

Characterizing Particulate and Condensable Emissions from a Wood-Burning Insert

Clara Akl¹, Julie Schobing², Cornelius Schönnenbeck², Gwenaëlle Trouvé²

^{1,2}Laboratoire Gestion des Risques et Environnement, Université de Haute-Alsace

3b rue A. Werner, 68093 Mulhouse Cedex, France

clara.akl@uha.fr ; julie.schobing@uha.fr

cornelius.schonnenbeck@uha.fr ; gwenaelle.trouve@uha.fr

Abstract - Wood combustion in domestic appliances is a key contributor to renewable energy in France, accounting for 31% of primary renewable energy production in 2023. However, it remains a significant source of particulate and gaseous emissions, including total suspended particles (TSP), volatile organic compounds (VOCs), and polycyclic aromatic hydrocarbons (PAHs). This study evaluates particulate and condensable organic emissions from a modern wood-burning insert under nominal and degraded combustion conditions. The experimental setup accords to NF EN 16510-1 standards, using gravimetric methods for TSP quantification and gas chromatography coupled to mass spectrometry (GCMS) for chemical characterization of organic compounds. TSP concentrations mainly remained low under both combustion conditions. Higher filter temperatures (180 °C) lead to lower TSP collected mass by inhibiting the condensation of volatile species. Condensable compounds were captured using a series of impingers. Chemical analysis revealed the presence of heavy alkanes, PAHs, and oxygenated PAHs (O-PAHs), especially during degraded combustion. Phenolic compounds, indicative of lignin degradation, were also identified. Temperature and combustion phase significantly influence the partitioning of organic molecules between particle and gas phases. This study highlights the importance of characterizing condensable organic compounds to better understand their role in air quality and health impacts. The findings emphasize the need for further studies on the chemical composition of emissions from wood-burning appliances, especially under real-world conditions, to optimize their environmental performance and compliance with evolving regulations.

Keywords: Wood combustion, TSP, condensable organic compounds.

1. Introduction

The law of 17th August 2015 on energy transition for green growth committed France to reduce its final energy consumption by 50% by 2050 compared with 2012, and to increase the share of renewable energies to 32% of gross final energy consumption by 2030 [1]. In this context, solid biomass has emerged as the fuel of choice for heat production, driven by cultural preferences and its reputation as carbon neutral [2].

In 2023 in France, primary production of renewable energies was still dominated by wood energy, which accounted for 31%. Wood energy also made up 58% of the primary consumption of renewable energy for domestic heating purposes [1]. During the 2022/2023 heating season, 7.5 million of primary residences in France heated with wood [3].

Although wood energy was considered renewable, its combustion at domestic scale emits gaseous and particulate pollutants such as carbon monoxide (CO), total suspended particles (TSP), PM_{2.5} (particle with aerodynamic diameter below 2.5 µm), black carbon (BC), volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAH) [4-8]. In France 2022, CO emissions were mainly from wood combustion (43%). Around 20% of total non-methane volatile organic compound (NMVOC) emissions were emitted from fuels, 68% of which from the use of wood combustion, which represents 14% of total national emissions. 80% of PAH emissions were attributed to wood combustion mainly (benzo(a)pyrene, benzo(k)fluoranthene, benzo(a)anthracene, benzo(g,h,i)perylene, benzo(b)fluoranthene, indeno(1,2,3-cd)pyrene, dibenzo(a,h)anthracene and fluoranthene). In France, wood combustion was the main emitter of TSP and PM_{2.5}, representing 86% and 87% respectively of fuel combustion emissions in 2022 and domestic wood stove appliances were the main contributors, 46% of TSP emissions and 43% for PM_{2.5} emissions [9]. Studies on particle emission factors have identified residential wood combustion (RWC) as a significant contributor to atmospheric fine particulate matter (PM_{2.5}) worldwide and especially in Europe [5].

The marketing of domestic wood heating appliances is controlled by different regulations to limit these emissions. In France, the 'Label Flamme Verte' was in place since 2000. The updated version in 2022 requires an emission limit of CO of 1500 mg/Nm³ and the sum of both emissions of TSP and VOC of 150 mg/Nm³, these concentrations referred at 13% O₂ in fumes [10]. Currently, while identifying organic molecules is not a regulatory standard, their identification is important to understand the relationship between their chemical composition and toxicity.

Since the 2010s, significant studies have focused on the chemical composition of carbonaceous particles. Particular attention has been given to elemental carbon (EC) and organic carbon (OC), which together can account for up to 90% of aerosol mass, depending on the region and sources. These components are commonly quantified using thermo-optical transmission techniques, both in ambient air and directly at combustion sources, such as domestic fireplaces. EC is a product of incomplete combustion, and OC, a mix of primary and secondary components. Their ratio (EC/OC) has proven critical for distinguishing primary emissions from secondary organic aerosol formation [11-14]. However, the chemical characterization of the organic fraction in TSP remains limited. While some studies have identified PAHs and oxygenated organics as contributors to the toxic and radiative properties of OC in biomass combustion, studies and comprehensive analyses of this fraction are lacking. Addressing this gap could clarify the role of condensable organic compounds in air quality and climate modeling, where significant uncertainty persists. Condensable particulate matter (CPM) lacks a universal definition. The U.S. Environmental Protection Agency (EPA) first identified CPM emissions in 1983 [15]. CPM are particles that exist in a gaseous state within chimney flues but rapidly condense or react to form liquid or solid particles when released into the atmosphere, driven by cooling and dilution processes [16].

Therefore, this paper evaluates the particulate and condensable emissions from a modern wood-burning insert under different combustion conditions. By combining advanced gravimetric methods and gas chromatography coupled to mass spectrometry (GCMS), the study identifies TSP emission trends and analyses condensable organic compounds in both the gas and particle phases, emphasising the impact of temperature and combustion phases.

2. Experimental Section

2.1. Combustion Platform

Combustion tests were conducted using a domestic wood-burning insert (model XP-68 IN), commercialized by LORFLAM with a nominal output of 11 kW. It is labelled Flamme Verte 5 étoiles (2012) but meets the requirements of the latest updated version Label Flamme Verte 2022. The insert uses hornbeam wood of 33 cm length at 12% moisture content. This insert features three air inlets, one under the grate where the logs are placed for primary combustion, the other inlet is behind the combustion chamber for post-combustion and another at the glass level to reduce soot deposits and maintain glass cleanliness. The air inlet is adjustable via a front-panel knob and residual ashes are collected in a compartment below the combustion chamber. Combustion tests were divided into different phases: ignition, flaming phase (nominal and degraded) and char combustion. During degraded combustion regime, the air flow is reduced into the stove.

2.2. Measurement of Particulate Mass Emissions

TSP were collected by gravimetric method based on NF EN 16 510-1 (2018) [16] and NF EN 16510-1 (2022) [17] standards. Filter used were Whatman EPM 2000 borosilicate glass fiber filters with a porosity of less than 2 µm at a flow rate of 0.5 Nm³/h. These filters were prepared by drying them in an oven at 105°C and cooling them in a desiccator prior to use. During the tests, the filter was placed in a support heated at 110°C based on NF EN 16 510-1 (2018) or at 180°C and placed at the end of a 2 m long heated line (also at 180°C) connecting the filter holder to the conduit. According to the 2018 standard, TSP were collected for 30 minutes starting from 3 minutes after the load began and for the full combustion duration (around 45 min) according to the 2022 standard. At the end of each combustion test, the filters were placed in a desiccator for 24 h and then weighted. These filters were extracted either by Soxhlet (4 cycles) or ultrasound (at room temperature) using 50/50 dichloromethane/acetone solvent mixture.

2.3. Measurement of Condensable Compounds

The method for measuring condensable compounds was inspired from the protocol developed by Ineris (Institut National de l'Environnement Industriel et des Risques, France). The method involves capturing condensable compounds in two impingers containing 150 mL isopropanol (IPA) (VWR isopropanol with a purity of 99.7%) and placed in a cold-water bath maintained at 5°C. In this study, additional empty impingers were placed before and after the IPA impingers. The first empty impinger served to condense water vapor from the combustion gases, while the final one prevented the IPA from escaping into the pump. After some experiences, a second filter maintained at ambient temperature was placed after the fourth impinger to capture any remaining molecule that was not condensed in the impingers. After testing, the water condensed in the first impinger was extracted using liquid-liquid extraction with dichloromethane at room temperature, the solution was filtered and then evaporated at ambient temperature under the hood. The IPA in the impinger was also evaporated and the filter after the impingers was extracted by Soxhlet procedure and then the solution was evaporated. Figure 1 below shows the whole experimental sampling systems

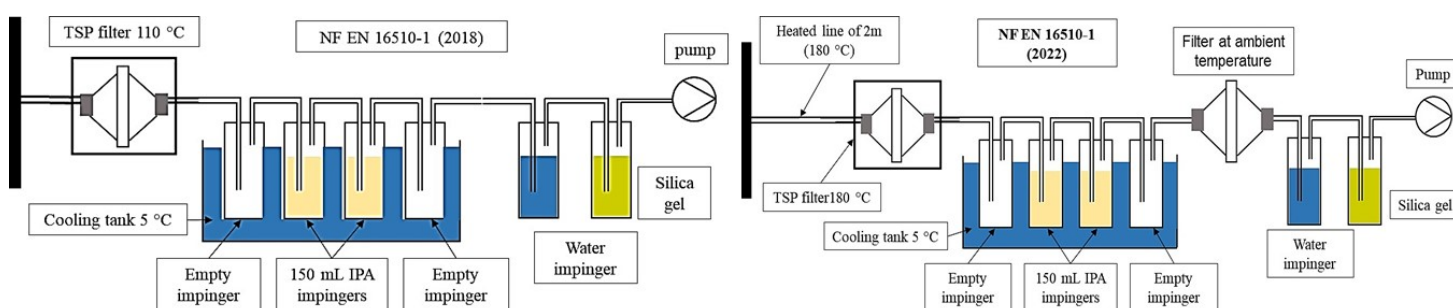


Fig. 1: Schematic diagram of the sampling train of TSP and condensable compounds

2.4. Analysis by Chromatography Coupled to Mass Spectrometry (GCMS)

In addition to the Ineris protocol, all extracts from TSP, water, IPA and second filter after evaporation were dissolved in 1 mL dichloromethane for identification of the organic molecules present using GCMS coupling. Chromatographic conditions used were as follows: the column was a apolar phase TR 5MS column from THERMOSCIENTIFIC, with dimensions of 60 m * 0.25 mm * 0.25 µm; several chromatographic separation conditions were tested, using methods available in the laboratory and temperature-programmed according to the nature of the sample (bubblers, filters, etc.); ion detection by mass spectrometry was carried out in SCAN mode: detection of ions by scanning in mass ranges between 10 and 300 amu or 10-650 amu (range used to detect heavy alkanes); identification of PAHs and Oxygenated Polycyclic Aromatic Hydrocarbons (O-PAHs) molecules was carried out in SIM mode by exclusive molecular ion search.

3. Results and Discussion

3.1 Particle Mass Emissions

Experiments were conducted with a wood insert under both nominal and degraded load conditions. Average masses and concentrations of TSP filters were calculated.

The average masses of the filters heated at 110°C for 30 minutes, according to NF EN 16510-1 (2018) standard, were 3.3 mg and 3.7 mg for nominal and degraded load tests, respectively. These low filter masses correspond to low TSP concentrations. This is because the NF EN 16510-1 (2018) standard requires gravimetric TSP measurement after the ignition phase (t = 3 min) and over a 30-min period, during which CO and total hydrocarbons compounds (THC) concentrations are particularly low as shown in Figure 2.

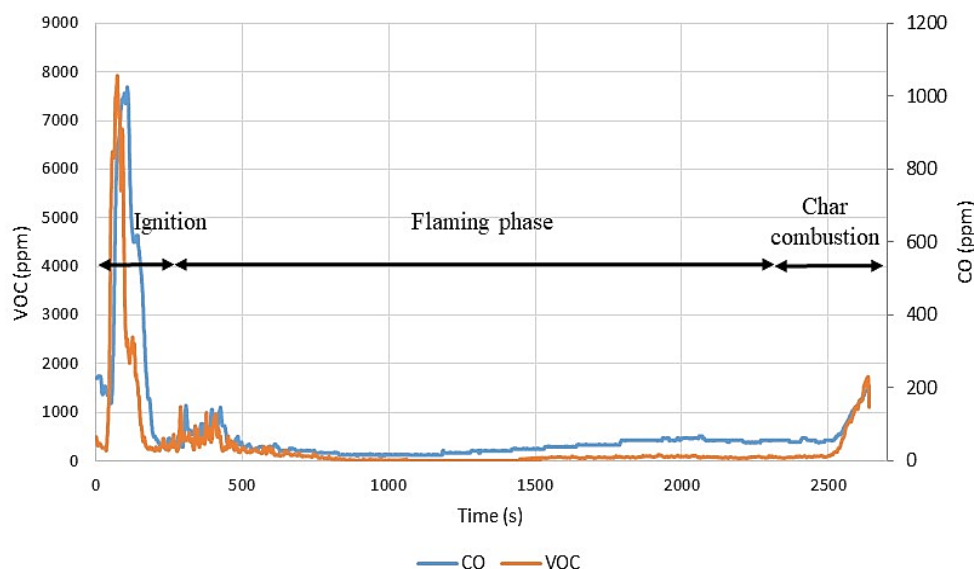


Fig. 2 : Evolution of CO and THC emissions during a nominal load phase

A comparison of TSP emission factors (in mg/Nm^3) during nominal conditions with available literature data on modern wood-burning devices in the European market shows that our results ($10.6 \text{ mg}/\text{Nm}^3$) are significantly lower—at least by a factor of 10. For example, Ozgen et al. [18] reported values of $150 \text{ mg}/\text{Nm}^3$ for both advanced and traditional wood stoves burning beech logs. These lower values may reflect the efficiency of the high performance insert we have. Tschamber et al. found TSP ranged between 5 to $20 \text{ mg}/\text{Nm}^3$ with the same device for nominal loads [11].

Lower TSP masses and concentrations were observed when filters were heated at 180°C for 30 minutes compared to those heated at 110°C . The recorded average masses were 3 mg for nominal load tests and 3.4 mg for degraded load tests. But regards to deviation standards, values remain in the same order of magnitude. This slight reduction is attributed to higher filter temperatures, which prevent certain species from condensing and allow them to remain in gaseous form, thereby bypassing the filter [6].

According to the updated NF EN 16510-1 (2022) standard, TSP measurements were captured on filters heated at 180°C for the entire duration of the test. In this case, TSP masses and concentrations were a bit higher than those obtained during the 30-min measurements. The average filter masses were 6.5 mg during nominal load tests and 5.2 mg during degraded load tests—approximately double the mass values measured over the 30 min' measurement duration. As expected, the measurement over the entire combustion process includes the ignition and heterogeneous combustion phases that emit the highest CO and THC emissions as shown in Fig. 2.

Furthermore, no significant difference in TSP mass and concentration was observed between nominal and degraded load conditions, as both test conditions yielded similar results. This lack of difference indicates that the combustion process was not significantly degraded during the tests with the close of air intakes.

3.2 Condensable mass measurements

3.2.1 Water and IPA impingers

The water and IPA in the impingers after the tests were extracted and evaporated, and the difference in mass before and after evaporation was used to determine the mass of condensable residues trapped in the impingers. Table 1 summarizes the residue masses obtained after evaporation.

Table 1: Mass of the residue after evaporation of the water extract and IPA in the impinger

mass in mg	Water impinger		IPA impinger		TSP filter	
	Nominal	Degraded	Nominal	Degraded	Nominal	Degraded
30 min without heated line (TSP at 110°C)	1.0 ± 0.1	2.0 ± 0.1	6.0 ± 0.1	5.0 ± 0.1	3.3 ± 0.4	3.7 ± 1.6
30 min with heated line and TSP at 180°C	3.5 ± 0.1	4.0 ± 0.1	8.0 ± 0.1	8.0 ± 0.1	3.0 ± 0.9	3.4 ± 1.1
Full test duration with heated line and TSP at 180°C	4.5 ± 0.1	3.5 ± 0.1	8.0 ± 0.1	9.0 ± 0.1	6.5 ± 2.3	5.2 ± 1.4

Whatever the experimental samplings, condensation of compounds mainly occurs in IPA than water impingers. As for TSP, no significant difference was observed for both nominal and degraded loads. However, when the impingers were placed after the heated line at 180°C, masses of residue increased for both the 30-min and full test measurements. This indicates that when the impingers were positioned directly after the TSP filter heated at 110°C, fewer condensable compounds were trapped and more TSP mass collected on the filter. In contrast, when the heated line was used and the filter temperature was raised to 180°C, the TSP masses on the filter decreased, while the condensable mass collected in the impingers increased. The higher temperature avoids the condensation of some of the hydrocarbons prior to the filter, preventing them from being trapped and allowing them to condense later in the impingers. Additionally, when comparing the masses collected during tests with the heated line over different measurement durations, no significant differences in mass were observed, despite the full test measurement encompassing phases with higher emissions. This is because during the ignition phase, high concentrations of low molecular weight VOCs as BTEX (Benzene, Toluene, Ethylbenzenes) are emitted having at sampling temperatures high saturated vapor pressures that maintains them in their gaseous state [19]

3.2.2 Condensable filters

A filter was placed at ambient temperature after the impingers to capture any molecules that were not condensed within the impingers. After calculating the mass difference of this filter, a very small residue was observed: 0.2 mg for the 30-min tests and 0.4 mg for the full-duration tests for both nominal and degraded load conditions. However, when comparing the two measurement durations, a higher mass was found on the filters collected during the longer tests. These results suggest that only a minimal number of molecules were not condensed in the impingers and were subsequently captured by the filter at the outlet.

In the literature, many studies measured condensable from residential combustion without using impingers. In those studies, the flue gas was diluted and cooled via a dilution, and the condensable were subsequently trapped on a filter. Claudia Schön (2023) conducted combustion tests on log stoves to measure condensable particles. In her study, condensable residues were trapped on a filter maintained at 40 °C, placed after a porous tube used to dilute and cool the flue gas [20]. To compare literature data, the condensable concentrations collected during our tests in the water and IPA impingers, along with the residue on the filter, were summed to obtain total condensable concentrations equivalent to that was reported. Schön found a condensable concentration on the filter at 35 mg/Nm³. For the tests conducted in our study more condensable compounds were trapped in both the water and IPA impingers, as well as on the afterward filter, resulting in a total condensable concentration of 65 mg/Nm³. The mass difference could be attributed to the sampling temperatures. Because we have sampled at lower temperatures than Schön, condensation processes were favoured in both impingers and ambient filter.

3.3 Condensable organic compounds analysis

To identify the presence of organic molecules in the particles, GCMS analyses were performed. Whatever the sampling conditions, only few traces of molecules were detected in all chromatograms. Because traces were detected, the reproducibility of results from the analysis of a large number of filters and impingers has not been achieved with a simple quadrupole mass spectrometer and sometimes, the identification is random depending on runs. No molecules were detected

4. Conclusion

This study provides valuable insights into the particulate and condensable organic emissions from a modern wood-burning insert under various combustion conditions. The gravimetric and GCMS methods have enabled the quantification of TSP and the identification of condensable organic compounds in both gas and particle phases, respectively. The results revealed that TSP emissions measured were significantly influenced by temperature, with higher filter temperatures (180°C) reducing particle collected mass by preventing the condensation of volatile compounds and favouring transfer of semi-volatile molecules in the gas phase.

Notably, a distinct profile of PAHs and O-PAHs which are indicators of incomplete combustion was observed, particularly under degraded combustion conditions. The main findings indicated that, water vapor trapped the majority of the organic compounds during full duration tests measurement in the selected chromatographic conditions and only few molecules were identified in IPA. The chemical characterization of condensable organic compounds, especially the presence of phenolic compounds from lignin degradation, provides crucial information for understanding their potential environmental and health impacts. They also promote important insights into the mechanisms governing particulate and condensable organic emissions at the duct at low temperature from wood-burning appliances. By highlighting the influence of temperature and combustion conditions on emissions, this study underscores the importance of adapting measurement protocols to account for real-world scenarios. Future measurements planned at the chimney outlet, where lower temperatures prevail, will provide a more comprehensive understanding of condensation phenomena and emissions transfer to ambient air. These results will enhance the relevance of emission data for regulatory frameworks and environmental modelling.

Acknowledgements

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