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Characterization of aerosols emissions from the combustion of dead shrub twigs and leaves using a cone calorimeter

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ABSTRACT

This work is a contribution to the understanding of wildfire smoke emissions. It focuses on the characterization of aerosols emitted by the combustion of dead shrub leaves and twigs with different thickness (range of 0.75–20 mm). The experiments were carried out at bench scale with a cone calorimeter for the burning of *Cistus monspeliensis* leaves and twigs. Auto-ignition of the samples was obtained by heating their surface with a radiant heat flux of 50 kW/m^2. The smoke and aerosols emitted before ignition during pre-heating were analysed separately from the smoke and aerosols emitted during the flaming phase. Heat release rate (HRR) was also measured and we observed two different behaviours depending on the diameter of the twigs. Fuel samples with diameter smaller than 4 mm exhibit a single peak HRR whereas two peaks were observed for the twigs with larger diameters. The smoke production rate (SPR) was also measured and it showed that smoke was mainly emitted during the pre-heating phase. We also obtained a strong correlation between HRR and SPR during the flaming phase but no smoke was emitted during the glowing phase. Emission factors of aerosols were calculated depending on these combustion phases (pre-ignition and flaming) and for the range of thickness of the samples. The observations of the aerosols were performed by scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The chemical composition of organic carbon (OC) aerosols, emitted during the pre-ignition phase, were analysed using gas chromatography (GC) coupled with mass spectrometry (MS). Some carcinogenic compounds were identified. The sizes of OC and black carbon (BC) aerosols emitted were measured with an optical device. Most of the BC were $PM_{0.3}$, which corresponds to the alveolar fraction of particles.

1. Introduction

Wildfires produce significant levels of smoke particles. Among them, some present particular risks. Indeed, aerosols, also named particulate matter (PM) are an important ambient air pollutant and present serious health hazards. PM can also be emitted from other different sources like wind-blown dust, sea salt, or road transport (fossil fuel combustion). They are now classified as carcinogenic to humans (Group 1) by the International Agency for Research on Cancer (IARC) and the World Health Organization (WHO) considers that at least 1.4% of deaths worldwide are caused by ambient air pollution in particulate matter, including all sources [1]. Wildfires contributes to global PM emissions up to 35% [2], and smoke from biomass combustion are dominated by submicron particles (< 1 μm) [3]. Among those carbonaceous aerosols, black carbon (BC) and organic carbon (OC) are distinguished. The BC or elemental carbon is formed by a mass of carbon atoms with a graphitic-like structure that is black in colour and is usually named as soot. The OC is formed by a complex mixture of organic compounds [4]. Organic matter is an important fraction of the atmospheric aerosols, contributing with 10–70% of their mass [5]. Fine particles and the associated organic compounds present health hazard because of their size for the BC and because of their chemical composition for the OC [6,7].

The behaviour of those particles within the human respiratory system depends on their size, varying from a few nanometres to tens of micrometres. The respirable particles have aerodynamic diameter of 10 μm or less (PM_{10}), and they are not retained by the upper airways (nose, mouth). Fine particles, with diameter of 2.5 μm or less ($PM_{2.5}$) represent the thoracic fraction of particles. The very fine particles, with diameter of 1 μm or less (PM_1) penetrate into the alveoli. And the ultrafine particles, with diameter of 0.1 μm or less ($PM_{0.1}$) are the greatest number of particles. This alveolar fraction (PM_1 including $PM_{0.1}$) arise largely from primary combustion emissions, as wildfires. They present a particular health threat because their small size allows

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Nomenclature

d	Diameter of the twigs
E	Heat release per unit mass of O_2 consumed (MJ/kg)
EF	Emission factor (g/kg)
k	Extinction coefficient of smoke (m^{-1})
HRR	Heat release rate per unit area (kW/m^2)
m	Mass (g)
$\dot{m}$	Mass loss rate (g/s)
n	Number of moles (mol)
$\dot{q}$	Heat release rate (kW)
SEA	Smoke extinction area (m^2/kg)
SPR	Smoke production rate (m^2/s)
$\dot{V}$	Volume flow rate (m^3/s)
W	Molecular weight (g/mol)

Greek symbols

$\Delta H_{c,net}$	Net heat of combustion (kJ/kg)
σ	Specific extinction coefficient (m^2/kg)
φ	Combustion phase

Subscripts

O_2	Oxygen
$^\circ$	Before combustion

deep penetration into the lungs and onward passage across the air-blood barrier [8]. However, few studies on biomass burning targeting this ultrafine fraction has been identified in the literature [9]. And more generally, current knowledge does not yet provide precise quantification or definitive ranking of the effects of particulate matter emissions on human health.

A fairly substantial literature provides values of emission factors (or concentrations) from wildfire or prescribed burns for PM_{10} and $PM_{2.5}$, by distinguishing between OC and BC quantities among those particles [10–12]. Some of these previous studies have provided chemical analyses of the particles like TEM analyses of the particle chemistry [10] and identification of the inorganic (water-soluble ions and trace elements) and organic (chemical families) composition of the PM [11]. Studies on aerosols emissions from the combustion of biomass in laboratory have also been conducted. Some were carried out in a large-scale combustion chamber (≈3000 m^3) [13,14] and other in a wood-stove (1.75×10^{-2} m^3) [4,15]. They investigated the PM_{10} and $PM_{2.5}$, by distinguishing between OC, BC and ion species quantities. A recent study [16] conducted at bench scale with a combustion pan, gave a more detailed differentiation of the OC composition, by distinguishing the water-insoluble OC, the water-soluble hydrophilic OC and the water-soluble hydrophobic OC. Few studies have investigated the individual speciation of OC [15]. This is probably due to analytical difficulties, complexity of phenomena and huge number of compounds that are present [17]. It should be mentioned that for all the studies cited here above, the sampling of the aerosols was performed without differentiating pre-heating stage on one side and flaming and glowing stages on the other side.

The final goal of our study is to determine the toxicity level and the degree of risks for health related to smoke exposure, according to the composition and the size of the aerosols emitted from wildfires. As a first step, this paper details the results of an experimental investigation conducted to characterize smoke aerosols for size and composition, according to the combustion phases (pre-ignition, flaming and glowing) and for different thickness of vegetation fuel samples. In order to better understand the process of emission the HRR and the SPR were also measured to investigate the relation between SPR and HRR. Indeed, the HRR is a fundamental variable of fire with which almost all other emission properties are highly correlated. Although there is an abundant literature in the context of fire safety in buildings concerning the measurement of HRR [18], SPR [19], and smoke extinction area (SEA) [20], these quantities have been poorly related to emissions in the wildfire research community. Some works can be mentioned that have investigating the burning of vegetative fuels by using the oxygen consumption calorimetry principle under laboratory conditions [21,22]. However, these last works mainly investigated the ignition time and the combustion dynamics (HRR and mass loss). For instance the relationship between HRR and SPR was never investigated for vegetative fuels before a recent study on pine needles [23].

The experiments presented in the present paper were carried out with a cone calorimeter (in auto-ignition mode). A radiant heat flux of 50 kW/m^2 was imposed at the top of the fuel samples composed of dead shrub leaves and twigs with varying diameter to study their burning and emissions. The aim was to examine whether the emission factors vary with the thickness of the wood samples. Indeed, the size of the twigs burnt in forest fires depend on the intensity of the fire front and different emission factors could be expected as function of the size of the fuels involved. Aerosols emitted from these combustion tests were quantified from their extinction properties, using a He-Ne laser with a wavelength of 633 nm. Their size was provided by electronic microscopy (SEM and TEM) and their distribution was measured with an optical device. The chemical composition of OC aerosols was determined by gas chromatography coupled with mass spectrometry. The following section describes the equipment and methods used to determine the combustion parameters, smoke opacity, size and chemical composition of aerosols. Then, the results of this experimental work are presented and discussed. Heat release rate and smoke production rate are examined. Finally, the investigation of aerosols at the macroscopic, microscopic and molecular scales are detailed.

2. Methodology

2.1. Fuel samples

Dead and dry leaves and twigs of *Cistus monspeliensis* (a typical shrub of the Mediterranean maquis) were used in this study. To remove

Fig. 1. Sample holder with a) leaves and twigs with diameter of b) 2 mm, c) 4 mm, d) 8 mm and e) 15 mm.