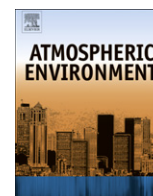


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Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Odor, gaseous and PM₁₀ emissions from small scale combustion of wood types indigenous to Central Europe

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ARTICLE INFO

Article history:

Received 7 April 2011

Received in revised form

18 January 2012

Accepted 19 January 2012

Keywords:

Wood smoke

Odor

Biomass combustion

Particulate emissions

Pellet stove

Log wood stove

ABSTRACT

In this study, we investigated the emissions, including odor, from log wood stoves, burning wood types indigenous to mid-European countries such as Austria, Czech Republic, Hungary, Slovak Republic, Slovenia, Switzerland, as well as Baden-Württemberg and Bavaria (Germany) and South Tyrol (Italy). The investigations were performed with a modern, certified, 8 kW, manually fired log wood stove, and the results were compared to emissions from a modern 9 kW pellet stove. The examined wood types were deciduous species: black locust, black poplar, European hornbeam, European beech, pedunculate oak (also known as “common oak”), sessile oak, turkey oak and conifers: Austrian black pine, European larch, Norway spruce, Scots pine, silver fir, as well as hardwood briquettes. In addition, “garden biomass” such as pine cones, pine needles and dry leaves were burnt in the log wood stove. The pellet stove was fired with softwood pellets.

The composite average emission rates for log wood and briquettes were 2030 mg MJ⁻¹ for CO; 89 mg MJ⁻¹ for NO_x, 311 mg MJ⁻¹ for C_xH_y, 67 mg MJ⁻¹ for particulate matter PM₁₀ and average odor concentration was at 2430 OU m⁻³. CO, C_xH_y and PM₁₀ emissions from pellets combustion were lower by factors of 10, 13 and 3, while considering NO_x – comparable to the log wood emissions. Odor from pellets combustion was not detectable. C_xH_y and PM₁₀ emissions from garden biomass (needles and leaves) burning were 10 times higher than for log wood, while CO and NO_x rise only slightly. Odor levels ranged from not detectable (pellets) to around 19,000 OU m⁻³ (dry leaves). The odor concentration correlated with CO, C_xH_y and PM₁₀. For log wood combustion average odor ranged from 536 OU m⁻³ for hornbeam to 5217 OU m⁻³ for fir, indicating a considerable influence of the wood type on odor concentration.

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1. Introduction

In the current climate issue, biomass combustion is one of the recommended technologies for reducing fossil fuel consumption. Biomass combustion, however, is a source of fine particles (PM), carbon monoxide (CO), nitrogen oxides (NO_x) and volatile organic compounds (VOC), including toxic and carcinogenic constituents, such as polycyclic aromatic hydrocarbons (PAH). The emissions depend crucially on fuel type, combustion technology, combustor

type and individual parameters, such as lighting the fuel and operation habits.

As far as the combustion technology is concerned, highest emitters are manually operated small units (fireplaces, stoves) when fired with wood logs or “primitive fuel” (e.g. leaves, twigs). Fireplaces and stoves emissions have been reported for US wood types (e.g. Fine et al., 2004 and other references cited therein); Northern European wood (e.g. Johansson et al., 2003; Sippula et al., 2007; Boman et al., 2011; Pettersson et al., 2011), Western Mediterranean wood types (Gonçalves et al., 2010), wood from European Alpine regions (e.g. Schmidl et al., 2008, 2011; Bari et al., 2009), and for fuels used in Asia for cooking and heating (e.g. Sheesley et al., 2003).

Little attention has been directed to an additional problem encountered with biomass combustion, the odor. In literature pertaining to odor prevention, biomass combustion is considered as a source, but quantitative data are practically absent. For the

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improvement of working and living conditions, many countries apply regulations to limit odorous emissions from various sources (Henshaw et al., 2006). Nicell (2009) describes facilities, activities and chemicals identified as source of potentially offensive odors. This study does not explicitly mention wood smoke as a potential odorant, however, phenols, which are important constituents of wood smoke (Kjällstrand et al., 2000, Kjällstrand and Petersson, 2001), are considered. Bari et al. (2010), report about wood smoke smell, however not in a quantitative manner. The first source with quantitative odor data from biomass combustion so far appears to be a master thesis (Ebbinghaus, 1993), reporting data from combustion of mixed mid-European wood types with different moisture content in a 40 kW boiler. It reports five-minute averages maximum peaks for dry log wood of 30,000–105,000 OU m⁻³ in wood smoke, with accompanying very high CO emissions of around 4% v/v., indicating prevalence of smoldering combustion conditions.

Around 26% domestic applied thermal energy in EU households comes from renewable sources, among which biomass combustion is predominant (98%) (European Biomass Association, 2007). Being aware of the potential increase in emissions of major and minor pollutants, including odorous substances, we launched a series of investigations including olfactometric measurements, focused on burning wood types from Central Europe, of which several have not been tested until now. Logwood combustion tests were performed on a test stand with a manually operated, typical modern stove used for space heating of homes in the region. The results were compared with an automated, small pellet combustor of newest technology.

This paper reports the results of odor-, gas- and PM₁₀ mass emissions and the dependence of odor levels on gas or particulate matter concentrations. The detailed chemical analyses of particulate emissions including wood smoke tracers and PAH are to be presented in further communications.

2. Experimental

2.1. Stove and fuel choice

Tests with wood logs and “garden biomass” were performed with a modern, 8 kW “chimney type” iron, log wood stove with fireclay lining (“A”). This manually loaded stove is certified to fulfill the current Austrian emission standards defined in state laws, e.g. for Vienna in LGBl. (2005). Combustion air is provided through a grate in the bottom (primary air) and a slit in the back wall (secondary air). Exhaust gases are redirected two times prior to entering the chimney. Airflow is controlled manually via rotary knob connected to a valve system that adjusts both primary and secondary combustion airflow. Our test procedure for log wood and briquettes included two fuel loads starting from the cold stove and adding the second load after burn down of the previous load (when CO₂ concentration in exhaust falls below 3%). Each load consisted of around 1.3 kg of wood (2 or 3 wood logs sized on the average 3 × 5 × 25 cm, assuring a full power output), totaling around 2.6 kg fuel per test (Table 2). Ignition was performed with commercial lighter cubes (2–3 cubes, trade name “ECOMA”). The test ended when the CO₂ concentration in exhaust dropped below 3%. The total duration of a test procedure was 50–80 min. The tests with garden biomass were performed with around 1.2 kg of fuel and the test duration was 20–30 min (Table 2). The stove was filled for the first load, then ignited and several times refueled.

Due to sampling over the whole burning (inclusive start up phase) the test procedure is close to “reality” since the products of incomplete combustion of the ignition are included in our measurement cycle.

The second test stove (“B”) is a modern automatically fired pellet burner with internal pellet storage. The fuel is supplied to the combustion chamber via an “auger screw”. Combustion air supply is adjusted adequately for the selected thermal output by a fan situated in the flue gas stream. Air enters through holes under the fuel bed. Both fuel load and fan speed are controlled automatically, only the percentage of power output (30–100%) is set by the user. Our test consisted of part load runs of about 75 min and full load runs of around 80–370. Ignition took place with an electrical resistance heater and air supply from a blower. The ignition phase was not included in tests with pellets.

Fuel woods were selected among tree species indigenous for Austria (AT), and neighboring countries or regions: Czech Republic (CZ), Hungary (HU), Slovak Republic (SK), Slovenia (SI), Switzerland (CH), Baden-Württemberg and Bavaria (South Germany, DE) and South Tyrol (Northern Italy, IT). Tree species with abundance of more than 5% of the forest area in each country have been chosen for the test burns and are given in Table 1. The data were obtained from national forest inventories: Austrian Forest Inventory, 2010; Forest Inventory, Germany, 2010; Italian National Forest Inventory, 2010; Swiss Forest Inventory, 2010; as well as from national forest reports: Forest Report Hungary, 2009; Forest Report Czech Republic, 2008; Cop, 2007; Moravcik, 2007. Cylindrical hardwood briquettes (“Forest briketi” 28 cm, 8 cm diameter) were obtained from “Istrabenz Energetski Sistemi”, Slovenia.

In the series of 44 measurements seven types of hardwood, five types of softwood, briquettes (beech) and wood pellets were burned. In addition, combustions of pine cones, dry pine needles and dry leaves (orchard leaves and maple) were performed to obtain emissions from non conventional material, sometimes used for starting the firing of wood logs. The fuels were obtained from local markets or from private forest owners in Austria and Hungary, “garden biomass” (cones, needles and leaves) from a private garden. Logs were stored in a dry place to achieve relatively stable water content recommended for fuel wood. Elemental composition, ash and water content of tested fuels are listed in Table 1s in the supplement. Sampling and calculation details are presented in Table 2.

2.2. Gas emission measurement

Gas emission measurements started with the ignition of the fuel and were performed at a test stand (Notified Testing Laboratory for Combustion Systems, Institute of Chemical Engineering Vienna University of Technology). The gaseous emissions were determined continuously over the test period with a time resolution of 10 s. The test stand maintains a static pressure of 12 Pa in the chimney during the test by means of a blower. Measurements of exhaust gases followed in general the standards for automatically fired stoves (DIN EN 14785, 2006) and for manually fired systems (DIN EN 13240, 2001). Carbon monoxide (CO), nitrogen oxides (NO_x) and total gaseous organic compounds (C_xH_y) were assessed, as these emissions are regulated by legislation in Austria. The amount of oxygen (O_{2meas}) and carbon dioxide (CO₂) in the exhaust gas were determined in order to assess the combustion conditions.

The calculation procedure of the normalized concentrations is described in the supplement.

2.3. Particulate matter sampling

Particulate matter (PM_{2.5}, PM₁₀ and total particulate fraction) was collected on filters after pre-dilution of the exhaust, over the whole combustion time (two continuous full-load runs). The dilution apparatus was constructed for previous studies and consists of two sampling lines, which were modified, for the current project purposes. The detailed characterization of the

Table 1
Share of tree species in forests of selected Central European countries. (Sources: see text).

Wood	AT	DE	CZ	HU	SK	SI	IT	CH
Total forest cover [% of land area]	47	37	36	20	41	60	45	30
Botanic name	Common name [% of woodland area]							
<i>Carpinus betulus</i>	European hornbeam							
<i>Fagus sylvatica</i>	10	16	7	5	6	31	31	1
<i>Populus nigra</i>	Black poplar							
<i>Quercus cerris</i>	2^b	7^b	7^b	4(11)^d	11	7^b		2^b
<i>Quercus petraea</i>	Sessile oak							
<i>Quercus robur</i>	Pedunculate oak							
<i>Robinia pseudoacacia</i>	Black locust							
Other deciduous	12	13	9	11	7	14	2	13
Total deciduous	24 ^e	36	24	88	59	52	7	34
<i>Abies alba</i>	Silver fir							
<i>Larix decidua</i>	European larch							
<i>Picea abies</i>	54	42	53	23	32	32	53	42
<i>Pinus nigra</i>	Austrian black pine							
<i>Pinus sylvestris</i>	Scots pine							
Other coniferous	1	1		12	1	2	11	1
Total coniferous	67 ^e	64	75	12	41	48	93	66

^a 4% correspond to Black poplar, while 11% is a contingent of all poplar types (including Hybrid poplar).

^b Sum of all oak species.

^c Sum of temperate oak species (*Q. petraea*, *Q. robur*).

^d Sum of all pine species.

^e Remaining 9% is dedicated to blanks, gaps and shrubs, for abbreviations see text.

original particle sampling system is given in Schmidl et al. (2011). Our modifications included two separate lines for PM₁₀ and PM_{2.5} and a line without a pre-separation stage. The internal volume of dilution tunnel is 0.04 m³, resulting in a residence time in the tunnel of 60 s. The sampling flow velocity into the PM₁₀/PM_{2.5} sampling heads was higher than the flow in the dilution tunnel with $U_o/U = 0.3$, leading to super-isokinetic conditions. Sampling occurs at near ambient temperatures.

Most of the European studies so far used quartz, glass fiber or organic based front-filters for sampling without a back-filter for sampling artifact corrections (e.g. Kocbach et al., 2006; Boman et al., 2011). Jordan and Seen (2005) used a quartz front – quartz back up filter combination and observed 4% of the mass of the front filter at the back up filter.

Thus we notice that we have not used a back up filter to infer positive artifacts for the deposit on the front filter. This type of sampling, however, is until now quite common in Europe and thus

our results are comparable to the results from earlier European studies. Details and appropriate discussion are in the [on-line supplement](#).

2.4. Odor sampling

For the olfactometric analyses, about 20 liters of flue gas was collected continuously behind the dilution tunnel (same as for particulate matter sampling), during the entire burning process. Pre-dilution factors of 10–20 assured that no condensation of water vapor occurred before and during the measurements. According to the sampling standards for odor assessment given in European Union norm DIN EN 13725 (2003), a smell- and tasteless, liquid- and temperature resistant (–60–220 °C) Nalophan (polyethylene terephthalate, Kalle) sampling bag was used. The inlet of the sampling bag was equipped with an odorless Teflon tube. The whole equipment was placed in a tight Plexiglas pipe. To achieve

Table 2
Sampling and calculation details.

Fuel name	Index	Stove	Burning type	Number of tests	Dilution factor	Wet fuel mass [kg]	Burn rate [kg h ⁻¹]	LHV [kJ kg ⁻¹]	Average T [°C] in flue gas	MCE factor [%]	O ₂ [% v/v]	CO ₂ [% v/v]
Wood pellets	WP	B	FL1	2	3	2.3	1.4	17,706	132	99.8	14.2	6.2
	WP	B	FL2	1	3	12.6	1.9	17,706	130	99.7	13.9	6.4
	WP	B	PL	2	3(4) ^a	1.00	0.7	17,706	92	99.6	17.5	3.1
Wood briquettes	BR	A	FL	3	10	2.65	2.0	17,177	183	97.5	15.6	4.8
European hornbeam	EH	A	FL	3	10	2.63	2.5	16,117	184	98.0	14.4	6.0
European beech	EB	A	FL	3	10	2.63	2.8	15,857	195	97.8	13.2	7.1
Black poplar	BPop	A	FL	3	10	2.59	2.2	16,778	109	96.8	15.4	5.0
Turkey oak	TO	A	FL	3	10	2.61	2.7	16,709	196	96.9	13.8	6.5
Sessile oak	SO	A	FL	3	10	2.64	2.1	16,164	145	93.9	16.9	3.5
Pedunculate oak	PO	A	FL	2	10	2.63	2.2	15,750	154	94.8	16.7	3.7
Black locust	BL	A	FL	3	10	2.66	1.9	16,676	149	96.5	16.3	4.2
Silver fir	SF	A	FL	3	10	2.61	2.0	16,686	175	94.3	15.6	4.6
European larch	EL	A	FL	1	10	2.63	2.5	17,162	176	97.7	13.8	6.5
Norway spruce	NS	A	FL	3	10	2.61	2.1	16,481	174	96.9	15.3	5.0
Black pine	BP	A	FL	3	10	2.61	2.3	17,610	179	97.1	14.7	5.4
Scots pine	SP	A	FL	3	10	2.63	2.1	–	170	97.9	14.4	5.8
Dry leaves	L	A	ST	1	15	1.25	5.3	17,364	196	95.5	13.6	6.3
Pine cones	C	A	ST	1	15	1.14	3.3	17,265	175	95.4	11.7	8.3
Pine needles	N	A	ST	1	20	1.14	4.6	18,761	192	95.7	11.7	8.0

FL, full-load operation; PL, part-load operation; ST, special test; LHV, lower heating value; MCE, modified combustion efficiency calculated as a ratio $c_{CO_2}/(c_{CO_2} + c_{CO})$ where c_{CO_2} and c_{CO} are the measured volume mixing ratios of CO₂ and CO.

^a Different dilution factors of both tests.

a stable flow of the sample into the sampling bag, air was pumped constantly out of the pipe. The flow rate, set and regulated with mass flow controllers (MFC, Bronkhorst), varied between 0.25 and 0.35 L min⁻¹, depending on combustion time. After a single measurement, the sampling bag was purged with purified air. Being used for one fuel type, it was replaced by a new one.

2.5. Analytical procedures

2.5.1. Filter weighing and sample preparation

Previous to sampling, quartz fiber filters were baked for 5 h at 550 °C, cooled in a desiccator with water vapor saturated atmosphere, equilibrated for 48 h in an air conditioned room (20 ± 1 °C, 50 ± 5% relative humidity) and weighed with a microbalance (Sartorius M5P with range up to 1 g ± 0.5 µg). The same procedure was repeated with loaded filters. Loaded filters were stored at -18 °C.

2.5.2. Olfactometry

Olfactometry is a widely used method, which allows measuring of odor concentration (given as Odor Units "OU" per cubic meter) and deriving the odor thresholds (Hudson and Ayoko, 2008a,b).

The measurement of odor was performed by dynamic dilution olfactometry, involving a self-made diluting device (olfactometer, Fig. 1s in supplement) and a panel of four people, who determine the odor threshold, i.e. the dilution ratio, where no smell is experienced.

The pre-diluted odor sample was mixed with purified air (active carbon, particulate filter and silica gel), in ratios ranging between 1:10 and 1:10,000. Usually 10–20 dilutions, in a random order, were presented to panelists during a single measurement. Binary answers (yes/no) were collected discretely.

The dilution was performed with four suspended-body flow meters. One of them controlled the neutral air flow, which was constant during the whole measurement (~15 L min⁻¹). The other three flow meters worked in different ranges and were adjusted manually to set a desired dilution. The dilutions achieved with the device are comparable to short-time dilutions in ambient air.

In order to estimate the odor thresholds the answers of the panel members were subjected to a probit analysis, which is a well established method for modeling probabilities of binary outcomes (Finney, 1971). We decided to use a probit instead of a logit model solely for convenience reasons, since probit models are easier to interpret. Our choice of the probit model is also supported by a work of Chambers and Cox (1967), who showed that extremely large samples, are necessary, in order to distinguish between probit and logit models.

3. Results and discussion

The study shows a wide diversity of emission factors according to fuel, appliance and combustion mode. Expectations were to investigate situations, which happen in real life and not only under laboratory conditions. For this reason no special treatment of fuels and stoves was performed. Emphasis was put on keeping the firing chamber clean and to assure the recommended wood volume and proper air supply. In consequence, quite high relative standard deviations between different tests of a single wood type under identical firing conditions were observed.

To obtain information about the combustion efficiency we derived the modified combustion efficiency factor (MCE) given in Table 3. MCE values >90% suggest that more than 50% of the emissions were produced by flaming combustion, whereas when MCE <90%, more than 50% of the emissions originated in the smoldering phase (Ward and Hardy, 1991). All tests in this study

Table 3
Average emission factors.

Fuel	CO [mg MJ ⁻¹]	NO _x [mg MJ ⁻¹]	C _x H _y [mg MJ ⁻¹]	PM10 [mg MJ ⁻¹]	Odor [OU]
WP FL1	118	94	58	21	ND
WPFL2	188	131	5	31	ND
WP PL	245	74	8	16	ND
BR	1482	63	163	32	1804
EH	1234	110	462	41	536
EB	1410	95	234	66	1563
BPop	1856	65	216	20	2843
TO	1816	88	206	59	1781
SO	3681	131	657	222	4226
PO	3253	104	452	57	1973
BL	2000	118	239	67	1689
SF	3497	105	581	100	5217
EL ^a	1263	58	179	21	2422
NS	1901	69	267	53	3815
BP	1710	64	243	101	1589
SP	1189	70	109	53	2134
L ^a	2249	132	1543	626	18963
C ^a	2821	89	1106	<LOD	3524
N ^a	2204	111	1424	85	7346

<LOD, under limit of detection; ND, not detected, ranges given in supplementary material in Table 2s.

^a Single test.

represent emissions predominantly from the flaming combustion, with an average MCE of 96.8%.

3.1. Gas phase and particle emissions

Emission factors obtained in the study are presented in Table 3. Observed ranges for the individual tests are given in Table 2s in the supplement. In case of pine cones the PM₁₀ emissions were, due to relatively fast (in comparison with wood) and clean (in comparison to other garden waste) combustion, not detectable. The emission factors ranged from 16 to 625 mg MJ⁻¹ with surprisingly low values for poplar and larch, similar to the emissions from the pellet stove, relatively high emissions for sessile oak (221 mg MJ⁻¹) and an extremely high value for unconventional fuel (dry leaves) combustion. Pine needles smoke PM₁₀ was well in the range of log wood. CO and NO_x emissions of the garden biomass samples were within the range of the emissions from log wood, while C_xH_y emissions were higher for all types of garden biomass.

Automated regulation of fuel and combustion air supply in the pellet stove assured efficient and relatively clean combustion with low particulate emissions. The tested device showed a burning rate dependent PM₁₀ emission – increasing PM₁₀ with increasing burning rates: 16 mg MJ⁻¹ PM₁₀ at 0.7 kg h⁻¹; 20 mg MJ⁻¹ at 1.4 kg h⁻¹ and 30 mg MJ⁻¹ at 1.9 kg h⁻¹. A previous study with the same appliance reported even lower PM₁₀ emissions from wood pellets of around 3 mg MJ⁻¹ for full load and 10 mg MJ⁻¹ for part-load (Schmidl et al., 2011). Scandinavian studies reported higher PM emissions from pellet stoves. Sippula et al. (2007) obtained a PM₁ emission rate of 58 mg MJ⁻¹ for commercial pellets fired in an 8 kW automated German pellet stove ("Wodtke GmbH"). Boman et al. (2011) investigated PM_{tot} emissions from a Scandinavian 6 kW and a North American 9.5 kW "exempt" pellet stove and reported 16 and 23 mg MJ⁻¹ for full load respectively 34 and 40 mg MJ⁻¹ for part load operation.

Interestingly, two wood types tested here (poplar and larch), as well as briquettes emitted equally low amounts of PM₁₀, as observed for pellets. This result is surprising, because pellets combustion was considered consistently "cleaner" than log wood. Log wood exhibited, depending on the wood type, a wide range of PM₁₀ emissions (20–221 mg MJ⁻¹) what is depicted in Fig. 1. The

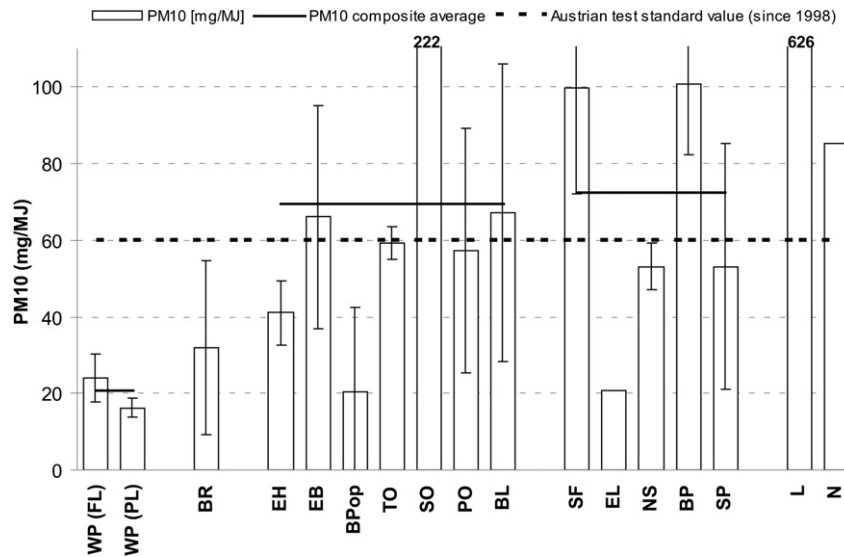


Fig. 1. Average emission rates of PM_{10} . Error whiskers in terms of standard deviation. Dashed line represents the Austrian emission limit for standardized test conditions.

lowest emissions obtained are around two orders of magnitude higher if compared to the PM_{10} emission from combustion of extra light fuel oil in domestic boilers (Hübner et al., 1996; Austrian Energy Report, 2003).

Relatively low emissions in the pellets tests during both burning modes (full- or part-load) were obtained for CO (142 and 245 $mg MJ^{-1}$) and C_xH_y , (40 and 8 $mg MJ^{-1}$). Contrary to pellets, wood logs burned in full-load mode in the manually fired stove caused in many cases much higher emissions. Among 12 wood types and briquettes, the highest values for CO and C_xH_y were obtained for sessile oak (3681 $mg MJ^{-1}$ CO and 657 $mg MJ^{-1}$ C_xH_y), and the lowest values (1189 CO $mg MJ^{-1}$ and 109 $mg MJ^{-1}$ C_xH_y) for Scots pine. The most common fuel woods in Austria, spruce and beech show likewise an emission of 1900 and 1400 $mg MJ^{-1}$ CO, 267 and 234 $mg MJ^{-1}$ C_xH_y , respectively of 53 and 66 $mg MJ^{-1}$ PM_{10} .

The PM_{10} emission rates depend on the burning quality (expressed in terms of MCE factor) which is strongly influenced by the relation of smoldering and flaming combustion phase (Fig. 1s in supplementary material).

Nitrogen oxides emissions show a different behavior than other determined components. Test averages range between 58 and 131 $mg MJ^{-1}$, with lowest values for larch (58 $mg MJ^{-1}$) and highest for sessile oak (131 $mg MJ^{-1}$) and dry leaves (132 $mg MJ^{-1}$) smoke. The NO_x emissions from pellets combustion were close to the median of the tested fuels. However, there is a statistically significant difference of 30% for the NO_x emissions from hard- and softwood combustion (102 and 75 $mg MJ^{-1}$ NO_x). There is an obvious influence of the nitrogen content in the fuel with 0.16% N for hardwood, 0.08% N for softwood and 0.95% for dry leaves (averaged data from Table 1s), however also combustion conditions

e.g. temperature seem to play a significant role (Fig. 3s in supplementary material).

Summarized composite data for pellets, wood fuels with briquettes, hardwood and softwood, as well as “garden biomass” (excluding cones) in different units simplifies comparison with results from other studies, and are of interest for emission inventories and modeling purposes (Table 3s supplement, data discussed therein).

In 1997 and 1998, a comprehensive emission test of Austrian domestic combustion units was performed at 180 randomly chosen households, thus considering “real world” conditions in a large variety of boilers and stoves. This data is compiled in Austrian Energy Report (2003). The average emission rates for log wood stove survey are included in Table 4. The PM_{tot} results, however, were obtained by “hot” sampling, which underestimates the particle concentrations occurring in the ambient air. Pettersson et al. (2011) discussed two results from Scandinavian sources (Bäfver, 2008; Ortega, 2008) where diluted mass emissions from wood stoves were 3–9 and 5–12 times higher, than for undiluted “hot” sampling.

CO, C_xH_y and PM_{10} emissions from the modern, certified “chimney” stove chosen for this study are around a factor of two lower than the respective emissions from the “Austrian” stoves collective. We can conclude from this result, that modern, certified stoves tend to emit notably less CO, C_xH_y and PM_{10} compared to the typical “Austrian average” which included a wide variety and age range of devices under undefined operation conditions.

The emission limits of the Austrian certification procedure (beech wood, two combustion cycles, standardized conditions, hot start), mentioned above are presented in Table 4. Comparing our test results, we have to consider the differences to the standard test

Table 4

Comparison of emission rates for small log wood stoves from an emission survey of Austrian log wood stoves from 1997/98, averaged results for modern log wood stove tests, and the emission limits for new log wood stoves in Austria.

	CO [$mg MJ^{-1}$]	NO_x [$mg MJ^{-1}$]	C_xH_y [$mg MJ^{-1}$]	PM [$mg MJ^{-1}$]
Austrian test 1997/98 log wood stoves	4463	106	664	148 PM_{tot} “hot”
This test – 8 kW stove, wood logs and BR	2030	89	311	67 PM_{10}
Schmidl et al. (2011), (EB, PO, SP, BR) average from 2 stoves (6 and 6.5 kW)	1454	91	125	81 PM_{10}
Pettersson et al. (2011) birch, NS, pine 9 kW stove, average from 3 modes	3600	47	820	140 PM_{tot}
New Austrian Standard 1998 (beech, “hot start”)	1100	150	80	60 PM_{tot} “hot”

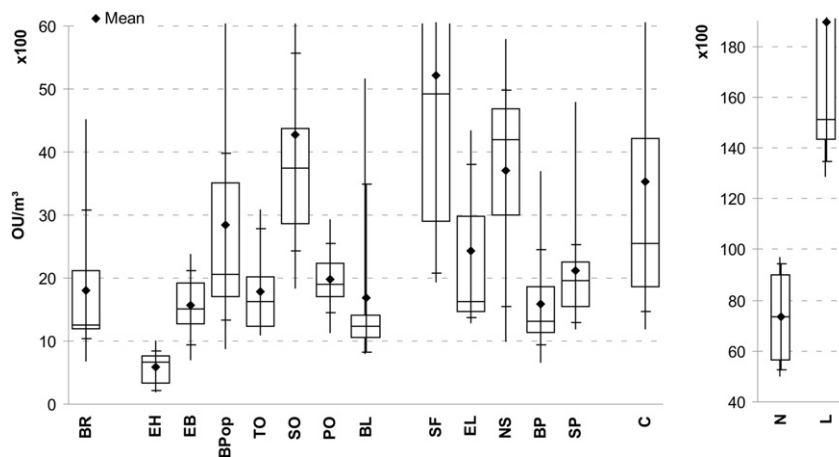


Fig. 2. Odor thresholds (OU m^{-3}). Whiskers present 10 and 90 percentile as well as maximal and minimal values. Garden biomass – needles and dry leaves are presented with separate scaling.

procedure (i.e. including most of the incensing phase and choosing other wood types). Average emissions of CO and C_xH_y , are exceeding the emission limit for 90% of tested fuels, PM_{10} – for 39% of the tested fuels. In case of NO_x average results for individual wood types were within the limit. Averaged CO and C_xH_y emission rates are considerably above the standard limits: CO around a factor of two, C_yH_y nearly a factor of four higher. Certain wood types exhibit the emission rates even higher than the average obtained in the current test, which allows the conclusion, that country specific emission rates have to be derived considering the different wood types available in different countries of Europe.

3.2. Odorous emissions from wood combustion

Our aim was to create a matrix of odorous substances emission data for different wood types to derive information about the occurrence of wood smoke odor dependent on other wood smoke parameters, which are currently used in dispersion models in order to derive information about the odor nuisance potential in wood burning communities. Due to this intention, our study concentrated on average odor thresholds for the complete burning processes and not on individual combustion phases. Odor phenomena are

considerable for manually fired stoves, while no odor was detected for pellet combustion in the automatically fired stove. The odor thresholds for briquettes, wood logs and garden biomass combustion range between 536 and 18,963 OU m^{-3} (Table 3s). A high variability among each measurement relates to the individual character of the test with panelists. This is reflected by box plot (Fig. 2).

Odor is a sensorial signal caused by compounds, which are volatile at room temperature (Rossiter, 1996), thus odor threshold is considered to correlate with the concentration of gaseous organic compounds in the exhaust.

The highest odor concentration was observed by burning dry leaves. The result differs strongly (one order of magnitude) from wood smoke. The highest wood smoke odor threshold (5217 OU m^{-3}) was observed for fir smoke, the lowest, 536 OU m^{-3} for hornbeam smoke. The average odor threshold of hardwood smoke (2087 OU m^{-3}) is lower than that of softwood smoke (3036 OU m^{-3}), although among both fuel groups there are high- and low emitting species. This behavior of smoke from different wood types can be observed in the correlation with CO emissions depicted in Fig. 3. The correlation is significant at the P95 level. Contrary to odor, the average emissions of C_xH_y show the reversed

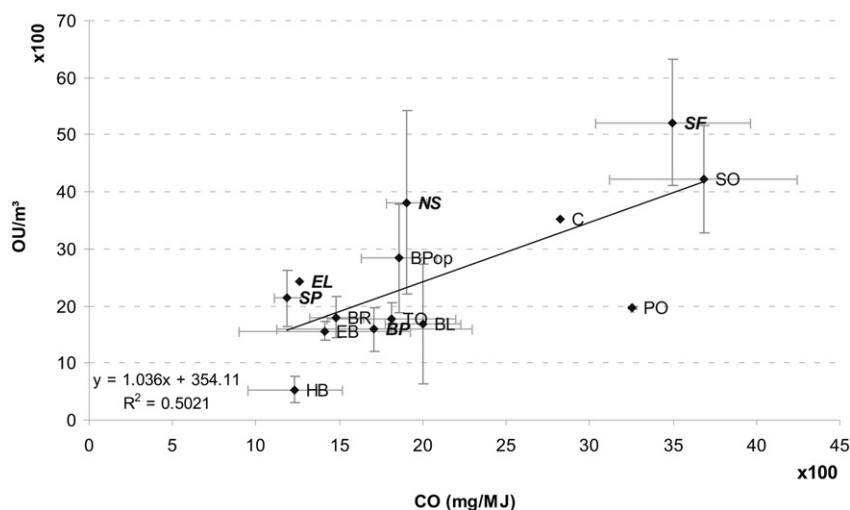


Fig. 3. Correlation of odor and CO showing high and low emitters among fuels. Softwood types are marked with bold italics. Whiskers present standard deviations. The correlation is significant at the P95 level.

order (Table 3). For CO and PM₁₀ the emissions from hard- and softwood were similar.

The emission rates of measured pollutants depend closely on combustion efficiency (in terms of MCE). Positive slopes are reported for emissions generated mainly by flaming combustion, while negative slopes originate from emissions by smoldering processes (Sinha et al., 2003). Current sampling was conducted over a complete burning process thus negative slopes for the odor, PM₁₀ and C_xH_y emissions in the plot against MCE (Fig. 2s in the supplementary material), which were observed indicate that the emissions of a complete burning cycle are dominated by smoldering that occurs during the start-up and end phases of burning, when no more flames are observable.

Relying on the fact that odor is mainly emitted during smoldering we are able to explain the differences between odor concentrations in our experiment and the study of Ebbinghaus (1993). The highest reported values for log wood, from 30,000–105,000 OU m⁻³ were recorded in 5 min intervals during start- and end – phase at extremely high CO levels, while we report an averaged odor threshold over the whole combustion cycle.

Although odor thresholds among different wood types do not vary too much, for most softwood species the odor concentration was higher than for hardwood species. A potential explanation might give the guaiacol (2-methoxyphenol) content in the exhaust gas. Guaiacol is possibly the leading odorous substance in wood smoke, considered to be responsible for smoky smell (Czerny and Buettner, 2009) and it occurs generally in higher amounts in softwood than in hardwood smoke (Kjällstrand and Petersson, 2001). A combined study on methoxyphenols and wood smoke odor relationship is still required to prove this hypothesis.

An interesting diagnostic ratio in wood smoke is its relationship to particles. This ratio can be applied to derive the ambient PM concentration from wood smoke, which is accompanied by exceeding the odor threshold, i.e. the potential for an odor perception. It gives also a possibility to estimate the odor concentration if ambient wood smoke concentrations are known from tracer measurements. Table 5 compiles the averaged ratios PM₁₀ [μg m⁻³]/Odor [OU m⁻³] for tested fuels. The “Austrian mix” is based on estimated fuel data given in Schmidl et al. (2008) (70% spruce, 20% beech and 10% briquettes) and is derived according to Equation (1), where C_x is an ambient PM concentration for selected species, which allows odor perception.

$$C_{\text{Austrian mix}} = 0.7C_{\text{spruce}} + 0.2C_{\text{beech}} + 0.1C_{\text{briquettes}} \quad (1)$$

Applying the relationship of odor threshold and PM₁₀ emission rates to PM ambient concentrations, we obtain that for the “Austrian mix” smoke odor could be perceived at the ambient wood smoke concentration of already about 30 μg m⁻³.

High wood smoke concentrations reported for suburban and urban sites in pre-Alpine forested environments, e.g. in Styria and

Salzburg on days exceeding the EU daily mean limit value for PM₁₀ of 50 μg m⁻³ (EC, 2008) were in the range of 20–30 μg m⁻³ as average over a period of 2–7 days (Caseiro et al., 2009). Due to the daily variation of domestic emissions occurring from chimneys at low elevation above ground, namely in stagnant periods, hourly maxima of wood smoke PM are expected to exceed considerably the daily averages. Thus, wood smoke odor levels above the threshold level are likely during winter days in wood burning communities situated in areas with reduced ventilation.

4. Conclusions

Among fuels chosen for combustion tests are species dominant in lowland as well as in mountainous forests of Europe, thus the emission data are of relevance for many countries. Furthermore, combustion emissions of the main tree types of the Pannonian lowlands have not been reported before.

Results showed a large variation among fuel types, e.g. PM₁₀ emissions from European larch and black poplar were a factor of ten lower than from sessile oak; likewise odor concentration in European hornbeam smoke were a factor of ten lower than in silver fir smoke. The variability of emission rates combined with the diversity in the tree species distribution in European countries appeals to the presumption of different averaged emission rates of wood smoke components for different European countries.

The data of this and comparable studies show, that modern wood combustion technologies for small scale appliances, are lowering the emission of particles for manually fired log wood stoves to around 70–140 mg MJ⁻¹. This is in the lower part of the range indicated in the review by Kocbach Bølling et al. (2009). The observed PM emissions for small pellet stoves were quite in the range given in the review (10–50 mg MJ⁻¹). The emission of wood and pellet stoves is predominantly in the fine (<2.5 μm) size range, thus a similar emission of PM_{2.5} can be anticipated. Considering a typical heat demand of 100 GJ/Home a pellet appliance equipped home would emit around 1–6 kg of fine particles per year. This is still 1–2 orders of magnitude higher than emissions from gas or oil (extra light, low sulfur) based heat supply. The planned large scale introduction of wood combustion based domestic heating systems in Austria (planned for up to 400,000 households) would increase the Austrian PM_{2.5} emission by around 6% based on an average emission of 30 mg MJ⁻¹, if the old “high emitting” solid fuel based heating systems are not replaced. As most of the wood smoke emissions are occurring during the cold season, their impact during the cold period would range up to a 12% increase for ambient PM_{2.5} (PM₁₀) concentrations.

From the dependence of C_xH_y, PM₁₀, odor and the MCE it could be derived, that the respective emissions during the complete burning cycle as sampled in our study, are dominated by the smoldering phases that are occurring during the start-up and end phases of burning when no more flames are observable. We have started the log wood combustion tests with relatively smokeless lighter cubes. In practice, wood combustion is often started with waste paper, kindle wood, twigs and other igniting material. This causes far higher fine particles emissions in the ignition phase, as can be anticipated from our tests with garden leaves and needles. Thus, the emission rates observed, though considered to be close to “real world” conditions are at the low end of the actual occurring emissions.

The relationship of PM₁₀ and odor concentration was used to derive a “critical” ambient PM₁₀ level from wood smoke where wood smoke odor becomes perceivable. The PM₁₀ level, where wood smoke odor exceeds the odor threshold was around 40 μg m⁻³ for softwood and 60 μg m⁻³ for hardwood. Peak emissions during the incensing phases are, however, a factor of 10–50 higher than the burning cycle averages, as derived from the

Table 5
PM₁₀ [μg m⁻³]/Odor [OU m⁻³] ratios for composite “hardwood”, “softwood” and “Austrian mix”.

	PM [μg m ⁻³]/odor [OU m ⁻³]	
	Average	Median
Wood pellets	–	–
Wood logs and briquettes	53.4	43.0
Hardwood logs	66.4	62.1
Softwood logs	40.4	28.8
Needles and leaves	39.6	27.7
Austrian mix ^a	29.8	–

^a 70% spruce, 30% beech and 10% briquettes (Equation (1)).

results of the Ebbinghaus (1993) study. Thus frequent short term occurrences of wood smoke odor is likely for many communities with traditional and modern wood stove use.

The odor units can be used in dispersion models to estimate the impact of odor nuisance from wood combustion.

Acknowledgements

This is a part of the project “Wood smoke impact on particulate matter (PM) and odor levels in Austria” funded by Austrian Science Foundation (FWF) Project Nr. P20567-N19. We thank Richard Triebaumer, Carlos Ramirez – Santa Cruz and the family Bognar for fuel supply and Dr. Josef Rendl for collecting garden biomass.

Appendix. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.atmosenv.2012.01.044.

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