeks storage
P1	2297	1699	1444
P2	3444	2421	2071
P3	3587	2804	2141
P4	3501	2463	2091
P5	3239	2480	2091
P6	2461	1567	1284
P7	736	1503	955
P8	836	724	832
P9	819	854	832
P10	739	814	878
P11	2017	2221	2250

Table 4. Total concentration of aldehydes/ketones ($\mu\text{g g}^{-1}$) emitted from newly produced pellets and after 2 and 4 weeks of storage in 11 piles

Experiment no.	Fresh	2 Weeks storage	4 Weeks storage
P1	1806	288	—
P2	1235	523	—
P3	2903	377	422
P4	1730	289	—
P5	1996	2878	1900
P6	2045	1129	1809
P7	1244	1165	—
P8	1092	556	—
P9	1113	936	1056
P10	1003	1603	1183
P11	2017	944	—

components explaining 97.1% of the total sum of squares. The first component explained 85.4% and the second one 11.7%.

The experimental runs are shown in a score plot, Fig. 2a. The horizontal direction in the score plot is a reduction in spruce content or an increase in pine content. Cluster C on the left side shows that 100% spruce samples have very low variation over time. Also, the center points cluster together (Cluster B). The high pine fraction samples with fresh pellets are in Cluster A. The second component (direction toward D) is the difference between fresh and stored pellets, most marked for the high pine samples and the center points. The loadings are shown in Fig. 2b. The strongest variables in component one (furthest

away from the center) are 7-oxo-dehydroabietic acid and dehydroabietic acid. The second component explaining the difference in storage time is made up of a contrast between 7-oxo-dehydrodiabetic acid and 11-octadecenoic acids. Exposed to air and heat, dehydroabietic acid is oxidized to 7-oxo-dehydroabietic acid. 7-Oxo-dehydroabietic acid is generally not further oxidized (Anderson *et al.*, 1992), see Fig. 3. This oxidation reaction has been used as an indicator for pellet maturation (Arshadi and Gref, 2005; Arshadi *et al.*, 2007). In Fig. 2b, one can see a negative second component loading for 11-octadecenoic acid as this compound is missing in spruce. Figure 3 shows that in pine, the concentration of resin acids generally decreased during storage. In pellets made of spruce,

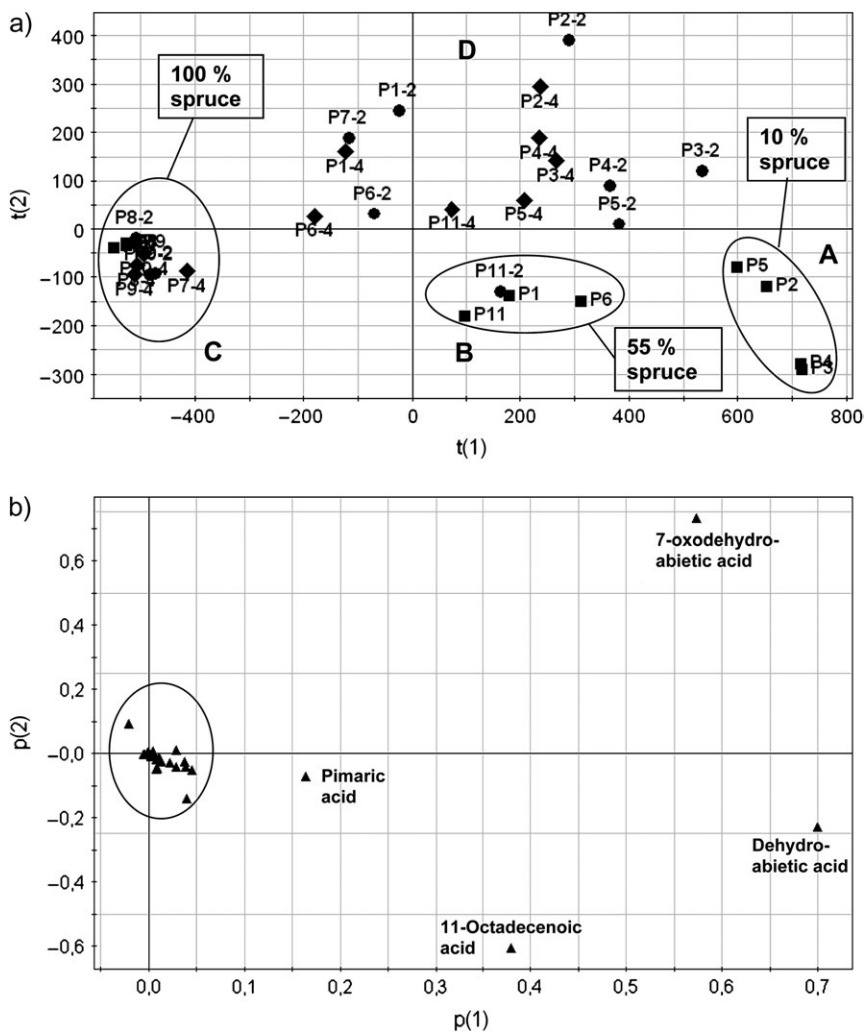


Fig. 2. (a) Score plot of the first two components from the PCA model. A clear clustering as a function of raw material and storage times can be observed. A, fresh pellets with 90% pine fraction; B, fresh pellets center points; C, 100% fresh and stored spruce fraction pellets; D, direction of aged pellets containing mainly pine. P1–P11, the runs of the experimental design after 0 weeks; P1-2 to P11-2 after 2 weeks; and P1-4 to P11-4 after 4 weeks. (b) Loading plot of the first two components from the PCA model. The cluster of variables close to zero has no influence on the two-component model. The variables that are far from zero are important ones for the model.

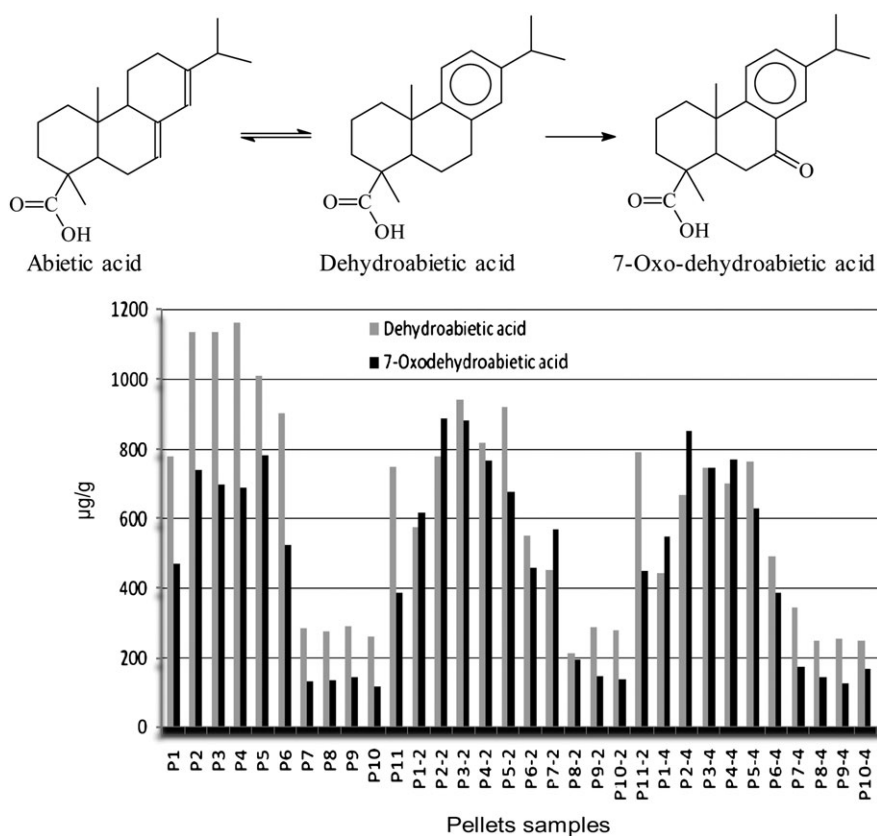


Fig. 3. Concentration of dehydroabietic acid and 7-oxo-dehydroabietic acid in pellets: newly produced (P1 to P11), stored 2 weeks (P1-2 to P11-2), and stored 4 weeks (P1-4 to P11-4). The oxidation path is also shown in the same figure.

the changes in concentration of resin acids are about the same as the variation in the analytical method.

The aldehyde/ketone data set was originally 27 objects and 17 variables. The first 11 objects were the fresh sample runs and the objects 12–22 were runs for 2 weeks of storage. Two outliers were removed ending up with a 25×17 matrix. This matrix was mean-centered and scaled to equal variance. A PCA model gave two components explaining 54 and 11% of the sum of squares. The first component explains mainly the difference between high and low spruce levels with a small effect of drying gas temperature for the fresh samples. For the stored pellets, a high variation in the results was found. Therefore, the data set was reduced to only contain fresh samples and the results of the PCA analysis are shown in Fig. 4. The first and second component explains 58 and 17% of the sum of squares. Separation can be seen in the first component direction with spruce samples clustered at low score values and pellets containing pine spread out at higher score values.

CONCLUSIONS

The results of the experimental designs implemented showed that the content of fatty/resin acids

in pellets decreased in a large-scale storage situation over 4 weeks. The only significant factor in this experimental design was spruce content, while the other factors were insignificant. The response variable aldehydes/ketones had drying gas temperature as a significant factor for fresh pellets. For stored pellets, no factors were significant. For both the fatty/resin acids and aldehydes/ketones, substantial decreases over time of the average levels were noted. The multivariate data analysis of the individual fatty/resin acid concentrations showed the importance of both fatty acid and resin acids. For the aldehydes/ketones, the multivariate data analysis showed that the content of pine and drying gas temperature are the two factors that have a positive effect on the formation of all detected aldehydes.

In the piles of pellets made of pine, the temperature evolved over storage time because of fatty/resin acid oxidation as shown in Fig. 5. The reduction in fatty/resin acids in pellets made mostly of pine caused lower emission of aldehydes/ketones after 2 weeks of storage. The decrease in dehydroabietic acid corresponded to an increased content of 7-oxo-dehydroabietic acid. So the accumulation of 7-oxo-dehydroabietic acid could be used as an indicator of pellet maturity. A possible and reasonable

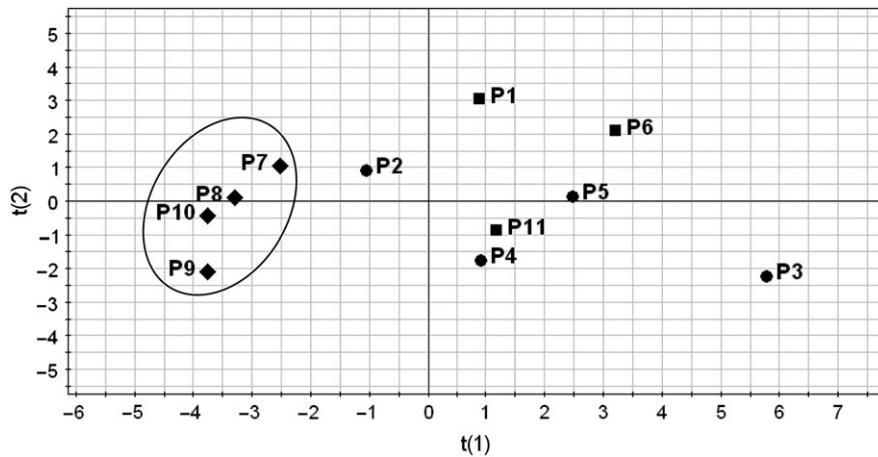


Fig. 4. Score plot of the first two components from the PCA model for concentration of emitted aldehydes/ketones from newly produced pellets. Pellet samples made of 100% spruce (P7 to P10) are separated from the other pellet samples which are mixtures of pine and spruce. Filled circles indicate P2 to P5 samples with 10% spruce; filled squares indicate P1, P6, and P11 samples with 55% spruce; filled diamonds indicate P7 to P10 samples with 100% spruce.

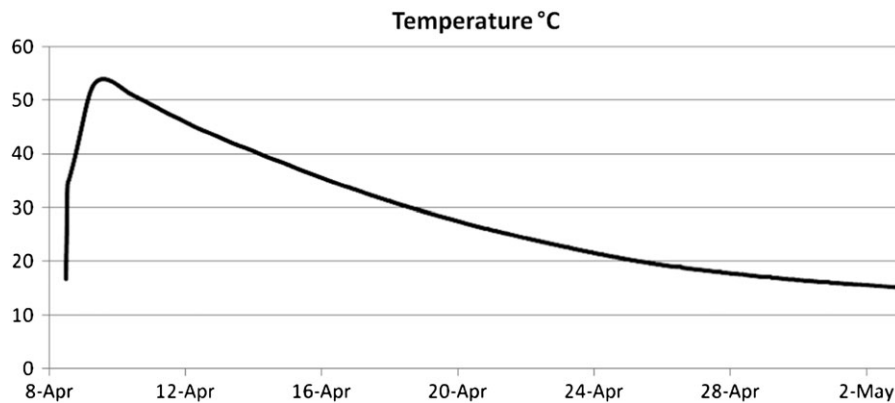


Fig. 5. A typical temperature logger profile as measured in a wood pellets pile.

definition of maturity is that fatty acid and resin acid concentrations have stabilized (Arshadi *et al.*, 2007). The oxidation of unsaturated fatty acids gives rise to aldehyde/ketone emissions from pellets. In pellets made from spruce, the emissions of aldehydes and ketones are significantly lower than in pellets made from pine due to the lower amount of fatty acids in spruce. By choosing the optimal raw material mixture of pine and spruce and process parameters, it should be possible to reduce the aldehyde emissions from pellets. Emissions of aldehydes/ketones cause unpleasant odors in storage rooms. A high drying gas temperature used for drying the sawdust leads to higher emissions of aldehydes/ketones from pellets. The high drying gas temperature initiates break down of fatty/resin acids resulting in emissions from wood pellets after production and during storage. In the worst case, an emission of total aldehydes of 7.5 g h^{-1} is possible from fresh pine pellets. After 4 weeks, this emission is reduced to 13%. The results

from this study show that with optimal production parameters it is possible to reduce the amount of these aldehydes during storage of wood pellets and also reduce any possible risk to the health of workers in the pellets industry and users in the households.

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