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Research paper

Investigation of the organic carbon ratio analysis on particles from biomass combustion and its evolution in three generations of firewood stoves

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ABSTRACT

In this study, the emissions of three different generations of firewood stoves were characterized to test their sustainability with this new problematic. Organic Carbon (OC) and Elemental Carbon (EC) emissions were particularly targeted. After an investigation of the OC/EC measurement method, a specific protocol was developed, validated and optimized in order to measure OC/EC from direct samplings in the flue gas, without diluting or cooling systems. Carbon monoxide (CO), Total Suspended Particles (TSP) and Total Hydrocarbon (THC) emissions were followed too. To go further in the characterization and in the differentiation of the particles emitted from each kind of stoves, a Field Emission Gun Scanning Electron Microscope (FEG SEM) was used. The medium generation emitted 40% less mass of OC than the previous one. Concerning the new generation, emissions were reduced in mass about 98% for the EC and 40% for the OC compared to the old generation.

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1. Introduction

Limiting and regulating the atmospheric pollution is one of the biggest challenges of the 21st century. Particulate matter is one of the most problematic pollutants. On the other hand, develop and improve renewable energy, like domestic wood stoves, can be one response among others to avoid a global warming higher than 2 °C. Biomass burning was described as a significant source of particulate matter emissions in many studies [1–4]. Herich et al. [5] demonstrated that during winter, the contribution of wood-burning appliances to ambient levels of black carbon (BC) was significant (about 30%). Dejmek et al. [6], Penttinen et al. [7], Moshammer et al. [8] and Ghio et al. [9] demonstrated that the mass, the number of particles, as well as surface and elemental compositions were related to respiratory diseases. Indeed, Stieb et al. [10] demonstrated that the atmospheric pollution was associated with all-cause mortality. In 2012, the World Health Organization [11] showed that the organic carbon and elemental carbon (OC and EC), contained in particulate matter, had different health effects. Lim et al. [12] and Bond et al. [13] showed that OC and EC played different role in global climate change by modifying the chemistry of clouds and the radiative balance of the earth. Both types of carbon were usually measured in ambient air with a thermal-optical device [12,14–16]. Previous studies sampled particles on quartz filters after diluting and cooling the flue gas to avoid a black surface. Indeed, McDonald et al. [4] and Rau et al. [17] explained that a black surface limited the optical measurement. However, Lipsky et al. [18] established that this type of protocol (dilute or cool down the flue gas) did not allow accurate and realistic characterizations due to physicochemical modifications of the particles by diluting and cooling. In order to evaluate the emissions of each generation of firewood stoves, the OC/EC ratio had to be measured in the flue gas chimney. Moreover, measuring OC ratio without any dilution and cooling system was the only way to know the real composition of the particles emitted by firewood stoves and how they were impacted by the improvement of the stove. The evolution of the standards and labels led to the improvement of the stoves, in terms of yield, carbon monoxide, particulate matter and volatile organic compounds emissions. The aim of this study was to assess if the emissions of OC and EC followed the same trend and then to validate the sustainability of the new generation stoves.

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2. Materials and methods

2.1. Nomenclature

OC Organic Carbon
EC Elemental Carbon
PC Pyrolytic Carbon
CC Carbon Carbonates
OC mass ratio: $(OC.TC^{-1})*100$ *With:OC and TC in mg m*$^{-3}$
TC mass ratio: $(TC.TSP^{-1})*100$ *With:TC and TSP in mg m*$^{-3}$
OC total mass ratio: $(OC.TSP^{-1})*100$ *With:OC and TSP in mg m*$^{-3}$
FOG Firewood stove Old Generation
FMG Firewood stove Medium Generation
FNG Firewood stove New Generation
CO Carbon Monoxide
TSP Total Suspended Particles
THC Total Hydrocarbon
FEG SEM Field Emission Gun Scanning Electron Microscope
NCV_{wb} Net Calorific Value wet based

2.2. Wood stoves and experimental procedure

2.2.1. Description of the stoves and wood

For this study, three stoves were studied:

(i) Firewood stove Old Generation (FOG) of 12 kW, without any insulation nor secondary air (2000, SUNFLAM, Seguin Duteriez);
(ii) Firewood stove Medium Generation (FMG) of 12 kW, with firebrick and secondary air (2006, AX F 700, AXIS);
(iii) Firewood stove New Generation (FNG) of 6 kW, with vermiculite and secondary air (2012, WABI, D2I INVICTA).

For each firewood stove, five tests in steady state at the nominal thermal heat output according to the manufacturer settings were achieved.

The applied fuel was split beech (*Fagus sylvatica*) logs of 12 cm of diameter from a 40-year-old beech. The logs were selected without nodes and bark to get a higher repeatability. Their water mass fraction was stabilized in a regulated enclosed chamber and was measured following the standard EN 14774. The average result is 0.12. The wet base Net Calorific Value (NCV_{wb}) was 16.7 MJ kg^{-1} (following the standard EN 14918). The ash mass fraction of the dried material was 2.7 g kg^{-1} at 550 °C and 1.4 g kg-1 at 815 °C (following the standard EN 14775). In order to obtain a representative result of the elemental composition, a complete log was crushed. Three samples of 1 g arbitrary sampled in the mixed mash were analysed. The results are in average: 0.495 kg kg^{-1} of C; 0.059 kg kg^{-1} of H; 0.438 kg kg^{-1} of O; less than 0.003 kg kg^{-1} of N; 108 mg kg^{-1} of S; 64 mg kg^{-1} of Cl.

2.2.2. Experimental platform and sampling procedure

The experimental platform is described in Fig. 1. The stove was on a balance, and the duct was separated from the balance with a water seal. The molecular oxygen and carbon monoxide in the flue gas were measured in continuous with photo-chemical cells. The temperatures of the flue gas and the ambient were also measured. For the TSP, an isokinetic system was used. The sampling was performed at 160 °C on a quartz filter with a diameter of 90 mm with a flue gas flow of 17 dm^3 min^{-1}. The THC were sampled in a Flame Ionization Detector (FID) at 160 °C. The Organic Carbon and Elemental Carbon analyser (OC, EC) were sampled at 10 dm^3 min^{-1} at 160 °C on a quartz filter of 90 mm of diameter. This sampling was held 10min for FNG and only 5min for FOG and FMG during the homogeneous combustion in any case (15 min after the loading). This short sampling allowed avoiding an overload of particles on the filter. The analysis protocol is further described in the next part.

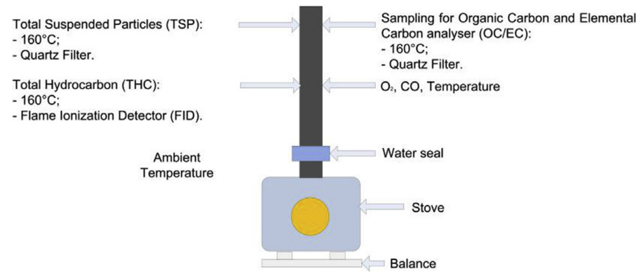

Fig. 1. Description of the experimental platform.

2.3. OC/EC analyse

Thermo-optical analysis can be performed based on the fact that OC is volatile and non-black and EC is non-volatile and black. Moreover, OC can be transformed in Pyrolytic Carbon (PC) which presents the same characteristics as native EC if it receives enough energy. Fig. 2 shows the resulting curve obtained with this type of analyser. The first analysis step consists in a laser measurement in order to determine the initial transmittance or absorbance of the filter. Then the filter is heated with a helium flow at a precise temperature, depending on the chosen standard. Chow [19] proved that the protocol used for the analysis had a significant effect on the results. The first peak corresponds to the OC volatilization. Since the filter became blacker, transmittance decreases due to the production of PC. The measurement of the PC formation is the main point to determine precisely the discrimination between OC and EC [20]. At 870 °C (for NIOSH 870 standards) all the OC is transformed in PC. During this isothermal step Carbon Carbonates (CC, second peak) are emitted. The temperature drops to 500 °C, and when the transmittance is stable, an oxidizer flow goes through the sample. The OC transformed in PC is then oxidized (third peak), leading to a brighter filter and thus to an increase in transmittance. Once the transmittance comes back to its initial level, all the carbon still present on the filter corresponds to the initial quantity of EC. This point is called the split point. After, EC is also burnt (fourth peak). The last step is a calibration of the FID with a known volume of CH_4 (fifth peak). This kind of analyser is usually used for ambient sampling. For analysing the filters directly sampled in the flue gas, a specific protocol has to be developed as the filter receives too many particles.

Four different analysis standards exist: NIOSH 870, NIOSH 930, EUSAAR 2, IMPROVE. Cavalli et al. [21] showed that EUSAAR 2 improves the pyrolysis, but NIOSH 870 and NIOSH 930 heat the filter to higher temperatures. Each standard measures the same Total Carbon (TC, TC = OC + CC + EC) mass but not the same OC mass fraction [22–24]. Table A1 shows the results of each standard achieved for three different kinds of filters. The IMPROVE protocol was not tested in this study. In our case, NIOSH 870 always presents a higher accuracy in measurements of OC and EC. For these reasons, this analysis standard was used for this study.

2.3.1. Accuracy of OC and EC measurements

To analyse the accuracy of the thermo-optical analyser (Sunset Laboratory Inc.) with black filters, the measurement accuracy of the OC alone, as well as that of a black powder (to simulate a real filter) containing OC and EC, were studied. Both types of filters were analysed on the same device. A sucrose ($C_{12}H_{22}O_{11}$) solution with an elemental carbon concentration of 3.504 kg m^{-3} was used to study the measurement accuracy of the OC alone and a charcoal

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