

Emission of Volatile Organic Compounds from the Residential Combustion of Pyrenean Oak and Black Poplar

M. Evtugina, C. A. Alves, A. I. Calvo, T. Nunes, L. Tarelho, M. Duarte, and S. O. Prozil

Abstract—Smoke from domestic wood burning has been identified as a major contributor to air pollution, motivating detailed emission measurements under controlled conditions. A series of experiments was performed to characterise the emissions from wood combustion in a fireplace and in a woodstove of two common species of trees grown in Spain: Pyrenean oak (*Quercus pyrenaica*) and black poplar (*Populus nigra*). Volatile organic compounds (VOCs) in the exhaust emissions were collected in Tedlar bags, re-sampled in sorbent tubes and analysed by thermal desorption-gas chromatography-flame ionisation detection. Pyrenean oak presented substantially higher emissions in the woodstove than in the fireplace, for the majority of compounds. The opposite was observed for poplar. Among the 45 identified species, benzene and benzene-related compounds represent the most abundant group, followed by oxygenated VOCs and aliphatics. Emission factors obtained in this study are generally of the same order than those reported for residential experiments in the USA.

Keywords—Fireplace, VOC emissions, woodstove.

I. INTRODUCTION

RESIDENTIAL biomass burning has been pointed out as one of the main sources of wintertime atmospheric pollutants, including in Mediterranean countries [1]-[4]. Emission factors (EFs) from this source are required not only for international reporting obligations, but also for applying source apportionment methodologies and for implementation of mitigation measures. As in other countries, in Spain there is a lack of information concerning the characteristics of emissions from biomass combustion systems. The emission profiles published in the literature up to now are almost exclusively focused on particles, CO and CO₂ from the residential combustion of typical Portuguese wood species [5]-[7] and, as far as we know, only one study devoted to the determination of volatile organic compounds (VOCs) from biomass burning of Mediterranean vegetation species has been carried out to simulate forest fires in a fireplace [8]. However,

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it has been demonstrated that the characteristics of biofuels and the type of combustion appliance used can have a huge influence on both gaseous and particle emissions [9]. Thus, any significant differences in emissions from diverse fuel types and combustion devices should be accounted for in regional control strategies focused on residential wood burning. The aim of this study was to characterise the VOC emissions from the combustion in a fireplace and in a woodstove of common woods grown in Spain.

II. METHODOLOGIES

A. Biofuels

Wood from two typical Spanish tree species, Pyrenean oak (*Quercus pyrenaica*) and black poplar (*Populus nigra*), was used as fuel (Table I). The wood was cut into logs of 0.3 to 0.4m in length with a total biomass burned during each cycle of around 1.1 to 1.8kg. The combustion of a batch of fuel lasted between 45 and 60min.

B. Biomass Combustion Facility and Sampling

Two different equipments were used in this work: i) a cast iron woodstove (Solzaima, model Sahara), operated manually in batch mode with handheld control of combustion air, and ii) a traditional brick open fireplace, also operated manually in batch mode, and with no control of combustion air. Both devices are equipped with a vertical exhaust duct (chimney) with 0.20m internal diameter and 3.30m height. A detailed description of the combustion facility (configuration, operating control, etc.) can be found elsewhere [10], [11]. Collection of VOCs was performed in a dilution tunnel, which is similar to other systems reported in previous studies [12]-[16].

TABLE I
CHEMICAL CHARACTERISTICS OF BIOFUELS

Proximate analysis (% wt, as received)	Moisture	<i>P. nigra</i>		<i>Q. pyrenaica</i>	
		WS	6.6	9.7	FP
	Ash		0.53		2.94
	C		48.86		47.22
	H		6.26		5.94
Ultimate analysis (% wt, dry basis)	N		0.07		0.20
	S		bdl		bdl
	O (by difference)		44.28		43.70
	Extractives		1.42		6.07
	Cellulose		50.9		38.8
	Lignin		31.1		28.1
	Hemicellulose		16.1		24.1

WS – Woodstove; FP – Fireplace, bdl – below detection limit

Sampling was made at a flow rate of about 400mL/min into Tedlar bags after pre-removal of particles in a filter and water vapour by condensation in a U-tube immersed in an ice/water bath. The dilution tunnel consisted of a cylindrical tube with 0.20m internal diameter and 11m length. The dilution ratios applied to the flue gases from the woodstove were around 25:1. In the case of the fireplace, 4 to 5 times dilution ratios were applied. Three or four replicate tests were performed for each wood species in every combustion appliance.

C. Analytical Determination of VOCs

The C>5 VOCs were desorbed and analysed by a thermal desorption/cryogenic concentration method on a Trace Ultra (Thermo scientific) gas chromatograph (GC) equipped with a thermal desorption injector Master TD (DANI) and a flame ionisation detector (FID). The VOCs contained in the adsorbent tubes were thermally desorbed at 250°C with pure helium at 8psi for 15min and cryofocused in a cold trap of the thermal desorber at -30°C. The trap was connected to the GC split/splitless injector by a transfer line heated to 250°C. Compounds were injected into a column (split ratio 25) by fast heating of the trap to 250°C using helium as a carrier gas (8psi). A TRB-1MS capillary column (50m×0.20mm i.d., 0.50µm) was used for separation of VOCs. The GC oven temperature programme was as follows: 40°C – 3min; from 40 to 160°C at 4°Cmin⁻¹; from 160 to 250°C at 10°Cmin⁻¹; 250°C –4min. Considering that additional calibrations are going on in order to quantify supplementary VOCs, the results presented here should be taken as preliminary.

III. RESULTS AND DISCUSSION

Around 45 individual VOCs have been identified and quantified in the exhaust fumes from the fireplace and woodstove (Tables II and III). Benzene was the compound with highest EFs for both wood species. Although both species are hardwoods, the magnitude of these and other VOC emissions is different when comparing fireplace and woodstove. In fact, Pyrenean oak presented substantially higher emissions in the woodstove than in the fireplace, for the majority of compounds. An opposite trend was observed for black poplar. Among the identified species, benzene and benzene-related compounds represent the most abundant

group, followed by oxygenated VOCs and aliphatics (Fig. 1).

Theoretically, one would expect that the high temperatures and long smouldering conditions encountered during a woodstove cycle favour the emission and formation of aromatic species. Taking into account that different proportions were observed between the VOCs from the combustion of oak and poplar in the fireplace and in the woodstove, the characteristics of firewood (Table I) may also exert an important effect on emissions. The combustion process is initiated when firewood is ignited using an external heat source. As the wood is heated, the compounds that makes up wood start to hydrolyse, oxidise, dehydrate, and pyrolyse. Combustible volatiles tarry substances and highly reactive carbonaceous char begin to form. The highest emissions are generated during this stage as the temperatures are insufficient to ignite the volatile gases. After ignition of volatiles and tarry substances, flaming combustion begins. The heat generated during flaming combustion evaporates any moisture contained in the wood, and drives out the volatiles arising from the decomposition products of the primary constituents – cellulose, hemicellulose, lignin and extractives. Thus, differences in the composition of the woods (such as those of this study) will be reflected in qualitative and quantitative differences in emissions. The volatilisation process continues until the amount of volatiles required to maintain flaming combustion decreases, and oxidation of the reactive char (or remaining charcoal) begins. Fixed carbon mixes with oxygen at high temperatures and CO₂ and heat are released. The heat is sufficient to continue the oxidising process and additional volatile compounds are produced, although at lower quantities than during the earlier stages of combustion [17].

In general, the emission factors obtained in this study are of the same order or slightly higher than those reported in the USA for the residential combustion in a woodstove and a fireplace of different hardwood and softwood species (Table IV). The exceptions encompass some of the higher molecular weight compounds. 3-Carene is emitted in lower proportions during the combustion of both poplar and oak than found in the smoke of American wood species. Whereas EFs of 18 and 190g/kg (dry biofuel burned) have been, respectively, reported in the USA for the combustion of hard- and softwood in a fireplace, in the present study, values always lower than 0.2g/kg (dry basis) were obtained in all tests. Much higher average EFs, between 21 and 55g/kg (dry basis), were also determined for naphthalene in the combustion experiments carried out in the USA, while the Spanish oak and poplar emitted 3 to 15g/kg (dry basis). Also, there is a considerable difference in the amount of limonene emitted from hardwood (3g/kg) and softwood (35g/kg) combustion of American species as compared to emissions of poplar and oak (<1g/kg, dry basis). Finally, acetophenone is present at lower quantities (<3g/kg) in the smoke of the Spanish biofuels, but emitted at 11-65g/kg (dry basis) in the case of the American wood species. Differences may be related not only to the specificities of each combustion device and characteristics of biofuels, but also to the adoption of different sampling and

analytical strategies.

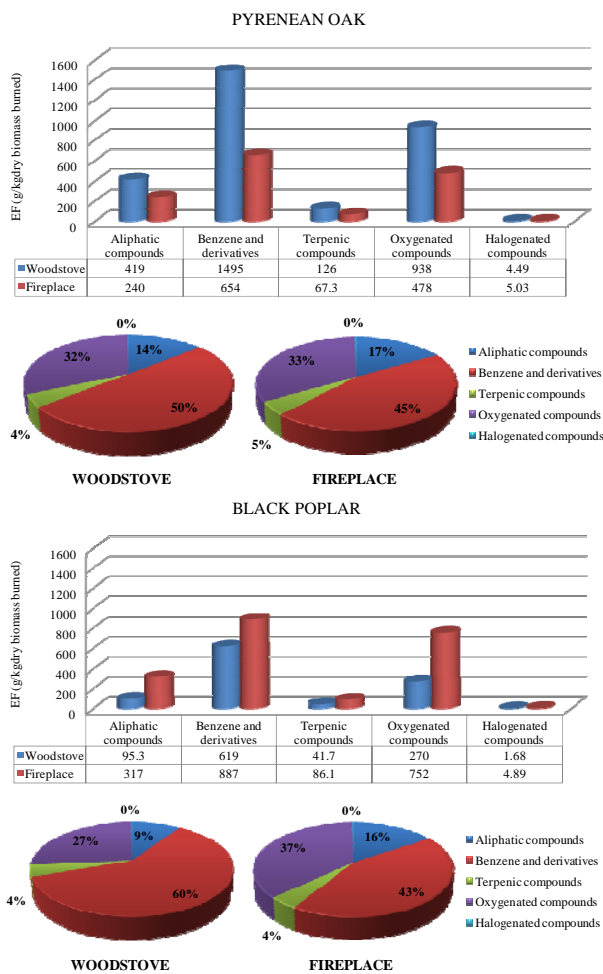


Fig. 1 Emissions of identified VOCs grouped into classes

TABLE II
EMISSION FACTORS OF VOCs FROM COMBUSTION OF PYRENEAN OAK

Compound	EF (g/kg dry biomass burned)	
	Woodstove	Fireplace
<i>n</i> -Pentane	23.4 ± 5.2	12.9 ± 3.1
Isoprene	111 ± 16	56.5 ± 7.3
Methyl acetate	356 ± 95	174 ± 25
2,2-Dimethylbutane	161 ± 23	86.6 ± 15.1
<i>i</i> -Hexane	152 ± 30	94.4 ± 20.2
Butanal	21.3 ± 3.7	12.9 ± 3.0
2-Butanone	158 ± 18	101 ± 20
<i>n</i> -Hexane	15 ± 4.6	8.61 ± 3.03
Ethyl acetate	24.9 ± 7.4	11.0 ± 3.0
1-Butanol	13.1 ± 3.4	8.42 ± 2.63
Benzene	901 ± 131	390 ± 60
Cyclohexane	11.2 ± 0.9	5.55 ± 1.58
1,2-Dichloropropane	4.49 ± 0.68	3.54 ± 0.78
1-Heptene	23.9 ± 2.7	14.9 ± 3.7
<i>n</i> -Heptane	13.0 ± 3.0	6.82 ± 1.97
Toluene	326 ± 47	156 ± 17
Hexanal	10.8 ± 2.6	6.25 ± 1.52
<i>n</i> -Octane	7.31 ± 0.78	4.86 ± 1.19
2-Furaldehyde	599 ± 186	445 ± 81
Ethylbenzene	52.9 ± 5.9	22.4 ± 2.7
<i>m,p</i> -Xylene	39.7 ± 7.1	22.5 ± 4.4
Styrene	114 ± 12	39.8 ± 4.1
<i>o</i> -Xylene	26.7 ± 2.1	12.0 ± 2.3
<i>n</i> -Nonane	2.94 ± 0.20	1.75 ± 0.49
Isopropylbenzene	3.10 ± 0.49	1.33 ± 0.30
Bromobenzene	nd	1.21 ± 0.31
4-Methylcyclohexanol	nd	0.945 ± 0.215
α -Pinene	9.02 ± 4.42	6.54 ± 2.37
Benzaldehyde	27.8 ± 6.2	8.83 ± 0.78
<i>n</i> -Propylbenzene	3.25 ± 0.64	1.62 ± 0.40
1,3,5-Trimethylbenzene	1.66 ± 0.38	0.830 ± 0.200
<i>tert</i> -Butylbenzene	10.5 ± 1.8	4.55 ± 1.00
1,3-Dichlorobenzene	nd	0.276 ± 0.046
<i>n</i> -Decane	2.26 ± 0.31	0.942 ± 0.211
<i>sec</i> -Butylbenzene	1.62 ± 0.37	0.639 ± 0.100
3-Carene	0.16 ± 0.03	0.083 ± 0.023
<i>p</i> -Isopropyltoluene	0.781 ± 0.096	0.375 ± 0.102
Limonene	0.855 ± 0.137	0.967 ± 0.280
Eucalyptol	2.45 ± 0.36	2.50 ± 0.76
Acetophenone	2.93 ± 0.60	0.589 ± 0.244
γ -Terpinene	5.30 ± 1.02	1.29 ± 0.12
3-Methylbenzaldehyde	1.11 ± 0.25	0.286 ± 0.055
<i>n</i> -Undecane	0.479 ± 0.096	0.113 ± 0.300
Naphtalene	15.4 ± 2.6	2.64 ± 0.33

nd – not detected; bdl – below detection limit

TABLE III

EMISSION FACTORS OF VOCs FROM COMBUSTION OF BLACK POPLAR

Compound	EF (g/kg dry biomass burned)	
	Woodstove	Fireplace
<i>n</i> -Pentane	4.22 ± 1.00	13.53 ± 2.75
Isoprene	38.1 ± 10.9	77.52 ± 17.64
Methyl acetate	78.4 ± 6.6	316 ± 116
2,2-Dimethylbutane	43.9 ± 8.9	115 ± 19
<i>i</i> -Hexane	28.3 ± 2.8	141 ± 53
Butanal	6.74 ± 1.51	24.3 ± 8.5
2-Butanone	35.5 ± 8.4	148 ± 43
<i>n</i> -Hexane	3.39 ± 0.64	7.60 ± 1.68
Ethyl acetate	6.69 ± 1.11	14.50 ± 4.17
1-Butanol	2.17 ± 0.72	19.66 ± 7.35
Benzene	401 ± 89	532 ± 76
Cyclohexane	3.57 ± 0.55	7.15 ± 2.39
1,2-Dichloropropane	1.03 ± 0.35	3.25 ± 0.55
1-Heptene	3.68 ± 1.15	11.53 ± 1.83
<i>n</i> -Heptane	2.51 ± 0.69	6.87 ± 1.59
Toluene	126 ± 20	216 ± 25
Hexanal	2.72 ± 1.87	8.85 ± 1.75
<i>n</i> -Octane	1.42 ± 0.64	5.15 ± 1.28
2-Furaldehyde	73.1 ± 19.3	30.0 ± 17.2
Ethylbenzene	16.9 ± 2.1	28.2 ± 3.1
<i>m,p</i> -Xylene	14.5 ± 2.2	34.3 ± 3.3
Styrene	40.0 ± 8.0	49.7 ± 7.8
<i>o</i> -Xylene	7.22 ± 1.10	15.92 ± 1.35
<i>n</i> -Nonane	0.723 ± 0.193	1.929 ± 0.434
Isopropylbenzene	1.365 ± 0.331	2.021 ± 0.429
Bromobenzene	0.647 ± 0.116	1.214 ± 0.572
4-Methylcyclohexanol	0.843 ± 0.114	0.976 ± 0.167
α -Pinene	1.64 ± 0.27	5.46 ± 4.06
Benzaldehyde	10.89 ± 2.48	7.99 ± 1.41
<i>n</i> -Propylbenzene	0.766 ± 0.137	1.73 ± 0.24
1,3,5-Trimethylbenzene	0.697 ± 0.122	1.143 ± 0.161
<i>tert</i> -Butylbenzene	3.07 ± 0.47	4.90 ± 0.60
1,3-Dichlorobenzene	bld	0.427 ± 0.076
<i>n</i> -Decane	0.745 ± 0.121	0.867 ± 0.155
<i>sec</i> -Butylbenzene	bld	0.809 ± 0.178
3-Carene	bld	0.202 ± 0.107
<i>p</i> -Isopropyltoluene	0.155 ± 0.029	0.262 ± 0.116
Limonene	0.207 ± 0.026	0.547 ± 0.424
Eucalyptol	0.955 ± 0.131	2.41 ± 1.43
Acetophenone	0.923 ± 0.246	0.736 ± 0.099
γ -Terpinene	1.64 ± 0.14	bld
3-Methylbenzaldehyde	0.489 ± 0.043	0.217 ± 0.024
<i>n</i> -Undecane	0.136 ± 0.957	0.158 ± 0.034
Naphtalene	7.39 ± 1.75	2.85 ± 0.92

nd – not detected; bdl – below detection limit

TABLE IV

EMISSION FACTORS FOR COMBUSTION TESTS IN THE USA [18]

Compound	EF (g/kg dry biomass burned)		
	Hardwood (woodstove)	Hardwood (fireplace)	Softwood (fireplace)
<i>n</i> -Pentane	13.12 ± 7.81	11.85 ± 5.89	8.69 ± 0.84
Isoprene	43.25 ± 31.86	27.04 ± 13.39	38.74 ± 14.63
Butanal	36.49 ± 26.56	22.48 ± 14.61	18.64 ± 1.26
2-Butanone	172.7 ± 96.7	138.8 ± 44.2	80.40 ± 15.47
<i>n</i> -Hexane	12.30 ± 7.47	7.02 ± 3.77	6.01 ± 2.03
Benzene	1190 ± 875	312.2 ± 83.1	225.0 ± 40.3
1-Heptene	6.70 ± 4.45	5.15 ± 3.94	3.22 ± 0.77
<i>n</i> -Heptane	4.60 ± 2.04	5.36 ± 3.23	3.71 ± 0.26
Toluene	320.0 ± 223.6	141.5 ± 44.0	130.6 ± 21.1
<i>n</i> -Octane	14.94 ± 21.78	3.90 ± 3.00	2.54 ± 0.71
2-Furaldehyde	99.71 ± 80.13	445.3 ± 473.8	317.6 ± 258.2
<i>m,p</i> -Xylene	71.78 ± 47.67	40.91 ± 12.22	49.63 ± 16.95
Styrene (+heptanal)	117.2 ± 91.5	34.51 ± 22.29	40.17 ± 13.15
<i>o</i> -Xylene	27.28 ± 16.80	19.15 ± 9.71	16.12 ± 4.19
<i>n</i> -Nonane	5.13 ± 3.56	2.31 ± 1.90	1.09 ± 0.26
α -Pinene	3.69 ± 2.55	4.01 ± 4.39	53.59 ± 24.05
<i>n</i> -Decane	1.67 ± 0.71	2.10 ± 1.94	0.92 ± 0.26
3-Carene	0.00 ± 0.01	18.15 ± 27.13	189.6 ± 56.4
Limonene	0.06 ± 0.13	2.89 ± 1.15	35.36 ± 8.71
Acetophenone	10.90 ± 4.95	56.63 ± 48.87	64.95 ± 30.03
<i>n</i> -Undecane	1.29 ± 1.28	2.92 ± 2.57	2.27 ± 0.93
Naphtalene	28.06 ± 19.06	54.62 ± 18.32	21.42 ± 5.13

IV. CONCLUSION

Data concerning the emissions of VOCs from residential wood combustion are currently sparse. This is one of the very few studies reporting emission profiles for the combustion of wood species in woodstove and fireplace. The comprehensive database allowed the observation of significant differences in emissions, both between wood species, or between combustion devices. Such data are important due to the toxicity of many compounds and/or the presence of highly reactive species. Information collected in the current study is potentially useful to model the impact of residential wood combustion on tropospheric ozone formation. In addition, the characterisation of gas-phase emissions from wood combustion provided in this study, when combined with particle-phase wood smoke tracers, makes it easier to estimate the contribution of domestic biomass burning to ambient VOC concentrations.

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