

## FLUE GAS ANALYSIS OF APPLE AND GREY ALDER WOOD PELLETS IN A MEAT SMOKEHOUSE CHAMBER

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### Abstract

The use of the smoke released during the wood burning process to prepare food products is a centuries-long tradition, practically all over the world. However, during the combustion process, a group of compounds called polyaromatic hydrocarbons (PAHs) are formed in the flue gases, which are carcinogenic and condense during the smoking process and diffuse into the smoked food product. Therefore, permissible PAH norms have been set for food producers, which significantly complicate the use of wood. In the study, using a gas analyzer, we measured the flue gases released during the burning of specially made, apple and gray alder wood pellets, with and without enrichment of the supplied air with ozone. The use of ozone does not ensure a higher burning temperature of pellets, but it stabilizes it – temperature fluctuations are significantly wider using non-ozonated air (697 to 817 and 611 to 817 °C, respectively). The content of CO<sub>2</sub>, CO, as well as CH<sub>4</sub> and N<sub>2</sub>O increases significantly in apple wood flue gases using ozonated air, while CH<sub>4</sub> increases and N<sub>2</sub>O decreases in gray alder smoke. Which generally indicates specific reactions with ozone during combustion. Comparing the flue gases released during the burning of apple and grey alder wood pellets, grey alder smoke contains significantly more N<sub>2</sub>O and CO<sub>2</sub> than apple wood pellet flue gases. On the other hand, using ozonated air in the combustion process increases N<sub>2</sub>O significantly in the flue gas of apple tree pellets compared to white alder.

**Key words:** ozone, smoking meat, polyaromatic hydrocarbons (PAHs).

### Introduction

Apple and grey alder wood are used to produce smoked foods using traditional technologies (Stumpe & Viksna, 2009). However, during the smoking process on the surface products condensate and absorb polyaromatic hydrocarbons (PAHs) that are present in smoke, followed by the diffusion of PAHs within the product. PAH compounds are cancerogenic and in order to protect consumers, norms have been introduced that food manufacturers must fulfill. As a result, technologies were introduced into production that use different smoking liquids (liquid smoke) with different tastes and smells, or use short, intense periods of smoking. However, the problem is relevant both for manufacturers who stick to traditional technologies and for all home manufacturers. The PAH content of smoked foods depends on the humidity, burning temperature, oxygen concentration and air exchange fan rate of wood used in the smoking (Hitzel *et al.*, 2013, Shkaljac *et al.*, 2014).

Wood burning process is the oxidation of the organic part of the solid – conversion to combustion products of flue gas. The combustion theory states that the final products for the total combustion of wood are carbon dioxide (CO<sub>2</sub>), water vapor H<sub>2</sub>O, nitrous monoxide (NO), nitrous oxide (NO<sub>2</sub>), which is also, of course, ash and mixed minerals of different origin, as well as metals: KCl, SO<sub>2</sub>, HCl, Cu, Pb, etc., while wood is not completely burned in addition to Carbon monoxide (CO), CxHy, tar, soot, H<sub>2</sub>, N<sub>2</sub>O, polyaromatic hydrocarbons, etc. (Williams *et al.*, 2012; Van Loo *et*

*al.*, 2008; Obernberger *et al.*, 2006; Nussbaumer *et al.*, 2003). This points to the importance of ensuring the complete burning of wood for smoked products, since the least of the compounds produced by PAHs are released. Only nitrogen compounds formed by the conversion of wood nitrogen to N<sub>2</sub> and nitrogen oxides (NO, NO<sub>2</sub>) remain in practice (Williams *et al.*, 2012; Van Loo *et al.*, 2008).

In order to ensure the complete burning of wood, it is necessary to ensure burning temperature is above 850 °C and air supply (higher than the calculated consumption factor) and good gas mixing (Van Loo *et al.*, 2008). The inflow of air into the combustion chamber shall be provided artificially by means of fans, providing continuous oxygen supply directly in the combustion area. However, wood burning at a high and uniform temperature is difficult to achieve, due to the heterogeneous properties of wood (wood density, moisture, etc.), which today is successfully solved by wood pelletizing. Studies have found that adding ozone is a promising method for improving and controlling burning processes. In recent decades, the impact of the addition of ozone on combustion processes has been widely studied, ranging from simple heating burners to internal combustion engines (Wenting *et al.*, 2019). Ozone has demonstrated the ability to accelerate ignition and control the timing of the ignition, to improve the spread and stabilization of the flame, affecting emissions of flue gases and reducing the formation of certain pollutants (Docquier & Candell, 2002; Wenting *et al.*, 2019). This

improvement is closely linked to ozone chemistry, particularly ozone decomposition, to obtain atomic oxygen and rapid exothermic ozonizes reactions with unsaturated hydrocarbons (Greene & Atkinson, 1992; Atkinson & Carter, 1984). Ozone decomposition at relatively low temperatures is responsible for most improvements in combustion achieved through  $O_3$ . Such reactions release reactive atom oxygen and accelerate the chain's branching process.

The aim of the study was to find out the composition of the flue gases released during the combustion process of grey alder and apple wood pellets in the chamber of the smokehouse, enriching the air supplied during the combustion process with and without ozone.

### Materials and Methods

The wood pellets are made of apple and grey alder wood without bark, from each tree species of  $6 \pm 1$  and  $8 \pm 1$  mm in diameter, moisture content respectively apple  $6.5 \pm 0.3$  and  $6.9 \pm 0.2$ , grey alder  $6.2 \pm 0.4$  and  $7.0 \pm 0.2$ , length from 5 to 35 mm, mechanical strength  $\geq 97.5\%$ . The produced pellets corresponded to the quality indicators of class EN2 (ENplus Handbook).

Each wood pellet type depending on trees species and size were burned in an experimental furnace (Figure 1) in three repetitions, total 12 experiments. Each experiment started from 'cold' burning furnace. Pellets from the tank (volume 3 liters) were transferred to a combustion chamber with a thread-type rotating shill of 40 mm diameter, a rotation rate of 6 rotations per minute. The air supply in the combustion chamber was constant – provided with a fan (airflow  $10 \text{ m}^3 \text{ h}^{-1}$ ). In addition, the possibility of aerial enrichment with ozone is provided. We used 6 high-voltage discharge elements with a nominal capacity of  $60 \text{ g h}^{-1}$  to generate ozone. The ozone generators were queued in a stainless metal housing and paired. Respectively, each pair of ozone generators, worked in 15 second intervals to prevent them from overheating.

The flue gases released during the pellets burning were passed through the flue-canal into the experimental smoker. Flue gases have been measured in a meat smoking chamber (Figure 1) by pumping a flue gas through a special type of tube that is built into a door and ensures the absorption of flue gases from the middle of the chamber. Flue gas measurements were started when the temperature of the chamber reached a working temperature of  $\sim 90 \pm 5 \text{ }^\circ\text{C}$ .

For the flue gas analysis, we used Gasmeter DX4040. The calibration is made before starting the measurements and is valid for a maximum of 24 hours or until a sudden change in the temperature of the environment where gas analyzer is located. The gas used to perform the calibration is nitrogen 5.0. The

analyzer is cleaned out of this gas until stable values are achieved, which is close to zero to be obtained for the gases to be analyzed. A measurement of the ambient air is carried out first in order to identify the starting level. For example,  $\text{CO}_2$  should be around 400 ppm. If this is not the case, the smoking chamber must be ventilated. Probe installation. The probe is introduced into the chamber through the door close to the center of the chamber. The pipe connecting the probe to the analyzer passes through a cooling circuit in order to force the condensation of water vapor before entering the analyzer. The gas analyzer sample cell is sensitive to water and could be damaged.

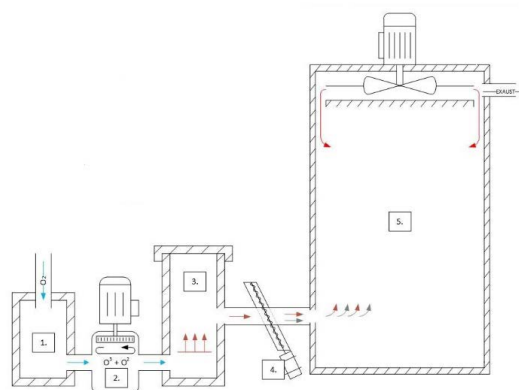


Figure 1. An experimental combustion chamber with a smokehouse. (1 ozone generator; 2 air flow fan; 3 burning camera; 4 additional smoke generator; 5 smokehouse chamber).

The analyzer pump is started in a continuous cycle to perform periodic measurements. The measurements were done every 120 seconds at least 30 minutes. The spectrum is recorded and interpreted in real time and checked in post processing.

Additional temperature measurements have been performed in the combustion chamber using temperature sensors of type pt100 in the smoking chamber. The estimated marginal means for the levels of significant effects were compared using the Tukey's HSD multiple comparison test. The data analysis was performed in R v. 4.1.2. (R Core Team, 2018).

### Results and Discussion

The burning temperature of the apple and grey alder wood pellets did not differ significantly if in burning chamber supplies atmospheric air or enriching it with ozone (Table 1). However, the range of combustion temperature with  $O_3$  enriched air is significantly shorter (697 to 817 and 611 to 817  $^\circ\text{C}$ , respectively) compared by using atmospheric air. On the other hand, the flue gases analyzed differed significantly. As a result of the introduction of ozonated air, significantly less  $\text{CO}_2$ ,  $\text{CH}_4$  and CO

were detected in the combustion gases, while N<sub>2</sub>O differ depends on the tree species used to make the wood pellets (Table 1). Ozone reactions with fuel release of atomic oxygen resulting in an increase in chain reaction response rates at relatively low temperatures (Wenting *et al.*, 2019), resulting in a rapid oxidation of released pyrolysis gases at higher initial temperatures, and consequently in differences in flue gas concentrations (Wenting *et al.*, 2019).

As predicted, the different pellets diameter of the tree species did not significantly affect the composition of the smoke gases analyzed, so they were analyzed together. However, we observed significant differences in the composition of flue gases, depending on the tree species wood used in the manufacture of granules, which are directly related to differences in the chemical composition of apple and grey alder wood (Anderson *et al.*, 2009; Cichy *et al.*, 2018). A different pyrolysis gas composition is released during the combustion process for each tree species, which also affects reactions with O<sub>3</sub> and further differences in flue gas composition (Zandersons *et al.*, 2009). By burning apple wood pellets, it releases significantly higher CO<sub>2</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O, as well as water vapors

are released by the use of ozonated air. On the other hand, when burning grey alder wood pellets with ozonated air, only CH<sub>4</sub> is released significantly more, but N<sub>2</sub>O is reduced, while other gases did not differ significantly (Table 1). Long-term storage of grey alder wood is characterized by natural processes that reduce the amount of organic acids, found in wood during studies of pyrolysis processes (Zandersons *et al.*, 2009). During the production of pellets, the most likely amount of organic acids in grey alder wood has decreased, which also explains only the increase in CH<sub>4</sub> in the flue gases.

On the other hand, when analyzing the differences in the composition of the flue gases between tree species, using shaded air during the combustion process, apple wood granules emit more N<sub>2</sub>O and H<sub>2</sub>O in the flue gases than grey alder pellets. Without the use of ozonated air in the combustion process, the opposite effect is that grey alder pellets emit more N<sub>2</sub>O and CO<sub>2</sub> than apple-wood pellets. Which, in general, refers to differences in the wood chemical properties which have a significant impact on the composition of flue gases.

Table 1

**Mean values of the analysed flue gases when burning apple and grey alder wood pellets, with and without enrichment of the air entering in the combustion chamber with ozone**

Species	Air supply	Burning temperature, °C	Water vapour H <sub>2</sub> O, ppm	Carbon dioxide CO <sub>2</sub> , ppm	Carbon monoxide CO, ppm	Nitrous oxide N <sub>2</sub> O, ppm	Methane CH <sub>4</sub> , ppm
Apple	O <sub>3</sub>	767±3.7	1.14±0.02 <sup>a</sup>	11962±525 <sup>a</sup>	1376±61 <sup>a</sup>	0.47±0.11 <sup>a</sup>	49.6±3.2 <sup>a</sup>
Grey alder	O <sub>3</sub>	768±2.8	1.09±0.04 <sup>b</sup>	11974±1351 <sup>ab</sup>	1396±256 <sup>ab</sup>	0.13±0.05 <sup>b</sup>	43.6±3.2 <sup>ab</sup>
Apple	Air	766±3.9	1.07±0.04 <sup>bc</sup>	6895±1325 <sup>b</sup>	1069±121 <sup>b</sup>	0.18±0.02 <sup>b</sup>	38.6±1.7 <sup>bc</sup>
Grey alder	Air	771±4.1	0.95±0.09 <sup>c</sup>	9891±1580 <sup>c</sup>	1121±219 <sup>bd</sup>	0.22±0.01 <sup>c</sup>	36.8±3.4 <sup>c</sup>

a,b – different letters in superscript represent significant differences (p<0.05) between light treatments for a concrete morphological variable of a concrete clone according to Tukey’s HSD test results. Numbers after plus-minus signs represent 95% confidence intervals.

**Conclusions**

1. Enrichment of the air supplied to the combustion chamber with O<sub>3</sub> did not affect the average burning temperature of apple and grey alder wood pellets. However, the use of O<sub>3</sub> ensures a smoother combustion process – smaller temperature fluctuations in the combustion chamber.
2. Using ozone-enriched air when burning apple wood pellets increases flue gases CO<sub>2</sub>, CO, CH<sub>4</sub>,

N<sub>2</sub>O, as well as water vapor than when burning them without ozone supply. On the other hand, when burning grey alder wood pellets with ozonated air, only CH<sub>4</sub> is emitted significantly more, while N<sub>2</sub>O decreases.

3. When comparing between tree species, burning grey alder wood pellets released more CO<sub>2</sub> and N<sub>2</sub>O in the flue gas than burning apple wood pellets, while the use of ozonated air in the

combustion process increased  $N_2O$  in the flue gas burning apple wood pellets.

4. By introducing ozonated air into the combustion chamber, it is possible to influence the combustion process, as a result of which the composition of flue gases changes significantly, but further practical research must be carried out in order to start the practical use of ozone.

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#### References

- Andersons, I., Andersons, B., Čirkova, J., Biziks, V., Irbe, I., Kurnosova, N., & Grīniņš, J. (2009). Alkšņu koksnes ilgizturības uzlabošanas iespējas hidrotermiskās modifikācijas ceļā. Valsts pētījumu programma 2005-2009. Rakstu krājums, Lapu koku audzēšanas un racionālās izmantošanas pamatojums, jauni produkti un tehnoloģijas. (Possibilities of improving the durability of alder wood through hydrothermal modification. State research program 2005–2009. Rationale cultivation and use of deciduous trees, new products and technologies). Latvijas Valsts koksnes ķīmijas institūts, Rīga, 2009, pp. 105–109. (in Latvian).
- Cichy, W., Witzak, M., & Walkowiak, M. (2017). Fuel Properties of Woody Biomass from Pruning Operations in Fruit Orchards. *Bioresources* 12(3): 6458–6470.
- Docquier, N., & Candel, S. (2002). Combustion control and sensors: a review. *Prog Energy combust Sci* 2002; 28: 107–50.
- ENplus Handbook, Part 3 – Pellet Quality Requirements. European Pellet Council (EPC) c/o AEBIOM – European Biomass Association Place du Champ de Mars 2 1050 Brussels, Belgium.
- Greene, C.R., & Atkinson, R. (1992). Rate constants for the gas-phase reactions of  $O_3$  with a series of alkenes at  $296 \pm 2$  K. *Int J Chem Kinet.* 24: 803–11. DOI: 10.1002/kin.550240905.
- Hitzel, A., Pöhlmann, M., Schwägele, F., Speerb, K., & Jira, W. (2013). Polycyclic aromatic hydrocarbons (PAH) and phenolic substances in meat products smoked with different types of wood and smoking spices. *Food Chemistry Vol. 139, Issues 1–4, 15*, pp. 955–962. DOI: 10.1016/j.foodchem.2013.02.011.
- Nussbaumer, T. (2003). Combustion and Co-combustion of Biomass: Fundamentals, Technologies, and Primary Measures for Emission Reduction // *Energy&Fuels*. Vol. 17. pp. 1510–1521.
- Obernberger, I., Brunner, T., & Barnthaler, G. (2006). Chemical properties of solid biofuels – significance and impact // *Biomass and Bioenergy*. No. 30. pp. 973–982. DOI: 10.1016/j.biombioe.2006.06.011.
- R Core Team. (2018). R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. Retrieved March 5, 2023, from <https://www.R-project.org/>.
- Skaljac, S., Petrovic, L., Tasic, T., Ikonc, P., Jakonovic, M., Tomovic, V., Džinic, N., Šojic, B., Tjapkin, A., & Škrbic, B. (2014). Influence of smoking in traditional and industrial conditions on polycyclic aromatic hydrocarbons content in dry fermented sausages (Petrovska klobasa) from Serbia. *Food Control Vol. 40*. 12–18. DOI: 10.1016/j.lwt.2017.08.038.
- Stumpe-Viksna, I., Bartkevičs, V., Kukāre, A., & Morozovs, A. (2008). Polycyclic aromatic hydrocarbons in meat smoked with different types of wood. *Food Chemistry*, 110 (3), pp. 794–797.
- Van Loo, S., & Koppejan, J. (2008). *The Handbook of Biomass Combustion & Co-firing*. – UK: CPI Antony Rowe. 442 p.
- Wenting, S., Xiang, G., Bin, W., & Ombrello, T. (2019). The effect of ozone addition on combustion: Kinetics and dynamics. *Progress in Energy and Combustion Science* 73 (2019) 1–25. DOI: 10.1016/j.pecs.2019.02.002.
- Williams, A., Jones, J.M., Ma, L., & Pourkashanian, M. (2012). Pollutants from the combustion of solid biomass fuels, *Progress in Energy and Combustion Science // Progress in Energy and Combustion Science*. Vol. 38. pp. 113–137.
- Zandersons, J., Dobeles, G., Jurkjane, V., Tardenaka, A., Spince, B., Rizhikovs, J., & Zhurinsh, A. (2009). Pyrolysis and smoke formation of grey alder wood depending on the storage time and the content of extractives. *J. Anal. Appl. Pyrol.* Vol. 85, pp. 163–170. DOI: 10.1016/j.jaap.2008.11.036.