Emission factors from small scale appliances burning wood and pellets

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1. Introduction

Residential wood combustion (RWC) is a major source of air pollutants with potential health hazards (Bølling et al., 2009). Its impacts on local air quality are confirmed in numerous studies by different methods such as emission inventories, air quality data analysis and modeling, and source receptor modeling (Hellén et al., 2008; Glasius et al., 2006; Gianelle et al., 2013).

In addition to a high level of particulate matter (PM), RWC produces volatile organic compounds (VOC) with a high content of various toxic and carcinogenic compounds such as polycyclic aromatic hydrocarbons (PAH; Ravindra et al., 2008) and dioxins (Lavric

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Table 1
Description of the tested appliances.

Appliance	Fuel	Nominal heat output [kW]	Energy efficiency [%]	Air regulation	Combustion air	Heat transfer
Open fireplace	Log wood	8	51	Manual	Natural draft	Natural convection
Closed fireplace	Log wood	11	82	Manual	Primary and secondary	Forced air and natural convection
Traditional stove	Log wood	6	70	Manual	Primary	Natural convection
Advanced stove	Log wood	8	76	Manual	Primary and secondary	Natural convection
Stove	Pellets	8	91	Automatic	Primary and secondary	Forced air
Boiler	Pellets	25	93	Automatic	Lambda probe	Water

et al., 2004). RWC is also an important source of black carbon (BC) and organic carbon (OC) emissions (Caserini et al., 2013) and thus has an impact on climate, given the potential of BC and OC to alter the Earth's energy balance through a complex net of processes (US-EPA, 2012).

Regarding the greenhouse gas (GHG) emissions, despite the debate going on over the validity of the assumption of carbon neutrality for biomass from forestry where the carbon cycles can last centuries (McKechnie et al., 2011; Zanchi et al., 2011), net GHG emission savings are expected substituting biomass for fossil fuels in various combustion processes (e.g. Caserini et al., 2010). Besides, the combination of high fossil prices and international efforts to decrease GHG together with associated incentives for bioenergy is expected to increase the use of different types of biomass in the near future (Beurskens et al., 2011).

Although the impacts of RWC on air quality have been studied for three decades (Dasch, 1982), there is still a substantial uncertainty on the emission assessment of RWC at a local scale (Pastorello et al., 2011). This is not only due to the uncertainty related to the activity data (i.e., amount of fuel burnt), but also to the lack of emission factors (EFs) able to represent actual combustion conditions. In fact, the magnitude of emissions from RWC depends heavily on combustion device, fuel quality and operating conditions (Johansson et al., 2004; EEA, 2013; Win et al., 2012; Orasche et al., 2013).

Emissions caused by incomplete combustion are mainly a result of combustion conditions such as inadequate mixing of combustion air and fuel in the combustion chamber, an overall lack of available oxygen, too low over all combustion temperatures, non-homogeneous temperature distribution in the combustion chamber (cold zones), as well as too short residence times, they thus depend on operational practices that could differ significantly between countries and appliances (Van Loo and Koppejan, 2008; EC DG TREN, 2009; Nussbaumer, 2010). This point is not formally addressed in standard methods used for testing small combustion devices; the methods for non-heat storing appliances require a combustion process as constant as possible, not considering the transient phases such as the initial kindling when the fuel temperature is locally raised up to several hundred degrees, and the refueling onto an existing fire bed which is likely to be repeated several times per burning session.

The present work aimed to determine average EFs for combustion devices, types of fuel and firing behavior typical of Italy and European countries. The work assessed the EFs of manual appliances burning log wood and automatic devices burning pellets, investigating the influence of the fuel type and burning cycle on the emissions in order to support the new policy frameworks needed to lower the wood burner emissions (i.e., programs of incentives for the substitutions of most polluting devices) (Fuller et al., 2013). Differently from the standard emission testing methods, user habits were simulated in a schematic way in the laboratory employing different combustion cycles that represent a realistic user behavior. The experimental results are compared with the reference values proposed in the EMEP/EEA Air Pollutant Emission Inventory Guidebook (AEIG; EEA, 2013) used for local and national inventory in Europe.

2. Experimental

2.1. Tested appliances

Technology and characteristics of the six tested appliances are summarized in Table 1. The tested devices are representative of the residential heating appliances burning woody biomasses commonly used in Italy (Caserini et al., 2007) and in Europe (EEA, 2013). Among them 4 manual appliances burning wood logs and 2 automatic appliances fed with wood pellets were selected (detailed information in the Supplementary Material). In particular, an open fireplace (8 kW heat output) was tested to represent low efficiency old fireplaces; while a close fireplace (11 kW nominal output) provided with a forced air convection heat exchanger was considered representative of newly featured models, it is also equipped with a separated manual control system for primary and secondary combustion air. The same combustion regulation is used in the advanced stove while the traditional stove has only primary air control. The automatic pellet stove (8 kW heat output) represents one of the most common appliance types in the Italian market while the pellet boiler provided with a lambda probe is a technologically advanced model.

Table 2
Characteristics of the tested woody biomass.

	Beech	Hornbeam	Oak	False acacia	Spruce	High quality pellet	Low quality pellet
Moisture ($\%_w$)	9.5	9.8	10	9.2	9.3	6.8	7.1
Ashes ($\%_w$)	0.5	0.5	1.4	0.8	0.4	0.4	0.8
Carbon (% kg kg^{-1})	44.9	45.3	44.8	45.2	46	47.9	48.1
Hydrogen (% kg kg^{-1})	5.4	5.3	5	5.6	5.3	5.5	5.5
Nitrogen (% kg kg^{-1})	0.1	<0.1	<0.1	0.4	0.65	0.3	0.35
Chlorine (mg kg^{-1})	<10	30	<10	170	20	30	85
Sulfur (mg kg^{-1})	70	150	105	355	40	55	110
NCV (MJ $kg^{-1}{}_{wet\ basis}$)	16.5	16.4	16.2	16.2	16.9	17.5	17.6

2.2. Fuels

Five types of firewood (beech, false acacia, hornbeam, oak, and spruce) were used for the feeding of manual appliances; these types were selected for their large market penetration, especially in northern Italy, where firewood use is most common. Table 2 summarizes the main characteristics of the tested woods determined based on the international standard methods UNI EN 15104-2011 for elemental analysis (C, H, N), UNI EN 14918-2012 for NCV (net calorific value) determination, ASTM D7582 for moisture and ash content and UNI 15289-2011 for chlorine and sulfur concentration.

Moisture content, ranging between 9.2%$_w$ for false acacia and 10%$_w$ for oak, was below the limit values defined for type testing for all wood types, and it was lower than the threshold indicated in the best practices for a correct use of heating systems fed with woody fuels as stated by UNI EN 14961-5 (first class for wood logs <20%), DIN 51731 (<12%) and Austrian classification ÖNORM M7132 (<10%). Beech wood is generally considered the reference fuel for type testing and the other woods are comparable with it in terms of composition except for hornbeam and false acacia woods which are characterized by high amounts of chlorine and sulfur. The wood logs selected for the tests had length and diameter suggested by the appliance user manual which guarantee a good handling during the combustion chamber loading. No log debarking has been carried out to simulate, as far as possible, real operating conditions.

Two types of pellets (i.e., low-quality cheap pellets and high-quality pellets with DIN-PLUS certification) were selected for the experiments in the automatic stove and boiler; their characteristics are reported in Table 2. The cheap pellets have higher ash, chlorine and sulfur content with respect to the certified pellets, whereas NCV is similar for both types.

2.3. Combustion cycle

European standards regarding type testing of residential solid fuel appliances (EN 13240:2001, EN 13229:2001, EN 14785:2006) require the use of predefined, well controlled combustion cycles. The difference of appliance behavior between well-controlled and real-world usage is significant, especially in batch working devices where the combustion process shows great variability throughout the main phases (i.e., drying, pyrolysis/gasification, combustion) caused by the batch feeding of the appliance (Van Loo and Koppejan, 2008). The definition of the actual combustion cycle, in terms of duration of start-up and stopping phases, or of refueling frequency, is thus crucial for the evaluation of EFs for manually fed heating systems under real-world operating conditions.

Field measurements of the temporal variation of the combustion temperature inside the firebox were carried out with a temperature probe and a data logger in 13 different appliances in 12 houses in northern Italy, with the purpose of obtaining a realistic estimation of the main characteristics of the actual combustion cycle (i.e., frequency of refueling). Nineteen periods of measurement (10–15 days of duration on average), for a total of about 1300 combustion hours, together with interviews to the owners through a questionnaire, allowed to assess an average time interval between two refueling operations of about 60 min for open fireplaces, 80 min for closed fireplaces and 90 min for wood stoves; intervals from one wood load to the following resulted larger in comparison with the time needed to consume the load.

Aiming to be representative of the average user behavior, the “real life” cycle (Cycle A) has been defined as follows and was subsequently reproduced in laboratory:

- the ignition phase consisted of the load of a small amount of wood sticks (0.7 kg) keeping the air regulation valve completely open throughout the phase,
- after 20 min from the ignition an amount of wood logs equal to the nominal fuel load (defined by the manufacturer) was added and the air valve was partially closed,
- after 1 h a second load was added,
- after another hour a final full load was added.

The duration of each load was limited to a maximum of 60 min, which is already higher than the measurement duration during the standard cycles; the fire has been stoked if necessary not more than once in the time between one load and the following.

Furthermore, regarding the closed combustion chamber appliances (i.e., closed fireplaces, traditional or advanced stoves), during the late evening hours the users were typically observed to add one last large batch and to close the air inlet completely, trying to make the combustion last as long as possible. This behavior was simulated through a second type of cycle (Cycle B): this cycle is similar to Cycle A except that the final of the three consecutive loads is increased by 50% and simultaneously the air valve is totally closed.

The Cycle A in the open fireplace gave many problems in terms of smoke leaks directly from the open combustion chamber. To avoid this inconvenience the nominal load was divided into three batches and the pollutants have been sampled during nine consecutive refueling periods (Cycle C).

The manual appliances were tested also during the operating conditions described in standard methods to compare the emissions from aforementioned “user behavior” cycles with those from standard cycles. The two stoves and the closed fireplace were tested according to EN-13240 where a pre-test period is imposed to guarantee stable appliance temperature which enables stable operating conditions and an acceptable repeatability of the emission measurements. The emissions have been measured during three consecutive loadings with each measurement period not less than 45 min (Cycle EN). The open fireplace was tested according to EN-13229 which required a pre-test period before starting the loading cycles used for emission monitoring without any limit on the measurement period.

For automatic appliances (pellet stove and pellet boiler) the combustion cycle used (Cycle P) starts after 1 h of appliance operation and lasts for 1 h at nominal heat output.

A summary of the number of test runs per cycle type used for the estimation of the EFs related to different appliances is shown in Table 3.

Table 3
Number of test runs per cycle type used for the estimation of the emission factors.

	Cycle type	Number of EF			
		CO	NOx	NMHC	PM
Open fireplace	C	15	15	15	15
	EN	1	1	1	1
Closed fireplace	A	5	5	4	5
	B	13	13	13	10
	EN	1		1	1
Traditional stove	A	5	5	5	5
	B	10	10	10	10
	EN	1	2	2	1
Advanced stove	A	5	5	4	5
	B	10	10	8	10
	EN	1	1	2	5
Pellets stove	P	6	6	6	6
Pellets boiler	P	2	2	2	6

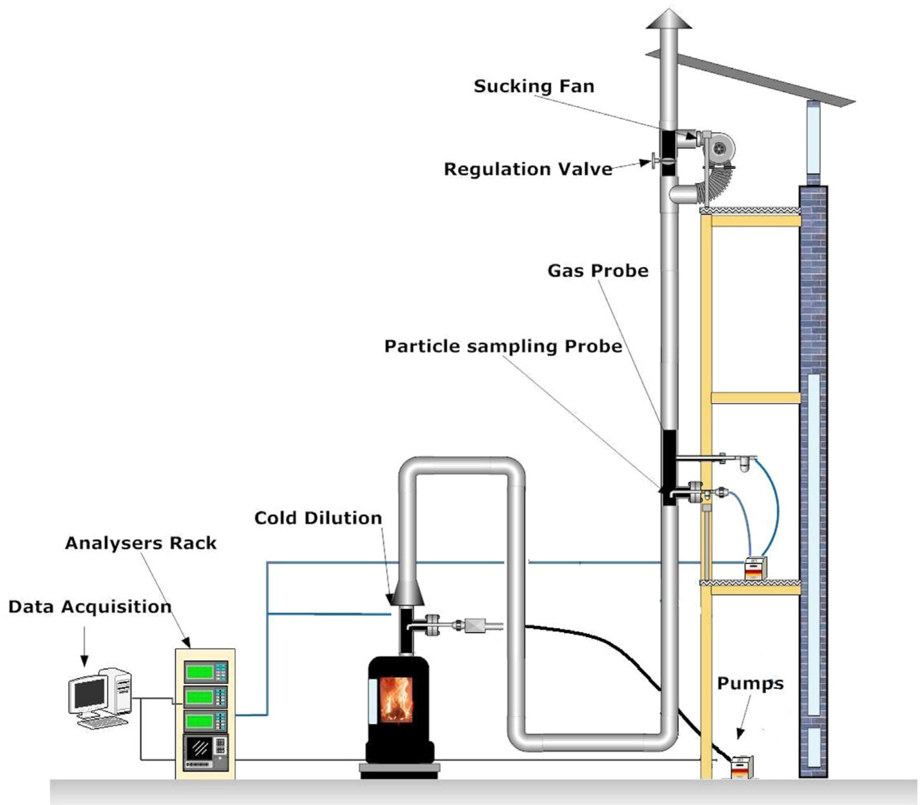

Fig. 1. Scheme of the test bench.

2.4. Sampling systems

In order to obtain a realistic evaluation of ambient emissions and to measure the condensable particles produced by heating systems, the emission sampling was performed in a dilution tunnel (Fig. 1) according to the NS3058-2 the Norwegian standard method for the determination of PM emissions produced by small appliances fed with woody biomass (Standard Norge, 1994). Gases coming from wood combustion are diluted about 10 times with ambient air by means of a manually regulated extraction fan. In this way, the flue gas was quenched up to a temperature around 30–35 °C promoting the condensation of the volatile organic

Table 4
Average CO, NO*x*, NMHC and PM emission factor (g GJ^{-1}) and 95th confidence intervals.

	Appliance type	Number of experimental data	Average EF – experimental	95th Confidence intervals	
				Lower	Upper
CO (g GJ^{-1})	Open fireplace	15	5048	4417	5828
	Closed fireplace	18	4471	3949	5030
	Traditional stove	15	7681	6059	11131
	Advanced stove	15	6232	4885	7829
	Pellets stove	6	88	73	108
	Pellets boiler	2	350		
NO*x* (g GJ^{-1})	Open fireplace	15	134	121	148
	Closed fireplace	18	120	105	140
	Traditional stove	15	100	91	110
	Advanced stove	15	132	99	182
	Pellets stove	6	60	32	90
	Pellets boiler	2	71		
NMHC (g GJ^{-1})	Open fireplace	15	1011	853	1219
	Closed fireplace	17	548	445	677
	Traditional stove	15	243	197	352
	Advanced stove	12	366	266	701
	Pellets stove	6	9.0	3.0	17
	Pellets boiler	2	1.3		
PM (g GJ^{-1})	Open fireplace	15	512	434	611
	Closed fireplace	15	183	152	219
	Traditional stove	15	178	140	225
	Advanced stove	15	143	120	176
	Pellets stove	6	109	75	139
	Pellets boiler	6	61	30	103

compounds. The calculation of the dilution ratio is based on simultaneous carbon dioxide (CO_2) concentration measurements in the flue gas upstream and downstream the dilution. The appliances were placed on a scale to measure the weight variation during the test periods. A thermocouple was located just after the combustion chamber to measure the flue gas temperature.

2.5. Measurement techniques

2.5.1. Gases

The exhaust gases were extracted from the dilution tunnel by means of a heated probe at 160 °C, to be distributed to the following flue gas analyzers: a flame ionization detector (CAI 600 M-HFID) equipped with a catalytic cutter for non-methane hydrocarbons (NMHC), a chemiluminescence analyzer (Thermo Environmental Instr. 42H) for nitrogen oxides (NO_x), and a non-dispersive infra-red gas analyzers for carbon monoxide (CO) and carbon dioxide (Fisher Rosemount NGA 2000). Flue gas O_2 and CO_2 were measured before the dilution tunnel with a multiparameter analyzer (Horiba PG-250).

2.5.2. Particulate matter (PM)

For each test total PM emissions were measured after the flue gas dilution according to NS:3058-2 sampling method (Standard Norge, 1994b). The gas temperature at the filter holder was between 32 and 35 °C. Prebaked (3 h at 850 °C), preweighed 47 mm quartz fiber filters were used for all tests. The filters were conditioned in a desiccator at ambient temperature before weighing and were stored at −20 °C till use. After sampling they were conditioned in the desiccator for 24 h and then weighed to determine the amount of PM.

Some additional tests were performed on the advanced stove under the EN cycle with simultaneous sampling from the hot (i.e., just after the combustion chamber) and diluted (i.e., from the dilution tunnel) flue gas in order to determine the influence of the sampling conditions.

2.5.3. PAH and dioxins

PAH and dioxins were measured respectively according to the ISO standard 11338-1 and UNI EN 1948-1. Both pollutants were collected with the same sampling system which is composed of a titanium sampling probe heated at 120 °C, a quartz fiber filter to collect the particulate matter, a cooling system at 2 °C that condenses the condensable gas and polyurethane foam (PUF) to fix the gas phase. The filter and the PUF were extracted and analyzed together with the condensate by High Resolution GC/MS. PAH samples were analyzed for benzo(a)pyrene (B(a)P), benzo(b)fluoranthene (B(b)F), benzo(k)fluoranthene (B(k)F) and indeno(1,2,3−cd)pyrene (IP).

2.6. Emission factor calculation

The EFs in a sampling period may be estimated from average pollutant concentration and average flue gas volume flow rate in relation to the amount of fuel consumed (e.g., g $kg^{-1}_{dry\ fuel}$), or in relation to the energy input to the combustion process (e.g., g MJ^{-1}). While the method may work quite well with stable combustion processes, assuming constant flue gas flow rate, deviations from the average emission factor calculated in this way may occur in batch combustion installations with variations among different combustion phases. In these cases, for continuous gaseous pollutant measurements, in alternative to considering the average conditions, the emission factor may be calculated instant by instant based on observed concentration values weighted by the specific flue gas volume, $V_{spec,i}$ (i.e., dry flue gas volume produced per kg dry fuel, $m^3\ kg^{-1}_{fuel}$):

$$EF = \frac{1}{n}\sum_i C_i \cdot V_{spec_i} \Big/ NCV*10^3 \quad (1)$$

where EF is the emission factor (g GJ^{-1}), C_i is the observed pollutant concentration (g m^{-3}) at instant i of n observations, and NCV is the fuel net calorific value (MJ $kg^{-1}_{dry\ fuel}$). If the flue gas CO_2 content is known, the dry flue gas volume produced per kg of dry fuel may be calculated with Eq. (2) assuming a complete combustion:

$$V\text{spec}_i = X_C \cdot V_{mol} / (M_C \cdot CO_{2i}) \quad (2)$$

where, X_C is the fuel carbon content ($kg_C\ kg^{-1}_{fuel}$), V_{mol} is gas molar volume (22.4 l at NTP), M_C is carbon molar weight (12 g mol^{-1}), CO_{2i} is flue gas CO_2 concentration at instant i (%$_v$ of dry gas) (Van Loo and Koppejan, 2008).

For PM, EFs (in terms of g GJ^{-1}) for all tested appliances are calculated by means of fuel consumption rate, NCV and emission

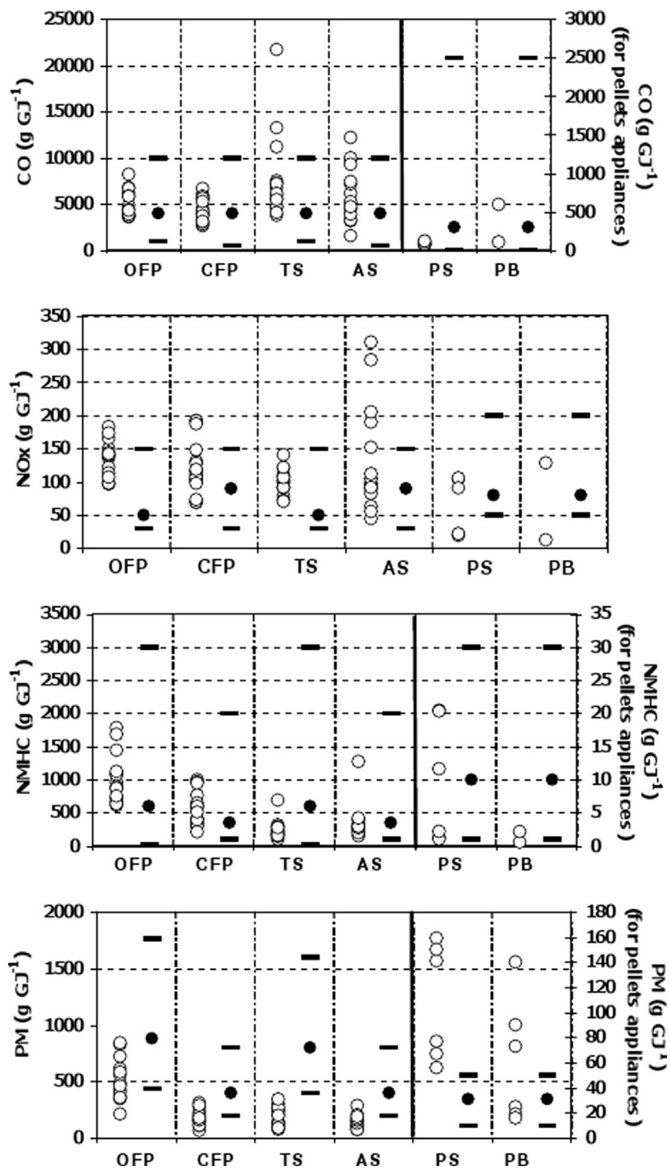

Fig. 2. Experimental emission factor results (white circles) from batch working and automatic appliances in comparison with AEIG average EF (black dots) and the 95th confidence interval (black lines). All data in g GJ^{-1} (OFP: open fireplace, CFP: closed fireplace, TS: traditional stove, AS: advanced stove, PS: pellets stove, PB: pellets boiler).

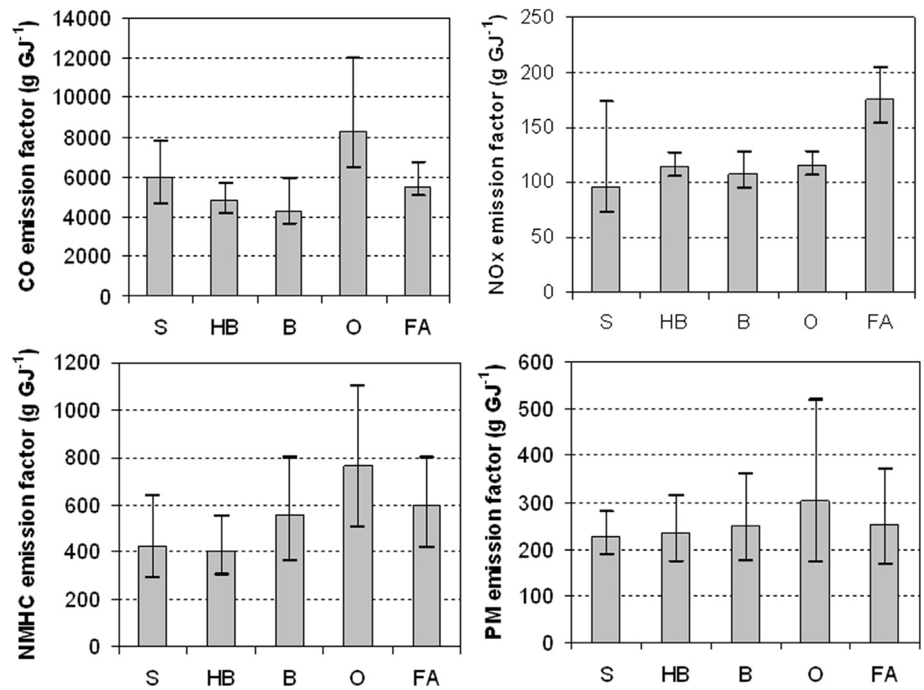

Fig. 3. Average emission factors (gray bars) and the 95% confidence interval of the mean (error bars) for manually fired appliances (open fireplace, closed fireplace, traditional stove and advanced stove) grouped by fuel type (S: spruce, HB: hornbeam, B: beech, O: oak, FA: false acacia).

ratio (g h^{-1}) measured according to the NS:3058-2 sampling method.

3. Results and discussion

3.1. Gaseous compounds and PM emission factors

CO, NO*x*, NMHC and PM EFs in g GJ^{-1} from all tested appliances are provided in the Supplementary Material (Tables S1–S4); average results and 95th bootstrap percentile confidence intervals (c.i.) are reported in Table 4 (and in Table S5 in g $kg^{-1}_{\text{dry basis}}$) and the experimental results are compared with AEIG (EEA, 2013) average Tier 2 EFs and the 95th c.i. in Fig. 2. Based on the characteristics of the appliances, results for closed fireplace and advanced stove were compared with AEIG values for energy efficient stoves.

3.1.1. Influence of appliance type

Experiments showed that gaseous emissions from batch working appliances are highly variable during operation with peaks of incomplete combustion products in correspondence of the fuel feed to the combustion chamber; on the other hand, automatically fired systems exhibit relatively constant emissions. This fact reflects on CO and NMHC EFs, which result to be one to two orders of magnitude lower in automatically fed appliances with respect to manually fed systems; while for NO*x* EFs the influence of the appliance type (i.e., batch vs. continuous), has a minor effect; NO*x* EFs for automatic appliances are lower by only a factor of 2.

The closed and the open fireplaces had lower CO emissions than traditional and advanced stoves; average experimental EFs for open and closed fireplaces are within 25% of the Tier 2 average value suggested in the AEIG, whereas higher values have been measured for the traditional and advanced stoves. An opposite trend is observed for NMHC emissions for which the stoves present a better performance with respect to the fireplaces, and experimental EFs are very close to the proposed AEIG values for the advanced stove, higher for the open and the closed fireplaces and lower for the traditional stove which is the appliance with the best NMHC performance among the tested appliances. Regarding the NO*x* emissions, all batch working appliances have a comparable performance with the lowest emissions from the traditional stove. However, the experimental average EFs are about two times higher than the AEIG values for manually fed appliances. Fig. 2 shows also that average experimental EFs for the automatic appliances are generally lower than or in some cases comparable to the proposed AEIG values.

PM EFs from all tested appliances are provided in Table S4 (Supplementary Material). Average results and 95th confidence intervals, reported in Table 4 and Fig. 2, show that the experimental EFs for the manually fed appliances are in general lower than Tier 2 AEIG values and below the lower value of confidence interval (except for open fireplace). This can be due to the low humidity of the fuel (as reported in paragraph 2.2) that may be cause of a decrease in EF (Shen et al., 2013; Hays et al., 2003; Gras et al., 2002). In any case, no definitive conclusions are drawn due to relatively limited number of samples.

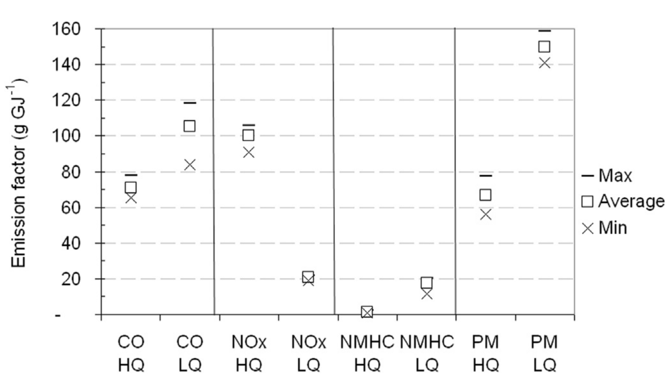

Fig. 4. Emission factors (g GJ^{-1}) for pellet stove for high quality pellets (HQ) and low quality pellets (LQ).

In the case of automatic appliances, EFs for PM are higher than values proposed by AEIG; results are highly influenced by pellet quality, as will be shown in the next paragraph.

3.1.2. Influence of fuel type

The summary of the average EFs for the manually fired systems is shown in Fig. 3 grouped for different fuel types (i.e., spruce, hornbeam, beech, oak, false acacia). Average CO emissions relative to the five wood types are in the same order with a maximum difference of two times between beech wood presenting the lowest and oak the highest EF value. Similarly for NO*x*, the EFs are in the same order of magnitude, with a maximum difference of two times between the highest average for false acacia and the lowest for spruce. NMHC emissions result to be highly variable among the fuel types with a difference of two times between oak and spruce, however largely overlapping 95% confidence intervals suggest that the differences may not be significant. PM EFs are very similar (1.3 times between the minimum average EF observed for spruce and the maximum for oak); the confidence intervals are largely overlapping suggesting that the differences for the wood types may not be significant. Several studies in literature (Hays et al., 2003; Purvis et al., 2000; McDonald et al., 2000) have investigated the influence of wood type on EFs, and in general softwood is considered to have the highest PM emissions (Meyer, 2012). However, this was not the case of the present study, since average emissions obtained with spruce are lower than those from oak.

Regarding the automatic appliances, better pellet quality (i.e., lower ash, Cl and S content) corresponds to lower incomplete combustion product emissions (i.e., CO and NMHC) and higher NO*x* emissions (Fig. 4). Fig. 4 shows the marked dependence of PM EFs on the type of pellets combusted in the pellet stove, with an increase of 124% in PM emissions; the influence is even larger for pellet boiler (+400%, see Table S4). As shown in Table 2 the main difference between the two pellet types is in the ash, sulfur and chlorine content, since moisture and calorific value are similar. The enhancement in PM emissions related to the used high-ash-content pellets is connected also to the less efficient combustion process as indicated by the higher CO emission factor (1.5 times on average), higher NMHC and lower NO*x* emission factors.

3.1.3. Influence of sampling conditions

In some selected cases (namely manual appliances fed with beech wood), PM sampling was carried out also in the hot flue gas prior to the dilution tunnel (see EN[a] values in Table S4) as prescribed by some national standards. In each case, the PM EF for the measurement in hot flue gas under EN cycle ($EF_{hot,EN}$) is lower than the corresponding average EFs for the measurement in the dilution tunnel under real-life cycles A and B ($EF_{dil,\ real}$). The average $EF_{dil,\ real}$ is 1.5 times the $EF_{hot,EN}$ for the open fireplace, about 4 times for the closed fireplace and traditional stove, and 11 times for the advanced stove. It is well known that PM sampled in a dilution tunnel includes also the condensable species, which are present in gaseous form in the hot gases, increasing the overall measured PM concentration (Nussbaumer et al., 2008). So, the differences observed may be associated to the combined effect of the increased NMHC levels during real-life cycles and the enhanced gas-to-particle partitioning when the flue gas is diluted and cooled. The smaller difference between the EFs in the case of the open fireplace is probably due to the fact that the flue gas produced by this kind of appliance is already highly diluted and cooled hence quite similar to the conditions encountered in the dilution tunnel (except for the temperature which is higher: about 100 °C in the stack and about 35 °C in the dilution tunnel).

In order to better understand the influence of the sampling conditions, a set of simultaneous sampling were performed in the hot and diluted flue gas from the advanced stove fed with beech wood under EN cycle (Table S6). In these tests, the diluted EFs are on average 4 times the hot EFs, confirming that the sampling conditions can influence substantially the PM emission factors.

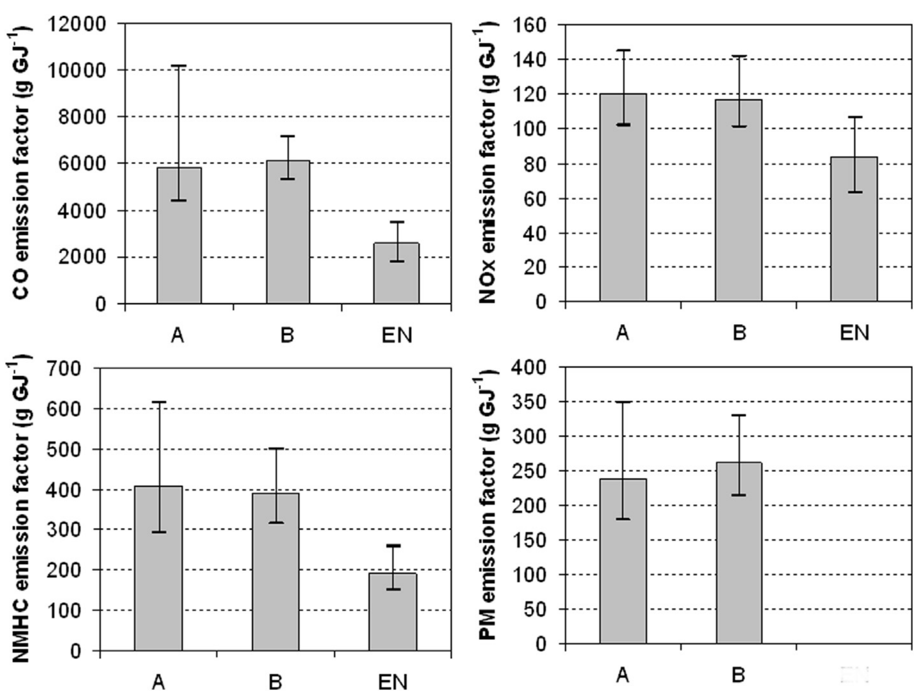

Fig. 5. Average emission factors (gray bars) and the 95% confidence interval of the mean (error bars) for different type of combustion cycle (for A, B, EN specifications, see par. 2.3) for manually fired appliances (closed fireplace, traditional stove and advanced stove).

Table 5
PAH emission factors (mg GJ^{-1}) measured in the study and comparison with average values proposed by AEIG.

Appliance	Fuel type	B(a)P	B(b)F	B(k)F	IP
Open fireplace	Beech	22	30	10	13
	Spruce	32	41	14	21
	Hornbeam	10	13	4.7	4.9
	False acacia	18	22	7.6	11
	Oak	17	22	6.3	10
	Average – this study	20	25	8.5	12
	AEIG suggested	121	111	42	71
	AEIG (95% c.i.)	(12–1210)	(11–1210)	(4–420)	(7–710)
Closed fireplace	Beech	9.5; 8.3	5.6; 5.3	11; 8.3	5.5; 4.4
	Spruce	12; 15	13; 12	7.2; 14	9.2; 15
	Hornbeam	4.7; 8.5	6.8; 13	1.9; 4.4	2.9; 4.7
	False acacia	25; 18	35; 20	12; 14	15; 10
	Oak	25.8; 12.2	33.1; 18	12.1; 5.3	17.3; 8.6
	Average – this study	14	16	9.0	9.2
	AEIG suggested	121	111	42	71
	AEIG (95% c.i.)	(12–1210)	(11–1210)	(4–420)	(7–710)
Traditional wood stove	Beech	56; 52	83; 61	35; 28	30; 40
	Spruce	67	114	72	60
	Hornbeam	566; 44	941; 34	233; 29	442; 30
	False acacia	25	72	35	23
	Oak	66; 100	113; 181	24; 58	92; 61
	Average – this study	122	200	64	97
	AEIG suggested	121	111	42	71
	AEIG (95% c.i.)	(12–1210)	(11–1210)	(4–420)	(7–710)
Advanced wood stove	Beech	204; 94	247; 86	113; 96	116; 64
	Spruce	456; 36	574; 90	252; 29	311; 34
	Hornbeam	153	210	105	90
	False acacia	76	112	43	48
	Oak	48	65	27	26
	Average – this study	152	198	95	99
	AEIG suggested	121	111	42	71
	AEIG (95% c.i.)	(12–1210)	(11–1210)	(4–420)	(7–710)
Pellets stove	High quality pellet	1.9	5.7	2.2	1.4
	Low quality pellet	1.1	2.2	0.8	0.9
	Average – this study	1.5	4.0	1.5	1.1
	AEIG suggested	10	16	5	4
	AEIG (95% c.i.)	(5–20)	(8–32)	(2–10)	(2–8)
Pellets boiler	High quality pellet	0.08	0.10	0.07	0.04
	Low quality pellet	0.04	0.04	0.03	0.02
	Average – this study	0.06	0.07	0.05	0.03
	AEIG suggested	10	16	5	4
	AEIG (95% c.i.)	(5–20)	(8–32)	(2–10)	(2–8)

3.1.4. Influence of fuel load and air supply for batch working (e.g., manually fed) appliances

The closed manual appliances (i.e., closed fireplace, traditional stove, and advanced stove) were tested for the influence of increased fuel load and decreased air supply (Fig. 5) through the application of cycles A and B. As previously described, both cycles consist of an ignition phase and three consecutive fuel loadings, and differ only by the last fuel feed being 1.5 times higher than the nominal value in B cycles together with the complete closure of the primary air valve.

The difference in this last batch does not seem to influence the overall cycle emissions of gaseous compounds. No significant difference is observed between A and B cycles. On the other hand, with respect to EN cycle averages, both cycles present an average increase in the emissions of about 2 times for CO and NMHC emissions and 1.4 times for NO_x emissions.

Regarding the PM emissions a marked difference is observed between cycles A and B for the advanced stove (50% of average PM). For the traditional stove and the closed fireplace, the difference is not so marked in terms of average emissions but median values lower than mean values indicate the presence of very high EFs during some test runs. However, no particular difference is observed between cycles A and B considering the average for all the closed manual appliances.

3.2. PAHs and dioxins emission factors

PAH emissions reported include particulate and semivolatile PAH. B(a)P, B(b)F, B(k)F and IP emissions were determined on the collected samples for all the appliance and fuel types. The calculated EFs in mg GJ^{-1} are reported in Table 5 (and in Table S7 in μg $kg^{-1}{}_{dry\ basis}$) and compared with average Tier 2 values (not detailed for wood type) suggested by AIEG. Experimental results show a prevalence of B(b)F for all the appliances, whereas in the AIEG B(a)P is supposed to give the major contribution. EFs show larger variations according to the appliance type and less variation with fuel type for the same device. Very high EFs (e.g., for hornbeam in the traditional stove, from spruce in the advanced stove) are obtained during some test runs with difficult ignition phase. The repeated testing with the same device and fuel type gave an order of magnitude lower EF. Average PAH emissions from the advanced and traditional stoves were similar and were higher than the closed and open fireplaces.

Emissions for pellet burners are substantially lower compared to other appliances and average data proposed by AIEG. For pellet boilers, after 3 h of PAH sampling the concentrations determined were quite near the detection limit of the instrument used to analyze the extracted material.

EFs for PCDD were measured only for the closed fireplace fed with beech wood and spruce; the average emission factor for two tests is 170 ng I-TEQ GJ^{-1} for spruce and 77 ng I-TEQ GJ^{-1} for beech wood. These values are lower than average Tier 2 values suggested by AIEG.

4. Conclusions

About 300 emission factors for 6 appliances and 5 types of wood were measured mostly in real-world operating conditions. The composite EFs of macropollutants for manually fed appliances are 5858 g GJ^{-1} for CO, 122 g GJ^{-1} for NO*x*, 542 g GJ^{-1} for NMHC, 254 g GJ^{-1} for PM, whereas emissions result to be much lower for automatic pellets appliances: CO 219 g GJ^{-1}, NO*x* 66 g GJ^{-1}, NMHC 5 g GJ^{-1}, PM 85 g GJ^{-1}. Benzo(b)fluoranthene is the PAH with the highest contribution (110 mg GJ^{-1} for manual appliances and 2 mg GJ^{-1} for automatic devices) followed by benzo(a)pyrene (77 mg GJ^{-1} for manual appliances and 0.8 mg GJ^{-1} for automatic devices).

The impact of the parameters outlined in this study (i.e., fuel type, appliance type, combustion cycle) on actual emissions suggests that the emissions in the batch working process are strictly related to the combustion cycle (i.e., real-world cycle vs. type testing cycle) and the seasoning of the firewood rather than the fuel type, because the general emission performance of the manual appliances is observed not to differ notably between different types of commercially available firewood. However the same thing can not be said for continuous pellets burning appliances where the pellets quality (i.e., ash and chlorine content) drastically influences the emission levels, especially of the pellet boiler.

It is known that the design of a heating appliance (i.e., primary and secondary air supply) directly influences the emission characteristics, however the reported results indicate that NMHC, NO*x* and PAH emissions from the advanced stove are similar or higher than the traditional stoves, and are only slightly lower for CO and PM. This finding highlights the importance of the real-world emission factors in the evaluation of the environmental performance of the heating appliances. A partly unexpected result is the lower PAH emissions of the open and closed fireplaces with respect to the investigated stoves.

The EFs proposed by the AEIG and used in the local inventory do not reflect completely the real-world emissions of the manual appliances as the experimental EFs are in most cases higher than what is proposed regarding the major gaseous pollutants; despite this fact the experimental findings for PM emissions are lower by what is expected according to AEIG for the same appliances.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.05.032.

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