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# Emission factors and chemical characterization of particulate emissions from garden green waste burning

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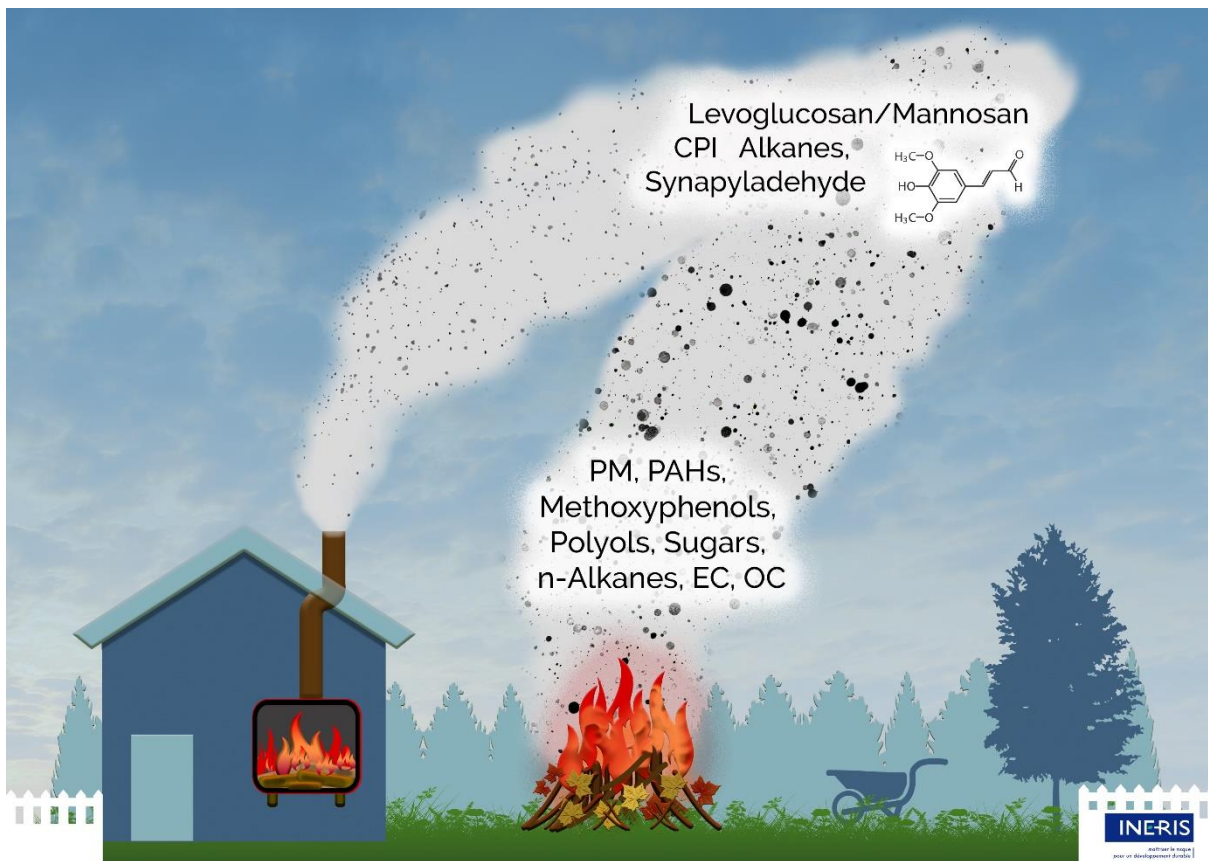
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## Abstract

This work provides an evaluation of the emission factors (EFs) of typical garden waste burning (fallen leaves and hedge trimming) in terms of particulate matter (PM), elemental and organic carbon (EC-OC) together with a detailed chemical characterization of 88 particle-bound organic species including polycyclic aromatic hydrocarbons (PAHs), levoglucosan and its isomers, lignin breakdown products (methoxyphenols), cholesterol, alkanes, polyols and sugars. Furthermore, wood-log based burning experiments have been performed to highlight key indicators or chemical patterns of both, green waste and wood burning (residential heating) sources, that may be used for PM source apportionment purposes. Two residential log wood combustion appliances, wood stove (RWS) and fireplace, under different output conditions (nominal and reduced) and wood log moisture content (mix of beech, oak and hornbeam), have been tested. Open wood burning experiments using wood logs were also performed. Green waste burning EFs obtained were comparable to the available literature data for open-air biomass burning. For PM and for most of the organic species studied, they were about 2 to 30 times higher than those observed for wood log combustion experiments. Though, poor performance wood combustions (open-air wood log burning, fireplace and RWS in reduced output) showed comparable EFs for levoglucosan and its isomers, methoxyphenols, polyols, PAHs and sugars. Toxic PAH equivalent benzo[a]pyrene EFs were even 3-10 times higher for the fireplace and open-air wood log burning. These results highlighted the impact of the nature of the fuel burnt and the combustion performances on the emissions. Different chemical fingerprints between both biomass burning sources were highlighted with notably a predominance of odd high-molecular weight n-alkanes (higher carbon preference index, CPI), lower levoglucosan/mannosan ratio and lower sinapylaldehyde abundance for green waste burning. However, the use of such indicators seems limited, especially if applied alone, for a clear discrimination of both sources in ambient air.



## 1. Introduction

The impacts of airborne particles (particulate matter, PM) on air quality, and therefore on health, is widely recognized (Brook et al., 2010; Brunekreef and Holgate, 2002; Delfino et al., 2005; Donaldson et al., 2001; Heal et al., 2012; Pope et al., 2009, 2002; Rajagopalan et al., 2018; WHO (REVIHAAP), 2013). PM are regulated pollutants in several countries (European Official Journal, 2008; Heal et al., 2012) and, in order to implement efficient actions to reduce their emission and ambient air concentration levels, the knowledge of their sources is of major concern for air quality policy-makers.

One of the most critical PM source in ambient air is biomass burning notably in winter period when wood burning is used for residential heating purposes. Many previous studies have already shown the major impact of this source on ambient air PM concentration levels in rural, suburban and urban areas worldwide (Chen et al., 2017; Crippa et al., 2013; Denier van der Gon et al., 2015; Favez et al., 2009, 2010; Fuller et al., 2014; Herich et al., 2014; Kotchenruther, 2016; Lanz et al., 2010; Maenhaut et al., 2012; Petit et al., 2014; Puxbaum et al., 2007; Saarikoski et al., 2008; Srivastava et al., 2018; Viana et al., 2016; Vicente and Alves, 2018; Weber et al., 2019; Zhang et al., 2019, 2020). The apportionment of biomass burning sources is usually achieved thanks to typical source marker (tracer) species such as levoglucosan, potassium or retene (Bhattacharai et al., 2019; Hopke et al., 2020; Karagulian et al., 2015; Ramdahl, 1983; Simoneit et al., 1999<<). This latter is not highly specific and can be also emitted by coal combustion (Shen et al., 2012). Acetonitrile (CH<sub>3</sub>CN) and chloromethane (CH<sub>3</sub>Cl) are also considered as gas-phase tracers for biomass burning (Edgerton et al., 1984; Khalil et al., 1983; Holzinger et al., 1999). Key non-methane volatile organic compounds (VOC) fingerprints can be also use for such purpose (Kumar et al., 2020). In some places, such chemical compounds also showed substantial concentrations out of the heating season (Bhattacharai et al., 2019; Puxbaum et al., 2007; Vicente and Alves, 2018; Weber et al., 2019; Zhang et al., 2019). In these cases, combustion emission sources can be linked to the emissions from forest fires (wildfires), agricultural open burning (crop field residues), but also from garden green waste burning (garden bonfires or backyard burning) (Akagi et al., 2011; Alves et al., 2010, 2019; Andreae, 2019; Andreae and Merlet, 2001; Crutzen and Andreae, 1990; Hays et al., 2005; Iinuma et al., 2007; Pio et al., 2008; Sommers et al., 2014; Wiedinmyer et al., 2014).

Although the collection of garden green waste (trimming of trees, shrubs or hedges, fallen leaves, grass clippings...) and composting are quite well organized and efficient in many developed countries,

their burning is still a common practice even if it is forbidden for instance in several European countries, in the USA or in Canada (Cogut, 2017; Eades et al., 2020; Mihai et al., 2019; Wiesen and Ciceu, 2018). For examples, in France, such practice is currently estimated to account for about a million tons of green waste burnt each year, and in England and Romania to about 0.2-0.4 kg household<sup>-1</sup> day<sup>-1</sup> burnt in rural areas (Eades et al., 2020; INDDIGO et al., 2008; Mihai et al., 2019).

Due to poor combustion performances, open-air combustion leads to significant gaseous and particulate pollutant emissions which can differ depending on the fuel burnt. Numerous studies have reported pollutant emission factors from agricultural waste burning (Akagi et al., 2011; Alves et al., 2019; Andreae, 2019; Andreae and Merlet, 2001; Chen et al., 2017; Estrellan and Iino, 2010; Hays et al., 2005; Jenkins et al., 1996a; Jenkins et al., 1996b; Lemieux et al., 2004; Li et al., 2007; Turn et al., 1997; Wiedinmyer et al., 2014; Zhang et al., 2011, 2007). By comparison, the chemical characterization of the emissions from garden green waste burning (garden bonfires) is less documented even if this practice induces large emissions of several toxic compounds such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/F), polychlorinated biphenyls (PCBs), etc. (Alves et al., 2019; Andreae, 2019; Hays et al., 2002; Hedman et al., 2005; Kannan et al., 2005; Kaufmann, 1997; Lutes and Kariher, 1996; Schmidl et al., 2008a; Wardoyo, 2007; Wevers et al., 2004; Wiedinmyer et al., 2014). In addition, the discrimination between residential wood burning and green waste burning aerosol sources in ambient air is difficult and rarely achieved. Thus, a better knowledge of green waste burning PM chemical composition, notably investigating potential specific chemical fingerprint, would be of great value for PM source apportionment purposes.

One of the main objectives of this work was to document PM emissions factors (EFs) for some typical garden green waste burnings (hedge trimming and fallen leaves), as well as their carbonaceous fraction (elemental carbon (EC) and organic carbon (OC) together with a detailed chemical organic speciation. The second main objective was to search for some specific organic molecular markers and/or chemical patterns of both, green waste and wood burning (residential heating) sources, that can be further used for PM source apportionment purposes.

In this context, organic species such as well-known biomass burning markers (monosaccharide anhydrides including levoglucosan, mannosan and galactosan) (Bhattarai et al., 2019; Simoneit et al., 1999; Simoneit, 2002), lignin breakdown products (methoxyphenols including guaiacyl and syringyl

types) (Hawthorne et al., 1988, 1989; McDonald et al., 2000; Nolte et al., 2001; Oros and Simoneit, 1999; Simoneit et al., 1993; Simpson et al., 2005), n-alkanes, known for their typical source combustion patterns linked to their molecular weight and parity (predominance of high molecular weight n-alkanes with odd carbon number - n-C<sub>27</sub>, n-C<sub>29</sub>, n-C<sub>31</sub> – in green waste burning) (Simoneit et al., 1991; Hays et al., 2002), and PAHs, toxic compounds largely emitted by biomass burning (IARC, 2010; Kim et al., 2013; Ravindra et al., 2008), have been targeted. In addition, as several authors also highlighted their presence in the water-soluble organic aerosol fraction from biomass burning emissions (Xu et al., 2019; Graham et al., 2002; Medeiros and Simoneit, 2008), the analysis of polyols, simple sugars and sugar alcohols has been also performed.

## **2. Material and Methods**

### **2.1 Biomass burning experiments**

Garden green waste burning experiments were performed in simulated real-world conditions in a large combustion chamber (1000 m<sup>3</sup>) to simulate the ambient air dilution conditions and account for post-combustion processes (condensation, evaporation/desorption and chemical reactions) occurring in close field after introduction of the emissions into the atmosphere (Nalin et al., 2016; Nussbaumer, 2008). Such large inner dimensions minimized wall effects (Fig. S1 of the Supplementary Material, SM). The system includes several air flow and velocity measuring locations (Mac Caffrey and Pitot probes) as well as temperature probes. The aerualic of the combustion chamber is thus well known and controlled in terms of dilution rate and smoke residence time. Smoke emissions were collected in the upper part of the combustion chamber through a duct and different air flow extractions have been applied (constant flows from  $25 \times 10^3$  to  $85 \times 10^3$  Nm<sup>3</sup> h<sup>-1</sup>, depending on the experiment) in order to obtain the required dilution factors (Table 1). Note that, smoke temperatures recorded were related to the outside ambient air temperatures observed during the experiments performed in winter season (from 13/02 to 01/03/2018). They were about 3 to 6°C higher than the ambient air ones due to the warm air input.

Garden green waste burning experiments were performed on a sand-bed located at the center of the combustion chamber. Two types of fuels were burnt, namely hedge trimming, with branches and leaves (mix of local species including cedar, bay leaf, privet...) and fallen tree leaves (local species). After fire ignition using a butane gas burner ( $\approx$  1 min), combustion experiments were successively

carried out with known amount of fuel heap (4-5 kg of leaves or 25 kg of hedge trimming), in order to have combustion durations of about 1 h.

Complementary combustion experiments conditions were also performed using two residential wood log heating appliances (fireplace and residential wood stove - RWS). A cast-iron RWS, with only one air inlet, was used and experiments were performed under three different output conditions (nominal, reduced and brisk). The reduced output was obtained by closing the air inlet while the brisk one was achieved by fully opening it. The tests at the different outputs were carried out with similar wood loadings to obtain combustion duration of about 1 h. Both the, RWS and fireplace were equipped with a 4m-height heat-insulated chimney pipe with a diameter of 250 mm (CEN (European Committee for Standardization), 2018).

Furthermore, to evaluate the impact of the combustion quality on the emitted PM chemical composition, open-air wood burning experiments, using wood logs, were also performed following the same procedure as for green waste burning tests. A mix of three wood species (oak, hornbeam and beech) was used. As the moisture content of green waste burning is significant (45 to 60%), two wood log moistures have been tested (15 and 25%, determined using a conductimetry probe) for the residential heating experiments to evaluate the impact of this parameter on the PM chemical composition observed. The fire in the RWS or fireplace was initiated using a small quantity of wood. Both residential heating appliances were preheated systematically before each test with a first wood load. Once the preload fully burnt ( $\text{CO}_2 < 4\%$  measured at the emission), a weighted wood load (3-4 kg of wood logs) was added into the residential heating appliance to start the experiment in agreement with the device power, the test duration (around 1 h), and to reach a minimum temperature in the exhaust pipe ( $> 200\text{ }^\circ\text{C}$  for the RWS and  $> 90\text{ }^\circ\text{C}$  for the fireplace). All burning experiments were conducted at least in triplicates except for brisk fire output (Table 1). Additional details are available in the SM (Table S1, section 2).

## 2.2 *Samplings and measurements*

Samplings and measurements were performed at two different locations: at the emission source (only for fireplace and RWS by instrumenting the chimney pipe) and at 20 m from the emission exhaust, over a straight length of the combustion chamber smoke extraction duct (close field, after dilution) (Fig.



S2). At this location, the flow is a laminar, homogeneous and the dilution factors of the emissions were in the range of 300 - 1000, representative of close field conditions (Table 1). Dilution factors were determined using air flow ratios or gaseous compounds (CO, CO<sub>2</sub>) concentration ratios measured at each sampling point (see SM section 4 for details). In the experimental conditions used, the residence time of the smoke between the emission and the close field locations was about 10 to 30 s. Note that, as the emissions were not directly channeled for open-air biomass burning, samplings or measurements at the emission, and dilution ratio, have not been determined.

Following a 1 min-fire ignition using a gas burner for open-air burning (Fig. S1), and after the wood load addition for residential heating appliances, manual measurements and filter samplings began immediately and ended when the initial test conditions were observed (CO<sub>2</sub> concentration < 4% and no visible flames) assuming the end of the combustion. Several parameters such as atmospheric pressure, smoke temperature, air flow as well as NO<sub>x</sub>, CO, O<sub>2</sub>, CO<sub>2</sub> concentrations, were monitored continuously (1 min time resolution) by using automatic sensors or analyzers at both, emission and close field locations (Tables S3-S4). Total PM mass concentrations were also determined by gravimetric measurements (filter weighting) at both locations. In addition, the non-volatile PM mass concentrations (solid fraction measured by online weighing at 50°C) were monitored continuously (1 min time resolution) at close field using a TEOM-50 (tapered element oscillating microbalance, 1400a series, Rupprecht & Patashnick). PM samples for chemical characterization purposes were collected on quartz fiber filters (Pallflex Tissuquartz, Ø = 150 mm, using a Digitel DA-80 high volume sampler (30 m<sup>3</sup> h<sup>-1</sup>) directly connected to the combustion chamber smoke extraction duct (Fig. S2). For all PM measurements and samplings, no PM size cut was applied and total suspended particles (TSP) were collected assuming that biomass burning emissions particles were mainly associated (> 90%) to the fine aerosol fraction (D<sub>p</sub> < 2.5 µm) (Andreae, 2019; Hays et al., 2005; Janhäll et al., 2010; Nussbaumer, 2008). Further details on PM mass measurement and applied sampling procedures are available in the SM (section 3 and Table S2).

Before sampling, quartz fiber filters were heated for 12 h at 500°C to remove any organic contaminants and stored in pre-cleaned glass petri dishes prior to use. Emission sampling probes and glassware (impingers) were pre-washed using deionized water and organic solvents (acetone, dichloromethane) prior to use. Blanks correspond to the sampling of dilution air in the same conditions as the exhaust samples. Overall, about 36 biomass combustion samples and 11 field blanks (collected in the same

instrumental conditions as the combustion samples) dedicated to the chemical characterization ( $\varnothing = 150$  mm) were collected. After collection, these samples were wrapped in aluminum foil, sealed in polyethylene bags, and stored at  $-18$  °C until analysis.

### 2.3 *PM chemical characterization*

Overall, about 88 particle-bound chemical species (Tables 2, S5 and S6 for details) were analyzed using different analytical procedures briefly described hereafter.

EC-OC and total carbon (TC) were measured on  $1.5$  cm<sup>2</sup> quartz fiber filter punches by thermo-optical method (in transmission) using a Sunset Lab analyzer following the EUSAAR-2 protocol (Cavalli et al., 2010; CEN (European Committee for Standardization), 2017). Cellulose combustion markers, levoglucosan and its isomers, were analyzed by IC-PAD (ion chromatography followed by pulsed amperometric detection) after filter extraction of a  $22$  mm diameter filter punch by sonication using ultra-pure water (Verlhac et al., 2013; Yttri et al., 2015). This method allows the quantification of simple sugars, sugar alcohols and polyols including glucose, threulose, rhamnose, arabitol, sorbitol, glycerol, erythritol, inositol, xylitol, and mannitol. Anions and cations ( $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{NH}_4^+$ ,  $\text{Mg}^{2+}$ ) were also analyzed from the same water extract by IC (Jaffrezou et al., 2005)  $47$  mm diameter punches were extracted by pressurized liquid extraction (Dionex, ASE 200,  $100$  °C,  $100$  bars, 2 cycles with methanol/dichloromethane (10/90, v/v) and acetone/dichloromethane (50/50, v/v), 5 min) for an extended chemical characterization. Once reduced under a nitrogen flow (Zymark II, Caliper), the sample extracts were divided into sub-sample fractions for analysis using several analytical protocols, depending on the targeted compounds, detailed previously (Golly et al., 2015; Nalin et al., 2016). Briefly, 16 PAHs and 14 methyl-PAHs were quantified by liquid chromatography coupled with fluorescence detection (HPLC-Fluo) and gas chromatography coupled with mass spectrometry (GC-MS), respectively. The quantification of 30 n-alkanes, from  $\text{C}_{11}$  to  $\text{C}_{40}$ , pristane and phytane, was done by GC-MS. Lignin combustion by-products including, 6 guaiacyl and 7 syringyl derivatives, and cholesterol, were analyzed by GC-MS after a derivatization step using BSTFA (N,O-Bis(trimethylsilyl)trifluoroacetamide). Prior extraction, known amounts of labelled surrogate standards (tetracosane- $\text{d}_{50}$ , levoglucosan- $\text{d}_6$  and guaiacol- $\text{d}_4$ ) were added to the sample extracts for further quantification purposes by internal calibration.

## 2.4 *Quality assurance/quality control*

Analysis of the collected field blanks showed very low contamination and below the quantification limits (LQ) for most of them. Major contaminations accounted for less than 7 % of the average concentration levels observed during the biomass combustion experiments. No blank correction has been applied to the sample measurements.

Quality control for the quantification of PAHs and levoglucosan, mannosan and galactosan was achieved by the analysis of the NIST standard reference material SRM 1649b (urban dust). The results obtained were in good agreement with the certified, reference and indicative values and with those available in the literature for substances not referenced in the certificate of analysis such as levoglucosan and its isomers (Favez et al., 2021; Verlhac et al., 2013). In addition, every two years, EDYTEM and Ineris participate in national and European PAH analytical inter-laboratory comparison exercises (ILC). The last exercise showed results, for both laboratories in good agreement, with reference or consensus values (Bailleul and Albinet, 2018). The same applies for EC-OC analyses and gravimetric measurements of PM concentrations after dilution, which were carried out in agreement with the European standard procedures CEN (European Committee for Standardization), 2014, 2017) and for which satisfactory results are obtained during European ILCs (Chiappini et al., 2014; Panteliadis et al., 2015; Lagler et al., 2019).

For the RWS and fireplace experiments, PM mass concentration measurements have been performed at both, the emission and after dilution (close field) locations, a comparison of the results obtained has been performed (see SM section 5). Total PM (solid + semi-volatile fractions) emission factors differed by a factor 2 to 3 with lower concentrations determined in close field (Fig. S3). By comparison, a good agreement has been observed for the PM solid fraction (Fig. S4) highlighting a good mass transfer of species mainly associated with the particulate phase from the emission to the close field. In addition, total PM EFs determined after dilution by gravimetry and by chemical reconstruction from EC-OC measurements were in good agreement (Fig. S5) further confirming the robustness of the measurements performed after dilution. It is well-known that large dilution induces significant evaporation and desorption of semi-volatile organic species which then form, in the atmosphere, secondary organic aerosols (SOA) (Nussbaumer, 2008; Robinson et al., 2007; Donahue et al., 2012,

2009). SOA fraction was not accounted here explaining the differences observed between both measurement methods. However, as the differences observed were larger than usually observed using dilution tunnels (dilution ratios about 20-30) and during previous experiments with similar dilution ratios obtained here (Nalin et al., 2016), we cannot rule out additional losses of semi-volatile species. The combustion chamber configuration and in particular the smoke extraction duct (bends, pressure drop between the combustion chamber and the extraction duct, wall-effects enhanced by the presence of soot or other combustion residues, impact of low ambient air temperatures) may have played a role in this potential loss of semi-volatile species.

### 3. Determination of the dilution factors

Dilution factors for residential appliances experiments (RWS and fireplace) have been determined from gaseous compound (CO, CO<sub>2</sub>) concentration ratios measured at each sampling point. An example of the calculation of the dilution factor based on gaseous compounds (DF) is given in Equation (1)

$$DF = \frac{[CO_2]_E - [CO_2]_{AA}}{[CO_2]_{CF} - [CO_2]_{AA}} \quad \text{Equation (1)}$$

With:

E: emission, AA: ambient air, CF: close field (= after dilution).

The ratios between the air flow rates of the 1000 m<sup>3</sup> combustion chamber and the smoke emission rates from the heating appliances have been also used for such purposes. The air flow rates from the combustion chamber were determined using a Mc-Caffrey probe, while residential heating appliance smoke emission flows were determined by calculation following the EN 16510-1 reference method specifications (CEN (European Committee for Standardization), 2018) (see SM, section 4).

### 4. Calculation of the emission factors

The emission factors of the particulate compounds were determined from the measurements after dilution (close field) using Equation (2):

$$EF_i = \frac{[i] \times FCC \times CD}{\text{Dry mass of fuel burnt}} \quad (2)$$

With:

$EF_i$ : emission factor of the species  $i$  in  $\text{mg kg}^{-1}$  of fuel burnt (dry mass basis),

$[i]$ : concentration of the species  $i$  in particulate phase in  $\text{mg m}^{-3}$ ,

FCC: extraction flow rate from the combustion chamber,

CD: combustion duration in hours.

## 5. Results and discussion

### 5.1 Green waste burning emission factors

EFs of the particulate compounds obtained for garden green waste burning are presented in Table 2 and are discussed in detail below. Results obtained for residential appliances and open-air wood log burning are available in Tables S5 and S6.

#### 5.1.1 Particulate matter (PM)

The average total PM EFs obtained for garden green waste burning were comparable with average values of about  $33.4 \pm 5.7$  and  $34.7 \pm 9.2 \text{ g kg}^{-1}$  of dry fuel burnt for hedge trimming and fallen leaves, respectively. The non-volatile fraction accounted for about 50-60% of the total PM mass for fallen leaves and about 80% for hedge trimming. By contrast, open-air wood log burning EFs were twice lower,  $13.3 \pm 2.2 \text{ g kg}^{-1}$  for total PM mass on average, with a similar proportion of the solid fraction as fallen leaves ( $\approx 50\text{-}60\%$ ) (Table S6). Compared to green waste burning, EFs observed for residential heating appliances were 4 to 30 times lower for the fireplace ( $7.7 \pm 1.3 \text{ g kg}^{-1}$ ) and RWS ( $1.1 - 5.5 \text{ g kg}^{-1}$ ) respectively, with solid fraction accounting for about 50 - 70% (Tables S5 and S6). Note that, if fireplace and open-air wood log burning EFs were in a similar range, EFs for the fireplace were about 1.7 times lower. As no significant differences were observed between the dry and humid wood used with the RWS, the impact of wood log moisture on PM emissions seemed limited (Table S5). All these results highlighted the influence of the nature of the fuel and the combustion performances on the PM emissions.

Data available in the literature for green waste burning is scarce. However, several authors have reported PM EFs for various biomass burning materials including agricultural or crop residues, land and stubble clearing burning, and forest or savanna wildfires (Table S7). As these data are usually reported based on the mass of raw fuel burnt, we may compare our findings on the same basis, and without

considering the fuel moisture content. Average total PM EFs were then about  $13.3 \pm 2.3 \text{ g kg}^{-1}$  for hedge trimming and  $19.1 \pm 5.1 \text{ g kg}^{-1}$  for fallen leaves. These emission factors were consistent with results found in the literature, either for leaf burning (Andreae, 2019; Collet, 2011; Hays et al., 2002; Kannan et al., 2005; Lutes and Kariher, 1996; Wardoyo, 2007; Wiedinmyer et al., 2014) or tree pruning burning (Alves et al., 2019; Jenkins et al., 1996a, b). They were in the high range of values reported for the agricultural and crop residues burning (Akagi et al., 2011; Alves et al., 2019; Andreae, 2019; Chen et al., 2017; Hays et al., 2005; Jenkins et al., 1996a; Jenkins et al., 1996b; Lemieux et al., 2004; Li et al., 2007; Wiedinmyer et al., 2014), stubble-burnings or for the burning of different grasses (Akagi et al., 2011; Andreae, 2019; Christian et al., 2003; Dhammapala et al., 2007; Iinuma et al., 2007; Janhäll et al., 2010; Lemieux et al., 2004; Rennie et al., 2020; Wiedinmyer et al., 2014). Finally, our results fell within the wide range of EFs reported in the literature for forest fires (Akagi et al., 2011; Alves et al., 2011a, 2010; Andreae, 2019; Janhäll et al., 2010).

#### 5.1.2 Carbonaceous matter (TC, EC, and OC)

Average TC EFs for both green waste fuels tested were comparable and about  $21.8 \pm 9.6$  and  $24.7 \pm 9.7 \text{ gC kg}^{-1}$  of dry fuel burnt for hedge trimming and fallen leaves, respectively. Wood log combustion experiments resulted in 2 to 20 times lower average TC EFs, with an average value of  $9.0 \pm 1.2 \text{ gC kg}^{-1}$  of dry fuel for open-air wood log burning and values between 1 to 5.5 g.eg C  $\text{kg}^{-1}$  of dry fuel for residential wood heating appliances (Tables S5 and S6). Here again, no significant impact of the log wood moisture has been observed on the TC, as well EC and OC, emissions. (Table 5).

Average emission factors of EC and OC obtained for leaf burning were of about  $0.2 \pm 0.1$  and  $24.5 \pm 9.7 \text{ gC kg}^{-1}$  of dry fuel ( $0.1 \pm 0.0$  and  $13.5 \pm 5.3 \text{ gC kg}^{-1}$ , considering raw material), respectively, corresponding to an average OC/EC ratio of about to  $120.4 \pm 50.9$ . For hedge trimming experiments, the distinction between EC and OC has not been achieved due to measurement limitations related to very high sample filter loading. For open-air wood burning and fireplace combustion tests, average OC/EC ratios were largely lower ( $26.9 \pm 6.7$  and  $17.1 \pm 2.8$ , respectively). EC and OC EFs obtained for both combustions were similar ranging from 5.8 to 8.6  $\text{gC kg}^{-1}$  of dry fuel for OC and of about 0.3  $\text{gC kg}^{-1}$  for both, for EC EFs (Table S6). For the RWS, the highest OC/EC ratios were observed for the RWS in reduced output (13.1 to 20.1 on average) and were comparable to the ones obtained for the fireplace and open-air wood burning (Table S5). In nominal or brisk output conditions, such ratios were about 1

to 2 on average. All these results highlighted the impact of the nature of the fuel burnt and of the combustion performances on the carbonaceous aerosol fraction emissions.

When considering raw material, TC EFs were  $8.7 \pm 3.8$  and  $13.6 \pm 5.3$  gC kg<sup>-1</sup> for hedge trimming and leaves, respectively. These EFs values are similar to the reported values in the literature for green waste, tree pruning, crop residues burning as well for forest fires (1.1 to 31.0 g C kg<sup>-1</sup>) (Table S8) (Akagi et al., 2011; Alves et al., 2019; Christian et al., 2003; Oros et al., 2006; Oros and Simoneit, 1999; Wiedinmyer et al., 2014; Schmidl et al., 2008b; Andreae, 2019; Alves et al., 2011a). Lower TC EFs have been reported for stubble burning, grassland, savanna and pasture burning (1.4 to 5.8 gC kg<sup>-1</sup>) (Akagi et al., 2011; Alves et al., 2019; Andreae, 2019; Christian et al., 2003; Iinuma et al., 2007; Oros et al., 2006; Rennie et al., 2020; Wiedinmyer et al., 2014). Finally, the high OC/EC ratio found here for leaf burning is only comparable to maximum values reported in the literature for forest wildfires or prescribed fires (77 to 90) (Akagi et al., 2011; Alves et al., 2011a; Andreae, 2019; Robinson et al., 2011; Zhang et al., 2013; Dambruoso et al., 2014; Robinson et al., 2004).

### 5.1.3 Polycyclic aromatic hydrocarbons (PAHs)

Average  $\Sigma_{30}$ PAH emission factors for hedge trimming and leaves were  $85.3 \pm 35.2$  and  $45 \pm 22.9$  mg kg<sup>-1</sup> of dry fuel, respectively (Table 3). If considering the compounds mainly associated with the particulate phase (PAH<sub>p</sub>) in ambient air or in close field conditions (from retene to coronene and from 4-methylpyrene to methylchrysene / methylbenzo[a]anthracene (Albinet et al., 2008; Huang et al., 2014; Nalin et al., 2016; Odabasi et al., 1999; Tomaz et al., 2016), average EFs for  $\Sigma$ PAH<sub>p</sub> ranged from 6 to 29 mg kg<sup>-1</sup>, with lower values obtained for leaf burning. The same observation can be made for individual PAHs with EFs always higher for hedge trimming, except for phenanthrene. Overall, compounds with 3 or 4 aromatic rings were predominant as previously reported for the combustion of biofuels (Venkataraman et al., 2002; Keshtkar and Ashbaugh, 2007).

PAH emission factors values available in the literature can be extremely variable according to the number of PAHs considered. When considering the eight PAHs usually quantified (fluoranthene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenzo[a,h]anthracene, indeno[1,2,3-cd]pyrene, benzo[g,h,i]perylene), EFs obtained ( $8.0$  mg kg<sup>-1</sup> for hedge trimming and  $2.3$  mg kg<sup>-1</sup> for leaves, considering raw material) were consistent with results found in the literature for green waste (Lutes and Kariher, 1996), leaves (Collet, 2011), and tree pruning (Alves

et al., 2019; Collet, 2011; Jenkins et al., 1996a, 1996b; Keshtkar and Ashbaugh, 2007) (Table S9). Furthermore, they were similar with EF values reported for grassland, pasture, savanna and stubble burning (Akagi et al., 2011; Andreae, 2019; Collet and Fiani, 2006; Dhammapala et al., 2007; Iinuma et al., 2007; Lemieux et al., 2004; Oros et al., 2006) and they were in the lowest range of results obtained for forest fires and agricultural residues burning (Akagi et al., 2011; Andreae, 2019; Chen et al., 2017; Jenkins et al., 1996a, 1996b; Lemieux et al., 2004; Oros et al., 2006; Shen et al., 2011, 2013; Zhang et al., 2011).

It should be highlighted that the fireplace and open-air wood log burning displayed the highest particle-bound PAH emissions in the present study (4 to 12 times higher than any of the other fuels tested, Table S6). For the RWS,  $\Sigma\text{PAH}_p$  EFs were slightly lower than values obtained for hedge trimming but comparable to the ones observed for leaf burning (Table S5). Such results are consistent with the higher PAH EFs usually reported for smoldering and slow combustion conditions (Collet, 2011; Jenkins et al., 1996a; Jenkins et al., 1996b; Kim et al., 2018). Interestingly, the same was observed for benzo[a]pyrene (B[a]P) emissions with EFs 3 to 10 times higher for the fireplace and open-air wood log burning (9 and 3.2 mg kg<sup>-1</sup> of raw material, respectively), compared to green waste burning (0.2 – 0.8 mg kg<sup>-1</sup>, raw material). Low B[a]P EFs have been reported in the literature for the burning of green waste (Lemieux et al., 2004; Lutes and Kariher, 1996), tree pruning (Alves et al., 2019; Collet, 2011; Jenkins et al., 1996a; Jenkins et al., 1996b; Keshtkar and Ashbaugh, 2007; Lemieux et al., 2004), crop residues, savanna and peat fires (Dhammapala et al., 2007; dos Santos et al., 2002; Iinuma et al., 2007; Lemieux et al., 2004).

Following these results, a comparison of the PAH related toxic potential of the different biomass burning emissions was performed (Fig. 1). The individual PAH EFs have been converted in B[a]P equivalent EFs (B[a]P eq. EF) using toxic equivalent factors (TEFs), with B[a]P as the reference compound with conventional value of 1 (OEHHA, 2011), and following Equation (2). In order to avoid any bias in the estimations, only compounds mainly associated with the particulate phase (PAH<sub>p</sub>) in ambient air or close field conditions were considered (Table S12).

$$B[a]P \text{ eq. EF} = \left[ \sum_i EF_{PAH(i)} \times TEF(i) \right] \quad (2)$$

Results obtained highlighted the higher toxic potential of the PM emitted by the fireplace and open-air wood log burning. As previously reported by several authors, the emission of toxic compounds is highly



dependent on the nature of the fuel and the quality of the combustion (Jenkins et al., 1996b; Kim et al., 2018). Note, that no significant impact of the log wood moisture has been observed on the PAH emissions (Table S5 and Fig. 1 showing low standard deviation).

#### 5.1.4 Levoglucosan and its isomers

EFs observed for levoglucosan, mannosan and galactosan were similar for both, hedge trimming and fallen leaves. Levoglucosan was the dominating compound with EFs of about 1300 - 1600 mg kg<sup>-1</sup> of dry fuel burnt while values observed for both isomers were about 7 to 8 times lower (180 - 240 mg kg<sup>-1</sup> of dry fuel burnt (Table 2). EFs obtained for open-air wood log burning and for the fireplace were of the same order of magnitude than values observed for green waste burning (on average, 754 - 1600, 50 - 90, 40 - 60 mg kg<sup>-1</sup> of dry fuel burnt for levoglucosan, mannosan and galactosan, respectively) (Table S6). By contrast, values obtained for the residential wood heating were on average 2 to 5 times lower for the reduced output and up to 200 times lower for the nominal and brisk output operating conditions (Table S5). Only the values observed for the RWS in nominal and brisk output conditions were significantly lower (7 - 41 mg kg<sup>-1</sup> of dry fuel burnt) in agreement with the higher combustion temperatures and performances inducing lower monosaccharide anhydride species emissions (Albinet et al., 2015; Calvo et al., 2015; Kuo et al., 2008; Viana et al., 2016; Vicente and Alves, 2018). This was also supported by the large differences in levoglucosan/K<sup>+</sup> ratio values observed between nominal or brisk output and the reduced output, fireplace or open-air biomass burning experiments (Fig. S6). Potassium is known to be predominantly emitted from the flaming phase of a fire, as in nominal or brisk output conditions, whereas levoglucosan is emitted during both, the flaming and smoldering fire phases (Echalar et al., 1995; Sullivan et al., 2014; Lee et al., 2010). Finally, impact of the log wood moisture seemed really limited (Table S5 and Fig. S6). Overall, differences observed between green waste and residential appliances, as well with open-air wood log burning, were linked to the combustion conditions and fuel used (Albinet et al., 2015; Mazzoleni et al., 2007; Schmidl et al., 2008a; Viana et al., 2016; Vicente and Alves, 2018). Finally, whatever the type of combustion considered, levoglucosan accounted for more than 90% of the emissions of the monosaccharide anhydrides.

The levoglucosan EFs considering raw material were about 540 ± 60.0 and 900 ± 441.2 mg kg<sup>-1</sup> for hedge trimming and leaves, respectively. These results were within the range of the emission factors found in the literature for green waste, (Andreae, 2019; Medeiros and Simoneit, 2008; Schmidl et al.,

2008b), forest fires (Akagi et al., 2011; Alves et al., 2011a; Andreae, 2019), stubble burning, grassland and peat fires (Akagi et al., 2011; Andreae, 2019; Christian et al., 2003; Iinuma et al., 2007; Oros et al., 2006; Rennie et al., 2020; Wiedinmyer et al., 2014) and agricultural residues (Akagi et al., 2011; Christian et al., 2003; Oros et al., 2006; Oros and Simoneit, 1999; Wiedinmyer et al., 2014). However, they were 2 to 5 times higher than those indicated for tree pruning (Alves et al., 2019) (Table S10). Mannosan and galactosan EFs based on raw material were about  $93 \pm 7.2$  and  $71 \pm 7.6$  mg kg<sup>-1</sup>, respectively, for hedge trimming and  $99 \pm 38.6$  and  $130 \pm 53.1$  mg kg<sup>-1</sup> for leaves. As for levoglucosan, these EFs were within the range of literature data for green waste burning (159 - 599 and 177 - 439 mg kg<sup>-1</sup>, respectively), savanna, grassland and peat fires (22 – 650 and 20 - 670 mg kg<sup>-1</sup>, respectively) (Iinuma et al., 2007; Medeiros and Simoneit, 2008) while lower values have been reported for forest fires (46 and 31 mg kg<sup>-1</sup>), tree pruning (42 and 21 mg kg<sup>-1</sup>) and grassland fires (0.2 - 10 mg kg<sup>-1</sup> and 0.2 - 66 mg kg<sup>-1</sup>) (Alves et al., 2019, 2011b; Oros et al., 2006).

#### 5.1.5 Lignin breakdown products and cholesterol

The average EFs of the total lignin breakdown ( $\Sigma$ Methoxyphenols) products were in the range 250 - 358 mg kg<sup>-1</sup> of dry fuel burnt for green waste burning (Table 2). The proportions of both, guaiacyl and syringyl derivatives, were quite similar in both cases (50/50), hedge trimming and leaves. Syringol and methylsyringol dominated the emissions of the syringyl derivatives while vanillin and guaiacylacetone were the most abundant over the guaiacyl ones. Average  $\Sigma$ Methoxyphenols EFs were about 2 to 4 times higher ( $808 \pm 488.8$  mg kg<sup>-1</sup>) for open-air wood log burning or comparable for the fireplace ( $394 \pm 257.7$  mg kg<sup>-1</sup>) and RWS in reduced output (160-195 mg kg<sup>-1</sup>) while they were significantly lower in nominal or brisk outputs (1.3 - 10.4 mg kg<sup>-1</sup>) (Tables S5 and S6). Considering the standard deviations obtained from the different replicate experiments, no impact of the log wood moisture was observed on the RWS EFs (Table S5). These results highlighted the significant impact of the poor combustion performances on the large emissions of such species. Furthermore, green foliage contains lower density of lignin than wood, explaining also the differences observed in terms of EFs (Hays et al., 2002). By comparison to green waste, syringyl derivatives were largely predominant for wood log-based combustions and accounted for 60 to 80% of the total methoxyphenols emissions. Note, that lignin of hardwoods, as used here, consists of both guaiacyl and syringyl types structural units in similar proportions, in contrast to softwoods in which guaiacyl types are predominant (McDonald et al., 2000).

As the nature and number of methoxyphenols reported in the literature for similar green waste fuels are highly variable, it is not possible to make a direct comparison of the  $\Sigma$ Methoxyphenols EFs. Considering the main compounds emitted such as vanillin (11 - 12 mg kg<sup>-1</sup> raw material), syringol (20 - 43 mg kg<sup>-1</sup>) and methylsyringol (24 - 33 mg kg<sup>-1</sup>), EFs were in comparable to the values reported for foliar fuels (vanillin, 15 - 100; syringol, 80 – 325 and methylsyringol, 36-274 mg kg<sup>-1</sup>) (Hays et al., 2002), while significantly lower EFs were observed for different grasses, bamboo and sugarcane (vanillin, 0.2 - 0.3 and syringol, 0.2 - 10 mg kg<sup>-1</sup>) (Oros et al., 2006).

Interestingly, cholesterol was only emitted by leaf burning (14.6 ± 10 mg kg<sup>-1</sup> dry-mass basis) and was not detected or in very low amount (0.3 mg kg<sup>-1</sup>) for all other biomass combustions (Tables S5 and S6). Cholesterol has been reported to play a crucial role in the cellular growth and development of plants and is therefore associated to plant debris (Nolte et al., 2002; Rogge et al., 1994; Schaller, 2003). However, only very few studies have reported its emission from green waste or crop residues burning (Fushimi et al., 2017) as this compound is usually associated to cooking emissions (Schauer et al., 2002; Zhao et al., 2007; Rogge et al., 1991; Nolte et al., 1999).

#### 5.1.6 Alkanes

Average total n-alkanes EFs (from C<sub>11</sub> to C<sub>40</sub>) for hedge trimming and leaves were, respectively, 83 ± 43.9 and 255 ± 163.2 mg kg<sup>-1</sup> of dry fuel burnt with the highest EFs for high molecular weight odd alkanes C<sub>25</sub>, C<sub>27</sub> and C<sub>29</sub> (Table 2).  $\Sigma$ n-Alkanes EFs were about 10 times lower for wood log-based combustions with values ranging from 8 to 30 mg kg<sup>-1</sup> for the RWS, with no impact from log wood moisture (Table S5), and from 14 to 16 mg kg<sup>-1</sup> for the fireplace or open-air wood log burning (Table S6). Pristane and phytane, well-known compounds emitted by vehicular engines (Rogge et al., 1993; Didyk et al., 2000; Simoneit, 1984; Alves, 2008), were below the detection limits for green waste burning and have been only quantified with low EFs (0.0 - 1.0 mg kg<sup>-1</sup>) for wood log-based combustions.

Alkane EFs reported in the literature for the particulate phase usually considered compounds from C<sub>21</sub> to C<sub>31</sub>.  $\Sigma$ n-Alkanes C<sub>21</sub>-C<sub>31</sub> EFs obtained were about 24 and 128 mg kg<sup>-1</sup> raw material, for hedge trimming and leaves, respectively. These values were comparable to the ones obtained for green waste and leaf burning (15 - 180 mg kg<sup>-1</sup>) and in the same range as for stubble, peat, savanna and grassland burning (1 – 688 mg kg<sup>-1</sup>) (Hays et al., 2005, 2002; Iinuma et al., 2007; Medeiros and Simoneit, 2008; Oros et al., 2006) (Table S11).

### 5.1.7 Polyols, simple sugars and sugar alcohols

$\Sigma$ Polyols + sugars EFs were 10 times higher for hedge trimming ( $1987 \pm 596.6 \text{ mg kg}^{-1}$  of dry fuel burnt on average) compared to leaves ( $236 \pm 204.6 \text{ mg kg}^{-1}$ ). Erythritol and sorbitol were the most emitted polyols for hedge trimming ( $1470 \pm 472.4 \text{ mg kg}^{-1}$  and  $300 \pm 68.2 \text{ mg kg}^{-1}$ ), while these compounds were not detected from leaf burning. Glycerol was the most abundant in this latter and EFs were comparable for both biomass types ( $163 \pm 36.5$  and  $217 \pm 197.6 \text{ mg kg}^{-1}$ ). Xylitol, threulose and rhamnose have not been detected in any case and glucose was only emitted during hedge trimming combustion. Finally, EFs for arabitol ( $6 - 8 \text{ mg kg}^{-1}$ ) and mannitol ( $3 - 8 \text{ mg kg}^{-1}$ ) were comparable for both burning materials. For wood log-based combustions, comparable total EFs ( $55 - 310 \text{ mg kg}^{-1}$ ) were observed for poor performance combustions such as open-air, fireplace and RWS in reduced output (Tables S5 and S6). At nominal and brisk outputs, the emissions of polyols and sugars were significantly lower and most of the time  $< \text{LD}$ . Impact of the log wood moisture on the emissions seemed limited (Table S5). As for green waste burning, glycerol and erythritol showed the higher EFs for all wood log-based combustions and xylitol, sorbitol, mannitol, threulose, rhamnose and glucose were all  $< \text{LD}$ . All these results showed the impact of the quality of the combustion as well of the biomass burnt on the emissions of polyols, simple sugars and sugar alcohols.

EF data of these species for green waste burning, or similar materials, are scarce in the literature. However, several authors reported different clues on their presence in such biomass and in emissions during the burning process. High leaf glycerol concentration contents have been reported in crop plants due to glycerol synthesis in the root and transport to the leaves together with part of the production in the leaves themselves (Gerber et al., 1988). Similarly, sorbitol is found in the leaves and fruit of higher plants (Lewis and Smith, 1967) and therefore emitted during leaf burning (Schmidl et al., 2008b). Arabitol and mannitol constitute energy reserve materials in fungi (Zhang et al., 2010) and are highly correlated with fungal activity (Samaké et al., 2019b, a), which could be enhanced in fallen leaves and green waste (Bauer et al., 2002, 2008; Schmidl et al., 2008b). Inositol was previously associated to biomass burning from forest and pasture fires (Graham et al., 2002; Medeiros and Simoneit, 2008). Finally, as reported here, emission of erythritol was only observed during the combustion of tree pruning or green waste and not for leaf burning (Alves et al., 2019; Medeiros and Simoneit, 2008).

## 5.2 *Source specific molecular markers or chemical patterns*

### 5.2.1 *PAHs and methyl-PAHs*

The PAHs and methyl-PAHs (only PAH<sub>p</sub> compounds considered) chemical profiles obtained for the different type of combustion tested are compared on Fig. S7. Retene was the dominating compound whatever the combustion considered confirming the well-known specificity of this compound largely emitted by biomass burning (Ramdahl, 1983; Simoneit, 2002). The contribution observed to total PAH<sub>p</sub> was even up to 80% for fireplace and open-air wood log burning while for RWS and green waste burning, its contribution was about 30 - 40%. Besides, no characteristic chemical pattern for one or the other combustion tested can be highlighted as the PAH chemical profiles were all quite similar. The same conclusion can be made based on the methyl-PAH chemical profiles. No significant predominance of any specific compound can be highlighted for either residential wood heating or green waste burning (garden bonfires). Finally, the PAHs do not allow the discrimination between both types of biomass burning. Note that, even PAH chemical patterns and/or PAH diagnostic ratios are commonly used in the literature for source apportionment purposes (Nalin et al., 2016; Ramdahl, 1983; Robinson et al., 2006), their source specificity is questionable (Dvorská et al., 2011; Katsoyiannis et al., 2011; Wu et al., 2021) as shown also here.

### 5.2.2 *Polyols, simple sugars and sugar alcohols*

Chemical profiles obtained for polyols, sugar and sugar alcohols are presented on Fig. 2 (only the 7 compounds, over the 10 analyzed, systematically > LQ were considered). Overall, glycerol and erythritol were the major compounds accounting for about 40 - 80 % and 20 - 40 % to the sum of the species considered. Similar results were reported in literature in the case of tree pruning burning, forest, pasture or temperate climate green vegetation fires (Alves et al., 2019; Graham et al., 2002; Medeiros and Simoneit, 2008). To our knowledge, no data is available in the literature for residential wood combustion. A higher contribution of erythritol and sorbitol can be highlighted for green waste burning. However, as both compounds were not detected from fallen leaves burning (see section 4.1.7 and Table 2), the resulting standard deviations were significant. Such results contrast with previous studies suggesting the use of sorbitol (and arabitol) as marker of leaf burning in ambient air (Schmidl et al., 2008b). In addition, sorbitol content in leaves depends on several parameters like tree species, season and fruit removal. Higher sorbitol concentrations have been reported in fruit tree but fruit removal

induces a decrease in the observed in leaf content (Nii, 1997). Therefore, it seems difficult to use these compounds to discriminate between both biomass burning sources, residential wood heating and green waste burning (garden bonfires), in ambient air especially as such species mainly arise from primary biogenic emissions (fungal spores) (Medeiros et al., 2006; Samaké et al., 2019b, a; Bauer et al., 2008).

### 5.2.3 Lignin breakdown products: methoxyphenols

Chemical profiles obtained for methoxyphenols are shown on Fig. 3. Regardless the combustion experiment, the major contributions for syringyl derivatives were observed for syringol, methylsyringol and acetosyringone, ranging from 10 to 20% of the total methoxyphenol emissions, while for guaiacyl derivatives, vanillin and guaiacylacetone were the dominating compounds with methoxyphenol contributions ranging from 5 to 12%. Given the large experimental standard deviations obtained, no significant differences between residential wood heating and green waste burning chemical profiles have been observed. Only a lower contribution of sinapylaldehyde could be highlighted for green waste burning. This compound has been observed previously in wood combustion emissions (Fine et al., 2001, 2002a, 2002b, 2004; Schauer et al., 2001). However, as it is rarely analyzed and reported in the literature for other biomass burning types including green waste. It seems difficult to conclude on its residential wood heating specificity and/or on its actual minor contribution in green waste burning emissions. It might be used as a complementary indicator, together with other key molecular markers or ratios, for PM source apportionment purposes.

### 5.2.4 *n*-Alkanes

*n*-Alkanes chemical profiles are shown on Fig. S8. As already mentioned in the literature, a predominance of high molecular weight alkanes (C<sub>21</sub>-C<sub>31</sub>) for all biomass combustion conditions was also observed here (bin Abas et al., 1995; Hays et al., 2002, 2005; Iinuma et al., 2007; Oros et al., 2006; Oros and Simoneit, 2001a, 2001b; Rogge et al., 1998). In addition, a higher contribution of both odd alkanes, C<sub>27</sub> and C<sub>29</sub>, was observed for green waste burning. These results agreed with the ones reported previously (Hays et al., 2002). The predominance of high molecular weight *n*-alkanes with an odd number of carbons in green waste burning emissions is linked to plant waxes present on the leaves (Simoneit et al., 1991, 1993). The cuticular plant wax is a complex mixture of long-chain aliphatic

compounds and one of the most abundant compound groups is odd carbon number n-alkanes from C<sub>25</sub>–C<sub>33</sub> (Kolattukudy, 1970).

The influence of carbon number parity in the chemical signature of n-alkanes can be represented by the carbon preference index (CPI). This indicator is obtained by the ratio of concentrations of odd alkanes to even alkanes following Equation (2) (Marzi et al., 1993). A predominance of odd carbon number alkanes is observed for CPI values greater than 1.

$$\text{CPI} = \left( \frac{\sum_{i=n}^m C_{2i+1} + \sum_{i=n+1}^{m+1} C_{2i+1}}{2 \cdot \sum_{i=n+1}^{m+1} C_{2i}} \right) \quad (2)$$

with here, n = 9 et m = 17.

Resulting CPI values (calculated from C<sub>18</sub> to C<sub>35</sub>), for green waste burning, were about 2.5 to 4 (Fig. 4A) while values observed for the RWS and fireplace were twice lower (CPI = 1.1 - 1.9). Furthermore, the CPI value observed for open-air wood log burning was similar and about 1.5. These two latter ones were comparable to those available in the literature for residential heating appliances (0.9 - 1.9, C<sub>19</sub>-C<sub>30</sub>) (Rogge et al., 1998). The CPI values obtained here for green waste burning were comparable to those available in the literature for the combustion of foliar fuels (1.6 - 4.4, C<sub>16</sub>-C<sub>35</sub>) (Hays et al., 2002), forest fires (3.6, C<sub>17</sub>-C<sub>35</sub>) (bin Abas et al., 1995) or savanna fires (2.9, C<sub>20</sub>-C<sub>34</sub>) (Iinuma et al., 2007) and the burning of grasses (2.1 - 11.6, C<sub>16</sub>-C<sub>35</sub>) (Oros et al., 2006). Other studies, with test conditions close to open-air burning, showed CPI values for conifers (dry and green needles, branches) in the range of 0.9 - 3.4 and for deciduous trees (dry and green leaves, branches) in the range of 2.6 - 6.8 (C<sub>14</sub>-C<sub>34</sub> or C<sub>35</sub>) (Oros and Simoneit, 2001a, b). By comparison, the CPI for leaf abrasion products (vegetable waxes) is in the range of 12.1 - 12.4 (C<sub>19</sub>-C<sub>36</sub>) (Rogge et al., 1993). The presence of leaves, and thus the emission of vegetable waxes, explained the higher CPI values obtained here for green waste burning. The alkane chemical patterns, and associated CPI, seem characteristic of each biomass burning source. However, as for the literature data, the low differences observed considering the experimental standard deviations obtained, make their use difficult to discriminate efficiently between residential wood heating and green waste burning sources in ambient air especially if CPI of alkanes is used alone.

### 5.2.5 *Levoglucosan/mannosan ratios*

Several authors have reported the use of the levoglucosan/mannosan (L/M) ratio to discriminate between hardwood (angiosperm) and softwood (gymnosperm) burning (Kawamura et al., 2012; Kirchgeorg et al., 2014; Kuo et al., 2011; Verma et al., 2015). High L/M ratios are indicative of the combustion of hardwoods while lower values are typical of the combustion of softwoods. For the latter, average L/M ratios reported in the literature are about 3 - 5 (with values ranging from 0.6 to 14, depending on the study) while for hardwoods, average values are about 13 - 14 (with values ranging from 3 to 32) (Engling et al., 2009, 2006; Fine et al., 2004, 2002a; Nolte et al., 2001; Oros and Simoneit, 2001a, 2001b; Schmidl et al., 2008b).

Here, high L/M ratios (19 to 30 on average, RWS NO HW excluded) have been observed for residential wood heating appliances, with values (Fig. 4B). Given the wood species burnt (mixture of oak, beech and hornbeam) these results are consistent with those previously reported in the literature for such hardwoods. L/M ratios seemed slightly higher in the case of humid wood combustion tests, but the standard deviations between replicates were also higher, especially for RWS in nominal output. The combustion performances did not show any significant impact on the L/M ratio. Indeed, the values obtained for the RWS at nominal and reduced outputs (not determined for brisk output as mannosan was < LQ), the fireplace and open-air wood log burning, were all similar. In comparison, the L/M ratios obtained for green waste burning were significantly lower (7 for leaves and 5 for hedge trimming, on average). These results are in agreement with those available in the literature for leaf burning (5.1 - 5.9) (Schmidl et al., 2008b), grasses (tundra, pampas, ryegrass...) (2 - 9) (Oros et al., 2006) (tree pruning (olive, acacia, vine, willow) (4.2 - 8.8) (Alves et al., 2019) and for several different fuels that might burn during wildfires or prescribed fires in Western and Southeastern USA (branches, straws, needles, duffs, leaves, grasses) (10.6 on average, 5.6 on median, but with a wide range from 0.8 to 55) (Sullivan et al., 2008). Results reported in literature for crop residues burning (sugar cane, cotton, maize, rice, wheat, etc.) are much higher (13-55) (dos Santos et al., 2002; Sheesley et al., 2003; Zhang et al., 2007) highlighting the influence of the nature of the fuel on this ratio. Thus, if the L/M ratio may discriminate between residential heating and green waste burning (garden bonfires) emissions, its use alone seems limited, especially when other combustion processes are present such as agricultural waste burning or the use of softwoods for residential heating (lower L/M ratio).



## 6. Conclusions

This study provides emission factors for key particulate pollutants and chemical species (PM, EC, OC and 88 particle-bound organic compounds) for the burning of typical garden waste such as fallen leaves and hedge trimming (backyard burning). EFs factor obtained, from this still common although forbidden practice, have been compared to literature data from open-air biomass burning or from similar burning material as well as to emissions from wood log-based combustion experiments including open-air and two residential appliances (RWS, fireplace) working in different conditions in terms of outputs or wood moisture. The database obtained may contribute to improve emission inventories and to more accurately apportion biomass combustion sources in ambient air.

Overall, green waste burning EFs obtained here agreed well with the few data available in the literature and emissions were about 2 to 30 times higher than wood-log based combustions for PM and most of the species studied in this work. However, lower or poor-quality wood log-based combustions such as open-air wood log burning, fireplace or RWS in reduced output showed comparable EFs for levoglucosan and its isomers, lignin breakdown derivatives, polyols, sugar and sugar alcohols. Furthermore, EFs of toxic compounds such as PAHs were comparable or significantly lower (4-12 times) than the RWS and fireplace/open-air wood log burning ones. All these results obtained highlighted the impact of the nature of the fuel burnt and the combustion performance on the emissions.

Differences between both biomass burning sources in terms of chemical patterns or key species, such as the CPI of n-alkanes, levoglucosan/mannosan ratio or the abundance of sinapylaldehyde within the guaiacyl and syringyl derivatives, could also be observed. However, results obtained here pointed the limits of using these indicators for a clear discrimination of these sources in ambient air especially if used alone. Significant variability in the results have been observed (large standard deviations) and possible confusing factors in ambient air could also exist with other combustion processes not considered here (for example softwood combustion). These elements should be only used as complementary indicators, or as cross-validation, for source apportionment purposes based on statistical source-receptor modelling for instance.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at XXX.

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**Table 1**

Summary of the biomass burning experiment conditions.

Combustible	Output	Number of replicates	Fuel load (kg)	Fuel humidity (%)	Combustion duration (h)	Emission flow (Nm <sup>3</sup> h <sup>-1</sup> )	Extraction flow (Nm <sup>3</sup> h <sup>-1</sup> )	Smoke temperature after dilution (°C) <sup>d</sup>	Dilution factor <sup>e</sup>
<b>Open-air burning</b>									
Leaves	-	4	4.5	45 <sup>b</sup>	0.80	nd <sup>c</sup>	50 × 10 <sup>3</sup>	1.2	nd
Hedge trimming	-	5	25.0	60 <sup>b</sup>	0.62	nd	50 × 10 <sup>3</sup>	3.5	nd
Wood logs <sup>a</sup>	-	4	3.5	15	0.73	nd	51 × 10 <sup>3</sup>	0.9	nd
<b>Residential wood stove</b>									
Wood logs <sup>a</sup>	Nominal	5	3.5	15	0.87	30.4	25 × 10 <sup>3</sup>	4.3	831
Wood logs <sup>a</sup>	Nominal	4	3.3	25	0.77	36.6	29 × 10 <sup>3</sup>	-1.4	781
Wood logs <sup>a</sup>	Reduced	5	3.8	15	1.00	30.2	25 × 10 <sup>3</sup>	4.1	840
Wood logs <sup>a</sup>	Reduced	4	3.1	25	0.99	31.2	29 × 10 <sup>3</sup>	-2.6	916
Wood logs <sup>a</sup>	Brisk fire	2	3.6	15	0.69	nd	25 × 10 <sup>3</sup>	5.0	nd
<b>Fireplace</b>									
Wood logs	-	3	4.0	15	0.96	260	85 × 10 <sup>3</sup>	-3.0	329

<sup>a</sup> Mix of oak, hornbeam and beech (1 log of each)<sup>b</sup> Estimated (Collet, 2011).<sup>c</sup> Not determined. Not measurement possible for open burning experiments or for short experiments like brisk fire.<sup>d</sup> Determined using a temperature probe located inside the smoke extraction duct at the close field location (after dilution).<sup>e</sup> Dilution factors determined using air flow ratios, CO or CO<sub>2</sub> concentration ratios measured at the emission and after dilution. Calculations using Equation (S1). Results obtained were all in good agreement.



**Table 2**

Average emission factors  $\pm$  standard deviation (n = 4 or 5, see Table 1) of particle-bound species obtained for garden green waste burning (hedge trimming and fallen leaves). Results from samplings and measurements performed after dilution (close field).

Species	Hedge trimming	Fallen leaves
<b>Particulate matter mass (g kg<sup>-1</sup> fuel burnt, dry-mass basis)</b>		
Total PM	33.4 $\pm$ 5.7	34.7 $\pm$ 9.2
Non-volatile PM (solid fraction)	25.6 $\pm$ 5.7	17.8 $\pm$ 5.1
<b>Carbonaceous fraction (gC kg<sup>-1</sup> fuel burnt, dry-mass basis)</b>		
TC	21.8 $\pm$ 9.6	24.7 $\pm$ 9.7
OC	- <sup>a</sup>	24.5 $\pm$ 9.7
EC	-	0.2 $\pm$ 0.1
OC/EC	-	120.4 $\pm$ 50.9
<b>Chemical speciation (mg kg<sup>-1</sup> fuel burnt, dry-mass basis)</b>		
<i>Monosaccharide anhydrides</i>		
Levogucosan	1340.8 $\pm$ 150.1	1624.0 $\pm$ 802.1
Mannosan	233.6 $\pm$ 18.2	180.1 $\pm$ 70.1
Galactosan	177.1 $\pm$ 21.3	235.6 $\pm$ 96.7
<i>Alkanes</i>		
C <sub>11</sub>	< LD	0.9 $\pm$ 0.3
C <sub>12</sub>	< LD	0.2 $\pm$ 0.1
C <sub>13</sub>	< LD	< LD
C <sub>14</sub>	9.3 $\pm$ 6.4	0.6 $\pm$ 0.2
C <sub>15</sub>	0.3 $\pm$ 0.1	< LD
C <sub>16</sub>	2.1 $\pm$ 1.2	1.6 $\pm$ 0.6
C <sub>17</sub>	< LD	1.8 $\pm$ 0.6
C <sub>18</sub>	0.9 $\pm$ 0.0	2.4 $\pm$ 0.6
C <sub>19</sub>	2.1 $\pm$ 0.4	3.6 $\pm$ 1.0
C <sub>20</sub>	1.7 $\pm$ 0.4	4.0 $\pm$ 1.1
C <sub>21</sub>	2.8 $\pm$ 0.6	5.0 $\pm$ 1.5
C <sub>22</sub>	3.5 $\pm$ 0.9	6.6 $\pm$ 2.8
C <sub>23</sub>	4.2 $\pm$ 1.7	9.6 $\pm$ 4.6
C <sub>24</sub>	3.4 $\pm$ 1.7	9.9 $\pm$ 4.5
C <sub>25</sub>	4.7 $\pm$ 1.6	24.8 $\pm$ 14.7
C <sub>26</sub>	3.4 $\pm$ 1.8	7.4 $\pm$ 4.9
C <sub>27</sub>	13.4 $\pm$ 6.0	113.2 $\pm$ 80.0
C <sub>28</sub>	5.1 $\pm$ 2.7	10.0 $\pm$ 7.2
C <sub>29</sub>	15.1 $\pm$ 10.7	38.5 $\pm$ 28.7
C <sub>30</sub>	1.6 $\pm$ 1.2	4.2 $\pm$ 3.4
C <sub>31</sub>	3.5 $\pm$ 4.6	3.2 $\pm$ 2.8
C <sub>32</sub>	0.6 $\pm$ 0.1	0.7 $\pm$ 0.7
C <sub>33</sub>	< LD	0.2 $\pm$ 0.2
C <sub>34</sub>	0.4 $\pm$ 0.0	0.2 $\pm$ 0.0
C <sub>35</sub>	< LD	< LD
C <sub>36</sub>	0.1 $\pm$ 0.0	< LD
C <sub>37</sub>	< LD	< LD
C <sub>38</sub>	< LD	< LD
C <sub>39</sub>	0.1 $\pm$ 0.0	2.7 $\pm$ 1.9
C <sub>40</sub>	0.1 $\pm$ 0.0	0.1 $\pm$ 0.1
Pristane	4.4 $\pm$ 1.9	0.8 $\pm$ 0.8
Phytane	< LD	< LD
$\Sigma$ n-Alkanes <sup>b</sup>	82.8 $\pm$ 43.9	255.4 $\pm$ 163.2
<i>Polycyclic aromatic hydrocarbons (PAHs)</i>		
Phenanthrene	17.3 $\pm$ 7.3	29.3 $\pm$ 15.2
Anthracene	3.3 $\pm$ 1.7	0.3 $\pm$ 0.1
Fluoranthene	7.8 $\pm$ 3.1	2.0 $\pm$ 0.8
Pyrene	6.7 $\pm$ 2.7	2.1 $\pm$ 0.9
Triphenylene	9.5 $\pm$ 4.6	2.6 $\pm$ 1.1
Retene	10.9 $\pm$ 5.4	1.7 $\pm$ 0.8
Benzo[a]anthracene	4.4 $\pm$ 2.7	1.2 $\pm$ 0.6
Chrysene	2.7 $\pm$ 1	0.5 $\pm$ 0.3
Benzo[e]pyrene	2.0 $\pm$ 1.8	0.5 $\pm$ 0.2
Benzo[b]fluoranthene	1 $\pm$ 0.5	0.2 $\pm$ 0.1
Benzo[k]fluoranthene	0.7 $\pm$ 0.3	0.1 $\pm$ 0.1
Benzo[a]pyrene	1.5 $\pm$ 0.6	0.3 $\pm$ 0.2
Benzo[g,h,i]perylene	1 $\pm$ 0.5	0.2 $\pm$ 0.1
Dibenzo[a,h]anthracene	0.2 $\pm$ 0.0	0.1 $\pm$ 0.0
Indeno[1,2,3-cd]pyrene	0.6 $\pm$ 0.3	0.1 $\pm$ 0.0
Coronene	< LD	0.0 $\pm$ 0.0

**Table 2.** (continued)

Species	Hedge trimming	Fallen leaves
<i>Polycyclic aromatic hydrocarbons (PAHs) (continued)</i>		
1-Methylfluorene	0.1 ± 0.0	0.0 ± 0.0
3-Methylphenanthrene	2.0 ± 0.4	0.4 ± 0.3
2-Methylphenanthrene	3.1 ± 0.5	0.7 ± 0.5
2-Methylanthracene	1.6 ± 0.2	0.4 ± 0.3
4- + 9-Methylphenanthrene <sup>c</sup>	1.2 ± 0.2	0.3 ± 0.2
1-Methylphenanthrene	2.9 ± 0.5	0.5 ± 0.3
4-Methylpyrene	0.9 ± 0.1	0.3 ± 0.2
1-Methylpyrene	0.7 ± 0.1	0.3 ± 0.2
1- + 3-Methylfluoranthene <sup>c</sup>	0.6 ± 0.1	0.2 ± 0.1
2-Methylfluoranthene	0.7 ± 0.2	0.2 ± 0.1
2-Methylpyrene	0.4 ± 0.1	0.1 ± 0.1
3-Methylfluoranthene	0.9 ± 0.2	0.2 ± 0.1
3-Methylchrysene	0.4 ± 0.1	0.1 ± 0.0
Methylchrysene / Methylbenzo[a]anthracene <sup>c, d</sup>	0.2 ± 0.1	0.1 ± 0.0
∑PAH <sub>b</sub> <sup>e</sup>	29.1 ± 14.1	6.4 ± 3.2
∑ <sub>30</sub> PAHs <sup>f</sup>	85.3 ± 35.2	45.0 ± 22.9
<i>Methoxyphenols</i>		
Vanillin	29.7 ± 6.1	19.8 ± 12.3
Homovanillic acid	2.2 ± 0.3	1.3 ± 0.8
Acetovanillone	16.5 ± 5.4	14.1 ± 4.5
Guaiacylacetone	43.4 ± 8.1	33.3 ± 13.8
Coniferylaldehyde	15.6 ± 5.3	5.3 ± 5.5
Vanillic acid	23.5 ± 29.7	5.0 ± 4.8
Syringol	108.0 ± 28.0	36.9 ± 23.9
Methylsyringol	58.8 ± 18.3	60.8 ± 34.7
Acetosyringone	10.4 ± 1.1	13.3 ± 5.6
Syringylacetone	22.7 ± 3.1	28.5 ± 9.3
Sinapylaldehyde	5.4 ± 2.8	3.9 ± 4.7
Propenylsyringol	17.4 ± 2	21.2 ± 8.8
Syringic acid	4.7 ± 1.2	7.1 ± 4.1
∑Guaiacyl derivatives <sup>g</sup>	130.9 ± 54.9	78.6 ± 41.6
∑Syringyl derivatives <sup>h</sup>	171.5 ± 90.9	91.0 ± 90.9
∑Methoxyphenols <sup>i</sup>	358.3 ± 111.4	250.1 ± 132.6
Cholesterol	< LD	14.6 ± 10.0
<i>Polyols, simple sugars and sugar alcohols</i>		
Inositol	32.2 ± 11.1	4.6 ± 1.9
Glycerol	162.7 ± 36.5	217.1 ± 197.6
Erythriol	1469.6 ± 472.4	< LD
Xylitol	< LD	< LD
Arabitol	8.1 ± 0.7	6.1 ± 2.8
Sorbitol	297.4 ± 68.2	< LD
Mannitol	3.4 ± 4.8	8.2 ± 2.3
Threulose	< LD	< LD
Rhamnose	< LD	< LD
Glucose	7.8 ± 3.0	< LD
∑Polyols + sugars <sup>j</sup>	1987.4 ± 596.6	235.9 ± 204.6

<sup>a</sup> Distinction of EC and OC not possible for hedge trimming due to filter sample overloading.

<sup>b</sup> Sum of all *n*-alkanes from C<sub>11</sub> to C<sub>40</sub>.

<sup>c</sup> Not separated by chromatography and quantified as a single compound.

<sup>d</sup> Not identified (native standard compounds not available).

<sup>e</sup> Sum of PAHs and methyl-PAHs mainly associated to the particulate phase in close field or ambient air conditions (from retene to coronene and from 4-methylpyrene to methylchrysene / methylbenzo[a]anthracene).

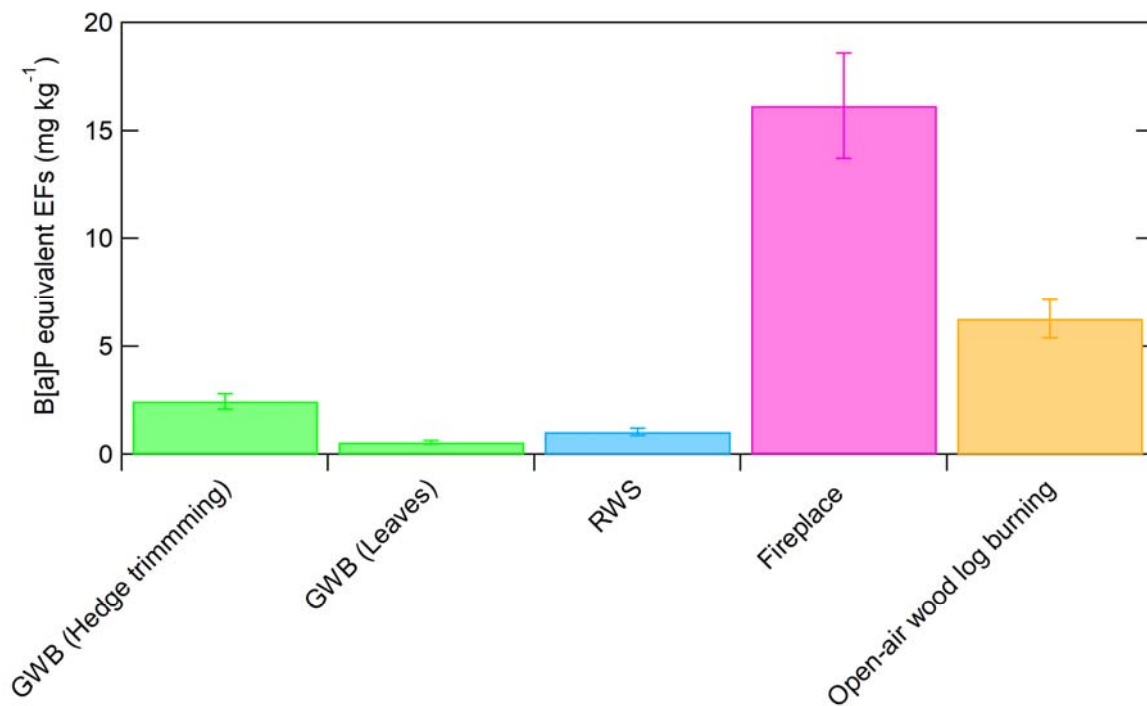
<sup>f</sup> Sum of all PAHs and methyl-PAHs.

<sup>g</sup> Sum of all guaiacyl derivatives: vanillin, homovanillic acid, acetovanillone, guaiacylacetone, coniferylaldehyde, vanillic acid.

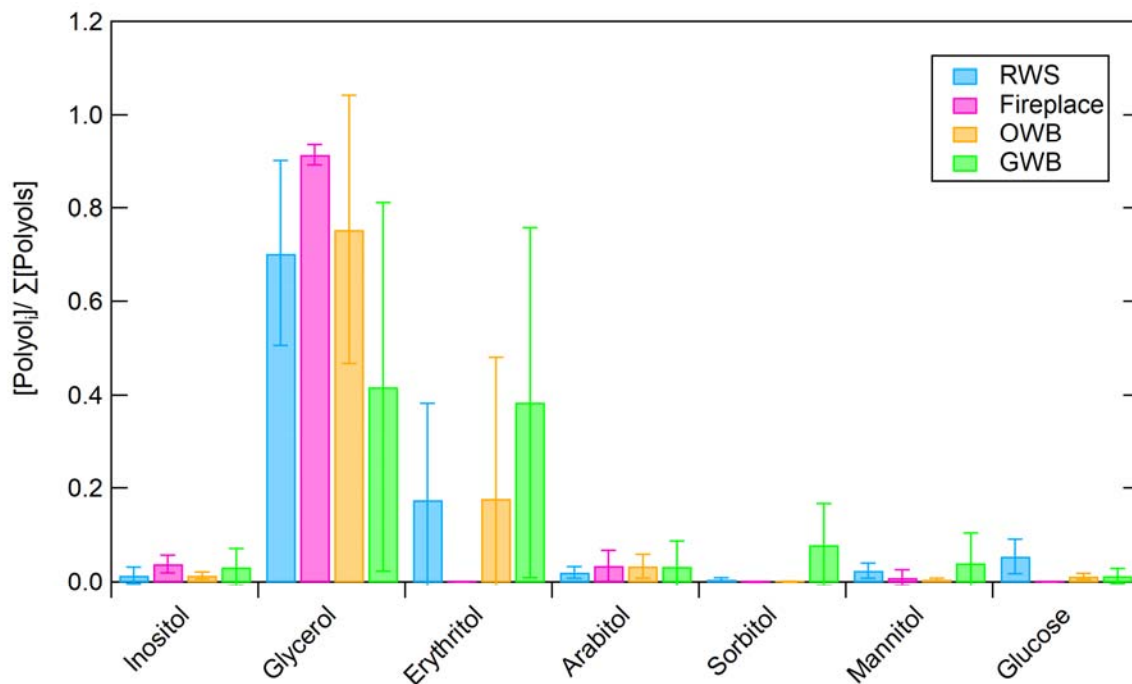
<sup>h</sup> Sum of all syringyl derivatives: syringol, methylsyringol, acetosyringone, syringylacetone, sinapylaldehyde, propenylsyringol, syringic acid

<sup>i</sup> Sum of all methoxyphenols.

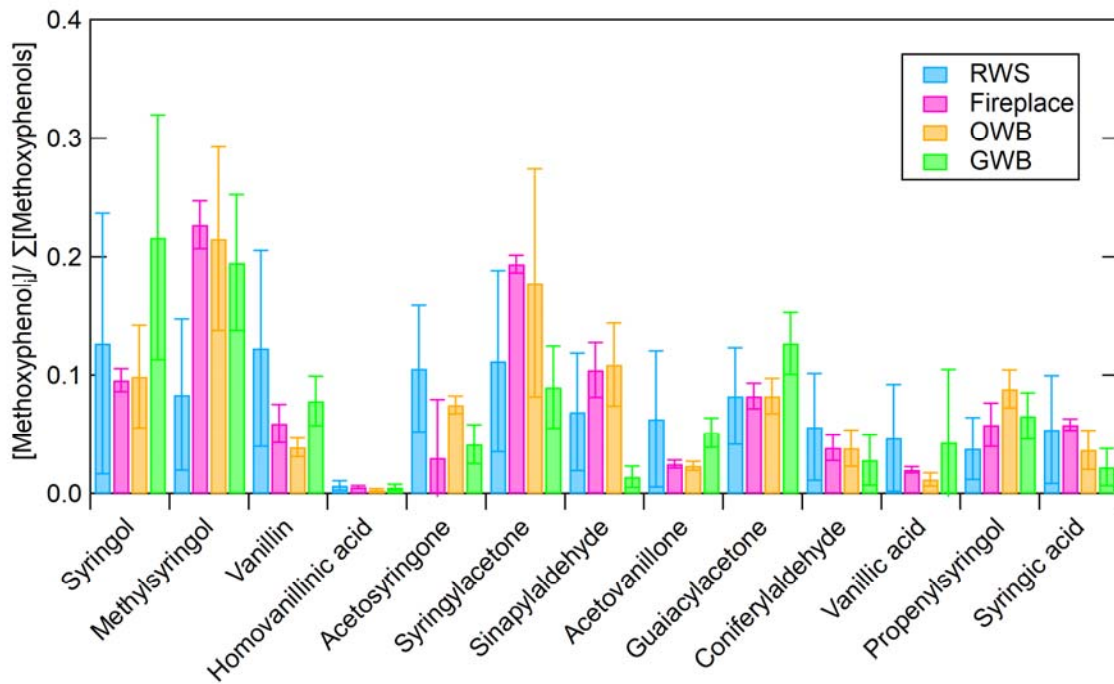
<sup>j</sup> Sum of all polyols, simple sugars and sugar alcohols.



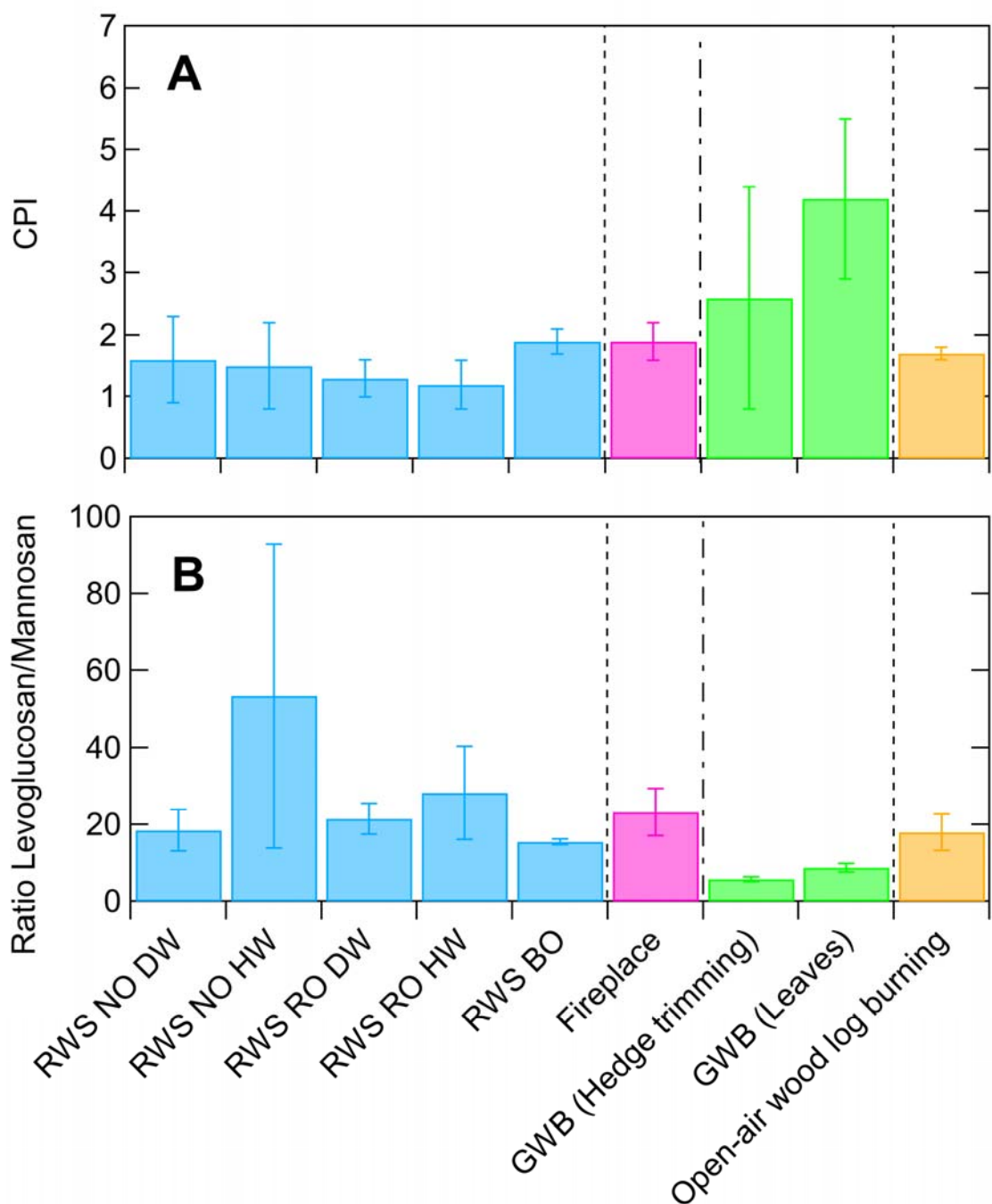
**Fig. 1.** Comparison of the B[a]P equivalent emission factors (EFs) obtained for the different biomass burning conditions tested (n= 2 to 5). RWS: residential wood stove in nominal (NO), reduced (RO) or brisk (BO) outputs using dry (DW) or humid wood (HW); OWB: open-air wood log burning; GWB: garden green waste burning. The error bars correspond to the standard deviation ( $\pm 2 \sigma$ ) for the total number of experiments performed.



**Fig. 2.** Polyol, sugar and sugar alcohol chemical profiles obtained for the different biomass burning conditions tested (n=4, 5 or 7). RWS: residential wood stove combining nominal and reduced outputs, dry and humid wood all together; OWB: open-air wood log burning; GWB: garden green waste burning with hedge trimming and fallen leaves combined. The error bars correspond to the standard deviation ( $\pm 2 \sigma$ ) for the total number of experiments performed.



**Fig. 3.** Methoxyphenol chemical profiles obtained for the different biomass burning conditions tested (n=4, 5 or 7). RWS: residential wood stove combining nominal and reduced outputs, dry and humid wood all together; OWB: open-air wood log burning; GWB: garden green waste burning with hedge trimming and fallen leaves combined. The error bars correspond to the standard deviation ( $\pm 2 \sigma$ ) for the total number of experiments performed.



**Fig. 4.** CPI (carbon preference index) of alkanes (A) and levoglucosan/mannosan ratios (B) obtained for the different biomass burning conditions tested (n=4, 5 or 7). RWS (residential wood stove) nominal (NO), reduced (RO) or brisk outputs (BO) using dry (DW) or humid wood (HW); open-air wood log burning; GWB: garden green waste burning with hedge trimming and fallen leaves combined. The error bars correspond to the standard deviation ( $\pm 2 \sigma$ ) for the total number of experiments performed.

# SUPPLEMENTARY MATERIAL

## Emission factors and chemical characterization of particulate emissions from garden green waste burning

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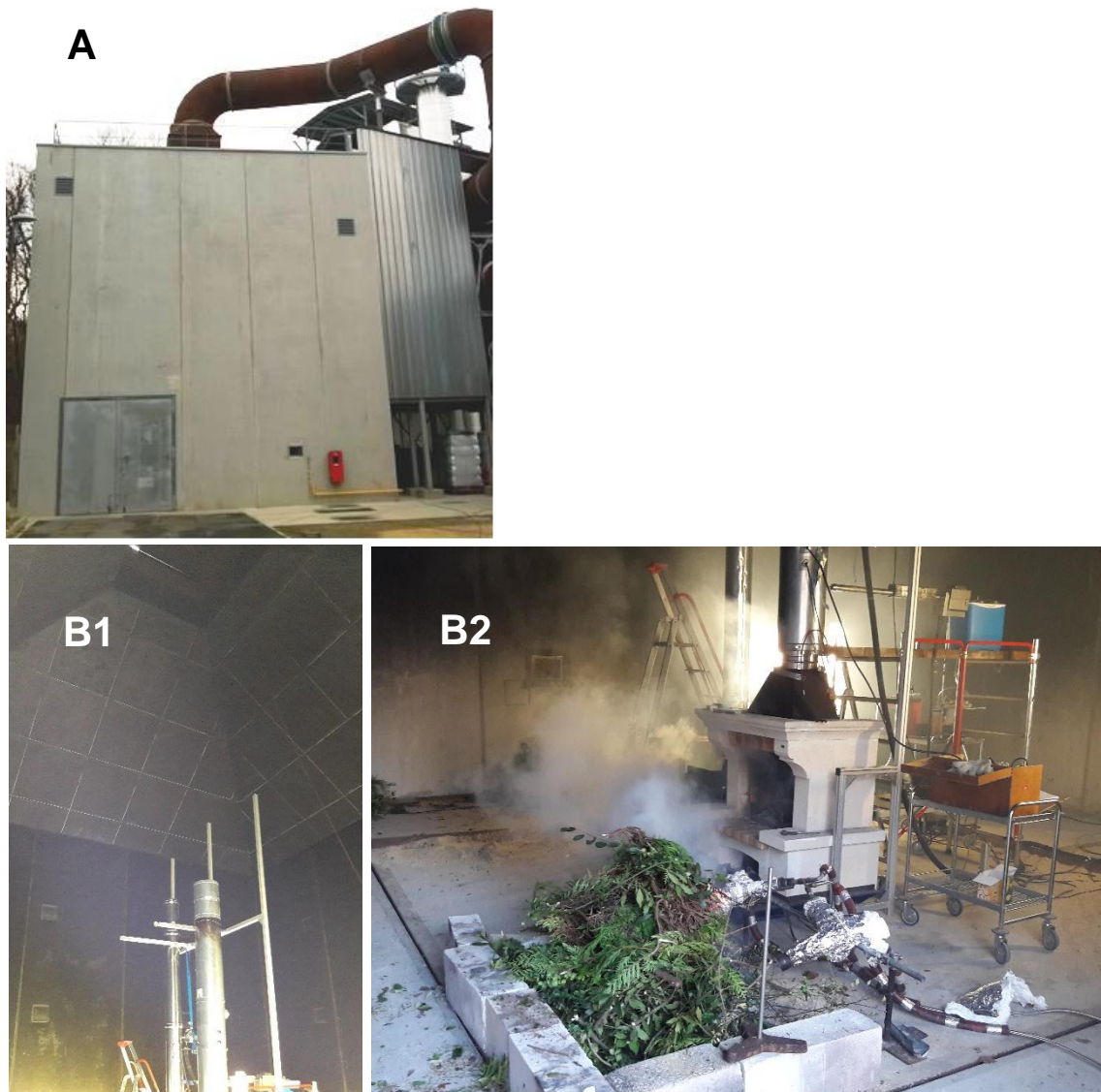
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## 1. Combustion chamber



**Fig. S1.** External view of the 1000 m<sup>3</sup> combustion chamber with its smoke extraction duct (A) and internal view showing the top of the combustion chamber (B1) and a garden green waste burning experiment (hedge trimming here) with also the residential heating appliances (fireplace in the front and RWS in the back) (B2).

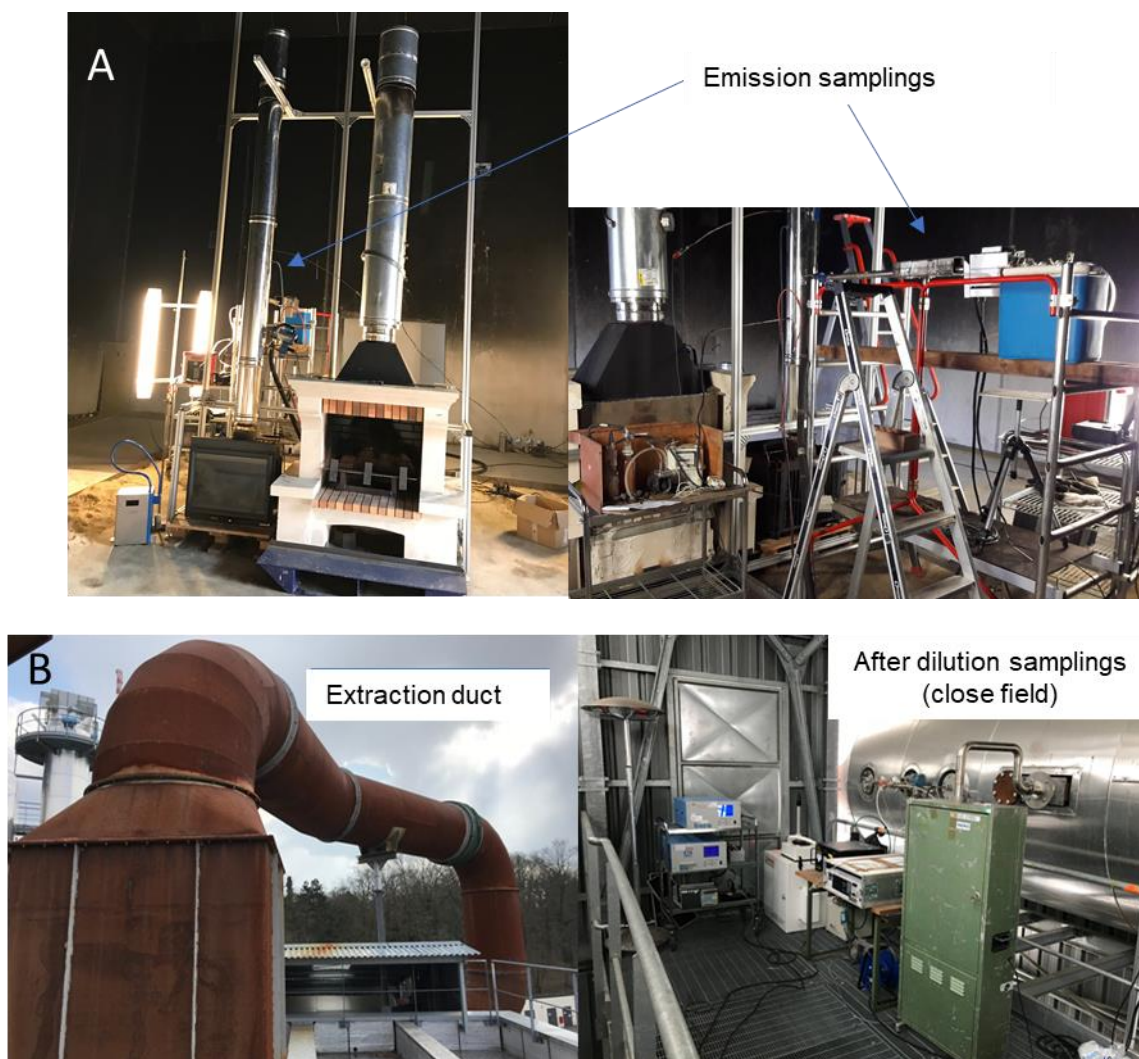


## 2. Residential heating appliances description

**Table S1.** Main characteristics of the residential heating appliances used in this study.

<b>4* RWS “Flamme verte” labelled</b>	<b>Characteristics</b>
Marketing year	1998
Power (kW)	10.5
Log dimension (cm)	50
Efficiency (%)	74.5
CO [% (v/v) at 13 % of O <sub>2</sub> (v/v)]	0.25
PM [mg Nm <sup>-3</sup> at 13 % of O <sub>2</sub> (v/v)]	60
<b>Fireplace</b>	
Material	Gard stones with an inner red clay bricks furnace
Dimensions (cm)	80 (width) × 60 (depth) × 60 (height)

### 3. Sampling and measurements locations, instrumentations and details



**Fig. S2.** Emission sampling locations (for the RWS and the fireplace) (A). Sampling location in the extraction duct after dilution (close field) of combustion smokes (B).

**Table S2.** Parameters measured at the emission (E) and in close field (CF) and corresponding reference methods.

Parameters (sampling location)	Method of measurement	References (sampling location)
<b>Continuous measurements (automatic analyzers)</b>		
CO <sub>2</sub> (E <sup>a</sup> , CF <sup>b</sup> )	Non-dispersive infrared	CEN/TS 17405
O <sub>2</sub> (E)	Paramagnetism	EN 14789
CO (E, CF)	Non-dispersive infrared	EN 15058 (E) EN 14626 (CF)
NO <sub>x</sub> (NO and NO <sub>2</sub> ) (E, CF)	Chemiluminescence	EN 14792 (E) EN 14211 (CF)
PM mass (non-volatile = solid fraction)	Micro-balance	EN 16450
Temperature (E, CF)	K-type thermocouple	-
Flow (gas velocity) (E, CF)	Mac Caffrey probe	-
<b>Manual measurements and chemical analyses</b>		
<b>Emission (E)</b>		
Humidity	Condensation and/or absorption	EN 14790
PM mass (solid and condensable fractions)	Gravimetry: weighting of filter samples (quartz, Pallflex, Tissuquartz, Ø = 75 mm) (solid fraction) + dry residue from isopropanol impinger samplings (in the temperature regulated bath at 2°C) (condensable fraction). 10 L min <sup>-1</sup> , isokinetic sampling conditions. Heated sampling probe and filter holder at 120 °C.	(PEREN2BOIS, 2012)
<b>Close field (CF, after dilution)</b>		
Total PM mass concentrations	Gravimetry: weighting of glass-teflon fiber filter samples (Pallflex, Emfab, Ø = 47 mm); Collection using a Partisol sampler (Thermo, Model 2025) at 20 L min <sup>-1</sup> )	EN 12341
PAHs and methyl-PAHs	Extraction by solvent using PLE. PAHs analyses by HPLC-Fluorescence. Methyl-PAHs quantified by GC-MS	(Golly et al., 2015; Nalin et al., 2016)
Alkanes	Extraction by PLE (same extraction as PAHs). Quantification by GC-MS	(Golly et al., 2015; Nalin et al., 2016)
Methoxyphenols, guaiacyl and syringyl derivatives, cholesterol	Extraction by PLE (same extraction as PAHs). Quantification by GC-MS after a derivatization step (silylation using BSTFA)	(Golly et al., 2015; Nalin et al., 2016)
Levoglucosan, mannosan, galactosan, polyols sugars and sugar alcohols	Extraction by sonication with ultra-pure water and analyses by IC-PAD	(Verlhac et al., 2013; Yttri et al., 2015)
EC/OC/TC	Analyses using thermal optical method (Sunset Laboratory, EUSSAR 2 protocol)	(Cavalli et al., 2010; EN 16909; Chiappini et al., 2014)

<sup>a</sup>E: emission.

<sup>b</sup>CF: close field.

CEN/TS 17405: Stationary source emissions - Determination of the volume concentration of carbon dioxide - Reference method: infrared spectrometry.

EN 12341: Ambient air - Standard gravimetric measurement method for the determination of the PM<sub>10</sub> or PM<sub>2.5</sub> mass concentration of suspended particulate matter.

EN 14211: Ambient air. Standard method for the measurement of the concentration of nitrogen dioxide and nitrogen monoxide by chemiluminescence EN 14626: Ambient air quality. Standard method for the measurement of the concentration of carbon monoxide by non-dispersive infrared spectroscopy.

EN 14789: Stationary source emissions. Determination of volume concentration of oxygen (O<sub>2</sub>).

EN 14790: Stationary source emissions. Determination of the water vapor in ducts Reference method. Paramagnetism.

EN 14792: Stationary source emissions. Determination of mass concentration of nitrogen oxides (NO<sub>x</sub>). Reference method: Chemiluminescence.

EN 15058: Stationary source emissions. Determination of the mass concentration of carbon monoxide (CO). Reference method: non-dispersive infrared spectrometry.

EN 16450: Ambient Air - automated measuring systems for the measurement of the concentration of particulate matter (PM<sub>10</sub>, PM<sub>2.5</sub>).

EN 16909 - Ambient air - Measurement of elemental carbon (EC) and organic carbon (OC) collected on filters.

**Table S3.** Instrumentation used for gaseous phase emission (E) measurements.

Parameters measured	Analyzer model
O <sub>2</sub>	Multigas analyzer PG250 (Horiba) Range: 0-25% volume
CO	Multigas analyzer PG250 (Horiba) Range: 0-1000 ppm
NO / NO <sub>x</sub>	Multigas analyzer PG250 (Horiba) Range: 0-100 ppm
CO <sub>2</sub>	VA 3000 (Horiba) Range: 0-5000 ppm

**Table S4.** Instrumentation used for gaseous phase close field (CF) measurements.

Parameters measured	Analyzer model
CO	Model 48i CO Analyzer (Thermo) Range: 0-10000 ppm
NO / NO <sub>x</sub>	Model 42i NO-NO <sub>2</sub> -NO <sub>x</sub> Analyzer (Thermo) Range: 0-100 ppm
CO <sub>2</sub>	VA 3000 (Horiba) Range: 0-500 ppm
PM (non-volatile = solid fraction)	TEOM-50 (Rupprecht & Patashnick) Range: 5 µg m <sup>-3</sup> - up to g m <sup>-3</sup>

#### 4. Determination of the dilution factors (DF) using air flow ratios

The determination of the smoke emission flow rates from domestic wood-burning appliances is based on the fuel characteristics and the amount of wood burnt during the test, taking into account the different phases of combustion (from fuel addition until the end of the combustion) (CEN (European Committee for Standardization), 2018).

$$DF = \frac{\Phi_{cc}}{\Phi_{v,s}} \quad \text{Equation (S1)}$$

With:

$\Phi_{cc}$ : combustion chamber air flow rate ( $\text{m}^3 \text{h}^{-1}$ ),

$\Phi_{v,s}$  (dry gas): smoke volumetric emission flow rate ( $\text{m}^3 \text{h}^{-1}$ ).

The smoke volumetric emission flow rate ( $\Phi_{v,s}$  dry gas,  $\text{m}^3 \text{h}^{-1}$ ) was calculated following (Equation (S2)):

$$\Phi_{v,s} \text{ (dry gas)} = \frac{\frac{\Phi_{m,s} \times 3600}{1000}}{\varphi_{m,s}} \times \frac{273}{273+T^\circ\text{C}} - \left( \frac{\frac{\Phi_{m,s} \times 3600}{1000}}{\varphi_{m,s}} \times \frac{273}{273+T^\circ\text{C}} \times \frac{\text{humidity}}{100} \right) \quad \text{Equation (S2)}$$

With:

$\Phi_{m,s}$ : smoke mass flow rate ( $\text{g s}^{-1}$ ),

$\varphi_{m,s}$ : smoke density ( $\text{kg m}^{-3}$ ).

The smoke mass flow rate ( $\Phi_{m,s}$ ,  $\text{g s}^{-1}$ ) and the smoke density ( $\varphi_{m,s}$ ,  $\text{kg m}^{-3}$ ) are calculated following Equations (S3) and (S4).

$$\Phi_{m,s} = \frac{M_h \times 1.3 \times \left( \frac{C_r - C_f}{0.536 \times (CO_2 + CO)} + 1.244 \times \frac{9H_f + W_f}{100} \right)}{3.6} \quad \text{Equation (S3)}$$

With:

$M_h$ : mass of fuel burnt per hour ( $\text{kg h}^{-1}$ ),

$C_f$ : fuel carbon content (raw fuel) (% of mass),

$C_r$ : residues carbon content (related to the mass of fuel burnt) (% of mass),

H<sub>r</sub>: fuel hydrogen content (raw fuel) (% of mass),

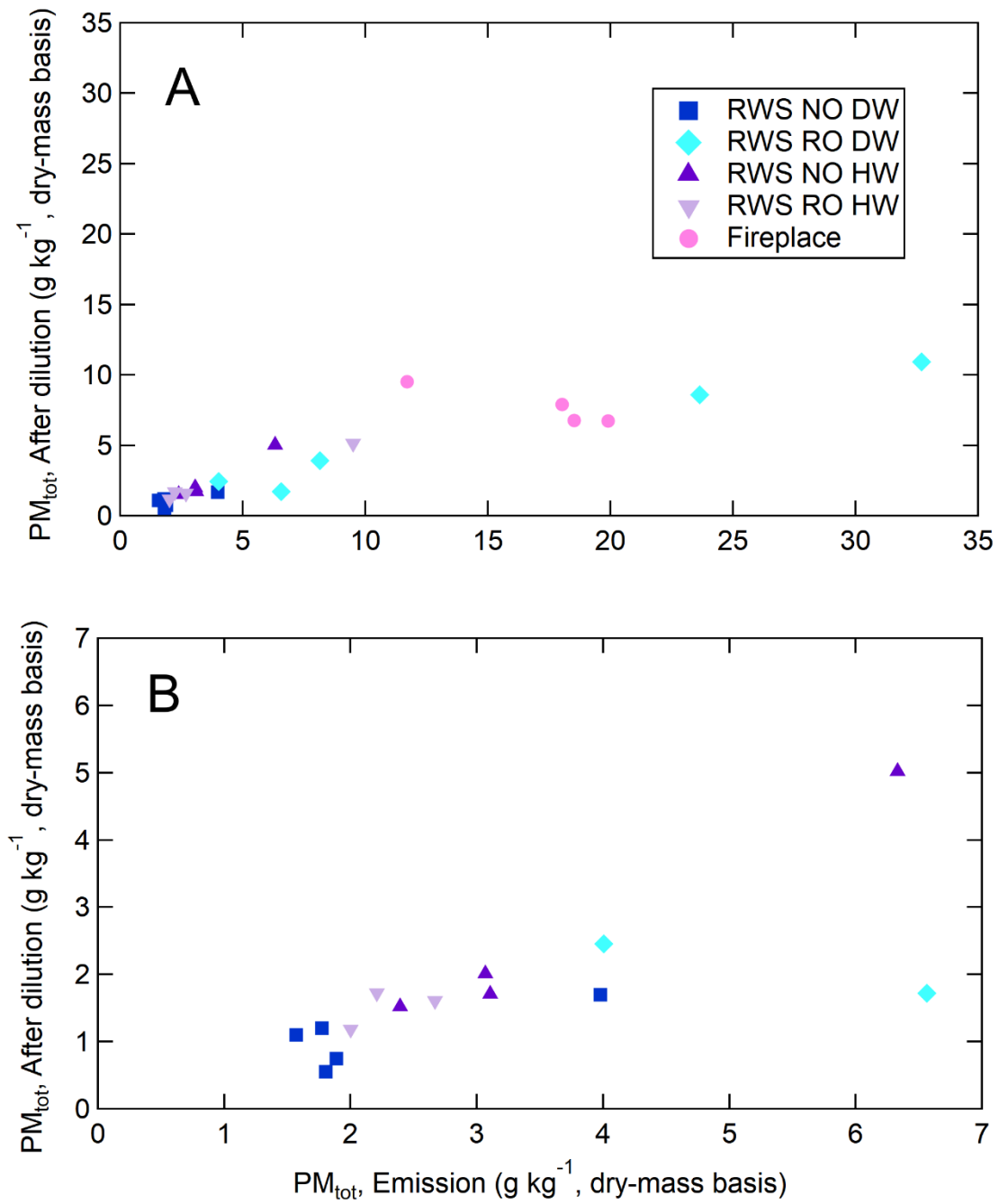
W<sub>r</sub>: fuel water content (raw fuel) (% of mass).

$$\Phi_{m,s} = Pressure \times \frac{0.000001 \times \left( 32 \times O_2 \times \left( 1 - \frac{humidity}{100} \right) \right) + \left( 44 \times CO_2 \times \left( 1 - \frac{humidity}{100} \right) \right) + (18 \times humidity) + 28 \times \left( 100 - O_2 \times \left( 1 - \frac{humidity}{100} \right) - CO_2 \times \left( 1 - \frac{humidity}{100} \right) - humidity \right)}{8.314 \times (273 + T^{\circ}C)}$$

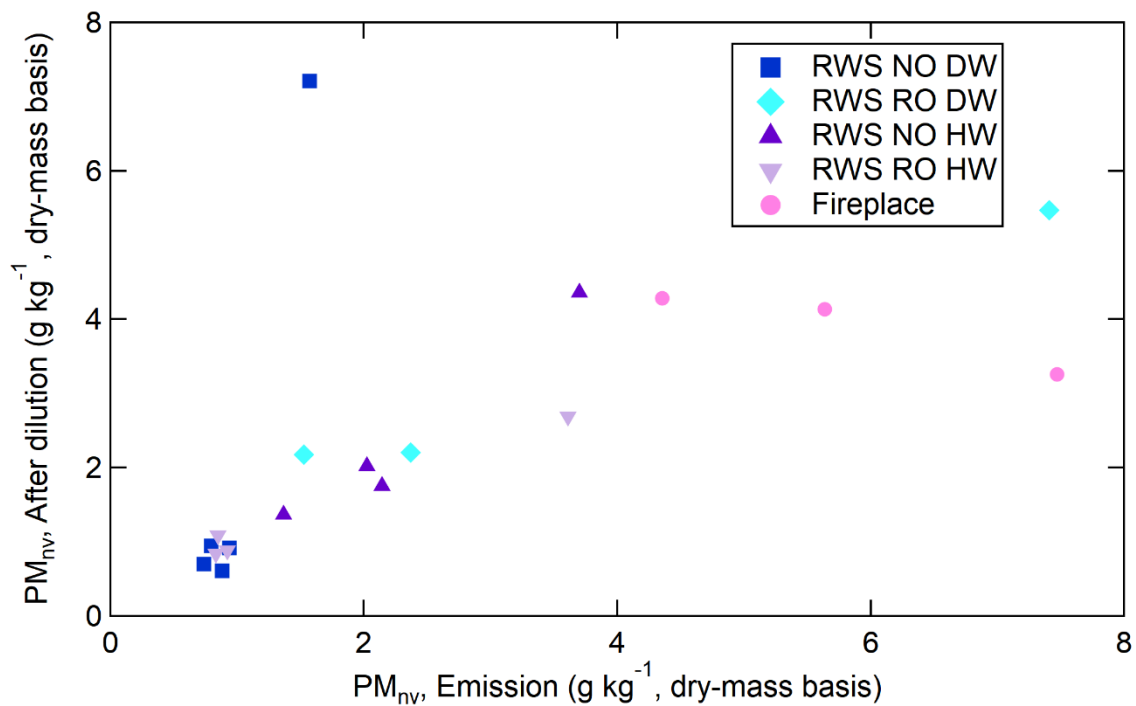
Equation (S4)

with pressure in Pa, O<sub>2</sub> and CO<sub>2</sub> concentrations in % (vol) of dry gas.

### 5. Comparison of the PM EFs obtained from emission and after dilution measurements

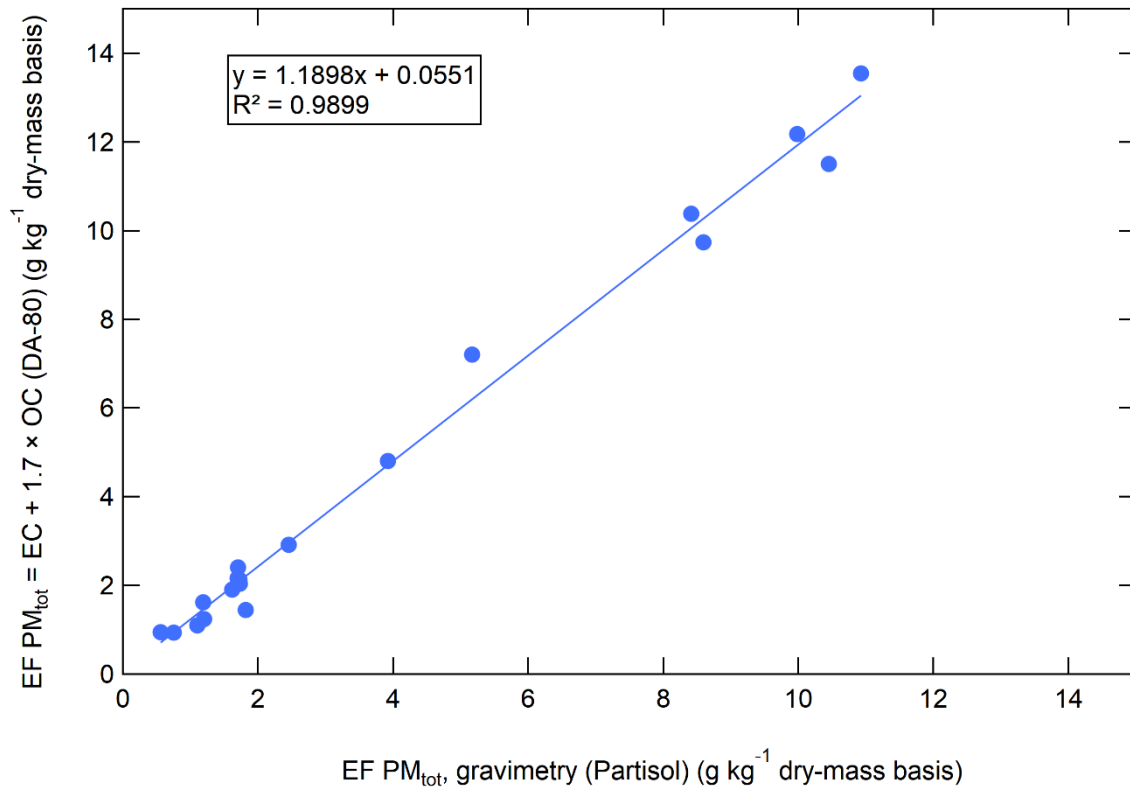


**Fig. S3.** Comparison of total PM emission factors (EFs) determined by gravimetry from emission and after dilution measurements (A). Focus on low EF values (B).



**Fig. S4.** Comparison of non-volatile (solid fraction) PM emission factors determined by gravimetry (filters only) at emission and from TEOM-50 measurements after dilution (close field). Outliers are highlighted by red circles.





**Fig. S5.** Comparison of total PM emission factors determined from measurements after dilution by gravimetric method (Partisol) and by reconstruction based on EC/OC measurements (DA-80) (all residential wood heating appliances and operating conditions combined).  $PM_{tot} = EC + 1.7 \times OC$ , conversion factor 1.7 between OC and OM for biomass combustion (Puxbaum et al., 2007).

## 6. Emission factors for RWS, fireplace and open-air wood log burning

**Table S5.** Average emission factors  $\pm$  standard deviation ( $n = 2$  to  $5$ , see Table 1) of particulate-bound species obtained for the residential wood stove in nominal, reduced and brisk output conditions and with both wood log moistures tested (15 and 25%). Results from samplings and measurements performed after dilution (close field).

Species	RWS NO DW	RWS RO DW	RWS NO HW	RWS RO HW	RWH Brisk
<b>Particulate matter mass (<math>g\ kg^{-1}</math> fuel burnt, dry-mass basis)</b>					
Total PM	$1.1 \pm 0.4$	$5.5 \pm 4.0$	$2.6 \pm 1.7$	$2.4 \pm 1.8$	$2.4 \pm 1.8$
Non-volatile PM (solid fraction)	$1.1 \pm 2.9$	$3.9 \pm 1.9$	$2.4 \pm 1.4$	$1.4 \pm 0.9$	$1.6 \pm 0.0$
<b>Carbonaceous fraction (<math>gC\ kg^{-1}</math> fuel burnt, dry-mass basis)</b>					
TC	$0.9 \pm 0.3$	$4.0 \pm 2.9$	$2.5 \pm 1.5$	$2.0 \pm 1.6$	$1.2 \pm 0.3$
OC	$0.6 \pm 0.3$	$3.8 \pm 2.9$	$0.2 \pm 0.4$	$1.8 \pm 1.6$	$0.3 \pm 0.4$
EC	$0.3 \pm 0.1$	$0.2 \pm 0.1$	$0.2 \pm 0.4$	$0.2 \pm 0.1$	$0.2 \pm 0.3$
OC/EC	$2.0 \pm 1.2$	$20.1 \pm 17.6$	$1.0 \pm 0.4$	$13.1 \pm 11.0$	$1.5 \pm 1.0$
<b>Chemical speciation (<math>mg\ kg^{-1}</math> fuel burnt, dry-mass basis)</b>					
<i>Monosaccharide anhydrides</i>					
Levoglucosan	$40.7 \pm 36.7$	$726.1 \pm 590.4$	$17.5 \pm 10.1$	$298.1 \pm 347.9$	$6.7 \pm 0.7$
Mannosan	$2.3 \pm 2.2$	$33.6 \pm 27.3$	$0.7 \pm 0.0$	$12.4 \pm 14.7$	< LD
Galactosan	$1.8 \pm 1.5$	$22.1 \pm 16.9$	$0.7 \pm 0.1$	$10.3 \pm 12.7$	$0.5 \pm 0.1$
<i>Alkanes</i>					
C <sub>11</sub>	$1.0 \pm 0.5$	$7.5 \pm 14.8$	$1.1 \pm 1.1$	$1.8 \pm 1.3$	< LD
C <sub>12</sub>	< LD	$2.6 \pm 0.0$	< LD	$0.6 \pm 0.0$	< LD
C <sub>13</sub>	$0.3 \pm 0.1$	$5.1 \pm 4.8$	$0.5 \pm 0.1$	$1.2 \pm 1.1$	< LD
C <sub>14</sub>	$0.3 \pm 0.1$	$2.0 \pm 2.7$	$0.3 \pm 0.2$	$0.2 \pm 0.1$	$0.2 \pm 0.1$
C <sub>15</sub>	$0.1 \pm 0.1$	$0.5 \pm 0.8$	$0.2 \pm 0.2$	$0.7 \pm 0.5$	$0.1 \pm 0.0$
C <sub>16</sub>	$0.2 \pm 0.2$	$2.0 \pm 2.7$	$0.4 \pm 0.2$	$0.8 \pm 1.2$	$0.2 \pm 0.0$
C <sub>17</sub>	$0.2 \pm 0.1$	$2.0 \pm 3.6$	$0.3 \pm 0.3$	$0.2 \pm 0.1$	$0.2 \pm 0.0$
C <sub>18</sub>	$0.3 \pm 0.2$	$1.1 \pm 1.7$	$0.4 \pm 0.2$	$0.3 \pm 0.1$	$0.4 \pm 0.2$
C <sub>19</sub>	$0.2 \pm 0.1$	$0.7 \pm 1.1$	$0.2 \pm 0.1$	$0.1 \pm 0.0$	$0.2 \pm 0.0$
C <sub>20</sub>	$0.1 \pm 0.0$	$0.5 \pm 0.7$	$0.2 \pm 0.2$	$0.1 \pm 0.0$	$0.1 \pm 0.1$
C <sub>21</sub>	$0.2 \pm 0.1$	$0.5 \pm 0.5$	$0.1 \pm 0.1$	$0.1 \pm 0.1$	$0.1 \pm 0.1$
C <sub>22</sub>	$0.2 \pm 0.1$	$0.6 \pm 0.5$	$0.1 \pm 0.1$	$0.1 \pm 0.0$	$0.1 \pm 0.1$
C <sub>23</sub>	$0.3 \pm 0.1$	$0.4 \pm 0.2$	$0.3 \pm 0.1$	$0.2 \pm 0.1$	$0.2 \pm 0.1$
C <sub>24</sub>	$0.6 \pm 0.2$	$0.5 \pm 0.2$	$0.8 \pm 0.2$	$0.3 \pm 0.2$	$0.5 \pm 0.3$
C <sub>25</sub>	$1.0 \pm 0.3$	$0.6 \pm 0.2$	$1.3 \pm 0.3$	$0.4 \pm 0.1$	$0.8 \pm 0.6$
C <sub>26</sub>	$1.1 \pm 0.4$	$0.5 \pm 0.1$	$1.3 \pm 0.2$	$0.4 \pm 0.2$	$0.7 \pm 0.4$
C <sub>27</sub>	$1.5 \pm 0.6$	$0.9 \pm 0.5$	$2.6 \pm 1.6$	$0.5 \pm 0.1$	$1.1 \pm 0.6$
C <sub>28</sub>	$0.9 \pm 0.3$	$0.3 \pm 0.1$	$1.1 \pm 0.3$	$0.3 \pm 0.2$	$0.5 \pm 0.4$
C <sub>29</sub>	$0.9 \pm 0.4$	$0.6 \pm 0.5$	$1.4 \pm 0.9$	$0.3 \pm 0.1$	$0.6 \pm 0.5$
C <sub>30</sub>	$0.4 \pm 0.2$	$0.1 \pm 0.0$	$0.5 \pm 0.2$	$0.2 \pm 0.0$	$0.6 \pm 0.0$
C <sub>31</sub>	$0.5 \pm 0.2$	$0.1 \pm 0.1$	$0.5 \pm 0.4$	$0.2 \pm 0.1$	$0.5 \pm 0.0$
C <sub>32</sub>	$0.3 \pm 0.2$	< LD	$0.2 \pm 0.2$	< LD	< LD
C <sub>33</sub>	$0.2 \pm 0.1$	$0.0 \pm 0.0$	$0.3 \pm 0.3$	< LD	< LD
C <sub>34</sub>	< LD	$0.1 \pm 0.0$	$0.1 \pm 0.0$	< LD	< LD
C <sub>35</sub>	< LD	$0.0 \pm 0.0$	< LD	< LD	< LD
C <sub>36</sub>	< LD	< LD	< LD	< LD	< LD
C <sub>37</sub>	< LD	< LD	< LD	< LD	< LD
C <sub>38</sub>	$0.2 \pm 0.1$	$0.3 \pm 0.7$	$0.1 \pm 0.0$	$0.0 \pm 0.0$	< LD
C <sub>39</sub>	$0.3 \pm 0.4$	$0.3 \pm 0.3$	$0.1 \pm 0.1$	$0.0 \pm 0.0$	$0.5 \pm 0.0$
C <sub>40</sub>	< LD	< LD	< LD	< LD	$0.1 \pm 0.0$
Pristane	$0.2 \pm 0.1$	$0.1 \pm 0.1$	$0.1 \pm 0.1$	$1.0 \pm 0.0$	$0.1 \pm 0.0$
Phytane	$0.1 \pm 0.1$	$0.3 \pm 0.3$	$0.1 \pm 0.1$	$0.0 \pm 0.0$	< LD
$\Sigma n$ -Alkanes <sup>a</sup>	$11.3 \pm 5.0$	$29.8 \pm 36.8$	$14.7 \pm 7.4$	$8.9 \pm 5.7$	$7.7 \pm 3.4$
<i>Polycyclic aromatic hydrocarbons (PAHs)</i>					
Phenanthrene	$0.2 \pm 0.1$	$7.6 \pm 9.2$	$4.2 \pm 0.6$	$6.7 \pm 4.9$	$0.2 \pm 0.0$
Anthracene	$0.0 \pm 0.0$	$0.1 \pm 0.1$	$0.5 \pm 0.6$	$0.1 \pm 0.0$	$0.0 \pm 0.0$
Fluoranthene	$0.4 \pm 0.5$	$3.4 \pm 1.7$	$5.0 \pm 2.2$	$2.5 \pm 0.7$	$0.5 \pm 0.6$
Pyrene	$0.4 \pm 0.5$	$5.0 \pm 2.9$	$4.8 \pm 1.4$	$2.4 \pm 0.7$	$0.1 \pm 0.0$
Triphenylene	$0.9 \pm 0.7$	$2.6 \pm 1.0$	$4.6 \pm 0.9$	$1.9 \pm 0.5$	$3.7 \pm 5.1$
Retene	$0.8 \pm 0.5$	$3.0 \pm 2.1$	$2.8 \pm 1.2$	$0.6 \pm 0.1$	$3.4 \pm 4.7$
Benzo[a]anthracene	$0.3 \pm 0.2$	$2.4 \pm 1.4$	$1.3 \pm 0.6$	$0.6 \pm 0.3$	$1.6 \pm 2.2$
Chrysene	$0.2 \pm 0.2$	$0.8 \pm 0.5$	$1.2 \pm 0.1$	$0.3 \pm 0.1$	$1.5 \pm 2.1$
Benzo[e]pyrene	$0.2 \pm 0.2$	$0.6 \pm 0.5$	$0.9 \pm 0.2$	$0.2 \pm 0.1$	$0.8 \pm 1.1$
Benzo[b]fluoranthene	$0.2 \pm 0.1$	$0.4 \pm 0.1$	$1.5 \pm 0.7$	$0.2 \pm 0.1$	$0.9 \pm 1.3$
Benzo[k]fluoranthene	$0.1 \pm 0.1$	$0.4 \pm 0.2$	$0.8 \pm 0.4$	$0.2 \pm 0.1$	$0.5 \pm 0.7$
Benzo[a]pyrene	$0.2 \pm 0.1$	$1.1 \pm 0.6$	$1.3 \pm 0.1$	$0.4 \pm 0.2$	$1.2 \pm 1.7$
Benzo[g,h,i]perylene	$0.2 \pm 0.1$	$0.7 \pm 0.4$	$1.5 \pm 1.0$	$0.2 \pm 0.1$	$1.0 \pm 1.4$

**Tableau S5.** (continued)

Species	RWS NO DW	RWS RO DW	RWS NO HW	RWS RO HW	RWH Brisk
<i>Polycyclic aromatic hydrocarbons (PAHs) (continued)</i>					
Dibenzo[a,h]anthracene	0.0 ± 0.0	0.2 ± 0.2	0.0 ± 0.0	0.0 ± 0.0	0.1 ± 0.1
Indeno[1,2,3-cd]pyrene	0.1 ± 0.1	0.5 ± 0.2	1.4 ± 1.1	0.2 ± 0.1	0.7 ± 0.9
Coronene	0.0 ± 0.0	0.2 ± 0.2	0.7 ± 1.0	0.0 ± 0.0	0.5 ± 0.0
1-Methylfluorene	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	< LD
3-Methylphenanthrene	0.0 ± 0.0	0.3 ± 0.5	0.4 ± 0.4	0.0 ± 0.0	< LD
2-Methylphenanthrene	0.0 ± 0.0	0.4 ± 0.6	0.5 ± 0.4	0.1 ± 0.0	< LD
2-Methylanthracene	0.0 ± 0.0	0.3 ± 0.4	0.1 ± 0.2	0.0 ± 0.0	< LD
4- + 9-Methylphenanthrene <sup>b</sup>	0.0 ± 0.0	0.3 ± 0.4	0.3 ± 0.2	0.0 ± 0.0	< LD
1-Methylphenanthrene	0.0 ± 0.0	0.5 ± 0.6	0.6 ± 0.5	0.1 ± 0.1	< LD
4-Methylpyrene	0.0 ± 0.0	0.4 ± 0.2	0.5 ± 0.7	0.1 ± 0.0	< LD
1-Methylpyrene	0.4 ± 0.1	0.3 ± 0.1	0.4 ± 0.6	0.1 ± 0.0	< LD
1- + 3-Methylfluoranthene <sup>b</sup>	0.0 ± 0.0	0.2 ± 0.1	0.3 ± 0.4	0.0 ± 0.0	0.1 ± 0.1
2-Methylfluoranthene	0.0 ± 0.0	0.3 ± 0.2	0.3 ± 0.6	0.0 ± 0.0	< LD
2-Methylpyrene	0.0 ± 0.0	0.1 ± 0.1	0.2 ± 0.3	0.0 ± 0.0	< LD
3-Methylfluoranthene	0.0 ± 0.0	0.3 ± 0.2	0.3 ± 0.5	0.1 ± 0.1	< LD
3-Methylchrysene	0.0 ± 0.0	0.1 ± 0.0	0.2 ± 0.3	0.0 ± 0.0	< LD
Methylchrysene / Methylbenzo[a]anthracene <sup>b,c</sup>	0.0 ± 0.0	0.0 ± 0.0	0.1 ± 0.1	0.0 ± 0.0	< LD
∑PAH <sub>16</sub> <sup>d</sup>	2.5 ± 1.6	12.1 ± 7.2	15.7 ± 9.9	3.3 ± 1.6	13.7 ± 16.4
∑ <sub>30</sub> PAHs <sup>e</sup>	4.5 ± 3.4	32.7 ± 24.8	36.7 ± 17.3	17.0 ± 8.6	18.3 ± 22.2
<i>Methoxyphenols</i>					
Vanillin	0.9 ± 0.8	7.0 ± 10.3	0.3 ± 0.1	10.8 ± 13.4	0.2 ± 0.0
Homovanillic acid	0.1 ± 0.0	0.9 ± 0.9	< LD	0.6 ± 0.8	< LD
Acetovanillone	0.3 ± 0.2	5.3 ± 5.5	0.2 ± 0.2	3.5 ± 5.3	< LD
Guaiacylacetone	0.8 ± 0.4	20.9 ± 24.1	0.3 ± 0.1	0.3 ± 0.1	0.1 ± 0.0
Coniferylaldehyde	0.7 ± 0.0	12.8 ± 15.3	0.2 ± 0.1	5.0 ± 8.7	0.2 ± 0.0
Vanillic acid	0.3 ± 0.2	3.4 ± 3.1	0.2 ± 0.1	2.6 ± 3.7	0.1 ± 0.0
Syringol	1.7 ± 2.2	11.0 ± 20.9	0.4 ± 0.1	35.8 ± 58.3	0.0 ± 0.0
Methylsyringol	1.2 ± 1.6	19.8 ± 30.6	0.2 ± 0.1	40.8 ± 77.1	0.1 ± 0.0
Acetosyringone	0.6 ± 0.5	20.3 ± 21.3	0.2 ± 0.0	8.5 ± 14.0	0.1 ± 0.0
Syringylacetone	0.8 ± 0.8	38.5 ± 38.2	0.2 ± 0.1	20.8 ± 36.1	0.1 ± 0.0
Sinapylaldehyde	0.7 ± 0.0	27.0 ± 28.8	0.2 ± 0.0	12.6 ± 23.2	< LD
Propenylsyringol	0.4 ± 0.5	16.7 ± 19.6	0.2 ± 0.0	6.2 ± 10.6	< LD
Syringic acid	0.5 ± 0.4	11.0 ± 11.0	0.2 ± 0.1	6.1 ± 10.8	< LD
∑Guaiacyl derivatives <sup>f</sup>	3.2 ± 1.5	50.2 ± 59.2	1.2 ± 0.6	30.1 ± 44.2	0.6 ± 0.0
∑Syringyl derivatives <sup>g</sup>	6.9 ± 6.0	144.5 ± 170.5	1.7 ± 0.0	130.7 ± 230.1	0.3 ± 0.0
∑Methoxyphenols <sup>h</sup>	10.4 ± 7.6	194.7 ± 229.6	3.1 ± 0.0	160.8 ± 274.3	1.3 ± 0.0
Cholesterol	0.3 ± 0.0	< LD	0.3 ± 0.0	< LD	0.3 ± 0.0
<i>Polyols, simple sugars and sugar alcohols</i>					
Inositol	< LD	2.3 ± 2.3	< LD	2.3 ± 2.2	< LD
Glycerol	34.3 ± 2.9	57.0 ± 16.6	< LD	52.1 ± 2.5	< LD
Erythriol	< LD	233.6 ± 183.8	< LD	< LD	< LD
Xylitol	< LD	< LD	< LD	< LD	< LD
Arabitol	0.7 ± 0.0	2.1 ± 1.3	< LD	1.0 ± 0.0	< LD
Sorbitol	< LD	< LD	< LD	< LD	< LD
Mannitol	< LD	< LD	< LD	< LD	< LD
Threulose	< LD	< LD	< LD	< LD	< LD
Rhamnose	< LD	< LD	< LD	< LD	< LD
Glucose	< LD	< LD	< LD	< LD	< LD
∑Polyols + sugars <sup>i</sup>	35.0 ± 2.9	296.1 ± 203.9	< LD	55.4 ± 4.6	< LD

<sup>a</sup> Sum of all *n*-alkanes from C<sub>11</sub> to C<sub>40</sub>.

<sup>b</sup> Not separated by chromatography and quantified as a single compound.

<sup>c</sup> Not identified (native standard compounds not available).

<sup>d</sup> Sum of PAHs and methyl-PAHs mainly associated to the particulate phase in close field or ambient air conditions (from retene to coronene and from 4-methylpyrene to methylchrysene / methylbenzo[a]anthracene).

<sup>e</sup> Sum of all PAHs and methyl-PAHs.

<sup>f</sup> Sum of all guaiacyl derivatives: vanillin, homovanillic acid, acetovanillone, guaiacylacetone, coniferylaldehyde, vanillic acid.

<sup>g</sup> Sum of all syringyl derivatives: syringol, methylsyringol, acetosyringone, syringylacetone, sinapylaldehyde, propenylsyringol, syringic acid

<sup>h</sup> Sum of all methoxyphenols.

<sup>i</sup> Sum of all polyols, simple sugars and sugar alcohols.

**Table S6.** Average emission factors  $\pm$  standard deviation ( $n = 3$  to  $4$ , see Table 1) of particulate-bound species obtained for the fireplace and open-air wood log burning. Results from samplings and measurements performed after dilution (close field).

Species	Fireplace	Open-air wood log burning
<b>Particulate matter mass (<math>g\ kg^{-1}</math> fuel burnt, dry-mass basis)</b>		
Total PM	$7.7 \pm 1.3$	$13.3 \pm 2.2$
Non-volatile PM (solid fraction)	$3.9 \pm 0.6$	$5.5 \pm 1.3$
<b>Carbonaceous fraction (<math>gC\ kg^{-1}</math> fuel burnt, dry-mass basis)</b>		
TC	$5.6 \pm 0.7$	$9.0 \pm 1.2$
OC	$5.8 \pm 0.8$	$8.6 \pm 1.2$
EC	$0.3 \pm 0.1$	$0.3 \pm 0.1$
OC/EC	$17.1 \pm 2.8$	$26.9 \pm 6.7$
<b>Chemical speciation (<math>mg\ kg^{-1}</math> fuel burnt, dry-mass basis)</b>		
<i>Monosaccharide anhydrides</i>		
Levoglucosan	$754.8 \pm 71.6$	$1600.0 \pm 523.0$
Mannosan	$50.0 \pm 17.5$	$90.5 \pm 22.0$
Galactosan	$38.0 \pm 7.0$	$58.3 \pm 13.1$
<i>Alkanes</i>		
C <sub>11</sub>	$0.1 \pm 0.1$	$0.1 \pm 0.1$
C <sub>12</sub>	$0.2 \pm 0.0$	$0.2 \pm 0.0$
C <sub>13</sub>	< LD	< LD
C <sub>14</sub>	< LD	< LD
C <sub>15</sub>	< LD	$0.8 \pm 0.4$
C <sub>16</sub>	$2.4 \pm 1.4$	$4.5 \pm 3.3$
C <sub>17</sub>	$0.5 \pm 0.6$	$0.7 \pm 0.4$
C <sub>18</sub>	$0.2 \pm 0.1$	$0.1 \pm 0.1$
C <sub>19</sub>	$0.1 \pm 0.0$	$0.3 \pm 0.1$
C <sub>20</sub>	$0.6 \pm 0.2$	$0.3 \pm 0.1$
C <sub>21</sub>	$0.6 \pm 0.1$	$0.5 \pm 0.1$
C <sub>22</sub>	$0.6 \pm 0.2$	$0.6 \pm 0.2$
C <sub>23</sub>	$0.7 \pm 0.2$	$0.8 \pm 0.1$
C <sub>24</sub>	$0.9 \pm 0.1$	$1.0 \pm 0.2$
C <sub>25</sub>	$1.2 \pm 0.2$	$1.1 \pm 0.1$
C <sub>26</sub>	$0.7 \pm 0.1$	$0.7 \pm 0.2$
C <sub>27</sub>	$2.8 \pm 0.5$	$2.0 \pm 0.4$
C <sub>28</sub>	$0.4 \pm 0.1$	$0.5 \pm 0.2$
C <sub>29</sub>	$0.8 \pm 0.3$	$1.0 \pm 0.5$
C <sub>30</sub>	$0.1 \pm 0.1$	$0.1 \pm 0.5$
C <sub>31</sub>	$0.3 \pm 0.2$	$0.5 \pm 0.4$
C <sub>32</sub>	$0.2 \pm 0.1$	$0.3 \pm 0.0$
C <sub>33</sub>	< LD	$0.2 \pm 0.1$
C <sub>34</sub>	$0.1 \pm 0.0$	$0.0 \pm 0.0$
C <sub>35</sub>	$0.1 \pm 0.0$	< LD
C <sub>36</sub>	$0.1 \pm 0.1$	$0.3 \pm 0.1$
C <sub>37</sub>	< LD	$0.1 \pm 0.0$
C <sub>38</sub>	< LD	< LD
C <sub>39</sub>	$0.1 \pm 0.1$	$0.1 \pm 0.0$
C <sub>40</sub>	$0.03 \pm 0.0$	$0.0 \pm 0.0$
Pristane	$0.8 \pm 0$	$0.2 \pm 0.2$
Phytane	$0.1 \pm 0.0$	$0.1 \pm 0.0$
$\Sigma n$ -Alkanes <sup>a</sup>	$13.7 \pm 2.7$	$15.7 \pm 5.5$
<b>Polycyclic aromatic hydrocarbons (PAHs)</b>		
Phenanthrene	$11.1 \pm 1.3$	$16.6 \pm 1.1$
Anthracene	$0.4 \pm 0.0$	$0.9 \pm 0.2$
Fluoranthene	$7.5 \pm 5.8$	$11.4 \pm 4.9$
Pyrene	$18.7 \pm 2.9$	$15.9 \pm 2.7$
Triphenylene	$17.4 \pm 2.7$	$17.6 \pm 3.9$
Retene	$225.5 \pm 63.8$	$125.4 \pm 36.9$
Benzo[a]anthracene	$18.9 \pm 5.7$	$8.9 \pm 2.0$
Chrysene	$11.7 \pm 3.7$	$5.4 \pm 1.6$
Benzo[e]pyrene	< LD	< LD
Benzo[b]fluoranthene	$3.4 \pm 3.4$	$2.2 \pm 1.3$
Benzo[k]fluoranthene	$3.2 \pm 0.9$	$1.2 \pm 0.3$
Benzo[a]pyrene	$10.6 \pm 3.4$	$3.8 \pm 0.8$
Benzo[g,h,i]perylene	$5.0 \pm 3.7$	$2.7 \pm 0.7$
Dibenzo[a,h]anthracene	$2.1 \pm 1.2$	$0.9 \pm 0.3$
Indeno[1,2,3-cd]pyrene	$5.0 \pm 2.2$	$1.5 \pm 0.8$
Coronene	$1.7 \pm 1.2$	$0.1 \pm 0.0$

**Table S6.** (continued)

Species	Fireplace	Open-air wood log burning
<i>Polycyclic aromatic hydrocarbons (PAHs) (continued)</i>		
1-Methylfluorene	0.0 ± 0.0	0.0 ± 0.0
3-Methylphenanthrene	0.0 ± 0.0	0.1 ± 0.0
2-Methylphenanthrene	0.0 ± 0.0	0.1 ± 0.0
2-Methylanthracene	0.0 ± 0.0	0.1 ± 0.0
4- + 9-Methylphenanthrene <sup>b</sup>	0.0 ± 0.0	0.1 ± 0.0
1-Methylphenanthrene	0.0 ± 0.0	0.1 ± 0.0
4-Methylpyrene	0.3 ± 0.1	0.3 ± 0.0
1-Methylpyrene	0.4 ± 0.1	0.3 ± 0.0
1- + 3-Methylfluoranthene <sup>b</sup>	0.2 ± 0.1	0.2 ± 0.0
2-Methylfluoranthene	0.3 ± 0.1	0.2 ± 0.0
2-Methylpyrene	0.1 ± 0.0	0.1 ± 0.0
3-Methylfluoranthene	0.3 ± 0.1	0.3 ± 0.0
3-Methylchrysene	0.1 ± 0.0	0.1 ± 0.0
Methylchrysene / Methylbenzo[a]anthracene <sup>b,c</sup>	0.1 ± 0.0	0.1 ± 0.0
∑PAH <sub>p</sub> <sup>d</sup>	288.9 ± 89.7	153.7 ± 44.7
∑ <sub>30</sub> PAHs <sup>e</sup>	344.0 ± 102.4	216.6 ± 57.5
<i>Methoxyphenols</i>		
Vanillin	8.3 ± 1.0	34.0 ± 26.4
Homovanillic acid	< LD	2.0 ± 0.5
Acetovanillone	9.3 ± 3.7	20.3 ± 14.8
Guaiacylacetone	43.0 ± 16.4	71.0 ± 52.3
Coniferylaldehyde	14.0 ± 8.7	27.1 ± 5.8
Vanillic acid	5.0 ± 3.7	8.7 ± 3.1
Syringol	17.8 ± 12.8	81.1 ± 52.3
Methylsyringol	49.6 ± 37.6	175.3 ± 104.8
Acetosyringone	33.3 ± 20.8	59.4 ± 28.7
Syringylacetone	125.7 ± 99.5	151.2 ± 131.4
Sinapylaldehyde	27.9 ± 19.0	77.1 ± 10.4
Propenylsyringol	53.9 ± 25.6	75.7 ± 54.4
Syringic acid	6.0 ± 8.8	25.1 ± 3.9
∑Guaiacyl derivatives <sup>f</sup>	79.0 ± 33.5	163.0 ± 103.0
∑Syringyl derivatives <sup>g</sup>	308.2 ± 215.3	619.8 ± 381.9
∑Methoxyphenols <sup>h</sup>	393.7 ± 257.7	808.1 ± 488.8
Cholesterol	< LD	< LD
<i>Polyols, simple sugars and sugar alcohols</i>		
Inositol	0.6 ± 5.4	3.1 ± 1
Glycerol	62.4 ± 0.0	130.1 ± 2.7
Erythriol	144.4 ± 17.6	171.7 ± 83.9
Xylitol	< LD	< LD
Arabitol	3.0	4.8
Sorbitol	< LD	< LD
Mannitol	<LD	<LD
Threalose	<LD	<LD
Rhamnose	<LD	<LD
Glucose	<LD	<LD
∑Polyols + sugars <sup>i</sup>	210.4 ± 80.1	309.7 ± 188.7

<sup>a</sup> Sum of all *n*-alkanes from C<sub>11</sub> to C<sub>40</sub>.

<sup>b</sup> Not separated by chromatography and quantified as a single compound.

<sup>c</sup> Not identified (native standard compounds not available).

<sup>d</sup> Sum of PAHs and methyl-PAHs mainly associated to the particulate phase in close field or ambient air conditions (from retene to coronene and from 4-methylpyrene to methylchrysene / methylbenzo[a]anthracene).

<sup>e</sup> Sum of all PAHs and methyl-PAHs.

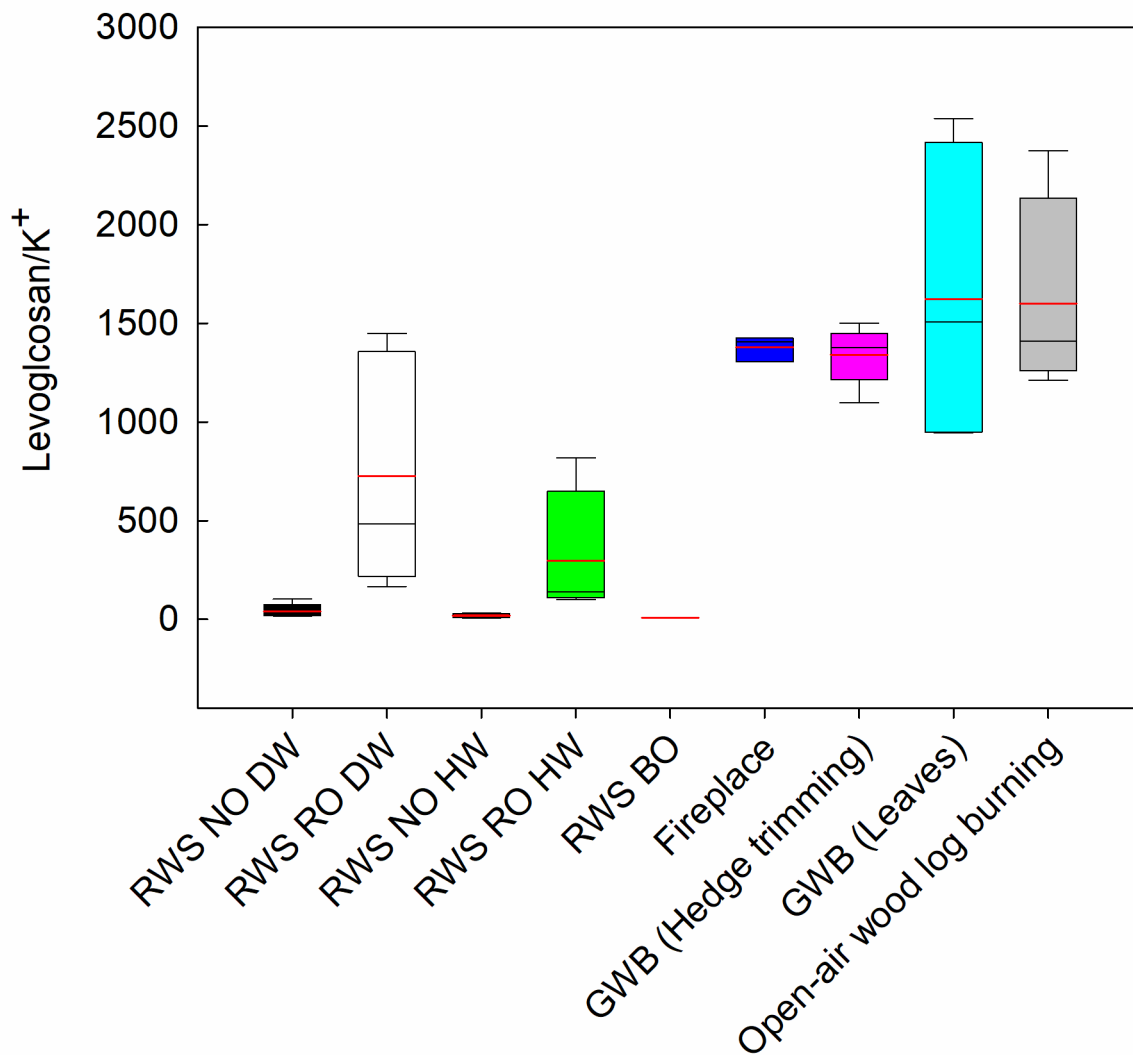
<sup>f</sup> Sum of all guaiacyl derivatives: vanillin, homovanillic acid, acetovanillone, guaiacylacetone, coniferylaldehyde, vanillic acid.

<sup>g</sup> Sum of all syringyl derivatives: syringol, methylsyringol, acetosyringone, syringylacetone, sinapylaldehyde, propenylsyringol, syringic acid

<sup>h</sup> Sum of all methoxyphenols.

<sup>i</sup> Sum of all polyols, simple sugars and sugar alcohols.

## 7. Ratio levoglucosan to potassium



**Fig. S6.** Ratios levoglucosan/ $K^+$  obtained for the different biomass burning conditions tested ( $n=4, 5$  or  $7$ ). RWS (residential wood stove) nominal (NO), reduced (RO) or brisk outputs (BO) using dry (DW) or humid wood (HW); open-air wood log burning; GWB: garden green waste burning with hedge trimming or fallen leaves. The boxplots are built using the 15<sup>th</sup>, 25<sup>th</sup>, 50<sup>th</sup>, 75<sup>th</sup> and 85<sup>th</sup> percentiles as respectively the lower whisker, the bottom, middle and top of the box and the upper whisker. The reds lines represent the average values.

## 8. Emission factors from the literature

**Table S7.** Emission factors ( $\text{g kg}^{-1}$  of raw fuel burnt) for open-air burning of different biomasses reported in the literature for total PM ( $\text{PM}_{2.5}$  in most cases).

Biomass burnt	$\text{PM}_{\text{tot}}$	References
Fallen leaves	10.8 - 13.0	(Collet, 2011; Hays et al., 2002)
Green waste	1.0 - 32.3	(Andreae, 2019; Collet, 2011; Hays et al., 2002; Kannan et al., 2005; Lutes and Kariher, 1996; Wardoyo, 2007; Wiedinmyer et al., 2014)
Forest wildfires	5.0 - 55.0	(Akagi et al., 2011; Alves et al., 2011, 2010; Andreae, 2019; Janhäll et al., 2010)
Tree pruning	8.8 - 16.9	(Alves et al., 2019; Jenkins et al., 1996a, b)
Agricultural and crop residues (corn, rice, wheat, sugar cane...)	2.2 - 19.5	(Akagi et al., 2011; Alves et al., 2019; Andreae, 2019; Chen et al., 2017; Hays et al., 2005; Jenkins et al., 1996a, b; Lemieux et al., 2004; Li et al., 2007; Wiedinmyer et al., 2014)
Stubble burning, grassland, savanna, pasture	2.2 - 18.0	(Akagi et al., 2011; Andreae, 2019; Christian et al., 2003; Dhammapala et al., 2007; Iinuma et al., 2007; Janhäll et al., 2010; Lemieux et al., 2004; Rennie et al., 2020; Wiedinmyer et al., 2014)

**Table S8.** Emission factors ( $\text{g eq.C kg}^{-1}$ , raw fuel burnt) for open-air burning of different biomasses reported in the literature for EC, OC and OC/EC.

Biomass burnt	EC	OC	OC/EC	References
Green waste	1.4 - 6.5	5.3 - 10.8	1.7 - 10.2	(Andreae, 2019; Schmidl et al., 2008)
Forest wildfires	0.1 - 1.0	1.0 - 30.0	4.4 - 90.0	(Akagi et al., 2011; Alves et al., 2011; Andreae, 2019)
Tree pruning, prescribed fires	0.3 - 1.2	2.7 - 7.4	5.2 - 77.0	(Alves et al., 2019; Robinson et al., 2011; Zhang et al., 2013; Dambruoso et al., 2014; Robinson et al., 2004; Yan et al., 2008)
Agricultural and crop residues (corn, rice, wheat, sugar cane...)	0.1 - 2.2	1.0 - 12.0	9.8 - 10.0	(Akagi et al., 2011; Alves et al., 2019; Christian et al., 2003; Oros et al., 2006; Oros and Simoneit, 1999; Wiedinmyer et al., 2014)
Stubble burning, grassland, savanna, pasture	0.4 - 1.4	1 - 3.4	0.7 - 93	(Akagi et al., 2011; Alves et al., 2019; Andreae, 2019; Christian et al., 2003; Iinuma et al., 2007; Oros et al., 2006; Rennie et al., 2020; Wiedinmyer et al., 2014)

**Table S9.** Emission factors (g kg<sup>-1</sup>, raw fuel burnt) for open-air burning of different biomasses reported in the literature for PAHs.

Biomass burnt	$\sum_8$ PAH <sup>a</sup>	References
Leaves	6.3	(Collet, 2011)
Green waste	2.9 - 6.3	(Lutes and Kariher, 1996)
Forest fires	8.7 - 15.0	(Akagi et al., 2011; Andreae, 2019; Collet and Fianni, 2006)
Tree pruning	0.7 - 3.5	(Alves et al., 2019; Collet, 2011; Jenkins et al., 1996a, b; Keshtkar and Ashbaugh, 2007)
Agricultural and crop residues (corn, rice, wheat, sugar cane...)	1.0 - 35.0	(Akagi et al., 2011; Chen et al., 2017; Jenkins et al., 1996b, a; Lemieux et al., 2004; Oros et al., 2006; Shen et al., 2011, 2013; Zhang et al., 2011)
Stubble burning, grassland, savanna, pasture	3.6 - 12.0	(Akagi et al., 2011; Andreae, 2019; Dhammapala et al., 2007; Iinuma et al., 2007; Lemieux et al., 2004; Oros et al., 2006, 2006; Venkataraman et al., 2002)

<sup>a</sup> Sum of the 8 PAHs commonly quantified: fluoranthene, benzo[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenzo[a,h]anthracene, indeno[1,2,3-cd]pyrene, benzo[g,h,i]perylene.

**Table S10.** Emission factors (mg kg<sup>-1</sup>, raw) for open-air burning of different biomasses reported in the literature for levoglucosan.

Biomass burnt	Levoglucosan	References
Green waste	400 - 2150	(Andreae, 2019; Medeiros and Simoneit, 2008; Schmidl et al., 2008)
Forest wildfires	420 - 2540	(Akagi et al., 2011; Alves et al., 2011; Andreae, 2019)
Tree pruning	50 - 215	(Alves et al., 2019)
Agricultural and crop residues (corn, rice, wheat, sugar cane...)	400 - 1200	(Akagi et al., 2011; Alves et al., 2019; Christian et al., 2003; Oros et al., 2006; Oros and Simoneit, 1999; Wiedinmyer et al., 2014)
Stubble burning, grassland, savanna, pasture, peat	400 - 500	(Akagi et al., 2011; Alves et al., 2019; Andreae, 2019; Christian et al., 2003; Iinuma et al., 2007; Oros et al., 2006; Rennie et al., 2020; Wiedinmyer et al., 2014)

**Table S11.** Emission factors (mg kg<sup>-1</sup>, raw) for open-air burning of different biomasses reported in the literature for n-alkanes.

Biomass burnt	$\sum_{C_{21} - C_{31}}$ alkanes	References
Green waste	23 - 116	(Medeiros and Simoneit, 2008)
Leaves	15 - 180	(Hays et al., 2002)
Agricultural and crop residues (rice, wheat)	42 - 58	(Hays et al., 2005)
Stubble burning, peat, grassland, savanna	1 - 688	(Iinuma et al., 2007; Oros et al., 2006)

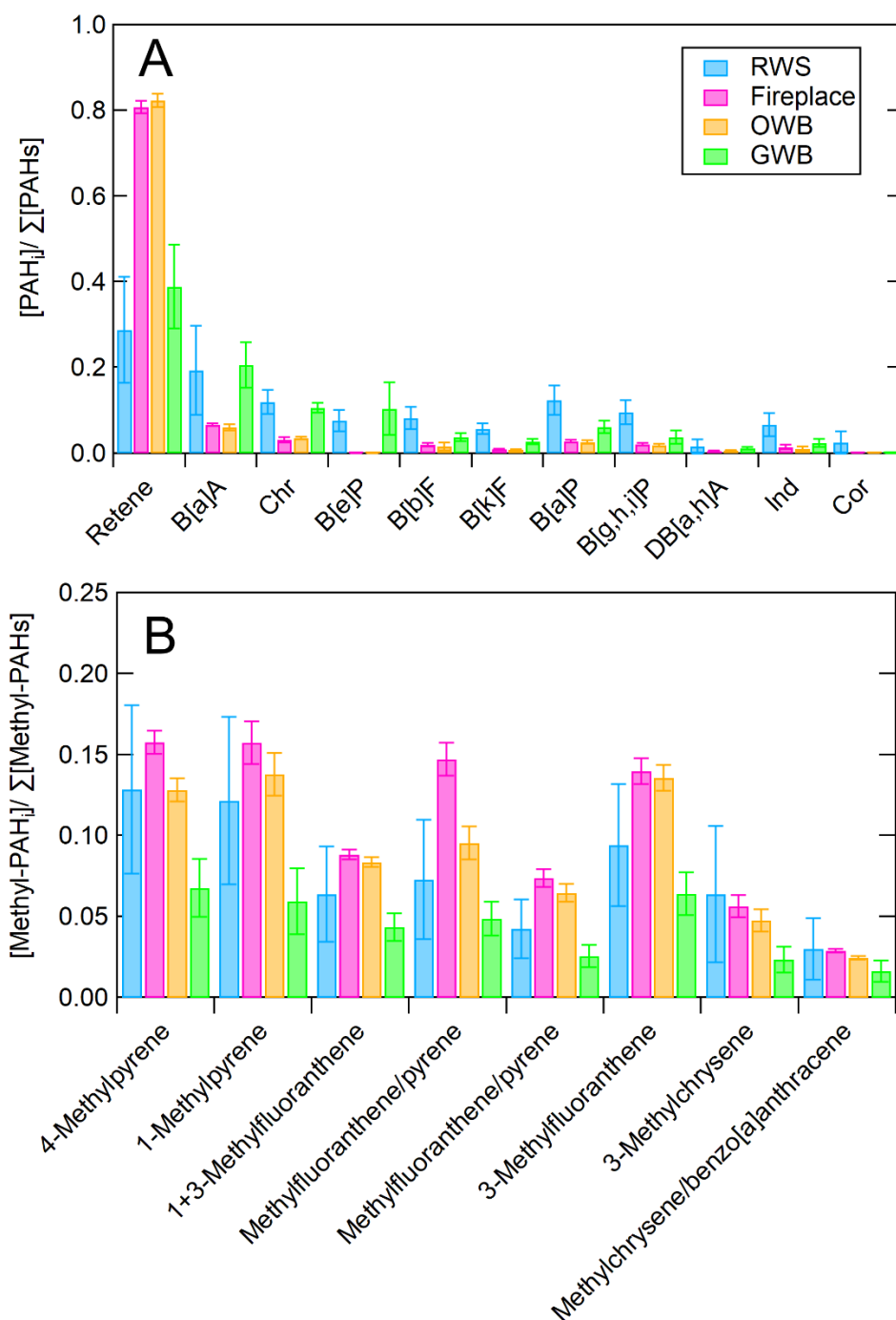


## 9. PAH toxic equivalent factors (TEF) values

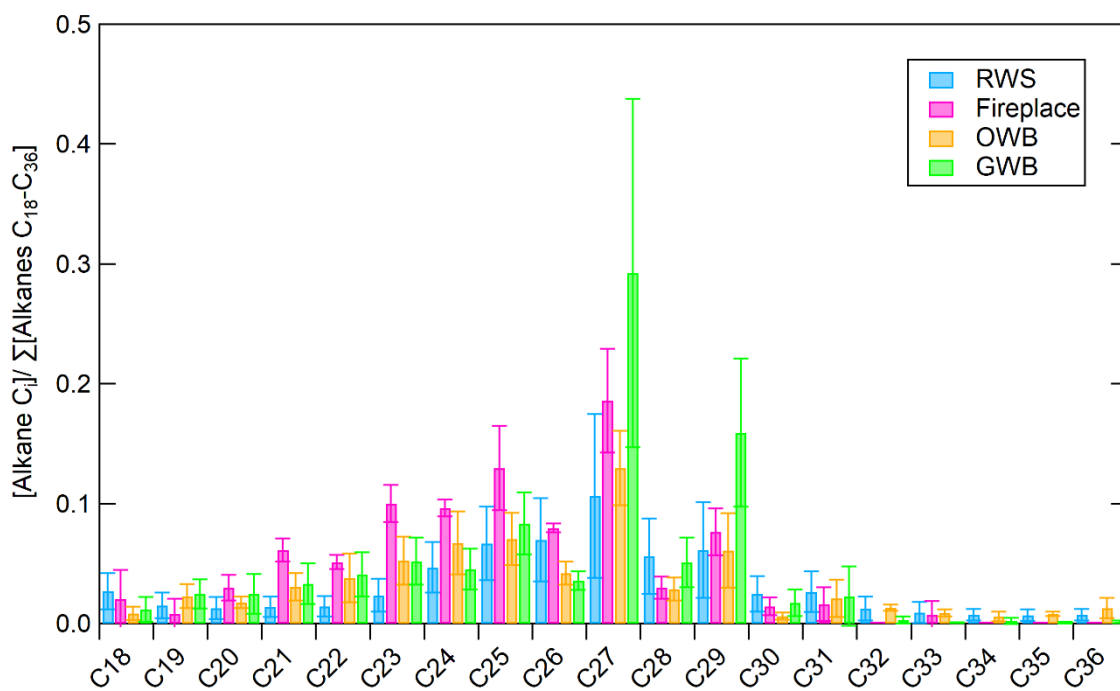
**Table S12.** Toxic equivalent factors (TEF) used for B[a]P equivalent concentration calculations.

PAHs	TEF	References
Retene	0.001	(Samburova et al., 2017)
Benz[a]anthracene	0.1	(Doornaert and Pichard, 2005)
Chrysene	0.01	(Doornaert and Pichard, 2005)
Benzo[e]pyrene	0.002	(OEHHA, 2011)
Benzo[b]fluoranthene	0.1	(OEHHA, 2011)
Benzo[k]fluoranthene	0.1	(Hester and Harrison, 1998)
Benzo[a]pyrene	1	(OEHHA, 2011)
Benzo[g,h,i]perylene	0.01	(Doornaert and Pichard, 2005)
Dibenzo[a,h]anthracene	1	(Doornaert and Pichard, 2005)
Indeno[1,2,3-cd]pyrene	0.1	(Doornaert and Pichard, 2005)
Coronene	0.001	(Doornaert and Pichard, 2005)
1-Methylphenanthrene	0.001	(Samburova et al., 2017)
4-Methylpyrene	0.001	(Samburova et al., 2017)
1-Methylpyrene	0.001	(Samburova et al., 2017)
1- + 3-Methylfluoranthene <sup>h</sup>	0.001	(Samburova et al., 2017)
2-Methylfluoranthene	0.001	(Samburova et al., 2017)
2-Methylpyrene	0.001	(Samburova et al., 2017)
3-Methylfluoranthene	0.001	(Samburova et al., 2017)
3-Methylchrysene	0.01	(Samburova et al., 2017)
Methylchrysene / Methylbenzo[a]anthracene	0.01	(Samburova et al., 2017)

## 10. Chemical profiles obtained for PAHs and n-alkanes.



**Fig. S7.** PAH (A) and methyl-PAH (B) chemical profiles obtained for the different biomass burning conditions tested ( $n=4, 5$  or  $7$ ). RWS: residential wood stove combining nominal and reduced outputs, dry and humid wood all together; OWB: open-air wood log burning; GWB: garden green waste burning with hedge trimming and fallen leaves combined. The error bars correspond to the standard deviation ( $\pm 2\sigma$ ) for the total number of experiments performed.



**Fig. S8.** n-Alkane chemical profiles obtained for the different biomass burning conditions tested (n=4, 5 or 7). RWS: residential wood stove combining nominal and reduced outputs, dry and humid wood all together; OWB: open-air wood log burning; GWB: garden green waste burning with hedge trimming and fallen leaves combined. The error bars correspond to the standard deviation ( $\pm 2 \sigma$ ) for the total number of experiments performed.

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